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Publication date: 2014

Document Version
Peer reviewed version

Citation (APA):

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Modelling the Pultrusion Process of Off Shore Wind Turbine Blades

by

Ismet Baran

Ph.D. Thesis

Department of Mechanical Engineering
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May, 2014
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TM xx-xx

ISBN xxx-xx-xxxxx-xx-x
To the memory of my beloved grandmother Meliha
Preface

This PhD thesis is a part of DeepWind project which has been granted by the European Commission (EC), Grant 256769 FP7 Energy 2010, under the platform Future Emerging Technology. The studies have been carried out under the Work Package 2 (WP2) entitled “Development of modelling tool for manufacturing process” at the “Process Modelling Group” headed by Professor Jesper H. Hattel, Department of Mechanical Engineering (MEK), Technical University of Denmark (DTU) during the period 2011-2014.

First and foremost I would like to express my deepest appreciation to my main supervisor Professor Jesper H. Hattel for his continuous support and perfect guidance from beginning to end of my PhD work. His friendship and encouragement throughout the PhD work were the key to experience a rewarding and a memorable research. I appreciate all his contributions of time, ideas, and funding to make my PhD experience productive and stimulating. I would like to express my sincere thanks and deep gratitude to my co-supervisor Dr. Cem C. Tutum, who as a good friend, for his constant motivation and untiring help during the course of my PhD. He was always willing to give his best suggestions on both academic and personal level. I would also like to thank my external co-supervisor Per Hørlyk Nielsen for his support and help during my PhD.

I am indebted to Professor Remko Akkerman, who is the head of the “Chair of Production Technology” at University of Twente (UT), The Netherlands, for his supervision in a calm and truly inspiring atmosphere. I thank him for fruitful scientific discussions and outstanding guidance during my external research state at UT.

I would like to express my sincere gratitude to Dr. Pierpaolo Carlone for our deep discussions and constructive collaborations on the pultrusion process.

Many thanks to my colleagues and fellow PhD students at DTU MEK and I would like to give special thanks to current and former Process Modelling Group members for creating an inspiring environment. Moreover, I acknowledge the Production Technology research group members at UT.

I would like to thank administrative staff members of DTU. Special thanks goes to Pia Holst Nielsen and Anette Fournaïs Kaltoft for their efforts to make life easy during my PhD.

Many thanks to Dr. Cüneyt Baykal for his sincere friendship and neighbourhood while living together at the same building at Daugaardsvej.

I would like to dedicate this thesis to the memory of my beloved grandmother Meliha
whose role in my life was, and remains, immense. Last but not least, I would like to extend my deep appreciation to my wonderful wife Didem for her love, patience and understanding and to my angel mother Türkan for believing in me and encouraging me to follow my dreams.

Ismet Baran
Kgs. Lyngby, May 2014
Abstract

This thesis is devoted to the numerical modelling of the pultrusion process for industrial products such as wind turbine blades and structural profiles. The main focus is on the thermo-chemical and mechanical analyses of the process in which the process induced stresses and shape distortions together with the thermal and cure developments are addressed. A detailed survey on pultrusion is presented including numerical and experimental studies available in the literature since the 1980s. Keeping the multi-physics and large amount of variables involved in the pultrusion process in mind, a satisfactory experimental analysis for the production requires considerable time which is obviously not a cost-efficient approach. Therefore, the development of suitable computational models is highly desired in order to analyse the process for different composite manufacturing aspects such as heat transfer, curing and solid mechanics.

In order to have a better understanding of the processing polymer behaviour in pultrusion, the chemo-rheology of an industrial “orthophthalic” polyester resin system specifically prepared for a pultrusion process has been characterized. The curing behaviour is first characterized using differential scanning calorimetry (DSC). Isothermal and dynamic scans are performed to develop a cure kinetics model which accurately predicts the cure rate evolutions. The resin viscosity and the gelation point are subsequently obtained from rheological experiments using a rheometer. Based on this, a resin viscosity model as a function of temperature and degree of cure is developed and it is found to predict the measured viscosity correctly. The temperature- and cure-dependent elastic modulus of the resin is determined using a dynamic mechanical analyzer (DMA) in tension mode. A cure hardening and thermal softening model is developed and a least squares non-linear regression analysis is performed. The predicted best fit results are found to agree quite well with the measured data.

The temperature and degree of cure distributions inside the processing material have been calculated using the developed thermo-chemical numerical process models and subsequently used in the mechanical analysis of the pultrusion. The effects of the thermal contact resistance (TCR) at the die-part interface of a pultruded part are investigated using a two dimensional (2D) thermo-chemical model. It is found that the use of a variable TCR is more convenient than the use of a constant TCR for the simulation of the process. The 3D thermo-chemical modelling strategies of a thermosetting pultrusion process are investigated considering both transient and steady state approaches. So far in the literature, the pultrusion process of a relatively thick composite having a curved cross sectional geometry such as the NACA0018 blade profile has not been modelled numerically. Hence, a numerical simulation tool embracing the blade manufacturing process has been developed in this thesis. The effects of the heater configuration and pulling speed on the pultruded blade profile have been addressed by means of the devised numerical
In addition to the efficient thermo-chemical models developed in this thesis, state-of-the-art mechanical models have also been developed by the author to predict the process induced stresses and shape distortions in the pultrusion process. Together these models present a thermo-chemical-mechanical model framework for the process which is unprecedented in literature. In this framework, the temperature and degree of cure fields already calculated in the thermo-chemical model are mapped to the quasi-static mechanical model in which the finite element method is employed. In the mechanical model, the composite part is assumed to advance along the pulling direction meanwhile tracking the corresponding temperature and degree of cure profiles. Modelling the pultrusion process containing both uni-directional (UD) roving and continuous filament mat (CFM) layers has not been considered in the literature up to now. A numerical simulation tool embracing the thermo-chemical and mechanical aspects of the pultrusion for industrial, pultruded products is hence developed in the present work.

Various case studies have been carried out using the devised numerical simulation tool. The residual stresses and shape distortions in pultrusion of an industrial rectangular hollow profile and L-shaped product are predicted. The deformation pattern as well as the corresponding magnitudes are found to agree with the real pultruded profiles. In addition, the internal stresses at the web flange junction of a pultruded I-beam are addressed which includes a more complex layer orientation. The manufacturing aspects of the pultrusion process such as the residual stresses and distortions are combined with the subsequent service loading scenario for a pultruded wind turbine blade profile (NACA0018). The effects of the residual stresses on the internal stress level after the loading analysis are investigated.

A pulling force model has specifically been analysed including gelation effects and the shrinkage induced effects. The compaction, viscous and frictional forces have been predicted for a pultruded composite rod. The viscous drag is found to be the main contribution in terms of the frictional force to the overall pulling force, while the contribution due to material compaction at the inlet is found to be negligible.

Process optimization studies have been carried out in order to improve the production rate and the quality. For this purpose, the mixed integer genetic algorithm (MIGA) is developed to optimize the process by finding the optimum heater configuration. Moreover, a multi-objective optimization problem (MOP) is implemented to the thermo-chemical analysis to minimize the energy consumption and maximize the productivity of the process simultaneously. Probabilistic analyses are also performed to investigate the effect of uncertainties in the process parameters on the product quality by using Monte Carlo simulations, response surface method and first order reliability method.
Resumé


Den grundlæggende numeriske model, der er udviklet, er en termo-kemisk model, som kan forudsige temperatur- og hårdningsgrads (cure degree) fordelinger i pultrusionprocessen. Denne (i 2D) er bl.a. blevet anvendt til at undersøge indflydelsen af kontaktmodstanden mellem emner og form på de resulterende temperatur- og hårdningsprofiler og her viste det sig, at en variabel kontaktmodstand bedre beskriver hold Fastighedene end en konstant værdi. Forskellige strategier til termo-kemisk modellering af pultrusion blev også undersøgt, herunder specielt tidsafhængige vs. stationære beregninger, og det blev her fundet, at sidstnævnte ofte giver en hurtigere numerisk løsning end at køre førstnævnte til stationaritet. Herefter blev pultrusion af et NACA0018 vindmølleprofil modelleret med den termo-kemiske model (3D), og effekten af forskellige varelegemekonfigurationer samt trækningstastigheden blev undersøgt.

Den termo-kemiske model er blevet udbygget til at også at medtage mekanisk forhold, således at procesinducerede spændinger og deformationer kan forudsiges. Der er herved opbygget en samlet termo-kemisk-mekanisk model, som må siges at være den absolute state-of-the-art i litteraturen for så vidt angår numerisk modellering af pultrusion. Modellen bruger de udregnede temperatur og hårdningsprofiler fra den termo-kemiske model i en kvasi-stationær mekanisk analyse, hvori det betragtede tværsnit bevæger sig
i langsgående retning i processen. Modellen har bl.a. været brugt på industrielle, pultruderede emner, der indeholder uni-directional (UD) roving og continuous filament mat (CFM) lag, noget der ikke er gjort før i litteraturen.


Trækkraften hvormed profilet trækkes igennem pultruderen er en vigtig parameter, og dens indflydelse er også blevet undersøgt i kombination med forhold som gelering, termisk sammentrækning og kemisk krymp med de udviklede modeller.

Der er endvidere blevet udført matematiske optimeringsstudier på basis af de udviklede modeller med henblik på at forbedre produktiviteten og kvaliteten af de pultruderede emner. Disse omfatter bl.a. anvendelse af en mixed integer genetic algorithm (MIGA) til at finde den optimale konfigurering af varmelegemerne. Derudover blev den termo-kemiske model brugt som simulator i et multi-objective optimization problem (MOOP), hvor målet var at minimere energiforbruget og samtidig øge produktiviteten (her udtrykt ved trækningshastigheden). Endelig blev der også udført probabilistiske analyser for at undersøge processens følsomhed overfor uundgåelige variationer i proces- og materialeparametre. De anvendte metoder var her Monte Carlo simulering, response surface metoder og first order reliability method (FORM).
Publications

The following publications are appended to the thesis:

PAPER1

PAPER2

PAPER3

PAPER4

PAPER5

PAPER6
Baran I, Hattel JH, Akkerman R. Investigation of process induced warpage for pultrusion of a rectangular hollow profile. 2014 (*submitted*).

PAPER7
Baran I, Akkerman R, Hattel JH. Modelling the pultrusion process of an industrial L-shaped composite profile. 2014 (*submitted*).

PAPER8

PAPER9


The non-appended publications are:


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Chapter 1

Introduction

This chapter serves as an introduction to the present thesis. The motivation and the objectives of the thesis are presented together with the main challenges faced in the pultrusion industry in terms of processing issues. Afterwards, a theoretical description of the pultrusion process is made and a detailed literature survey including the author’s own contributions is presented in the field of numerical modelling of the pultrusion process. Following this, an overview of the thesis structure is given by briefly explaining the content of each included chapters.

1.1 Motivation and Background

In DeepWind, a novel concept for a floating offshore vertical axis wind turbine (VAWT) based on the Darrieus design is being developed [1,2]. The Darrieus concept for VAWT is shown in Fig. 1.1. The main objective of DeepWind is to develop more cost-effective MW-scale wind turbines through innovative technologies for the sea environment rather than advancing existing concepts (i.e. either a horizontal or a vertical axis wind turbine) that are based on onshore technology. Hence the main challenges of the project are to increase the simplicity of the design and the manufacturing techniques as well as to reduce the total cost of an installed offshore wind farm. The concept is aiming at large-scale wind turbines for deep water. The up-scaling potential of the project is 20 MW wind turbines. It is expected that the structural design can be improved to have a higher strength-to-weight ratio for larger chord lengths, e.g. 10-20 m, with a deep water offshore floating system.

The blade cross section for the VAWT can be constant along the length of the blade. The pultrusion technology is foreseen to be one of the most efficient and suitable methods to manufacture such a composite blade with a constant profile having a large chord. A VAWT blade has already been manufactured by using the pultrusion process, as reported in [3,4]. Producing large blades in one piece using a single die will lead to cost reduction for large series production. The pultruded blades can achieve very high stiffness and resistance against aerodynamic loads as well as vibrations. In principle, a production facility with a relatively short die length, e.g. 1-3 m, can be put near to the location of the wind turbine installation which will alleviate transportation issues for these large constructions.

The use of pultruded profiles in several industries such as construction, transportation and marine has grown significantly. They are foreseen to have potential for the
replacement of some of the conventional materials used in the construction industry due to their main advantages over traditional materials such as high strength-to-weight ratio, high corrosion resistance as well as good electrical and thermal insulation properties. An example of this is the increased application of structural pultruded profiles for bridge constructions. Therefore, in order to increase the product quality as well as their reliability, a series of processing design challenges must be tackled. Firstly, premature cracking due to inter-laminar residual stresses have been experienced in the pultrusion industry for pultruded I-beams even before service loading. This shows that residual stresses might play a vital role for load carrying parts such as pultruded wind turbine blade reinforcements and structural profiles in the construction industry. Secondly, unwanted residual distortions may lead to not meeting the desired geometrical tolerances e.g. warpage of pultruded window frames and hollow profiles as well as spring-in of L-shaped profiles, etc.

The thermal and cure history together with highly non-linear resin phase transitions (viscous-rubbery-glassy) make the composite manufacturing process complex to control. During phase transitions, the resin undergoes large changes in its material properties, most significantly in its thermal expansion and elastic modulus. The main mechanisms generating the process induced stresses and shape distortions in pultrusion are summarized in the following:

i. Different coefficient of thermal expansion (CTE) for the thermosetting resin and reinforcements (micro level). The resin systems have usually much higher CTE as compared to the reinforcement fiber.

ii. Mismatch in the ply-level CTEs (macro level). The expansion behaviour differs in the longitudinal and transverse directions (in-plane or out-of-plane).

iii. Chemical shrinkage of the resin system during curing.

iv. The temperature and cure gradients inside the composite promote internal constraints during cure of especially thick parts.

v) The interaction between the tool (heating die) and the part.

Keeping the multi-physics and large amount of variables involved in the pultrusion process in mind, a satisfactory experimental analysis for the production requires considerable time which is obviously not a cost-efficient approach. Therefore, a numerical process simulation tool is essential to address the main challenges in pultrusion such as process induced residual stresses and shape distortions together with the prediction of the thermal and cure history. The expensive trial-and-error approaches for designing new products and process conditions can be avoided using the developed process models.

In order to have a better understanding of the mechanical response or the failure mechanisms of pultruded structures under service loading conditions, the process induced residual stresses have to be characterized since they may lead to cracking during curing of the thermosetting composites. In addition, the dimensional changes during processing have to be controlled in order to improve the product quality in terms of geometrical tolerances. In this Ph.D. thesis, state-of-the-art numerical simulation tools have been developed to model the multiphysics taking place in pultrusion. The main challenges in pultrusion such as process induced stresses and shape distortions have been addressed by employing the developed techniques and approaches. For this purpose, not only generic
simple geometries but also industrially pultruded parts have been considered. Moreover, the pulling force and its components are predicted using the devised numerical simulation tool. In addition, advanced thermo-chemical models have been proposed to calculate the temperature and the degree of cure distributions during the process. Using these process models, novel optimization studies have been carried out in order to improve the production rate and quality. Probabilistic analyses have also been performed to investigate the effect of uncertainties in the process parameters on product quality. In this thesis, the focus is particularly on the heating die section of the pultrusion process.

1.2 The Pultrusion Process: Theoretical Description

Pultrusion is in principle a simple process to manufacture constant cross sectional composite profiles. The process has a low labour content and a high raw material conversion efficiency since it is a continuous processing technique. There is little waste material being produced at the start up and the end of the process. Pultruded products have consistent quality and there is no need for any secondary finishing steps before the usage in service. A schematic view of a pultrusion process is shown in Fig. 1.2. The reinforcements in the form of continuous unidirectional (UD) roving or continuous filament mats (CFM) are held on creel racks and fed continuously through a guiding system. These reinforcements are impregnated with the desired matrix system in a resin bath. The wetted-out reinforcement pack is then collimated into a preformed shape before entering the heating die. A polymerization takes place inside the die with the help of the heat coming from the heaters. The cured profile is advanced via a pulling system to the cut-off saw where the finished product is cut to desired lengths.

The chemical exothermic reaction of the resin starts when the composite reaches the reaction initiation temperature during pultrusion of thermosetting resin composites. At this point the gelation of the resin is also observed. After some point in time the direction of the heat flux is inverted such that the heat flow is transmitted from the composite to
the die owing to the internal heat generation. A chemical shrinkage takes place during the curing of the composite resulting in separation of the part from the die at the die-part interface. At this instant, the volumetric shrinkage of the resin has a higher contribution to the deformation of the cured resin than the thermal expansion coefficient since the temperature difference is getting smaller while the process tends to reach the steady state. A schematic representation of the phase change of a thermosetting composite is seen in Fig. 1.3.

![Figure 1.2: Schematic view of a pultrusion process.](image)

![Figure 1.3: Representation of the phase change of a thermosetting composite (liquid-gel-solid) inside a heating die.](image)

In general, industrially pultruded parts contain UD roving and CFM layers impregnated by a thermosetting resin. The CFM consists of long, continuous lengths of fibre strands overlying each other in a totally random swirl pattern. The UD roving provides longitudinal tensile strength in the length of the profile. On the other hand, the CFM provides transverse strength across the width of the profile. A UD layer is transversely isotropic (TI), whereas the CFM layer can be considered as quasi isotropic (QI) since it consists of long swirled fibers randomly oriented in the plane of the mat. Therefore, the CFM layer has equal material properties in the in-plane directions \[10\] and the out-of-plane properties are different than the in-plane properties \[11\].

Among the matrix materials used in the pultrusion industry, polyester and epoxy resins are some of the most common. These two types of resin system behave differently in terms of curing dynamics. Both systems have inherent characteristics such as chemical shrinkage and reactivity which are crucial for the pultrusion process. A comparison of the properties of these resin system is shown in Table 4.1. It is seen that the polyester resin is more reactive than the epoxy and hence higher pulling speeds can be used for the pultrusion of polyester based composites. Moreover, the gelation occurs at lower conversion rates or degree of cure values for the polyester as compared to the epoxy and the volumetric shrinkage varies between 6-12% for the polyester resins. This value can be further decreased to 2% by mixing the unsaturated polyester with “low profile” or “shrink-reducing” additives \[12\].
Table 1.1: Comparison of the characteristics for the traditional polyester and epoxy thermosetting resin systems [12].

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Polyester</th>
<th>Traditional epoxy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Viscosity [µPas]</td>
<td>Low (500-2000)</td>
<td>High (&gt;3000)</td>
</tr>
<tr>
<td>Cure rate</td>
<td>Fast</td>
<td>Slow</td>
</tr>
<tr>
<td>Gel time</td>
<td>Short (Seconds)</td>
<td>Long (minutes)</td>
</tr>
<tr>
<td>Conversion at gelation</td>
<td>0.1-0.3</td>
<td>&gt;0.5</td>
</tr>
<tr>
<td>Volumetric shrinkage [%]</td>
<td>6-12</td>
<td>1-6</td>
</tr>
<tr>
<td>Typical processing rates [mm/min]</td>
<td>600-1500</td>
<td>70-100</td>
</tr>
</tbody>
</table>

In recent years, several scientific studies have been carried out for pultrusion and presented in literature in order to understand and control the process. Here, these studies are grouped under six main research fields and presented in the following. In each of the fields, a general survey of existing literature is presented first and this is followed by a presentation of the author’s own contributions in the field.

1.2.1 Thermo-chemical Modelling

Thermo-chemical numerical models have been developed and used for the simulation of the pultrusion process since the 1980s [13–15]. Transient and steady state simulations have been applied by using numerical techniques such as the finite difference method (FDM) and the finite element method (FEM) with control volume (CV) as well as the nodal control volume (NCV) method. A one dimensional (1D) heat transfer model of the pultrusion process for a thermosetting resin composite was developed in [13,14] employing the FEM. In [15,16] a mathematical model for heat transfer and cure inside the heating die was developed utilizing the FDM in which the time stepping was carried out implicitly using the Crank-Nicolson method. In these studies, the assumption of no axial conduction and negligible bulk flow simplified the 2D pultrusion model into a 1D transient heat transfer model. A comprehensive 2D axisymmetric pultrusion model of a graphite/epoxy composite rod in cylindrical coordinates was developed in which a control volume based finite difference method (CV/FD) was implemented in [17,18]. In [19], the pultrusion process of composite profiles was simulated in 2D in which the solution of the thermo-chemical equations was carried out using the alternating direction implicit (ADI) method. In 2D, the ADI method is an unconditionally stable finite difference time domain method of second order accuracy in both time and space [20]. 2D process simulations of pultruded profiles were carried out in [21,22] using the FEM.

Apart from 1D and 2D process models, 3D thermo-chemical simulations of the pultrusion process for different unidirectional (UD) composite cross sections were performed in [23–32]. The pultrusion of various irregular Cartesian geometries such as U, S, rectangular and hollow-square shaped products were analyzed in [23,25] in essence using Patankar’s finite volume method (FVM) [46]. 3D FE/NCV techniques were utilized, with the use of the general purpose finite element software LUSAS, for transient thermo-chemical modelling of UD pultruded composites in [26,28]. The effects of resin shrinkage and temperature-dependent material properties on temperature and cure degree distributions have been discussed in [29,31]. In [31], a thermal pultrusion simulation of multi materials i.e. a foam/glass fiber reinforced polymer (GFRP) sandwich panel has been...
In addition to the numerical thermo-chemical modelling of the pultrusion process, experimental studies of various UD composite profiles have been carried out in [33][40]. In these studies, experimental temperature data were obtained by using thermocouples inside the composite or the die and the resin kinetic parameters used in the numerical models were obtained from differential scanning calorimetry (DSC) scans of the resin. A 3D thermal model (including curing kinetics) of pultrusion of a flat plate was given in [33]. In addition to the composite plate, the die block, heating platens, insulators and cooling channels were also included in the numerical model. Predicted centerline temperatures were validated experimentally. Die and post die analysis was performed numerically in [34] for the temperature and cure degree profiles of a composite rod and the results were validated experimentally. The effect of convection cooling after the die exit was also considered. The post die curing and temperatures are vital for the analysis of the residual stresses developing during the cooling of the composite after the exit from the die. The numerical validation of the study in [34] was performed by using the ABAQUS and LUSAS general purpose finite element packages in [21] and [28], respectively. In [37], the transient temperature and cure degree distributions for the pultrusion of a glass/epoxy I-beam were obtained both numerically and experimentally. In [38][39], the heat transfer and curing during pultrusion of a UD glass/vinyl ester I-beam were simulated by using LUSAS. The predicted temperature profiles matched well with the experimental ones. In [40], the temperature and the cure degree distributions were predicted for a soy based thermosetting composite by using ABAQUS and the experimental results were found to be in good agreement with the numerical predictions.

In addition to these studies in the literature, the author has contributed substantially with efficient thermo-chemical process models for pultrusion [41][45]. In [41], the effects of the thermal contact resistance (TCR) at the die-part interface on the pultrusion process of a composite rod have been investigated by using the CV/FD method and it was found that the use of a variable TCR is more reliable than the use of a constant TCR for simulation of the process (PAPER1). In [42] (PAPER2), 3D numerical modeling strategies of a thermosetting pultrusion process are investigated considering both transient and steady state approaches. So far, the pultrusion process of a relatively thick composite having a curved cross sectional geometry such as the NACA0018 blade profile has not been described in the literature. A numerical simulation tool embracing the blade manufacturing process is hence being developed by the author in [44][45] (PAPER3).

According to the results obtained from the various numerical and experimental work mentioned above, similar temperature and cure degree behaviours have been found all showing the general trend that the temperature inside the composite is initially lagging behind the die temperature, however later during the curing the temperature exceeds the die temperature due to the internal heat generation of the resin.

1.2.2 Thermo-mechanical Modelling

Prior to the present work, there has been no contributions in literature in the field of mechanical modelling of the pultrusion process for the calculation of residual stresses
and shape distortions. Therefore, state-of-the-art process models based on a thermo-
chemical-mechanical analysis of the pultrusion process have recently been proposed by
the author [47–54]. The development of the process induced stresses and distortions were
specifically addressed in [47] in which a 3D transient thermo-chemical model was sequen-
tially coupled with a 2D quasi-static mechanical model using the FEM (PAPER4). The
proposed model in [47] was found to be computationally fast for the calculation of the
process induced deformations in the transverse directions. A more advanced 3D mechanical
analysis has been proposed by the author in [55] using 3D quadratic elements which
is a novel application for the numerical modelling of the pultrusion process (PAPER5).
In addition to the transverse directions, the residual stresses in the longitudinal (pulling)
direction were also addressed in [55]. Note that the material composition considered
in [47–55] (only the UD roving case) was relatively simple as compared to real industrially
pultruded parts which generally contain the combination of UD roving and CFM layers
as aforementioned. At present, no contribution has been given in the literature regarding
this; however, the author has proposed a novel numerical simulation tool addressing the
thermo-chemical and mechanical aspects of the pultrusion for industrial pultruded parts
containing both UD and CFM layers [56–58] (PAPER6, PAPER7). As a part of the work
done in [57] (PAPER7), the processed resin system was characterized in terms of curing,
rheological and mechanical behaviour under different thermal conditions in [59] (PAPER8).

In [48,49], an integrated modelling of the pultrusion process of a NACA0018 blade
profile was carried out by the author. The calculated residual stresses were transferred to
the subsequent bending simulation of the pultruded blade profile and the internal stress
distribution was evaluated in [49] (PAPER8) which is the second part of the work in [45]
(PAPER3). The effects of varying process conditions on the part quality are investigated
for two different heater configurations and with three different pulling speeds.

1.2.3 Process Optimization

Numerical optimization based on thermo-chemical analysis of the pultrusion process was
carried out in the literature [60–64]. The quality of the pultruded product and the pro-
ductivity of the process are improved by minimizing the power consumption of the process
or maximizing the pulling speed as well as the final degree of cure. Optimum process
parameters such as the pulling speed, the temperature of the heaters and the coolers are
obtained using different optimization approaches while satisfying certain specific process
constraints. In [60,62], the mathematical relationship between the cure degree at the die
exit and the design parameters such as the temperature of heaters, the pulling speed and
the power of the heaters were investigated for the pultrusion of a C-shaped product. The
optimization was performed by using the steepest descend algorithm. The same model,
i.e. the pultrusion of a C-shaped cross section [32], was optimized by means of a genetic
algorithm (GA) and the simplex method in [63]. The GA was utilized to find a suitable
starting point for the simplex method which highly depends on the starting point and a
poor value may result in finding a local minimum rather than the global minimum.
The variance of the cure degree evaluated at the exit cross section was minimized by an
iterative procedure based on the combination of the above techniques. Multi-objective
optimization was performed on a pultrusion process model utilizing finite element and
finite difference methods in [64] in which the multi-objective problem was reduced to a
single objective problem by using proper weightings between the objectives. In this opti-
mization problem, the combination of an artificial neural network (ANN) and a GA was
proposed to find the optimal solution. The objectives were reducing the power of the die and improving the productivity i.e increasing the pulling speed, while guaranteeing the quality expressed in terms of a target degree of cure of the composite product.

In addition to the optimization works mentioned above, the author has also carried out novel optimization studies \cite{65,66} based on thermo-chemical analysis of the pultrusion process. In \cite{65}, the productivity of the pultrusion process for a composite rod was improved by using a mixed integer genetic algorithm (MIGA) such that the total number of heaters was minimized while satisfying the constraints for the maximum composite temperature and the pulling speed as well as the mean of the cure degree at the die exit. The second heater (a total of five heaters were placed equidistant along the die) close to the die inlet was found to be the optimal heater location according to the optimization study (PAPER\textsuperscript{10}). In \cite{66}, a thermo-chemical simulation of the pultrusion process was integrated with a well-known evolutionary multi-objective (EMO) algorithm, i.e. the non-dominated sorting genetic algorithm (NSGA-II), to simultaneously maximize the pulling speed and minimize the total energy consumption.

\subsection*{1.2.4 Pulling Force Calculation}
Besides the degree of cure and the temperature distributions, the pulling force which results from the integration of several resistance forces along the heating die is also an important issue which needs to be addressed in the design of the pultrusion manufacturing line. In literature, the pulling force is modelled and experimentally validated by taking into account phenomena such as \textit{i}) the viscous drag before the resin transforms into the gel state, \textit{ii}) the collimation force related to the impregnation, \textit{iii}) the preforming processes and \textit{iv}) the bulk compaction force as well as \textit{v}) the friction force between the die and the composite \cite{67,68,69,70}. In \cite{67}, a rheological model was developed using a temperature- and cure-dependent viscosity. This allows to evaluate the viscous pulling force arising from the shear stresses acting between the internal die walls and the processing material. Evaluation of the pulling force was considered in \cite{68,69,70,71}. The resistance force was calculated using an analytical model. In addition to that, the thermal expansion-polymerization shrinkage model and a friction coefficient model were also developed in addition to obtaining the temperature and resin conversion profiles. In \cite{71}, the pulling force model was validated by conducting experiments. An improved pulling force model was specifically analysed in detail \cite{72} including gelation effects and the shrinkage induced effects. The compaction, viscous and frictional forces were predicted in \cite{74} (PAPER\textsuperscript{11}) for a pultruded composite rod. The viscous drag was found to be the main contribution in terms of the frictional force to the overall pulling force, while the contribution due to material compaction at the inlet was found to be negligible.

\subsection*{1.2.5 Probabilistic Modelling}
Composite materials have large statistical variations in their mechanical properties \cite{75} which may be due to the uncertainties in volume fraction of the resin content, degree of cure and process induced residual stresses during processing as well as the probability of defects and void formations inside the composites, etc. Hence, there is a need for a probabilistic or reliability analysis of composite failure and the product quality. Such analyses play a vital role in the strength evaluation of composite structures. In contrast to the deterministic analysis of the composite materials, the probabilistic analysis gives a better
understanding of the effect of the variations inherently being present in the geometry, material properties or manufacturing process. This makes it easier and more practical to predict how sensitive the scatter of the output parameters (e.g. the performance of the composite, failure criterion, degree of cure etc.) with respect to the scatter in the input design parameters is. In other words, this provides a way for an evaluation of the robustness of the process.

Apart from the deterministic process models for pultrusion reported in literature, the author has carried out probabilistic numerical simulations based on thermo-chemical and thermo-mechanical analysis of pultrusion\cite{76, 79}. The reliability estimation of the pultrusion process of a flat plate was analyzed in\cite{76} using Monte Carlo simulations (MCS) and the first order reliability method (FORM) (\textit{PAPER}12). The degree of cure at the die exit and the maximum composite temperature were selected as the random output variables. The statistical variation in the activation energy and the heater temperature multiplier were found to have the highest effect on the variation in the degree of cure and the maximum composite temperature, respectively. A new application for the probabilistic analysis of the pultrusion process is introduced using the Response Surface Method (RSM) in\cite{77} (\textit{PAPER}13). The results obtained from the RSM are validated by employing the Monte Carlo simulation (MCS) with Latin Hypercube Sampling (LHS) technique. According to the results obtained from both methods, the variations in the activation energy as well as the density of the resin are found to have a relatively stronger in influence on the centerline degree of cure at the exit. Moreover, different execution strategies are examined for the MCS to investigate their effects on the accuracy of the random output parameter.

1.2.6 Modelling the Resin Flow

It is very important to obtain complete wet-out of the reinforcement in the pultrusion process to avoid any formation of voids inside the product. Several researchers have investigated the impregnation of the reinforcements. A 2D finite element model based on Darcy’s law was developed for porous media in\cite{80, 81}. The fluid resin pressure rise was predicted inside the pultrusion die. A 3D axisymmetric model was used to calculate the pressure rise in a pultrusion die for a graphite/epoxy composite in\cite{82, 84}. The FVM was utilized based on Darcy’s law for the flow simulations. A variable viscosity and an anisotropic permeability model were employed for calculating the permeability values in the axial and radial directions. The effects of pulling speed, fiber volume fraction, resin viscosity and compression ratio of the injection chamber on resin fiber wet out were investigated in\cite{85, 86}. 
1.3 Structure of the Thesis

This thesis consists of 6 chapters followed by 13 appended papers. The content of the chapters are explained in the following:

Chapter-1: Introduction

- This chapter focuses the motivation and background of the thesis. A detailed theoretical description of modelling the pultrusion process is presented based on the scientific studies carried out in literature.

Chapter-2: Numerical Implementation

- The thermo-chemical and mechanical process model formulations are described in detail. An overview of the governing equations used for the heat transfer, resin cure kinetics (PAPER\textsuperscript{1}-PAPER\textsuperscript{3}) and mechanical constitutive models (PAPER\textsuperscript{4}, PAPER\textsuperscript{5}) are given.

Chapter-3: Constitutive Material Behaviour

- In this chapter, the summary of the results in PAPER\textsuperscript{8} which has recently been published are presented. The chemo-rheology of an industrial “orthophthalic” polyester system specifically prepared for a pultrusion process is characterized.

Chapter-4: Summary of Results on Thermo-chemical Modelling

- The summary of the main results and discussions based on the thermo-chemical analysis of pultrusion (PAPER\textsuperscript{1}-PAPER\textsuperscript{3}) are given in this chapter. Moreover, the optimization studies (PAPER\textsuperscript{10}) and the probabilistic modelling work (PAPER\textsuperscript{12}, PAPER\textsuperscript{13}) based on the thermo-chemical analysis of the pultrusion process are presented.

Chapter-5: Summary of Results on Thermo-mechanical Modelling

- This chapter consists of the main outcomes of the studies carried out in (PAPER\textsuperscript{4}-PAPER\textsuperscript{7}, PAPER\textsuperscript{9}) in terms of the thermo-chemical-mechanical analysis of the pultrusion process. The focus is here on the prediction of the process induced stresses and shape distortions. In addition, the evaluation of the total pulling force for a pultruded rod (PAPER\textsuperscript{11}) is also presented.

Chapter-6: Conclusion and Future Work

- Some concluding remarks on the obtained results as well as the applicability of the developed process models for the future challenges in pultrusion industry are given in this chapter.
Chapter 2

Numerical Implementation

In this chapter, the implementations of the numerical methods are introduced in detail. First, the governing equations for the thermo-chemical modelling of pultrusion are presented. The numerical solution strategies are investigated considering both transient and steady state approaches. Afterwards, the details of the process induced stress calculation used in the thermo-mechanical model are presented. In addition, the calculation of the effective mechanical properties of the processing composite together with the thermal and chemical strains are presented in detail. Finally, the details of the pulling force calculations are presented.

2.1 Thermo-chemical Analysis

The three dimensional (3D) transient energy equations for the composite and the die block are given in Eq. 2.1 and Eq. 2.2 respectively in a Cartesian coordinate system. Here, $x_1$ is the pulling or longitudinal direction; $x_2$ and $x_3$ are the transverse directions. In the energy equation, the convective ($u\partial T/\partial x_1$) and the source ($q$) terms are present for the composite part only due to the advection of the material and the internal heat generation of the resin system, respectively [41,42] (PAPER1, PAPER2).

\[
(\rho C_p)c \left( \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x_1} \right) = k_{x_1,c} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2,c} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3,c} \frac{\partial^2 T}{\partial x_3^2} + q \tag{2.1}
\]

\[
(\rho C_p)d \frac{\partial T}{\partial t} = k_{x_1,d} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2,d} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3,d} \frac{\partial^2 T}{\partial x_3^2} \tag{2.2}
\]

where $T$ is the temperature, $u$ is the pulling speed, $\rho$ is the density, $C_p$ is the specific heat and $k_{x_1}$, $k_{x_2}$ and $k_{x_3}$ are the thermal conductivities in the $x_1$-, $x_2$- and $x_3$-directions, respectively. The subscripts $c$ and $d$ correspond to the composite layer and the die, respectively. It should be noted that the composite layer can be either unidirectional (UD) roving or continuous filament mat (CFM). Therefore, Eq. 2.1 has to be considered separately for the UD roving and CFM layers [56,57] (PAPER6, PAPER7). Lumped material properties are used and assumed to be constant. The source term $q$ in Eq. 2.1 is related to the internal heat generation due to the exothermic reaction of the resin and expressed as [28]:

\[
q = (1 - V_f)\rho r H_{tr} R_r(\alpha, T) \tag{2.3}
\]
where \( H_{tr} \) is the total heat of reaction for the resin during the exothermic reaction, \( \rho_r \) is the resin density, \( V_f \) is the fiber volume fraction, \( \alpha \) is the degree of cure and \( R_r(\alpha, T) \) is the reaction of cure which can also be defined as the rate of the cure degree, i.e. \( d\alpha/dt \).

In composite manufacturing processes, the rate of cure degree is usually assumed to be proportional to the rate of heat flow \( (dH/dt) \) \(^87\) and expressed as:

\[
\frac{d\alpha}{dt} = \frac{1}{H_{tr}} \frac{dH}{dt}
\]  
(2.4)

In literature, several kinetic models have been proposed and analysed to describe the resin curing reactivity \(^88\) \(^94\). In general, Arrhenius-type equations are employed for most of the cure kinetics models. An example of a well known semi-empirical autocatalytic model \(^95\) \(^98\) is expressed as:

\[
R_r(\alpha, T) = \frac{d\alpha}{dt} = A_0 \exp(-\frac{E_a}{RT}) \alpha^m (1 - \alpha)^n
\]  
(2.5)

where \( A_0 \) is the pre-exponential constant, \( E_a \) is the activation energy, \( R \) is the universal gas constant and \( m \) and \( n \) are the orders of reaction (kinetic exponents). On the other hand, \( n \)\textsuperscript{th}-order cure models are particularly used for epoxy systems \(^33\) \(^34\) since they experience no autocatalyzation. The corresponding expression is given as:

\[
R_r(\alpha, T) = \frac{d\alpha}{dt} = A_0 \exp(-\frac{E_a}{RT})(1 - \alpha)^n
\]  
(2.6)

The material derivative of the degree of cure field can be translated into a partial derivative form in a Eulerian frame of reference in the pulling direction. Using the “chain rule”, the rate of cure rate is expressed as \(^47\) \( \text{PAPER}^4 \):

\[
R_r(\alpha, T) = \frac{d\alpha}{dt} = \frac{\partial\alpha}{\partial t} + \frac{\partial\alpha}{\partial x_1} \frac{dx_1}{dt} = \frac{\partial\alpha}{\partial t} + u \frac{\partial\alpha}{\partial x_1}
\]  
(2.7)

and from Eq. 2.7, the relation of the resin kinetics equation can be expressed as:

\[
\frac{\partial\alpha}{\partial t} = R_r(\alpha, T) - u \frac{\partial\alpha}{\partial x_1}
\]  
(2.8)

which is used in the thermo-chemical model. The equations above are solved using two different techniques in the present thesis: the nodal control volume based finite element (NCV-FE) method and the control volume based finite difference (CV-FD) method. The details of these methods are explained in the following.

**CV-FD Technique:**

The discretization of the energy and cure kinetics equations in the space domain is obtained by employing the CV-FD technique in the mathematical computing environment MATLAB \(^99\). The total thermal resistances (K/W) being the sum of the single resistances coupled in series between the two adjacent control volumes are used \(^100\). The CV-FD approach has already been used in numerical modeling of the pultrusion process \(^41\) \(^42\) \(^76\) (\text{PAPER}^2). The representation of the thermal resistances in the \( x \)-, \( y \)- and \( z \)-directions for an internal CV, i.e. the node \((i,j,k)\), is seen in Fig. 2.1. Here, the thermal resistances in the \( x-y \) plane and the \( y-z \) plane are seen in Fig. 2.1 (left) and Fig. 2.1 (right), respectively. The first order “upwind” scheme is used for the convective
term \((u\partial T/\partial x_1)\) in the energy equation (Eq. 2.1) and for the space discretization of the cure degree \((u\partial \alpha/\partial x_1)\) in the resin kinetics equation (Eq. 2.8) in order to obtain a stable solution for high “Peclet” \((P_e)\) numbers, i.e. \(P_e > 2\). It should be noted that the dimensionless \(P_e\) number indicates the strength between the convective and the conductive terms in the flow simulation. In theory, \(P_e\) should be less than 2 in order to get stable results when using a central finite difference discretization [46]. However, the “upwind” scheme avoids the possible oscillations during the simulation for large \(P_e\). The details of the CV-FD implementation can be found in [42] (PAPER2).

Figure 2.1: Schematic view of the thermal resistances for a 3D CV on \(x-y\) plane (left) and \(y-z\) plane (right) [42] (PAPER2).

NCV-FE Method:

The NCV-FE approach as described in [28] is implemented in a commercial FE software, e.g. ANSYS [101] and ABAQUS [102], to model the pultrusion process. The representation of the NCV-FE grid is illustrated in Fig. 2.2. CVs are defined at the nodes of each finite element and the temperature is calculated using the finite element method. The degree of cure, on the other hand, is calculated in the NCVs using the user defined subroutines in ABAQUS or ANSYS. The temperature and cure degree profiles at steady state are needed for the evaluation of the pultrusion since it is a continuous process; the composite part entering the heating die keeps tracking these steady state profiles during processing. In order to reach the steady state, the transient and steady state solution techniques are investigated in this thesis. The details are presented in the following.

2.1.1 Transient Approach

The transient solution is suitable for the simulation of the pultrusion process in which the material properties are a function of time, temperature, etc. Additionally, it is also convenient to simulate the transient pultruder operation in which the heaters operate with a heating power and a feedback thermocouple controls the heater temperature within a prescribed tolerance [28][33].
Chapter 2. Numerical Implementation

The non-linear internal heat generation (Eq. 2.3) together with the cure kinetics equation (Eq. 2.5 or Eq. 2.6) are coupled with the energy equation (Eq. 2.1) in an explicit manner in order to obtain a straightforward and fast numerical procedure. The degree of cure is subsequently updated explicitly for each CV using Eq. 2.8 in its discretized form \[42\] (PAPER\(^2\)). This time-stepping procedure is illustrated in the flowchart in Fig. 2.3. The criteria for reaching the steady state is defined as the maximum temperature and cure degree increments between the new time step \((n + 1)\) and the old time step \((n)\), i.e \(\Delta T = \max(T^{n+1} - T^n)\) and \(\Delta \alpha = \max(\alpha^{n+1} - \alpha^n)\), respectively. The values for \(\Delta T\) and \(\Delta \alpha\) are specified as 0.001°C and 0.0001, respectively (see Fig. 2.3).

Figure 2.2: Schematic view of the FE-NCV grids in the pulling direction \[44,45\].

Figure 2.3: Flowchart of the time-stepping procedure in the transient approach to reach the steady state solution for the temperature and the degree of cure \[42\] (PAPER\(^2\)).
2.1.2 Steady State Approach

The energy equations at steady state are obtained by discarding the time dependent term \( \partial T / \partial t \) from the energy equations (Eq. 2.1 and Eq. 2.2). Similarly, the term \( \partial \alpha / \partial t \) is discarded from the cure kinetics equation (Eq. 2.8) in the steady state approach. The steady state solution is convenient for the numerical model when having constant processing conditions throughout the process. A similar iteration procedure as given in Fig. 2.3 is used to obtain a converged steady state solution. However, it should be noted that there is no time step in this case; instead, an iteration loop is utilized to obtain the converged results, i.e. \( T \) and \( \alpha \) as well as \( q \) (Fig. 2.4) are updated until the steady state conditions are satisfied (\( \Delta T < 0.001^\circ \text{C} \) and \( \Delta \alpha < 0.0001 \)).

![Flowchart of the iteration procedure to solve the equation system for the temperature and the degree of cure at steady state.](image)

2.2 Mechanical Analysis

A novel approach is developed by the author to predict the stresses and displacements evolving during the pultrusion process. A 3D thermo-chemical model is coupled with a 2D quasi-static plane strain mechanical model using the FEM. In this 2D mechanical model, the cross section of the composite is moved through the pulling direction during the process meanwhile tracking the corresponding temperature and degree of cure profiles. A generic representation of this sequential coupling procedure is shown in Fig. 2.5 [47]. Since the length of the pultruded profile is generally much larger than the cross sectional dimensions, a plain strain assumption is convenient for the mechanical analysis of the pultrusion [47]. A more advanced 3D mechanical model is also developed by the author in which the process induced stresses and distortions are calculated in a 3D domain [55] (PAPER5). This provides a better understanding of the stresses and distortions in the longitudinal direction. In the 3D mechanical model, instead of the cross section of the part which is used in the 2D mechanical model (see Fig. 2.5), the entire 3D part is assumed to move along the pulling direction of the process while tracking...
the corresponding temperature and degree of cure profiles calculated in the 3D thermo-chemical simulation. In other words, a 3D Eulerian thermo-chemical model is coupled with a 3D quasi-static Lagrangian mechanical model (see Fig. 2.6). The details of the numerical implementations in the mechanical analysis are presented in the following.

Figure 2.5: Representation of the coupling of the 3D Eulerian thermo-chemical model with the 2D Langrangian plain-strain/generalized-plane-strain mechanical model including the rigid body surfaces and the mechanical BCs [47] (PAPER^4).

Figure 2.6: Representation of the coupling of the 3D thermo-chemical model with the 3D mechanical model [55] (PAPER^5).
2.2.1 Elastic Modulus of the Resin

The stiffness of the resin significantly depends on the degree of cure ($\alpha$). The cure dependent instantaneous isotropic resin modulus ($E_r$) was proposed in [8] and expressed as:

$$E_r = (1 - \alpha)E^0_r + \alpha E^\infty_r$$  \hspace{1cm} (2.9)

where $E^0_r$ and $E^\infty_r$ are the initial (i.e. uncured) and fully cured resin moduli, respectively. It should be noted that $E^0_r$ is generally assumed to be $E^\infty_r/1000$ as a first approximation [6–8]. Eq. 2.9 has been modified by incorporating the temperature dependency as suggested in the cure hardening instantaneous linear elastic (CHILE) approach [95,96] which exhibits the cure hardening and also thermal softening as shown in Eq. 2.10.

$$E_r = \begin{cases} 
E^0_r & T^* \leq T_{C1} \\
E^0_r + \frac{T^* - T_{C1}}{T_{C2} - T_{C1}}(E^\infty_r - E^0_r) & T_{C1} < T^* < T_{C2} \\
E^\infty_r & T_{C2} \leq T^* 
\end{cases}$$  \hspace{1cm} (2.10)

where $T_{C1}$ and $T_{C2}$ are the critical temperatures at the onset and completion of the glass transition, respectively and $T^*$ represents the difference between the instantaneous glass transition temperature $T_g$ and the temperature $T$ of the resin, i.e. $T^* = T_g - T$ [95,96]. The evolution of the $T_g$ with the degree of cure is modelled by the Di Benedetto equation [97,98] and expressed as:

$$\frac{T_g - T_{g0}}{T_{g\infty} - T_{g0}} = \frac{\lambda \alpha}{1 - (1 - \lambda)\alpha}$$  \hspace{1cm} (2.11)

where $T_{g0}$ and $T_{g\infty}$ are the glass transition temperatures of uncured and fully cured resin, respectively and $\lambda$ is a constant used as fitting parameter [97]. Moreover, the dependence of glass transition on the degree of cure was estimated using the experimental data and the corresponding relation is given as [7,95]:

$$T_g = T_g^0 + a_T g \alpha$$  \hspace{1cm} (2.12)

where $T_g^0$ is the glass transition temperature at $\alpha = 0$ and $a_T g$ is a constant.

In this thesis, a temperature- and cure-dependent resin modulus is developed using a modified CHILE model [95,97]. This model captures the modulus variation due to the phase changes (viscous-rubbery-glassy) during processing. The corresponding expression for the modulus is given as:

$$E_r = \begin{cases} 
E_0 & T^* \leq T_{C1} \\
A_e \exp(K_e T^*) & T_{C1} < T^* < T_{C2} \\
E_1 + \frac{T^* - T_{C2}}{T_{C3} - T_{C2}}(E_{\infty} - E_1) & T_{C2} < T^* < T_{C3} \\
E_{\infty} & T_{C3} \leq T^* 
\end{cases}$$  \hspace{1cm} (2.13)

where $A_e$ and $K_e$ are the constants for the exponential term. The other model constants indicate the phase transition zones [59] (PAPER8) and are schematically shown in Fig. 2.7.
2.2.2 Effective Mechanical Properties

2.2.2.1 UD Laminate

The effective mechanical properties of the transversely isotropic UD layer are calculated using the self consistent field micromechanics (SCFM) approach which is a well known and documented technique in the literature [8].

The mechanical properties of the fiber reinforcements are assumed to be transversely isotropic which is described by 5 independent elastic constants, instead of 9 for fully orthotropic materials. These 5 elastic constants are the Young’s modulus and the Poisson’s ratio in the transverse direction ($E_{2f}$ and $\nu_{23f}$) and in the longitudinal direction ($E_{1f}$ and $\nu_{12f}$) and the shear modulus in the longitudinal direction ($G_{12f}$). The resin has an isotropic Young’s modulus ($E_r$), Poisson’s ratio ($\nu_r$) and a shear modulus ($G_r$). Based on the fiber volume fraction ($V_f$), the effective mechanical properties of the composite are calculated in the following by using the SCFM approach proposed in [8].

**Longitudinal Young’s Modulus:**

$$E_1 = E_{1f}V_f + E_r(1 - V_f) + \left[\frac{4(\nu_r - \nu_{12f})k_fk_r(1 - V_f)V_f}{(k_f + G_r)k_r + (k_f - k_r)G_rV_f}\right]$$ (2.14)

where $k_f$ and $k_r$ are the isotropic plane strain bulk modulus for fiber and resin, respectively and expressed as [8]:

$$k_f = \frac{E_{1f}}{2(1 - \nu_{12f} - 2\nu_{12f}^2)}$$ (2.15)

$$k_r = \frac{E_r}{2(1 - \nu_r - 2\nu_r^2)}$$ (2.16)

**Shear Modulus:**

$$G_{12} = G_{13} = G_r\left[\frac{(G_{12f} + G_r) + (G_{12f} - G_r)V_f}{(G_{12f} + G_r) - (G_{12f} - G_r)V_f}\right]$$ (2.17)
2.19$ G_{23} = \frac{G_r [k_r (G_r + G_{23f}) + 2G_{23f} G_r + k_r (G_{23f} - G_r)V_f]}{k_r (G_r + G_{23f}) + 2G_{23f} G_r - (k_r + 2G_r)(G_{23f} - G_r)V_f}$  

(2.18)

where

$$G_{23f} = \frac{E_{3f}}{2(1 + \nu_{23f})}$$

(2.19)

Transverse Young’s Modulus:

$$E_2 = E_3 = \frac{1}{(4k_T)^{-1} + (4G_{23})^{-1} + (\nu_{12}^2/E_1)}$$

(2.20)

where $k_T$ is the effective plain strain bulk modulus and calculated as $[8]$:

$$k_T = \frac{(k_f + G_r)k_r + (k_f - k_r)G_r V_f}{(k_f + G_r) - (k_f - k_r)V_f}$$

(2.21)

Poisson’s Ratios:

$$\nu_{12} = \nu_{13} = \nu_{12f} V_f + \nu_r (1 - V_f) + \left[ \frac{(\nu_r - \nu_{12f})(k_r - k_f) G_r (1 - V_f)V_f}{(k_f + G_r)k_r + (k_f - k_r)G_r V_f} \right]$$

(2.22)

$$\nu_{23} = \frac{2E_1 k_T - E_1 E_2 - 4\nu_{12}^2 k_T E_2}{2E_1 k_T}$$

(2.23)

2.2.2.2 Quasi-isotropic Laminate

The mechanical properties of the quasi isotropic (QI) laminate (CFM layer) is calculated considering the effective mechanical properties of the UD layer obtained by the SCFM for the same $V_f$ as the QI layer $[11]$. The details of the calculations for the QI CFM layer are given in following.

For any orientation of the in-plane coordinate axes $x$ and $y$, the in-plane stiffness constants $(E_x, E_y, \nu_{xy}$ and $G_{xy})$ for a QI laminate can be expressed as $[11][103]$:

$$E_x = E_y = 2(1 + \nu_{xy}) G_{xy}$$

(2.24)

$$\nu_{xy} = \frac{-1}{2} \frac{G_{12}}{E_1 - \nu_{12}^2} + \frac{1}{8} \frac{E_1 (E_1 + E_2 + 6\nu_{12} E_2)}{E_1 - \nu_{12}^2} E_2$$

(2.25)

$$G_{xy} = \frac{1}{2} \frac{G_{12}}{E_1 - \nu_{12}^2 E_2} + \frac{1}{8} \frac{E_1 (E_1 + E_2 - 2\nu_{12} E_2)}{E_1 - \nu_{12}^2 E_2}$$

(2.26)

where $E$ is the elastic modulus, $G$ is the shear modulus, $\nu$ is the Poisson’s ratio, subscripts 1 and 2 are the longitudinal and transverse directions, respectively, for a UD laminate having the same $V_f$ as the QI laminate (the CFM layer).
The corresponding out-of-plane elastic properties in the $z$ direction for the QI laminate were obtained in [11] using the averaging method [104] and expressed as:

$$E_z = \frac{E_1 + (1 + 2\nu_{12})E_2}{(1 - \nu_{23}^2)E_2 + (1 + 2\nu_{12} + 2\nu_{12}\nu_{23}) - \nu_{12}^2E_1}$$

$$G_{xz} = 2 \left( \frac{G_{12}G_{23}}{G_{12} + G_{23}} \right)$$

$$\nu_{xz} = \nu_{yz} = \frac{E_x (\nu_{12} + \nu_{23} + \nu_{12}\nu_{23}) + \nu_{12}^2E_2}{1 + (1 + 2\nu_{12})E_2E_1}$$

for any in-plane coordinate $x$ for the QI laminate. It should be noted that for this set of constants the transverse Poisson’s ratio $\nu_{23}$ for the UD laminate is also required which is generally higher than the in-plane Poisson’s ratio $\nu_{12}$ [11].

### 2.2.3 Thermal Strain

The effective coefficient of thermal expansion (CTE) ($\alpha_i$) of the UD laminate is obtained as [8]:

$$\alpha_1 = \frac{\alpha_{1f}E_1V_f + \alpha_2E_r(1 - V_f)}{E_1V_f + E_r(1 - V_f)}$$

$$\alpha_2 = \alpha_3 = (\alpha_{2f} + \nu_{12f}\alpha_{1f})V_f + (\alpha_r + \nu_\alpha_\alpha_r)(1 - V_f) - (\nu_{12f}V_f + \nu_r(1 - V_f))\alpha_1$$

Similarly the in- and out-of-plane CTEs are also derived in a similar manner [11] and given as:

$$\alpha_x = \alpha_y = \frac{(E_1 + \nu_{12}E_2)\alpha_1 + (1 + \nu_{12})E_2\alpha_2}{E_1 + (1 + 2\nu_{12})E_2}$$

$$\alpha_z = \frac{\nu_{12}E_2 - \nu_{23}E_1}\alpha_1 + ((1 + \nu_{23})E_1 + (1 + \nu_{12})E_2)\alpha_2}{E_1 + (1 + 2\nu_{12})E_2}$$

The incremental effective thermal strains of the composite ($\dot{\varepsilon}_i^{th}$) are then calculated considering a temperature increment ($\Delta T$) and the effective CTEs ($\alpha_i$):

$$\dot{\varepsilon}_i^{th} = \alpha_i \cdot \Delta T$$

### 2.2.4 Chemical Strain

The chemical shrinkage of the resin is expressed via the total volumetric shrinkage ($V_{sh}$) as explained in the following. Assuming a uniform contraction for a unit cell in the resin, the isotropic incremental resin shrinkage strain ($\dot{\varepsilon}_r$) is calculated as [8]:

$$\dot{\varepsilon}_r = \frac{3}{\sqrt{1 + \Delta V_r}} - 1$$
where $\Delta V_r$ is the incremental specific volume shrinkage of the resin expressed as a function of change in the degree of cure ($\Delta \alpha$) and $V_{sh}$ \cite{8}:

$$\Delta V_r = \Delta \alpha \cdot V_{sh} \quad (2.36)$$

Using the SCFM approach, the effective incremental chemical shrinkage strain of a UD composite laminate is written as \cite{8}:

$$\varepsilon_{ch}^1 = \frac{\varepsilon_r E_r (1 - V_f)}{E_{1f} V_f + E_r (1 - V_f)}$$  \quad (2.37)

$$\varepsilon_{ch}^2 = \varepsilon_{ch}^3 = (\varepsilon_r + \nu_r \varepsilon_r)(1 - V_f) - (\nu_1 V_f + \nu_r (1 - V_f))\varepsilon_{ch}^1$$  \quad (2.38)

In the present study, the in- and out-of-plane incremental chemical strains ($\varepsilon_{ch}^i$) for the QI laminate are obtained using the similar expressions given in Eq. 2.32 and Eq. 2.33 since the chemical contraction or shrinkage has the similar physical considerations with thermal expansion/contraction. As a consequence $\varepsilon_{ch}^i$ for the QI laminate is expressed as:

$$\varepsilon_{ch}^x = \varepsilon_{ch}^y = \frac{(E_1 + \nu_1 E_2)\varepsilon_{ch}^1 + (1 + \nu_1) E_2 \varepsilon_{ch}^2}{E_1 + (1 + \nu_1) E_2}$$  \quad (2.39)

$$\varepsilon_{ch}^z = \frac{(\nu_1 E_2 - \nu_2 E_1)\varepsilon_{ch}^1 + ((1 + \nu_2) E_1 + (1 + \nu_1) E_2) \varepsilon_{ch}^2}{E_1 + (1 + \nu_1) E_2}$$  \quad (2.40)

### 2.2.5 Stress Calculation

Process induced stresses and displacements are incrementally solved using the FEM. The total incremental strain ($\varepsilon_{tot}$), which is composed of the incremental mechanical strain ($\varepsilon_{mech}$), thermal strain ($\varepsilon_{th}$) and chemical strain ($\varepsilon_{ch}$), is given in Eq. 2.41. Here, the incremental process induced strain ($\varepsilon_{pr}$) is defined as the summation of $\varepsilon_{th}$ and $\varepsilon_{ch}$ as also done in e.g. \cite{8,95,96}. The incremental stress tensor ($\sigma_{ij}$) is calculated using the material Jacobian matrix ($J$) based on the incremental mechanical strain tensor ($\varepsilon_{mech}$) (Eq. 2.42). The corresponding expression is given in Eq. 2.43 for a 3D analysis and Eq. 2.46 for a 2D analysis in which an orthotropic material is considered.

$$\varepsilon_{ij} = \varepsilon_{mech} + \varepsilon_{th} + \varepsilon_{ch}$$  \quad (2.41)

$$\varepsilon_{pr} = \varepsilon_{th} + \varepsilon_{ch} \quad \varepsilon_{mech} = \varepsilon_{tot} - \varepsilon_{pr}$$

$$\sigma_{ij} = J \varepsilon_{mech}$$  \quad (2.42)

$$\begin{pmatrix}
\dot{\sigma}_{11} \\
\dot{\sigma}_{22} \\
\dot{\sigma}_{33} \\
\dot{\tau}_{12} \\
\dot{\tau}_{13} \\
\dot{\tau}_{23}
\end{pmatrix} = \begin{pmatrix}
J_{11} & J_{12} & J_{13} & 0 & 0 & 0 \\
J_{21} & J_{22} & J_{23} & 0 & 0 & 0 \\
J_{31} & J_{32} & J_{33} & 0 & 0 & 0 \\
0 & 0 & 0 & J_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & J_{55} & 0 \\
0 & 0 & 0 & 0 & 0 & J_{66}
\end{pmatrix} \begin{pmatrix}
\dot{\varepsilon}_{11} \\
\dot{\varepsilon}_{22} \\
\dot{\varepsilon}_{33} \\
\dot{\gamma}_{12} \\
\dot{\gamma}_{13} \\
\dot{\gamma}_{23}
\end{pmatrix}^{mech}$$  \quad (2.43)
where

\[
\mathbf{J} = \begin{bmatrix}
1 - \nu_{23}\nu_{32} & \nu_{21} + \nu_{31}\nu_{23} & \nu_{31} + \nu_{21}\nu_{32} & 0 & 0 & 0 \\
\frac{E_2E_3\Delta}{\nu_{12} + \nu_{13}\nu_{32}} & \frac{E_2E_3\Delta}{1 - \nu_{31}\nu_{13}} & \frac{E_2E_3\Delta}{\nu_{32} + \nu_{31}\nu_{12}} & 0 & 0 & 0 \\
\frac{E_1E_3\Delta}{\nu_{13} + \nu_{12}\nu_{23}} & \frac{E_1E_3\Delta}{\nu_{23} + \nu_{13}\nu_{21}} & \frac{E_1E_3\Delta}{1 - \nu_{12}\nu_{21}} & 0 & 0 & 0 \\
\frac{E_1E_2\Delta}{\nu_{12} + \nu_{13}\nu_{32}} & \frac{E_1E_2\Delta}{\nu_{23} + \nu_{13}\nu_{21}} & \frac{E_1E_2\Delta}{1 - \nu_{12}\nu_{21}} & 0 & 0 & 0 \\
0 & 0 & 0 & 2G_{12} & 0 & 0 \\
0 & 0 & 0 & 0 & 2G_{13} & 0 \\
0 & 0 & 0 & 0 & 0 & 2G_{23}
\end{bmatrix}
\]  

(2.44)

and

\[
\Delta = \frac{1 - \nu_{12}\nu_{21} - \nu_{13}\nu_{31} - \nu_{23}\nu_{32} - 2\nu_{12}\nu_{23}\nu_{31}}{E_1E_2E_3}
\]  

(2.45)

\[
\begin{pmatrix}
\dot{\sigma}_{11} \\
\dot{\sigma}_{22} \\
\dot{\sigma}_{33} \\
\dot{\gamma}_{12}
\end{pmatrix} = \begin{pmatrix}
1 - \nu_{23}\nu_{32} & \nu_{21} + \nu_{31}\nu_{23} & \nu_{31} + \nu_{21}\nu_{32} & 0 & 0 & 0 \\
\frac{E_2E_3\Delta}{\nu_{12} + \nu_{13}\nu_{32}} & \frac{E_2E_3\Delta}{1 - \nu_{31}\nu_{13}} & \frac{E_2E_3\Delta}{\nu_{32} + \nu_{31}\nu_{12}} & 0 & 0 & 0 \\
\frac{E_1E_3\Delta}{\nu_{13} + \nu_{12}\nu_{23}} & \frac{E_1E_3\Delta}{\nu_{23} + \nu_{13}\nu_{21}} & \frac{E_1E_3\Delta}{1 - \nu_{12}\nu_{21}} & 0 & 0 & 0 \\
\frac{E_1E_2\Delta}{\nu_{12} + \nu_{13}\nu_{32}} & \frac{E_1E_2\Delta}{\nu_{23} + \nu_{13}\nu_{21}} & \frac{E_1E_2\Delta}{1 - \nu_{12}\nu_{21}} & 0 & 0 & 0 \\
0 & 0 & 0 & 2G_{12} & 0 & 0 \\
0 & 0 & 0 & 0 & 2G_{13} & 0 \\
0 & 0 & 0 & 0 & 0 & 2G_{23}
\end{pmatrix}\begin{pmatrix}
\dot{\varepsilon}_{11} \\
\dot{\varepsilon}_{22} \\
\dot{\varepsilon}_{33} \\
\dot{\gamma}_{12}
\end{pmatrix}_{\text{mech}}
\]  

(2.46)

The stress and strain tensors are updated at the end of the each time increment as in Eq. 2.47 and Eq. 2.48 respectively.

\[
\sigma_{ij}^{n+1} = \sigma_{ij}^n + \dot{\sigma}_{ij}^n
\]  

(2.47)

\[
\varepsilon_{ij}^{n+1} = \varepsilon_{ij}^n + \dot{\varepsilon}_{ij}^n
\]  

(2.48)

Equilibrium Equations:

The variation of the stress components as functions of position within the interior of a body is determined in the stress analysis. This can be considered as a type of boundary value problem often encountered in the theory of differential equations, in which the gradients of the variables, rather than the explicit variables themselves, are specified. In the case of stress, the gradients are governed by conditions of static equilibrium. Let the surface traction at any point on a surface \( S \) be the force \( \mathbf{t} \) per unit of area, and let the body force at any point within the volume of material \( V \) under consideration be \( \mathbf{f} \) per unit of current volume. Then, the force equilibrium for this volume \( V \) is written as:

\[
\int_S \mathbf{t} \, dS + \int_V \mathbf{f} \, dV = 0
\]  

(2.49)

The “true” or Cauchy stress matrix \( \mathbf{\sigma} \) at a point of \( S \) is defined by

\[
\mathbf{t} = \mathbf{n} \cdot \mathbf{\sigma}
\]  

(2.50)
where \( \mathbf{n} \) is the unit outward normal to \( S \) at the considered point. This yields in:

\[
\int_S \mathbf{n} \cdot \mathbf{\sigma} dS + \int_V \mathbf{f} dV = 0 \tag{2.51}
\]

Using Gauss’s theorem, the surface integral can be rewritten as a volume integral. Hence, the equilibrium equation given in Eq. 2.51 is expressed as:

\[
\int_S \mathbf{n} \cdot \mathbf{\sigma} dS = \int_V \nabla \mathbf{\sigma} dV = 0 \tag{2.52}
\]

Since the volume \( V \) is arbitrary, this requires that the integrand be zero:

\[
\nabla \mathbf{\sigma} = 0 \tag{2.53}
\]

For Cartesian problems in 3D, the governing differential equations for the static equilibrium state are written by an extension of Eq. 2.53 as:

\[
\frac{\partial \sigma_x}{\partial x} + \frac{\partial \tau_{xy}}{\partial y} + \frac{\partial \tau_{xz}}{\partial z} = 0
\]

\[
\frac{\partial \tau_{xy}}{\partial x} + \frac{\partial \sigma_y}{\partial y} + \frac{\partial \tau_{yz}}{\partial z} = 0 \tag{2.54}
\]

\[
\frac{\partial \tau_{xz}}{\partial x} + \frac{\partial \tau_{yz}}{\partial y} + \frac{\partial \sigma_z}{\partial z} = 0
\]

### 2.2.6 Pulling Force Calculation

The pulling force consists of different contributions, such as i) collimation force \( (F_{col}) \) due to resistances arising from the creel to the die inlet, ii) the bulk compaction force \( (F_{bulk}) \) due to the pressure increase in the tapered portion of the die, iii) the viscous drag \( (F_{vis}) \) acting in the liquid zone, and iv) the frictional force due to the contact between the internal surface of the die and the solidified processing material \( (F_{fric}) \). The first contribution \( F_{col} \) is generally assumed to be negligible, hence the total pulling force \( F_{pull} \) can be expressed as follows [71,73]:

\[
F_{pull} = F_{col} + F_{bulk} + F_{vis} + F_{fric} \approx F_{bulk} + F_{vis} + F_{fric} \tag{2.55}
\]

These pulling force contributions are strictly related to the geometrical features of the die-part interactions as well as the resin phase transitions (liquid-gel-solid). A schematic view of the pulling force contributions are depicted in Fig. 2.8. The compaction force is related to the increase in the resin pressure typically observed in the initial part of the die (tapered) at which the resin is in liquid phase. The compaction pressure allows the resin to completely impregnate the fiber reinforcements. Defining the local resin pressure as \( p \), the die taper angle as \( \theta \), and the inlet surface area as \( A_1 \), the bulk compaction force can be written as follows [74]:

\[
F_{bulk} = \iint_{A_1} p \sin \theta dA_1 \tag{2.56}
\]

Here, \( p \) is obtained from the 2D mechanical analysis of the pultrusion process in which a mechanical contact formulation is defined at the die-part interface providing the pressure
rise. The viscous drag occurs at the die-part interface before the gelation point of the resin. This resistance is imputable to the presence of a thin liquid layer between the travelling fibers and the stationary die surface. The viscous force can be written analytically as follows [74]:

\[
F_{\text{vis}} = \frac{v_{\text{pull}}}{\lambda} \int_{A_2} \eta(\alpha, T) dA_2
\]  

(2.57)

where \( \lambda \) is the thickness of the resin layer between the solid boundary and the moving fibers, \( \eta(\alpha, T) \) denotes the resin viscosity as a function of degree of cure and temperature, \( v_{\text{pull}} \) is the pulling speed, and \( A_2 \) is the surface on which the viscous effects take place and is determined by the gelation point. After the gelation point, the resin flow and the viscous effects are obviously inhibited and the composite is mechanically pulled through the die. Consequently, the interaction between the processing material and the die surface is mainly characterized by frictional effects. Generally, the entity of the frictional force can be inferred by considering the friction coefficient \( \mu \) and the contact pressure \( \sigma \), according to the following equation [74]:

\[
F_{\text{fric}} = \int_{A_3} \mu \cdot \sigma dA_3
\]  

(2.58)

where \( A_3 \) is the die surface from the gelation point to the detachment (separation) point (see Fig. 2.8). The frictional resistance vanishes when the shrinkage effect prevails, inducing the detachment of the material from the die.

Figure 2.8: Pulling force contributions in pultrusion [74].
This chapter is dedicated to the experimental work conducted to characterize an industrial polyester resin system specifically prepared for a pultrusion process \cite{8} (PAPER\textsuperscript{8}). The chemo-rheology of the “orthophthalic” polyester system is characterized using a differential scanning calorimetry (DSC) and a rheometer. The curing behaviour as well as the viscosity and the gelation of the resin are obtained. A modified temperature- and cure-dependent elastic modulus model is developed using a dynamic mechanical analyser (DMA). In addition, the ultimate glass transition temperature of a fully cured sample is also obtained from the DMA. A least squares non-linear regression analysis is carried out to find the parameters in the material constitutive models.

3.1 Cure Kinetics

The temperature and the degree of cure distributions inside the processing material have to be analysed in order to investigate the pultrusion process effectively. The chemical exothermic reaction takes place when the composite reaches the reaction initiation temperature inside the die. In order to capture the thermo-chemical aspects in pultrusion, the cure behaviour has to be characterized.

The cure characteristics of an industrial pultrusion polyester resin are obtained from the DSC tests \cite{8} (PAPER\textsuperscript{8}). The difference in the heat flows from the sample and the reference side of the sensor is measured in the DSC as a function of temperature or time. A variation in the heat flow arise when the resin sample absorbs or releases heat due to the thermal effects such as exothermic reaction during curing. The temperature spectrum used in the DSC is determined based on the processing conditions provided by a commercial pultrusion company. Three different dynamic and isothermal experiments are performed in order to specify the parameters used in the cure kinetics model which describes the curing behaviour of the resin over a wide range of different processing conditions.

Isothermal experiments are carried out at temperatures 120°C, 130°C and 140°C using a resin sample of 10 mg. On the other hand, dynamic scans are performed by heating the sample from 25°C to 200°C with a heating rate of 5°C/min, 7.5°C/min and 10°C/min. It should be noted that high heating rates are employed in pultrusion such that the peak temperature (∼170°C) is obtained by heating the sample from room temperature
(~25°C) within 1-2 min inside the heating die.

The total exothermic heat of reaction ($H_{tr}$) released during cure is calculated approximately as 175±15 kJ/kg by obtaining the integral of the heat flow-time plots for the dynamic DSC experiments. Here, a straight baseline between the onset and the end of the reaction is considered as suggested in [97]. The parameters in the autocatalytic cure kinetics equation (Eq. 2.5) are obtained using a weighted least squares non-linear regression analysis including the experimental data from both isothermal and dynamic DSC scans. The estimated best fit parameters are given in Table 3.1. As seen from the best fit between the experimental data and the predictions in Fig. 3.1, the autocatalytic model (Eq. 2.5) accurately predicts the degree of cure as well as the cure rate evolutions for all three isothermal temperatures. Fig. 3.2 shows that a reasonably good fit is also obtained between the measured and the estimated degree of cure as well as cure rate for different heating rates in dynamic DSC scans.

Table 3.1: The estimated cure kinetics parameters of the polyester (Eq. 2.5) [59].

<table>
<thead>
<tr>
<th>$A_0$ [1/s]</th>
<th>$E_a$ [kJ/mol]</th>
<th>$m$</th>
<th>$n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.5581×10$^9$</td>
<td>82.727</td>
<td>0.63</td>
<td>1.847</td>
</tr>
</tbody>
</table>

Figure 3.1: The experimental and predicted (best fit) degree of cure evolutions (left) and the cure rate as a function of degree of cure (right) for the isothermal DSC scans [59].

3.2 Rheological Behaviour

In pultrusion, the rheological behaviour of the processing resin system directly affects the viscous force development at the die-part interface. The viscous force is one of the contributions (the collimation, the bulk compaction, the viscous and the frictional force) to the pulling force [74]. Before the gelation point, a viscous drag occurs at the die-part interface. The viscous force can be defined as a function of viscosity for pultrusion [67,74]. In order to predict the viscous force correctly, the rheological properties of the processing
resin has to be characterized.

In this thesis, rheological measurements are carried out using a rheometer in “plate-plate” mode for the polyester resin system \(^{59}\) (PAPER\(^8\)). The neat polyester is prepared in liquid form with dimensions of ∼30 mm (diameter) and ∼2 mm (thickness). Circular plates are used in oscillatory mode at 1% strain and 1 Hz. Two temperature cycles (Cycle-1 and Cycle-2) having different heating rates are used in the rheometer. A hold temperature of 100°C and 130°C are considered in Cycle-1 and Cycle-2, respectively. The viscosity evolutions as well as the variations in the storage modulus (\(G'\)) and the loss modulus (\(G''\)) are measured. A well known temperature- and cure-dependent viscosity model is implemented to predict the viscosity development and expressed as \(^{72,74}\):

\[
\eta(\alpha, T) = \eta_\infty \exp\left(\frac{\Delta E_\eta}{RT} + K \alpha\right)
\]  

(3.1)

where \(\Delta E_\eta\) is the viscous activation energy, \(\eta_\infty\) is the initial viscosity, \(K\) is a constant, \(R\) is the universal gas constant, \(T\) is the absolute temperature. A least squares non-linear regression analysis is performed upon the measured data in order to determine the constants in the viscosity model. Fig. 3.3 shows the viscosity evolutions as a function of time and temperature. The estimated model constants are given in Table 3.2. It is seen that a good agreement is found between the measured and estimated (best fit) viscosity profiles for the two different temperature scans. The estimated initial viscosity slightly deviates from the measured one, nevertheless this does not have an important effect on the thermo-mechanical analysis of the pultrusion process since the stiffness of the liquid resin is not high enough to build up stresses.

Table 3.2: The estimated constants used in the viscosity model (Eq. 3.1) \(^{59}\) (PAPER\(^8\)).

<table>
<thead>
<tr>
<th>(\eta_\infty) [Pas]</th>
<th>(\Delta E_\eta) [kJ/mol]</th>
<th>(K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1.08\times10^{-6})</td>
<td>40.147</td>
<td>83.39</td>
</tr>
</tbody>
</table>
3.3 Elastic Modulus

A proper modulus model is required for the calculation of the process induced stresses and the dimensional variations in pultrusion. A temperature- and cure-dependent modulus model is considered in the present study. A DMA is utilized in tension mode by applying a sinusoidal deformation/strain to the sample. The stiffness (modulus) and damping ($\tan \delta$) are measured as a response in the DMA. The modulus can be described in two components: an in-phase component, the storage modulus (elastic behaviour) ($E'$), and an out-of-phase component, the loss modulus ($E''$). $\tan \delta$ is defined as the ratio of the loss modulus to the storage modulus and represents the energy dissipation in the sample. The peak of $\tan \delta$ at which the difference between $E'$ and $E''$ is minimum indicates the glass transition temperature $T_g$.

The DMA experiments are conducted to obtain the evolution of the elastic modulus of the polyester. The DMA is utilized in tension mode by applying a sinusoidal deformation/strain to the sample. The stiffness (modulus) and damping ($\tan \delta$) are measured as a response. The measured modulus can be described in two components: an in-phase component, the storage modulus (elastic behaviour) ($E'$), and an out-of-phase component, the loss modulus ($E''$). $\tan \delta$ is defined as the ratio of the loss modulus to the storage modulus and represents the energy dissipation in the sample. The peak of $\tan \delta$ at which the difference between $E'$ and $E''$ is minimum indicates the glass transition temperature $T_g$.

The neat resin is first cured in the form of rectangular stripes using an oven. Dynamic heating scans are performed from 25°C to 190°C with a heating rate of 5°C/min and a frequency of 6.22 Hz [105]. The static and dynamic load strains are set to 1% and 0.1%, respectively.

Using the modified CHILE model Eq. 2.13 given in Chapter 2, a least squares non-linear regression analysis is performed to obtain the constants in Eq. 2.13 which gives the best agreement with the measured data. The estimated parameters used in the modified
CHILE model are given in Table 3.3. The calculated elastic modulus evolution (best fit) is compared with the experimental data seen in Fig. 3.4. A good agreement is found between the measured and the predicted modulus evolution.

Table 3.3: The estimated constants used in the modulus model (Eq. 2.13) [59].

<table>
<thead>
<tr>
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</thead>
<tbody>
<tr>
<td>-60</td>
<td>30</td>
<td>110</td>
<td>0.0195</td>
<td>0.73</td>
<td>3.76</td>
<td>0.20</td>
<td>0.043</td>
</tr>
</tbody>
</table>

Figure 3.4: Comparison of the measured (DMA) and predicted elastic modulus developments (log scale) [59].
Chapter 4

Summary of Results on Thermo-chemical Modelling

The main results obtained from the thermo-chemical analysis of the pultrusion process are summarized in this chapter. The work presented in [41] (PAPER1) deals with the effect of the thermal contact resistance (TCR) on the thermosetting pultrusion. The thermo-chemical modelling strategies are described in detail in [42] (PAPER2). A 3D thermo-chemical simulation of the pultrusion process for a NACA0018 blade profile is presented in [45] (PAPER3). Novel optimization studies are presented in which single-objective [65] (PAPER10) and multi-objective [66] genetic algorithms are employed based on the thermo-chemical model. Moreover, the results obtained from the probabilistic modelling of the pultrusion process [76,77] (PAPER12, PAPER13) using Monte Carlo simulations (MCS), response surface method (RSM) and first order reliability method (FORM) are also presented.

4.1 Thermal Contact Resistance in the Pultrusion Process

The effects of the thermal contact resistance (TCR), which can also be expressed by the heat transfer coefficient (HTC), on the pultrusion process are investigated in PAPER2 [41]. The pultrusion of a composite rod (initially without the die block) taken from the literature [34] is simulated as a validation case by using the CV-FD method in a 2D domain (see Chapter 2). After validating the thermo-chemical model, a cylindrical die block with heaters is included. A schematic view of the die, the composite and the heaters is seen in Fig. 4.1. In this new configuration, the TCR at the die-composite interface is taken into account for the process simulation. The aim is to investigate the significance of the TCR for the pultrusion process, while having the same centerline temperature and cure degree profile as found in the validation case [34]. The MATLAB computing environment is used for both simulations and temperature fitting procedure.

In order to obtain the same centerline temperature profile of the composite within this pultrusion simulation domain (Fig. 4.1), a curve fitting procedure (i.e. inverse modeling) is performed using the data composed of 15 centerline temperature values measured from the validation case. The TCR values (design variables in the curve-fitting procedure) are predicted by minimizing the difference between the measured (the validation case) and the calculated (the new configuration) centerline temperatures, i.e. $\sum (T_{\text{meas}} - T_{\text{cal}})^2$, for certain die radii. The constrained minimization function “fmincon” is employed using
MATLAB which finds the minimum of a multi-variable problem. The temperature curve fitting procedure is repeated with 5 different die radii \(r_d\) selected as 10, 25, 50, 75 and 100 mm, thereby considering possible die designs for the composite rod in the validation case. Two different optimization case studies are performed:

a) Constant TCR (Case-1): In this case study only a single TCR value (one design variable) through the axial direction is optimized to minimize the error, \(\sum (T_{\text{meas}} - T_{\text{cal}})^2\). A schematic view of the single TCR case can be seen in Fig. 4.1. The single TCR along the axial direction at the die-part interface is indicated with the red color.

b) Variable TCR (Case-2): In this problem 9 equally spaced (each of ~100 mm) TCR regions are defined along the interface. The configuration of the variable TCR regions can be seen in Fig. 4.1. The discrete TCR regions are indicated with the blue color.

The minimum error \(\sum (T_{\text{meas}} - T_{\text{cal}})^2\) values, which give the optimum TCR values for both cases, are found to be 6957.5, 7979.8, 8250.7, 297.5 and 8275.9 in Case-1 and 6.7, 5.2, 3.7, 7.4 and 19.4 in Case-2 for the die radii of 10, 25, 50, 75 and 100 mm, respectively. The minimum error for Case-1 in which a single TCR is used is significantly higher than the error for Case-2 with respect to all die radii. This shows that the use of a single TCR is not suitable for the thermo-chemical simulation of the pultrusion process as compared with the use of variable TCR at the die-part interface. The discrete TCR can be adjusted according the curing as well as the chemical shrinkage of the part inside the heating die. The centerline temperature and the cure degree profiles of the composite rod for a die radius of 10 mm are seen in Fig. 4.2. The temperature and the cure degree profiles obtained with the use of variable TCRs are almost the same as those given in the validation case. However the results obtained by using a single TCR deviate considerably with respect to the centerline temperature and the cure profiles of the composite rod.

![Figure 4.1: Schematic representation of the pultrusion domain of the composite rod including the cylindrical die block and the heaters with TCR regions.](image-url)
4.2 Strategies for Thermo-Chemical Analysis of Pultrusion

In PAPER$^2$ $^{42}$, 3D numerical modelling strategies of a thermosetting pultrusion process are investigated considering both transient and steady state approaches as described in Chapter$^2$ $^2$. For the transient solution, an unconditionally stable alternating direction implicit Douglas-Gunn (ADI-DG) scheme is implemented as the first contribution of its kind in this specific field of application. The results are compared with corresponding results obtained from i) the transient fully implicit scheme, ii) the straightforward extension of the 2D ADI and iii) the steady state approach. A design of experiments is also carried out for the curing characteristic of the product based on the pulling speed and the part thickness.

A schematic view of the 3D thermo-chemical model is depicted in Fig. 4.3. The temperature and degree of cure developments are firstly calculated and validated by comparing the results from literature as seen in Fig. 4.4. The computational details of the proposed approaches are given in Table 4.1. It is seen that the ADI-DG method is approximately 2 times faster than the transient implicit method even though the numbers of time increments for reaching the steady state condition are very close to each other. On the other hand, the steady state approach is extremely fast since it only requires 6 iterations to satisfy the steady state conditions and it takes only 0.43 s in computational time for the same problem having the same discretization and $Pe$ as used in the transient approach.

The transient solution is suitable for the simulation of the pultrusion process when the transient process conditions are pronounced such as time and/or temperature dependent material properties, time dependent “switch on/off” type heaters etc. Unlike the transient solution, the steady state solution is more convenient for the simulation of pultrusion having constant processing conditions throughout the process in terms of
Figure 4.3: Pultrusion domain of the composite flat plate. A mesh of $61 \times 10 \times 11$ CVs which corresponds to 6710 CVs is used for the numerical model.

Figure 4.4: The predicted centerline temperature (left) and cure degree (right) profiles. It should be noted that the present results (ADI-DG, Implicit and Steady-state) are very close to each other and hence practically overlapping.

computational efficiency. The proposed transient solver (ADI-DG) implemented for the pultrusion process simulation is found to be relatively faster as compared to the transient fully implicit method. Note that the ADI-DG solver includes the advection term and the non-linear internal heat generation in the energy equation. The steady state solution is found to be extremely fast as compared to the transient approaches. The increase in absolute computational time is very small for the steady state solution with an increase in the CVs as compared to the transient solutions. This makes the difference between steady state and transient solution even more pronounced for larger meshes than the one considered here.
Table 4.1: Comparison of the proposed transient and steady state approaches with 61 CVs in the pulling direction.

<table>
<thead>
<tr>
<th></th>
<th>Transient (ADI-DG)</th>
<th>Transient (Implicit)</th>
<th>Steady-state approach</th>
</tr>
</thead>
<tbody>
<tr>
<td>Computational time</td>
<td>5.26</td>
<td>10.82</td>
<td>0.43</td>
</tr>
<tr>
<td>[s]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of time</td>
<td>168</td>
<td>175</td>
<td>6</td>
</tr>
<tr>
<td>increments/iterations to reach the steady state</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\Delta t) [s]</td>
<td>4.5</td>
<td>4.5</td>
<td>-</td>
</tr>
<tr>
<td>(P_e)</td>
<td>92.0</td>
<td>92.0</td>
<td>92.0</td>
</tr>
<tr>
<td>(C_r)</td>
<td>1.0</td>
<td>1.0</td>
<td>-</td>
</tr>
</tbody>
</table>

4.3 Process Simulation of a Pultruded NACA0018 Blade Profile

A simulation is performed for the pultrusion of a NACA0018 blade profile having a curved geometry in [45]. The evolutions of the temperature and cure degree distributions are predicted inside the heating die and in the post-die region where convective cooling prevails. The effects of varying process conditions on the part quality are investigated for two different heater configurations and with three different pulling speeds.

The model geometry is shown in Fig. 4.5. Only half of the cross section has been considered due to the symmetry. Three heating platens are assumed to be placed on the top surface of the die block and the remaining platens are placed on the bottom surface of the die. The cross sectional details of the die and the composite including the FE mesh are illustrated in Fig. 4.6. Two case studies were carried out based on the set temperature of the heaters which are given in Table 4.2. Three different pulling speeds were used for both cases: 2.3 mm/s, 3 mm/s and 5 mm/s, implying total pultrusion process times of approximately 73 min, 56 min and 34 min, respectively, based on the total length of the profile which is approximately 10 m.

The predicted temperature and cure degree profiles for Case-1 and Case-2 are presented in Fig. 4.7 and Fig. 4.8 respectively. In both cases, the temperature at the die-part interface (e.g. at point B in Fig. 4.7 and Fig. 4.8) remains almost the same for different pulling speed values due to the prescribed temperature of the heaters. The temperature profile at point A is more sensitive to the increase in pulling speed. The profile shifts to the right for both cases when the pulling speed is increased. The degree of cure distributions follow the same trend: both the temperature and the cure degree profiles at point A shift more to the right than for point B for the two cases as the pulling speed increases. Obviously, also the cure degree profile at point A is more sensitive to the pulling speed than the cure degree profile at point B.

Table 4.2: The set temperatures of the heaters (°C) used in the validation case.

<table>
<thead>
<tr>
<th>Heater-1</th>
<th>Heater-2</th>
<th>Heater-3</th>
<th>Heater-4</th>
<th>Heater-5</th>
<th>Heater-6</th>
</tr>
</thead>
<tbody>
<tr>
<td>105.5</td>
<td>148.5</td>
<td>200.0</td>
<td>115.5</td>
<td>146.5</td>
<td>200.0</td>
</tr>
</tbody>
</table>
In addition, the maximum temperature values at point A are found to be higher than those at point B for all cases. This results in through-thickness temperature and cure degree gradients which together with the heater configuration would have a direct effect on the process induced residual stresses and distortions. The temperature may still increase after the profile has left the die due to exothermic internal heat generation as the curing process continues. In other words, an increase in the pulling speed promotes larger through-thickness gradients of temperature and degree of cure in both cases. The pulling speed has a negative effect on the cure degree at the die exit, i.e. it decreases with an increase in speed; however, this effect vanishes at the end of the process where almost the same cure degree values are obtained for different pulling speeds in both cases.

Using the proposed numerical tool, designs of various internal cross-section configurations, e.g. reinforcements, stiffeners, spars, etc., can be analysed including the effects of processing parameters on the expected performance. In this way, the simulation can be used to optimize the process in order to obtain the desired mechanical properties of the product.
Figure 4.7: Temperature (top) and cure degree (bottom) profiles at points A and B with zoomed plots (right) between 0-2 m from the heating die entrance for Case-1 (105.5-148.5-200 °C).
Figure 4.8: Temperature (top) and cure degree (bottom) profiles at points A and B with zoomed plots (right) between 0-2 m from the heating die entrance for Case-2 (171-188-188 °C).
4.4 Optimization of the Pultrusion Process

The productivity of the process is improved by using a mixed integer genetic algorithm (MIGA) in [65] (PAPER10) and this is a novel single objective optimization problem for pultrusion to optimize the process. The total number of heaters is minimized while satisfying the constraints for the maximum composite temperature, the mean of the cure degree at the die exit and the pulling speed. For this purpose, the 2D CV-FD thermochemical model is employed including the die block and the heaters.

In the MIGA approach, there are 5 equally spaced cylindrical heaters attached to the die block as seen in Fig. 4.9. The objective is to minimize the number of heaters while increasing the speed of the process as well as the mean of the cure degree at the die exit. Sets of different sequential combinations of active heaters are also considered in the optimization procedure. The MIGA is a mixed integer problem containing both continuous and discrete design variables. In the optimization study, the temperature of each heater \( T_i \) is varying between 150°C and 250°C. The pulling speed \( u \) is changing between 5 mm/s (validation case) and 13 mm/s. The constraints are determined as the following:

- The maximum temperature of the composite should not exceed the degradation temperature which is taken to be 240°C \( (T_{max} < 240°C) \).
- The mean cure degree at the die exit should be higher than 0.85 \( (\alpha > 0.85) \). This value is chosen considering the mean of cure in the validation case.
- The pulling speed should be higher than 5 mm/s \( (u > 5\text{mm/s}) \).

![Figure 4.9: Schematic view of the pultrusion process used in the MIGA.](image)

In the MIGA, the best fitness of the population is stored while considering the pulling speed of the process. The total number of active heaters is minimized such that the optimum configuration is found as “01000” (i.e. the total number of heaters is 1) which shows that only the second heater is used while satisfying the constraints. The convective cooling boundary condition is applied to the rest of the heater regions. The corresponding temperature of the second heater is found to be 233.7°C. The pulling speed is increased to 11.1 mm/s and the centerline cure degree of the composite rod at the die exit is enhanced to 0.869. The mean of the cure degree at the die exit and maximum composite temperature are found as 0.856 and 239.3°C, respectively, which shows that the constraints are satisfied.
Corresponding centerline temperatures and cure degree profiles are seen in Fig. 4.10 for the optimum process configuration. The centerline temperature and cure degree profiles are shifted to the right which shows that the temperature is advected along the axial distance faster than in the validation case since the pulling speed is increased. Moreover, the behavior of the convective cooling boundaries at the outermost surface of the die geometry can also be seen in Fig. 4.10 (left). The first, third, fourth and fifth heater regions are exposed to ambient temperature, so only the second heater is active and this is easily seen on the temperature of the path A-A (see Fig. 4.9 (left)).

As briefly mentioned in Chapter 1, there is a limited number of studies in numerical optimization aspects of the pultrusion process. The limitations can in general be related to the nature of the process simulation or the challenges in the field of numerical optimization. The more physics involved with the use of integrated models, the more set of parameters and interestingly even more objectives need to be considered for the optimization procedure. Current common attempts in optimizing manufacturing processes (based on simulations) with the design of experiments techniques will eventually be replaced by the automated and/or interactive optimization procedures due to the need for efficient ways of calculation of the immense number of parameter combinations. In the optimization problems specific for the pultrusion process, as common in most engineering optimization applications, these desired goals are often conflicting with each other, which in turn involves multiple trade-off solutions. Having more than one solution, hence having an overall idea about the trends of gain and sacrifices, gives engineers the opportunity to select the best “available” solution among those multiple competitive choices.

In addition to the MIGA, a multi-objective problem (MOP) considering thermochemical aspects of the pultrusion process has been formulated in [66]. In this study, the pulling speed is maximized and the heating power is minimized simultaneously without defining any preference between them. An evolutionary multi-objective optimization (EMO) algorithm, non-dominated sorting genetic algorithm (NSGA-II [106]), has been
used to solve this MOP in an ideal way. The outcome is a set of multiple solutions (i.e. Pareto-optimal solutions) and each solution is theoretically an optimal solution corresponding to a particular trade-off among objectives \[107\,109\]. Following the solution process, in other words obtaining the Pareto-optimal front, a further post-processing study has been performed to unveil some common principles existing between the variables.

A 3D thermo-chemical analysis of the pultrusion process for a UD flat plate is considered for the MOP in \[66\]. The pultrusion domain as well as the cross sectional details are shown in Fig. 4.11. The details of the parameters seen in Fig. 4.11 are listed in Table 4.3. The constrained MOP is formulated in Eq. 4.1. The first objective \(f_1(x)\) is to maximize the pulling speed in order to increase the production rate. However, the optimization algorithm is implemented to minimize the given objective, therefore maximization problem is equally converted to a minimization problem by considering the negative of the original function (i.e. design variable in this particular case). The second objective \(f_2(x)\) is the minimization of a so-called total energy consumption (TOC) criterion. This is defined as a function of the total heating area \(\text{Area}_i\) and the active heater temperatures \(T_i\), i.e. the number of heaters is also variable. This MOP is constrained with one geometrical and two process response related constraints as well as the side constraints for the design variables. The geometrical constraint makes sure that the length of the die is bigger than the total length of initial free space, heater and spacing between the active heaters. The process related constraints are defined as follows: i) the maximum temperature in the composite should not exceed 240°C, ii) the average cure degree of the composite cross section at the die exit should be higher than 0.9.

Figure 4.11: Schematic view of the pultrusion domain for the MOP.
Table 4.3: The details of the parameters used in the thermo-chemical analysis [66].

<table>
<thead>
<tr>
<th>#</th>
<th>Parameter</th>
<th>Description</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$L_{ini}$</td>
<td>Initial free length</td>
<td>60 mm</td>
</tr>
<tr>
<td>2</td>
<td>$L_1$</td>
<td>Length of the 1st heater</td>
<td>270 mm</td>
</tr>
<tr>
<td>3</td>
<td>$L_2$</td>
<td>Length of the 2nd heater</td>
<td>270 mm</td>
</tr>
<tr>
<td>4</td>
<td>$L_3$</td>
<td>Length of the 3rd heater</td>
<td>270 mm</td>
</tr>
<tr>
<td>5</td>
<td>$s_1$</td>
<td>Spacing between the 1st and 2nd heaters</td>
<td>15 mm</td>
</tr>
<tr>
<td>6</td>
<td>$s_2$</td>
<td>Spacing between the 2nd and 3rd heaters</td>
<td>15 mm</td>
</tr>
<tr>
<td>7</td>
<td>$T_1$</td>
<td>Set temperature of the 1st heater</td>
<td>171°C</td>
</tr>
<tr>
<td>8</td>
<td>$T_2$</td>
<td>Set temperature of the 2nd heater</td>
<td>188°C</td>
</tr>
<tr>
<td>9</td>
<td>$T_3$</td>
<td>Set temperature of the 3rd heater</td>
<td>188°C</td>
</tr>
<tr>
<td>10</td>
<td>$n_h$</td>
<td>Number of heaters</td>
<td>3</td>
</tr>
<tr>
<td>11</td>
<td>$L_{die}$</td>
<td>Length of the die</td>
<td>915 mm</td>
</tr>
<tr>
<td>12</td>
<td>$u$</td>
<td>Pulling speed</td>
<td>200 mm/min</td>
</tr>
<tr>
<td>13</td>
<td>$2w$</td>
<td>Width of the die</td>
<td>76.2 mm</td>
</tr>
<tr>
<td>14</td>
<td>$2h$</td>
<td>Height of the die</td>
<td>76.2 mm</td>
</tr>
</tbody>
</table>

Maximize : $f_1(x) = -u$,  
Minimize : $f_2(x) = TOC = \sum_{i=1}^{n_{heaters}} \text{Area}_i T_i = \sum_{i=1}^{n_{heaters}} (wL_i)T_i$,  
subject to :  
$g_1(x) = L_{die} > L_{ini} + \sum_{i=1}^{n_{heaters}} L_i + \sum_{j=1}^{n_{heaters}-1} s_j$,  
$g_2(x) = T_{max} < 240^\circ\text{C}$,  
$g_3(x) = \alpha_{avg,exit} > 0.9$,  
$g_4(x) = 60 \leq L_{ini} \leq 240 \text{ mm}$,  
$g_{5-7}(x) = 60 \leq L_1, L_2, L_3 \leq 360 \text{ mm}$,  
$g_{8-9}(x) = 15 \leq s_1, s_2 \leq 250 \text{ mm}$,  
$g_{10-12}(x) = 150 \leq T_1, T_2, T_3 \leq 250^\circ\text{C}$,  
$g_{13}(x) = 1 \leq n_{heaters} \leq 3$,  
$g_{14}(x) = 750 \leq L_{die} \leq 1500 \text{ mm}$,  
$g_{15}(x) = 100 \leq u \leq 700 \text{ mm/min}$,  
$g_{16-17}(x) = 45.4 \leq w, h \leq 200 \text{ mm}$.

The results of the MOP based on the 3D thermo-chemical analysis of the pultrusion are shown in Fig. 4.12 (left). From the course of evolution it is clearly seen that the initial design (black diamond) results in much higher energy consumption ($f_2$: TOC). There are total of 9,936 feasible (blue markers), 164 infeasible solutions (red markers) and 133 of these feasible solutions form the Pareto-optimal front (green circles). The Pareto solutions, which are also called non-dominated solutions, are better performing solutions with respect to both of the objectives as compared to the rest while satisfying the constraints. The leftmost solution can be considered to be an outlier. The maximum pulling speed is found to be approximately 540 mm/min. However it should be kept in mind that the obtained front can be improved with further iterations (“generations” in evolutionary computing terminology) as well as incorporating a local search capability.
This preliminary study is performed to provide the first ideal solution methodology for a real multi-objective optimization problem in this particular manufacturing process. The heater configuration is given in Fig. 4.12 (right). If the solutions are sorted in the same order with the Pareto solutions from left to right (i.e. from highest pulling speed to lowest pulling speed), the number of heaters being used is varying between 3 and only 1 which indicates that there is need for more heat to cure the composite at higher pulling speeds as expected.

Figure 4.12: Distribution of all solutions during the course of evolution (left): Blue markers show the feasible solutions, red markers show the infeasible solutions, green circles indicate the Pareto-optimal solutions and black diamond stands for the initial design [66]. Variation of the number of heaters along the Pareto front (right).

4.5 Probabilistic Modelling and Reliability Analysis

A reliability assessment of the pultrusion process is investigated in PAPER[12] by using the first order reliability method (FORM) and the Monte Carlo simulations (MCS). Pultrusion of a flat plate used in [42] (PAPER[2]) is considered. The CV-FD approach is employed and the predicted temperature and cure degree profiles of the composite are found to match well those in similar analyses from literature [28,33]. The cumulative distribution functions (CDFs) of the centreline degree of cure at the exit (CDOCE) and the maximum composite temperature ($T_{\text{max}}$) during the process are calculated by using the FORM and the results are compared with the MCS.

For the probabilistic analysis of the pultrusion process, the pulling speed, fiber volume ratio, inlet temperature, heater temperature multiplier, all the characteristic material properties and the resin kinetic parameters are considered as the random input parameters (RIPs). The random variables are listed in PAPER[12] have been utilized to predict and assess the reliability of the pultrusion process. In a pultrusion process, the CDOCE is desired to be sufficiently high while the maximum composite temperature ($T_{\text{max}}$) is expected not to be higher than the degradation temperature of the epoxy resin. Hence, the CDOCE being less than a certain value (i.e. critical cure degree, $\alpha_{\text{crit}}$) and $T_{\text{max}}$ being greater than another value (i.e. critical temperature, $T_{\text{crit}}$) are selected as the limit.
The CDF of the CDOCE and $T_{\text{max}}$ are calculated by using the FORM based on the random variables given in Table 3. The CDFs can be expressed as the probability of the CDOCE being less than $\alpha_{\text{crit}}$, $P(\text{CDOCE} \leq \alpha_{\text{crit}})$ and the probability of $T_{\text{max}}$ being greater than $T_{\text{crit}}$, $P(T_{\text{max}} \geq T_{\text{crit}})$. In order to validate the implementation of the FORM, the same CDFs are also predicted by using the MCS with the LHS. The MCS is one of the most common techniques used for uncertainty analyses. The LHS technique is selected for the sampling method since it has a sample memory which avoids the repetition of the samples. However, its accuracy depends on the sample size and computational expense is much higher as compared with that of FORM for a certain number of random variables.

A total of 1000 samples (function evaluations) are used in the MCS analysis. On the other hand, it requires approximately 50 function evaluations for the FORM to calculate the probability of failure for this problem which has 14 design variables. The FORM is performed for 14 different $\alpha_{\text{crit}}$ and 8 different $T_{\text{crit}}$ values based on the equivalent division of the CDOCE and $T_{\text{max}}$ intervals. The results are given in Fig. 4.13(left) and Fig. 4.13(right) for the CDF of the CDOCE and $T_{\text{max}}$, respectively. It is seen that the FORM gives accurate results as compared with the ones obtained from the MCS. For instance, the probability of the CDOCE being less than 0.89, i.e. $P(\text{CDOCE} \leq 0.89)$ is approximately 20\% (i.e. indicated with a solid line in Fig. 4.13(left)). The probability of $T_{\text{max}}$ being greater than 195\degree C, i.e. $P(T_{\text{max}} \geq 195\degree \text{C})$, is approximately 7\% (100-93=7) (i.e. similarly in Fig. 4.13(right)).

Regarding the sensitivities of the random variables with respect to the CDOCE and $T_{\text{max}}$, similar results are obtained from the FORM and the MCS. The sensitivities are taken from the gradient information of the random variables for the FORM. The normalized magnitude vector is called the sensitivity indicator. On the other hand the sensitivities are obtained by calculating the linear correlation coefficients for the MCS. The sensitivities of the random input variables are given in Fig. 4.14 and Fig. 4.15 as a pie chart for the CDOCE and the $T_{\text{max}}$, respectively. It is seen from Fig. 4.14 that the FORM and the MCS results are very close to each other based on the most sensitive 4 random variables, namely the activation energy ($E$), the heater temperature multiplier.
(cons), the order of reaction (n) and the pulling speed (u) which cover almost 90% of the sensitivity distribution. According to the sensitivity indicator for the FORM as a bar plot in Fig. 4.14 (left) and the linear correlation coefficients for the MCS in Fig. 4.14 (right), the statistical variation in E and cons has the highest effect on the variation in the CDOCE. The linear correlation coefficient or the sensitivity indicator value for E and cons are calculated as approximately (-0.78) and (0.61), respectively. Here, a positive correlation indicates that an increase in the input parameter provides an increase in the output parameter and vice versa. According to Fig. 4.15 cons has the highest sensitivity indicator value or correlation coefficient (positive) and the magnitude is close to 1 (0.98). This indicates that the variation in the heater temperatures is strongly correlated with the $T_{\text{max}}$. The corresponding sensitivities are given in a pie chart in Fig. 4.15 (left) and Fig. 4.15 (right) for the FORM and the MCS, respectively.

![Figure 4.14: The sensitivity indicators and the linear correlation coefficients calculated by using the FORM (left) and the MCS (right), respectively, for the CDOCE.](image1)

![Figure 4.15: The sensitivity indicators and the linear correlation coefficients calculated by using the FORM (left) and the MCS (right), respectively, for the $T_{\text{max}}$.](image2)

In [77] (PAPER\textsuperscript{13}), a new application for the probabilistic analysis of the pultrusion process is presented. The effects of the uncertainties in the pultrusion process parameters
on the degree of cure at the die exit are investigated. The deterministic analysis is based on a thermo-chemical model of the pultruded AS4/Epon 9420/9470/537 carbon fiber/epoxy composite rod taken from the literature [34]. The model geometry and the boundary conditions are shown in Fig. 4.16.

For the probabilistic analysis, the response surface method (RSM), which is the first contribution of its kind in the numerical modelling of pultrusion process, have been performed. For this purpose, the probabilistic design system (PDS) toolbox of ANSYS [101] is utilized incorporating with a parametric deterministic model developed by the author using its own scripting language APDL (ANSYS Parametric Design Language). The results obtained from the RSM are verified by performing MCS. Fig. 4.17 shows the Gauss plots of the CDFs considered in the probabilistic analysis of the pultrusion process. It is seen that the overall MCS results agree with the RSM results. The variations in the activation energy as well as the density of the resin are found to have a relatively stronger influence on the centerline degree of cure at the exit. Moreover, different execution strategies are examined for the MCS to investigate their effects on the accuracy of the random output parameter (see PAPER$^{13}$).

Figure 4.17: The Gauss plots of the CDFs calculated in Case-1 and Case-2 (see PAPER$^{13}$).
Chapter 5

Summary of Results on Thermo-mechanical Modelling

This chapter focuses on the main results obtained from the novel thermo-chemical-mechanical analyses of the pultrusion process developed in this PhD work. The state-of-the-art numerical approaches already presented in Chapter 2 are employed to address the process induced stresses and shape distortions in pultrusion. The 2D plane strain mechanical model presented in PAPER\textsuperscript{4} \cite{47} is further improved in PAPER\textsuperscript{5} \cite{55} by using the full 3D mechanical model. Modelling the pultrusion process containing both UD and CFM layers has not been considered in the literature up to now. A numerical simulation tool embracing the thermo-chemical and mechanical aspects of the pultrusion for an industrial rectangular hollow profile (PAPER\textsuperscript{6}) \cite{56} and L-shaped product (PAPER\textsuperscript{7}) \cite{57} is hence being developed by the author. In addition, the internal stresses at the web flange junction of a pultruded I-beam profile are investigated using the devised numerical tool. An integrated modelling study is carried out (PAPER\textsuperscript{9}) \cite{49} in which the manufacturing aspects of the pultrusion process are associated with the preliminary subsequent service loading scenario for a pultruded NACA0018 blade profile. Finally, the main outcomes of the work in \cite{74} (PAPER\textsuperscript{11}) based on the modelling the pulling force in pultrusion are presented.

5.1 State-of-the-art Models for Prediction of the Residual Stresses and Shape Distortions in Pultrusion

The temperature and degree of cure distributions are first calculated using the 3D transient thermo-chemical analysis of a pultruded square product. Afterwards, these profiles are mapped to the 2D (PAPER\textsuperscript{4}) \cite{47} and 3D (PAPER\textsuperscript{5}) \cite{55} quasi-static mechanical analyses of the pultrusion process. The already developed 2D plane strain mechanical model in \cite{47} (PAPER\textsuperscript{5}) is further improved using generalized plane strain elements. 3D quadratic brick elements are used in the 3D model for the calculation of the process induced longitudinal stresses as well as transverse stresses. The evolution of the transient stresses and distortions are captured and the obtained results are compared with each other using the three different mechanical models (i.e. 2D plane strain, 2D generalized plane strain and 3D models).

A glass/epoxy is considered for the pultruded product in which the fiber reinforcement orientation is UD along the pulling direction and steel is used for the die. Only a quarter
of the pultrusion domain, seen in Fig. 5.1, is modelled due to symmetry. The details of the cross section are also shown in Fig. 5.1. In order to reflect the 3D mechanical behaviour of the process more precisely, the mid section of the composite part is considered since the pultrusion is a continuous process, i.e. there is always material present inside the heating die during the process. The details of the mid section are shown in Fig. 5.2 in which a schematic view of the movement of the 3D part in the pulling direction is also depicted.

<table>
<thead>
<tr>
<th>TH1 [°C]</th>
<th>TH2 [°C]</th>
<th>TH3 [°C]</th>
</tr>
</thead>
<tbody>
<tr>
<td>171</td>
<td>188</td>
<td>188</td>
</tr>
</tbody>
</table>

Figure 5.1: Schematic view of the pultrusion domain for the pultruded square beam. All dimensions are in mm.

The contour plots of the in-plane stresses at the end of the process are seen in Fig. 5.3. It is found that the stress distributions in the transverse directions over the cross section of the part have almost the same pattern for these three different types of analysis. For the longitudinal stress distribution, the generalized plane strain results and the 3D results have similar distribution trends such that tension and compression stresses in the pulling direction are predicted (see \textsuperscript{55} \textsc{Paper}\textsuperscript{5}). On the other hand, the plane strain solution predicts unrealistic longitudinal stress values such that the whole cross section is under tension showing that self static equilibrium in the longitudinal direction is not fulfilled as expected. The reason for this is that there is no strain in the longitudinal direction for the plane strain assumption. This may not be a valid assumption for the calculation of the longitudinal stresses, since there is an existing strain in the pulling direction. In addition to the stress predictions, the evolution of the displacement is also predicted for the top in the $x_2$-direction (U2) (see \textsuperscript{55} \textsc{Paper}\textsuperscript{5}). It is found that there is a good match between the U2 evolutions obtained using the three different element types.
Figure 5.2: Schematic view of the movement of the 3D part in the pulling direction and the positioning of the mid section. The sizes of the die and the part are not scaled.

Figure 5.3: Undeformed contour plots of the normal stress $S_{11}$ calculated using the plane strain elements (a), the generalized plane strain elements (b) and the 3D elements (c) at the end of the process ($x_3 \approx 14.6$ m). All units are in Pa.
Chapter 5. Summary of Results on Thermo-mechanical Modelling

5.2 Warpage in a Rectangular Hollow Profile

A novel thermo-chemical-mechanical analysis of the pultrusion process is presented in [56] PAPER. A process simulation is performed for an industrially pultruded rectangular hollow profile containing both unidirectional (UD) roving and continuous filament mat (CFM) layers. The reinforcements are impregnated with a commercial polyester resin mixture (Atlac 382 [105]). The reactivity of the resin is obtained from gel tests performed by the pultruder. The cure kinetics parameters are estimated from a fitting procedure against the measured temperature. Two different micromechanics approaches are used to calculate the instantaneous mechanical properties of the UD and the CFM layers as discussed in Chapter 2.

The 3D thermo-chemical analysis implemented in ABAQUS [102] is used to simulate the pultrusion process of a rectangular hollow profile. A glass/polyester is considered for the UD and the CFM layers and steel is used for the die. The details of the process set-up is taken from a commercial pultrusion company. A schematic representation of the model is seen in Fig. 5.4. Only a quarter model is used due to symmetry conditions. The 3D thermo-chemical model is subsequently coupled with a 2D quasi-static mechanical model in which quadratic generalized plane strain elements are utilized in ABAQUS. The cross sectional details including the meshing are given in Fig. 5.5. It is seen that the dimension of the processing hollow rectangular profile is 64×27×3 mm. A local material orientation is employed for the CFM layer as seen in Fig. 5.5 in order to reflect the in-plane (x1- and x2-direction) and the out-of-plane (x3-direction) properties correctly. For the round corner, a cylindrical local coordinate system is used in which the x2- and x3-directions are taken as the tangential (T) and radial (R) directions, respectively. The die and the mandrel are also included in the 2D mechanical model (see Fig. 5.5) in which a mechanical contact formulation is defined at the die-part and mandrel-part interface. By using this contact formulation, a separation due to the thermal contraction of the part and/or the resin shrinkage at the interface is allowed and any expansion of the composite beyond the tool interfaces is restricted. The friction force at the contact surfaces is assumed to be zero (sliding condition). Symmetry mechanical BCs are applied at the symmetry lines and the die is assumed to be fixed at the outer regions as shown in Fig. 5.5.

![Figure 5.4: Schematic view of the quarter pultrusion domain for the pultruded rectangular hollow profile. All dimensions are in mm.](image)
The predicted vertical displacement development for point A (die-part interface) in the $x_3$-direction is depicted in Fig. 5.6 for various total volumetric shrinkage values ($V_{sh} = 0.02, 0.04, 0.06, 0.08, 0.10$ and $0.12$). The trend of the displacement development is found to be almost the same for different $V_{sh}$ values. However, the magnitude of the displacement decreases with a decrease in $V_{sh}$ which is expected. The detachment at the die-part interface is captured using the mechanical contact formulation at the interface as seen from Fig. 5.6. The part separates from the die due to the chemical shrinkage. The detachment point where the displacement starts being negative shifts left towards the die inlet as the $V_{sh}$ increases.

The deformed contour plots in Fig. 5.7 shows the predicted displacement field in the $x_3$-direction for $V_{sh}$ values of 0.02, 0.06 and 0.10 at the end of the process. The deformation or warpage pattern is found to be almost the same for various $V_{sh}$ values, however the magnitudes are different as aforementioned. The predicted warpage patterns (Fig. 5.7) match well with the warpage observed in the real pultruded rectangular hollow products seen in Fig. 5.7(right). The magnitude of the warpage ($w$) at point A is defined as the difference between the magnitude of the displacements at point A ($w_a$) and point C ($w_c$) seen in Fig. 5.7(left), i.e. $w = w_a - w_c$. The warpage magnitudes of the pultruded products are also measured at the room temperature. The predicted and the measured warpage values are given in Table 5.1. A good agreement is found between the predicted
and the measured values such that the predicted warpage value for $V_{sh}$ of 0.10 and 0.12 are in the range of measured values.

Figure 5.6: The displacement evolution at point A (die part interface) in the $x_3$-direction (left) and the corresponding zoomed plot inside the die (0-1 m) (right) for various $V_{sh}$ values.

Figure 5.7: Deformed contour plots of the residual displacement field in the $x_3$-direction and the initial geometry of the cross section (undeformed frame) (left). Scale factor for the deformed shape is 10. The warpage formation observed in the real pultruded hollow rectangular profiles (right).
Table 5.1: The predicted and measured values for the warpage defined as \( w = w_A - w_C \) (see Fig. 5.7).

<table>
<thead>
<tr>
<th>( w_A ) [mm]</th>
<th>( w_C ) [mm]</th>
<th>Warpage ( w = w_A - w_C ) [mm]</th>
<th>( w_A - w_C ) [mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>( V_{sh} = 0.02 )</td>
<td>0.101</td>
<td>0.049</td>
<td>0.052</td>
</tr>
<tr>
<td>( V_{sh} = 0.04 )</td>
<td>0.111</td>
<td>0.051</td>
<td>0.060</td>
</tr>
<tr>
<td>( V_{sh} = 0.06 )</td>
<td>0.122</td>
<td>0.052</td>
<td>0.070</td>
</tr>
<tr>
<td>( V_{sh} = 0.08 )</td>
<td>0.140</td>
<td>0.053</td>
<td>0.087</td>
</tr>
<tr>
<td>( V_{sh} = 0.10 )</td>
<td>0.164</td>
<td>0.061</td>
<td>0.103</td>
</tr>
<tr>
<td>( V_{sh} = 0.12 )</td>
<td>0.192</td>
<td>0.075</td>
<td>0.117</td>
</tr>
<tr>
<td>( V_{sh} = 0.15 )</td>
<td>0.15 ± 0.05</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

5.3 Spring-in in a Pultruded L-shaped Product

A numerical simulation tool is developed to calculate the process induced stresses and dimensional variations in an industrially pultruded L-shaped profile [57] (PAPER7). An “orthophthalic” polyester resin system presented in Chapter 3 is utilized to wet-out the reinforcements. The spring-in formation is predicted and compared with the real pultruded products. The residual spring-in angle is further calculated using the developed simulation tool for different pulling speed values. The classical laminate theory (CLT) [110] is employed to verify the predicted through-thickness residual stress field.

An L-shaped profile was pultruded in a commercial pultrusion company. The pictures of the product are shown in [57] PAPER7. The cross sectional dimensions of the part were 50×50×5 mm and it contained glass/polyester based UD roving and CFM layers. A CFM having a density of 450 g/m² was used in the process. The heating die was made of chrome steel. A schematic representation of the pultrusion model is seen in Fig. 5.8. Only half of the model is used due to symmetry.

![Figure 5.8: Schematic view of the half pultrusion domain for the L-shaped pultruded profile. All dimensions are in mm.](image)

The cross sectional details are shown in Fig. 5.9. Local material orientations (Local CS-1 and Local CS-2) are defined for the CFM layer in order to reflect the in-plane (\( x_1 \)- and \( x_2 \)-direction) and the out-of-plane (\( x_3 \)-direction) properties correctly. A cylindrical
local coordinate system (Local CS-1) is used for the round corner in which $x_2$- and $x_3$-direction are taken as the tangential (T) and radial (R) directions, respectively.

Figure 5.9: The cross sectional details of the pultruded L-shaped profile and the die. All dimensions are in mm.

Fig. 5.10 shows the predicted displacements as a deformed contour plot in the global $x_2$-direction ($U_{22}$) and $x_3$-direction ($U_{33}$) at the end of the process ($x_1 = 6$ m). It is seen that a spring-in formation is found according to the predicted deformation pattern. This phenomenon agrees quite well with the spring-in pattern observed in the real pultruded L-shaped profiles. The evolution of the spring-in angle denoted as $\theta$ is shown in Fig. 5.11 (left). It is seen that the movement of the part is restricted by the die due to the employed mechanical contact formulation at the die-part interface. The spring-in angle starts increasing after die exit where the mechanical contact is deactivated. The magnitude of $\theta$ is calculated approximately as 0.45° at the end of the process. The spring-in angle is also measured from the pultruded L-shaped products and the predicted value for the spring-in angle ($\theta = 0.45^\circ$) is found to be very close to the measured range 0.5±0.05°. The residual spring-in angle is further calculated using the developed simulation tool for different pulling speed values and the results are depicted in Fig. 5.11 (right). It is seen that $\theta$ increases with the pulling speed since the glass transition point shifts towards the process end as the pulling rate increases. Therefore, a wider rubbery zone appears, thus providing larger deformations for the part.

The details of the predicted process induced residual stresses are presented in PAPER7. The through-thickness stress distributions ($S_{11}$, $S_{22}$ and $S_{33}$) at section BB are depicted Fig. 5.12. A uniform stress distribution is obtained at section BB as seen from Fig. 5.12 for which the boundary effects are less pronounced. It is seen that there is a discontinuity in the stress at the UD-CFM interface. The CFM layers are under compression ($S_{22} < 0$) whereas a tension zone ($S_{22} > 0$) exists for the UD layer in the $x_2$-direction. The other way around is the case for $S_{11}$, i.e. the UD roving layer is under compression and the CFM layers are under tension (Fig. 5.12).
Figure 5.10: The deformed contour plots of the L-shaped profile in the global $x_2$-direction (left) and $x_3$-direction (right) at the end of the process $x_1 = 6$ m. Scale factor for the deformed shape is 10.

Figure 5.11: The evolution of the spring-in angle $\theta$ (left) for a pulling rate of 600 mm/min. Predicted residual spring-in angles ($\theta$) for different pulling speed values.

The obtained stress pattern is verified using the CLT \[110\]. A composite laminate having CFM and UD layers similar to the pultruded product (5 mm thick) is considered in the ply stress calculation using the CLT. The process induced stresses are built up significantly after the part reaches the peak temperature during pultrusion \[47\]. Hence, a thermal load ($\Delta T = -100^\circ C$) is applied to the laminate which corresponds to the cooling of the part from peak temperature to ambient temperature. Knowing the elastic modulus and CTE of the layers in glassy state, the in-plane ply strains and stresses are calculated using the CLT. A schematic view of the laminate is shown in Fig. 5.13. Here, $x$-direction is defined as the fiber direction for the UD roving layer. The predicted in-plane stresses ($S_{xx}$ and $S_{yy}$) are also depicted in Fig. 5.13. The stress distributions are found to be similar as compared to the in-plane stresses ($S_{11}$ and $S_{22}$) in Fig. 5.12, e.g. the CFM layer is under compression for $S_{yy}$ and tension for $S_{xx}$. Nevertheless, the magnitudes deviate since the phase changes of the resin influence the stress evolutions during pultrusion. Note that the counter parts of $S_{11}$ and $S_{22}$ (Fig. 5.12) are $S_{xx}$ and $S_{yy}$ from the CLT analysis, respectively.
5.4 Internal Stress Evaluation at the Web-flange Junction of a pultruded I-beam

A numerical simulation of the pultrusion of a glass/epoxy I-beam profile is carried out in this thesis which has not been considered in the literature up to now. The pultruded I-beam is particularly used in the construction industry. The process induced residual stresses together with the temperature and degree of cure fields are predicted using the proposed thermo-chemical and mechanical process models (see Chapter 2).

The details of the process set-up as well as the cross section is seen in Fig. 5.14. Only a quarter model is used due to symmetry conditions. It is seen that the dimension of the processing I-beam is 200×100×10 mm (height×width×thickness). The length of the die is specified as 915 mm and the length of the profile at the post-die region is taken approximately as 6.4 m. For the sake of convenience and simplicity, a perfect thermal contact is assumed at the die-part interface in the 3D thermo-chemical model. The employed thermal properties and resin kinetics parameters can be found in \cite{42}.
The pulling speed is set to 200 mm/min in the pulling direction ($x_3$-direction). Similar thermal and mechanical boundary conditions are utilized as in PAPER\textsuperscript{2}.

The configuration of the UD and CFM layers are taken from an industrially pultruded I-beam reported in PAPER\textsuperscript{111}. The details of the layer orientations in the $x_1$-$x_2$ plane are depicted in Fig. 5.15. A local material orientation is employed for the CFM layer as in PAPER\textsuperscript{6}-PAPER\textsuperscript{7}. The thickness of the pultruded CFM layers is assumed to be 0.762 mm according to the data given in PAPER\textsuperscript{12}.

Figure 5.14: Schematic view of the quarter pultrusion model for the I-beam profile and the details of the cross section.

Figure 5.15: The configuration of the UD and CFM layers employed in the present FE model (left). The layer orientations reported in PAPER\textsuperscript{111} (right).
The temperature and the cure degree distributions are first obtained at steady state in the 3D thermo-chemical analysis and the results are depicted in Fig. 5.16 at the end of the process (at \( x_3 = 7.32 \) m). It is seen that the part cools down to 37-53°C and the degree of cure over the cross section is found to be higher than 0.91. The calculated temperature and cure degree profiles are used in the mechanical analysis in which the process induced residual stresses and distortions in the transverse directions are evaluated. The predicted residual stresses in the transverse directions (global coordinate system) are depicted in Fig. 5.17. Here, S11 and S22 are the normal transverse stresses in the \( x_1 \)- and \( x_2 \)-directions, respectively and S12 is the shear stress in the \( x_1-x_2 \) plane. It is seen that the UD roving layer is found to be under tension, whereas compression stresses prevail for the CFM layers in terms of normal stresses. A through-thickness stress variation is obtained for the shear stress S12 obtained at the web flange junction (WFJ) of the pultruded I-beam. This in combination with the normal stresses (S11 and S22) has an important effect of the mechanical respond and failure mechanism of the WFJ under service loading.

The mechanical behaviour of the WFJ of industrially pultruded I-beams was investigated in [112]. Pull-out tests were performed to evaluate the performance of the WFJ of the I-beam in terms of strength and stiffness. The failure behaviour of two samples are depicted in Fig. 5.18. It is seen that two different failure mechanisms were obtained under the same pull-out test. The process induced residual stresses (Fig. 5.17) obtained from the proposed thermo-chemical-mechanical model in this thesis might play an important role for the crack initiation at the WFJ seen in Fig. 5.18. Therefore, the residual stress has to be taken into account for the investigation of the internal stresses at the WFJs under service loading. It should also be noted that residual stresses have the potential to promote or demote the internal stress levels rise in the pultruded profile.

![Temperature and degree of cure distributions at the end of the process (at \( x_3=7.32 \) m).](image-url)
5.5 Integrated Modelling for the Pultrusion of a Wind Turbine Blade

An integrated modelling of a pultruded product particularly combining the manufacturing simulation with the subsequent service loading scenario is described in [49] for a NACA0018 blade profile which is the second part of the work in [45] (PAPER³). The residual stresses together with the final mechanical properties of the transversely isotropic pultruded product are subsequently transferred to the loading analysis in which a non-linear bending simulation of a NACA0018 profile is carried out. A unidirectional (UD) glass/epoxy composite is considered for the process simulation. A schematic view of the process set-up is shown in Fig. 5.19. The details of the cross section and the meshing are depicted in Fig. 5.20 for the part and the die.

The process induced residual stresses and distortions are predicted in a 2D quasi-static mechanical analysis in which the generalized plane strain elements are utilized in ABAQUS. The corresponding contour plots of $S_{11}$, $S_{22}$ and $S_{33}$ together with $S_{23}$ (transverse shear stress on the $x_2 - x_3$ plane) are shown in Fig. 5.21 at the end of the process.
In the subsequent loading scenario the transversely isotropic pultruded blade profile is assumed to be bent into Darrieus shape (i.e. arched-blades) taking the residual stresses into account. A schematic view of the bent-in place simulation of the blade is shown in Fig. 5.22. The bent shape is obtained by applying a displacement on one end of the profile having an initial total length of 3.7 m and keeping the other end fixed (i.e. hinged BC). In Darrieus type VAWTs, the length/diameter ratio of the rotor, i.e. $h/(2r)$ seen in Fig. 5.22, has an important effect on the aero-dynamical behaviour of the turbines. In the present study, $h/(2r) \approx 2.64$ is used by applying a displacement ($u$ in Fig. 5.22) value of approximately 0.3 m. The length ($h$) and the radius ($r$) of the rotor at the end of the simulation are approximately 3.4 m and 0.645 m, respectively. The obtained $h/(2r)$ is found to be very close to the one used in [4] as 2.7 for a 3-bladed VAWT.

A 3D non-linear structural static analysis is performed by using the quadratic solid elements in ABAQUS. The residual stresses together with the final mechanical properties of the profile predicted in the pultrusion process simulation are transferred to the bending simulation. The residual stresses are treated as a pre-stress condition utilizing the user defined routines in ABAQUS before the bending simulation. Subsequently, the loading scenario depicted in Fig. 5.22 is carried out and the internal stress levels of the bent profile are evaluated.
Figure 5.21: Undeformed contour plots of the longitudinal normal stress ($S_{11}$), the transverse normal stresses ($S_{22}$ and $S_{33}$) and the transverse shear stress ($S_{23}$) at the end of the pultrusion process. Note that the legend of the plots is not same.

Figure 5.22: A schematic view of the subsequent loading scenario (i.e. bending of the blades).

The maximum normal stresses are expected to be built up at the center of the blade profile according to the loading scenario (section “M” depicted in Fig. 5.22) in the longitudinal direction. Hence, the evaluation of the internal stresses at section M are analysed in detail. The contour plots of the normal stresses at section M at the end of the bending simulation are shown in Fig. 5.23 with and without taking the residual stresses into account. It is seen that the effect of the residual stresses are more dominant for the transverse directions (i.e. for $S_{22}$ and $S_{33}$) as compared with the longitudinal component ($S_{11}$) which is obviously more critical for this type of loading scenario. The magnitude of $S_{11}$ is found to be much larger than $S_{22}$ and $S_{33}$ as expected. After obtaining the equilibrium state, it is found that the maximum compression stress for $S_{11}$ is increased approximately from 216 MPa to 220 MPa with the residual stresses. However, the $S_{11}$ value for the maximum tensile stress is decreased from approximately 213 MPa to 210 MPa. The stress levels for $S_{22}$ and $S_{33}$ are found to be relatively small as compared to $S_{11}$. Nevertheless, the residual stresses have more significant effect on the transverse
stresses ($S_{22}$ and $S_{33}$) for this specific loading scenario as it is seen from Fig. 5.23. The residual stresses have the potential to promote or demote the internal stress levels arise in the pultruded profile as aforementioned. Moreover, they would have more important influence for the upscaled profiles or specific applications such as the pull out tests on pultruded L-shaped products (see Fig. 5.17 and Fig. 5.18) in which the residual stresses in the transverse directions are crucial.

Figure 5.23: Contour plots showing the stress distribution with/without residual stresses for section M at the end of the bending simulation. Note that the legend of the plots is not same.

5.6 Modelling of the Pulling Force in Pultrusion

Pultrusion process simulations are performed for a unidirectional (UD) graphite/epoxy composite rod in [74] Paper11. The temperature and cure degree profiles are calculated using developed 3D thermo-chemical models. The viscosity profile is subsequently predicted and used for the calculation of the viscous pull force. The pressure rise at the die-part interface is calculated in the 2D thermo-mechanical model in which a mechanical
contact behaviour is defined at the die-part interface.

A schematic view of the simulation domain is depicted in Fig. 5.24. The radius of the processing rod is 4.75 mm, while the length of the die is 915 mm, which are adopted from the numerical and experimental analysis provided in [34]. It should be noted that, in the performed simulations, the temperature distribution on the internal die surface is used to provide the required closure of the above described thermochemical problem. The inlet temperature is assumed to be equal to the resin bath temperature (38°C) while the matrix material is assumed to be totally uncured ($\alpha = 0$) at the same cross section. Only a quarter of the 3D model has been considered due to the symmetry and in order to reduce the computational effort.

According to the calculated viscosity and pressure profiles in the thermo-chemical analysis the total pulling force together with its components (see Fig. 2.8) is predicted and the results are depicted in Fig. 5.25. The friction coefficient $\mu$ in Eq. 2.58 has been assumed to be 0.25. Numerical outcomes show that, the viscous force represents the principal amount of the total resistance, being compaction force $F_{\text{bulk}} = 4.9$ N, viscous force $F_{\text{vis}} = 313.7$ N, and frictional force $F_{\text{fric}} = 184.1$ N. Relatively smaller frictional force (112.9 N) is predicted by the FE mechanical model as compared with the semi-analytical model (SAM), due to the lower contact pressure profile obtained using FE (see [74] PAPER11 for details). The key role played by the viscous drag with respect to the frictional force can be related to the reduced die length affected by the frictional phenomena and to the delayed development of the resin (and the composite) modulus. The contribution due to the material compaction is found to be not significant as compared to other amounts, being less than 1% of the total load.
Figure 5.25: Contribution to the pulling force as provided by the impregnation, viscous, and frictional models (SAM), assuming chemical shrinkage and initial viscosity to be 4% and 1.05 Pas, respectively (\textsuperscript{[74] PAPER\textsuperscript{11}}).
Chapter 6

Conclusions and Future Work

In this chapter, the main concluding remarks extracted from the current work are summarized. This is followed by recommendations for the future work in terms of the numerical modelling of the pultrusion process.

6.1 Conclusions

This thesis focuses on the thermo-chemical and thermo-mechanical modelling of the pultrusion process for industrial products such as wind turbine blades and structural profiles. In particular, the main challenges in pultrusion such as process induced residual stresses and shape distortions together with the prediction of the thermal and cure history are addressed using the developed numerical process simulation tools. A detailed literature review was conducted providing a summary of the major theoretical contributions in the field of the pultrusion manufacturing process. Numerical modelling of pultrusion is a challenging task due to the multi-physics involved in the process. Therefore it is necessary to develop process models in order to avoid the expensive trial-and-error approaches for designing new products and optimizing the process conditions.

The chemo-rheology of an industrial “orthophthalic” polyester system specifically prepared for a pultrusion process was characterized in this thesis. The curing behaviour was first obtained using differential scanning calorimetry (DSC). Isothermal and dynamic scans were performed to develop a cure kinetics model which accurately predicted the cure rate evolutions. The resin viscosity and the gelation point were subsequently obtained from rheological experiments using a rheometer in “plate-plate” mode. Based on this, a resin viscosity model as a function of temperature and degree of cure was developed and predicted the measured viscosity correctly. A temperature- and cure-dependent elastic modulus was determined using a dynamic mechanical analyzer (DMA) in tension mode. A modified cure hardening instantaneous linear elastic (CHILE) model was developed for the resin elastic modulus evolution during curing and a least squares non-linear regression analysis was performed. The predicted best fit results were found to agree quite well with the measured data. The modified CHILE approach, which is a valid pseudo-viscoelastic approximation of the linear viscoelasticity [113], captured the variations in the elastic modulus of the resin during curing.

Novel applications were carried out in terms of efficient thermo-chemical analysis of the pultrusion. In particular, the processing characteristics at the heating die and the post
die regions were studied intensively. The effects of the thermal contact resistance (TCR) at the die-part interface of a composite rod were investigated. It was found that the use of a variable TCR is more convenient than the use of a constant TCR for the simulation of the process since a variable TCR can emulate the thermal effect of the chemical shrinkage on the curing. The 3D numerical modelling strategies of a thermosetting pultrusion process were investigated considering both transient and steady state approaches. A numerical simulation tool embracing the blade manufacturing process (NACA0018 profile) was developed to tailor the effects of the heater configuration and pulling speed on the pultruded blade profile.

State-of-the-art process models were developed by the author to predict the process induced stresses and shape distortions together with the temperature and degree of cure fields. A 3D thermo-chemical model was coupled with 2D and 3D quasi-static mechanical models using the FEM. In the mechanical model, the composite part was assumed to advance through the pulling direction meanwhile tracking the corresponding temperature and degree of cure profiles already calculated in the thermo-chemical analysis (see Fig. 2.5 and Fig. 2.6). Various case studies were carried out including industrial pultruded products using the proposed thermo-chemical-mechanical numerical simulation tool. As aforementioned, modelling the pultrusion process containing both UD and CFM layers has not been considered in the literature up to now. A numerical simulation tool embracing the residual stresses and shape distortions in pultrusion of an industrial rectangular hollow profile and L-shaped product was hence developed by the author. The warpage in the rectangular hollow profile and the spring-in formation in the L-shaped product were predicted correctly as compared with the real pultruded profiles. In addition, the internal stresses at the web flange junction of a pultruded I-beam were investigated using the devised numerical tool. An integrated modelling study was carried out in which the predicted residual stresses and the final mechanical properties are transferred to the preliminary subsequent service loading scenario for a pultruded NACA0018 blade profile. It is found that the residual stresses have the potential to promote or demote the internal stress levels arise in the pultruded profile and they would have more important influence for the upscaled profiles or specific applications such as the pull out tests on pultruded L-shaped products (see Fig. 5.17 and Fig. 5.18).

The pulling force model was developed to predict the component of the total resistance forces. The viscous drag is found to be the main contribution to the overall pulling force, while the contribution due to material compaction at the inlet was found to be negligible.

Using the process models, novel optimization studies were carried out in order to improve the production rate and the quality. The mixed integer genetic algorithm (MIGA) was developed to optimize the process by finding the optimum heater configuration. Moreover, a multi-objective problem (MOP) was implemented to the thermo-chemical analysis to minimize the energy consumption and maximize the productivity of the process simultaneously. Probabilistic analyses were also performed to investigate the effect of uncertainties in the process parameters on the product quality by using Monte Carlo simulations (MSC), response surface method (RSM) and first order reliability method (FORM).

The proposed thermo-chemical-mechanical process models are found to be efficient
and computationally fast for the calculation of the residual stresses and distortions together with the temperature and cure distributions. They have a great potential for the future investigation of the residual stresses and shape distortions for more complex pultruded profiles. More specifically, the distortions might be important for rigid polymer structures such as window frames and fencing panels due to their desired high geometrical precision. The residual stresses would have an important effect on the mechanical behaviour of the pultruded structural profiles such as wind turbine blades and I-beam (especially at the web-flange junction (WFJ)) since residual stresses have the potential to promote or demote the internal stress levels arise in the pultruded products. Therefore, the residual stresses and shape distortions arise in the pultrusion process have to be analysed carefully.

6.2 Future Work

In addition to the research studies presented in this thesis, there exists issues needed to be pinpointed as future challenges for the pultrusion process. The current mechanical process models can be further improved by imposing damage formulations which would provide a better idea in terms of delamination resistance/strength and premature cracking during processing. Moreover, the bonding quality of the UD roving and CFM layers can be investigated by taking the possible fracture of the matrix material at the UD-CFM interface into account. The interlaminar and interfacial failures occurring in a particular layer can be addressed by modelling the corresponding failure mechanisms during processing.

The impregnation models can be coupled with the thermo-chemical and thermo-mechanical models in order to tailor the effects of the initial pressure condition at the die inlet on the residual stress and pulling force. It is a fact that there is a direct relation between the residual stresses, resistance forces inside the die and the initial pressure condition.

A more advanced industrial pultruded products containing woven reinforcements with an angle e.g. ±45 or ±90 together with the UD roving and CFM layers can be modelled numerically and analysed during processing. This would bring technical solutions for the practical problems existing in advanced pultruded products. Moreover, the process can be optimized utilizing probabilistic modelling and optimization algorithms simultaneously. This would provide a robust way to obtain the optimum process conditions taking the probability density functions into account.
Bibliography


Appendixes
A PAPER

“The Effect of Thermal Contact Resistance on the Thermosetting Pultrusion Process”

Ismet Baran, Cem C. Tutum, Jesper H. Hattel

Composites Part B: Engineering 2013; 45:995-1000.
The effect of thermal contact resistance on the thermosetting pultrusion process

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Abstract

In the present study the control volume based finite difference (CV/FD) method is utilized to perform thermo-chemical simulation of the pultrusion process of a composite rod. Preliminary, the model is applied for a simple setup without die and heaters and the results match well with those obtained experimentally in the literature. In order to study the effects of the thermal contact resistance (TCR), which can also be expressed by the heat transfer coefficient (HTC), on the pultrusion process, a cylindrical die block and heaters are added to the original problem domain. The significance of using the TCR in the numerical model is investigated by comparing constant and variable TCR (i.e. position dependent) at the interface. Results show that the use of a variable TCR is more reliable than the use of a constant TCR for simulation of the process.

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1. Introduction

The pultrusion process is one of the most cost efficient methods for production of composite materials having constant cross-sectional profiles such as beams, stiffeners, and tubes. This process has been widely used for manufacturing highly strengthened and continuous composite structures, e.g. the application of pultruded rods is significantly increasing for reinforcements of concrete elements in the construction industry. The fascinating advantage of the process is the high productivity and the low cost. Since it is a continuous process, there is little waste material arising at the end of the production. A schematic view of the pultrusion process can be seen in Fig. 1. Fibers and resin matrix are continuously pulled through a heating die by a pulling mechanism. The composite part gradually cures and solidifies inside the heating die and finally a saw mechanism cuts the product into the desired length.

1.1. Related work

In recent years, several experimental studies and numerical simulations for the pultrusion process have been performed in order to investigate the effects of the process parameters on the quality of the manufactured part and the productivity of the process. Since the present work first of all is based on the thermo-chemical modeling of the pultrusion process, studies in this field are reviewed in detail.

Thermo-chemical models of the pultrusion process have been developed and used for the simulation of the process since the 1980s [1–17]. Transient and steady state simulations have been applied by using numerical techniques such as the finite difference method (FDM) and the finite element method (FEM) with control volume (CV) method as well as the nodal control volume (NCV) method. A basic one dimensional (1D) numerical heat transfer model of the thermosetting pultrusion process was developed in [1]. In [2,3], a 2D model in cylindrical coordinates was developed by using the CV/FD method for fiberglass/epoxy and graphite/epoxy composites. The pultrusion of various irregular Cartesian geometries such as U, S, rectangular and hollow-square shaped cross-sections were analyzed and moreover the temperature and the cure degree characteristics of the graphite/epoxy composite were discussed by Chachad et al. [4–6]. 3D FE/NCV techniques were utilized, with the use of the general purpose finite element software LUSAS, for transient thermo-chemical model of thermoplastic composites by Ahmed et al. [10] and Joshi et al. [11]. The effects of the resin shrinkage and the temperature-dependent material properties on temperature and cure degree distributions have been discussed in [12–14]. In [14], the thermal pultrusion simulation of multi-materials i.e. a foam/glass fiber reinforced polymer (GFRP) sandwich panel has been performed using a multi-heater environment. In [15], two different three dimensional pultrusion simulations of a C-shaped composite have been performed by using the Alternating Direction Implicit method (ADI)-FDM and the FEM. It was concluded that similar results were obtained from both methods. According to the results obtained by using the different numerical methods mentioned above, similar temperature and cure degree behaviors have been found, e.g. the
centerline temperature of the composite is initially lagging behind the die temperature, however during the curing the composite temperature exceeds the die temperature due to the internal heat generation of the resin. As a means of supporting the numerical thermo-chemical modeling of the pultrusion process, experimental studies of various composite profiles have been carried out in [19–27].

1.2. The effect of TCR

In pultrusion of thermosetting resin composites, the chemical exothermic reaction of the resin starts when the composite reaches the reaction initiation temperature at which the gelation of the resin is also observed. Because of the internal heat generation, after some point in time the direction of the heat flux is inverted such that the heat flow is transmitted from the composite to the die. During the curing of the composite, the shrinkage, which causes the separation of the composite profile from the die, is observed at the die-part interface where the degree of cure is higher than inside the composite. At this instant, the volumetric shrinkage has a higher contribution to the deformation of the cured resin than the thermal expansion of the resin since the temperature difference is getting smaller while the process tends to reach the steady state. Hence an air gap is initiated at the die-part interface which affects directly the heat transfer, the total pulling force and the pressure inside the die. A schematic representation of the phase change of a thermosetting composite is seen in Fig. 1.

In [1,16] the effects of the resin shrinkage and the HTC on the temperature distribution were mentioned but were not taken into account for the simulation of the pultrusion process. As a consequence of this, in [16], the numerical temperature distribution was over estimated as compared with the experimental data measured especially near the last section of the die where the chemical shrinkage of the resin plays a vital role. On the other hand it was stated in [1] that the HTC at the die-part interface changes significantly from liquid to solid phase of the composite part.

In order to compensate for the decrease in the amount of heat flux at the die-part interface during curing of the resin which consequently shrinks and separates away from the die, an empirical relation adjusting the Nusselt number (Nu) for the convective boundary condition at the interface was used in [29]. The effect of the chemical shrinkage on the pulling force was investigated in [30] such that the friction force at the interface occurs only after the gel phase and the friction vanishes after the profile shrinks and separates away from the wall. Similarly it was observed experimentally and numerically in [30] that the resin pressure increased rapidly at the die inlet because of the back flow of the liquid resin, and subsequently the pressure slightly increased due to the thermal expansion of the resin. When the resin started solidifying, a subsequent decrease in the pressure was observed due to the shrinkage of the resin during curing.

In order to improve the efficiency of the pultrusion process, there is a need for an accurate and precise numerical model. In this study, the pultrusion of a composite rod (initially without the die block) taken from the literature [20] is simulated as a validation case by using the CV/FD method for the transient and the steady state solutions of the energy equation. After validating the model, a cylindrical die block with heaters is included. In this new configuration, the TCR at the die-composite interface is taken into account for the process simulation. The aim is to investigate the significance of the TCR for the pultrusion process, while having the same centerline temperature and cure degree profile as found in the validation case [20]. The MATLAB computing environment is used for both the simulations and the temperature fitting procedure. The proposed model is generic and can easily be adapted to other pultrusion process applications such as profiles for bridges, window frames, reinforcement profiles for turbine blades, gratings etc.

2. Governing equations

2.1. Energy equation

The transient heat transfer equations in the cylindrical coordinate system for the composite rod (Eq. (1)) and the die (Eq. (2)) are derived from the conservation of energy principle which states that the rate of the energy stored in a control volume is equal to the sum of the net rate of energy transferring into the control volume and the internal heat generation (for the composite part). The subscripts $c$ and $d$ correspond to the composite and the die, respectively. In general, the material properties of the resin change during curing and the material properties of the fiber remain almost same compared to resin. Thus, the resin material properties depend on the temperature and the degree of cure. On the other hand, there is a considerable number of well-known studies in this specific field [i.e. pultrusion [3–6,11–20]] in which the material properties are assumed to be constant and the simulation results matched well with corresponding experiments. Therefore, in the present study a similar methodology of assuming constant material properties has been followed.

The steady state energy equations can be obtained by neglecting the time dependent terms i.e. $\partial T/\partial t$. It should be noted that there is no convective ($u\partial T/\partial z$) and heat source ($q$) term in the energy equation for the die (Eq. (2)).

\[
\rho_c\mathcal{C}_p\frac{\partial T}{\partial r} = \frac{k_c}{\mathcal{C}_p} \frac{\partial}{\partial r}\left(r \frac{\partial T}{\partial r}\right) + q \tag{1}
\]

\[
\rho_d\mathcal{C}_p\frac{\partial T}{\partial r} = \frac{k_d}{\mathcal{C}_p} \frac{\partial}{\partial r}\left(r \frac{\partial T}{\partial r}\right) \tag{2}
\]

where $T$ is the temperature, $u$ is the pulling speed, $t$ is the time, $\rho$ is the density, $\mathcal{C}_p$ is the specific heat, $k_c$ and $k_d$ are the thermal conductivities in the axial direction ($z$) and in the radial direction ($r$) respectively. Lumped material properties are used for the composite.

The internal heat generation ($q$) [W/m$^3$] due to the exothermic reaction of the epoxy resin is expressed as

\[
q = (1 - V_f) \rho_f Q \tag{3}
\]

where $V_f$ is the fiber volume ratio and $Q$ is the specific heat generation rate [W/kg] due to the resin exothermic cure reaction.
2.2. Resin Kinetics

Resin kinetics is an important phenomenon which is related to the exothermic chemical reaction of the resin inside the die. This directly affects the curing of the resin. Several kinetic models can be found in the literature to describe the cure process; generally kinetic models relate the rate of resin reaction (\(R_r\)) to the temperature (\(T\)) and the degree of cure (\(x\)). The degree of cure can be written as the ratio of the amount of heat generated (i.e. \(H(t)\)) as a function of time) during curing, to the total heat of reaction \(H_0\):

\[
x = \frac{H(t)}{H_0}
\]

(4)

\(R_r\) can be written as an Arrhenius equation [18],

\[
R_r(x) = \frac{dx}{dt} = \frac{1}{H_0} \frac{dH(t)}{dt} = K_o \exp \left( \frac{E}{RT} \right) (1 - x)^n
\]

(5)

where \(K_o\) is the pre-exponential constant, \(E\) is the activation energy, \(R\) is the universal gas constant and \(n\) is the order of reaction (kinetic exponent). \(K_o, E, H_0\) and \(n\) can be experimentally evaluated using DSC [20]. As a result, \(Q (Eq. (3))\) can be expressed as

\[
Q = \frac{dH(t)}{dt} = H_0 R_r(x)
\]

(6)

By using the chain rule, the rate of cure degree can be expressed as,

\[
\frac{d\alpha}{dt} = \frac{\partial \alpha}{\partial x} \frac{dx}{dt} + \frac{\partial \alpha}{\partial T} \frac{dT}{dt}
\]

(7)

\[
\frac{\partial \alpha}{\partial T} = R_r(x) - \frac{\partial \alpha}{\partial T}
\]

(8)

where it is the expression in Eq. (8) which is used in the numerical model.

3. Numerical implementation

Transient and steady state numerical schemes are used for the validation case in which only the composite part without the die block is simulated. On the other hand, only the steady state numerical scheme is utilized for the simulation of the new configuration involving the composite part with the die block including the TCR at the interface. The CV/FD method is used for the discretization of the energy equations in which the resin kinetics is coupled with the heat conduction/convection terms. The internal heat generation inside the composite is highly non-linear such that it depends on both the degree of cure and the temperature which are the parameters in the Arrhenius equation (Eq. (5)). Therefore, in order to develop a stable and fast numerical procedure, the resin kinetics equations are decoupled from the energy equation for the composite part and hence the degree of cure is updated explicitly for each control volume using Eq. (8) in its discretized form.

The transient time integration schemes for the energy equation of the composite and the degree of cure are given in Eqs. (9) and Eq. (10) respectively where \(n\) is the time step index. The alternating direction implicit (ADI) method is utilized for the transient simulation of the pultrusion process. In the first half of the time step \((n+1/2)\) the temperature distribution is calculated implicitly in the axial direction and explicitly in the radial direction (first part of Eq. (9)). For the second half of the time step \((n+1/2-n+1)\) the directions are reversed (second part of Eq. (9)). In Eq. (10), it is seen that the time integration of the cure degree relation is handled explicitly over the time step, \(n-n+1\), as mentioned before.

\[
\rho \cdot C_p \left( \frac{\partial x}{\partial t} \right)_{n+1/2} + \left[ u \frac{\partial x}{\partial z} \right]_{n+1/2} = \left[ k_x \frac{\partial^2 x}{\partial z^2} \right]_{n} + \left[ k_z \frac{\partial x}{\partial z} \right]_{n} - \frac{H(t)}{H_0}
\]

(9)

\[
\rho \cdot C_p \left( \frac{\partial x}{\partial t} \right)_{n+1/2} + \left[ u \frac{\partial x}{\partial z} \right]_{n+1/2} = \left[ k_x \frac{\partial^2 x}{\partial z^2} \right]_{n+1/2} + \left[ k_z \frac{\partial x}{\partial z} \right]_{n+1/2} + \frac{H(t)}{H_0}
\]

(10)

Similarly steady state numerical schemes are obtained by discarding the time dependent terms in Eqs. (9) and (10). In the CV/FD numerical scheme the total thermal resistances (K/W) being the sum of the single resistances coupled in series between the two adjacent control volumes are used [31]. In order to overcome the oscillatory behavior in the numerical implementation the upwind scheme is used for the convective term \((u \cdot T/\partial x)\) in the energy equation and for the space discretization of the cure degree \((u \cdot \partial x/\partial x)\) in the resin kinetics equation. In the solution of the numerical schemes the tri-diagonal matrix algorithm (TDMA) is used.

4. Validation case

Pultrusion of a composite rod without the die block is simulated for the purpose of validating the model. The transient and the steady state solutions of the temperature distribution and the cure degree profiles are obtained with a given die wall (die-part interface) temperature profile based on the experimental work of Valliappan et al. [20]. Since the die wall temperature profile is constant throughout the periphery (cylindrical surface) of the composite rod cross-section, the axisymmetric numerical model is used. The graphite fiber reinforcement (Hercules AS4–12 K) and epoxy resin (SHELL EPON9420/9470/537) system are used for the composite. The thermo-physical properties of the composite (composed of graphite and resin) as well as the die block are given in Table 1. The resin kinetic parameters are [20]: \(K_o = 191400 \text{ (1/s)}\), \(E = 60500 \text{ (J/mol)}\) and \(n = 1.69\).

4.1. Problem description

The model geometry and the boundary conditions are shown in Fig. 2. At the die inlet the degree of cure is assumed to be equal to zero and the temperature of the composite (\(T_{app}\)) is taken as the resin bath temperature (\(-38\,^\circ\text{C}\)). The initial temperature of the die and the composite are taken as \(27\,^\circ\text{C}\) (ambient temperature) for the transient simulation. The initial cure degree of the composite is taken as 0 for both transient and steady state simulations. The convergence limits of the temperature and the degree of cure for both solutions are set to 0.001 °C and 0.0001, respectively.

4.2. Results and discussion

The temperature and the cure degree profiles are obtained with a pulling speed of 30 cm/min. 4 (radial) \times 150 (axial) control volumes having 6 (radial) \times 152 (axial) nodes are used for the thermo-chemical simulation of the composite rod. The final centerline temperature and the cure degree profiles are found to be almost similar for different Peclet numbers, \(Pe (2, 23, 92)\) used in the simulations. It should be noted that \(Pe\) indicates the strength between the convective and the conductive terms in the flow simulation. In theory \(Pe\) should be less than 2 in order to get stable results when using a central finite difference discretization [32]. However, since the upwind scheme is utilized in the numerical implementation of the pultrusion, no oscillation has occurred.

<table>
<thead>
<tr>
<th>Material</th>
<th>(\rho) (kg/m³)</th>
<th>(C_p) (J/kg K)</th>
<th>(k_x) (W/m K)</th>
<th>(k_z) (W/m K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Epoxy resin</td>
<td>1260</td>
<td>1255</td>
<td>0.2</td>
<td>0.2</td>
</tr>
<tr>
<td>Graphite</td>
<td>1790</td>
<td>712</td>
<td>66.0</td>
<td>11.6</td>
</tr>
<tr>
<td>Lumped ((V_r = 62.2%))</td>
<td>1589.66</td>
<td>874.69</td>
<td>0.6628</td>
<td>0.6416</td>
</tr>
<tr>
<td>Steel die</td>
<td>7833</td>
<td>460</td>
<td>40</td>
<td>40</td>
</tr>
</tbody>
</table>
during the simulation. A time step of 0.1 s is used for the transient simulation. The transient and the steady state centerline temperature and the cure degree distributions are seen in Fig. 3 for $P_e = 92$. It is seen that both results are matched well with those in [20].

This shows that the developed numerical scheme is stable and converged to a reliable solution. It is seen in Fig. 3 that after around 380 mm from the die inlet, the centerline temperature of the composite becomes higher than the die wall temperature due to the internal heat generation and the peak temperature of the composite is reached around 205 °C. The centerline exit cure degree is calculated as 0.84.

5. Effect of TCR

5.1. Problem description

A cylindrical die block is added to the previous pultrusion domain investigated in the validation case together with three cylindrical heating pads mounted on top of it. A schematic view of the die, the composite and the heaters is seen in Fig. 4. The set temperatures of the heaters are given in [20] from left to right as 171–188–188 °C, but the location and the length of the heaters are not given. The boundary conditions for the composite part are assumed to be the same as in the validation case except at the die-part interface where the non-perfect thermal contact between the die and the composite is now modeled by a thermal contact resistance (TCR) which is either constant or taken as a function of the axial distance at the interface boundary. For the die block all the exterior surfaces except those on which the heating pads are located are exposed to the ambient temperature (27 °C) with a convective heat transfer coefficient of 10 W/m² K. Since it is not given in [20], cooling channels located at the initial die section are not considered in this model. 20 (radial) × 150 (axial) control volumes corresponding to 22 (radial) × 152 (axial) nodes are used for the space discretization of the die block.

In order to obtain the same centerline temperature profile of the composite within this new pultrusion simulation domain, a curve fitting procedure (i.e. inverse modeling) is performed using the data composed of 15 centerline temperature values measured from the validation case by equally spaced thermocouples (every 60 mm through the axial direction). The TCR values (design variables in the curve-fitting procedure) are predicted by minimizing the difference between the measured (the validation case) and the calculated (the new configuration) centerline temperatures, i.e. $\sum (T_{meas} - T_{cal})^2$, for certain die radii. For this purpose the constrained minimization function "fmincon" in MATLAB [33] which finds the minimum of a multivariable problem is applied. The temperature curve fitting procedure is repeated with five different die radii ($r_2$) selected as 10, 25, 50, 75 and 100 mm, there by considering possible die designs for the composite rod in the validation case. Two different optimization case studies are performed:

(a) Constant TCR (Case-1) In this case study only a single TCR value (one design variable) through the axial direction is optimized to minimize the error, $\sum (T_{meas} - T_{cal})^2$. A schematic view of the single TCR case can be seen in Fig. 4. The single TCR along the axial direction at the die-part interface is indicated with the red color.

(b) Variable TCR (Case-2) In this problem 9 equally spaced (each of ~100 mm) TCR regions are defined along the interface. The configuration of the variable TCR regions can be seen in Fig. 4. The discrete TCR regions are indicated with the blue color.

5.2. Results and discussions

As aforementioned, the optimum TCR values are found by using the fmincon function for the specific (predefined) die radii as shown in Fig. 5 (left) and Fig. 5 (right) for Case-1 and Case-2, respectively. According to Fig. 5 (left), as the die radius increases the constant TCR value used in Case-1 also increases which shows that the total amount of heat input (total area of the heaters) is increased. This can be explained since the steady state numerical model is used such that there is no time limitation for the temperature of the heaters to reach the die-part interface. In other words, in order to obtain the same centerline temperature for a transient case as the die radius increases, more time would be required however this is not the case for the steady state analysis. In addition to this, the temperature gradient in radial direction at the die-part interface is only slightly affected as the die radius increases because of the increase in the conductive area between the die-part interface and the heaters. However, this would not be the case if a Cartesian two dimensional pultrusion domain was used instead of the current cylindrical one since the total area of the heaters...
as well as the conductive area then would be constant as the die radius increases. It is seen from the shape of the curves in Fig. 5 (right) that the behavior or characteristics of the variable (set of discrete values) TCR regions defined in Case-2 is similar for all the die radii used. There is a decreasing trend in the TCR values for the initial regions (TCR regions /C24 1–3 in Fig. 5 (right)); however for the last region this trend is reversed. Although there is no shrinkage at the initial regions and the resin is in liquid state, large TCR values are found for the first three regions (/C24 0–300 mm.). The reason for this observation is that the thermal effect of the cooling channels inside the cylindrical die block in the validation case has to be compensated in order not to obtain any degradation for the composite part. Therefore it can be considered that these TCR regions, i.e. TCR regions 1–3, to some extent undertake the role of

Fig. 4. Schematic representation of the pultrusion domain of the composite rod including the cylindrical die block and the heaters with TCR regions.

Fig. 5. Optimum constant TCR values found in Case-1 (left) and optimum variable TCR values found in Case-2 (right) at the interface for different die radii.

Fig. 6. The centerline temperature (left) and the cure degree (right) distributions obtained by using single and variable TCR at the interface and the measured centerline temperature and cure degree profiles obtained from the validation case (rd = 10 mm).
the cooling channels in the simulation. This also shows that the first heater may not be needed depending on the inlet temperature of the composite \(T_{in}\). The effects of the curing and therefore the chemical shrinkage of the resin can be related to the last TCR region, i.e. 9th region (see Fig. 5 (right)), since higher TCR values are obtained at the last region. It must be noted that the TCR values are strongly dependent on the pultrusion domain, i.e. the location and the temperature of the heaters, the die geometry, the pressure inside the die, inlet temperature etc. It is seen in Fig. 5 (right) that the TCR value at the last region decreases as the die radius increases. This shows that the temperature characteristics of the last region differ from the entrance region. This can be explained since the peak temperature of the composite (being higher than the heater temperature) becomes smaller as the die radius increases. This should be combined with the effect of the internal heat generation which plays an important role for the temperature in the mid region as well as the exit region.

The optimum TCR values for both cases, i.e. the minimum error \(\left(\sum_{j=1}^{n} (T_{cal_j} - T_{exp_j})^2\right)^{1/2}\), are found to be 6957.5, 7979.8, 8250.7, 297.5 and 8275.9 in Case-1 and 6.7, 5.2, 3.7, 7.4 and 19.4 in Case-2 for the die radii of 10, 25, 50 and 100 mm, respectively. The minimum error for Case-1 in which a single TCR is used is significantly higher than the error for Case-2 with respect to all die radii. This shows that the use of a single TCR is not suitable for the thermo-chemical simulation of the pultrusion process as compared with the use of variable TCR at the die-part interface. For instance the centerline temperature and the cure degree profiles of the composite rod for a die radius of 10 mm are seen in Fig. 6. The temperature and the cure degree profiles obtained with the use of variable TCRs are almost the same as those given in the validation case. However the results obtained by using a single TCR deviate considerably with respect to the centerline temperature and the cure profiles of the composite rod.

6. Conclusion

In the present study a numerical model for the simulation of pultrusion of a composite rod is presented. In the validation case, the utilized numerical model was found stable and accurate as compared with the results from the literature. In order to obtain the same centerline temperature for the new configuration (i.e. including the die block and the heaters) the TCR was taken into account for predicting the same temperature profiles. The application of variable TCRs gave much better results than the application of a single TCR at the interface. In addition to that the TCR takes the role of the shrinkage and also the cooling channels which are not included in the numerical model. According to the TCR values found at the initial regions (\(\sim 1\)–3) it can be concluded that the first heater may not be needed in the numerical simulation depending on the inlet temperature of the composite. It is also concluded that the TCR has an important effect on the deterministic thermo-chemical simulation of the thermostetting pultrusion process. Hence, a probabilistic modeling approach, in which the variations in the process parameters or the working conditions inherently involved in the manufacturing process are taken into account, would be very useful to investigate the effect of scatter in the TCR on the thermal field, and hence the degree of cure distribution in the part. Besides this, the product size would have also an effect on the variation of the TCR at the interface, primarily because the potential air gap between the composite and the die inherently is affected by the radius of the composite part.

References

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“Thermo-chemical Modelling Strategies for the Pultrusion Process”

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Thermo-Chemical Modelling Strategies for the Pultrusion Process

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Received: 11 April 2013 / Accepted: 17 June 2013 / Published online: 4 July 2013
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Abstract In the present study, three dimensional (3D) numerical modeling strategies of a thermosetting pultrusion process are investigated considering both transient and steady state approaches. For the transient solution, an unconditionally stable alternating direction implicit Douglas-Gunn (ADI-DG) scheme is implemented as a first contribution of its kind in this specific field of application. The corresponding results are compared with the results obtained from the transient fully implicit scheme, the straightforward extension of the 2D ADI and the steady state approach. The implementation of the proposed approach is described in detail. The calculated temperature and cure degree profiles at steady state are found to agree well with results obtained from similar analyses in the literature. Detailed case studies are carried out investigating the computational accuracy and the efficiency of the 3D ADI-DG solver. It is found that the steady state approach is much faster than the transient approach in terms of the computational time and the number of iteration loops to obtain converged results for reaching the steady state. Hence, it is highly suitable for automatic process optimization which often involves many design evaluations. On the other hand sometimes the transient regime may be of interest and here the proposed ADI-DG method shows to be considerably faster than the transient fully implicit method which is generally used by the general purpose commercial finite element solvers. Finally, using the proposed steady-state approach, a design of experiments is carried out for the curing characteristic of the product based on pulling speed and part thickness.

Keywords Numerical modeling · ADI · Finite difference method · Pultrusion · Curing

1 Introduction

Pultrusion is one of the most cost efficient manufacturing processes for producing continuous lengths of fiber reinforced composite shapes with a constant cross section. The process is basically the same although the pultrusion machines vary in their design. The fibers and mats are first pulled into a resin bath or resin injection chamber in which the reinforcements are saturated or wetted out. The wet out reinforcements then enter the heating or forming die in which an exothermic reaction
takes place inside the thermosetting resin. This reaction generates an internal heat and the resin cures. The cured profile is advanced via a pulling system to the cut-off saw where it will be cut to its final length. A schematic view of a pultrusion process is shown in Fig. 1.

In recent years, several numerical modeling studies of pultrusion have been performed in the literature in order to investigate the dynamics of the process, e.g. the temperature and cure degree developments inside the heating die. For this purpose, well known numerical methods such as the finite difference method (FDM) or the finite element method (FEM) have been utilized.

A one dimensional (1D) heat transfer model of the pultrusion process for a thermosetting resin composite was developed in [2, 3] employing the FEM. In [4, 5] a mathematical model for heat transfer and cure inside the heating die was developed utilizing the FDM in which the time stepping was carried out implicitly using the Crank-Nicolson method. In these studies, the assumption of no axial conduction and negligible bulk flow simplified the 2D pultrusion model into a 1D transient heat transfer model. A comprehensive 2D axisymmetric pultrusion model of a graphite/epoxy composite rod in cylindrical coordinates was developed in which a control volume based finite difference method (CV/FD) was implemented in [6–8]. In [9, 10], the pultrusion process of composite profiles was simulated in 2D in which the solution of the thermo-chemical equations was carried out using the alternating direction implicit (ADI) method. In 2D, the ADI method is an unconditionally stable finite difference time domain method of second order accuracy in both time and space [11]. 2D process simulations of pultruded profiles were carried out in [12–14] using the FEM. In addition to the temperature and cure calculation, the effect of the uncertainties in the material and resin kinetic properties on the cure degree of the composite at the die exit was investigated by using a probabilistic model based on the 2D FEM in [14].

Apart from 1D and 2D process models, 3D thermo-chemical simulations of the pultrusion process for different composite cross sections were performed in [15–22] in essence using Patankar’s finite volume method (FVM) [23]. In [24], the reliability of the pultrusion process was estimated using the first order reliability method (FORM) in which the process was modeled by implementing the 3D CV/FD approach. A multi objective problem was defined in [25] for the pultrusion of a thick flat profile which was modeled by using the 3D CV/FD method. The non-dominated sorting genetic algorithm (NSGA-II) was utilized in [25] to simultaneously maximize the pulling speed and minimize a so-called criterion of ‘total energy consumption’. As a result, a set of optimal solutions were obtained for different trade-offs between these conflicting objectives. In

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**Fig. 1** Schematic of the pultrusion process [1]
The finite element nodal control volume (FE/NCV) method was implemented for the 3D simulation of temperature and cure profiles for the pultrusion process. It was shown that the procedure was numerically stable and the results agreed with corresponding experimental studies published in the literature. The process induced residual stresses and distortions were calculated in [32, 33] using a coupled 3D transient Eulerian thermo-chemical analysis together with a 2D plane strain Lagrangian mechanical analysis of the pultrusion process. For the thermo-chemical analysis, the 3D FE/NCV method was implemented in [32, 33]. A straightforward extension of the symmetrical 2D ADI time integration scheme was utilized in [34] and the results were found to agree with those obtained from using the FEM. However, it was also concluded that the FEM required less computational time as compared with the FDM, i.e. the 3D extension of the 2D ADI (denoted the “conventional 3D ADI” henceforth in this paper), using the same Peclet number as in the FEM. This is expected since the unconditional stability of the 2D ADI scheme becomes conditional in the 3D ADI case which is of first order accurate in time [35, 36]. Therefore, the conventional 3D ADI method requires relatively small time steps in order to ensure a converged solution.

In the present study, a 3D thermo-chemical analysis of the pultrusion process for a composite flat plate is carried out using both transient and steady state approaches. For the transient approach, an unconditionally stable 3D ADI Douglas-Gunn (ADI-DG) method [37] is used for the numerical model of the process and it constitutes the first contribution of its kind in this specific field of application. The novelty for this approach lies in combining the ADI-DG solver with the advection of the composite due to pulling, the nonlinear internal heat generation together with the cure degree evolution as well as using the upwind scheme and applying the whole procedure to the pultrusion process. The implementation of the method, i.e. the time marching and the space discretization, are presented in detail. The results are compared with the transient fully implicit scheme, the conventional ADI scheme and the steady state solution. A unidirectional glass/epoxy composite is considered for the flat plate. The temperature and cure degree distributions at steady state are calculated for the die and the part. Detailed case studies are carried out examining the computational accuracy of the 3D ADI-DG solver, and hence investigating the convergence and stability, expressed in terms of the Courant and Peclet numbers which are essential in stability analyses of coupled transient diffusion-convection problems. Finally, a case study is carried out in which a design of experiments (DOEs) has been performed for the curing characteristics of the product based on pulling speed and part thickness.

2 Governing Equations

The transient heat transfer equations in a Cartesian coordinate system for the composite and the die block are given in Eqs. 1 and 2, respectively. Here, \( x \) is the pulling (axial or longitudinal) direction; \( y \) and \( z \) are the transverse directions in height and width, respectively. In the energy equation, the convective (\( u \partial T/\partial x \)) and the source (\( q \)) terms are present for the composite part only due to the advection of the material and the internal heat generation of the resin, respectively. Similarly, the steady state equations can be obtained by discarding the time dependent term in Eqs. 1 and 2.

\[
\rho_c C_p c \left( \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} \right) = k_{x,c} \frac{\partial^2 T}{\partial x^2} + k_{y,c} \frac{\partial^2 T}{\partial y^2} + k_{z,c} \frac{\partial^2 T}{\partial z^2} + q
\]  

\[
\rho_d C_p d \frac{\partial T}{\partial t} = k_{x,d} \frac{\partial^2 T}{\partial x^2} + k_{y,d} \frac{\partial^2 T}{\partial y^2} + k_{z,d} \frac{\partial^2 T}{\partial z^2}
\]

where \( T \) is the temperature, \( t \) is the time, \( u \) is the pulling speed, \( \rho \) is the density, \( C_p \) is the specific heat and \( k_x, k_y \) and \( k_z \) are the thermal conductivities. The subscripts \( c \) and \( d \) correspond to the composite and the die, respectively. Lumped material properties are used and assumed to be
constant throughout the process as in [19, 24, 30]. The internal heat generation \( q \) [W/m³] due to the exothermic reaction of the epoxy resin can be expressed as

\[
q = (1-V_f)\rho_r Q
\]

where \( V_f \) is the fiber volume ratio and \( Q \) is the specific heat generation rate [W/kg] due to the exothermic cure reaction of the thermosetting resin.

The expression of the degree of cure (\( \alpha \)) can be written as the ratio of the amount of heat generated (\( H(t) \)) during curing, to the total heat of reaction \( H_{tr} \), i.e. \( \alpha = \frac{H(t)}{H_{tr}} \). The rate of the cure degree, \( R_r \), can be written as an Arrhenius equation [19],

\[
R_r(\alpha) = \frac{d\alpha}{dt} = \frac{1}{H_{tr}} \frac{dH(t)}{dt} = K_o \exp \left( \frac{-E}{RT} \right) (1-\alpha)^n
\]

where \( K_o \) is the pre-exponential constant, \( E \) is the activation energy, \( R \) is the universal gas constant and \( n \) is the order of reaction (kinetic exponent). \( K_o, E, H_{tr} \) and \( n \) can be obtained by a curve fitting procedure applied to the experimental data evaluated using differential scanning calorimetry DSC [19]. As a result, \( Q \) (Eq. 3) can be expressed as

\[
Q = \frac{dH(t)}{dt} = H_{tr} R_r(\alpha)
\]

The transient time integration scheme for the rate of cure degree can be derived by using the chain rule. From this, the rate of cure degree (Eq. 4) can be expressed as:

\[
\frac{d\alpha}{dt} = \frac{\partial \alpha}{\partial t} + \frac{\partial \alpha}{\partial x} \frac{dx}{dt} = \frac{\partial \alpha}{\partial t} + \frac{u}{\partial x}
\]

and from Eq. 6, the time integration scheme of the resin kinetics equation can be expressed as

\[
\frac{\partial \alpha}{\partial t} = R_r(\alpha) - u \frac{\partial \alpha}{\partial x}
\]

where it is the expression in Eq. 7 which is used in the transient numerical model. For the steady state model, the time dependent term in Eq. 7 is equated to zero, i.e. \( \frac{\partial \alpha}{\partial t} = 0 \).

3 Numerical Implementation

Temperature and cure degree profiles at steady state are needed for the evaluation of the pultrusion since it is a continuous process; the composite part entering the heating die keeps tracking these steady state profiles during processing. In order to reach steady state, the 3D transient and steady state solution techniques are investigated in the present work.

3D discretization in the space domain is obtained by the CV/FD technique in which the total thermal resistances (K/W) being the sum of the single resistances coupled in series between the two adjacent control volumes are used [38]. The CV/FD method has already been used in similar studies (i.e. numerical modeling of the pultrusion process) in the literature, e.g. in [8, 9, 24]. The representation of the thermal resistances in \( x, y \) and \( z \) directions for an internal CV, i.e. the node \((i,j,k)\), is seen in Fig. 2. Here, the resistances in the \( x-y \) plane and the \( y-z \) plane are seen in Fig. 2 (left) and Fig. 2 (right), respectively. The upwind scheme is used for the convective term \((u\partial T/\partial x)\) in the energy equation and for the space discretization of the cure degree \((u\partial \alpha/\partial x)\) in the resin kinetics equation in order to obtain a stable solution for high Peclet (Pe) numbers,
i.e. $Pe>2$. It should be noted that the dimensionless $Pe$ number indicates the strength between the convective and the conductive terms in the flow simulation as expressed in Eq. 8. In theory, $Pe$ should be less than 2 in order to get stable results when using a central finite difference discretization [23]. However, the upwind scheme avoids the possible oscillations during the simulation for large $Pe$. The internal heat generation together with the resin kinetics equation (Eq. 4) is coupled with the energy equation (Eq. 1) in an explicit manner to get a fast numerical solution since the internal heat generation is highly nonlinear. This time-stepping procedure can be seen as a flowchart in Fig. 3. It is also seen from Fig. 3 that the degree of cure is subsequently updated explicitly for each CV by using Eq. 7 in its discretized form as given in Eq. 9. To reach steady state, the convergence limits are defined as the maximum temperature and cure degree difference between the new time step ($n+1$) and the old time step ($n$), i.e. $\Delta T = \max(T^{n+1} - T^n)$ and $\Delta \alpha = \max(\alpha^{n+1} - \alpha^n)$ and these are set to 0.001 °C and 0.0001, respectively (Fig. 3).

$$Pe = \frac{u \Delta x}{\frac{\rho_c C_p c}{k_{x,c}}}$$

$$\frac{\alpha_{i,j,k}^{n+1} - \alpha_{i,j,k}^n}{\Delta t} = [R_r(\alpha)]^n u \frac{\alpha_{i,j,k}^n - \alpha_{i-1,j,k}^n}{\Delta x}$$

3.1 Transient Approach

The transient solution is suitable for the simulation of the pultrusion process in which the material properties are a function of time, temperature, etc. Additionally, it is also convenient to simulate the transient pultruder operation in which the heaters operate with a heating power and a feedback thermocouple controls the heater temperature within a prescribed tolerance [19, 30].

As earlier stated, the ADI-DG method is used to solve the 3D transient energy equations (Eqs. 1 and 2). The details of the implementation of the 3D ADI-DG method for the energy equation of the composite, i.e. the time marching and the spatial discretization, are explained in the following. Since the same time integration schemes are used for the energy equation of the die block without having the convective term and the source, the details are not given for the die.
3.2 Steady State Approach

The steady state discretization is obtained by discarding the time dependent term \( \frac{\partial T}{\partial t} \) from the energy equations and given in Appendix A. A steady state solution is convenient for the numerical model when having constant processing conditions throughout the process. A similar iteration procedure as given in Fig. 3 is used to obtain a converged steady state

**Fig. 3** Flowchart of the time-stepping procedure in the transient approach to reach the steady state solution for the temperature and the degree of cure
solution. However, it should be noted that there is no time step in this case; instead, an iteration loop is utilized to obtain the converged results, i.e. $T$ and $\alpha$ as well as $q$ (Fig. 3) are updated until the steady state conditions are satisfied.

4 Test Case: Pultrusion of a Flat Beam

4.1 Problem Description

Pultrusion of a flat beam (thickness of 1/8 in. = 3.18 mm) is simulated as a validation case. A fiberglass/Shell EPON 9420 epoxy based composite is used for the plate and steel is used for the die block [19]. The material properties of the composite and resin kinetic parameters are given in Tables 1 and 2, respectively, and assumed to be constant. The geometrical domain of the pultrusion process is given in Fig. 4. Three heating zones having set temperatures of 171-188-188 °C (in the order of the pulling direction) are defined [19, 24] and assumed to be constant throughout the process.

Only one quarter of the pultrusion domain which is seen in Fig. 4 is modeled due to symmetry. A schematic view of the discretization of the cross section of the composite plate and die block is given in Fig. 5. Note that the total thickness of the composite part is indicated as $2t$ in Fig. 5. A mesh of 61×10×11 CVs which corresponds to 6710 CVs is used in the numerical model. Perfect thermal contact is assumed at the die-part interface as in [24]. Initially ($t=0$) the temperature of all nodes are assigned to ambient temperature (27 °C) and the cure degree of all composite nodes are assigned to 0. For $t>0$ the temperature and cure degree of all composite nodes at the die inlet are set to the resin bath temperature (30 °C) and 0, respectively. At the symmetry surfaces adiabatic boundaries are defined across which no heat is transmitted. The remaining exterior surfaces of the die are exposed to ambient temperature with a convective heat transfer coefficient of 10 W/(m² K) except for those located at the heating regions. At the die exit, an adiabatic boundary condition is applied to the composite surface. As in [24], cooling channels are located at the first 100 mm under the first heating region. Hence, in this region, all nodes at the layers A-A and B-B in Fig. 4 are set to the cooling temperature (50 °C) during the whole process.

4.2 Results and Discussion

The thermo-chemical simulations are performed using the proposed transient and steady state approaches in MATLAB and executed on a PC with 2.30 GHz Intel 4 processors and 16 GB of memory. The computational efficiency of the ADI-DG method is compared with the transient fully implicit method (which in general is used by general purpose FE softwares but with an FE spatial discretization, of course) as well as the conventional 3D ADI method.

Table 1  Material properties [19]

<table>
<thead>
<tr>
<th>Material</th>
<th>$\rho$ (kg/m³)</th>
<th>$C_p$ (J/kg K)</th>
<th>$k_x$ (W/m K)</th>
<th>$k_y$, $k_z$ (W/m K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Epoxy resin</td>
<td>1260</td>
<td>1255</td>
<td>0.21</td>
<td>0.21</td>
</tr>
<tr>
<td>Fiberglass</td>
<td>2560</td>
<td>670</td>
<td>11.4</td>
<td>1.04</td>
</tr>
<tr>
<td>Lumped (V_f=63.9 %)</td>
<td>2090.7</td>
<td>797.27</td>
<td>0.905</td>
<td>0.559</td>
</tr>
<tr>
<td>Steel die</td>
<td>7833</td>
<td>460</td>
<td>40</td>
<td>40</td>
</tr>
</tbody>
</table>
and the steady state solution. The temperature and cure degree distributions are predicted for a pulling speed of 200 mm/min. A time step (\( \Delta t \)) of 4.5 s is used for the transient solution. The length of the CV in the pulling direction is determined as 15 mm which gives a Pe of 92. The corresponding Courant number (\( Cr \)) is calculated as 1.0. Cr is a dimensionless number and in general, the necessary condition for obtaining a stable (without any oscillations in the solution) and converged solution to a flow problem solved by the FD method is that \( Cr \leq 1 \) \([40, 41]\). The expression for \( Cr \) is given in Eq. 10.

\[
Cr = \frac{u \Delta t}{\Delta x}
\]  

(10)

The calculated centerline temperature and cure degree profiles (at steady state) for the composite are shown in Fig. 6. It is seen that the results obtained using the three different techniques agree well with those calculated for the similar pultrusion set-ups in \([19, 24, 30]\). The present results are much closer to the results in \([24]\) as compared to \([19, 30]\). The reason for that is the usage of prescribed heater temperatures (constant throughout the process) for the heating zones as in \([24]\) instead of using the heat fluxes for those zones which are switched on and off depending on the temperature of the control node as described in \([19, 30]\). Due to the exothermic internal heat generation inside the thermosetting resin, the centerline composite temperature (~190 °C) exceeds the heater temperature (188 °C). The centerline degree of cure at the exit of the die (i.e. at \( x=0.915 \) m) is calculated as approximately 0.91.

The computational details of the proposed approaches are given in Table 3. It is seen that the ADI-DG method is approximately 2 times faster than the transient implicit method even though the numbers of time increments for reaching the steady state condition are very close to each other. On the other hand, the steady state approach is extremely fast since it only requires 6

<table>
<thead>
<tr>
<th>( H_tr ) (J/kg)</th>
<th>( K_0 ) (1/s)</th>
<th>( E ) (J/mol)</th>
<th>( n )</th>
</tr>
</thead>
<tbody>
<tr>
<td>323700</td>
<td>191400</td>
<td>60500</td>
<td>1.69</td>
</tr>
</tbody>
</table>

Fig. 4 Pultrusion domain of the composite flat plate \([24]\). A mesh of 61 \( \times \) 10 \( \times \) 11 CVs which corresponds to 6710 CVs is used for the numerical model.
iterations to satisfy the steady state conditions and it takes only 0.43 s in computational time for the same problem having the same discretization and $Pe$ as used in the transient approach.

The computational time comparison of the process simulations with the ADI-DG, the implicit and the steady state approaches for different number of CVs in the pulling direction is illustrated in Fig. 7. Here, a time step of $\Delta t=0.5$ s is used for the transient approaches, i.e. the ADI-DG and the implicit. As can be seen in Fig. 7, the computational time of the ADI-DG method is almost linearly proportional to the number of CVs. However, it increases dramatically for the implicit method. This shows that more CVs make the ADI-DG solver relatively faster as compared to the fully implicit method. On the other hand, the increase in the computational time with an increase in the number of CVs for the steady state solution is very small as compared to the transient solutions. It should be noted that as expected, the trend of the increase in computational time for the steady state approach (Fig. 7 (right)) is similar to the one for the transient implicit approach which of course is expected.

The evolution of $\max(T^{n+1} - T^n)$ and $\max(\alpha^{n+1} - \alpha^n)$ with time inside the composite part is shown in Fig. 8. Here, the results obtained from conventional ADI and ADI-DG are presented. The simulations are carried out for the time steps of $\Delta t=1$ s and $\Delta t=0.5$ s in order to compare the stability of the two methods with relatively small time steps. It should be noted that the steady state is reached when $\max(T^{n+1} - T^n)<10^{-3}$ and $\max(\alpha^{n+1} - \alpha^n)<10^{-4}$ (see Fig. 3). It is seen from Fig. 8 that both methods have converged after 1,000 time increments for $\Delta t=0.5$ s, however, the conventional ADI method diverged for $\Delta t=1$ s. The corresponding $Cr$ is calculated as 0.22 and 0.11 for $\Delta t=1$ s and $\Delta t=0.5$ s, respectively. Although $Cr=0.22<1$, it is found that the conventional ADI scheme diverges because of the first order accuracy in time, which is a well known drawback of the conventional 3D ADI (straightforward extension of the 2D ADI) [35, 36]. A similar diverging behavior is also obtained for different CV sizes while using the conventional 3D ADI method and the corresponding results are not presented in this work. On the other hand, the 3D ADI-DG approach gives stable and converged results for $Cr=0.22$, ...
since the Crank-Nicolson scheme is used in the ADI-DG instead of fully implicit scheme as in the conventional 3D ADI. However, as expected the ADI-DG method starts showing

Table 3  Comparison of the proposed transient and steady state approaches with 61 CVs in the pulling direction

<table>
<thead>
<tr>
<th></th>
<th>Transient (ADI-DG)</th>
<th>Transient (Implicit)</th>
<th>Steady state approach</th>
</tr>
</thead>
<tbody>
<tr>
<td>Computational time [s]</td>
<td>5.26</td>
<td>10.82</td>
<td>0.43</td>
</tr>
<tr>
<td>Number of time increments/iterations to reach the steady state</td>
<td>168</td>
<td>175</td>
<td>6</td>
</tr>
<tr>
<td>(\Delta t) [s]</td>
<td>4.5</td>
<td>4.5</td>
<td>–</td>
</tr>
<tr>
<td>(Pe)</td>
<td>92.0</td>
<td>92.0</td>
<td>92.0</td>
</tr>
<tr>
<td>(Cr)</td>
<td>1.0</td>
<td>1.0</td>
<td>–</td>
</tr>
</tbody>
</table>
oscillations in the solution for \( Cr > 1 \). This confirms the expectation that a much faster and more efficient solution can be obtained using the 3D ADI-DG method as compared to the conventional 3D ADI method.

5 Case Study: Effect of Pulling Speed and Part Thickness

5.1 Problem Description

In order to show the potential of the steady state approach, thermo-chemical simulations are performed for different pulling speeds and thickness values of the flat beam. In addition to the temperature and cure degree distributions, the maximal through thickness degree of cure difference during the process are also investigated by using the proposed steady state approach since it is relatively faster than the ADI-DG approach. The through thickness cure gradients are important for pultrusion since they result in non-uniform curing and hence, at the end of the process, manufacturing induced residual stresses and distortions may arise [32].

The same meshing as given in Section 3 is used except for the number of CVs in the thickness direction (\( z \)-direction in Fig. 5) for the composite, i.e. in this case 10 CVs are used instead of 1 CV which was used in the validation case in Section 3. Hence, a mesh of
61×10×20 (x×y×z directions) CVs which corresponds to a total of 12,200 CVs are used in this case study. It should be noted that the width of the part as well as the dimensions of the die are kept constant.

5.2 Results and Discussion

The temperature and degree of cure distributions together with the maximal through thickness cure gradients are calculated for a total of 12 different pulling speeds varying between 100–1,200 mm/min which provide relatively high values of $Pe$ ($92 < Pe < 550$). For each pulling speed, a total of 18 flat beams having different thickness values ($3.18 \text{ mm} < t < 45 \text{ mm}$) are considered. Therefore, a total of $12 \times 18 = 196$ simulations are performed using the proposed steady state approach which means that this can also be considered as a design of experiments for this specific process set-up and material.

The contour plot of the centerline degree of cure at the die exit is shown in Fig. 9 as a function of pulling speed and part thickness. Note that stable results are obtained even for high $Pe$. It is seen from Fig. 9 that the degree of cure decreases as the speed increases for all the thickness values. The rate of this decrease is relatively larger for large thicknesses as compared to small thicknesses because it is more difficult to cure the thicker profile which requires more curing time. For lower speeds, the degree of cure increases with an increase in the part thickness ($2t$) up to a certain value and decreases after that point. For instance, the degree of cure at the exit as a function of $2t$ for a pulling speed of 200 mm/min is shown in Fig. 10 in which the degree of cure increases up to approximately $2t = 15 \text{ mm}$ because the contribution of the exothermic reaction of the resin increases up to $2t = 15 \text{ mm}$. After that point, the degree of cure at the exit starts decreasing due to the decrease in curing rate.

The corresponding maximal through thickness degree of cure difference $\Delta \alpha$ (between center point and top points, see Fig. 9) during the process is depicted in Fig. 11 as a function of pulling speed and part thickness ($2t$). Note that Fig. 11 should be seen together with the degree of cure distribution as a function of pulling speed and part thickness (Fig. 9). It is seen from Fig. 11 that there is a nonlinear relationship between $\Delta \alpha$ and the speed and thickness. $\Delta \alpha$ becomes larger as the part thickness increases for lower pulling speeds (approximately between 100–300 mm/min). On the other hand $\Delta \alpha$ remains almost constant for part thickness values up to approximately 10 mm for all the pulling speeds even though there is a gradual decrease in the degree of cure (see Fig. 9).


The performed design of experiments for the curing characteristics shows that it is not trivial to predict the optimum process parameters to get the desired manufacturing process conditions.

6 Conclusions

In the present study, 3D thermo-chemical simulation of the pultrusion process for a flat beam was performed using transient and steady state approaches. An unconditionally stable 3D ADI-DG solver was applied for the transient solution and compared with the steady state solution and the other transient solutions (implicit and conventional ADI). The results obtained from proposed

![Fig. 10](image-url) The centerline degree of cure as a function of part thickness (2 t) for a pulling speed of 200 mm/min

![Fig. 11](image-url) The contour plot of the maximal through thickness degree of cure difference ($\Delta \alpha$) as a function of pulling speed and part thickness (2 t)
approaches were validated with similar models given in literature, e.g. in [19, 24, 30]. The main outcomes of the present work are summarized as follows:

i. The transient solution is suitable for the simulation of the pultrusion process when the transient process conditions are pronounced such as time and/or temperature dependent material properties, time dependent “switch on/off” type heaters etc. Unlike the transient solution, the steady state solution is more convenient for the simulation of pultrusion having constant processing conditions throughout the process in terms of computational efficiency.

ii. The proposed transient solver (ADI-DG) implemented for the pultrusion process simulation which includes the advection term and the nonlinear internal heat generation in the energy equation was found to be relatively faster as compared to the transient fully implicit method.

iii. The steady state solution is found to be extremely fast as compared to the transient approaches. The increase in absolute computational time is very small for the steady state solution with an increase in the CVs as compared to the transient solutions. Using this approach a design of experiments was carried out for this specific process set-up in or to characterize the curing behavior of the product based on different pulling speeds and part thickness values.

The proposed 3D ADI-DG solver presented in this work has a great potential for the transient simulation of the pultrusion having time dependent processing conditions. On the other hand, the steady state solution would be useful for optimization studies or probabilistic analysis of the pultrusion process having constant processing conditions in which a large number of function evaluations are required.

Acknowledgments This work is a part of DeepWind project which has been granted by the European Commission (EC) under FP7 program platform Future Emerging Technology.

Appendix A

The detailed time integration and corresponding space discretization of the energy equation (Eq. 1) for the 3D ADI-DG approach are expressed in the following.

Step I: \( n \rightarrow n+1/3 \)

\[
\rho_c C_p \left\{ \frac{\partial T}{\partial t} \right\}^{n+\frac{1}{3}} + \frac{u}{2} \left[ \frac{\partial T}{\partial x} \right]^n + \frac{\left[ \frac{\partial T}{\partial x} \right]^n}{c_x} = k_{y,c} \left[ \frac{\partial^2 T}{\partial y^2} \right]^n + k_{z,c} \left[ \frac{\partial^2 T}{\partial z^2} \right]^n + \frac{1}{3} |q|^n
\]

\[
\rho_{i,j,k} C_p \left[ \frac{T_{i,j,k}^{n+1/3} - T_{i,j,k}^{n+1/3}}{\Delta t} + \frac{u}{2} \left( \frac{T_{i,j,k}^{n+1/3} - T_{i+1,j,k}^{n+1/3}}{\Delta x_{i,j,k}} + \frac{T_{i,j,k}^{n+1/3} - T_{i,j,k}^{n+1/3}}{\Delta x_{i,j,k}} \right) \right] =
\]

\[
\frac{1}{2} \left[ \frac{T_{i+1,j,k}^{n+1/3} - T_{i,j,k}^{n+1/3}}{R_{i+1,j,k} + R_{i,j,k}} + \frac{T_{i-1,j,k}^{n+1/3} - T_{i,j,k}^{n+1/3}}{R_{i+1,j,k} + R_{i,j,k}} \right] + \frac{T_{i,j,k+1}^{n+1/3} - T_{i,j,k}^{n+1/3}}{R_{i,j,k+1} + R_{i,j,k}} + \frac{T_{i,j,k-1}^{n+1/3} - T_{i,j,k}^{n+1/3}}{R_{i,j,k-1} + R_{i,j,k}} + \frac{T_{i,j,k}^{n+1} - T_{i,j,k}^{n}}{R_{i,j,k+1} + R_{i,j,k}} + \frac{T_{i,j,k}^{n-1} - T_{i,j,k}^{n}}{R_{i,j,k-1} + R_{i,j,k}} + \frac{1}{3} |q|^n
\]
Step II: \( n+1/3 \rightarrow n+2/3 \)

\[
\rho_c C_p C_i \left\{ \frac{\partial T^n_{i,j,k}}{\partial t} + \frac{u}{2} \left( \frac{\partial^2 T^n_{i,j,k}}{\partial x^2} \right) \right\} = \frac{k_{xc}}{2} \left( \frac{\partial^2 T^n_{i,j,k}}{\partial x^2} \right)^2 + \frac{k_{yc}}{2} \left( \frac{\partial^2 T^n_{i,j,k}}{\partial y^2} \right)^2 + \frac{k_{zc}}{2} \left( \frac{\partial^2 T^n_{i,j,k}}{\partial z^2} \right)^2 + \frac{1}{3} |q|^n
\]  

(A3)

\[
\rho_{i,j,k} C_{pi,j,k} \left[ \frac{T^n_{i+1,j,k} - T^n_{i,j,k}}{\Delta t} \right] + \frac{u}{2} \left( \frac{T^{n+1/3}_{i,j,k} - T^{n+2/3}_{i,j,k}}{\Delta x_{i,j,k}} \right) + \frac{T^n_{i+1,j,k} - T^n_{i,j,k}}{\Delta x_{i,j,k}} = \frac{T^n_{i,j,k} - T^n_{i-1,j,k}}{\Delta x_{i,j,k}}
\]

(A4)

Step III: \( n+2/3 \rightarrow n+1 \)

\[
\rho_c C_p C_i \left\{ \frac{\partial T^{n+1}_{i,j,k}}{\partial t} + \frac{u}{2} \left( \frac{\partial^2 T^{n+1}_{i,j,k}}{\partial x^2} \right) \right\} = \frac{k_{xc}}{2} \left( \frac{\partial^2 T^{n+1}_{i,j,k}}{\partial x^2} \right)^2 + \frac{k_{yc}}{2} \left( \frac{\partial^2 T^{n+1}_{i,j,k}}{\partial y^2} \right)^2 + \frac{k_{zc}}{2} \left( \frac{\partial^2 T^{n+1}_{i,j,k}}{\partial z^2} \right)^2 + \frac{1}{3} |q|^n
\]  

(A5)

\[
\rho_{i,j,k} C_{pi,j,k} \left[ \frac{T^{n+1}_{i+1,j,k} - T^{n+1}_{i,j,k}}{\Delta t} \right] + \frac{u}{2} \left( \frac{T^{n+1/3}_{i,j,k} - T^{n+2/3}_{i,j,k}}{\Delta x_{i,j,k}} \right) + \frac{T^{n+1}_{i+1,j,k} - T^{n+1}_{i,j,k}}{\Delta x_{i,j,k}} = \frac{T^{n+1}_{i,j,k} - T^{n+1}_{i-1,j,k}}{\Delta x_{i,j,k}}
\]

(A6)
where $\Delta t$ is the time step and the thermal resistances (here combined with the volume of the CV) $R_x$, $R_y$ and $R_z$ in the $x$, $y$ and $z$ directions, respectively, are written as:

\[
R_{x,i,j,k} = \frac{(\Delta x_{i,j,k})^2}{2k_{x,i,j,k}}, \quad R_{y,i,j,k} = \frac{(\Delta y_{i,j,k})^2}{2k_{y,i,j,k}}, \quad R_{z,i,j,k} = \frac{(\Delta z_{i,j,k})^2}{2k_{z,i,j,k}} \quad (A7)
\]

where $\Delta x_{i,j,k}$, $\Delta y_{i,j,k}$ and $\Delta z_{i,j,k}$ are the dimensions of the CV in the $x$, $y$ and $z$ directions, respectively (Fig. 2).

Similarly, for the steady state approach, the time integration of the energy equation (Eq. 1) and the space discretization of the degree of cure equation (Eq. 9) are given in the following

\[
\rho_{l,j,k}C_{l,j,k}\left[ u \left( \frac{T_{i,j,k}^n - T_{i,j,k}^{n-1}}{\Delta x_{i,j,k}} \right) \right] = \left( \frac{T_{i-1,j,k}^n - T_{i,j,k}^n}{R_{x,i-1,j,k} + R_{x,i,j,k}} + \frac{T_{i+1,j,k}^n - T_{i,j,k}^n}{R_{x,i+1,j,k} + R_{x,i,j,k}} \right) + \left( \frac{T_{i,j-1,k}^n - T_{i,j,k}^n}{R_{y,i,j-1,k} + R_{y,i,j,k}} + \frac{T_{i,j+1,k}^n - T_{i,j,k}^n}{R_{y,i,j+1,k} + R_{y,i,j,k}} \right) + [q]^n
\]

\[0 = [R_r(\alpha)]^n - u \frac{\alpha_{i,j,k}^n - \alpha_{i-1,j,k}^n}{\Delta x} \quad (A8)\]

\[0 = [R_r(\alpha)]^n - u \frac{\alpha_{i,j,k}^n - \alpha_{i-1,j,k}^n}{\Delta x} \quad (A9)\]

References

“Pultrusion of a Vertical Axis Wind Turbine Blade Part-I: 3D Thermo-chemical Process Simulation”

Ismet Baran, Cem C. Tutum, Jesper H. Hattel, Remko Akkerman

Pultrusion of a vertical axis wind turbine blade part-I: 3D thermo-chemical process simulation

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Received: 25 December 2013 / Accepted: 11 May 2014

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Abstract A novel three dimensional thermo-chemical simulation of the pultrusion process is presented. A simulation is performed for the pultrusion of a NACA0018 blade profile having a curved geometry, as a part of the DeepWind project. The finite element/nodal control volume (FE/NCV) technique is used. First, a pultrusion simulation of a U-shaped composite profile is performed to validate the model and it is found that the obtained cure degree profiles match with those given in the literature. Subsequently, the pultrusion process simulation of the NACA0018 profile is performed. The evolutions of the temperature and cure degree distributions are predicted inside the heating die and in the post-die region where convective cooling prevails. The effects of varying process conditions on the part quality are investigated for two different heater configurations and with three different pulling speeds. Larger through-thickness gradients are obtained as the pulling speed increases. This will affect the process induced residual stresses and distortions during manufacturing.

Keywords Pultrusion process · Curing · Finite element analysis · NACA0018 airfoil · Thermosetting resin

Introduction

The EU funded DeepWind Project [1, 2] develops a novel concept for a floating offshore vertical axis wind turbine (VAWT) based on the Darrieus design. The main objective of DeepWind is to develop more cost-effective MW-scale wind turbines through innovative technologies for the sea environment rather than advancing existing concepts (i.e. either a horizontal or a vertical axis wind turbine) that are based on onshore technology. Hence the main challenges of the project are to increase the simplicity of the design and the manufacturing techniques as well as to reduce the total cost of an installed offshore wind farm. The concept is aiming at large-scale wind turbines for deep water. The up-scaling potential of the project is 20 MW wind turbines. It is expected that the structural design can be improved to have a higher strength-to-weight ratio for larger chord lengths, e.g. 10–20 m, with a deep water offshore floating system (100–1000 m).

The blade cross section for the VAWT can be constant along the length of the blade. The pultrusion technology is foreseen to be one of the most efficient and suitable methods to manufacture such a composite blade with a constant profile having a large chord. A VAWT blade has already been manufactured by using the pultrusion process, as reported in [3]. Pultrusion is a continuous, automated closed-moulding process, and it is cost effective for high volume production of products with a constant cross section along the length of the part. Figure 1 presents a schematic view of the pultrusion process. Since it is a continuous process, there is little waste material being produced at the start up and the end of the process. It has been widely used for manufacturing highly strengthened continuous fibre composite structures, e.g. stiffeners in wind turbine blades, reinforcements of concrete elements in the construction industry,
Fig. 1 A schematic view of the pultrusion process with resin injection chamber (top) [4] and with open resin bath (bottom). Fibers and resin matrix are pulled together in the pultrusion direction by the pullers through the heated die and then the cured composite is cut by a saw system.

spars of ship hulls, thin wall panel joiners, door/window frames, drive shafts of vehicles, etc. Producing large blades in one piece using a single die will lead to cost reduction for large series production. The pultruded blades can achieve very high stiffness and resistance against aerodynamic loads as well as vibrations. In principle, a production facility with a relatively short die length, e.g. 1–3 m, can be put on a ship. Manufacturing near to the location of the wind turbine installation which will alleviate transportation issues for these large constructions.

Keeping the multi-physics and large amount of variables involved in the composite manufacturing processes in mind, a satisfactory experimental analysis for the production requires considerable time which is obviously not a cost-efficient approach. Therefore, the development of suitable computational models is highly desired in order to analyze the process for different processing conditions.

In literature, there have been several numerical analyses for the pultrusion of relatively simple geometries [4–19]. Generally, thermo-chemical analyses [5, 6, 8, 11], flow simulations [4, 18] and optimizations [7, 10, 13, 19] have been performed using numerical modelling techniques. Material behaviour, transport phenomena, material properties and process models need to be well defined in order to properly simulate the pultrusion process. The description of the material behaviour needs to include the resin kinetics and the chemorheology of the resin system. The transport phenomena involve the heat (and mass due to fluid flow) transfer during the process. A complete description of the pultrusion process requires a thorough understanding of the resin flow, heat transfer, pressure, force (pulling, friction forces etc.) as well as residual stresses and distortions via corresponding numerical models. In the following, some important numerical studies related to the pultrusion are reviewed in detail.

Transient and steady state temperature and cure degree profiles of the composite and the heating die systems during the pultrusion process have been simulated by using numerical techniques such as the finite difference (FD) method and the finite element (FE) method with the control volume (CV) technique or nodal control volume (NCV) technique [4–19]. In [5], the effects of the thermal contact resistance
The exothermic chemical reaction of the resin is described by the cure kinetics equation. The degree of cure ($\alpha$) of the resin is equal to the ratio of the amount of heat generated during curing, to the total heat of reaction $H_{tr}$ [15]:

$$\alpha = \frac{H(t)}{H_{tr}}$$

where $H(t)$ is the amount of heat generated during curing, and $H_{tr}$ is the total heat of reaction. The exothermic resin cure reaction is given by the exothermic chemical process simulation of a pultruded U-shaped cross section is first performed as a validation of the implemented FE/NCV method, using the data from similar analyses in the literature [7, 8]. Following the validation case, the same modelling strategy has been applied for the pultrusion of the NACA0018 profile in which the post-die region is also included in the model. The model predicts the temperature and the degree of cure distributions at steady state. The effects of the process parameters such as the heater configurations and the pulling speed on the quality of the profile are investigated.

Governing equations

Energy equations

The 3D transient heat transfer equations for the composite part and the die block can be written in a Cartesian coordinate system (Eulerian formulation) as, respectively,

$$\rho_c C_p \frac{\partial T}{\partial t} + u \cdot \nabla T = k_{x1,c} \frac{\partial^2 T}{\partial x_1^2} + k_{x2,c} \frac{\partial^2 T}{\partial x_2^2} + k_{x3,c} \frac{\partial^2 T}{\partial x_3^2} + q$$

(1)

$$\rho_d C_p d \frac{\partial T}{\partial t} = k_{x1,d} \frac{\partial^2 T}{\partial x_1^2} + k_{x2,d} \frac{\partial^2 T}{\partial x_2^2} + k_{x3,d} \frac{\partial^2 T}{\partial x_3^2}$$

(2)

where $T$ is the temperature, $t$ is the time, $u$ is the pulling speed, $\rho$ is the density, $C_p$ is the specific heat, and $k_{x1,c}$ and $k_{x3,c}$ are the thermal conductivities in the $x_1$- and $x_3$- direction, respectively. Here, $x_1$ is defined as the pulling direction, $x_2$ and $x_3$ are the transverse directions. The subscripts $c$ and $d$ correspond to the composite and the die, respectively. Lumped material properties are used and assumed to be constant. The volumetric internal heat generation ($q$) [W/m$^3$] due to the exothermic reaction of the epoxy resin is expressed as

$$q = (1 - V_f) \rho_r Q$$

(3)

where $V_f$ is the fiber volume fraction, $\rho_r$ is the resin density and $Q$ is the specific heat generation rate [W/kg] due to the exothermic resin cure reaction.

Cure kinetics

The exothermic chemical reaction of the resin is described by the cure kinetics equation. The degree of cure ($\alpha$) of the resin is equal to the ratio of the amount of heat generated during curing ($H(t)$) to the total heat of reaction $H_{tr}$ [15]:

$$\alpha = \frac{H(t)}{H_{tr}}$$

(4)

where $H(t)$ is the amount of heat generated during curing, and $H_{tr}$ is the total heat of reaction.
The reaction rate, \( R_r(\alpha, T) \), can be expressed for an \( n^{th} \) order reaction as [15]:

\[
R_r(\alpha, T) = \frac{d\alpha}{dt} = \frac{1}{H_{tr}} \frac{dH(t)}{dt} = K_o \exp\left(-\frac{E}{RT}\right)(1-\alpha)^n \tag{5}
\]

where \( K_o \) is the pre-exponential constant, \( E \) is the activation energy, \( R \) is the universal gas constant and \( n \) is the order of reaction (kinetic exponent). \( K_o, E, \) and \( n \) can be obtained by a curve fitting procedure applied to the experimental data evaluated using the differential scanning calorimetry (DSC) [20]. The specific heat generation rate, \( Q \) (in Eq. 3), is then calculated as [15]

\[
Q = H_{tr} R_r(\alpha, T) \tag{6}
\]

\(Q\) is used in the transient thermo-chemical model.

### Numerical implementation

The Eulerian formulation used here requires the spatial derivatives rather than the material derivatives as in Eq. 5 \((d\alpha/dt)\). The spatial derivatives can be found from relation given in Eq. 7.

\[
\frac{d\alpha}{dt} = \frac{\partial \alpha}{\partial t} + \frac{\partial \alpha}{\partial x_1} \frac{dx_1}{dt} = \frac{\partial \alpha}{\partial t} + u \frac{\partial \alpha}{\partial x_1} \tag{7}
\]

and from Eq. 7, the resin kinetics equation can be expressed as

\[
\frac{\partial T}{\partial t} = R_r(\alpha, T) - u \frac{\partial \alpha}{\partial x_1} \tag{8}
\]

which is used in the transient thermo-chemical model.

### Validation case

**Model description**

A 3D thermo-chemical simulation of the pultrusion process for a composite U-shaped profile was performed as a validation case. A UD glass/epoxy was used for the composite material and chrome steel was selected as the die material. The material properties and the cure kinetics parameters are given in Tables 1 and 2, respectively. The material properties of the composite are calculated by a rule of mixtures based on a fiber volume fraction of \( V_f = 63.9 \% \).

The model geometry, shown in Fig. 3, is taken from similar set-ups used in the literature [7, 8]. Only half of the cross section has been considered due to the symmetry, applying an adiabatic thermal boundary condition (BC) at the symmetry surface. Three heating platens are assumed to be
Table 1  Thermal properties used in the process simulation [7, 8]

<table>
<thead>
<tr>
<th>t1.2</th>
<th>ρ [kg/m³]</th>
<th>Cp [J/kg K]</th>
<th>(k_{x_1}) [W/m K]</th>
<th>(k_{x_2}, k_{x_3}) [W/m K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>t1.3</td>
<td>Composite</td>
<td>2090.7</td>
<td>797.27</td>
<td>0.9053</td>
</tr>
<tr>
<td>t1.4</td>
<td>Steel die</td>
<td>7833</td>
<td>460</td>
<td>40</td>
</tr>
<tr>
<td>t1.5</td>
<td>(V_f = 0.639)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 3  The set temperatures of the heaters (°C) used in the validation case

<table>
<thead>
<tr>
<th>t3.1</th>
<th>Heater-1</th>
<th>Heater-2</th>
<th>Heater-3</th>
<th>Heater-4</th>
<th>Heater-5</th>
<th>Heater-6</th>
</tr>
</thead>
<tbody>
<tr>
<td>105.5</td>
<td>148.5</td>
<td>200.0</td>
<td>115.5</td>
<td>146.5</td>
<td>200.0</td>
<td></td>
</tr>
</tbody>
</table>

[7, 8], perfect thermal contact is assumed between the die and the composite.

Results and discussions of the validation case

The degree of cure profiles at steady state shown in Fig. 5 are obtained with a pulling speed of 2.3 mm/s by using the FE/NCV approach inspired by [7, 8]. It is seen that the results match with those in [7, 8]. This shows that the implemented numerical scheme is stable and converged to the correct solution. It is seen from Fig. 5 that point B (along the pulling direction) starts curing before point A. The contribution of the die temperature to the cure degree is more dominant than the contribution of the internal heat generation at the die-part interface (e.g., point B) since point B is closer to the heaters. The reverse is the case for the inner regions of the composite (e.g., point A). However, approximately from \(x_1 = 0.6\) m the curing rate of point A is higher than at point B because the internal heat generation plays a more important role at the inner region of the composite. Therefore, although the curing at point A starts at a later stage, the cure degree of point A becomes almost the same as the cure degree of point B at the die exit which is predicted to be approximately 0.89.

Fig. 3  Symmetric pultrusion model geometry for the U-shaped composite and the corresponding FE mesh in the validation case. All dimensions are in mm.
Pultrusion of a NACA0018 profile

Model description

The thermo-chemical analysis implemented in ABAQUS was then used to simulate the pultrusion process of the NACA0018 profile. The schematic representation of the model is seen in Fig. 6. The heater locations are selected similarly to the validation case (in Section “Validation case”); however, in this case only three heating platens on one side are included in the numerical model due to the symmetry. The cross sectional details of the die and the blade including the FE mesh are seen in Fig. 7. The same material properties and BCs are used as in the validation case for the die and the composite. Convective boundaries are defined at the post die region for the exterior surfaces of the blade profile with a convective heat transfer coefficient of 10 W/(m² K). The length required to cool the surface of the profile down to room temperature (Lconv in Fig. 6) was determined by a trial-and-error analysis, leading to a value for Lconv of approximately 9.2 m. Two case studies were carried out based on the set temperature of the heaters which are given in Table 4. The set temperatures of the first case study (Case-1) are assumed to be the same as the first three set temperatures in the validation case. For the second case (Case-2), the corresponding temperatures were taken from [26]. Three different pulling speeds were used for both cases: 2.3 mm/s, 3 mm/s and 5 mm/s, implying total pultrusion process times of approximately 73 min, 56 min and 34 min, respectively, based on the total length of the profile which is approximately 10 m.
Fig. 7 Cross sectional details of the FE mesh for the die (top) and the blade (bottom). All dimensions are in mm

Table 4 The set temperatures (°C) of the heaters for the two cases

<table>
<thead>
<tr>
<th>Case</th>
<th>Heater-1</th>
<th>Heater-2</th>
<th>Heater-3</th>
</tr>
</thead>
<tbody>
<tr>
<td>t4.2</td>
<td>Case-1</td>
<td>105.5</td>
<td>148.5</td>
</tr>
<tr>
<td>t4.3</td>
<td>Case-2</td>
<td>171.0</td>
<td>188.0</td>
</tr>
</tbody>
</table>

Results and discussion

The predicted temperature and cure degree profiles for Case-1 and Case-2 are presented in Figs. 8 and 9, respectively.

In both cases, the temperature at the die-part interface (e.g. at point B in Figs. 8 and 9) remains almost the same for different pulling speed values due to the prescribed temperature of the heaters. The temperature profile at point A is more sensitive to the increase in pulling speed. The profile shifts to the right for both cases when the pulling speed is increased, illustrating that the convective term ($u \partial T / \partial x$) plays a more significant role at the inner regions. The degree of cure distributions follow the same trend: both the temperature and the cure degree profiles at point A shift more to the right than for point B for the two cases as the pulling speed increases. Obviously, also the cure degree profile at point A is more sensitive to the pulling speed than the cure degree profile at point B, higher temperature and final degree of cure values are obtained for this thick-walled section at point A as compared to point B at the end of the process.
as seen from Figs. 8 and 9. In addition, the maximum temperature values at point A are found to be higher than those at point B for all cases. This results in through-thickness temperature and cure degree gradients which together with the heater configuration would have a direct effect on the process induced residual stresses and distortions [28]. The temperature may still increase after the profile has left the die due to exothermic internal heat generation as the curing process continues. In other words, an increase in the pulling speed promotes larger through-thickness gradients of temperature and degree of cure in both cases. The pulling speed has a negative effect on the cure degree at the die exit, i.e. it decreases with an increase in speed; however, this effect vanishes at the end of the process where almost the same cure degree values are obtained for different pulling speeds in both cases (Figs. 8 and 9).

The curing starts earlier in Case-2 due to the higher set temperatures for the first and second heaters (Fig. 9) as compared to Case-1. Therefore, higher cure degree values prevail at the die exit for Case-2. The cure degree values at the die exit and also at the end of the process together with the corresponding % increase in the cure degree are given in Tables 5 and 6 for Case-1 and Case-2, respectively. It is seen that the increase in cure degree, from die exit to the end of the process, becomes larger with an increase in the pulling speed. It should be noted that, even though the cure degree at point A in Case-1 is relatively small at the die exit for a pulling speed of 5 mm/s (i.e. 0.13 in Table 5), surprisingly an increase of more than six times in the cure degree is found (see also Fig. 8(bottom)). This is due to the wall thickness differences leading to an additional variation through the profile cross section (along the chord). Some regions, particularly thinner, cure faster than the region where point A is located (i.e. the thickest region). In addition, high temperature of the regions close to the die surface act as a heater at the post die region.

The degree of cure values at the die exit (x1 = 0.915 m) and the end of the process (x1 ≈ 10 m) for Case-1 (105.5 - 148.5 - 200 °C) with three different pulling speeds

<p>| Table 5 | The degree of cure values at the die exit (x1 = 0.915 m) and the end of the process (x1 ≈ 10 m) for Case-1 (105.5 - 148.5 - 200 °C) with three different pulling speeds |
|---------|-------------------------------------------------|-------------------------------------------------|-------------------------------------------------|-------------------------------------------------|-------------------------------------------------|-------------------------------------------------|-------------------------------------------------|</p>
<table>
<thead>
<tr>
<th>Point A</th>
<th>Die exit</th>
<th>End of process</th>
<th>Increase (%)</th>
<th>Die exit</th>
<th>End of process</th>
<th>Increase (%)</th>
<th>Die exit</th>
<th>End of process</th>
<th>Increase (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.3 mm/s</td>
<td>0.87</td>
<td>0.99</td>
<td>13.5</td>
<td>0.90</td>
<td>0.98</td>
<td>9.1</td>
<td>0.90</td>
<td>0.98</td>
<td>9.1</td>
</tr>
<tr>
<td>3 mm/s</td>
<td>0.71</td>
<td>0.99</td>
<td>38.8</td>
<td>0.87</td>
<td>0.98</td>
<td>12.6</td>
<td>0.87</td>
<td>0.98</td>
<td>12.6</td>
</tr>
<tr>
<td>5 mm/s</td>
<td>0.13</td>
<td>0.99</td>
<td>658.4</td>
<td>0.77</td>
<td>0.98</td>
<td>27.1</td>
<td>0.77</td>
<td>0.98</td>
<td>27.1</td>
</tr>
</tbody>
</table>
Table 6: The degree of cure values at the die exit ($x_1 = 0.915$ m) and the end of the process ($x_1 = 10$ m) for Case-2 (171-188-188 °C) with three different pulling speeds.

<table>
<thead>
<tr>
<th>Point A</th>
<th>Point B</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Die exit</td>
</tr>
<tr>
<td>2.3 mm/s</td>
<td>0.95</td>
</tr>
<tr>
<td>3 mm/s</td>
<td>0.92</td>
</tr>
<tr>
<td>5 mm/s</td>
<td>0.52</td>
</tr>
</tbody>
</table>

Fig. 10: Temperature distributions inside the blade profile at the die exit (top) and at the end of the process (bottom) for Case-1 (105.5-148.5-200 °C) with a pulling speed of 5 mm/s.

Fig. 11: Cure degree distributions inside the blade profile at the die exit (top) and at the end of the process (bottom) for Case-1 (105.5-148.5-200 °C) with a pulling speed of 5 mm/s.
and hence, invoke the exothermic reaction at inner regions. This can be explained in the contour plots given in Figs. 10 and 11 for the temperature and the cure degree distributions, respectively (Case-1 for 5 mm/s). It is seen in Fig. 10 that the temperature in the thinner regions of the profile are found to be between approximately 208–214 °C at the die exit due the rapid curing whereas the temperature is only 142 °C at point A which shows that high through-thickness thermal gradients prevail at the cross section and the curing takes place at earlier stages at the thinner regions as compared with the point A (Fig. 11). Hence, at the post-die, the heat flow is transmitted from the thinner regions to the point A and at the end of the process the thickest section of the profile is found to be almost fully cured (Fig. 11). The large through-thickness variations in temperature and degree of cure may also lead to poor surface quality as well as unwanted surface defects at the end of the process.

Using the proposed numerical tool, designs of various internal cross-section configurations, e.g. reinforcements, stiffeners, spars, etc., can be analysed including the effects of processing parameters on the expected performance. In this way, the simulation can be used to optimize the process in order to obtain the desired mechanical properties of the product.

Conclusion

A thermo-chemical simulation was performed for the pultrusion process of a composite NACA0018 blade. First, a validation case was carried out in which the cure degree distribution of a pultruded U-shaped composite profile was simulated. The implemented numerical model was found to be stable and accurate as compared with the corresponding results in [7, 8]. Subsequently, the temperature and degree of cure evolutions were simulated for the pultrusion of the NACA0018 profile using two different set temperature schemes of the heaters, i.e. Case-1 (105.5-148.5-200 °C) [7] and Case-2 (171-188-188 °C) [26], for three different pulling speeds (2.3 mm/s, 3 mm/s and 5 mm/s). The main outcomes of this study are summarized as follows:

- It was found that higher final cure degree values prevail in Case-2 due to the higher set temperatures of the first and second heaters which invoke curing at early stages as compared with Case-1. Hence, the heater configurations as well as the set temperatures of the heaters must be determined to obtain the desired product quality which might require a more advanced process optimization study such as the work presented in [19].
- There is significant curing taking place in the post die region. This shows that high temperature of the regions close to the die surface acts as a heater at the post die region and hence, invokes the exothermic reaction at inner regions. In other words, the skin acts as an insulator for the core such that there exists a self accelerating cure reaction.

- Although the degree of cure at the die exit decreases with an increase in the pulling speed, almost the same degree of cure values are obtained at the end of the process for different pulling speeds in both cases due to the continued chemical reaction in the post die region.

This study clearly shows the importance of the process parameters such as the set temperatures of the heaters and the pulling speed on the quality, i.e. curing behaviour, in pultrusion of a composite NACA0018 blade. This directly affects the expected mechanical properties of the product as well as the possibility of the defects and process induced residual stresses. The results provide essential input to the prediction of process induced residual stresses and subsequent simulations of the mechanical performance of the pultruded wind turbine components under appropriate loading conditions and can hence be used to optimise the process, the design and the part performance.

Acknowledgements This work is a part of DeepWind project which has been granted by the European Commission (EC) under the FP7 program platform Future Emerging Technology.

References


Springer


“Process Induced Residual Stresses and Distortions in Pultrusion”

Ismet Baran, Cem C. Tutum, Michael W. Nielsen, Jesper H. Hattel

Process induced residual stresses and distortions in pultrusion

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ARTICLE INFO

Article history:
Received 14 November 2012
Received in revised form 17 February 2013
Accepted 10 March 2013
Available online 23 March 2013

Keywords:
A. Thermosetting resin
B. Residual/internal stress
C. Finite element analysis (FEA)
E. Pultrusion

ABSTRACT

In the present study, a coupled 3D transient Eulerian thermo-chemical analysis together with a 2D plane strain Lagrangian mechanical analysis of the pultrusion process, which has not been considered until now, is carried out. The development of the process induced residual stresses and strains together with the distortions are predicted during the pultrusion in which the cure hardening instantaneous linear elastic (CHILE) approach is implemented. At the end of the process, tension stresses prevail for the inner region of the composite since the curing rate is higher here as compared to the outer regions where compression stresses are obtained. The separation between the heating die and the part due to shrinkage is also investigated using a mechanical contact formulation at the die-part interface. The proposed approach is found to be efficient and fast for the calculation of the residual stresses and distortions together with the temperature and the cure distributions.

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1. Introduction

Pultrusion is a continuous, cost-effective manufacturing process for the production of composite structures having constant cross-sections. A schematic representation of the process with and without the use of a resin injection chamber is seen in Fig. 1. The fibers/mats and resin matrix are pulled together along the pultrusion direction by the pullers through the resin bath and the heating die. During this stage at some point in time, the phase of the resin is changed from liquid to gel state. Following this phase change, the resin is cured and finally the composite part is solidified before being pulled by the pulling mechanism out of the die exit. Then the cured component is cut by a saw system at the end of the process.

In literature, thermo-chemical analyses of the pultrusion process have been carried out both numerically and experimentally [2–11] in which the main aim was to provide a better understanding of the process by evaluating the development of the temperature and the degree of cure profiles inside the heating die. However, thermo-mechanical aspects of the pultrusion process such as the evolution of the mechanical properties, process induced stresses, and strains and deformations have not been considered up to now. In the following, some important thermo-mechanical studies related to composite manufacturing processes (CMPs) other than pultrusion such as autoclave, vacuum infusion and resin transfer molding (RTM) are reviewed in detail since the curing rate is higher here as compared to the outer regions where compression stresses are obtained. The separation between the heating die and the part due to shrinkage is also investigated using a mechanical contact formulation at the die-part interface. The proposed approach is found to be efficient and fast for the calculation of the residual stresses and distortions together with the temperature and the cure distributions.

In manufacturing processes of thermoset matrix composites, the phase of the resin is changed from liquid to rubbery state and this transition is referred to as gelation or the gel point. Following this transition, the resin is cured and finally the composite part is solidified, i.e. the resin reaches the glassy state where vitrification occurs when the resin glass transition temperature (Tg) becomes higher than the cure temperature. This complex thermo-chemical behavior is crucial for the thermoset processing and generally represented by time-temperature-transformation (TTT) diagrams [12]. During this curing process, the resin shrinks because of the chemical reaction (cross linking) of the resin which promotes a contraction of the part. Besides that, the part continues contracting due to the cooling effect, e.g. convective cooling at the post-die region in the pultrusion process. It should be noted that the general mechanical behavior of the composite material is orthotropic (transversely isotropic if only unidirectional fibers are used) and the coefficient of thermal expansion (CTE) of the polymer-matrix materials is usually much higher than that of the fibers. Hence, dimensional variations and internal stresses are induced mainly due to the curing shrinkage of resin and the mismatch in the CTE of the fibers and the resin matrix [13]. The difference in the ply-level CTEs in the fiber and transverse directions as well as the interaction between the tool and the composite part can also cause in-plane stresses and deformations in the laminates [14]. Moreover, the temperature and the degree of cure gradients through the composite thickness also promote the development of residual stresses in the manufactured part [15]. At the end of the process (i.e. when the composite cools down to ambient temperature), for some of the CMP types, the part is post-cured at an elevated temperature above Tg which may provide a significant relaxation of the process induced stresses since the resin changes...
phase to rubbery state above \( T_g \) [16]. Hence, after the post-cure process, the effect of cure shrinkage on the residual stresses and warpage in the part is normally said to be negligible [17].

In their well-known work from 1992, Bogetti and Gillespie [18] proposed a cure dependent resin modulus in combination with the chemical shrinkage for one dimensional (1D) thermo-mechanical simulation of the RTM processing of thick composite laminates. The SCFM approach was implemented for the calculation of the effective mechanical properties of the composite. The evolution of the residual stresses during the cure process was predicted using an incremental linear elastic approach built on classical laminate theory (CLT) and the relationship between the gradients in the thermal and the curing fields were investigated. In [19], a visco-elastic model was developed for the calculation of the residual moments and curvatures in unsymmetric cross-ply laminates. The verification of this model was accomplished in [20] in which the process induced residual curvatures were measured. As earlier mentioned, the residual stresses are developing due to the internal volume changes. The main factors for this are: (i) the anisotropic thermal expansion of the composite and (ii) the resin cure shrinkage, which cause micro- and macro-mechanical stresses leading to spring-in or spring-forward in curved laminates [21]. In [21], the process induced deformations of composite parts were predicted using a combined 2D/3D FE modeling technique in which a 2D thermo-chemical FE model was coupled with a 3D mechanical FE model. This technique was applied to a complex composite part where the predicted and measured spring-in angles were found to be relatively similar.

A plane-strain FE process model COMPRO was developed to simulate spring-in and warpage in the autoclave RTM process [22]. For the elastic modulus of the resin an extended version of the Bogetti and Gillespie’s approach [18] was used such that the resin modulus is not only a function of the degree of cure but also a function of the temperature. This procedure was called the cure hardening instantaneous linear elastic (CHILE) approach [22]. In [23], a 2D plane strain FE stress and deformation analysis model was integrated to COMPRO. This code, by utilizing the CHILE approach, can analyze the component temperature, the resin degree of cure, the resin flow and the fiber bed compaction, the process induced stresses and the deformations considering the effects of tooling. In [24], thermo-mechanical properties of 8552 resin and AS4/8522 composite were predicted using the SCFM approach and the Finite Element Based Micromechanics (FEBM) approach. It was found that the calculated values were matching well with the experimental data for a fully cured (glassy) composite and in addition to that it was also shown that reasonable resin properties in the rubbery state were estimated using the SCFM approach.

The movement of the molecules is limited when the resin is at the glassy state below \( T_g \). As a result, the cross-linking of the resin or the network formation has little effect on the expansion of the resin due to a lower CTE at this temperature region. On the other hand, the network formation has a significant effect on the expansion of the resin above \( T_g \) which allows the molecules inside the resin to move much easier as compared to the case below \( T_g \). Due to this higher mobility, the CTE at temperatures above \( T_g \) is higher than that at temperatures below \( T_g \) [25]. This information is supported by experimental data in which a non-contact video extensometer was used to capture the thermal expansion, the laminate consolidation throughout the cure process and also the cure shrinkage [26]. The through-thickness CTE of the cross-ply laminate before gelation was found to be higher than the one after gelation. In addition, it was also concluded in [26] that the cure shrinkage is only a function of the degree of cure regardless of time and temperature, which supports the approach by Bogetti and Gillespie [18].

In [27], the internal strain development in a composite manufactured by the RTM process was measured using the fiber Bragg grating (FBG) sensors. The measured strains were then compared with the FE simulations in which the CHILE [22] and the SCFM [18] approaches were implemented. In this RTM numerical process model, three different mechanical boundary conditions (BCs) were investigated: Free boundary, Tied (perfect) contact at tool/part interface and Frictional contact. The frictional contact model described the strain evolution in the composite during the RTM process qualitatively. The processing properties of the CYCOM 890RTM epoxy resin was characterized in [28]. The cure dependence of the resin above \( T_g \) was described using the Di Benedetto equation and taken into account for the evolution of the elastic modulus of the resin during curing. In addition, the CTE was also characterized based on the resin temperature below or above \( T_g \). In [29], the spring-in angles of curved AS4/8552 composite sections were predicted by using the general purpose FE software ABAQUS. The time dependent constitutive behavior of the resin (i.e. visco-elastic) was neglected. The CHILE and the SCFM approaches were implemented instead of a full visco-elastic model which is more complex and in general requires a higher amount of material data. The predicted spring-in angles for unidirectional and cross-ply parts were compared with measurements for different laminate thicknesses and fiber volumes.

The process induced deformations of a T-shape stiffened thermosetting composite panel were predicted using the general purpose FE software ANSYS for an autoclave co-curing process [32]. The SCFM approach was implemented using the FE method for the calculation of the resin and the composite material properties. The maximum warpage of the skin and the webs was found to be 0.337 mm for a 600 mm long stiffened panel. The process induced residual strains were predicted using the CHILE approach for a vacuum infusion process of a thick glass/epoxy laminate in [33,34] using the FE software ABAQUS. The calculated results were also compared with the monitored strains which were measured by embedded FBG sensors. Different tool-part interface conditions were considered in the process model and it was found that the predicted transverse strains using a tied (perfect) BC at the interface agreed with the measurements. This shows that, although the viscoelastic behavior is inevitably exhibited in the thermoset matrix for the long process times (several hours) in vacuum infusion at elevated temperatures, the linear elastic approach, i.e. the CHILE model, gives reasonable results in this case. In addition, it was also concluded that the magnitude of the process induced strains increased with an increase in the laminate thickness. In [35–37], process induced shape distortions were investigated both
numerically and experimentally. In the corresponding FE model, it was assumed that the mechanical properties of the composite were constant within the rubbery and glassy states such that there was a step change in the resin properties at $T_g$.

In the present study, the mechanical analysis together with the thermo-chemical analysis of the pultrusion process is carried out as a first contribution of its kind in this field. A thin (1/8 in. $= 3.175$ mm) and a thick (1 in. $= 25.4$ mm) unidirectional glass/epoxy composite beams are considered for the process simulations. The cure-dependent and temperature dependent resin modulus is modeled using the CHILE method [22] in which the instantaneous cure-dependent and temperature dependent resin modulus is modified conductivities along

\[ \rho \cdot C_p \cdot \frac{\partial T}{\partial t} = k_1 \frac{\partial^2 T}{\partial x_1^2} + k_2 \frac{\partial^2 T}{\partial x_2^2} + k_3 \frac{\partial^2 T}{\partial x_3^2} + q \]  

(2)

where $T$ is the temperature, $t$ is the time, $u$ is the pulling speed, $\rho$ is the density, $C_p$ is the specific heat and $k_1$, $k_2$, and $k_3$ are the thermal conductivities along $x_1$, $x_2$, and $x_3$ directions, respectively. The subscriptions $c$ and $d$ correspond to composite and die, respectively. Lumped material properties are used and assumed to be constant. The internal heat generation ($q$) [W/cm$^3$] due to the exothermic reaction of the epoxy resin can be expressed as:

\[ q = (1 - V_f) \rho_{r} Q \]  

(3)

where $V_f$ is the fiber volume fraction and $Q$ is the specific heat generation rate [W/kg] due to the resin exothermic cure reaction.

The expression of the degree of cure ($x$) can be written as the ratio of the amount of heat generated ($H(t)$) during curing, to the total heat of reaction $H_r$, i.e. $x = H(t)/H_r$. The rate of the degree of cure, $R$, can be written as an Arrhenius equation [9],

\[ R_i(x) = \frac{dx}{dt} = \frac{dH(t)}{dt} = K_e \exp \left( - \frac{E}{RT} \right) (1 - x)^n \]  

(4)

where $K_e$ is the pre-exponential constant, $E$ is the activation energy, $R$ is the universal gas constant and $n$ is the order of reaction (kinetic exponent). $K_e$, $E$, $H_r$, and $n$ can be obtained by a curve fitting procedure applied to the experimental data evaluated using differential scanning calorimetry DSC [9]. It should be noted that the increase in $x$ is slowing down after vitrification owing to the switch of the resin reaction from a kinetic form (as in Eq. (4)) to a diffusive form. This can also be explained such that the rate of bond formation decreases after vitrification where the resin diffusion takes place [22]. Therefore, in the present study the shift from a kinetics-dominated to a diffusion-dominated resin reaction near the point of vitrification is taken into account by modifying the resin kinetics-controlled equation (Eq. (4)) using the following expression [22]:

\[ R_i(x) = R_i(x) \cdot f(x, T) \]  

(5)

where $[R_i(x)]_e$ is the effective cure rate and $f(x, T)$ is the diffusion factor which accounts for the glass transition effect defined as [22]:

\[ f(x, T) = \frac{1}{1 + \exp[C(x - (x_{c1} + x_{c2} T))]} \]  

(6)

where $C$ is a diffusion constant, $x_{c1}$ is the critical degree of cure at $T = 0$ K and $x_{c2}$ is a constant for the increase in critical $x$ with $T$ [22]. As a result, $Q$ (in Eq. (3)) can be expressed as

\[ Q = \frac{dH(t)}{dt} = H_r \cdot [R_i(x)]_e \]  

(7)

The transient time integration scheme for the rate of the degree of cure can be derived by using the chain rule. Using this, the rate of the degree of cure can be expressed as:

\[ \frac{dx}{dt} = \frac{\partial x}{\partial t} + \frac{\partial x}{\partial x_1} \frac{dx_1}{dt} + \frac{\partial x}{\partial x_2} \frac{dx_2}{dt} + \frac{\partial x}{\partial x_3} \frac{dx_3}{dt} = \frac{\partial x}{\partial t} + u \frac{\partial x}{\partial x_1} \]  

(8)

and from Eq. (8), the relation of the effective resin kinetics equation can be expressed as

\[ \frac{\partial x}{\partial t} = [R_i(x)]_e - u \frac{\partial x}{\partial x_1} \]  

(9)

where it is the expression in Eq. (9) which is used in the transient numerical model.

2. Energy and resin kinetics equations

The 3D transient energy equations for the flat beam and the die block domains are given in Eqs. (1) and (2), respectively for the thermo-chemical simulation of the pultrusion process. Here, $x_3$ is the pulling (axial or longitudinal) direction; $x_1$ and $x_2$ are the transverse directions.
3. Numerical implementation

3.1. Thermo-chemical implementation

For the thermo-chemical simulation of the pultrusion process, two different numerical methods, namely the control volume based finite difference (CV/FD) method [5,6] and the finite element method with nodal control volume (FE/NCV) method [4,11], are used separately in order to ensure that the predicted temperature and degree of cure distributions are accurate and correct at the die and post-die regions. MATLAB and ABAQUS are used for the implementation of the CV/FD and the FE/NCV methods, respectively. In the CV/FD method, the concept of the total thermal resistance, $R$ [K/W], is applied [5,6,40]. In both methods, the nonlinear internal heat generation equation (Eq. (4)) together with the resin kinetics equation (Eq. (9)) is coupled with the energy equation (Eq. (1)) in an explicit manner in order to obtain a straightforward and fast numerical procedure. The degree of cure is subsequently updated explicitly and the solution is therefore limited to the fully cured state. These limitations are overcome in the FE/NCV method [4,11] by use of the nodal control volume (FE/NCV) method.

3.2. Incremental residual stress implementation

The incremental process induced stress calculation is performed using user-subroutines in ABAQUS. The details of the governing equations and the implementation are given subsequently.

3.2.1. Resin modulus development

The stiffness of the resin significantly depends on the degree of cure ($\alpha$). The instantaneous isotropic resin modulus ($E_i$) is expressed as a function of $\alpha$ [18]:

$$E_i = (1-\alpha)E_0^c + 2\alpha E_i^\infty$$

where $E_0^c$ and $E_i^\infty$ are the initial (i.e., uncured) and fully cured resin moduli, respectively. The values are given in Table 1 for polyester and epoxy resin. It should be noted that $E_i^\infty$ is generally assumed to be $E_0^c / 1000$ as a first approximation [18,35–37]. Eq. (10) has been modified by incorporating the temperature dependency as suggested in the CHILE approach [22] which exhibits the cure hardening and also thermal softening as shown in the following equation:

$$E_i = \begin{cases} E_0^c & \text{for } T < T_{C1} \\ E_0^c + \frac{T-T_{C1}}{T_{C2}-T_{C1}}(E_i^\infty - E_0^c) & \text{for } T_{C1} \leq T \leq T_{C2} \\ E_i^\infty & \text{for } T > T_{C2} \end{cases}$$

where $T_{C1}$ and $T_{C2}$ are the critical temperatures at the onset and completion of the glass transition, respectively, $T$ represents the difference between the instantaneous resin glass transition temperature ($T_g$) and the resin temperature, i.e., $T = T_g - T$ [22,23]. The evolution of the $T_g$ with the degree of cure is modeled by Di Benedetto equation [28] and expressed as

$$\frac{T_g - T_{go}}{T_{go} - T_{g0}} = \frac{\alpha}{1 - (1 - \alpha)\beta}$$

where $T_{go}$ and $T_{g0}$ are the glass transition temperatures of uncured and fully cured resin, respectively, $\beta$ is a constant used as fitting parameter [28]. Accordingly, the instantaneous resin shear modulus is calculated as

$$G_i = \frac{E_i}{2(1+\nu_i)}$$

where $\nu_i$ is the Poisson’s ratio of the resin in glassy state which is assumed to remain constant during the process.

3.2.2. Resin volumetric shrinkage model

Resin shrinkage strains ($\varepsilon_i$) dramatically depend on the degree of cure and the total specific volumetric shrinkage of the fully cured resin ($\varepsilon_{fc}$). Assuming uniform contraction for a unit cell in the resin, the incremental isotropic resin shrinkage strain ($\Delta e_i$) can be calculated as [18]

$$\Delta e_i = \sqrt{1 + \Delta V_f} - 1$$

where $\Delta V_f$ is the incremental specific volume shrinkage of the resin which is expressed as a function of the change in the degree of cure ($\Delta \alpha$) and $V_f$ [18]

$$\Delta V_f = \Delta \alpha \cdot V_{fi}$$

3.2.3. Effective thermal and chemical shrinkage strains

The incremental chemical shrinkage strain is a function of the incremental isotropic resin shrinkage strain ($\Delta e_i$) as well as the longitudinal elastic modulus and Poisson’s ratio of the fiber ($E_{11}$ and $v_{12}$) and the resin ($E_i$ and $\nu_i$). The incremental effective chemical shrinkage strain of the composite in the longitudinal direction ($\Delta e_{ch1}$) and the transverse directions ($\Delta e_{ch2}$ and $\Delta e_{ch3}$) for a fiber volume ratio of $V_f$ are given in Eqs. (16) and (17), respectively [18].

$$\Delta e_{ch1} = \frac{\Delta e_i E_i (1 - V_f)}{E_{11} V_f + E_i (1 - V_f)}$$

$$\Delta e_{ch2} = \frac{\Delta e_i E_i (1 - V_f)}{E_{12} V_f + E_i (1 - V_f)}$$

$$\Delta e_{ch3} = \frac{\Delta e_i E_i (1 - V_f)}{E_{13} V_f + E_i (1 - V_f)}$$

Similarly, the effective CTEs of the composite in the longitudinal direction ($\alpha_1$) and in the transverse directions ($\alpha_2$ and $\alpha_3$) are expressed in Eqs. (18) and (19), respectively [18].

$$\alpha_1 = \left[ x_1 E_{11} V_f + x_2 E_{12} (1 - V_f) \right] / E_{11} V_f + E_i (1 - V_f)$$

$$\alpha_2 = \left[ x_2 (1 - x_1) V_f + (x_1 + x_2) (1 - V_f) \right] / E_{12} V_f + E_i (1 - V_f)$$

$$\alpha_3 = \left[ x_3 (1 - x_1) V_f + (x_1 + x_3) (1 - V_f) \right] / E_{13} V_f + E_i (1 - V_f)$$

where $x_1$, $x_2$, and $x_3$ are the CTEs of the fiber in longitudinal and transverse direction, respectively and $\alpha_i$ is the CTE of the resin. The corresponding incremental thermal strains ($\Delta \alpha_i$) are calculated in Eq. (20) using the temperature increment between two consecutive time steps, ($\Delta T$).

$$\Delta \alpha_i = \alpha_i \cdot \Delta T \text{ for } i = 1, 2, 3$$

3.2.4. Incremental stress calculation

Process induced stresses and strains are incrementally solved in ABAQUS. The total incremental strain ($\Delta \varepsilon_i$), which is composed of
the incremental mechanical strain ($\Delta e_{ij}^{\text{mech}}$), thermal strain ($\Delta e_{ij}^{\text{th}}$) and chemical strain ($\Delta e_{ij}^{\text{ch}}$), is given in Eq. (21). Here, the incremental process induced strain ($\Delta e_{ij}^{\text{pr}}$) is defined as the summation of $\Delta e_{ij}^{\text{mech}}$ and $\Delta e_{ij}^{\text{pr}}$ as also done in e.g. [18,22,23]. The incremental stress tensor ($\Delta \sigma_{ij}$) is calculated using the material Jacobian matrix ($J$) based on the incremental mechanical strain tensor ($\Delta e_{ij}^{\text{mech}}$) and the corresponding expressions for the composite in 2D plane strain are given in Eqs. (22) and (23). It should be noted that there is no strain component present in the out of plane direction for the plane strain assumption that is considered for the pultrusion model in this paper.

$$\Delta \sigma_{ij} = \int \Delta e_{ij}^{\text{mech}}$$

$$\Delta e_{ij}^{\text{mech}} = \Delta e_{ij}^{\text{tot}} - \Delta e_{ij}^{\text{pr}}$$

$$\Delta \sigma_{ij} = \left[ \begin{array}{ccc} \Delta \sigma_{11} & 0 & 0 \\ 0 & \Delta \sigma_{22} & 0 \\ 0 & 0 & \Delta \sigma_{33} \end{array} \right] = \left[ \begin{array}{ccc} \Delta \sigma_{11} & 0 & 0 \\ 0 & \Delta \sigma_{22} & 0 \\ 0 & 0 & \Delta \sigma_{33} \end{array} \right]_{\text{mech}}$$

where

$$\Delta = \frac{(1 - V_{12} V_{21} - V_{22} V_{32} - V_{33} V_{13} - 2 V_{12} V_{32} V_{13})}{E_{11} E_{22} E_{33}}$$

The expressions of the effective moduli ($E_{11}$, $E_{22}$, $E_{33}$) and Poisson’s ratios ($V_{12}$, $V_{13}$, $V_{23}$) values of the composite are given in Appendix A. In order to ensure that the stiffness matrix is symmetric (transversely isotropic), the Poisson’s ratio values must satisfy the following relationships

$$V_{32} = V_{23}$$

$$V_{21} = \frac{E_{22} V_{12}}{E_{11}}$$

$$V_{13} = V_{23}$$

$$V_{31} = V_{32}$$

The stress and strain tensors are updated at the end of the time increment as in Eqs. (26) and (27), respectively.

$$\sigma_{ij}^{n+1} = \sigma_{ij}^{n} + \Delta \sigma_{ij}^{n}$$

$$\sigma_{ij}^{n+1} = \sigma_{ij}^{n} + \Delta \sigma_{ij}^{n}$$

where $n$ denotes the time marching.

A validation study for the implementation of the incremental stress calculation is being carried out with the current model and results in the reference paper [18] have been reproduced in Fig. 2.

In this validation, the RTM process in [18] is considered since no previous study in the literature related with the thermo-mechanical analysis of the pultrusion process currently exists. According to the results in Fig. 2, it can be concluded that the implementation of the 2D plane strain residual stress calculation in ABAQUS has been validated and is subsequently used in the pultrusion process model.

### 4. Thermo-chemical analysis of the pultrusion process

#### 4.1. Problem description

A 3D transient thermo-chemical analysis of the pultrusion process for a thin and a thick flat beam is carried out in a Eulerian frame. The pultrusion model is taken from similar set-ups available in the literature [4,6,9]. A glass/epoxy based composite and a steel die are used for the flat beam and the die block, respectively, in which the fiber orientation is unidirectional along the pulling direction. Material properties of the composite and the resin kinetic parameters are listed in Tables 1 and 2, respectively [9].

The parameters used for the diffusion factor (Eq. (6)) and given in Table 2 are determined based on a fitting analysis in which the same temperature and cure profiles inside the heating die are obtained for the thin beam as in [4] by imposing the effective resin kinetics equation (Eq. (5)). It should be mentioned that similar results as compared to those given in [22] are obtained. Only a quarter of the pultrusion domain, seen in Fig. 3, is modeled due to symmetry. Three heating zones having prescribed set temperatures of 171–188–188 °C [9] (see Fig. 3) are defined. The spacing between the heating zones is 15 mm.

#### Table 2

<table>
<thead>
<tr>
<th>$H_0$ (kJ/kg)</th>
<th>$K_0$ (1/s)</th>
<th>$E$ (kJ/mol)</th>
<th>$n$</th>
<th>$C$</th>
<th>$\alpha_C$</th>
<th>$\alpha_T$ (1/K)</th>
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<td>1.69</td>
<td>30</td>
<td>-1.5</td>
<td>0.0055</td>
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</table>
Details of the cross section and the finite element discretization are given in Fig. 4 for the thin beam (3.175 mm thick) and thick beam (25.4 mm thick) configurations. The element length in the pulling direction is 15 mm. Perfect thermal contact is assumed at the die-part interface as in [4,6]. Initially (\(t = 0\)), the temperature of all nodes are assigned to ambient temperature (27 °C) and the degree of cure of all composite nodes are assigned to 0. For \(t > 0\) the temperature of the composite nodes at the die inlet are set to resin bath temperature 30 °C and the degree of cure to 0, respectively. At the symmetry surfaces adiabatic boundaries are defined in which no heat flow is allowed across the boundaries. The remaining exterior surfaces of the die are exposed to ambient temperature with a convective heat transfer coefficient of 10 W/(m² K) except for the surfaces located at the heating regions. Similarly, at the post-die region, convective boundaries are defined for the exterior surfaces of the flat beam. In order to determine the required length of the post-die region (\(L_{\text{conv}}\) in Fig. 3) of the thin and thick beams for cooling down to the ambient temperature after die exit, a couple of trial analyses have been performed. As a result, \(L_{\text{conv}}\) is determined to be 3.7 m and 13.7 m for the thin and thick beams, respectively (Fig. 3). The difference between these values are expected since the thinner beam cools faster as compared to the thicker beam. At the end of the composite part, an adiabatic BC is applied to the composite surface. Cooling channels are located at the initial section 100 mm under the first heating region [4,6,9]. Hence, all the nodes at the layers A–A and B–B in Fig. 4 are set to the cooling temperature (50 °C) during the whole process. The convergence limits of the temperature and the degree of cure are set to 0.001 °C and 0.0001, respectively.

4.2. Results and discussion

The temperature and degree of cure profiles together with \(T_g\) development are predicted by using the CV/FD method which has already been validated in [5,6] and the FE/NCV method. It should be noted that the MATLAB and ABAQUS results are found to be very close to each other, hence only the results obtained from ABAQUS are given in Figs. 5 and 6. The pulling speed is set to 20 cm/min which indicates that the total process time is approximately 18.3 min and 68.6 min for the thin beam and the thick beam, respectively based on the total length. For the thin beam, it is seen that the results (up to the die exit, i.e. 0.915 m) agree well with those having the similar set-up in [4,9] by utilizing Eq. (5).

In Figs. 5 and 6, 'top' and 'center' denote the top line and the center line through the whole length of the part, respectively, as also shown in Fig. 3. The temperature, degree of cure and \(T_g\)
distributions over the cross section are almost uniform throughout the process for the thin beam, i.e. the center line and the top line have similar temperature, degree of cure and $T_g$ evolutions. However, for the thick beam, non-uniform distributions, i.e. larger through-thickness gradients, are obtained, especially inside the die, such that the top line cures earlier than the center line since it is closer to the die having heaters on top of it (Fig. 6). In other words, the contribution of the die temperature to the degree of cure is higher than the contribution of the internal heat generation at the top line for the thick beam and vice versa for the center line. Therefore, higher temperature and degree of cure values are obtained at the center line of the thick beam as compared to thin beam as it is seen from Fig. 5 (right). $T_g$ crosses the composite temperature of 159 °C at approximately 1.1 m from the die inlet for the thin beam, however for the thick beam $T_g$ crosses the composite temperature of 173 °C for the top and 182 °C for the center at approximately 1.6 m and 1.8 m from die inlet, respectively.

At the post-die region the degree of cure is increased slightly for both the thin and the thick beams which indicates that the curing still takes place after the die exit. This fact was also observed in [9]. The increase in the degree of cure at the post-die region is approximately 1% and 6% for the thin beam and the thick beam at top line, respectively. A slightly higher degree of cure is estimated for the center line as compared to the top line of the thick beam at the end of the process (i.e. at the end of the cooling). The reason is that the top line cools down faster than the center line. It is seen from Fig. 5 that the maximum center line temperature is predicted as around 190 °C and 217 °C for the thin and thick beams, respectively. These temperature values are higher than the heater temperature (i.e. 188 °C). This is due to the fact that an exothermic internal heat generation is taking place inside the thermosetting resin after some point in time while curing is still in progress. The center line degree of cure at the end of the cooling process is calculated to be approximately 0.91 and 0.97 for the thin and thick beams, respectively. However, for the thick beam, the degree of cure at the die exit is estimated approximately 0.9 and 0.82 for the top and the center lines, respectively.

5. Residual stress analysis of the pultrusion process

5.1. Problem description

In the 2D mechanical analysis of the pultrusion process, the cross section of the composite flat beam is moved through the pulling direction during the process (Lagrangian frame) meanwhile tracking the corresponding temperature and degree of cure profiles already calculated in the 3D thermo-chemical analysis (Eulerian frame) in Section 4. A detailed description of this procedure, i.e. the coupling of the 3D Eulerian thermo-chemical model with the 2D Langrangian plain-strain mechanical model, is shown in Fig. 7. Since the cross sectional dimensions are much smaller than the total length of the beam in the pulling direction ($x_3$-direction), as earlier mentioned, a plane strain assumption is made for the residual stress analysis in which no strain component is taken into account in the pulling direction (the out of plane strain is assumed...
The corresponding stresses, strains and the displacements are calculated based on the temperature and the cure cycles together with the corresponding $T_g$ of the cross section by using the quadratic plane-strain elements in ABAQUS.

For the mechanical analysis, the die is assumed to be rigid and therefore rigid body surfaces are added at the die-part interface instead of including the meshing for the whole die as was done in the thermo-chemical analysis in Section 4. Between the rigid surfaces and the composite beam, a mechanical contact condition is defined which allows separation at the interface. At the same time this restricts any expansion of the composite beyond the tool interface; however any separation due to resin shrinkage is allowed. The friction force at the contact condition is assumed to be zero (sliding condition). A generic view of the plane strain model including the rigid surfaces and the mechanical BCs is shown in Fig. 7.

The mechanical properties of the resin and the fiber are given in Table 3. It should be noted that the resin CTE in rubbery state ($T_g < T$) is known to be approximately 2.5 times larger than the CTE in glassy state ($T_g > T$) [33–35] given in Table 3. The constants in the CHILE model are assumed to be the same values as used in [22], i.e. $T_{C1} = -45.7$ °C and $T_{C2} = -12$ °C, since a high temperature cure cycle was used in [22] which is similar to pultrusion and there is no available data in the literature regarding the evolution of the resin modulus for pultrusion process. For the $T_g$ calculation in Eq. (12), $T_{g\infty}$ is set to 195 °C [41] and $T_{g0}$ is assumed to be 0 °C. The

<table>
<thead>
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<th>Properties</th>
<th>Glass</th>
<th>Epoxy</th>
</tr>
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<td>$E_1^r$ (MPa)</td>
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<td>Eq. (A.3)</td>
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<tr>
<td>$E_{22}$ (MPa)</td>
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<td>Eq. (A.3)</td>
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<td>0.30</td>
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<tr>
<td>$v_{23}$</td>
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<td>0.30</td>
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<td>$G_{12}$ (MPa)</td>
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<tr>
<td>$\alpha_2 (1/\text{C})$</td>
<td>$5.04 \times 10^{-6}$</td>
<td>$5.76 \times 10^{-5}$</td>
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</tbody>
</table>

Fig. 6. The zoomed plots of the temperature and $T_g$ (top) and the degree of cure (bottom) for the thick beam for the axial distance between 0 and 1.5 m.

Fig. 7. Representation of the coupling of the 3D Eulerian thermo-chemical model with the 2D Langrangian plain-strain mechanical model including the rigid body surfaces and the mechanical BCs.
fitting parameter in Eq. (12) is taken as \( k = 0.4 \) which is the expected value for thermosetting resins [28].

The range of the total volumetric shrinkage \( (V_{sh}) \) for the epoxy resin is in general between 2% and 7% as given in [42]. Similarly, in [43] \( V_{th} \) was predicted as 6.9% for an epoxy resin based on an experimental study. Therefore, in the present study \( V_{th} \) is assumed to be 6%.

A representation of the resin modulus \( (E_r) \) development which is calculated by using the CHILE model (Eq. (11)) is shown in Fig. 8 for the thick beam. It is seen that the top point starts gaining stiffness when \( x = 0.86 \) at approximately 0.78 m from the die inlet. On the other hand, the center point gains stiffness later on when \( x = 0.93 \) at approximately 1.13 m from the die inlet.

5.2. Results and discussions: thick beam

The evolution of the displacement profile at the top point in the \( x_2 \)-direction (U2) and at the corner point in the \( x_1 \)-direction (U1) are shown in Fig. 9. It is seen that there is no separation between the die and the beam at the top point until the die exit which indicates that the expansion of the part due to the temperature increase is prevented by the rigid surfaces. However, separation takes place at the corner point at approximately 0.88 m from the die inlet (35 mm from the die exit). It should be noted that the initial pressure condition of the part before entering the heating die is not taken into account which may affect the location of the separation. After die exit, the magnitude (absolute value) of the displacements increases subsequently because of the shrinkage.

The evolution of the linear process induced residual stresses and the corresponding strains in the transverse directions at the top line and the center line are given in Fig. 10. S11 and S22 are the normal stresses in the \( x_1 \)-direction (horizontal, transverse) and the \( x_2 \)-direction (vertical, transverse), respectively. \( \varepsilon^{\text{th}} \) is the linear thermal strain and \( \varepsilon^{\text{ch}} \) is the process induced strain in the transverse directions. The results are summarized and discussed in the following. Note that the results should be seen together with the temperature and the degree of cure curves given in Figs. 5 and 6 and the resin modulus development given in Fig. 8.

- For the top line, S22 is almost zero since there is no BC that constrains the top line in \( x_2 \)-direction (Fig. 10a).
- Inside the die, compressive (negative) S11 and S22 prevail since the expansion of the part is restricted by the die (Fig. 10c and d). These values are relatively small at that point since the resin has not enough stiffness to build up the stresses. However, after app. 0.88 m, a tensile S11 starts emerging due to the separation of the corner node in the \( x_1 \)-direction (U1) in Fig. 9.
- The top point starts gaining stiffness after app. 0.78 m from the die inlet when \( x = 0.86 \) and hence, there is a sharp increase in S11 (Fig. 10c). Similarly, there is also a sharp increase in S11 and S22 at the center point after approximately 1.13 m from the die inlet when \( x = 0.93 \) (Fig. 10b) since it gains stiffness after this point.
- The outer regions closest to the die cure first which make them little constrained by the inner region during shrinkage. Due to this, residual compression is found in these regions near the die which is coherent with the observations given in Bogetti and Gillespie’s work [18]. On the other hand, the interior regions cure and contract later but faster owing to more dominant exothermic reaction here (Fig. 6). When the internal region cures and contracts, it is constrained by the already cured exterior region which provides tension stresses at the center while upholding the self static equilibrium in which there is no applied external load [18].

![Fig. 8. Cure-dependent and temperature-dependent resin modulus (E_r) development for the thick beam. Blue color for the center line, red color for the top time. Arrows indicate the course of the process. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)](image1)

![Fig. 9. Displacement profile for top in the x_2-direction (U2) and for corner in the x_1-direction (U1) for the thick beam. (left). The corresponding zoomed plot of the displacement profiles (right) for the axial distance between 0 and 1.2 m.](image2)
The evolution of $S_{11}$ and $S_{22}$ and their values are almost the same at the center line due to the prescribed symmetry mechanical boundary conditions in both $x_1$ and $x_2$-directions.

The magnitude of the stress increase rate decreases after $T_g$ since the effective CTE is reduced after vitrification.

The corresponding contour plots of $S_{11}$ and $S_{22}$ at the end of the process are seen in Fig. 11. As expected, the $S_{11}$ distribution is almost symmetric with the $S_{22}$ distribution with respect to the diagonal of the thick beam, since all the mechanical boundary conditions are the same. The slight difference is due to the

---

**Fig. 10.** The transverse residual stress and strain evolutions at the top line (top) and at the center line (bottom) for the thick beam case.

**Fig. 11.** Undeformed contour plots of the normal stresses in $x_1$-direction ($S_{11}$, left) and $x_2$-direction ($S_{22}$, right) for the thick beam after cooling to ambient temperature. All units are in Pa.
non-uniform temperature and degree of cure evolution at the cross section (i.e. the heaters are located only at the top). The maximum compression and tensile stresses estimated for both S11 and S22 are approximately \(-16 \text{ MPa}\) and \(4.1 \text{ MPa}\), respectively.

The longitudinal and the transverse moduli at the end of the process are found to be 48.2 GPa and 12.4 GPa, respectively, which agrees well with typical values given in [44] for GFRP (glass fiber reinforced polymer) with a fiber volume fraction of 60%. In addition to that the range for the Young’s modulus of GFRP in the longitudinal direction is given as between 35 and 51 GPa [45] which is consistent with the predicted value (48 GPa) in the present work.

5.3. Results and discussions: thin beam

The displacement development of the top line is given in Fig. 12 in the \(x_2\)-direction. It is seen that up to the composite length of 0.8 m, there is no separation at the die-part interface because the die (rigid surfaces in Fig. 7) restricts the expansion of the beam even though the temperature increases. After 0.8 m from the die inlet, the shrinkage dominates and U2 decreases subsequently. In addition to that, the deformation at the end of the process is calculated approximately as \(-0.011 \text{ mm}\) for the top line which is smaller as compared to top line for the thick beam (\(-0.09 \text{ mm}\)). This is expected because for the thick beam the integration of the shrinkage strain is performed over a much longer distance than for the thin beam.

The predicted evolution of the residual stresses and strains are given in Fig. 13. Smaller process induced stress values (i.e. S11 and S22) are seen for the thin beam as compared to the thick beam.

Fig. 12. Displacement evolution of the top line in \(x_2\)-direction (U2) for the thin beam.

Fig. 13. The transverse residual stress and strain evolutions (profiles) at the top line (top) and the center line (bottom) for the thin beam case.
This is due to the fact that unlike for the thick beam, there is an almost uniform temperature and degree of cure distributions over the cross section of the thin beam (see Fig. 5), i.e. lower through-thickness gradients. In addition, the gradient of the heat flow along the thickness is very small. These promote almost no residual stresses at the end of the process. The compression stresses due to the expansion of the part can be seen clearly in Fig. 13c and d approximately up to 0.8 m where the separation takes place. After that point, similar trends are obtained for the S11 evolution as in the thick beam such that the compression and tensile residual stresses are present at the top line (Fig. 13a) and the center line (Fig. 13b), respectively at the end of the process. The variation in S11 is due to the competition between expansion and contraction of the part. However, the magnitude of S22 remains small and constant as compared to S11 at center. This is due to the fact that the ratio of the thickness and the width of the thin beam is 3.175/25.4 = 0.125 which shows that the plane stress approximation can be considered in the $x_2$-direction. Hence, there is almost no stress (S22) expected to be built-up in the plane stress direction, i.e. the $x_2$-direction (Fig. 13a and b) for both top and center lines. Contour plots of S11 and S22 are shown in Fig. 14. The maximum tensile and compression stresses are found to be approximately 0.55 MPa and –1.9 MPa, respectively for S11, 0.3 MPa and –0.2 MPa, respectively for S22. Regarding the final stiffness of the thin beam, the same effective longitudinal and transverse stiffness values are predicted as compared to the thick beam (48.2 GPa and 12.4 GPa, respectively).

6. Conclusions

In the present work, a 3D transient Eulerian thermo-chemical model was coupled with a 2D plane strain Lagrangian mechanical model of the pultrusion process. In this coupled model, a thin (1/8 in. = 3.175 mm) and a thick (1 in. = 25.4 mm) unidirectional glass/epoxy composite beam were used. For the 3D thermo-chemical simulation of the pultrusion process, the model was validated with similar models given in literature [4,6,9] using resin kinetics equation with diffusion factor. The incremental residual stress analyses of the process were performed using a 2D plane strain model based on the CHILE approach [22]. The SCFM approach from [18] was implemented for the calculation of the effective mechanical properties of the composite. In this 2D model, a mechanical contact formulation at the interface of the composite part and the heating die, which allows the separation due to shrinkage of the part, was implemented. The main outcomes of the present study are summarized as follows:

i. Non-uniform temperature and degree of cure distributions were obtained for the thick beam due to larger differential curing. On the other hand, uniform distributions are obtained for the thin beam since the top line and the center line cures almost at the same time.

ii. It was found that the curing continued at the post-die region of the pultrusion process since the temperature of the composite exiting the die was relatively high. Hence, the degree of cure increased approximately by 6% and 1% for the thick beam and the thin beam at top line, respectively at the post-die region.

iii. In the thermo-mechanical analysis, higher residual stress values were predicted for the thick beam as compared with the thin beam due to the development of higher temperature and degree of cure gradients over the thick beam cross-section. This indicates that the thickness of the composite part has an important effect on the residual stress evolution. Besides, it should be noted that the value of total volumetric shrinkage of the resin $V_{sh}$ also has a significant effect on the residual stresses.

iv. At the end of the process, tension stresses prevail for the center since it cured later and faster as compared to the outer regions where compression stresses were obtained while upholding the self static equilibrium.

v. It was found that the separation between the die and the part at top point due to the shrinkage took place after approximately 0.8 m for the thin beam depending on the initial pressure condition of the part before entering the heating die. For the thick beam, there is no separation observed at the top point until the die exit, however the corner point in Fig. 9 started separating after approximately 0.88 m from the die inlet.

vi. Furthermore, the proposed 3D/2D approach was found to be efficient and fast for the calculation of the residual stresses and distortions together with the temperature and the cure distributions.

vii. The presence of the process induced residual stresses most likely will have an important effect on the failure mechanisms of the pultruded composite profiles during in-service
loading. The pultrusion process model presented in this work has a great potential for future investigation of the process induced residual stresses and distortions of more complex pultruded profiles. More specifically, these distortions might be important for rigid polymer structures such as window frames, fencing panels etc. due to their desired high geometrical precision and the residual stresses might play a vital role for the load carrying parts such as pultruded wind turbine blade reinforcements, structural profiles in the construction industry etc.

Acknowledgements

This work is a part of DeepWind project which has been granted by the European Commission (EC) under the FP7 program platform Future Emerging Technology.

Appendix A.

The fiber mechanical properties are assumed to be transversely isotropic which is described by 5 independent elastic constants, instead of 9 for fully orthotropic materials. These 5 elastic constants are the Young’s modulus and the Poisson’s ratio in the transverse direction (E_{22} and ν_{23}) and in the longitudinal direction (E_{11} and ν_{12}) and the shear modulus in the longitudinal direction (G_{12}). The resin has an isotropic Young’s modulus (E_r), Poisson’s ratio (ν_r) and a shear modulus (G_r). Based on the fiber volume ratio (V_f), the mechanical properties of the composite are calculated in the following by using the self consistent field micromechanics (SCFM) model proposed by Boggetti and Gillespie [18].

Longitudinal Modulus:

\[
E_{11} = E_{11f}V_f + E_r(1 - V_f) + \frac{4(V_f - V_{12}^2)k_fk_rG_r(1 - V_f)V_f}{(k_f + G_r)(k_r + G_r)V_f} \quad (A.1)
\]

where k_f and k_r are the isotropic plane strain bulk modulus for fiber and resin, respectively and defined by [18]:

\[
k_f = \frac{E_{11}}{2(1 - \nu_{12}^2 - \nu_r^2)} \quad (A.2)
\]

Transverse Modulus:

\[
E_{22} = E_{33} = \frac{1}{(4k_f)^{-1} + (4G_{23})^{-1} + (V_f^2/E_{11})} \quad (A.3)
\]

where k_f is the effective plane strain bulk modulus and calculated as [18]:

\[
k_T = \frac{(k_f + G_r)k_r + (k_f - k_r)G_rV_f}{(k_f + G_r) - (k_f - k_r)V_f} \quad (A.4)
\]

Shear Modulus:

\[
G_{12} = G_{13} = G_r \left[ \frac{G_{23} + G_r}{G_{23} + G_r} + \frac{G_{12} - G_r}{G_{12} + G_r} \frac{1}{\nu_{12}} \right] \quad (A.5)
\]

\[
G_{23} = G_r \left[ (k_f + G_{23})k_r + 2G_{23}G_r + k_fG_{23} - G_rV_f \right] \quad (A.6)
\]

where

\[
G_{23} = \frac{E_{23}}{2(1 + \nu_{23})} \quad (A.7)
\]

Poisson’s ratios:

\[
\nu_{13} = \nu_{12} = \frac{1 + \nu_{12}}{1 - V_f} + \frac{(V_f - V_{12})k_rG_r(1 - V_f)V_f}{(k_r + G_r)(k_f + G_r)V_f} \quad (A.8)
\]

\[
\nu_{23} = \frac{2E_{23}k_f - E_{11}G_{23} - 4V_f^2k_fk_rG_r}{2E_{23}k_f} \quad (A.9)
\]

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“Mechanical Modelling of Pultrusion Process: 2D and 3D Numerical Approaches”

Ismet Baran, Jesper H. Hattel, Remko Akkerman, Cem C. Tutum

Mechanical Modelling of Pultrusion Process: 2D and 3D Numerical Approaches

Ismet Baran · Jesper H. Hattel · Remko Akkerman · Cem C. Tutum

Received: 21 March 2014 / Accepted: 7 April 2014 © Springer Science+Business Media Dordrecht 2014

Abstract The process induced variations such as residual stresses and distortions are a critical issue in pultrusion, since they affect the structural behavior as well as the mechanical properties and geometrical precision of the final product. In order to capture and investigate these variations, a mechanical analysis should be performed. In the present work, the two dimensional (2D) quasi-static plane strain mechanical model for the pultrusion of a thick square profile developed by the authors is further improved using generalized plane strain elements. In addition to that, a more advanced 3D thermo-chemical-mechanical analysis is carried out using 3D quadratic elements which is a novel application for the numerical modelling of the pultrusion process. It is found that the 2D mechanical models give relatively reasonable and accurate stress and displacement evolutions in the transverse direction as compared to the 3D model. Moreover, the generalized plane strain model predicts the longitudinal process induced stresses more similar to the ones calculated in the 3D model as compared with the plane strain model.

Keywords Pultrusion process · Finite element analysis · Residual/internal stress · Thermosetting resin

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Remko Akkerman Faculty of Engineering Technology, University of Twente, NL-7500AE, Enschede, The Netherlands

Cem C. Tutum Department of Electrical and Computer Engineering, Michigan State University, East Lansing, MI, USA
Nomenclature

\( \alpha \) Degree of cure
\( \alpha_{C0} \) Critical degree of cure at \( T = 0 \) K
\( \alpha_{CT} \) Constant used in the shift factor \( f(\alpha, T) \)
\( C \) Diffusion constant
\( C_{pc} \) Specific heat of the composite
\( C_{pd} \) Specific heat of the die
\( E \) Activation energy
\( E_r \) Instantaneous resin modulus
\( E_r^0 \) Initial (uncured) resin modulus
\( E_r^\infty \) Fully cured resin modulus
\( f(\alpha, T) \) Shift factor (from kinetics to diffusion region)
\( H_{tr} \) Total heat of reaction
\( k_{x1,c}, k_{x2,c}, k_{x3,c} \) Thermal conductivities in the \( x_1 \)-, \( x_2 \)- and \( x_3 \)-directions, respectively for the composite
\( k_{x1,d}, k_{x2,d}, k_{x3,d} \) Thermal conductivities in the \( x_1 \)-, \( x_2 \)- and \( x_3 \)-directions, respectively for the die
\( \lambda \) Constant used in the Di Benedetto equation
\( K_o \) Pre-exponential constant
\( n \) Order of the cure reaction (kinetic exponent)
\( q \) Heat source (internal heat generation)
\( \rho_c \) Density of the composite
\( \rho_d \) Density of the die
\( \rho_r \) Density of the resin
\( R \) Universal gas constant
\( R_r(\alpha, T) \) Reaction of cure
\( t \) Time
\( T \) Instantaneous temperature
\( T_g \) Glass transition temperature
\( T_g^0 \) Glass transition temperature of the uncured resin
\( T_{g^\infty} \) Glass transition temperature of the fully cured resin
\( T^* \) Difference between the glass transition temperature \( (T_g) \) and the instantaneous temperature \( (T) \)
\( T_{C1} \) Critical temperature at the onset of the glass transition
\( T_{C2} \) Critical temperature at the completion of the glass transition
\( u \) Pulling speed
\( V_f \) Fiber volume fraction

1 Introduction

Pultrusion is a continuous and a cost effective composite manufacturing process in which constant cross sectional profiles are produced. While pultrusion machines vary in design, the process is basically the same. Creels of unidirectional (UD) roving provide longitudinal tensile strength in the length of the profile. On the other hand, rows of continuous filament mat (CFM), woven roving or stitched fabrics provide transverse strength across the width of the profile. All reinforcements are first fed through the pre-forming guiders which start shaping the fiber reinforcements into the finished product. These reinforcements are then
pulled into a resin bath being wetted out and subsequently entering the heating die. The heaters initiate the exothermic reaction process in which the resin is being cured. The solidified and cured profile is advanced via a pulling system to the cut-off saw where it is cut to its final length. A schematic view of the pultrusion process is given in Fig. 1.

The process induced variations such as residual stresses and distortions are a critical issue in composite manufacturing [1–6], since they affect the structural behaviour and geometrical precision of the final product. More specifically, the residual stresses can lead to cracking during curing [1]. In order to capture and investigate these variations, a mechanical analysis should be performed.

In literature, thermo-chemical characteristics of the pultrusion process has been investigated numerically and experimentally [7–18] in which the temperature and degree of cure distributions inside the heating die were predicted. All these contributions have only been dealing with thermal modelling in which the temperature of the composite initially is lagging behind the heaters temperature; nevertheless during the curing it exceeds the die temperature due to the internal heat generation of the resin [7]. For this purpose, well known numerical methods such as the finite difference method (FDM) [7–10] and the finite element method (FEM) [11–13] have been utilized. Using these efficient thermo-chemical models, process optimization studies [14–16] as well as reliability analysis [17] have been performed. In [18], numerical modelling strategies for the thermo-chemical simulation of the pultrusion were investigated by the authors where the steady state approach was found to be computationally faster than the transient approach. In addition to these thermo-chemical studies in the literature, state-of-the-art models have recently been proposed by the authors for the thermo-chemical-mechanical analysis of the pultrusion [19] in which the thermo-mechanical aspects including the evolution of the process induced stresses and distortions in the transverse direction together with the mechanical properties were addressed. In this numerical model, a three dimensional (3D) transient thermo-chemical model is sequentially coupled with a 2D quasi-static plane strain mechanical model for the pultrusion process of a unidirectional (UD) fiber reinforced profile by using the FEM. The cure hardening instantaneous linear elastic (CHILE) approach [20, 21] was utilized for the resin modulus development. The proposed 3D/2D approach, which was found to be computationally efficient, provides an increased understanding of the process by evaluating the development of the stresses and distortions as well as the mechanical properties during processing. In [22], an integrated modelling of the pultrusion process of a NACA0018 blade profile was carried out by the authors using the proposed 3D/2D mechanical analysis in [19]. The calculated residual stresses were transferred to the subsequent bending simulation of the pultruded blade profile and the internal stress distribution was evaluated taking...
Table 1 Material Properties used in the thermo-chemical simulation of the pultrusion process [9]

<table>
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<tr>
<th>Material</th>
<th>$\rho$ [kg/m$^3$]</th>
<th>$C_p$ [J/kg K]</th>
<th>$k_{x_1}$ [W/m K]</th>
<th>$k_{x_2}$, $k_{x_3}$ [W/m K]</th>
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<td>Epoxy resin</td>
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<td>Glass fiber</td>
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<td>670</td>
<td>11.4</td>
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<td>7833</td>
<td>460</td>
<td>40</td>
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</tr>
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</table>

the process induced residual stresses into account.

3D thermo-chemical-mechanical analysis of the pultrusion process has not been considered in literature up to now. A novel 3D numerical simulation tool embracing the mechanical aspects of the pultrusion process is hence being developed in the present work. The temperature and degree of cure distributions at steady state are first calculated using the 3D transient thermo-chemical analysis of a pultruded square product. Afterwards, these profiles are mapped to the 2D and 3D quasi-static mechanical models. The already developed 2D plane strain mechanical model in [19] is further improved using generalized plane strain elements. Moreover, 3D quadratic brick elements are used for the 3D model for the calculation of the process induced longitudinal stresses as well as transverse stresses. In the 3D mechanical model, instead of the cross section of the part which is used in the 2D mechanical model (see Fig. 3 [19]), the entire 3D part is assumed to move along the pulling direction of the process while tracking the corresponding temperature and degree of cure profiles calculated in the 3D thermo-chemical simulation (see Fig. 4). Using these three different mechanical models (i.e. 2D plane strain, 2D generalized plane strain and 3D models), the evolution of the transient stresses and distortions are captured and the obtained results are compared with each other. The general purpose finite element software package ABAQUS [23] is utilized. The CHILE approach, which is a valid pseudo-viscoelastic approximation of the linear viscoelasticity [24], is considered for the resin modulus evolution as in [19].

Table 2 Epoxy resin kinetic parameters [9, 19]

<table>
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<tr>
<th>$H_{tr}$ [kJ/kg]</th>
<th>$K_0$ [1/s]</th>
<th>$E$ [kJ/mol]</th>
<th>$n$</th>
<th>$C$</th>
<th>$\alpha_{c0}$</th>
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<td>324</td>
<td>192,000</td>
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<td>30</td>
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</table>
Fig. 2 Schematic view of the pultrusion domain for the pultruded square beam. All dimensions are in mm

3D Transient thermo-chemical analysis
(Eulerian frame)

2D plane strain/ generalized plane strain model

2D plane strain quasi-static mechanical analysis
(Lagrangian frame)

Fig. 3 Representation of the coupling of the 3D Eulerian thermo-chemical model with the 2D Langrangian plain-strain/generalized-plane-strain mechanical model including the rigid body surfaces and the mechanical BCs
Table 3  Epoxy resin parameters used in the CHILE approach[19, 20]

<table>
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<tr>
<th></th>
<th>$T_{C1}$ [$^\circ$C]</th>
<th>$T_{C2}$ [$^\circ$C]</th>
<th>$E_r^0$ [MPa]</th>
<th>$E_r^\infty$ [MPa]</th>
<th>$\lambda$</th>
<th>$T_g^0$ [$^\circ$C]</th>
<th>$T_g^\infty$ [$^\circ$C]</th>
</tr>
</thead>
<tbody>
<tr>
<td>t3.1</td>
<td>-45.7</td>
<td>12</td>
<td>3.447</td>
<td>3.447e3</td>
<td>0.4</td>
<td>0</td>
<td>195</td>
</tr>
</tbody>
</table>

\[
\rho_d C_p d \left( \frac{\partial T}{\partial t} \right) = k_{x_1,d} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2,d} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3,d} \frac{\partial^2 T}{\partial x_3^2}
\]  

where $T$ is the temperature, $t$ is the time, $u$ is the pulling speed, $\rho$ is the density, $C_p$ is the specific heat, and $k_{x_1}$, $k_{x_2}$, and $k_{x_3}$ are the thermal conductivities along $x_1$, $x_2$, and $x_3$-directions, respectively. The subscripts $c$ and $d$ correspond to composite and die, respectively. Lumped material properties are used and assumed to be constant throughout the process [19]. The source term $q$ in Eq. 1 is related to the internal heat generation due to the exothermic resin reaction of the thermosetting epoxy resin and expressed as [11]:

\[
q = (1 - V_f) \rho_r H_{tr} R_r (\alpha, T)
\]  

where $H_{tr}$ is the total heat of reaction for the epoxy during the exothermic reaction, $\rho_r$ is the resin density, $V_f$ is the fiber volume fraction, $\alpha$ is the degree of cure, and $R_r (\alpha, T)$ is the reaction of cure which can also be defined as the rate of $\alpha$. $H_{tr}$ is calculated from the 3-D Thermo-chemical analysis (Eulerian frame) and 3-D quasi-static mechanical analysis (Lagrangian frame).

Fig. 4  Representation of the coupling of the 3D thermo-chemical model with the 3D mechanical model

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areas under the heat flow rate as a function of time obtained from the differential scanning calorimetry (DSC) tests. In literature, several kinetic models have been proposed and analyzed to describe the resin curing reactivity [25, 26]. In the present work, the well known modified $n^{th}$-order kinetic model is utilized in which the shift from a kinetics-dominated to a diffusion-dominated resin reaction near the point of vitrification (i.e. transition from rubbery to glassy state) is taken into account by a diffusion factor $f(\alpha, T)$ [19, 20, 27]. The corresponding expression is given as:

$$R_r(\alpha, T) = \frac{d\alpha}{dt} = K_\phi \exp \left( -\frac{E}{RT} \right) \left( 1 - \alpha \right)^n \cdot f(\alpha, T) \quad (4)$$

**Fig. 5** The temperature and $T_g$ distributions for center and top at the steady state (3D transient thermo-chemical approach)

**Fig. 6** The degree of cure profiles for center and top at the steady state (3D transient thermo-chemical approach)
Fig. 7 Schematic view of the movement of the 3D part in the pulling direction and the positioning of the mid section. The sizes of the die and the part are not scaled.

\[ f(\alpha, T) = \frac{1}{1 + \exp \left[ C(\alpha - (\alpha C_0 + \alpha C_T T)) \right]} \]  

where \( K_\phi \) is the pre-exponential constant, \( E \) is the activation energy, \( R \) is the universal gas constant and \( n \) is the order of reaction (kinetic exponent). \( C \) is a diffusion constant, \( \alpha C_0 \) is the critical degree of cure at \( T = 0 \) K and \( \alpha C_T \) is a constant for the increase in critical \( \alpha \) with \( T \) [20]. \( K_\phi \), \( E \), and \( n \) can be obtained by a curve fitting procedure applied to the experimental data evaluated using the DSC tests [9].

Fig. 8 Mapped temperature distributions for center and top at the mid section of the 3D part (left). The corresponding zoomed plot of the temperature profiles (right) for the axial distance between -2.5 m and 7.5 m.
Fig. 9 Mapped degree of cure distributions for center and top at the mid section of the 3D part (left). The corresponding zoomed plot of the degree of cure profiles (right) for the axial distance between -2.5 m and 7.5 m.

Fig. 10 The transient stress evolutions at center (S11 (a), S22 (b) and S33 (c)) which are calculated using the plane strain (PS), generalized plane strain (GPS) and 3D quadratic brick elements. The zoomed plot of S33 for 3D elements are depicted in (d).
The transient time integration scheme for the rate of the degree of cure can be derived by using the chain rule. Using this, the rate of the degree of cure can be expressed as:

\[
\frac{d\alpha}{dt} = \frac{\partial \alpha}{\partial t} + \frac{\partial \alpha}{\partial x_3} \frac{dx_3}{dt} = \frac{\partial \alpha}{\partial t} + u \frac{\partial \alpha}{\partial x_3}
\]  

(6)

and from Eq. 6, the relation of the resin kinetics equation can be expressed as:

\[
\frac{\partial \alpha}{\partial t} = R_r(\alpha, T) - u \frac{\partial \alpha}{\partial x_3}
\]  

(7)

where it is the expression in Eq. 7 used in the 3D transient thermo-chemical model. The equations presented above have been solved in a 3D domain by means of in-house developed routines implemented into the commercial software package ABAQUS.

Fig. 11  The transient stress evolutions at top (S11 (a), S22 (b) and S33 (c)) which are calculated using the plane strain (PS), generalized plane strain (GPS) and 3D quadratic brick elements. The zoomed plot of S33 for 3D elements are depicted in (d)
2.2 Thermo-chemical-mechanical Model

The temperature- and cure-dependent instantaneous resin modulus is defined as suggested in the CHILE approach [20]. The corresponding relation is given as:

\[
E_r = \begin{cases} 
E_r^0 & \text{for } T^* \leq T_{C1} \\
E_r^0 + \frac{T^* - T_{C1}}{T_{C2} - T_{C1}} (E_r^\infty - E_r^0) & \text{for } T_{C1} < T^* < T_{C2} \\
E_r^\infty & \text{for } T_{C2} \leq T^* 
\end{cases}
\]  \tag{8}

where \( E_r^0 \) and \( E_r^\infty \) are the initial (i.e. uncured) and fully cured resin moduli, respectively. \( T_{C1} \) and \( T_{C2} \) are the critical temperatures at the onset and completion of the glass transition, respectively, \( T^* \) represents the difference between the glass transition temperature \( T_g \) and the instantaneous resin temperature, i.e. \( T^* = T_g - T \) [20]. The evolution of the \( T_g \) with the degree of cure is modelled by the Di Benedetto equation [28] and expressed as:

\[
\frac{T_g - T_{g0}}{T_{g\infty} - T_{g0}} = \frac{\lambda \alpha}{1 - (1 - \lambda) \alpha} \tag{9}
\]

where \( T_{g0} \) and \( T_{g\infty} \) are the glass transition temperatures of uncured and fully cured resin, respectively, \( \lambda \) is a constant used as fitting parameter.

The instantaneous resin elastic modulus \( E_r \) is used in the calculation of the effective mechanical properties as well as the thermal and shrinkage strains of the composite part as reported in [19]. For this purpose, the self consistent field micromechanics (SCFM) approach is employed which is a well known and documented method in the literature to predict the effective mechanical properties of the UD composites [2, 19].

An incremental linear elastic approach is implemented utilizing user defined subroutines in ABAQUS for the calculation of the residual stresses and distortions in 2D and 3D mechanical analyses of the pultrusion process. As aforementioned, quadratic plane strain and generalized plane strain elements for the 2D analysis and brick elements for the 3D analysis are employed. The incremental total strain tensor \( \dot{\varepsilon}_{ij}^{tot} \) is composed of the incremental mechanical strain \( \dot{\varepsilon}_{ij}^{mech} \), thermal strain \( \dot{\varepsilon}_{ij}^{th} \) and chemical strain \( \dot{\varepsilon}_{ij}^{ch} \) tensors and expressed as:

\[
\dot{\varepsilon}_{ij}^{tot} = \dot{\varepsilon}_{ij}^{mech} + \dot{\varepsilon}_{ij}^{th} + \dot{\varepsilon}_{ij}^{ch} \tag{10}
\]

The incremental stress tensor \( \dot{\sigma}_{ij} \) is then calculated using the material Jacobian matrix \( \mathbf{J} \) based on the incremental mechanical strain tensor \( \dot{\varepsilon}_{ij}^{mech} \) [23] and the corresponding expression is given as:

\[
\dot{\sigma}_{ij} = \mathbf{J} \dot{\varepsilon}_{ij}^{mech} \tag{11}
\]
The details of the relation between the stress and strain tensors used in the present 2D finite element implementation can be found in [19]. The extension of the corresponding relation for the 3D mechanical analysis is presented in Eq. 12.

\[
\begin{pmatrix}
\dot{\sigma}_{11} \\
\dot{\sigma}_{22} \\
\dot{\sigma}_{33} \\
\dot{\tau}_{12} \\
\dot{\tau}_{13} \\
\dot{\tau}_{23}
\end{pmatrix}
= 
\begin{bmatrix}
J_{11} & J_{12} & J_{13} & 0 & 0 & 0 \\
J_{21} & J_{22} & J_{23} & 0 & 0 & 0 \\
J_{31} & J_{32} & J_{33} & 0 & 0 & 0 \\
0 & 0 & 0 & J_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & J_{55} & 0 \\
0 & 0 & 0 & 0 & 0 & J_{66}
\end{bmatrix}
\begin{pmatrix}
\dot{\varepsilon}_{11} \\
\dot{\varepsilon}_{22} \\
\dot{\varepsilon}_{33} \\
\dot{\gamma}_{12} \\
\dot{\gamma}_{13} \\
\dot{\gamma}_{23}
\end{pmatrix}_{mech}
\]  

(12)

where \( \dot{\tau}_{ij} \) is the incremental shear stress tensor, \( \dot{\gamma}_{ij} \) is the incremental shear strain tensor and \( J_{ij} \)'s are the elements of the material Jacobian matrix (\( J \)) [23].

### 3 Model Description

#### 3.1 Thermo-chemical Analysis

A 3D thermo-chemical analysis for the pultrusion of a relatively thick (25.4 mm) square profile is carried out in a Eulerian frame. The pultrusion model is taken from similar set-ups available in the literature [9, 11, 19]. A glass/epoxy is considered for the pultruded product in which the fiber reinforcement orientation is UD along the pulling direction and chrome steel is used for the die. Material properties and the resin kinetic parameters are listed in Table 1 and Table 2, respectively [9, 19]. Only a quarter of the pultrusion domain, seen in Fig. 2, is modelled due to symmetry. Three heating zones having prescribed set temperatures of 171-188-188°C (see Fig. 2) are defined and the spacing between these zones is defined as 15 mm [9]. The details of the cross section are also shown in Fig. 2. Perfect thermal contact is assumed at the die-part interface as in [11, 19]. Initially \( (t = 0) \), the temperature of all nodes are assigned to ambient temperature (27°C) and the degree of cure of all composite nodes are assigned to 0. For \( t > 0 \), the temperature of the composite nodes at the die inlet are set to the resin bath temperature 30°C and the degree of cure to 0, respectively. At the symmetry surfaces adiabatic boundaries are defined in which no heat flow is allowed across the boundaries. The remaining exterior surfaces of the die are exposed to ambient temperature with a convective heat transfer coefficient of 10 W/(m² K) except for the surfaces located at the heating regions. Similarly, at the post die region, convective boundaries are defined for the exterior surfaces of the pultruded square profile. The heat transfer coefficient utilized in the thermo-chemical analysis is an overall value which actually accounts for both convective and radiation heat transfer during the post-die cooling of the part. The choice of this value (within the realistic range) is not crucial for the results of the model. The length of the post die region \( L_{conv} \) is determined to be 13.7 m for cooling down to the ambient temperature after die exit. To reach steady state, the convergence limits are defined as the maximum temperature and cure degree difference between the new time step and the old time step and these are set to 0.001 °C and 0.0001, respectively.

#### 3.2 Thermo-chemical-mechanical Analysis: 2D Approach

In the 2D approach, the 3D thermo-chemical model defined in Section 3.1 is coupled with a 2D quasi-static mechanical model. Quadratic plane strain and generalized plane strain
elements are utilized separately for a comparison analysis of the process induced stress and distortion evolutions. In this 2D mechanical model, the cross section of the composite is moved through the pulling direction during the process (Lagrangian frame) meanwhile tracking the corresponding temperature and degree of cure profiles already calculated in the 3D thermo-chemical analysis (Eulerian frame). Since the length of the pultruded profile is generally much higher than the cross sectional dimensions, the plain strain assumption is convenient for the analysis of the pultrusion process [19]. For the mechanical analysis, the die is assumed to be rigid and therefore rigid body surfaces are added at the die-part interface instead of including the meshing for the whole die as was done in the thermo-chemical analysis. Between the rigid surfaces and the composite part, a mechanical contact formulation is defined which allows separation due to resin shrinkage at the interface and restricts any expansion of the composite beyond the tool interface. The friction force at the contact surfaces is assumed to be zero (sliding condition). A generic view of the plane strain model including the rigid surfaces and the mechanical boundary conditions (BCs) are shown in Fig. 3. The resin parameters used in the CHILE approach for the elastic modulus development are listed in Table 3.

3.3 Thermo-chemical-mechanical Analysis: 3D Approach

In 3D approach, the development of the process induced stresses and distortions are predicted using a 3D quasi-static mechanical model. In this model, the entire 3D composite part defined in Section 3.1 is assumed to be advanced through the pulling direction while tracking the mapped thermal and cure history. In other words, a 3D Eulerian thermo-chemical model is coupled with a 3D quasi-static Lagrangian mechanical model (see Fig. 4). As similar to the 2D approach, the die surfaces are modelled as rigid surfaces with a similar mechanical contact formulation defined in Section 3.2. Mechanical symmetry BCs are applied at the symmetry surface in the $x_1$- and $x_2$-directions. There is no

![Graph](image_url)

**Fig. 12** The process induced strain, which is the summation of the thermal strain ($\varepsilon^{th}$) and the chemical shrinkage strain ($\varepsilon^{ch}$), for center in the transverse and longitudinal direction.
applied mechanical BC in the $x_3$-direction, i.e. the part can freely move or rotate in the $x_3$-direction. The corresponding process induced stresses and distortions are calculated based on the temperature and the cure distributions together with the corresponding glass transition temperature ($T_g$) of the composite part. 3D quadratic brick elements are used in the 3D mechanical analysis. The effective mechanical properties of the pultruded part are calculated using the SCFM approach as aforementioned.

### 4 Results and Discussions

The temperature and the degree of cure profiles together with $T_g$ at steady state are first calculated in the 3D transient thermo-chemical analysis for the pultrusion of the square beam defined in Section 3.1. These results are depicted in Fig. 5 and Fig. 6. When the pultruded part enters the heating die, it follows these steady state profiles since pultrusion is a continuous process. The pulling speed is set to 20 cm/min [9, 19]. Here, “top” and “center” denote the top line and the center line through the whole length of the part, respectively, as also shown in Fig. 2. It is seen that non-uniform temperature (Fig. 5) and degree of cure (Fig. 6) distributions are obtained. The top cures earlier than the center since it is closer to the die having heaters on top of it (Fig. 2). The maximum temperature at the center is predicted as approximately $217^\circ$C which is higher than the heater temperature (i.e. $188^\circ$C)

![Undeformed contour plots of the normal stresses S11, S22 and S33 calculated using the plane strain elements at the end of the process ($x_3 \approx 14.6$ m). All units are in Pa](image-url)
due to the exothermic internal heat generation of the epoxy resin. \( T_g \) crosses the composite temperature of around 173\(^\circ\)C for the top and 182\(^\circ\)C for the center at approximately 1.6 m and 1.8 m from die inlet, respectively [19]. It is seen from Fig. 6 that at the post die region \((x_3 > 0.915 \text{ m})\) the degree of cure is increased slightly which indicates that the curing still takes place after the die exit. This fact was also observed in [8]. The degree of cure at the center is calculated to be approximately 0.97 at the end of the process (i.e. \( x_3 \approx 14.6 \text{ m} \)).

The obtained temperature and degree of cure distributions together with \( T_g \) are mapped to the 2D and 3D quasi-static mechanical analyses as described in Section 3.2 and Section 3.3. In order to reflect the 3D thermo-chemical-mechanical behaviour of the process more precisely, the mid section of the composite part is considered since the pultrusion is a continuous process, i.e. there is always existing material inside the heating die during the process. The mid section is far away from the end boundaries as seen in Fig. 7 in which a schematic view of the movement of the 3D part in the pulling direction is also depicted. It should be noted that the tracking of the mid section starts at \( x_3 \approx -7.3 \text{ m} \) and ends at \( x_3 \approx 7.3 \text{ m} \) (end of the process). At \( x_3 = 0 \text{ m} \), the mid section enters the heating die. The corresponding mapped temperature and degree of cure distributions at the mid section are shown in Fig. 8 and Fig. 9, respectively. It is seen that until the mid section enters the heating die, it is assumed that the temperature remains constant as the resin bath temperature of 30\(^\circ\)C and the material remains uncured, i.e. \( \alpha = 0 \). After entering the heating die, the mid

---

**Fig. 14** Undeformed contour plots of the normal stresses \( S_{11}, S_{22} \) and \( S_{33} \) calculated using the generalized plane strain elements at the end of the process \((x_3 \approx 14.6 \text{ m})\). All units are in Pa.
The evolution of the transient stresses are obtained using the 2D plane strain, generalized plane strain and 3D quadratic elements in ABAQUS. These results are depicted in Fig. 10 and Fig. 11 for center and top, respectively. Here, S11, S22 and S33 are the normal stresses in the $x_1$-direction (horizontal, transverse), the $x_2$-direction (vertical, transverse) and the $x_3$-direction (longitudinal), respectively. It is seen that the transverse normal stresses are found to be almost similar using both 2D and 3D approaches in terms of magnitude and evolution trend as compared with published data [19]. It is obvious that the 3D mechanical model is a more advanced and realistic numerical tool as compared to 2D models. Bearing this in mind, the plane strain assumption for the mechanical analysis of the pultrusion process gives accurate results and is a convenient way of modelling for the prediction of

Fig. 15 Undeformed contour plots of the normal stresses $S_{11}$, $S_{22}$ and $S_{33}$ for the mid section (see Fig. 7) calculated using the 3D elements at the end of the process ($x_3 \approx 7.3$ m). All units are in Pa
transverse stresses. For the 3D mechanical analysis, it is seen that until the mid section enters the die, there is almost no transverse stresses, i.e. $S_{11}$ and $S_{22}$ are zero before the die inlet. This shows that the already pulled material (e.g. the portion inside the heating die) has almost no effect on the stress development at the mid section before entering the die. Inside the die, the stress levels are relatively small because the matrix material has not enough stiffness to build up the stresses. The outer regions closest to the die cure first which make them constrained by the inner region during shrinkage. Due to this, a residual compression is found in these regions and tension prevails for the inner regions at the end of the process upholding the self static equilibrium.

Regarding the longitudinal stress development ($S_{33}$ for center in Fig. 10 and for top in Fig. 11) in the pulling or fiber direction, a different mechanical behaviour is obtained as compared to the development in the transverse directions ($S_{11}$ and $S_{22}$). It is seen that unrealistic and overestimated transient $S_{33}$ values are obtained using the plane strain elements. On the other hand “more” realistic stress values are calculated using the generalized plane strain elements since out-of-plane strain is now allowed to take place within certain restrictions. Therefore, the resulting $S_{33}$ values obtained by using the generalized plane strain elements are much closer to the values in the 3D mechanical analysis. Although the matrix material is in a liquid state before entering the die (the degree of cure is zero) for the 3D mechanical analysis, a non-zero $S_{33}$ (value of approximately 2 MPa) is found to exist at the mid section (center) while entering the die as seen in Fig. 10d. From modelling point of view, this is due to the stress gradient through the thickness in combination with a low shear modulus at the liquid state. Nevertheless, it will not appear in practice and the region before die is out of interest in terms of the mechanical variations in the processing material. As mentioned before, this 3D effect is not pronounced in the transverse directions. The longitudinal stress levels ($S_{33}$) calculated using the generalized plane strain and 3D elements are found to be relatively small as compared to $S_{11}$ and $S_{22}$ since the longitudinal component of the process induced strain is smaller than the transverse component (see Fig. 12 for the process induced strain development at center). The reason for that is the constraining fiber stiffness in the pulling direction which was also investigated in [2].

The corresponding contour plots of $S_{11}$, $S_{22}$ and $S_{33}$ at the end of the process are seen in Fig. 13 (plane strain solution), Fig. 14 (generalized plane strain solution) and Fig. 15 (3D solution). The stress distributions in the transverse directions (i.e. $S_{11}$ and $S_{22}$) over the cross section of the part have almost the same pattern for these three different types of analysis. As expected, the $S_{11}$ distribution is almost symmetric with the $S_{22}$ distribution with respect to the diagonal of the pultruded part, since all the mechanical boundary conditions are the same. The generalized plane strain results (Fig. 14) and the 3D results (Fig. 15) have the similar distribution trend for the $S_{33}$ distribution, such that tension and compression stresses are found to prevail in the pulling direction. On the other hand, the plane strain solution predicts unrealistic $S_{33}$ values such that the whole cross section is under tension showing that the self static equilibrium in the longitudinal direction is not fulfilled as expected. The reason for this is that there is no strain in the longitudinal direction for the plane strain assumption. This may not be a valid assumption for the calculation of the $S_{33}$ since there is an existing strain in the pulling direction.

The calculated maximum tension and compression stresses at the end of the process are given in Table 4. It is seen that the lowest stress levels are obtained for the 3D mechanical solution which is also intuitively expected. The maximum tension stress level is found to
Table 4 The maximum tension and compression stresses predicted at the cross section of the pultruded product using the plane strain (PS), generalized plane strain (GPS) and the 3D elements.

<table>
<thead>
<tr>
<th></th>
<th>S11 [MPa]</th>
<th>S22 [MPa]</th>
<th>S33 [MPa]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Max. tension</td>
<td>4.14 (PS)</td>
<td>4.15 (PS)</td>
<td>16.93 (PS)</td>
</tr>
<tr>
<td>3.70 (GPS)</td>
<td>3.68 (GPS)</td>
<td>2.54 (GPS)</td>
<td></td>
</tr>
<tr>
<td>3.48 (3D)</td>
<td>3.44 (3D)</td>
<td>1.74 (3D)</td>
<td></td>
</tr>
<tr>
<td>16.03 (PS)</td>
<td>16.00 (PS)</td>
<td>- (PS)</td>
<td></td>
</tr>
<tr>
<td>Max. compression</td>
<td>13.69 (GPS)</td>
<td>14.00 (GPS)</td>
<td>4.25 (GPS)</td>
</tr>
<tr>
<td>12.81 (GPS)</td>
<td>13.48 (3D)</td>
<td>3.65 (3D)</td>
<td></td>
</tr>
</tbody>
</table>

be approximately between 3-4 MPa and the maximum compression level is approximately between 12-16 MPa for S11 and S22. For S33, the levels are approximately between 1.7-2.5 MPa and 3.65-4.25 MPa, respectively, without considering the plane strain solution since it is overestimating S33.

The evolution of the displacement profile is also predicted using the devised numerical simulation tools for the top in the $x_2$-direction ($U_2$). The results are depicted in Fig. 16. It is found that there is a good match between the $U_2$ evolutions obtained using the three different element types. It should be noted that the initial pressure condition of the part before entering the heating die is not taken into account which may affect the stress and displacement field.

---

Fig. 16 Displacement evolution for top in the $x_2$-direction ($U_2$) calculated using the plane strain (PS), generalized plane strain (GPS) and 3D elements (mid section for the 3D model)
5 Conclusions

In the present work, the 2D quasi-static plane strain mechanical model developed by the authors for the pultrusion process is further improved using the generalized plane strain elements. In addition, 3D quadratic brick elements are also utilized for the 3D thermo-chemical-mechanical model for the calculation of the longitudinal stresses which has not been considered up to now in the field of numerical modelling of the pultrusion process. The temperature and degree of cure distributions at steady state are first calculated using a 3D transient thermo-chemical analysis. Afterwards, these profiles are mapped to the 2D and 3D quasi-static mechanical models. Using these three different mechanical models (i.e. 2D plane strain, 2D generalized plane strain and 3D models), the development of the process induced stresses and distortions are predicted for the pultrusion of a UD glass/epoxy square profile. The obtained results are compared with each other. The main outcomes are summarized as follows:

i) It is found that there is a good agreement between the calculated transverse stress evolutions (S11 and S22) together with the transverse displacement profiles (U2) using the 2D and 3D mechanical models. This shows that the plane strain assumption is a convenient and fast way of predicting the transverse stresses and displacements.

ii) For the calculation of the longitudinal stresses (S33), generalized plane strain and 3D elements predict “more” realistic stress values as compared to the plane strain elements in which overestimated S33 values are obtained.

iii) The predicted stress level for S33 is found to be lower than the S11 and S22 since the longitudinal component of the process induced strain is smaller than the transverse component.

iv) Lower stress values are obtained in the 3D mechanical analysis as compared with the 2D analyses which is intuitively expected.

The proposed 3D/3D thermo-chemical-mechanical model is found to be more suitable for advanced modelling of the pultrusion process, e.g. taking the effect of the frictional force inside the heating die on the longitudinal process induced stress levels into account. On the other hand, the 2D generalized plane strain mechanical model is found to be computationally fast as compared with the 3D model and is therefore more efficient and useful for internal stress investigation in the transverse directions of the pultruded products, e.g. the transverse residual stresses at the web-flange junctions of pultruded I-beam profiles or transverse warpage in pultruded hollow rectangular profiles.

6 Acknowledgements

This work is a part of the DeepWind project which has been granted by the European Commission (EC) under the FP7 program platform Future Emerging Technology.

References

“Investigation of Process Induced Warpage for Pultrusion of a Rectangular Hollow Profile”

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Submitted for publication, 2014.
Investigation of Process Induced Warpage for Pultrusion of a Rectangular Hollow Profile

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Abstract

A novel thermo-chemical-mechanical analysis of the pultrusion process is presented. A process simulation is performed for an industrially pultruded rectangular hollow profile containing both unidirectional (UD) roving and continuous filament mat (CFM) layers. The reinforcements are impregnated with a commercial polyester resin mixture (Atlac 382). The reactivity of the resin is obtained from gel tests performed by the pultruder. The cure kinetics parameters are estimated from a fitting procedure against the measured temperature. The cure hardening instantaneous linear elastic (CHILE) model is adopted for the evolution of the resin elastic modulus using the temperature-dependent elastic response provided by the resin supplier. The numerical model predictions for the warpage trend at the end of the process are found to agree well with the warpage observed in the real pultruded products. In addition, the magnitude of the warpage is found to be in the measured range.

Keywords: B. Cure behaviour; B. Thermomechanical; C. Computational modelling; E. Pultrusion.

1. Introduction

Pultrusion is a continuous and a cost effective composite manufacturing process in which constant cross sectional profiles are produced. While pultrusion machines vary in design, the process is essentially same. All reinforcements are first fed through the pre-forming guiders which start shaping the fiber reinforcements into the finished product. These reinforcements are wetted out in a resin bath and subsequently enter the heating die. The heaters initiate the exothermic chemical reaction during which the resin is being cured. The cured profile is advanced via a pulling system to the cut-off saw where it is cut to its final length. A schematic view of the pultrusion process is given in Fig. 1.

In general, industrially pultruded parts contain both unidirectional (UD) roving and continuous filament mat (CFM) layers impregnated by a thermosetting resin. The UD roving provides longitudinal tensile strength in the length of the profile. On the other hand, the CFM provides transverse tensile strength in the length of the profile. On the other hand, the CFM provides transverse tensile strength in the length of the profile. A UD layer is transversely isotropic (TI), whereas the CFM layer can be considered as quasi isotropic (QI) since
it consists of long swirled fibers randomly oriented in the plane of the mat. Therefore, the CFM layer has equal material properties in the in-plane directions [1] and the out-of-plane properties are different than the in-plane properties [2].

Among the matrix materials used in the pultrusion industry, polyester and epoxy resins are some of the most common materials. These two types of resin system behave differently in terms of curing dynamics. Both systems have inherent characteristics such as chemical shrinkage and reactivity which are crucial for the pultrusion. The polyester resin is more reactive than the epoxy and hence higher pulling speeds can be used for the pultrusion of a polyester based composites [3]. Moreover, the gelation occurs at lower conversion rates or degree of cure values for the polyester as compared to the epoxy and the volumetric shrinkage varies between 6-12% for the polyester resins. This value can be further decreased to 2% by mixing the unsaturated polyester with “low profile” or “shrink-reducing” additives [3].

The process induced mechanical variations are generated by various mechanisms inherently existing in composite manufacturing processes such as chemical shrinkage of the thermosetting resin and mismatch in the coefficient of thermal expansion (CTE) of the reinforcement and the resin [4–6]. In addition, the UD and the CFM layers in pultrusion also have different curing kinetics and mechanical responses which result in unwanted residual deformations. Therefore, the evolution of the process induced distortions must be well understood in order to have a better control of the mechanical behaviour of the pultruded profiles. Since running a production line by trial and error is an expensive and time consuming task, the development of a numerical simulation tool to predict the process induced deformations in terms of warpage and spring-in is highly required.

Several thermo-chemical analyses of the pultrusion have been carried in literature [7–18] in which the temperature and the degree of cure distributions inside the heating die were predicted. All these contributions have only been dealing with thermal modelling of pultruded UD profiles from which the temperature and the degree of cure distributions inside the part were calculated. In addition to these thermo-chemical studies in the literature, state-of-the-art process models based on a thermo-chemical-mechanical analysis of the pultrusion for UD profiles have recently been proposed by the authors [19, 20]. The development of the process induced stresses and distortions were specifically addressed in [19] in which a three dimensional (3D) transient thermo-chemical model was sequentially coupled with a 2D quasi-static plane strain mechanical model. The proposed model in [19] was found to be computationally fast for the calculation of the process induced deformations in the transverse directions.

Modelling the pultrusion process containing both UD and CFM layers has not been considered in the literature up to now. A numerical simulation tool embracing the thermo-chemical and mechanical aspects of the pultrusion for an industrial rectangular hollow profile is hence being developed in the present work. Two different micromechanics approaches are used to calculate the instantaneous mechanical properties of the UD and the CFM layers.
The Atlac 382 commercial polyester resin [21] is used to impregnate the reinforcements. The resin reactivity is obtained from gel test experiments performed by a commercial pultruder and a cure kinetics model is developed based on the test data. The cure kinetics model is consequently used in the 3D thermo-chemical pultrusion simulation to calculate the temperature and the degree of cure distributions in the part. The cure hardening instantaneous linear elastic (CHILE) model [22] is adopted for the evolution of the resin elastic modulus which is temperature- and degree of cure-dependent. The coefficients used in the CHILE model are fit to the temperature-dependent elastic response provided by the resin supplier [21]. The evolution of the distortions as well as the residual warpage formation is predicted using a 2D quasi-static mechanical model developed in the general purpose finite element software ABAQUS [23]. The warpage pattern is also measured in the real pultruded products and the predicted warpage magnitude is compared with the measured data.

2. Numerical Implementation

2.1. Energy and Cure Kinetics Equations

In the present work, a 3D thermo-chemical model is used to calculate the temperature and degree of cure for the pultruded part. Since pultrusion is a continuous equilibrium-based process, the part entering the heating die keeps tracking the temperature and degree of cure profiles at steady state. Therefore, the steady state approach [18], which is convenient for the pultrusion process, is utilized. The die is also included in this thermo-chemical model. The steady state energy equations are solved simultaneously in Cartesian coordinates for the UD layer (Eq. 1), the CFM layer (Eq. 2) and the die (Eq. 3). Here, $x_1$ is the pulling or longitudinal direction; $x_2$ and $x_3$ are the transverse directions for the UD layer. On the other hand, $x_1$ (pulling direction) and $x_2$ are the in-plane directions and $x_3$ is the out-of-plane direction for the CFM layer.

$$
\frac{(\rho C_p)_{UD}}{} \left( u \frac{\partial T}{\partial x_1} \right) = k_{x_1,UD} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2,UD} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3,UD} \frac{\partial^2 T}{\partial x_3^2} + q_{UD} \tag{1}
$$

$$
\frac{(\rho C_p)_{CFM}}{} \left( u \frac{\partial T}{\partial x_1} \right) = k_{x_1,CFM} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2,CFM} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3,CFM} \frac{\partial^2 T}{\partial x_3^2} + q_{CFM} \tag{2}
$$

$$
0 = k_{x_1,d} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2,d} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3,d} \frac{\partial^2 T}{\partial x_3^2} \tag{3}
$$

where $T$ is the temperature, $u$ is the pulling speed, $\rho$ is the density, $C_p$ is the specific heat and $k_{x_1}$, $k_{x_2}$ and $k_{x_3}$ are the thermal conductivities along $x_1$-, $x_2$- and $x_3$-directions, respectively. The subscripts $UD$, $CFM$ and $d$ correspond to the UD layer, the CFM layer and the die, respectively. Lumped material properties are used and assumed to be constant.
The source term \( q \) in Eq. 1 and Eq. 2 are related to the internal heat generation due to the exothermic reaction of the polyester resin and expressed as [11]:

\[
q_{UD} = (1 - V_f) UD \rho_r H_{tr} R_r(\alpha, T) \quad (4)
\]

\[
q_{CFM} = (1 - V_f) CFM \rho_r H_{tr} R_r(\alpha, T) \quad (5)
\]

where \( H_{tr} \) is the total heat of reaction for the polyester during the exothermic reaction, \( \rho_r \) is the resin density, \( V_f \) is the fiber volume fraction, \( \alpha \) is the degree of cure, and \( R_r(\alpha, T) \) is the reaction of cure which can also be defined as the rate of \( \alpha \), i.e. \( d\alpha/dt \). In literature, several kinetic models have been proposed and analyzed to describe the resin curing reactivity [24, 25]. In the present work, a well known semi-empirical autocatalytic model [22] which is an Arrhenius type of equation is utilized. The corresponding expression is given as:

\[
R_r(\alpha, T) = \frac{d\alpha}{dt} = A_0 \exp\left(\frac{-E_a}{RT}\right) \alpha^m (1 - \alpha)^n \quad (6)
\]

where \( A_0 \) is the pre-exponential constant, \( E_a \) is the activation energy, \( R \) is the universal gas constant and \( m \) and \( n \) are the orders of reaction (kinetic exponents).

The material derivative of the degree of cure field (Eq. 6) is translated into a partial derivative form in a Eulerian frame of reference in the pulling direction and expressed as [19]:

\[
R_r(\alpha, T) = \frac{d\alpha}{dt} = \frac{\partial \alpha}{\partial t} + \frac{\partial \alpha}{\partial x_1} \frac{dx_1}{dt} = \frac{\partial \alpha}{\partial t} + u \frac{\partial \alpha}{\partial x_1} \quad (7)
\]

and from Eq. 7, the relation of the resin kinetics equation for the steady state approach (i.e. \( \partial \alpha/\partial t = 0 \)) can be expressed as:

\[
0 = R_r(\alpha, T) - u \frac{\partial \alpha}{\partial x_1} \quad (8)
\]

which is used in the 3D steady state thermo-chemical model.

The equations above are solved using the commercial finite element code in ABAQUS. The evaluation of the degree of cure and the reaction rate has been obtained by means of in-house developed routines implemented in ABAQUS [19].

2.2. Thermo-chemical-mechanical Model

The state of the polyester resin changes from liquid to solid during processing, with the rubbery zone in between. In order to capture the effect of this transition mechanically, the
temperature- and cure-dependent instantaneous resin modulus is defined as proposed in the CHILE approach [22]:

$$E_r = \begin{cases} 
E^0_r & T^* < T_{C1} \\
E^0_r + \frac{T^* - T_{C1}}{T_{C2} - T_{C1}} (E^\infty_r - E^0_r) & \text{for } T_{C1} < T^* < T_{C2} \\
E^\infty_r & T_{C2} \leq T^* 
\end{cases}$$

(9)

where $E^0_r$ and $E^\infty_r$ are the uncured and fully cured resin moduli, respectively. $T_{C1}$ and $T_{C2}$ are the critical temperatures at the onset and completion of the glass transition, respectively and $T^*$ represents the difference between the instantaneous glass transition temperature ($T_g$) and the resin temperature, i.e. $T^* = T_g - T$ [22]. The evolution of $T_g$ as a function of degree of cure is modelled by the Di Benedetto equation [26] and expressed as:

$$\frac{T_g - T_{g0}}{T_{g\infty} - T_{g0}} = \frac{\lambda \alpha}{1 - (1 - \lambda) \alpha}$$

(10)

where $T_{g0}$ and $T_{g\infty}$ are the glass transition temperatures of uncured and fully cured resin, respectively and $\lambda$ is a constant used as a fitting parameter.

The chemical shrinkage of the resin is expressed via the total volumetric shrinkage ($V_{sh}$) as explained in the following. Assuming a uniform contraction for a unit cell in the resin, the isotropic incremental resin shrinkage strain ($\dot{\varepsilon}_r$) is calculated as [27]:

$$\dot{\varepsilon}_r = \sqrt{1 + \Delta V_r} - 1$$

(11)

where $\Delta V_r$ is the incremental specific volume shrinkage of the resin expressed as a function of change in the degree of cure ($\Delta \alpha$) and $V_{sh}$ [27]:

$$\Delta V_r = \Delta \alpha \cdot V_{sh}$$

(12)

The effective mechanical properties as well as the thermal and chemical strains for the UD layer are calculated using the self consistent field micromechanics (SCFM) approach which is a well known and documented technique in the literature [27]. On the other hand, the effective mechanical properties of the QI-CFM layer is calculated considering the material properties of the UD layer obtained by the SCFM approach for the same $V_f$ as the QI layer [2]. The details of the calculations for the QI laminate can be found in [2].

An incremental linear elastic approach is implemented utilizing the user defined subroutines in ABAQUS to calculate the displacements and stresses. For this purpose, quadratic generalized plane strain elements are considered [23]. The incremental total strain ($\dot{\varepsilon}_{tot}$) which is composed of the incremental mechanical strain ($\dot{\varepsilon}_{mech}$), thermal strain ($\dot{\varepsilon}_{th}$) and chemical strain ($\dot{\varepsilon}_{ch}$) due to the chemical shrinkage of the epoxy resin, is expressed as:

$$\dot{\varepsilon}_{ij}^{tot} = \dot{\varepsilon}_{ij}^{mech} + \dot{\varepsilon}_{ij}^{th} + \dot{\varepsilon}_{ij}^{ch}$$

(13)
The incremental stress tensor \( \dot{\sigma}_{ij} \) is calculated using the material Jacobian matrix (\( J \)) based on the incremental mechanical strain tensor \( \dot{\varepsilon}_{ij}^{\text{mech}} \) \cite{23} and the corresponding expression is given as:

\[
\dot{\sigma}_{ij} = J \dot{\varepsilon}_{ij}^{\text{mech}} \tag{14}
\]

The details of the relations between the stress and strain tensors used in the present finite element implementation can be found in \cite{19}.

3. Polyester Resin Reactivity

3.1. Experimental Tests

The reactivity of the polyester resin (Atlac 382 \cite{21}) used in the pultrusion process is determined by the commercial pultruder according to tests similar to the standard test method given by ASTM International \cite{30}. This method provides a guidance for measurement of the “Standard 82.2 °C Exotherm Curve” from which the performance of the resin can be predicted when used during elevated temperatures.

In this test, the neat unsaturated polyester resin is mixed with the initiators appropriate for the pultrusion in a circular metal cup at ambient temperature (\( T_{\text{amb}} \)) 25 °C. This mixture is then placed in a water bath in which the water temperature is kept constant at 82.2 ±0.5 °C. The heat accelerates the polymerization reaction and the exotherm is measured using a thermocouple as the resin mixture cures. It should be noted that this is the only available data provided by the pultruder regarding the curing kinetics of the polyester. A schematic view of this test and the location of the thermocouple are shown in Fig. 2. Note that the measurements are carried out at the center of the sample.

3.2. Cure Kinetics Model

The polyester reactivity is numerically simulated considering the test setup given in Fig. 2. In this simulation, an axisymmetric thermo-chemical model is used in which the transient heat conduction equation in the cylindrical coordinate system is solved together with the resin kinetics equation as in \cite{7, 15}. For this reactivity simulation, the alternating direction implicit (ADI) method \cite{7}, which is unconditionally stable in 2D, is utilized in the MATLAB computing environment \cite{31}. A schematic view as well as the boundary conditions (BCs) of the 2D model for the reactivity test are shown in Fig. 3. The resin temperature is assumed to be equal to the water temperature (\( T_{\text{water}} = 82.2 \) °C) at the circular metal cup boundaries (bottom and right BCs in Fig. 3). The top surface of the resin is assumed to be exposed to \( T_{\text{amb}} \) via a convective heat transfer coefficient (\( h_{\text{conv}} \)) of 10 W/m²-K. At \( t = 0 \) s, the temperature of the resin is equal to \( T_{\text{amb}} \) and the initial degree of cure is taken as 0. The simulations are performed for a total time of 1000 s since the experiments take approximately 900 s.
The cure kinetics parameters used in Eq. 6 together with $H_{tr}$ in Eq. 4 are estimated by performing a fitting analysis using the proposed 2D axisymmetric thermo-chemical model (Fig. 3) such that the numerical model predicts the measured temperature as well as the corresponding degree of cure at the specified thermocouple location. For this purpose, the constrained minimization function “fmincon” in MATLAB is employed which is a gradient-based mathematical programming algorithm. The objective is to minimize the sum of the square of difference between the measured and the calculated temperature, i.e. $\sum (T_{\text{meas}} - T_{\text{cal}})^2$. In addition to the peak temperature and the corresponding time, the degree of cure around the gelation point is also taken into account as a constraint in the fitting analysis. The gel time is defined as the period of time required for the temperature reaching 5.5 °C above the bath temperature (i.e. 87.7 °C) as described in [30]. The thermal properties of the polyester resin used in the numerical model are given in Table 1.

Fig. 4 depicts the best fit of the temperature development to the measured temperature and the corresponding cure degree at the specified thermocouple location. A good agreement between the temperature curves is observed. The estimated optimum cure kinetics parameters providing the temperature profile in Fig. 4 are given in Table 2. These values are found to be reasonable for the polyester resin as compared to the available data in the literature (see Table 2). The required time for reaching the peak exotherm (173 °C) is found to be approximately 734 s. A sharp increase in temperature of 82 °C is observed within approximately 80 s (from 654 s to 734 s) due to the exothermic reaction. This indicates that the resin mixture is highly reactive which is a desired resin performance for pultrusion. The corresponding degree of cure (Fig. 4(right)) is estimated using the parameters in Table 2. The predicted (and the measured) gel time is found to be approximately 554 s where the temperature is around 87.7 °C and at that time the degree of cure is calculated as 0.23 which is in the range of the conversion rate at gelation (0.1-0.3) for the polyester resin [3].

4. Pultrusion Model

4.1. Thermo-chemical Analysis

The 3D thermo-chemical analysis implemented in ABAQUS is used to simulate the pultrusion process of a rectangular hollow profile. A glass/polyester is considered for the UD and the CFM layers and steel is used for the die. The details of the process set-up is taken from a commercial pultrusion company. A schematic representation of the model is seen in Fig. 5. Only a quarter model is used due to symmetry conditions. The length of the die and the mandrel are specified as 1 m and 0.7 m, respectively, by the pultruder. Note that the mandrel has no heating elements. The length of the profile at the post-die region is taken as 5 m. According to production trials, it was found that the heater temperature close to the die exit should be 130°C which is lower than the heater temperature close to die inlet(140°C). The reason for that is to prevent an unwanted excess temperature in the part near die exit. In the thermo-chemical model, a perfect thermal contact is assumed at the die-part and mandrel-part interface. The employed thermal properties are listed in Table
1. As aforementioned, the estimated resin kinetic parameters in Table 2 are used in the thermo-chemical model of the pultrusion.

The cross sectional details including the meshing are given in Fig. 6. It is seen that the dimension of the processing hollow rectangular profile is 64×27×3 mm. A local material orientation is employed for the CFM layer as seen in Fig. 6 in order to reflect the in-plane ($x_1$- and $x_2$-) and the out-of-plane ($x_3$-) properties correctly. For the round corner, a cylindrical local coordinate system is used in which the $x_2$- and $x_3$-directions are taken as the tangential (T) and radial (R) directions, respectively. A CFM having a density of 450 g/m$^2$ is used. The overall fiber weight fraction ($W_f$) of the profile is determined to be approximately 60% according to weight measurements performed by the pultruder. Accordingly, the fiber volume fractions ($V_f$) are calculated as 56.5% and 23% for the UD layer and the CFM layers, respectively. The thickness of the pultruded CFM layers is assumed to be 0.762 mm according to the data given in [3]. To justify this, the layer configuration of a similar commercially pultruded product has been investigated in micro level. The details of the investigated product is shown in Fig. 6. It is observed that the thickness of the CFM layer varies between 0.6-0.9 mm having a mean value of approximately 0.75 mm which is very close to the thickness value (0.762 mm) given in [3]. Hence, 0.762 mm is used for the thickness of the CFM layer in the present work.

The composite temperature and degree of cure at the die inlet are set to the resin bath temperature 25 °C and 0, respectively, as a BC. Prescribed heater temperatures are applied at the heating regions shown in Fig. 5. At the symmetry surfaces adiabatic boundaries are defined in which no heat flow is allowed across the boundaries. The remaining exterior surfaces are exposed to ambient temperature with a convective heat transfer coefficient of 10 W/m$^2$-K except for the surfaces located at the heating regions.

4.2. Quasi Static Mechanical Analysis

The 3D thermo-chemical model defined in Section 4.1 is coupled with a 2D quasi-static mechanical model in which quadratic generalized plane strain elements are utilized in ABAQUS. In this 2D mechanical model, the cross section of the composite is moved through the pulling direction during the process meanwhile tracking the corresponding temperature and degree of cure profiles already calculated in the 3D thermo-chemical analysis. A generic representation of this sequential coupling procedure is shown in Fig. 7. Since the length of the pultruded profile is generally much larger than the cross sectional dimensions, a plain strain assumption is convenient for the mechanical analysis of the pultrusion [19]. The die and the mandrel are also included in the 2D mechanical model (see Fig. 6) in which a mechanical contact formulation is defined at the die-part and mandrel-part interface. By using this contact formulation, a separation due to the thermal contraction of the part and/or the resin shrinkage at the interface is allowed and any expansion of the composite beyond the tool interfaces is restricted. The friction force at the contact surfaces is assumed to be zero (sliding condition). Symmetry mechanical BCs are applied at the symmetry lines and the die is assumed to be fixed at the outer regions as shown in Fig. 6. Local material orientation
is employed for the CFM layer as described in Section 4.1.

The constituent mechanical properties of the glass fiber (E-glass) and the polyester resin (Atlac 382) are summarized in Table 3. It should be noted that the resin CTE in rubbery state \((T_g < T)\) is known to be approximately 2.5 times larger than the CTE in glassy state \((T_g > T)\) [6, 26]. The coefficients used in the CHILE model (Eq. 9) are fit to the fully cured temperature-dependent elastic response (normalized) provided by the resin supplier [21]. The cure- and temperature-dependent resin modulus overlaid the elastic response provided by the resin supplier, as shown in Fig. 8. A good agreement between the predicted resin modulus and the data from supplier is noted. The estimated material properties used in the CHILE approach are given in Table 4. The ultimate \(T_g\) is set to 147 °C [21] and the \(T_{g0}\) is assumed to be 0 °C in Eq. 10. The fitting parameter in Eq. 10 is taken as \(\lambda = 0.4\) which is the expected value for thermosetting resins [26]. A parametric study is carried out based on the total volumetric shrinkage \(V_{sh}\) used in Eq. 12 since the corresponding value is not available for Atlac 382 polyester. The \(V_{sh}\) range given in [21] is considered and six different values are selected for \(V_{sh}\) as 0.02, 0.04, 0.06, 0.08, 0.10 and 0.12.

5. Results and Discussions

The predicted temperature and degree of cure evolutions are depicted in Fig. 9 and Fig. 10, respectively, at the die-part (point A) and mandrel-part (point B) interface. The corresponding contour plots are shown in Fig. 11 for the profile cross section at the mandrel exit \((x_1 = 0.7 \, \text{m})\), die exit \((x_1 = 1 \, \text{m})\) and end of the process \((x_1 = 6 \, \text{m})\). The pulling speed is set to 650 mm/min for the pultrusion of the rectangular hollow profile.

It is seen from Fig. 9 that the composite temperature at the die-part interface (point A) is highly dominated by the prescribed heater temperatures. Hence, the temperature at point A remains almost the same as the heater temperatures. On the other hand, the internal heat generation plays a more significant role at the inner regions (e.g. point B at the mandrel-part interface) and therefore the temperature at the inner regions is found to be higher than the prescribed heater temperatures (140-130 °C). This can easily be seen from the contour plot in Fig. 11. The maximum temperature of the part is found to be approximately as 192.5 °C. The degree of cure development seen in Fig. 10 also shows a similar trend as the temperature such that curing starts earlier at point A than point B. The rate of curing is higher at point B owing to the dominant heat generation and therefore the degree of cure at point B becomes higher than point A near the mandrel end \((x_1 = 0.7 \, \text{m})\). This can also be seen from Fig. 11 in which the inner surface of the pultruded profile has a degree of cure value of approximately 0.86 near the mandrel end which is higher than the degree of cure value for the outer regions. However, it is found that the part is almost cured near the die exit (see Fig. 11) with a mean degree of cure value of approximately 0.96. The degree of cure keeps increasing at the post die region indicating that the curing still takes place after the die exit since the temperature of the part is still high enough to
promote the exothermic chemical reaction. As a consequence the mean degree of cure of the cross section increases to approximately 0.99 at the end of the process (Fig. 11). The part cools down to a temperature of approximately 56 °C at the end of the process due to the convective cooling at the inner (after mandrel end) and outer (after die exit) regions.

The predicted vertical displacement development for point A (die-part interface) in the $x_3$-direction is depicted in Fig. 12 for various $V_{sh}$ values ($V_{sh} = 0.02, 0.04, 0.06, 0.08, 0.10$ and 0.12). The trend of the displacement development is found to be almost the same for different $V_{sh}$ values. However, the magnitude of the displacement decreases with a decrease in $V_{sh}$ which is expected. The detachment at the die-part interface is captured using the mechanical contact formulation at the interface as seen from Fig. 12. The part separates from the die due to the chemical shrinkage. The detachment point where the displacement starts being negative shifts left towards the die inlet as the $V_{sh}$ increases.

The deformed contour plots in Fig. 13 shows the predicted displacement field in the $x_3$-direction for $V_{sh}$ values of 0.02, 0.06 and 0.10 at the end of the process. The deformation or warpage pattern is found to be almost the same for various $V_{sh}$ values, however the magnitudes are different as aforementioned. The predicted warpage patterns (Fig. 13) match well with the warpage observed in the real pultruded rectangular hollow products seen in Fig. 13(right). The magnitude of the warpage ($w$) at point A is defined as the difference between the magnitude of the displacements at point A ($w_a$) and point C ($w_c$) seen in Fig. 13(left), i.e. $w = w_a - w_c$. The warpage magnitudes of the pultruded products are also measured at the room temperature. The predicted and the measured warpage values are given in Table 5. A good agreement is found between the predicted and the measured values such that the predicted warpage value for $V_{sh}$ of 0.10 and 0.12 are in the range of measured values. It should be noted that the warpage values given in Table 5 are predicted at the end of the process (i.e. $x_1 = 6$ m) where the temperature of the part is approximately 56°C (see Fig. 11). In fact the predicted warpage magnitudes are expected to be slightly higher than the values given in Table 5 since there will be further contraction as the part cools down to room temperature of 25°C.

The predicted effective mechanical properties (glassy state) for the pultruded product are given in Table 6 and Table 7 for the UD layer ($V_f = 56.5\%$) and CFM layer ($V_f = 23\%$), respectively. Here, $E$ is the elastic modulus, $G$ is the shear modulus and $\nu$ is the Poisson’s ratio. These values are compared with another predicted values obtained from the University of Twente’s micromechanics model (“Micmec”) [35] and the available measured data (only for the CFM layer) [21]. It is seen that the predicted values using the SCFM approach agree well with the values obtained from “Micmec”. The predicted in-plane elastic modulus ($E_1 = E_2$) and CTE ($\alpha_1 = \alpha_2$) of the CFM layer are found to be very close to the measured values.
6. Conclusions

A thermo-chemical-mechanical analysis was performed for the pultrusion of an industrial rectangular hollow profile having both UD and CFM layers. The UD rovings and the CFM were impregnated with a polyester based resin mixture (Atlac 382). The reactivity of the resin was obtained from the gel tests performed by the pultruder. A numerical model was developed to fit the curing kinetics parameters for the polyester resin to the measured temperature data provided by the pultruder. The temperature- and cure-dependent resin modulus was utilized using the CHILE approach. The coefficients used in the CHILE model were fit to the temperature-dependent elastic response provided by the resin supplier [21].

A parametric study was performed based on the $V_{sh}$ value varying between 0.02-0.12. The displacement evolutions and the detachment points were calculated. The detachment point shifts towards die inlet as the $V_{sh}$ increases. The numerical model predictions for the process induced warpage formation at the end of the process were found to agree well with the warpage observed in the real pultruded products. In addition, the calculated magnitudes of the warpage for $V_{sh}$ of 0.10 and 0.12 were found to be in the measured range. This indicates that the proposed numerical simulation tool estimates the mechanical response of this specific process correctly. Moreover, the calculated effective mechanical properties at glassy state were compared with the available measured data (only for the CFM layer) and with the “Micmec”. Good agreement was obtained.

7. Acknowledgements

The author wishes to thank Dr. Joao Pedro Nunes and David Melo from University of Minho (Portugal) for guidance and valuable discussions of the experiments. This work is a part of DeepWind project which has been granted by the European Commission (EC) under the FP7 program platform Future Emerging Technology.

References


Figure 1: Schematic view of a pultrusion process.

Figure 2: Schematic view of the experimental set-up for the polyester reactivity tests carried out in the pultrusion company.
Figure 3: Schematic view of the axisymmetric thermo-chemical model for the estimation of the polyester resin reactivity.

Figure 4: The predicted best fit and the measured temperature (left) and cure degree (right) development during the reactivity test for the polyester resin.
Figure 5: Schematic view of the quarter pultrusion domain for the pultruded rectangular hollow profile. All dimensions are in mm.

Figure 6: The cross sectional details of the pultruded profile, die and mandrel used in the pultrusion process model. All dimensions are in mm.
3D Thermo-chemical analysis (Eulerian frame)  

2D Generalized plane strain quasi-static mechanical analysis (Lagrangian frame)  

Figure 7: Representation of the sequential coupling of the 3D thermo-chemical model with the 2D generalized plain-strain mechanical model.

Figure 8: Normalized elastic modulus of fully cured resin (Atlac 382) compared with the temperature- and cure-dependent resin modulus model given in Eq. 9 for various degree of cure values (α).
Figure 9: Steady state temperature evolution predicted at point A and B (left) and the corresponding zoomed plot inside the die (0-1 m) (right).

Figure 10: Steady state degree of cure evolution predicted at point A and B (left) and the corresponding zoomed plot inside the die (0-1 m) (right).
Figure 11: Contour plots showing the temperature (left) and degree of cure (right) distributions over the cross section at mandrel exit ($x_1 = 0.7$ m), die exit ($x_1 = 1$ m) and end of the process ($x_1 = 6$ m). Note that the legends of the plots are not same.

Figure 12: The displacement evolution at point A (die part interface) in the $x_3$-direction (left) and the corresponding zoomed plot inside the die (0-1 m) (right) for various $V_{sh}$ values.
Figure 13: Deformed contour plots of the residual displacement field in the $x_3$-direction and the initial geometry of the cross section (undeformed frame) (left). Scale factor for the deformed shape is 10. The warpage formation observed in the real pultruded hollow rectangular profiles (right).

Table 1: Thermal properties used in the thermo-chemical model [9, 21, 32].

<table>
<thead>
<tr>
<th>Material</th>
<th>$\rho$ [kg/m$^3$]</th>
<th>$C_p$ [J/kg-K]</th>
<th>$k_x$, $k_{x_2}$ [W/m-K]</th>
<th>$k_{x_2}$, $k_{x_3}$ [W/m-K]</th>
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<tr>
<td>Polyester</td>
<td>1100</td>
<td>1830</td>
<td>0.17</td>
<td>0.17</td>
</tr>
<tr>
<td>Glass fiber</td>
<td>2560</td>
<td>670</td>
<td>11.4</td>
<td>1.04</td>
</tr>
<tr>
<td>Steel die</td>
<td>7833</td>
<td>460</td>
<td>40</td>
<td>40</td>
</tr>
</tbody>
</table>
Table 2: The estimated cure kinetics parameters of polyester and the corresponding parameters taken from literature.

<table>
<thead>
<tr>
<th></th>
<th>(A_0) [1/s]</th>
<th>(E_a) [J/mol]</th>
<th>(m)</th>
<th>(n)</th>
<th>(H_{tr}) [kJ/kg]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Present Work</td>
<td>(1.895 \times 10^8)</td>
<td>74,967</td>
<td>0.288</td>
<td>1.302</td>
<td>321.1</td>
</tr>
<tr>
<td>Martin et al. [33]</td>
<td>(1.217 \times 10^{12})</td>
<td>90,933</td>
<td>0.302</td>
<td>1.343</td>
<td>350.7</td>
</tr>
<tr>
<td>Kosar and Gomzi [34]</td>
<td>(2.991 \times 10^3)</td>
<td>42,500</td>
<td>0.320</td>
<td>1.680</td>
<td>285.7</td>
</tr>
<tr>
<td>Bogetti and Gillespie [27]</td>
<td>(6.167 \times 10^{30})</td>
<td>164,740</td>
<td>0.524</td>
<td>1.476</td>
<td>77.5</td>
</tr>
</tbody>
</table>

Table 3: Mechanical properties of the glass fiber (E-glass) and the polyester resin (Atlac 382) [21, 27].

<table>
<thead>
<tr>
<th></th>
<th>Young’s Modulus [GPa]</th>
<th>Poisson’s Ratio</th>
<th>CTE [ppm/°C]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass fiber</td>
<td>73</td>
<td>0.22</td>
<td>5.04</td>
</tr>
<tr>
<td>Polyester resin</td>
<td>3.4</td>
<td>0.40</td>
<td>72</td>
</tr>
<tr>
<td>(glassy state)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 4: The estimated parameters used in the CHILE approach (Eq. 9) and in the calculation of the glass transition (Eq. 10).

<table>
<thead>
<tr>
<th>$T_{C1}$ [$^\circ$C]</th>
<th>$T_{C2}$ [$^\circ$C]</th>
<th>$E_r^0$ [MPa]</th>
<th>$E_r^\infty$ [MPa]</th>
<th>$\lambda$</th>
<th>$T_{g0}$ [$^\circ$C]</th>
<th>$T_{g\infty}$ [$^\circ$C]</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>25</td>
<td>3.4</td>
<td>3.4e3</td>
<td>0.4</td>
<td>0</td>
<td>147</td>
</tr>
</tbody>
</table>

Table 5: The predicted and measured values for the warpage defined as $w = w_A - w_C$ (see Fig. 13).

<table>
<thead>
<tr>
<th>$V_{sh}$</th>
<th>Predicted [mm]</th>
<th>Measured [mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$w_A$</td>
<td>$w_C$</td>
</tr>
<tr>
<td>0.02</td>
<td>0.101</td>
<td>0.049</td>
</tr>
<tr>
<td>0.04</td>
<td>0.111</td>
<td>0.051</td>
</tr>
<tr>
<td>0.06</td>
<td>0.122</td>
<td>0.052</td>
</tr>
<tr>
<td>0.08</td>
<td>0.140</td>
<td>0.053</td>
</tr>
<tr>
<td>0.10</td>
<td>0.164</td>
<td>0.061</td>
</tr>
<tr>
<td>0.12</td>
<td>0.192</td>
<td>0.075</td>
</tr>
</tbody>
</table>

$V_{sh} = 0.15 \pm 0.05$
Table 6: The mechanical properties of the UD layer with $V_f = 56.5\%$ ($W_f = 74.6\%$) at fully cured glassy state.

<table>
<thead>
<tr>
<th>UD</th>
<th>Micmec</th>
<th>SCFM</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_1$</td>
<td>42.43</td>
<td>43.14</td>
<td>GPa</td>
</tr>
<tr>
<td>$E_2 = E_3$</td>
<td>12.51</td>
<td>10.88</td>
<td>GPa</td>
</tr>
<tr>
<td>$G_{12} = G_{13}$</td>
<td>3.86</td>
<td>3.53</td>
<td>GPa</td>
</tr>
<tr>
<td>$G_{23}$</td>
<td>4.25</td>
<td>3.86</td>
<td>GPa</td>
</tr>
<tr>
<td>$\nu_{12} = \nu_{13}$</td>
<td>0.28</td>
<td>0.29</td>
<td>-</td>
</tr>
<tr>
<td>$\nu_{23} = \nu_{32}$</td>
<td>0.47</td>
<td>0.54</td>
<td>-</td>
</tr>
<tr>
<td>$\nu_{21} = \nu_{31}$</td>
<td>0.08</td>
<td>0.07</td>
<td>-</td>
</tr>
<tr>
<td>$\alpha_1$</td>
<td>7.34</td>
<td>7.36</td>
<td>ppm/°C</td>
</tr>
<tr>
<td>$\alpha_2 = \alpha_3$</td>
<td>45.19</td>
<td>45.13</td>
<td>ppm/°C</td>
</tr>
</tbody>
</table>

Table 7: The mechanical properties of the CFM layer with $V_f = 23\%$ ($W_f = 40\%$) at fully cured glassy state.

<table>
<thead>
<tr>
<th>CFM</th>
<th>Micmec</th>
<th>SCFM</th>
<th>Atlac 382</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_1 = E_2$</td>
<td>9.89</td>
<td>9.95</td>
<td>10.7</td>
<td>Gpa</td>
</tr>
<tr>
<td>$E_3$</td>
<td>6.74</td>
<td>6.60</td>
<td>-</td>
<td>GPa</td>
</tr>
<tr>
<td>$G_{12}$</td>
<td>3.67</td>
<td>3.70</td>
<td>-</td>
<td>GPa</td>
</tr>
<tr>
<td>$G_{23} = G_{13}$</td>
<td>1.87</td>
<td>1.82</td>
<td>-</td>
<td>GPa</td>
</tr>
<tr>
<td>$\nu_{12} = \nu_{21}$</td>
<td>0.35</td>
<td>0.34</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>$\nu_{31} = \nu_{32}$</td>
<td>0.26</td>
<td>0.26</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>$\nu_{13} = \nu_{23}$</td>
<td>0.38</td>
<td>0.40</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>$\alpha_1 = \alpha_2$</td>
<td>30.27</td>
<td>29.60</td>
<td>31</td>
<td>ppm/°C</td>
</tr>
<tr>
<td>$\alpha_3$</td>
<td>91.92</td>
<td>93.06</td>
<td>-</td>
<td>ppm/°C</td>
</tr>
</tbody>
</table>
“Modelling the Pultrusion Process of an Industrial L-shaped Composite Profile”

Ismet Baran, Remko Akkerman, Jesper H. Hattel

Submitted for publication, 2014.
Modelling the Pultrusion Process of an Industrial L-shaped Composite Profile

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Abstract
A numerical process simulation tool is developed for the pultrusion of an industrial L-shaped profile. The composite contains the combination of uni-directional (UD) roving and continuous filament mat (CFM) layers impregnated by a polyester resin system specifically prepared for the process. The chemo-rheology and elastic behaviour of the resin are obtained by applying a differential scanning calorimetry (DSC) and a dynamic mechanical analyser (DMA), respectively. The process induced stresses and shape distortions are predicted in a 2D quasi-static mechanical analysis. The numerical process model predicts the residual spring-in angle which is found to be close to the one measured from the real pultruded L-shaped products. The residual spring-in angle is further analyzed using the developed simulation tool for different pulling rates. Through-thickness stress variations are found to prevail inside the part such that the UD and CFM layers have different stress levels at the end of the process. The predicted stress pattern is verified by performing a stress calculation using the classical laminate theory (CLT).

Keywords: Spring-in, Internal stresses, Glass/Polyester, Finite element analysis (FEA), Pultrusion

1. Introduction
Pultrusion is in principle a simple process to manufacture constant cross sectional composite profiles. The process has a low labour content and a high raw material conversion efficiency since it is a continuous processing technique. The pultruded products have consistent quality and there is no need for any secondary finishing steps before the usage in service. A schematic view of the the pultrusion process is shown in Fig. 1. The reinforcements in the form of continuous unidirectional (UD) roving or continuous filament mat (CFM) are held on creel racks and fed continuously through a guiding system. These reinforcements are being impregnated with the desired matrix system in a resin bath. The wetted-out reinforcement pack is then collimated into a preformed shape before entering the heating die. A polymerization takes place inside the die with the help of the heat coming from the heaters. The cured profile is advanced via a pulling system to the cut-off saw where the
finished product is cut to desired lengths.

The CFM is generally used in combination with UD roving for pultruded industrial profiles. The CFM consists of long, continuous lengths of fibre strands overlying each other in a totally random swirl pattern. It provides stiffness and strength in the transverse as well as the pulling (longitudinal) directions. On the other hand, the UD roving provides longitudinal strength in the length of the profile. Therefore, the UD layer is transversely isotropic (TI), however the CFM layer can be considered as quasi-isotropic (QI) having equal material properties in the in-plane directions [1] and the out-of-plane properties are different than the in-plane properties [2].

The use of pultruded profiles in several industries such as construction, transportation and marine has grown significantly. Their main advantages over traditional materials are high strength-to-weight ratio, high corrosion resistance as well as good electrical and thermal insulation properties. In order to have a better understanding of the mechanical response or the failure mechanisms of pultruded structures under service loading conditions, the process induced residual stresses have to be characterized since they may lead to cracking during curing [3]. In addition, the dimensional changes during processing have to be controlled in order to improve the product quality in terms of geometrical tolerances. The thermal and cure history together with highly non-linear resin phase transitions (viscous-rubber-glassy) [4] make the process complex to control and have a significant influence on the quality of the final composite part. During phase transitions, the resin undergoes large changes in its material properties, most significantly in its thermal expansion and elastic modulus [5, 6]. The main mechanisms generating the process induced stresses and shape distortions in pultrusion are summarized in [3–7].

A numerical process simulation tool is essential to address the main challenges in pultrusion such as process induced residual stresses and shape distortions together with the prediction of the thermal and cure history. Expensive trial-and-error approaches for designing new products and process conditions can be avoided using the developed process models.

The temperature and cure evolutions have been investigated in detail for the pultrusion process in literature [8–16]. Both numerical simulations [8–13] and experiments [14, 15] were carried out to characterize the thermal and curing history for the pultruded products having simple cross sectional geometries such as rods and flat plates. Pulling force models were developed to calculate the viscous and frictional forces at the die-part interface [17, 18]. The effects of uncertainties in process parameters and material properties on the product quality were examined in [23] by means of a probabilistic simulation tool developed for pultrusion. The process parameters such as pulling speed and heater temperatures were optimized to maximize the cure quality or production rate by carrying out optimization studies based on thermo-chemical process models [19–22]. In addition to the thermo-chemical studies of pultrusion [8–23], state-of-the-art process models have recently been developed by the authors to calculate the process induced residual stresses and shape distortions in pultrusion of UD
profiles [24–27]. In this thermo-chemical-mechanical model, the temperature and the degree of cure fields were obtained in a three dimensional (3D) thermal model which was sequentially coupled with a 2D quasi-static mechanical model [24]. It should be noted that all these contributions have been dealing with pultruded products containing only UD reinforcements.

A novel thermo-chemical-mechanical analysis of the pultrusion process containing both UD and CFM layers is presented in this work. A numerical simulation tool is developed to calculate the process induced stresses and dimensional variations in an industrially pultruded L-shaped profile. An “orthophthalic” polyester resin system specifically prepared for the pultrusion process is utilized to wet-out the reinforcements. The resin system provided by the pultruder already contains the required fillers, initiators and chemical additives. The cure kinetics model was developed using isothermal and dynamic differential scanning calorimetry (DSC) experiments. The temperature- and cure-dependent resin modulus was defined by conducting dynamic mechanical analyser (DMA) tests in tension mode. A modified cure hardening instantaneous linear elastic (CHILE) model [28, 29] was employed for the modulus development during processing. The temperature and degree of cure distributions are calculated in a 3D thermo-chemical analysis formulated in a Eulerian frame. Subsequently, the 3D thermo-chemical model is coupled with a 2D quasi-static mechanical model to predict the stresses and displacements [24]. Two separate micromechanics approaches are utilized for the UD and the CFM layers. The spring-in formation is predicted and compared with measurements on real pultruded products. Subsequently, the residual spring-in angle is further calculated using the developed simulation tool for different pulling speed values. The classical laminate theory is employed to verify the predicted through-thickness residual stress field.

2. Numerical Implementation

2.1. 3D Thermal Model

The steady state energy equations are solved simultaneously for the UD layer (Eq. 1), the CFM layer (Eq. 2) and the die (Eq. 3) (see Fig. 10 for the positioning of the layers). Here, \( x_1 \) is the pulling or longitudinal direction; \( x_2 \) and \( x_3 \) are the transverse directions for the UD layer. On the other hand, \( x_1 \) (pulling direction) and \( x_2 \) represent the in-plane directions and \( x_3 \) is the out-of-plane direction for the CFM layer.

\[
\frac{(\rho C_p)_{UD}}{u} \left( \frac{\partial T}{\partial x_1} \right) = k_{x_1, UD} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2, UD} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3, UD} \frac{\partial^2 T}{\partial x_3^2} + q_{UD} \tag{1}
\]

\[
\frac{(\rho C_p)_{CFM}}{u} \left( \frac{\partial T}{\partial x_1} \right) = k_{x_1, CFM} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2, CFM} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3, CFM} \frac{\partial^2 T}{\partial x_3^2} + q_{CFM} \tag{2}
\]

\[
0 = k_{x_1, d} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2, d} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3, d} \frac{\partial^2 T}{\partial x_3^2} \tag{3}
\]
where $T$ is the temperature, $u$ is the pulling speed, $\rho$ is the density, $C_p$ is the specific heat and $k_{x1}$, $k_{x2}$ and $k_{x3}$ are the thermal conductivities along the $x_1$-, $x_2$- and $x_3$-directions, respectively. The subscripts $UD$, $CFM$ and $d$ correspond to the UD layer, the CFM layer and the die, respectively. Lumped material properties are used and assumed to be constant. The source terms $q$ in Eq. 1 and Eq. 2 are related to the internal heat generation due to the exothermic reaction of the polyester resin and expressed as [10]:

$$q_{UD} = (1 - V_f)_{UD} \rho_r H_{tr} R_r(\alpha, T)$$  \hspace{1cm} (4)

$$q_{CFM} = (1 - V_f)_{CFM} \rho_r H_{tr} R_r(\alpha, T)$$  \hspace{1cm} (5)

where $H_{tr}$ is the total heat of reaction for the polyester during the exothermic reaction, $\rho_r$ is the resin density, $V_f$ is the fiber volume fraction and $\alpha$ is the degree of cure. $R_r(\alpha, T)$ is the reaction of cure which can also be defined as the rate of $\alpha$, i.e. $d\alpha/dt$, which is assumed to be proportional to the rate of heat flow ($dH/dt$) [29] and expressed as:

$$\frac{d\alpha}{dt} = \frac{1}{H_{tr}} \frac{dH}{dt}$$  \hspace{1cm} (6)

In literature, several kinetic models have been proposed and analyzed to describe the resin curing reactivity [30, 31]. In the present work, a well known semi-empirical autocatalytic model [28] which is an Arrhenius type of equation is utilized. The corresponding expression is given as:

$$R_r(\alpha, T) = \frac{d\alpha}{dt} = A_0 \exp\left(\frac{-E_a}{RT}\right)\alpha^m(1 - \alpha)^n$$  \hspace{1cm} (7)

where $A_0$ is the pre-exponential constant, $E_a$ is the activation energy, $R$ is the universal gas constant, $m$ and $n$ are the orders of reaction (kinetic exponents).

The material derivative of the degree of cure field (Eq. 7) is translated into a partial derivative form in a Eulerian frame of reference in the pulling direction and expressed as [24]:

$$R_r(\alpha, T) = \frac{d\alpha}{dt} = \frac{\partial \alpha}{\partial t} + \frac{\partial \alpha}{\partial x_1} \frac{dx_1}{dt} = \frac{\partial \alpha}{\partial t} + u \frac{\partial \alpha}{\partial x_1}$$  \hspace{1cm} (8)

and from Eq. 7, the relation of the resin kinetics equation for the steady state approach (i.e. $\partial \alpha / \partial t = 0$) as suggested in [13] can be expressed as:

$$0 = R_r(\alpha, T) - u \frac{\partial \alpha}{\partial x_1}$$  \hspace{1cm} (9)

which is used in the 3D steady state thermo-chemical model. The equations above have been solved using a finite element approach by means of in-house developed routines implemented in ABAQUS [32].
2.2. 2D Mechanical Model

A temperature- and cure-dependent resin modulus is defined using a modified CHILE model [28, 29]. This model captures the modulus variation due to the phase changes (viscous-rubbery-glassy) during processing. It should be noted that the modulus is used in the material Jacobian matrix (\( J \)) [32] for the incremental stress calculation (i.e. \( \dot{\sigma} = J \dot{\varepsilon} \)). The corresponding expression for the modulus is given as:

\[
E_r = \begin{cases} 
E_0 & ; T^* \leq T_{C1} \\
A_e \exp(K_e T^*) & ; T_{C1} < T^* < T_{C2} \\
E_1 + \frac{T^* - T_{C2}}{T_{C3} - T_{C2}} (E_\infty - E_1) & ; T_{C2} < T^* < T_{C3} \\
E_\infty & ; T_{C3} \leq T^* 
\end{cases}
\] (10)

where \( T^* \) represents the difference between the instantaneous glass transition temperature \( (T_g) \) and the resin temperature \( T \), i.e. \( T^* = T_g - T \) [28]. \( A_e \) and \( K_e \) are the constants for the exponential term. The other model constants indicate the phase transition zones and are schematically shown in Fig. 2. Here, \( T_{C1}, T_{C2} \) and \( T_{C3} \) are defined as the critical temperatures and \( E_0, E_1 \) and \( E_\infty \) are the corresponding elastic modulus values, respectively, at which the modulus behaviour is changed due to the phase transitions of the resin (viscous-rubbery-glassy state). More specifically, \( E_0 \) and \( E_\infty \) can be considered as the elastic modulus in the viscous and glassy state, respectively. The glass transition temperature \( T_g \) can be defined as a function of cure degree [28, 33] and expressed as:

\[
T_g = T_g^0 + a_{T_g} \alpha 
\]

(11)

where \( T_g^0 \) is the glass transition temperature at \( \alpha = 0 \) and \( a_{T_g} \) is a constant.

The effective mechanical properties of the TI UD layer are calculated using the self consistent field micromechanics (SCFM) approach which is a well known and documented technique in the literature [7, 24]. On the other hand, the mechanical properties of the QI CFM layer is calculated considering the material properties of the UD layer obtained by the SCFM approach for the same \( V_f \) as the QI layer [2]. The details of the calculations for the QI laminate are given in [2].

An incremental linear elastic approach (Eq. 12) is implemented utilizing the user defined subroutines in ABAQUS to calculate the displacements and stresses. The details of the relations between the stress (\( \dot{\sigma}_{ij} \)) and strain (\( \dot{\varepsilon}_{ij} \)) tensors used in the present finite element implementation can be found in [24].

\[
\dot{\sigma}_{ij} = J \dot{\varepsilon}_{ij} 
\]

(12)

where \( J \) is the material Jacobian matrix [32]
3. Material Characterization of the Polyester

In this study, a summary of the results in [34] which has been carried out by the authors are presented. An industrial “orthophthalic” polyester system specifically prepared for the pultrusion process is considered.

3.1. Cure Kinetics

The curing behaviour of the polyester resin is characterized by performing DSC experiments. Isothermal scans are carried out at temperatures 120°C, 130°C and 140°C for a resin sample of ~10 mg. On the other hand, dynamic scans are performed by heating the sample from 25°C to 200°C with a heating rate of 5°C/min, 7.5°C/min and 10°C/min. The total exothermic heat of reaction ($H_{tr}$) released during cure is calculated approximately as 175±15 kJ/kg by integrating the heat flow-time plots for the dynamic DSC experiments [29].

The curing rate is obtained from the measured heat flow data by using Eq. 6. The experimental degree of cure is then calculated by calculating the area under $d\alpha/dt$ curves by integration. The parameters in Eq. 7 are obtained using a weighted least squares non-linear regression analysis including the experimental data from both isothermal and dynamic DSC scans. The estimated best fit parameters are given in Table 1. Fig. 3 shows the measured and estimated (best fit) evolutions for the degree of cure and the cure rate. It is seen that the autocatalytic model (Eq. 7) accurately predicts the degree of cure as well as the cure rate developments for all three isothermal temperatures. In addition, a reasonably good fit is also obtained considering different heating rates in dynamic DSC scans as seen in Fig. 4.

3.2. Elastic Modulus

The DMA experiments are conducted to obtain the evolution of the elastic modulus of the polyester. The DMA is utilized in tension mode by applying a sinusoidal deformation/strain to the sample. The stiffness (modulus) and damping ($\tan\delta$) are measured as a response. The measured modulus can be described by two components: an in-phase component, the storage modulus (elastic behaviour) ($E'$), and an out-of-phase component, the loss modulus ($E''$). $\tan\delta$ is defined as the ratio of the loss modulus to the storage modulus and represents the energy dissipation in the sample. The peak of $\tan\delta$ at which the difference between $E'$ and $E''$ is at its minimum indicates the glass transition temperature $T_g$.

The neat resin is first cured in the form of rectangular stripes using an oven. Dynamic heating scans are performed from 25°C to 190°C with a heating rate of 5°C/min and a frequency of 6.22 Hz [35]. The static and dynamic load strains are set to 1% and 0.1%, respectively.

Fig. 5 shows the measured $E'$ and $E''$ together with $\tan\delta$ evolutions. It is seen that the peak of $\tan\delta$ is obtained at ~135°C which corresponds to $T_g$ for fully cured resin ($\alpha=1.0$). In the present study, $T_g^0$ in Eq. 11 is assumed to be 0 which provides $a_{T_g}=135°C$. Using the modified CHILE model given by Eq. 10, a least squares non-linear regression analysis is
performed to obtain the constants in Eq. 10 which give the best agreement with the measured data. The estimated parameters used in the modified CHILE model are given in Table 2. The calculated elastic modulus evolution (best fit) is compared with the experimental data seen in Fig. 6. A good agreement is found between the measured and the predicted modulus evolution.

4. Validation

4.1. Experimental

An L-shaped profile was pultruded in a commercial pultrusion company. Some photographs of the product are shown in Fig. 7. The cross sectional dimensions of the part were $50 \times 50 \times 5$ mm and it contained glass/polyester based UD and CFM layers. A CFM having a density of 450 g/m$^2$ was used in the process. The heating die was made of chrome steel.

The die length was specified as 1 m and the heater temperatures were set to 110 °C (near die the inlet) and 140 °C (near the die exit) by the pultruder. The heating platens were located symmetrically on top and bottom of the pultrusion die. The length and width of the heaters were 275 mm and 120 mm, respectively. A spacing between the heaters were employed with a length of 150 mm. The pulling speed of the line was adjusted to 600 mm/min.

The overall fiber weight fraction ($W_f$) of the profile was determined approximately as 65% from the weight measurements performed by the pultruder. Accordingly, the fiber volume fractions ($V_f$) are calculated as 55.1% and 23% for the UD layer and the CFM layer, respectively. The thickness of the pultruded CFM layers (450 g/m$^2$) is assumed to be 0.762 mm according to the data given in [36]. To justify this, the layer configuration of the pultruded L-shaped profile has been investigated under a microscope and the details are shown in Fig. 8. It is observed that the thickness of the CFM layer varies between 0.6-0.9 mm having a mean value of approximately 0.75 mm which is very close to the thickness value (0.762 mm) given in [36]. Hence, 0.762 mm is specified for the CFM layer thickness in the present work.

4.2. Pultrusion Model

4.2.1. Thermo-chemical Analysis

The temperature and the degree of cure fields are predicted in the 3D thermo-chemical analysis. A schematic representation of the pultrusion model is seen in Fig. 9. Only half of the model is used due to symmetry. The length of the profile at the post-die region is taken as 5 m as seen also from Fig. 9. In the 3D thermo-chemical model, a perfect thermal contact is assumed at the die-part interface as in [10, 15, 24]. The employed thermal properties are listed in Table 3.

The cross sectional details are shown in Fig. 10. Local material orientations (Local CS-1 and Local CS-2) are defined for the CFM layer as seen in Fig. 10 in order to reflect the
in-plane ($x_1$- and $x_2$-direction) and the out-of-plane ($x_3$-direction) properties correctly. For the round corner, a cylindrical local coordinate system (Local CS-1) is used in which the $x_2$- and $x_3$-directions are taken as the tangential (T) and radial (R) directions, respectively.

The temperature and the degree of cure (only for the composite) of all nodes are assigned to the ambient temperature (25 °C) and 0, respectively at the beginning of the 3D thermo-chemical analysis. The composite temperature and degree of cure at the die inlet are likewise set to the resin bath temperature 25 °C and 0, respectively as a boundary condition (BC). Prescribed heater temperatures (110 °C and 140 °C) are applied at the heating regions as shown in Fig. 9. At the symmetry surfaces adiabatic boundaries are defined in which no heat flow is allowed across the boundaries. The remaining exterior surfaces of the die are exposed to ambient temperature with a convective heat transfer coefficient of 10 W/m\(^2\)-K except for the surfaces located at the heating regions. Similar convective boundaries are also defined for the outer surfaces of the pultruded profile at the post die region.

4.2.2. Quasi Static Mechanical Analysis

The process induced shape distortions (spring-in) and the stresses are predicted in the 2D quasi-static mechanical model. Quadratic generalized plane strain elements are utilized in ABAQUS. In this 2D mechanical model, the cross section of the composite is assumed to move through the pulling direction meanwhile tracking the corresponding temperature and degree of cure profiles already calculated in the 3D thermo-chemical analysis. A generic representation of this sequential coupling procedure is shown in Fig. 11. Since the length of the pultruded profile is generally much larger than the cross sectional dimensions, a plain strain assumption is convenient for the mechanical analysis of the pultrusion [24]. A mechanical contact formulation is defined at the die-part interface. By using this contact formulation, a separation due to the thermal contraction of the part and/or the resin shrinkage at the interface is allowed. However, any expansion of the composite beyond the tool interfaces is restricted. The friction force at the contact surfaces is assumed to be zero (sliding condition). Symmetry mechanical BCs are applied at the symmetry lines and the die is assumed to be fixed from the outer regions. A local material orientation is employed for the CFM layer as described in Section 4.2.1 (see Fig. 10).

The constituent mechanical properties of the glass fiber and the polyester resin are summarized in Table 4. It should be noted that the resin CTE in rubbery state is known to be approximately 2.5 times larger than the CTE in glassy state [6, 29]. Hence, a step change in the CTE is applied at $T^* = T_{C2}$ (see Fig. 2).

The polymerization shrinkage of a polyester system highly depends on the concentration, type and molecular weight of the chemical additives, the polyester structure as well as the processing conditions [38]. Therefore, the total volumetric shrinkage of a polyester resin system varies in a wide range, i.e. 2-12% [38–40]. In the present work, the total volumetric shrinkage is assumed to be 6% since the compounds of the polyester system are confidential and hence not provided by the pultruder.
5. Results and Discussions

Fig. 12 and Fig. 13 show the predicted temperature and degree of cure distributions, respectively, for the specified pulling speed (600 mm/min). The temperature evolution for a point inside the part is depicted in Fig. 12(left) and the corresponding contour plots at the die exit and end of the process are shown in Fig. 12(right). The temperature gradually increases inside heating die and decreases at the post die region owing to the convective cooling to the ambient temperature. It is seen that the composite temperature exceeds the heater temperature (140°C) near the die end due to the internal heat generation of the polyester system. In addition, the temperature of the inner regions is higher than the regions close to the die as seen from Fig. 12(right). The maximum composite temperature is calculated to approximately 170°C inside the die. The part cools down to a temperature around 60-70°C at the end of the process ($x_1 = 6$ m). The maximum temperature gradient over the cross section is calculated approximately as 30°C inside the die. The corresponding degree of cure development is depicted in Fig. 13(left) for the same point in Fig. 12. It is seen that the composite is cured inside the die with a conversion rate larger than 0.9 (90%). The inner region has a higher degree of cure value as compared to the outer regions since the internal heat generation is more dominant for the inner region [24]. Further curing (≈3%) takes place at the post die region since the composite temperature is still high enough to facilitate the exothermic chemical reaction. The maximum degree of cure gradient over the cross section is calculated approximately as 10% inside the die. It is found that the part is almost fully cured (>97%) at the end of the process as seen from Fig. 13(right). As aforementioned, the temperature and cure gradients play an important role for the development of residual stresses and distortions in the manufactured composite parts [7].

The obtained temperature and degree of cure distributions are mapped to the 2D quasi-static mechanical analyses for the calculation of the stresses and displacements. Fig. 14 shows the predicted displacements as a deformed contour plot in the global $x_2$-direction ($U_{22}$) and $x_3$-direction ($U_{33}$) at the end of the process ($x_1 = 6$ m). It is seen that a spring-in formation is found according to the predicted deformation pattern. This phenomenon agrees quite well with the spring-in pattern observed in the real pultruded L-shaped profiles. The maximum $U_{22}$ and $U_{33}$ in magnitude are calculated approximately as 0.32 mm and 0.16 mm, respectively. The evolution of the spring-in angle denoted as $\theta$ is shown in Fig. 15. It is seen that the movement of the part is restricted by the die due to the employed mechanical contact formulation at the die-part interface. The spring-in angle starts increasing after die exit where the mechanical contact is deactivated. The magnitude of $\theta$ is calculated approximately as 0.45° at the end of the process at which the temperature of the part is around 60-70°C. The spring-in angle is also measured from the pultruded L-shaped products and the predicted value for the spring-in angle ($\theta = 0.45^\circ$) is found to be very close to the measured range $0.5 \pm 0.05^\circ$. The residual spring-in angle is further calculated using the developed simulation tool for different pulling speed values and the results are depicted in Fig. 16. It is seen that $\theta$ increases with the pulling speed since the glass transition point shifts towards the process end as the pulling rate increases. Therefore, a wider rubbery
zone takes place providing larger deformations for the part. The processing conditions (line speed, temperature and degree of cure) together with different mechanical properties of the UD and CFM layers have an important effect on the shape distortions in pultrusion.

The predicted process induced residual stresses are shown in Fig. 17 as undeformed contour plots in the local material orientations (local coordinate systems seen in Fig. 10). Here, $S_{11}$, $S_{22}$ and $S_{33}$ are the normal stresses in the $x_1$-, $x_2$- and $x_3$-direction, respectively. It is seen that the normal stress in the in-plane direction for the UD and the CFM layer ($S_{22}$) has a higher stress level as compared to $S_{11}$ and $S_{33}$. The magnitude of the maximum tension and compression for $S_{22}$ are calculated approximately as 6.3 MPa and 18.3 MPa, respectively. Stress concentrations are found for $S_{33}$ through the thickness direction at the end boundaries. However, apart from these boundaries $S_{33}$ is almost zero and uniform over the cross section since the thickness (5 mm) is too small as compared with the width (50 mm) to build up the stresses ($S_{33}$) in the $x_3$-direction. The corresponding through-thickness stress distributions ($S_{11}$, $S_{22}$ and $S_{33}$) are depicted in Fig. 18 and Fig. 19 considering two different cross sections (AA and BB). A variation in stress prevails inside the UD layer and CFM layers for section AA as seen in Fig. 18 since section AA is located at the symmetry line. On the other hand, more uniform stress distributions are obtained at section BB as seen from Fig. 19 for which the boundary effects are less pronounced. It is seen that there is a discontinuity in the stress at the UD-CFM interface. The CFM layers are under compression ($S_{22} < 0$) whereas the tension ($S_{22} > 0$) exists for the UD layer in the $x_2$-direction. The other way around is the case for $S_{11}$, i.e. UD layer is under compression and the CFM layers are under tension (Fig. 19).

The obtained stress pattern shown in Fig. 19 is verified using the classical laminate theory (CLT) [41]. A composite laminate having CFM and UD layers similar to the pultruded product (5 mm thick) is considered in a ply stress calculation using the CLT. The process induced stresses are built up significantly after the part reaches the peak temperature during pultrusion [24]. Hence, a thermal load ($\Delta T = -100^\circ C$) is applied to the laminate which corresponds to the cooling of the part from peak temperature to ambient temperature. Knowing the elastic modulus and CTE of the layers in glassy state, the in-plane ply strains and stresses are calculated using the CLT. A schematic view of the laminate is shown in Fig. 20. Here, $x$-direction is defined as the fiber direction for the UD layer. The predicted in-plane stresses ($S_{xx}$ and $S_{yy}$) are also depicted in Fig. 20. The stress distributions are found to be similar as compared to the in-plane stresses ($S_{11}$ and $S_{22}$) in Fig. 19, e.g. the CFM layer is under compression for $S_{yy}$ and tension for $S_{xx}$. Nevertheless, the magnitudes deviate since the phase changes of the resin influence the stress evolutions during pultrusion. Note that the counterpart parts of $S_{11}$ and $S_{22}$ (Fig. 19) are $S_{xx}$ and $S_{yy}$ from the CLT analysis, respectively.

The variations and discontinuities in the residual stresses directly affects the quality of bonding between the UD and CFM layers. In addition, the failure modes and the delamination resistance are also influenced by the non-linear through-thickness residual stresses. The internal stress levels in the in-plane direction ($S_{22}$) are important especially for the web
flange junction (round corner) during service loading. Hence, the process induced stresses have to be taken into account during the structural analysis of the pultruded products.

6. Conclusions

A numerical process simulation was carried out for the pultrusion of an industrial L-shaped profile containing both UD and CFM layers. The reinforcements were impregnated using a polyester resin system specifically prepared for the process. The curing behaviour was characterized by performing DSC experiments for the resin sample. A cure kinetics model was developed using the isothermal and dynamic DSC data. A temperature- and cure-dependent resin modulus was defined by conducting DMA tests. A modified CHILE model was developed to capture the modulus variation due to the phase changes (viscous-rubbery-glassy) during processing. The temperature and the degree of cure distributions were calculated in a 3D thermo-chemical analysis of the process. Subsequently, the process induced stresses and shape distortions were predicted in a 2D quasi-static mechanical analysis. Different micromechanical approaches were employed for the UD and CFM layers since they have different mechanical behaviour.

The predicted spring-in formation was found to match well with the spring-in pattern observed in the real pultruded L-shaped profiles. In addition, it was found that the calculated spring-in angle was very close to the measured range. This shows that the proposed numerical simulation tool and approach estimate the mechanical response of this specific process correctly. The residual spring-in angle was further calculated for different pulling rates using the developed simulation tool. The spring-in angle was found to increase with the pulling speed. Through-thickness stress gradients were found to prevail at the end of the process. As expected, higher stress level was obtained for the in-plane normal stress ($S_{22}$) as compared to $S_{11}$ and $S_{33}$. It was also observed that there exists a discontinuity in the stress distribution at the UD-CFM interface. The overall characteristics of the predicted stress fields were verified by calculating the in-plane stresses using the CLT.

The stress variations may decrease the quality of bonding between the UD and CFM layers. In addition, the failure modes and the delamination resistance may also be affected by the through-thickness stress variations.

7. Acknowledgements

The author wishes to thank Dr. Roy Visser, Ivo Vrooijink and Nadia Vleugels from University of Twente for guidance and valuable discussions of the experiments. This work is a part of the DeepWind project which has been granted by the European Commission (EC) under the FP7 program platform Future Emerging Technology.

References


[26] Baran I, Hattel JH, Akkerman. Mechanical modelling of pultrusion process: 2D and 3D numerical


Figure 1: Schematic view of a pultrusion process.

Figure 2: Schematic representation of the elastic modulus evolution for the polyester resin.
Figure 3: The experimental and predicted (best fit) degree of cure evolutions (left) and the cure rate as a function of degree of cure (right) for the isothermal DSC scans.

Figure 4: The experimental and predicted (best fit) degree of cure evolutions (left) and cure rate as a function of temperature (right) for different heating rates used in dynamic DSC scans.
Figure 5: Measured storage modulus ($E'$) and loss modulus ($E''$) together with tan $\delta$ evolutions.

Figure 6: Comparison of the measured (DMA) and predicted elastic modulus developments (log scale).
Figure 7: Pultruded L-shaped profile.

Figure 8: Cross section of the pultruded L-shaped product showing the CFM and UD layer orientations.
Figure 9: Schematic view of the half pultrusion domain for the L-shaped pultruded profile. All dimensions are in mm.

Figure 10: The cross sectional details of the pultruded L-shaped profile and the die. All dimensions are in mm.
Figure 11: Representation of the sequential coupling of the 3D thermo-chemical model with the 2D quasi-static mechanical model.

Figure 12: The calculated temperature evolution at a point inside the part (left). The corresponding contour plots for the temperature at the die exit ($x_1 = 1$ m) and at the end of the process ($x_1 = 6$ m) (right). Note that the legends of the contour plots are not the same.
Figure 13: The calculated degree of cure evolution at a point inside the part (left). The corresponding contour plots for the degree of cure at the die exit ($x_1 = 1$ m) and at the end of the process ($x_1 = 6$ m) (right). Note that the legends of the contour plots are not the same.

Figure 14: The deformed contour plots of the L-shaped profile in the global $x_2$-direction (left) and $x_3$-direction (right) at the end of the process $x_1 = 6$ m. The scale factor for the deformed shape is 10.
Figure 15: The evolution of the spring-in angle ($\theta$).

Figure 16: Predicted residual spring-in angles ($\theta$) for different pulling speed values.
Figure 17: Undeformed contour plots of the normal stresses in the $x_1$-direction $S_{11}$ (a), $x_2$-direction $S_{22}$ (b) and $x_3$-direction $S_{33}$ (c) at the end of the process. Note that the directions are based on the local material orientations (Local CS-1 and Local CS-2) seen in Fig. 10.

Figure 18: The through-thickness stress distributions at section AA. Note that the coordinate "0 mm" indicates the part center.
Figure 19: The through-thickness stress distributions at section BB. Note that the coordinate "0 mm" indicates the part center.

Figure 20: The through-thickness in-plane stresses ($S_{xx}$ and $S_{yy}$) predicted using the CLT by applying a thermal load, i.e. $\Delta T = -100^\circ$C.
<table>
<thead>
<tr>
<th>$A_0$ [1/s]</th>
<th>$E_a$ [kJ/mol]</th>
<th>$m$</th>
<th>$n$</th>
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<td>$7.5581 \times 10^9$</td>
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<td>0.63</td>
<td>1.847</td>
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Table 2: The estimated constants used in the modified CHILE model (Eq. 10).

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<td>-60</td>
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<td>110</td>
<td>0.0195</td>
<td>0.73</td>
<td>3.76</td>
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<td>0.043</td>
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Table 3: Thermal properties used in the thermo-chemical model [15, 37].

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<th>$C_p$ [J/kg-K]</th>
<th>$k_x$ [W/m-K]</th>
<th>$k_{x1}$, $k_{x3}$ [W/m-K]</th>
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<td>1830</td>
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<td>0.17</td>
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<td>Glass fiber</td>
<td>2560</td>
<td>670</td>
<td>11.4</td>
<td>1.04</td>
</tr>
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<td>Steel die</td>
<td>7833</td>
<td>460</td>
<td>40</td>
<td>40</td>
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Table 4: Mechanical properties of the glass fiber and the polyester resin [7].

<table>
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<th>Poisson’s Ratio</th>
<th>CTE [ppm/°C]</th>
</tr>
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<tbody>
<tr>
<td>Glass fiber</td>
<td>73</td>
<td>0.22</td>
<td>5.04</td>
</tr>
<tr>
<td>Polyester resin (glassy state)</td>
<td>3.76</td>
<td>0.40</td>
<td>72</td>
</tr>
</tbody>
</table>
“Material Characterization of a Polyester Resin System for the Pultrusion Process”

Ismet Baran, Remko Akkerman, Jesper H. Hattel

Material characterization of a polyester resin system for the pultrusion process

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ARTICLE INFO
Article history:
Received 21 March 2014
Received in revised form 24 April 2014
Accepted 28 April 2014
Available online 6 May 2014

Keywords:
A. Thermosetting resin
B. Cure behaviour
B. Rheological properties
B. Mechanical properties

ABSTRACT
In the present work, the chemo-rheology of an industrial “orthophthalic” polyester system specifically prepared for a pultrusion process is characterized. The curing behaviour is first characterized using the differential scanning calorimetry (DSC). Isothermal and dynamic scans are performed to develop a cure kinetics model which accurately predicts the cure rate evolutions and describes the curing behaviour of the resin over a wide range of different processing conditions. The viscosity of the resin is subsequently obtained from rheological experiments using a rheometer. Based on this, a resin viscosity model as a function of temperature and degree of cure is developed and predicts the measured viscosity correctly. The evolution of the storage and loss moduli are also measured as a function of time using the rheometer which provides an information about the curing as well as the gelation. The temperature- and cure-dependent elastic modulus of the resin system is determined using a dynamic mechanical analyzer (DMA) in tension mode. A cure hardening and thermal softening model is developed and a least squares non-linear regression analysis is performed. The variation in elastic modulus with temperature and phase transition is captured for a fully cured resin sample.

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1. Introduction
Pultrusion is a continuous process in which constant cross sectional composite profiles are manufactured. The reinforcements can be in the form of uni-directional (UD) roving or continuous filament mat (CFM). A thermosetting resin system (containing the unsaturated resin, fillers and specialized additives) is used to impregnate the reinforcements in a resin bath. The wetted-out raw materials are pulled through a heated steel forming die using a continuous pulling mechanism. The curing as well as the exothermic chemical reaction is initiated by the heaters located on the die. The cured and solidified profiles are cut into the desired length by a cut-off saw at the end of the process. A schematic view of the pultrusion process is shown in Fig. 1.

Several studies with the aim of obtaining a better understanding of the pultrusion process can be found in literature [1–13]. The curing behaviour of an unsaturated polyester resin with mixed initiators was investigated both experimentally and theoretically and subsequently used in a thermo-chemical analysis of the pultrusion process [1,2]. Cure kinetics of the processing resin system (epoxy based) has been investigated experimentally in [3,4] to predict the temperature and degree of cure distributions inside the part. The temperature of the part exceeds the heater temperatures during the process due to the internal heat generation inside the resin [5–8]. In order to model this phenomenon, the total heat of reaction of the resin material has to be modelled correctly. Pulling force models were developed considering the viscosity evolution and the contact pressure at the die–part interface [9,10]. The process induced residual stresses and distortions were predicted in [11]. In [12], two different numerical simulations were performed for the pultrusion of a C-shaped composite using the finite difference method and the finite element method. It was concluded that similar results were obtained from both methods. The pultrusion process was optimized based on the numerical model in [12] by means of a genetic algorithm and the simplex method in [13]. The variance of the cure degree evaluated at the exit cross section was minimized by an iterative procedure based on the combination of the above techniques. In this work a temperature- and cure-dependent resin modulus was utilized for an epoxy resin system. The parameters in the modulus model were taken from an experimental analysis of a resin transfer moulding process (RTM) which provides an information about the curing as well as the gelation. The temperature- and cure-dependent elastic modulus of the resin system is determined using a dynamic mechanical analyzer (DMA) in tension mode. A cure hardening and thermal softening model is developed and a least squares non-linear regression analysis is performed. The variation in elastic modulus with temperature and phase transition is captured for a fully cured resin sample.

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http://dx.doi.org/10.1016/j.compositesb.2014.04.030
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A precise constitutive model is essential in order to numerically simulate and optimize the pultrusion process properly. The thermo-mechanical and rheological properties of the thermosetting resin change due to thermal softening and cure hardening. Hence, the cure kinetics and rheological behaviour together with the stiffness variations have to be addressed carefully for the processing resin system. However, the material properties of the reinforcements are in general assumed to remain constant throughout the composite manufacturing processes.

Among all the thermosetting resin systems, polyester resins play an important role in the pultrusion industry because of their versatility in properties, flexibility in processing and low cost of manufacture [14]. Unsaturated polyester resins (UPRs) are all based on the use of an unsaturated monomer within a polymer backbone, formed by reacting diacids and glycols which dissolve the UPR in a reactive diluent, typically a styrene monomer. Either maleic anhydride or fumaric acid are used as the unsaturated monomer for all commercial polyesters [15]. The major three types of UPR are the “orthophthalic” anhydride polyesters, “isophthalic” acid polyesters and “terephthalic” acid polyesters. Each system has specific chemical and mechanical properties [15].

The low viscosity of polyester resin systems promotes quick and total reinforcement wet-out during processing and their high polymerization reactivity allows for a fast but controllable cure within the pultrusion die. They are available in various formulations containing initiators, inhibitors, functional additives, etc. These formulations are prepared to meet the desired performance of the specific product and the pultrusion process. As a consequence, the processing conditions such as pulling rate, temperature of the heaters and the die geometry differ due to the specific resin system used in the pultrusion process. In addition, the thermal, chemical and mechanical behaviour of the processing composite also change depending on the resin system.

The cure kinetics, rheological and mechanical characteristics of polyester resins have been intensively studied in literature [14,16–30]. The curing behaviour of unsaturated polyester was investigated in [14,16–23] by performing differential scanning calorimetry (DSC) tests. Isothermal and dynamic scans were conducted using the DSC to measure the overall reaction rate and calculate the total heat of reaction during curing. It is assumed that the heat produced during cure is proportional to the extent of the curing reaction. The reaction and the conversion rates are defined based on the total heat calculated from the area under the measured heat flow curves. The glass transition temperature can also be determined from the DSC tests. The cure reaction of the unsaturated polyester resin is quite complex and an early formation of micro-gels occurs even at conversion levels as low as 3–4% due to the rapid build-up of a crosslinked network [14]. In addition to the DSC, Fourier transform infrared spectroscopy (FTIR) measurements were carried out to characterize the cure behaviour of the unsaturated polyester [19,22,23]. In general a good agreement was found between the conversion rates obtained from the DSC and the FTIR. A cure kinetics model was also developed to simulate the curing behaviour [17–21] and the parameters used in the model were calculated from the measured heat flow and conversion rates.

The phase of the polyester changes from a liquid (viscous) to a gel (rubbery) and then a stiff solid (glassy) state during curing. Thus, the rheological and mechanical properties change significantly during processing. The rheology of the polyesters have been studied using a rheological testing or dynamic mechanical analyser (DMA) [14,24–30]. Rheological measurements were employed to test the viscosity change during reaction of the polyester resin systems [14,27,28] using a rheometer in oscillatory mode. The gel point and the corresponding temperature can be determined at which the viscosity starts to grow and the hardening stage begins near gelation. In addition to the viscosity, the developments of the storage modulus and the loss modulus can also be obtained from rheological measurements. From DMA tests, the glass transition temperature is calculated from the ratio of the loss modulus to the storage modulus.

An industrial polyester resin system specifically prepared for a pultrusion process is characterized in the present work. The resin system is based on an “orthophthalic” polyester with medium reactivity and already contains the required fillers, initiators and chemical additives. From a process point of view, the resin is specific but can be considered generic for pultrusion. The curing, rheological and mechanical characteristics are analysed by performing experimental tests and developing the corresponding constitutive material models. A DSC is used to obtain the curing kinetics such as curing rate as a function of time and temperature. The total heat of reaction released during curing is also measured using the DSC. A weighted least squares non-linear analysis is performed to fit the autocatalytic cure kinetics model to the measured data. The viscosity evolution is obtained using a rheometer in oscillatory mode with circular plates. A temperature- and cure-dependent viscosity model is developed to predict the measured viscosity. The gelation point is also determined based on the viscosity profile together with the corresponding degree of cure profile calculated using the cure kinetics model. The modulus of the resin changes with the temperature and transitions (glassy to rubbery zone). The modulus development as well as the glass transition temperature at fully cured state is obtained by performing a DMA analysis in tension mode. A temperature- and cure-dependent modulus model is used to estimate the measured data.

2. Cure kinetics

The temperature and the degree of cure distributions inside the processing material have to be analysed in order to investigate the pultrusion process precisely. The chemical exothermic reaction takes place when the composite reaches the reaction initiation temperature inside the die. The direction of the heat flux from the heaters is inverted such that the heat flow is transmitted from the composite to the die due to the internal heat generation. Hence, at some point the composite temperature exceeds the die temperature during curing. In order to capture these thermo-chemical aspects in pultrusion, the cure behaviour has to be well identified.

The cure characteristics of an industrial pultrusion polyester resin are obtained by performing DSC tests using Mettler Toledo DSC822. The difference in the heat flows from the sample and the reference side of the sensor is measured in the DSC as a function of temperature or time with a sampling rate of 1 value per second. A variation in the heat flow arises when the resin sample absorbs or releases heat due to the thermal effects such as the exothermic reaction during curing. The temperature spectrum used in the DSC is determined based on the processing conditions provided by the pultruder. Three different dynamic and isothermal experiments are performed in order to specify the parameters used in
the cure kinetics model which describe the curing behaviour of the resin over a wide range of different processing conditions.

Isothermal experiments are carried out at temperatures 120 °C, 130 °C and 140 °C using a resin sample of 10 mg. On the other hand, dynamic scans are performed by heating the sample from 25 °C to 200 °C with a heating rate of 5 °C/min, 7.5 °C/min and 10 °C/min. It should be noted that high heating rates are employed in pultrusion such that the peak temperature (~170 °C) is obtained by heating the sample from room temperature (~25 °C) within 1–2 min inside the heating die. Figs. 2 and 3 show the heat rate evolved during the isothermal and dynamic experiments, respectively. It is seen from Fig. 2 that the reaction heat as well as the peak of the heat flow increases with the isothermal set temperature. On the other hand the duration of the exothermic reaction decreases with the temperature. Similarly, using a higher heating rate results in a higher reaction heat at earlier stages during the dynamic DSC scans as seen in Fig. 3. The total exothermic heat of reaction (Htr) released during cure is calculated approximately as 175 ± 15 kJ/kg by obtaining the integral of the heat flow-time plots for the dynamic DSC experiments. Here, a straight baseline between the onset and the end of the reaction is considered as suggested in [31]. The rate of degree of cure (dα/dt) is assumed to be proportional to the rate of heat flow (dH/dt) [31] and expressed in Eq. (1). The corresponding evolutions of dα/dt as a function of time are depicted in Fig. 4 for the isothermal DSC experiments.

\[
d\alpha = \frac{1}{Htr} \frac{dH}{dt} \tag{1}
\]

![Fig. 2. The evolution of the heat flow for the isothermal DSC experiments.](image)

![Fig. 3. The evolution of the heat flow for the dynamic DSC experiments.](image)

**Table 1**
The estimated cure kinetics parameters of the polyester (Eq. (2)).

<table>
<thead>
<tr>
<th>A0 (1/s)</th>
<th>Ea (kJ/mol)</th>
<th>m</th>
<th>n</th>
</tr>
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<tbody>
<tr>
<td>7.5581 \times 10^0</td>
<td>82,727</td>
<td>0.63</td>
<td>1.847</td>
</tr>
</tbody>
</table>

The degree of cure is then calculated by obtaining the areas under the curves depicted in Fig. 4 by integration. In literature, several cure kinetics models have been proposed and analysed to describe the curing of the thermosetting resin systems [32,33]. A well known semi-empirical autocatalytic model [34] which is an Arrhenius type of equation is utilized. The corresponding expression is given as:

\[
R_c(\alpha, T) = \frac{d\alpha}{dt} = \frac{A_0 \exp \left( \frac{-E_a}{RT} \right)}{Htr} \alpha^n (1 - \alpha)^m \tag{2}
\]

where \(A_0\) is the pre-exponential constant, \(T\) is the absolute temperature, \(E_a\) is the activation energy, \(R\) is the universal gas constant and \(m\) and \(n\) are the order of reaction (kinetic exponents). The parameters in Eq. (2) are obtained using a weighted least squares non-linear regression analysis including the experimental data from both isothermal and dynamic DSC scans. The estimated best fit parameters are given in Table 1. As seen from the best fit between the experimental data and the predictions in Fig. 5, the autocatalytic model accurately predicts the degree of cure as well as the cure rate evolutions for all three isothermal temperatures. Fig. 6 shows that a reasonably good fit is also obtained between the measured and the estimated degree of cure as well as cure rate for different heating rates in dynamic DSC scans.

**3. Rheological behaviour**

In pultrusion, the rheological behaviour of the processing resin system directly affects the viscous force development at the die–part interface. The viscous force is one of the contributions (the collimation, the bulk compaction, the viscous and the frictional force) to the pulling force [10]. Before the gelation point, a viscous drag occurs at the die–part interface. The viscous force can be defined as a function of the viscosity profile for pultrusion [35]. In order to predict the viscous force correctly, the rheological properties of the processing resin have to be characterized.
In this work, rheological measurements are carried out using Anton Paar-Physica MCR 501 rheometer in "plate–plate" mode for the polyester resin system and 200 measuring points are used in the experiments. The neat polyester is prepared in liquid form with dimensions of \( \phi 2430 \text{ mm (diameter)} \) and \( \phi 22 \text{ mm (thickness)} \). Circular plates are used in oscillatory mode at 1% strain and 1 Hz. Two temperature cycles (Cycle-1 and Cycle-2) having different heating rates are used in the rheometer as seen from Fig. 7(left). A hold temperature of 100 °C and 130 °C are considered in Cycle-1 and Cycle-2, respectively. The corresponding degree of cure
Evolutions are calculated using the developed cure kinetics model (Eq. (2)). It is seen from Fig. 7(right) that higher cure rate and degree of cure are obtained for temperature Cycle-2 (130 °C). The viscosity evolutions as well as the variations in the storage modulus \(G'\) and the loss modulus \(G''\) are measured. A well-known temperature- and cure-dependent viscosity model is implemented to predict the viscosity development and expressed as [36,37]:

\[
\eta = \eta_0 \exp \left( \frac{\Delta\varepsilon}{RT} + Kx \right)
\]

(3)

where \(\Delta\varepsilon\) is the viscous activation energy, \(\eta_0\) is the initial viscosity, \(K\) is a constant, \(R\) is the universal gas constant, \(T\) is the absolute temperature. A least squares non-linear regression analysis is performed upon the measured data in order to determine the constants in the viscosity model. Figs. 8 and 9 show the viscosity evolutions as a function of time and temperature, respectively. The estimated model constants are given in Table 2. It is seen that a good agreement is found between the measured and estimated (best fit) viscosity profiles for the two different temperature scans. The viscosity increases rapidly after some time owing to the curing of the sample. The viscosity for Cycle-2 starts increasing earlier (~4.6 min) than Cycle-1 (~6.7 min) since curing takes place at earlier stages in time for Cycle-2. The estimated initial viscosity slightly deviates from the measured one, nevertheless this does not have an important effect on the thermo-mechanical analysis of the pultrusion process since the stiffness of the liquid resin is not high enough to build up stresses. The gelation is defined as the point at which the state of the resin changes from a viscous liquid to a rubbery gel. As reported in [26], the gelation occurs when the viscosity of the resin increases to infinity. However, measuring an infinite viscosity is not practical and hence it is accepted to define the gel point when the viscosity reaches a value of \(\sim 10 \times 10^5\) Pas [26,38]. Keeping this in mind the degree of cure at gelation is calculated approximately as 0.12 and 0.14 for Cycle-1 and Cycle-2, respectively (Fig. 7(right)) by obtaining the gel time at \(\eta \approx 10 \times 10^4\) Pas from Fig. 8. This indicates that the polyester system used in this study is a highly reactive resin. The predicted degree of cure at gelation is found to be in the conversion range (0.1–0.3) provided in [15].

Fig. 10 shows the development of \(G'\) and \(G''\) as a function of time for Cycle-1 and Cycle-2. Both \(G'\) and \(G''\) increase as the crosslinking reaction occurs. The gel time is taken as the point at which the crossover of \(G'\) and \(G''\) takes place during curing [26,39], nevertheless, it may not be equal to the one calculated at \(G' = G''\) for several resin systems [26,40]. The gel time determined from \(G' = G''\) is found to be somewhat less than the one obtained from \(\eta \approx 10 \times 10^4\) Pas for unsaturated polyester resins [41]. As seen from Fig. 10, the gel time is determined to be ~4 min and ~6 min for Cycle-2 and Cycle-1, respectively which is less than the gel time measured from \(\eta \approx 10 \times 10^4\) Pas.

4. Elastic modulus

A proper mathematical description of the different mechanical moduli is required for the calculation of the process induced stresses and the dimensional variations in pultrusion. Hence, a temperature- and cure-dependent modulus model is considered in the present study. A DMA (Metravib Viscoanalyser VA2000) is utilized in tension mode by applying a sinusoidal deformation/strain to the sample. The stiffness (modulus) and damping (tan \(\delta\)) are measured as a response in the DMA and 200 measuring points are used in the experiments. The modulus can be described by two components: an in-phase component, the storage modulus (elastic behaviour)
(E'), and an out-of-phase component, the loss modulus (E''). The damping (\tan \delta) is defined as the ratio of the loss modulus to the storage modulus and represents the energy dissipation in the sample. The peak of tan \delta at which the difference between E' and E'' is minimum indicates the glass transition temperature T_g.

The neat resin is first cured in the form of rectangular stripes using an oven. Subsequently, the fully cured (\alpha = 1.0) samples are initially conditioned at room temperature (25 °C) for approximately 5 min in the DMA. The sample size is specified as 30 x 5 x 2 mm (length x width x thickness). Dynamic heating scans are performed from 25 °C to 190 °C with a heating rate of 5 °C/min and a frequency of 6.22 Hz [42]. The static and dynamic load strains are set to 1% and 0.1%, respectively.

Fig. 11 shows the measured E' and E'' as well as tan \delta evolutions. It is seen that the peak of tan \delta is obtained at \sim 135 °C which corresponds to T_g at \alpha = 1.0. The moduli change with temperature and a transition (glassy to rubbery zone) inside the sample. In order to capture these variations properly, a modified cure hardening instantaneous linear elastic (CHILE) model [31,34] is developed for the measured resin modulus. Note that the developed tangent modulus is convenient for the incremental stress calculation (i.e. \sigma = E\varepsilon) [11]. The corresponding expression is given as:

$$E_i = \begin{cases} E_0; & T^* \leq T_{C1} \\ A_i \exp(K_i T^*); & T_{C1} < T^* < T_{C2} \\ E_1 + \frac{T_{C2} - T_{C1}}{T_{C2} - T_{C1}} (E_a - E_1); & T_{C2} < T^* < T_{C3} \\ E_a; & T_{C3} \leq T^* \end{cases}$$

(4)

where $T^*$ represents the difference between the instantaneous glass transition temperature ($T_g$) and the resin temperature $T$, i.e. $T^* = T - T_g$ [34]. $A_i$ and $K_i$ are the constants for the exponential term. The other model constants indicate the transition zones and are schematically shown in Fig. 12. Here, $T_{C1}$, $T_{C2}$, and $T_{C3}$ are defined as the critical temperatures and $E_0$, $E_1$ and $E_a$ are the corresponding elastic modulus values, respectively, at which the modulus behaviour is changed due to the phase transitions of the resin (viscous–rubbery–glassy state). More specifically, $E_0$ and $E_a$ can be considered as the elastic modulus in the viscous and glassy state, respectively. The glass transition temperature $T_g$ can be defined as a function of the degree of cure [34] in the following way:

$$T_g = T_g^0 + a_{T_g} \alpha$$

(5)

where $T_g^0$ is the glass transition temperature at $\alpha = 0$ and $a_{T_g}$ is a constant. For the sake of convenience, $T_g^0$ is assumed to be a much lower value than $T_g = 135 °C$ obtained from DMA for $\alpha = 1.0$. In the

---

**Table 3**

<table>
<thead>
<tr>
<th>T_{C1} (°C)</th>
<th>T_{C2} (°C)</th>
<th>T_{C3} (°C)</th>
<th>E_0 (GPa)</th>
<th>E_1 (GPa)</th>
<th>E_a (GPa)</th>
<th>A_i (GPa)</th>
<th>K_i (°C)</th>
</tr>
</thead>
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<td>60</td>
<td>30</td>
<td>110</td>
<td>0.0195</td>
<td>0.73</td>
<td>3.76</td>
<td>0.20</td>
<td>0.043</td>
</tr>
</tbody>
</table>

---

**Fig. 12.** Schematic representation of the elastic modulus evolution for the polyester resin.

**Fig. 13.** Comparison of the measured and predicted elastic modulus developments (log scale).

**Fig. 14.** The Predicted Young's modulus evolutions for different degree of cure values at glassy state.
present work, $T_g^0$ is hence assumed to be 0 °C as in [43] providing $\alpha_{tg} = 135$ °C. The variation in $T_g^0$ would not have an important effect on mechanical response of the processing part during the pulling process since the stiffness of the viscous resin is relatively small to build up induced stresses. Using $T_g = 135$ °C at $\eta = 1.0$, a least squares non-linear regression analysis is performed to obtain the constants in Eq. (4) which give the best agreement with the measured moduli. The estimated parameters are given in Table 3. The calculated elastic modulus evolution (best fit) is compared with the experimental data seen in Fig. 13. A good agreement is found between the measured and the predicted modulus evolution. Eq. (4) is further used to generate the modulus evolutions for different degree of cure values as shown in Fig. 14. It is seen that there is a shift in modulus development as the degree of cure increases. This is an expected outcome and agrees quite well with the experimental observations in [34].

5. Conclusions

The material characterization of an industrial "orthophthalic" polyester resin system specifically prepared for a pultrusion process was investigated by using experimental tests and developing the corresponding constitutive material models. A least squares non-linear regression analysis was conducted to fit the material models to the measured data. The evolution of the degree of cure as well as the cure rate was obtained over a wide temperature range using the isothermal and dynamic DSC scans. A temperature- and cure-dependent viscosity model was developed to estimate the rheological behaviour of the polyester system. A good fit was established between the predicted and the measured viscosity data. The gelation point was determined according to the viscosity value around $\eta \approx 10 \times 10^4$ Pas. The corresponding degree of cure value at gelation was found to be in the range 0.1–0.3 [15] according to the degree of cure evolutions predicted using the cure kinetics model. The gel time determined from $G > G_0$ was found to be less than the gel time measured from $\eta \approx 10 \times 10^4$ Pas. The DMA analysis was carried out to investigate the modulus development as a function of temperature and time. A significant variation was found in modulus due to the temperature change and transition (glassy to rubbery zone). Hence, a cure hardening and thermal softening modulus model (modified CHILE approach) was implemented to predict the elastic modulus evolution.

The proposed constitutive models are crucial to analyze and subsequently control the pultrusion process correctly and can be used for targeted numerical process modelling. The degree of cure and temperature distributions inside the pultruded part can be evaluated by means of the developed cure kinetics model. This gives an idea about the quality of the cure at the end of the process. The developed temperature- and cure-dependent viscosity model can be used to calculate the viscous force at the die–part interface. The proposed cure hardening and thermal softening modulus model can be used for the calculation of the process induced stresses and distortions during processing. Moreover, the developed constitutive models are essential in order to optimize the process via process models.

Acknowledgements

The author wishes to thank Dr. Roy Visser, Ivo Vroooijink and Nadia Vleugels from University of Twente (The Netherlands) for guidance and valuable discussions of the experiments. This work is a part of DeepWind project which has been granted by the European Commission (EC). Grant 256769 FP7 Energy 2010, under the FP7 program platform Future Emerging Technology.

References


“Pultrusion of a Vertical Axis Wind Turbine Blade Part-II: Combining the Manufacturing Process Simulation with a Subsequent Loading Scenario”

Ismet Baran, Jesper H. Hattel, Cem C. Tutum, Remko Akkerman

Pultrusion of a vertical axis wind turbine blade part-II: combining the manufacturing process simulation with a subsequent loading scenario

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Received: 22 December 2013 / Accepted: 11 May 2014 © Springer-Verlag France 2014

Abstract This paper in particular deals with the integrated modeling of a pultruded NACA0018 blade profile being a part of EU funded DeepWind project. The manufacturing aspects of the pultrusion process are associated with the preliminary subsequent service loading scenario. A 3D thermo-chemical analysis of the pultrusion process is sequentially coupled with a 2D quasi-static mechanical analysis in which the process induced residual stresses and distortions are predicted using the generalized plane strain elements in a commercial finite element software ABAQUS. The temperature- and cure-dependent resin modulus is implemented by employing the cure hardening instantaneous linear elastic (CHILE) model in the process simulation. The subsequent bent-in place simulation of the pultruded blade profile is performed taking the residual stresses into account. The integrated numerical simulation tool predicts the internal stress levels of the profile at the end of the bending analysis. It is found that the process induced residual stresses have the potential to influence the internal stresses arise in the structural analysis.

Keywords Pultrusion process · Residual/internal stress · Finite element analysis · Integrated modelling · Thermosetting resin.

Introduction

Pultrusion is a continuous manufacturing process in which constant cross sectional composite profiles are produced. A schematic view of the process is depicted in Fig. 1. The reinforcements (roving, continuous filament mat, etc.) are pulled through the pre-formers and wetted out in a thermosetting resin bath. The part is cured inside the heating die and cut into desired lengths by a cutting mechanism at the end of the process.

Pultruded products are foreseen to have potential for the replacement of some of the structural profiles such as the VAWT blades/components and the steel reinforcement of the concrete. The process has to be well characterized to improve the product quality in terms of the internal stress level arising during the service loading. The process induced residual stresses are generated by various mechanisms that inherently exist in the composite manufacturing processes such as the chemical shrinkage of the thermosetting resin and the mismatch in the coefficient of thermal expansion (CTE) of the reinforcement and the resin [1–3]. Therefore, the evolution of the process induced stresses as well as the distortions must be well investigated in order to have a better understanding of the mechanical behaviour of the pultruded products under service loading conditions. Since running a production line by trial and error is an expensive and time consuming task, the development of a numerical simulation...
Pultrusion technology was used to manufacture a vertical axis wind turbine (VAWT) blade as reported in [4]. The pultruded blade was then shaped into a troposkien (Darrieus design). Similarly, pultrusion is currently being considered in EU funded DeepWind Project [5, 6] in which a novel design concept is developed for a floating offshore (VAWT) based on the Darrieus design.

In literature, the pultrusion process has been investigated both numerically and experimentally. The main aim has been to understand the process by evaluating the development of the resin flow [7–9], temperature [10–12] and degree of cure profiles [13–15]. In addition, the frictional force inside the heating die has also been analysed in [16–18]. In these studies [7–18], well known numerical methods such as the finite difference method (FDM), the finite volume method (FVM) or the finite element method (FEM) have been utilized. All these contributions have only been dealing mainly with thermal modelling thus being able to predict characteristic temperature and cure degree behaviours for pultrusion. Typically the temperature of the composite is initially lagging behind the die temperature and subsequently it exceeds the die temperature during curing due to the internal heat generation of the resin [13]. In addition to these studies in the literature, the authors have contributed substantially with state-of-the-art models for pultrusion. This includes efficient thermo-chemical models [19, 20] together with performing probabilistic analysis of the process [21] and optimization simulations [22, 23]. Moreover, the authors have proposed the first models for the thermo-mechanical aspects of the pultrusion process including the evolution of the process induced stresses, distortions and mechanical properties [24, 25]. In these works [24, 25], a three dimensional (3D) transient thermo-chemical model is sequentially coupled with a 2D quasi static plane strain mechanical model for the pultrusion process using the FEM. This proposed approach, which is found to be computationally efficient, provides an increased understanding of the process by evaluating the development of the stresses and distortions as well as the mechanical properties during processing.

So far, an integrated modelling of a pultruded product particularly combining the manufacturing simulation with the subsequent service loading scenario has not been described in literature. The manufacturing aspects of the pultrusion process are hence associated with the subsequent loading simulation for the pultruded NACA0018 blade profile in the present work. More specifically, the residual stresses together with the final mechanical properties of the transversely isotropic pultruded product are subsequently transferred to the loading analysis in which a non-linear bending simulation of the NACA0018 profile is performed. A unidirectional (UD) glass/epoxy composite is considered for the process simulation. The temperature and the degree of cure profiles are first calculated in the 3D thermo-chemical analysis of the pultrusion. The process induced residual stresses and distortions are predicted in a 2D quasi-static mechanical analysis in which the generalized plane strain elements are utilized in ABAQUS [26]. A 3D transient Eulerian thermo-chemical analysis is coupled with a 2D quasi-static Lagrangian plane strain mechanical analysis of the pultrusion process [24]. The temperature- and cure-dependent resin modulus is calculated using the cure hardening instantaneous linear elastic (CHILE) approach provided in [27].
Numerical implementation

Energy and cure kinetics equations

The 3D transient energy equations for the composite and the die are given in Eq. 1 and Eq. 2, respectively for the thermo-chemical simulation of the pultrusion process. Here, \( x_1 \) is the pulling (longitudinal) direction; \( x_2 \) and \( x_3 \) are the transverse directions.

\[
\rho_C C_p \left( \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x_1} \right) = k_{x_1,c} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2,c} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3,c} \frac{\partial^2 T}{\partial x_3^2} + q
\]  
\[
\rho_d C_p \frac{\partial T}{\partial t} = k_{x_1,d} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2,d} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3,d} \frac{\partial^2 T}{\partial x_3^2}
\]  

where \( T \) is the temperature, \( t \) is the time, \( u \) is the pulling speed, \( \rho \) is the density, \( C_p \) is the specific heat, \( k_{x_i} \) and \( k_{x_i,c} \) are the thermal conductivities in the \( x_i \), \( x_2 \), and \( x_3 \) direction, respectively. The subscripts \( c \) and \( d \) correspond to the composite and the die, respectively. Lumped material properties are used and assumed to be constant. The volumetric internal heat generation \( q \) \([\text{W/m}^3]\) due to the exothermic reaction of the epoxy resin can be expressed as [12]:

\[
q = (1 - V_f) \rho_C H_r, R_r(\alpha, T)
\]

Table 1 Thermal properties used in the process simulation [12, 14]

<table>
<thead>
<tr>
<th></th>
<th>( \rho ) [kg/m(^3)]</th>
<th>( C_p ) [J/kg K]</th>
<th>( k_{x_1} ) [W/m K]</th>
<th>( k_{x_2}, k_{x_3} ) [W/m K]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
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</tr>
<tr>
<td>t1.3</td>
<td></td>
<td></td>
<td></td>
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</tr>
</tbody>
</table>

and the corresponding relation is given in Eq. 4. \( dH(t)/dt \) is also obtained from the DSC analysis.

\[
R_r(\alpha, T) = \frac{d\alpha}{dt} = \frac{1}{H_r} \frac{dH(t)}{dt}
\]

where \( K_o \) is the pre-exponential constant, \( E \) is the activation energy, \( R \) is the universal gas constant and \( n \) is the order of reaction (kinetic exponent). \( K_o, E, \) and \( n \) can be obtained by a curve fitting procedure applied to the experimental data evaluated using the DSC [14].

The cure rate in (Eq. 4) can be written as:

\[
\frac{d\alpha}{dt} = \frac{d\alpha}{dx_1} + \frac{d\alpha}{dt} = \frac{d\alpha}{dx_1} + u \frac{d\alpha}{dx_1}
\]

and from Eq. 5, the resin kinetics equation can be expressed in the Eulerian frame as:

\[
\frac{\partial T}{\partial t} = R_r(\alpha, T) - u \frac{\partial \alpha}{\partial x_1}
\]

which is used in the thermo-chemical model.

The temperature and the degree of cure distributions at steady state are calculated in ABAQUS. The non-linear internal heat generation (Eq. 3) together with the resin kinetics equation (Eq. 6) is coupled with the energy equation (Eq. 1) in an explicit manner in order to obtain a straightforward and fast numerical procedure. This procedure is performed until the steady state conditions are satisfied. The corresponding procedure is given as a flowchart in Fig. 2.

The degree of cure is subsequently updated explicitly for each control volume using Eq. 6 in its discretized form. To reach the steady state conditions, the convergence limits of the temperature and the degree of cure are defined to be 0.001 °C and 0.0001, respectively as seen in Fig. 2. In order to obtain a stable result and to overcome the possible oscillatory behaviour in the numerical implementation, the upwind scheme is used for the term \((u \partial \alpha/\partial x_1)\) in the resin kinetics equation [20].

Incremental residual tress implementation

The CHILE model given in Eq. 7 [27] is implemented for the calculation of the instantaneous resin elastic modulus

Table 2 Epoxy resin kinetic parameters [12, 14]

<table>
<thead>
<tr>
<th>t2.2</th>
<th>( H_r ) [kJ/kg]</th>
<th>( K_o ) [s]</th>
<th>( E ) [kJ/mol]</th>
<th>( n )</th>
</tr>
</thead>
<tbody>
<tr>
<td>t2.3</td>
<td>324</td>
<td>192,000</td>
<td>60</td>
<td>1.69</td>
</tr>
</tbody>
</table>
where \( E_r^0 \) and \( E_r^\infty \) are the uncured and fully cured resin moduli, respectively. \( T_{C1} \) and \( T_{C2} \) are the critical temperatures at the onset and completion of the glass transition, respectively, \( T^* \) represents the difference between the instantaneous glass transition temperature \( (T_g) \) and the resin temperature, i.e. \( T^* = T_g - T \) [27]. The evolution of the \( T_g \) as a function of degree of cure is modelled by the Di Benedetto equation [28] and expressed as:

\[
\frac{T_g - T_{g0}}{T_{g\infty} - T_{g0}} = \frac{\lambda \alpha}{1 - (1 - \lambda) \alpha}
\]

(8)

where \( T_{g0} \) and \( T_{g\infty} \) are the glass transition temperatures of uncured and fully cured resin, respectively, \( \lambda \) is a constant used as a fitting parameter.

The effective mechanical properties including the CTEs and the chemical shrinkage strains together with the thermal strains are calculated using the self consistent field micromechanics (SCFM) approach provided by Bogetti and Gillespie [29]. Incremental linear elastic approach is used for the calculation of the residual stresses and distortions as suggested in [27]. The incremental process induced strain \( (\dot{\varepsilon}_{pr}) \), which is composed of the incremental thermal strain \( (\dot{\varepsilon}_{th}) \) and the chemical shrinkage strain \( (\dot{\varepsilon}_{ch}) \), is defined for the stress-strain relation [24]. The incremental total strain \( (\dot{\varepsilon}_{tot}) \) is defined as the sum of the incremental mechanical strain \( (\dot{\varepsilon}_{mech}) \), \( \dot{\varepsilon}_{th} \) and \( \dot{\varepsilon}_{ch} \) and written as:

\[
\dot{\varepsilon}_{tot} = \dot{\varepsilon}_{mech} + \dot{\varepsilon}_{th} + \dot{\varepsilon}_{ch}
\]

(9)

The incremental stress tensor \( \dot{\sigma}_{ij} \) is calculated using the material Jacobian matrix \( (J) \) based on the incremental mechanical strain tensor \( \dot{\varepsilon}_{mech} \) in ABAQUS [26] and the corresponding expression is given as:

\[
\dot{\sigma}_{ij} = J \dot{\varepsilon}_{mech}
\]

(10)

The details of the relations between the stress and strain tensors used in the present finite element implementation can be found in [24].

**Pultrusion process model**

Thermo-chemical analysis

A 3D transient thermo-chemical analysis of the pultrusion process is carried out for a UD NACA0018 blade profile using a Eulerian frame of reference. The pultrusion model is taken from similar set-ups available in the literature [12, 14, 24]. A glass/epoxy based composite and steel are used for the blade and the die block, respectively. The material properties of the composite and the resin kinetic parameters are listed in Table 1 and Table 2, respectively. The fiber volume fraction \( (V_f) \) of the composite is 0.639 [14]. A schematic view of the process set-up is shown in Fig. 3. The length of the die and the post die \( (L_{conv}) \) are determined to be 0.915 m and 9.15 m, respectively. Cooling channels are located 100 mm under the first heating regions [12, 14]. Hence, all the nodes at the layers D-D and E-E indicated in Fig. 3 are set to the temperature of the cooling water \( (50 \, ^\circ C) \) during the whole process. Three heating zones having prescribed set temperatures of 171-188-188 \(^\circ C\) [12, 14] are defined as seen in Fig. 3. The spacing between the heating zones is 15 mm. The temperature and the degree
of cure of the composite at the die inlet are set to 30 °C (resin bath temperature) and 0, respectively. The remaining exterior surfaces of the die are exposed to ambient temperature with a convective heat transfer coefficient of 10 W/m²K except for the surfaces located at the heating regions. Similar convective boundaries are also defined for the outer surfaces of the pultruded profile at the post die region. The details of the cross section and the meshing are depicted in Fig. 4 for the part and the die. It is seen that the chord length of the processed NACA0018 blade is 100 mm and the maximum thickness of the cross section is 18 mm. An element length of 15 mm is used in the pulling direction.

Thermo-chemical-mechanical analysis

In the 2D quasi-static mechanical analysis the cross section of the blade is assumed to be moved through the pulling direction (Lagrangian frame) meanwhile tracking the corresponding temperature and degree of cure profiles calculated in the 3D thermo-chemical analysis (Eulerian frame). A representation of the coupling of the 3D thermo-chemical model with the 2D generalized plane strain mechanical model is shown in Fig. 5 [24]. A plane strain assumption is considered in the analysis since the length of the processing profile (≈ 10 m) is much higher than the cross sectional dimensions (0.1 m). Quadratic generalized plane strain elements are employed in ABAQUS. The constituent mechanical properties are listed in Table 3 and the material properties used in the CHILE approach are given in Table 4.

The rigid body surfaces are defined instead of modelling the whole meshing of the die, since the die is assumed to be rigid as compared to the part. A mechanical contact formulation is employed between the rigid surfaces and the part which allows separation at the interface due to the chemical shrinkage or the thermal contraction. However, any expansion of the part beyond the tool interface is restricted. Note that the friction force at the contact condition is assumed to be zero (sliding condition). A schematic

Table 3 The mechanical properties of the glass fiber and the epoxy resin [29]

<table>
<thead>
<tr>
<th></th>
<th>Young’s Modulus [GPa]</th>
<th>Poisson’s Ratio</th>
<th>CTE [ppm/°C]</th>
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<tr>
<td>t3.3</td>
<td>Glass fiber</td>
<td>73</td>
<td>0.22</td>
</tr>
<tr>
<td>t3.4</td>
<td>Epoxy resin (glassy state)</td>
<td>3.447</td>
<td>0.35</td>
</tr>
</tbody>
</table>

Table 4 The parameters used in the CHILE approach (Eq. 7) and in the calculation of the glass transition temperature (Eq. 8) [24, 27–29]

<table>
<thead>
<tr>
<th></th>
<th>TC1 [°C]</th>
<th>TC2 [°C]</th>
<th>E0 [MPa]</th>
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<th>Tg∞ [°C]</th>
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<td>3.447e3</td>
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</tr>
</tbody>
</table>
view of the generalized plane strain model including the rigid surfaces and the mechanical boundary conditions (BCs) are shown in Fig. 5. A symmetric mechanical BC on the line C-C shown in Fig. 5 is applied in the $x_3$-direction. In addition, the point A shown in Fig. 5 is assumed to be also fixed in the $x_2$-direction based on the assumption that the innermost region of the composite behaves more stationary as compared with the outermost regions. In the present study the total volumetric shrinkage of the epoxy resin is assumed to be 6% [30].

Results and discussions

The temperature and the degree of cure developments are predicted in the 3D thermo-chemical analysis of the pultrusion and the results are depicted in Fig. 6 for certain points on the pultruded blade, i.e. point A and B. Note that these points are located at the thickest section of the part seen in Fig. 4. The corresponding contour plots are shown in Fig. 7 for the blade cross section at the die exit ($x_1 = 0.915$ m) and at the end of the process ($x_1 \approx 10$ m). The pulling speed is

Fig. 6 The temperature (top) and the degree of cure (bottom) developments at point A and B
Fig. 7 The contour plots of the temperature (left) and the degree of cure (right) at the die exit ($x_1 = 0.915$ m) and end of the process ($x_1 \approx 10$ m). Note that the legend of the plots is not same set to 200 mm/min. It is seen that a non-uniform temperature and degree of cure distributions are found to prevail over the cross section of the part. It is seen from Fig. 6 that point B cures earlier than point A since it is closer to the heaters. The thinner regions of the part have a higher degree of cure value as compared with the thicker regions as seen from the contour plot in Fig. 7 (the degree of cure at the die exit). At the post die region the curing continues increasing since the temperature of the blade is high enough to generate internal heat after the die exit. Therefore, the entire part is found to be almost fully cured at the end of the process, i.e. the mean degree of cure is found to be approximately 0.97 at the end of the process. The maximum temperature is calculated as around $215 \, ^\circ \text{C}$ near the die exit.

The calculated temperature and the degree of cure fields are mapped to the 2D quasi-static mechanical analysis. The development of the transverse process induced residual stresses are predicted and the results are depicted in Fig. 8.

Fig. 8 The development of the process induced stresses for point A and B.
Fig. 9 The undeformed contour plots of the longitudinal normal stress ($S_{11}$), the transverse normal stresses ($S_{22}$ and $S_{33}$) and the transverse shear stress ($S_{23}$) at the end of the pultrusion process. Note that the legend of the plots is not same

for the points A and B. Here, $S_{11}$ is the normal stress in the $x_1$-direction (longitudinal), $S_{22}$ and $S_{33}$ are showing the normal stresses in the $x_2$-direction (horizontal, transverse) and in the $x_3$-direction (vertical, transverse), respectively. Note that the results should be seen together with the temperature and the cure degree curves given in Fig. 6. The overall residual stress evolution at point A and B can be explained as follows: The region closest to the heaters (e.g. point B) cures first which poses a constraint against the inner regions (e.g. point A) of the pultruded profile. Due to this, compression is exhibited towards the regions near the die. When the internal region starts curing rapidly, due to the internal constraint against the outer region, tension and compression prevail at the inner and outer regions, respectively. This can be seen in Fig. 8b,d that point A is under tension and point B is under compression at the end of the process, while upholding the self static equilibrium in which there is no applied external load to the processing part. It is found that the longitudinal stress values are found to be higher than the transverse stress values inside the die. However, this turns out to be the other way around at the end of the process. Regarding the transverse stresses, the magnitude of the overall $S_{33}$ values are found to be smaller than the $S_{22}$ values since the thickness of the profile (18 mm) is thinner than the chord of the profile (100 mm). The corresponding contour plots of $S_{11}$, $S_{22}$ and $S_{33}$ together with $S_{23}$ (transverse shear stress on the $x_2$-$x_3$ plane) are shown in Fig. 9 for the end of the process. The magnitude of $S_{23}$ is found to be smaller than the magnitude of the normal stresses.

The deformation field is given as a deformed contour plot of the composite cross section at the end of the process in Fig. 10. The maximum process induced distortions are calculated approximately as 0.39 mm and 0.066 mm in the $x_2$- and $x_3$-direction, respectively.

The predicted effective mechanical properties at the end of the process are given in Table 5 for the pultruded NACA0018 blade which is in the glassy state. Here, $E$ is the elastic modulus, $G$ is the shear modulus, $\nu$ is the Poisson’s ratio and $\alpha_i$’s are the CTEs. It is seen that the fibers play more significant role for the development of the mechanical properties in the longitudinal direction such as $E_1$, $\nu_{12}$ and $\alpha_1$. On the other hand, $E_2$, $G_{12}$

Table 5 The mechanical properties of the UD composite at fully cured glassy state. $V_f$ is 0.639 for the predicted values and 0.6 for the measured values [31]

<table>
<thead>
<tr>
<th>Predicted (SCFM)</th>
<th>Measured [31]</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_1$</td>
<td>48.2</td>
<td>45.6</td>
</tr>
<tr>
<td>$E_2 = E_3$</td>
<td>12.4</td>
<td>16.2</td>
</tr>
<tr>
<td>$G_{12} = G_{13}$</td>
<td>4.42</td>
<td>5.83</td>
</tr>
<tr>
<td>$\nu_{12} = \nu_{13}$</td>
<td>0.261</td>
<td>0.278</td>
</tr>
<tr>
<td>$\alpha_1$</td>
<td>6.4</td>
<td>8.6</td>
</tr>
<tr>
<td>$\alpha_2 = \alpha_3$</td>
<td>30.3</td>
<td>26.4</td>
</tr>
</tbody>
</table>
Fig. 11 A schematic view of the subsequent loading scenario (i.e. bending of the blades)

and $\alpha_2$ are dominated by the resin matrix as expected. The predicted values are compared with the experimental values taken from literature [31] for a UD glass/epoxy laminate with $V_f = 60\%$. It is seen from Table 5 that a good agreement is found showing that the implementation of the SCFM approach gives reasonable results for the calculation of the effective mechanical properties of the processing composite. The slight difference between the predicted and the measured properties is due to the difference in $V_f$.

Preliminary structural bent-in place simulations

Model description

In the subsequent loading scenario the transversely isotropic pultruded blade profile is assumed to be bent into Darrieus shape (i.e. arched-blades) taking the residual stresses into account. A schematic view of the bent-in place simulation of the blade is shown in Fig. 11. The bent shape is obtained by applying a displacement on one end of the profile having an initial total length of 3.7 m and keeping the other end fixed (i.e. hinged BC). In Darrieus type VAWTs, the length/diameter ratio of the rotor, i.e. $h/(2r)$ seen in Fig. 11, has an important effect on the aero dynamical behaviour of the turbines. In the present study, $h/(2r) \approx 2.64$ is used by applying a displacement ($u$ in Fig. 11) value of approximately 0.3 m. The length $(h)$ and the radius $(r)$ of the rotor at the end of the simulation are approximately 3.4 m and 0.645 m, respectively. The obtained $h/(2r)$ is found to be very close to the one used in [4] as 2.7 for a 3-bladed VAWT.

A 3D non-linear structural static analysis is performed by using the quadratic solid elements in ABAQUS. The residual stresses together with the final mechanical properties of the profile predicted in the pultrusion process simulation are transferred to the bending simulation. The residual stresses are treated as a pre-stress condition utilizing the user defined routines in ABAQUS before the bending simulation. This procedure is given in Fig. 12 as a flowchart. Here, it should be noted that the primary equilibrium conditions has to be satisfied in the 3D bending simulation. Therefore, a 3D static equilibrium analysis is carried out without applying...
**Fig. 13** Contour plots showing the stress distribution with/without residual stresses for section M at the end of the bending simulation. Note that the legend of the plots is not same.

**Fig. 14** The through-thickness stress variation at section M with/without residual stresses.
any mechanical loads or BCs. The aim is to uphold the stress
equilibrium after invoking the pre-stress condition in which
the residual stresses are transferred from the process simula-
tion. Subsequently, the loading scenario depicted in Fig. 11
is carried out and the internal stress levels of the bent profile
are evaluated.

Results and discussions

The bending simulations are performed taking the residual
stresses into account as a pre-stress condition as aforemen-
tioned. According to the loading scenario, the maximum
normal stresses are expected to be built up at the center of
the blade profile (section “M” depicted in Fig. 11) in the lon-
gitudinal direction, i.e. \( x_1 \)-direction. Hence, the evaluation
of the internal stresses at section M are analysed in detail.

The contour plots of the normal stresses at section M at the
end of the bending simulation are shown in Fig. 13 with and
without taking the residual stresses into account. It is seen
that the effect of the residual stresses are more dominant for
the transverse directions (i.e. for \( S_{22} \) and \( S_{33} \)) as compared
with the longitudinal component (\( S_{11} \)) which is obviously
more critical for this type of loading scenario. The magni-
tude of \( S_{11} \) is found to be much larger than \( S_{22} \) and \( S_{33} \) as
expected. After obtaining the equilibrium state, it is found
that the maximum compression stress for \( S_{11} \) is increased
approximately from 216 MPa to 220 MPa with the residual
stresses. However, the \( S_{11} \) value for the maximum tensile
stress is decreased from approximately 213 MPa to 210
MPa. The stress levels for \( S_{22} \) and \( S_{33} \) are relatively small
as compared to \( S_{11} \). The through-thickness stress variations
are given in Fig. 14 for the thickest region of section M. It is
seen that the residual stresses promote the \( S_{22} \) level; on the
other hand the \( S_{33} \) level decreases with taking the residual
stresses into account. This shows that the residual stresses
have the potential to increase or decrease the internal stress
level depending on the service loading scenario.

The longitudinal tensile and compressive strength values
of the UD glass/epoxy composite for a \( V_f = 0.6 \) are given
as 1280 MPa and 800 MPa, respectively in [31]. The corre-
sponding transverse tensile and compression strength values
are 40 MPa and 145 MPa, respectively [31]. The predicted
primary internal stress levels shown in Fig. 13 are found
to be much smaller than the corresponding strength levels
given in [31] based on this specific NACA0018 profile.

Conclusions

A 3D thermo-chemical analysis of the pultrusion process for a
NACA0018 blade profile was first performed to obtain the
temperature and the degree of cure profiles during the pro-
cess. Afterwards, the process induced residual stresses and
distortions were predicted in a 2D quasi-static mechanical
analysis of the pultrusion process for the blade. The inte-
grated modelling of the pultruded NACA0018 blade profile
was carried out by combining the manufacturing process
simulation with the subsequent loading scenario. The pre-
dicted residual stresses together with the final mechanical
properties of the transversely isotropic pultruded product
were transferred to the loading simulation in which a non-
linear bent-in place simulation of the blade was performed.
The residual stresses were considered as a pre-stress con-
dition using a user defined routines in ABAQUS and the
quadratic elements were used in the bending simulation.

Non-uniform temperature and degree of cure distribu-
tions were obtained in the 3D thermo-chemical analysis of
the pultrusion process. It was found that the curing con-
tinued at the post-die region since the temperature of the
composite near the die exit was high enough to generate the
internal heat. At the end of the process, tensile and
compressive stresses were found to prevail at the inner and
outer regions of the pultruded profile. The proposed 3D/2D
approach was found to be computationally efficient and fast
for the calculation of the residual stresses and distortions
which are expected to be built up at the center of the blade
profile (section “M” depicted in Fig. 11) in the longitudinal
direction, i.e. \( x_1 \)-direction. Hence, the evaluation
of the internal stresses at section M are analysed in detail.

The contour plots of the normal stresses at section M at the
end of the bending simulation are shown in Fig. 13 with and
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Acknowledgments This work is a part of DeepWind project which
has been granted by the European Commission (EC) under the FP7
program platform Future Emerging Technology.

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“Optimization of the Thermosetting Pultrusion Process by Using Hybrid and Mixed Integer Genetic Algorithms”

Ismet Baran, Cem C. Tutum, Jesper H. Hattel

Optimization of the Thermosetting Pultrusion Process by Using Hybrid and Mixed Integer Genetic Algorithms

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Received: 1 June 2012 / Accepted: 21 June 2012 / Published online: 12 July 2012 © Springer Science+Business Media B.V. 2012

Abstract In this paper thermo-chemical simulation of the pultrusion process of a composite rod is first used as a validation case to ensure that the utilized numerical scheme is stable and converges to results given in literature. Following this validation case, a cylindrical die block with heaters is added to the pultrusion domain of a composite part and thermal contact resistance (TCR) regions at the die-part interface are defined. Two optimization case studies are performed on this new configuration. In the first one, optimal die radius and TCR values are found by using a hybrid genetic algorithm based on a sequential combination of a genetic algorithm (GA) and a local search technique to fit the centerline temperature of the composite with the one calculated in the validation case. In the second optimization study, the productivity of the process is improved by using a mixed integer genetic algorithm (MIGA) such that the total number of heaters is minimized while satisfying the constraints for the maximum composite temperature, the mean of the cure degree at the die exit and the pulling speed.

Keywords Pultrusion · Finite difference · Optimization · Genetic algorithms · Thermal contact resistance

1 Introduction

The pultrusion process which was conceptualized in the 1950s is one of the simplest manufacturing processes for high strength/weight ratio composite structural members having constant cross-sectional profiles such as stiffeners, door/window frames, hollow sections etc. Since it is a continuous process, i.e. creating continuous composite profiles, there is little waste material arising at the end of the process. A schematic view of the process is seen in Fig. 1. Fibers and resin matrix are pulled through a heated die by a pulling mechanism. At the end a saw mechanism cuts the product into the desired length.
The curing of the resin system is the primary mechanism which is related with the chemical behavior of the resin. After some point in time inside the heating die, the phase of the resin changes from liquid to gel. When the temperature of the composite reaches the exothermic reaction temperature of resin, the chemical reaction process is initiated in which internal heat is generated. Following this, curing of the composite continues and finally the cured product is obtained. However at some position inside the heating die, while curing of the composite is progressing, chemical shrinkage of resin which causes separation of the composite and die takes place. Hence an air gap might be observed at the die-part interface. This situation affects the temperature distribution and degree of cure inside the composite part since non-perfect thermal contact is obtained. For this reason the thermal contact resistance (TCR), which can also be expressed by the heat transfer coefficient (HTC), between the die and the composite part has to be taken into account to obtain reliable results from the numerical model. In [1] and [2] the significance of chemical shrinkage and HTC on the temperature distribution in the composite and die was mentioned but the HTC at the die-part interface was not taken into account in the actual simulation of the pultrusion process. As a consequence of this, higher numerical temperature levels were obtained as compared to experimental ones especially near the last section of the die/composite where the chemical shrinkage plays a vital role [2].

In the literature several studies have been investigated related with numerical modeling of the pultrusion process [3–8]. Three dimensional steady state die and post die analysis were performed numerically for temperature and cure degree profiles at the centerline of a composite rod and the results were experimentally validated by Valliappan et al. [3]. Transient pultrusion models of various irregular geometries such as U, S and a hollow section were analyzed in [4]. Pultrusion of a flat plate was studied numerically and experimentally in [5] and the same study was validated numerically in [6] by using the general purpose finite element software package LUSAS.

In addition to the modeling and experimental studies discussed above, process optimization studies in pultrusion have been performed by several researchers [9–13]. In order to improve the quality of the product and the efficiency of the process, i.e. through minimization of the power consumption in the process or maximization of the pulling speed as well as the exit degree of cure, optimum process parameters such as the pulling speed, the temperature of the heaters and the coolers, etc. satisfying certain specific process constraints are investigated with the use of different optimization approaches [9–13]. In [9–11], the mathematical relationship between the cure degree at the exit cross section and the design parameters such as the temperature of heaters as well as the pulling speed and the power of the heaters were investigated for the pultrusion simulation of a C-shaped cross section. The optimization was performed on this relationship by using the steepest descend algorithm. The same model in, i.e. the pultrusion of a C-shaped cross section [7], was optimized by the use of a genetic algorithm and the derivative-free simplex method [12]. The GA was utilized to find a suitable starting point for the simplex method due to the fact that the simplex method highly depends on the starting point and it may find a local minimum rather than the global minimum. The variation of the cure degree evaluated at the exit cross section was
minimized by an iterative procedure based on the combination of the above mentioned techniques. Multi-objective optimization was performed on a pultrusion process model utilizing finite element and finite difference methods by Chen et al. [13] in which the multi-objective problem was converted to a single objective problem by using predefined weightings between the objectives. In this scalarized optimization problem, the combination of the artificial neural network (ANN) and the GA was proposed to find the optimal solution. The goals were reduction of the power consumption of the die and improvement of the productivity, i.e. increase in the pulling speed, while guaranteeing the quality which can be determined by the degree of cure of the composite product.

In the present study, a steady state pultrusion simulation of a composite rod with a given prescribed steady state die-part interface temperature distribution, which has already been analyzed numerically and experimentally in [3] is used as a validation case. The control volume based finite difference (CV/FD) method is utilized for the numerical model of the pultrusion process. Following the validation case, a cylindrical die block is attached to the same composite geometry with cylindrical heaters. Two optimization case studies are performed by using this new configuration. In the first one the die radius and nine equally spaced TCR values at the die-part interface are optimized such that the same centerline temperature profile of the composite is obtained as in the validation case. For this purpose, the hybrid genetic algorithm based on a combination of a genetic algorithm and the constrained minimization subroutine “fmincon” in MATLAB [14] is used. After obtaining the unknown characteristics of the new pultrusion domain, i.e. the die radius and nine TCR values, the productivity of the process is optimized by using a mixed integer genetic algorithm (MIGA) where the total number of heaters is minimized (mixed integer problem). The MATLAB mathematical computing environment is used for both the simulations and the optimization procedures.

2 Governing Equations

2.1 Energy Equations

The steady state heat transfer equations in a cylindrical coordinate system for the composite rod (Eq. 1) and the die block (Eq. 2) express the conservation of energy principle such that the rate of the energy change in a point equals the sum of the net rate of energy transferred to the point and the internal heat generation (for the composite rod only).

\[
\rho_c C_p c \left( u \frac{\partial T}{\partial z} \right) = k_{c,\alpha} \frac{\partial^2 T}{\partial z^2} + \frac{k_{c,\beta}}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right) + q
\]

(1)

\[
0 = k_{d,\alpha} \frac{\partial^2 T}{\partial z^2} + \frac{k_{d,\beta}}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right)
\]

(2)

where \( T \) is temperature, \( u \) is the pulling speed, \( \rho \) and \( C_p \) are the density and the specific heat respectively, \( k_c \) and \( k_d \) are thermal conductivities in axial direction (z) and in radial direction (r), respectively. The subscripts \( c \) and \( d \) correspond to composite and die, respectively. Lumped material properties are used for the composite and all the material properties used in
the simulation remain constant. The internal heat generation \( q \) \([\text{W/m}^3]\) due to the exothermic reaction of epoxy resin is expressed as

\[
q = (1 - V_f) \rho_r Q
\]  

(3)

where \( V_f \) is the fiber volume ratio and \( Q \) is the specific heat generation rate \([\text{W/kg}]\) due to the resin exothermic cure reaction.

2.2 Resin Kinetics

Resin kinetics is an important phenomenon which is related with the exothermic chemical reaction of the resin inside the die. This phenomenon directly affects the curing of the resin. The degree of cure \( \alpha \) can be written as the ratio of the amount of heat generated \( H(t) \) during curing, to the total heat of reaction \( H_{tr} \):

\[
\alpha = \frac{H(t)}{H_{tr}}
\]  

(4)

Generally, resin kinetic models which relate the rate of resin reaction, \( R_r \), to the temperature \( T \) and degree of cure \( \alpha \) can be given by an Arrhenius equation (Eq. 5) and the specific heat generation rate, \( Q \) (in Eq. 3), can be calculated as shown in Eq. 6 [6].

\[
R_r(\alpha) = \frac{d\alpha}{dt} = \frac{1}{H_{tr}} \frac{dH(t)}{dt} = K_0 \exp\left(-\frac{E}{RT}\right) (1 - \alpha)^n
\]  

(5)

\[
Q = \frac{dH(t)}{dt} = H_{tr} R_r(\alpha)
\]  

(6)

where \( K_0 \) is a pre-exponential constant, \( E \) is the activation energy, \( R \) is the universal gas constant and \( n \) is the order of reaction (kinetic exponent). \( H_{tr}, K_0, E \) and \( n \) are experimentally evaluated parameters using differential scanning calorimetry (DSC) [3]. By using the chain rule the rate of cure degree can be expressed as,

\[
\frac{d\alpha}{dt} = \frac{\partial \alpha}{\partial t} + \frac{\partial \alpha}{\partial z} \frac{dz}{dt} = \frac{\partial \alpha}{\partial t} + u \frac{\partial \alpha}{\partial z}
\]  

(7)

\[
\frac{\partial \alpha}{\partial t} = R_r(\alpha) - u \frac{\partial \alpha}{\partial z}
\]  

(8)

where it is the expression in Eq. 8 which is used in the numerical model.

3 Numerical Implementation

The energy equations for the composite part and die block are discretized by using the CV/FD method. This discretization of Eq. 1 and Eq. 2 is shown in Eq. 9 and Eq. 10, respectively. In this numerical scheme the concept of the total thermal resistances \( R \) (K/W), which can be defined as the temperature difference divided by the heat flow between two adjacent control volumes, is used. Thermal resistances for an interior control volume are seen in Fig. 2. At the die-part interface the TCR value describing the thermal resistance is defined in addition to
the other thermal resistances in the radial direction. Since the internal heat generation inside the composite is highly nonlinear, i.e. it depends on the rate of resin reaction, the source term \( q \) in the energy equation is coupled with the energy equation in an explicit manner in order to obtain a stable and fast numerical procedure. The degree of cure (in Eq. 8) is calculated explicitly therefore. In order to overcome oscillatory behavior in the numerical implementation the upwind scheme is used for the convective term in the energy equation and for discretization of the cure degree in the resin kinetics equation. The built-in equation solver in MATLAB is used to solve the equation system arising from utilized numerical schemes.

\[
\rho_{i,j} C_{p_{i,j}} V_{i,j} \left( \frac{u T_{i,j} - T_{i-1,j}}{\Delta z_{i,j}} \right) = \left( \frac{T_{i-1,j} - T_{i,j}}{Rz_{i-1,j} + Rz_{i,j}} + \frac{T_{i+1,j} - T_{i,j}}{Rz_{i+1,j} + Rz_{i,j}} \right) + \left( \frac{T_{i,j-1} - T_{i,j}}{Rr_{i,j}^{up} + Rr_{i,j}^{down}} + \frac{T_{i,j+1} - T_{i,j}}{Rr_{i,j+1}^{up} + Rr_{i,j}^{down}} \right) + q 
\]

and

\[
0 = \left( \frac{T_{i-1,j} - T_{i,j}}{Rz_{i-1,j} + Rz_{i,j}} + \frac{T_{i+1,j} - T_{i,j}}{Rz_{i+1,j} + Rz_{i,j}} \right) + \left( \frac{T_{i,j-1} - T_{i,j}}{Rr_{i,j}^{up} + Rr_{i,j}^{down}} + \frac{T_{i,j+1} - T_{i,j}}{Rr_{i,j+1}^{up} + Rr_{i,j}^{down}} \right)
\]

\[
Rz_{i,j} = \frac{\Delta z_{i,j}}{2k_{z,i,j} A_{z,i,j}}, \quad Rr_{i,j}^{up} = \frac{\ln \left( \frac{r_{i+1,j}}{r_{i,j}} \right)}{2\pi k_{r,i,j}}, \quad Rr_{i,j}^{down} = \frac{\ln \left( \frac{r_{i,j-1}}{r_{i,j}} \right)}{2\pi k_{r,i,j}}
\]

where \( V_{i,j} = 2\pi r_{i,j} \Delta r_{i,j} \Delta z_{i,j} \) is the volume, \( A_{z,i,j} = 2\pi r_{i,j} \Delta r_{i,j} \) is the area of the interior control volume in the axial direction where the axial heat flux enters the interior control volume, \( Rz \)'s are the thermal resistances in the axial direction and \( Rr^{up} \) and \( Rr^{down} \) are the thermal
resistances in the radial direction [15]. The configuration of thermal resistances can be seen in Fig. 2. \( \Delta r_{i,j} \) and \( \Delta z_{i,j} \) are the dimensions of the control volume in radial and axial direction respectively. A steady state discretization of the cure degree relation (Eq. 8) can be written as

\[
u \frac{a_{i,j} - a_{i-1,j}}{\Delta z} = R_r(a)
\]  

(12)

4 Validation Case

Two dimensional axisymmetric pultrusion of a composite rod without die block is simulated as a validation case. The graphite fiber reinforcement (Hercules AS4-12K) and epoxy resin (SHELL EPON9420/9470/537) system is used for the composite. Material properties of the composite components and resin kinetic parameters are given in Tables 1 and 2 respectively. The lumped thermal conductivities are obtained by using the mass fraction technique where the fiber and resin conductivities are considered in parallel for both directions according to [3]. The model geometry and boundary conditions are seen in Fig. 3. The measured die wall temperature profile, \( T_w(z) \), is given in [3]. The length \( L \) and the radius of the composite rod \( r_c \) are 915 mm and 4.75 mm respectively. At the die inlet the degree of cure is equal to zero and the temperature of the composite \( T_{left} \) is taken as the resin bath temperature (~38 °C). Adiabatic boundaries are defined at the centerline and at the right side of the composite rod. The convergence limits for the temperature and degree of cure are set to 0.001 °C and 0.0001, respectively.

Steady state temperature and cure degree profiles are obtained with a pulling speed of 30 cm/min (5 mm/s) and a 62.2 % fiber volume ratio. 4 (radial)×150 (axial) control volumes and 6 (radial)×152 (axial) nodes are used for the thermo-chemical simulation of the composite rod. The steady state centerline temperature in Fig. 4 (top) and cure degree distributions in Fig. 4 (bottom) match well with those in [3]. This shows that the developed numerical scheme is stable and converged to the correct solution. The centerline temperature becomes higher than the die wall temperature after approximately 0.4 m due to the exothermic internal heat generation inside the composite. The centerline cure degree at the die exit is calculated as 0.84.

5 Optimization Case Studies

In the optimization case studies a cylindrical die block is added to the original composite geometry domain defined in the validation case (see Fig. 5). Nine equally spaced TCR regions (each of ~100 mm) along the axial direction are defined at the die-composite interface. Two different case studies are investigated. In the first one, optimal characteristics

**Table 1 Material properties**

<table>
<thead>
<tr>
<th>Material</th>
<th>( \rho ) (kg/m(^3))</th>
<th>( C_p ) (J/kg K)</th>
<th>( k_z ) (W/m K)</th>
<th>( k_r ) (W/m K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Epoxy Resin</td>
<td>1260</td>
<td>1255</td>
<td>0.2</td>
<td>0.2</td>
</tr>
<tr>
<td>Graphite</td>
<td>1790</td>
<td>712</td>
<td>66.0</td>
<td>11.6</td>
</tr>
<tr>
<td>Lumped (( V_f = 62.2 % ))</td>
<td>1589.66</td>
<td>874.69</td>
<td>0.6628</td>
<td>0.6416</td>
</tr>
<tr>
<td>Steel Die</td>
<td>7833</td>
<td>460</td>
<td>40</td>
<td>40</td>
</tr>
</tbody>
</table>

\[ \text{ Springer} \]
of the pultrusion domain i.e. die radius and TCR values are found by applying a hybrid genetic algorithm in which a combination of a single objective binary coded genetic algorithm (SOGA) and the “fmincon” gradient based minimization algorithm (Sequential Quadratic Programming—SQP) are used. SOGA is initially used for the global search purposes and eventually to determine the starting point for the “fmincon” function to increase the accuracy of the solution. In the second optimization problem, the productivity of the pultrusion process expressed in terms of the pulling speed is improved by using a mixed integer genetic algorithm (MIGA).

The details of hybrid programming and MIGA are explained in the following sections. Binary tournament selection (i.e. with a simple constraint handling strategy [16]) is implemented to both SOGA and MIGA in which every individual compete with each other only once within two consecutive tournaments.

5.1 Case-1: Hybrid Genetic Algorithm

In the first case study three cylindrical heating pads are attached on top of the die (see Fig. 5). The set temperatures of the heaters are given in [3] as 171–188–188 °C, but the location of the heaters is unknown. In this study the length of the heaters are taken as 225 mm. The boundary conditions for the composite part are taken from the validation case except the die-part interface. In addition to the conductive heat flow, the TCR is defined at the interface boundary. For the die block all the surfaces except those on which the heating pads are located are exposed to ambient temperature (27 °C) with a convective heat transfer coefficient of 10 W/m² K. Since no information regarding the presence of cooling channels and their positioning is given in [3], cooling channels at the initial part of the die are not considered in the present model. 20 (radial)×150 (axial) control volumes and 22 (radial)×152 (axial) nodes are used for discretization of cylindrical die block.

The aim is to find the optimum die radius and TCR values that give the same centerline temperature distribution as found in the validation case. For this purpose it is assumed that 15 centerline temperature data values are measured from the validation case by equally spaced thermocouples such that temperature data is taken at every 60 mm through the axial direction. The objective is to minimize the sum of the square of difference between the measured (validation case) and calculated (new configuration) centerline temperatures i.e. the error function, \(\sum (T_{meas} - T_{cal})^2\). 10 design variables are defined in total: The 9 TCR values (m² K/W) are changing between 0 and 0.1, corresponding to heat transfer coefficient (HTC) values (W/m² K) from 10 to infinity (TCR₀), and the die radius (mm) is changing between 10 and 50 mm.

![Fig. 3](image-url) Geometry and boundary conditions of pultrusion of the composite rod without die block. The pulling direction is from left to right

---

Table 2  Resin kinetic parameters

<table>
<thead>
<tr>
<th>(H_n) (J/kg)</th>
<th>(K_0) (1/s)</th>
<th>(E) (J/mol)</th>
<th>(n)</th>
</tr>
</thead>
<tbody>
<tr>
<td>323700</td>
<td>191400</td>
<td>60500</td>
<td>1.69</td>
</tr>
</tbody>
</table>

---

---
A single objective genetic algorithm (SOGA), a similar version of which has been applied in another optimization problem [16], is applied to determine the efficient starting points for the local search (SQP), i.e. for the “fmincon” function. The SQP is strongly influenced by the starting point but on the other hand, it in general finds the minimum very fast depending on the starting point and the complexity of the landscape. Hence in order to find the global
minimum of the error function accurately, the design variables giving the best fitness in the SOGA are used as the starting point for the SQP. The tolerance value for terminating the program used in the SQP is $10^{-6}$. A population size of 100, total number of generations of 100, string length for each design variable of 10, 40% elitism, two-point crossover with 80% probability and 5% probability of uniform mutation are applied in the SOGA. The details of these common genetic operators are given in [17].

5.1.1 Results and Discussions

A total of 10 runs are performed with the hybrid genetic algorithm to be sufficiently independent of statistical variations and two different die configurations are found (see Table 3). The reason for this is that the curve fitting procedure is a multi modal problem, i.e. there is more than one minimum point depending on the die configuration. Since the two configurations given in Table 3 approaches the same centerline temperature of the composite, one of the design sets having the minimum die radius (10.5 mm) is selected due to considering the die cost (i.e., the cost is assumed to be proportional to the die radius) and used in the second case study. The die is around 6 mm thick which might be too thin for practical industrial application however for the research objective of the present paper this is of less importance and it should mentioned that even though the specific value of the die thickness was not given in [3], a schematic of the experimental setup indicates a thickness similar to this value of 6 mm. The results for the 1st minimum point (Table 3) are depicted in Fig. 6 (TCR values) and Fig. 7 (centerline temperature).

Since the cooling channels at the inlet of the die section are not included in this study, the first 3–4 TCR values show that the heat flux coming from the die has to be reduced at these regions, hence it can be concluded that these regions to some extent act as the cooling channels. In addition, it can also be concluded that the first heater may not be needed depending on the inlet temperature of the composite. On the other hand, the contribution from curing becomes more important after approximately 0.4 m from the die exit and chemical shrinkage plays a vital role due to the initiation of the exothermic reaction. This effect can be seen at the last region, i.e. around 0.8–0.9 m, where a higher TCR value is found indicating the probability of a chemical shrinkage causing an air gap at the interface. The corresponding centerline temperature profile and 15 temperature data values taken from the validation case are seen in Fig. 7. It is seen that the obtained centerline temperature by using the new pultrusion domain, in which the error function is minimized to 6.57, matches well the one calculated in the validation case.

5.2 Case-2: MIGA

In the second case study, five equally spaced cylindrical heaters are attached to the die block (see Fig. 8). The distance from the die inlet to the first heater is 30 mm, the length of each heater is 165 mm and the spacing of the heaters is 12 mm. The optimal die radius (10.5 mm) and TCR values (Fig. 6) found in case-1 as well as the same boundary conditions are applied.

| Table 3 | Details of 10 runs for hybrid Genetic algorithm |
|-----------------|-----------------|-----------------|-----------------|
|                | 1st Minimum     | 2nd Minimum     |
| Error function  | 6.57            | 3.69            |
| Die radius (mm) | 10.5            | 42.5            |
The objective is to minimize the number of heaters while increasing the speed of the process and the mean of the composite degree of cure at the die exit. Sets of different sequential combinations of active heaters are also considered in the optimization procedure. An individual having 11 design variables is seen in Fig. 9. The first five design variables ($p_i$) consist of binary (boolean) values, i.e. they take only 0 or 1 indicating the decision whether the heater-$i$ will be used or not. The rest of the design variables ($T_i$’s and $u$) are encoded with binary strings having a string length of 10. Simply, there are two possibilities: a heater can be used ($p_i=1$) or cannot be used ($p_i=0$). In addition to that these five possibilities ($p_i$) control the temperature of the heaters, e.g. if $p_i=1$ then the corresponding temperature of the heater ($T_i$) is used in the thermo-chemical pultrusion model of the composite rod with die block. Otherwise if $p_i=0$, then instead of using the corresponding temperature of the heater, a convective boundary condition is applied to the nodes of this heater-$i$. For instance, if the combination of the heaters is “10101” (i.e. the number of active heaters is 3) for an individual in the GA population, then the first, third and fifth heaters and their temperatures ($T_1$, $T_3$ and $T_5$) are used in the already developed numerical model. Moreover, the second and the fourth heaters are not taken into account in this case instead a convective boundary condition is applied. 

Fig. 6 Optimum TCR values along the axial distance of the pultrusion domain for die radius 10.5 mm

Fig. 7 Calculated (for new configuration) and measured (from validation case) centerline temperatures for die radius 10.5 mm
exposed to ambient is utilized at the locations of the second and fourth heater. Since this case is a mixed integer problem (i.e. both continuous and discrete values are used), the classical optimization method “fmincon” is not used.

In the optimization study, the temperature of each heater \((T_i)\) is varying between 150 °C and 250 °C. The pulling speed \((u)\) is changing between 5 mm/s and 13 mm/s. The constraints are determined as the following:

- The maximum temperature of the composite should not exceed the degradation temperature which is taken to be 240 °C \((T_{\text{max}} < 240 \, ^\circ\text{C})\).
- The mean cure degree at the die exit should be higher than 0.85 \((\alpha > 0.85)\). This value is chosen considering the mean of cure in the validation case.
- The pulling speed should be higher than 5 mm/s \((u > 5 \, \text{mm/s})\).

All the constraints are normalized such that they vary between 0 and 1. It should be noted that the heaters are assumed to be active during the whole simulation of the process since the steady-state conditions are applied. The objective function is the minimization of the total number of heaters which would normally lead to reduction of the power consumption in the die and it can be mathematically expressed as

\[
\text{Minimize : } \sum p_i
\]

which lead to the following measure of the fitness

\[
\text{Fitness} = \text{The total number of heaters}
\]

Modified mutation is used for binary integer design variables (the first five design variables). In this operation a number \((n)\) is selected randomly between 1 and 5 at every individual. Then the values of \(n\) randomly selected genes \((p_i)\) are simply inverted (0 is flipped to 1 and vice versa). On the other hand, two point crossover and uniform mutation are sequentially applied for encoded \((T_i\) and \(u\)) design variables. A population size of 100, total number of generations of 100, 40 % elitism (i.e. 40 % of higher performing solutions are kept for the next generation and the rest is selected randomly to preserve the diversity in

\[
\text{String length } (p_i) = 1
\]

\[
\text{String length } (T_i, u) = 10 \quad \text{(Encoded)}
\]

\[
\text{Integer values (0 or 1)}
\]

\[
\text{Temperature of heaters}
\]

\[
\text{Speed}
\]

\[
\text{Fig. 9} \quad \text{Representation of design variables inside an individual. The total string length of an individual is } 5 \times 1 + 6 \times 10 = 66
\]
the population), two-point crossover with 80% probability and 5% probability of uniform mutation (for encoded design variables) are applied in the MIGA.

5.2.1 Results and Discussions

A total of 30 runs are performed for statistical assurance with the MIGA procedure and some information of these runs can be seen in Table 4. In the MIGA, the best fitness of the population is stored while considering the pulling speed of the process. Therefore in Table 4, the pulling speeds that correspond to the best fitness are also given. The total number of active heaters is minimized. The best and average fitness values as a function of generations is seen in Fig. 10 (i.e. the best performance out of 30 experiments). It is seen from Fig. 10 that the minimum fitness value is obtained at the fifth generation. Since the placements of the heaters ($p_i$) are design variables and at the same time they are used in the objective function, the average of the population changes significantly over the total generations. It should be noted that, although the MIGA has found the minimum total number of heaters which is 1 after five generations, it was not stopped. The reason is that besides the best fitness, the pulling speed is also maximized by considering a high constraint value. The optimum configuration is found as “01000” (i.e. the total number of heaters is 1) which shows that only the second heater is used while satisfying the constraints. The convective cooling boundary condition is applied to the rest of the regions. The corresponding temperature of the second heater is found to be 233.7 °C. The pulling speed is increased to 11.1 mm/s and the centerline cure degree of the composite rod at the die exit is enhanced to 0.869. The mean of the cure degree at the die exit and maximum composite temperature are found as 0.856 and 239.3 °C, respectively, which shows that the constraints are satisfied.

Table 4 Details of 30 runs (each with a population of 100 and 100 generation) for MIGA

<table>
<thead>
<tr>
<th></th>
<th>Best value</th>
<th>Average value</th>
<th>Standard deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Best fitness of the population</td>
<td>1</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>Best pulling speed (mm/s)</td>
<td>11.1</td>
<td>9.2</td>
<td>0.96</td>
</tr>
</tbody>
</table>

Fig. 10 Generations as a function of fitness values (total number of heaters) for best individuals and average individuals
Fig. 11 Outermost temperature profile of the die and centerline temperature distributions of the composite rod obtained by using MIGA and found in the validation case.

Corresponding centerline temperatures and cure degree profiles are seen in Figs. 11 and 12, respectively. In Fig. 11 the temperature profile of the outermost surface of the cylindrical die block (path A–A in Fig. 8) where the heaters are positioned is also seen. Compared to the validation case the number of heaters is decreased from 3 to 1, the pulling speed is increased from 5 mm/s to 11.1 mm/s and the centerline cure degree at the die exit is increased from 0.84 to 0.869. Since the speed of the pultrusion process is increased the centerline temperature and cure degree profiles are shifted to the right which shows that the temperature is advected along the axial distance faster than in the validation case. Moreover the behavior of the convective cooling boundaries at the outermost surface of the die geometry can also be seen in Fig. 11. The first, third, fourth and fifth heater regions are exposed to ambient temperature, so only heater 2 is active and this is easily seen on the temperature of the path A–A. It should also be noted that between 0.5 and 0.7 m the temperature is increased due to the exothermic internal heat generation of the composite rod. From Fig. 12, it is seen that the

Fig. 12 Centerline cure degree profile of the composite rod obtained by using MIGA and found in the validation case.
composite has not started curing up to around 0.3 m due to the presence of the TCR at the die interface and due to the presence of the convective boundary condition at the first heater region. It can be concluded that the operational cost, which can be associated with the total number of the heaters, is reduced while still having a substantial increase in the productivity of the pultrusion process.

6 Conclusion

In the present work the implemented thermo-chemical model for the pultrusion of a composite rod without die block was found to be numerically stable and good agreement with a validation case from literature regarding both centerline temperature and cure degree profiles of the composite. The optimal design parameters (die radius and TCR values) of the new pultrusion domain were found in Case-1 by using a hybrid optimization algorithm comprising a single objective genetic algorithm (SOGA) and a classical local search algorithm SQP (i.e. the “fmincon” function in MATLAB) for fitting the same centerline temperature as in the validation case. SOGA was first applied to find the near global optimum point serving as an initial solution for the “fmincon” which finds the minimum faster and more accurately. It should be noted that since all the design variables in Case-1 were continuous, the hybrid algorithm was utilized. On the other hand in Case-2, both discrete (boolean) and continuous variables were used, hence the MIGA was applied where the optimum heater configuration as well as the temperatures of the these active heaters and the pulling speed were found. A significant increase in the productivity of the process was obtained by using the MIGA. More specifically the speed of the process is increased approximately 120% compared to the validation case while decreasing the total number of heaters. It was shown that the genetic algorithms are very useful tools for optimizing manufacturing process parameters (i.e. discrete variables in particular) for the pultrusion process.

The optimization studies of pultrusion process parameters via evolutionary algorithms put forward in the present work can be further extended. The speed of the process can be maximized while minimizing the cost of the process simultaneously, i.e. a multi objective problem would be solved by using evolutionary algorithms [18]. This will in turn provide a deeper understanding of the behavior of the process.

Acknowledgment This work is a part of DeepWind project which has been granted by the European Commission (EC) under FP7 program platform Future Emerging Technology.

References

“Computational Approaches for Modelling the Multiphysics in Pultrusion Process”

Pierpaolo Carlone, Ismet Baran, Jesper H. Hattel, Gaetano S. Palazzo

Research Article

Computational Approaches for Modeling the Multiphysics in Pultrusion Process

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Received 30 August 2013; Accepted 4 November 2013

Academic Editor: Nao-Aki Noda

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Pultrusion is a continuous manufacturing process used to produce high strength composite profiles with constant cross section. The mutual interactions between heat transfer, resin flow and cure reaction, variation in the material properties, and stress/distortion evolutions strongly affect the process dynamics together with the mechanical properties and the geometrical precision of the final product. In the present work, pultrusion process simulations are performed for a unidirectional (UD) graphite/epoxy composite rod including several processing physics, such as fluid flow, heat transfer, chemical reaction, and solid mechanics. The pressure increase and the resin flow at the tapered inlet of the die are calculated by means of a computational fluid dynamics (CFD) finite volume model. Several models, based on different homogenization levels and solutions schemes, are proposed and compared for the evaluation of the temperature and the degree of cure distributions inside the heating die and at the postdie region. The transient stresses, distortions, and pull force are predicted using a sequentially coupled three-dimensional (3D) thermochemical analysis together with a 2D plane strain mechanical analysis using the finite element method and compared with results obtained from a semianalytical approach.

1. Introduction

Pultrusion is a continuous manufacturing process used to realize constant cross sectional composite profiles. In recent years, the pultrusion process experienced a remarkable growing within the composite industry, due to its cost-effectiveness, automation, and high quality of products. Nowadays, the process is widely used to manufacture highly strengthened structures such as wind turbine blades, window profiles, door panels, and reinforcing bars for concrete. Moreover, in some applicative sectors, such as in the automotive industry, the environmental impact of pultruded composite structures over the entire life cycles results is lower than other engineering materials [1]. A schematic view of the pultrusion process is depicted in Figure 1. During the process, the reinforcement fibers, in the form of rovings or mat, are pulled through guiders and impregnated by the resin material in an open bath or employing a resin injection chamber. Wetted out reinforcements are then pulled via a pulling mechanism through the heating die. The die inlet is typically characterized by a tapered or a conical convergent shape, in order to promote the desired impregnation and compaction of the reinforcement, the removal of the air and the excess resin. In the straight portion of the die, the heat provided by means of electrical heaters or hot oil activates the exothermic cure reaction of the thermoset resin. As a consequence, the material changes its status from reactive liquid to gel and then vitrified solid [2, 3]. The thermochemical behavior of the processing thermoset resin, generally represented by time-temperature-transformation (TTT) diagrams [2–4], is a crucial issue. During the curing process, the resin shrinks because of the chemical reaction (cross linking) promoting the contraction of the work piece. Besides that, the part continues contracting due to the cooling effect, for example, convective cooling at the postdie region. At the end of the process, the cured and solidified product is cut into desired lengths.

Even if the process is conceptually quite simple, the analysis of its dynamics and the definition of optimal processing parameters are a complex task, due to the mutual interactions between involved physical phenomena, mainly related to heat...
transfer, species conversion and phase changes, die-material contact, and stress-strain development. Several researchers have performed numerical and experimental investigations on different aspects inherent to the pultrusion process, mainly focusing on issues related to heat transfer and cure phenomena [5–11]. Pressure distribution [12, 13], and pulling force [14–20]. However, proposed models often neglect the interactions between involved phenomena, on the base of some simplifying assumptions. Most of the published works converge on the conclusion that the mechanical properties and the quality of the pultruded composite are strongly affected by the degree of cure (DOC) distribution and the applied pulling force. The aforementioned features, in turn, depend on the pull speed, die temperature, die geometry, constituents’ type, and volume fractions. Furthermore, the pulling force consists of different contributions, such as the bulk compaction force due to the pressure increase in the tapered portion of the die, the viscous drag acting in the liquid zone, and the frictional force due to the contact between the internal surface of the die and the solidified processing material [2, 3, 18–21].

Experimental outcomes reported in [14] by Price and Cupschalk showed the impact of the materials volume fractions, die temperature and pull speed on the pull force. It was also indicated that for a constant temperature, and pulling speed, the force increases exponentially with the volume of material. Lackey and Vaughan carried out an extensive experimental and statistical investigation on the influence of process parameters on the pulling force and flexural strength of pultruded products, employing a five-factor half-factorial central composite design (CCD). It was concluded that the process parameters affect the pulling force according to complex interactions whose overall effect may vary significantly using resin systems characterized by different cure kinetics [15]. Considering the elevated number of variables involved in the aforementioned problem, a satisfactory experimental analysis could result in undesired time and money spending. Furthermore, pure experimental tests may have no solid predictive capability. As a consequence, the development of suitable multiphysics models is highly required for composite manufacturing processes. In-plane stresses and deformations in composite laminates can also be related to the interaction between the tool and the part [22]. Besides, the temperature and the DOC gradients through the composite thickness also promote the development of residual stresses in the manufactured part [23]. A better understanding of these phenomena, which take place in the heating die as well as in the postdie region, is highly required to reduce process induced shape distortions and residual stresses and to obtain a realistic analysis of in-service loading scenarios and reliability assessments [24, 25].

In the present work, several processing models dealing with different phenomena are combined to simulate the manufacturing of a pultruded product. This approach has not been considered up to now for the analysis of the pultrusion, providing a better understanding of the entire process at a glance. A schematic representation of the implemented models, including outputs and relative connections, is depicted in Figure 2.

In particular, pultrusion process simulations are performed for a unidirectional (UD) graphite/epoxy composite rod including different processing physics with the aim to predict the pulling force and the stress/distortion evolutions in the processing material. All the contributions to the overall pulling force have been accounted for in the present work. The pressure increase, which is responsible for the bulk compaction force, has been derived by means of a computational fluid dynamics (CFD) model of the resin flow field at the inlet and solved using a finite volume (FV) approach. The reinforcing fibers have been modeled as an anisotropic porous media, with directional permeability in accordance with the Gebart model. The temperature and the DOC distributions inside the heating die and at the postdie region are obtained by means of a three-dimensional (3D) thermochemical analysis. Two different modeling approaches are implemented: a continuous finite element (FE) model and a porous FV model, based on different homogenization levels and solution schemes. Both models provide the viscosity field allowing one to infer the viscous drag acting in the liquid zone. Furthermore, two solution strategies have been developed and compared for the prediction of the normal pressure, which generates the frictional force, between the processing material and the internal die surface after resin gelation. In the first case, numerical outcomes provided by the FV porous model have been analytically processed considering the well-established relations of the continuous mechanics, resulting in a semianalytical method (SAM). In the second approach, the transient stresses, distortions, and frictional pull force are predicted using a sequentially coupled 3D thermochemical analysis together with a 2D plane strain mechanical analysis using the finite element method (FEM). In both cases the evolution of the mechanical properties of the processing material is computed using the cure hardening instantaneous linear elastic (CHILE) approach [25, 27]. The paper is organized as follows: in Section 2 the theoretical modeling and the governing equations are described in detail, while in Section 3 the obtained results are exposed and discussed. Finally, in Section 4, the relevant findings and the future perspectives of this research are highlighted.
2. Theoretical Modeling and Numerical Implementation

2.1. Pull Force Model. As aforementioned, four different contributions to the overall pulling force in pultrusion have been identified in the literature [2, 3, 14–21]: the collimation force $F_{\text{coll}}$, the bulk compaction force $F_{\text{bulk}}$, the viscous drag $F_{\text{vis}}$, and the frictional force $F_{\text{fric}}$. These contributions are strictly related to the geometrical features of the die-work piece system and to the resin transitions from liquid to gel and then solid status, as schematized in Figure 3.

The first contribution $F_{\text{coll}}$ is due to resistances arising from the creel to the die inlet and it is generally assumed to be negligible. As a consequence, the pulling force $F_{\text{pul}}$ can be expressed as follows:

$$F_{\text{pul}} = F_{\text{coll}} + F_{\text{bulk}} + F_{\text{vis}} + F_{\text{fric}} \approx F_{\text{bulk}} + F_{\text{vis}} + F_{\text{fric}}.$$  \hspace{1cm} (1)

The second contribution $F_{\text{bulk}}$ is related to the increase in the resin pressure typically observed in the initial part of the die, that is, when the resin is still in liquid phase. The die inlet is generally designed as tapered ($\theta \leq 10^\circ$) or rounded shapes [4] in order to promote the constituents compaction reducing fibers damage. Moreover, the resulting over-pressure allows the resin to completely fill the reinforcing material porosities. At the same time, this over-pressure forces the excess resin to flow back, as depicted in Figure 4. The excess resin is usually recovered and redriven to the open bath for the fiber impregnation. While the resin is in a liquid status at the die entrance, the force due to the applied pressure acts along a direction normal to the die surfaces. As a consequence, it does not affect the pulling force except at the tapered die entrance (Figure 4). Defining the local resin pressure as $p$, the die taper angle as $\theta$, and the inlet surface as $A_1$, the bulk compaction term can be written as follows:

$$F_{\text{bulk}} = \iiint_{A_1} p \sin \theta \, dA.$$  \hspace{1cm} (2)

In the straight portion of the die, the increase in the temperature of the processing resin, due to the heat provided by the heaters, activates the exothermic cure reaction. The crosslinking of the thermoset monomers, in conjunction with the existing temperature field, provides two relevant phenomena, namely, gelation and vitrification, in which the status of the resin is changed. The term gelation refers to the transition of the catalyzed resin from viscous liquid to gelled (rubbery) solid. This transition is associated with the achievement of a certain degree of cure or polymerization (degree of...
cure at gelation, $\alpha_{\text{gel}}$, which corresponds also to a sharp increase of the resin viscosity. Vitrification (glass transition) is not rigorously associated with a specific extent of the cure reaction, but with the ($\alpha$-dependent) glass transition temperature ($T_g$). If the resin temperature is below $T_g$, it behaves as a vitrified (glassy) solid. Differently from gelation, vitrification is a reversible phase change.

Before the gel point, viscous drag occurs at the die wall. This resistance is imputable to the presence of a thin liquid layer between the travelling fibers and the stationary die surface. Thus, a plane Couette flow is induced, in which the reinforcing fibers are assumed to be the moving plate translated at a constant pull speed and the die surface as the fixed plate. A schematic view is shown in Figure 5 [17]. The viscous force can be written analytically as follows:

$$F_{\text{vis}} = \frac{V_{\text{pul}}}{\lambda} \int_{A_3} \eta(\alpha, T) \, dA,$$

where $\lambda$ is the thickness of the resin layer between the solid boundary and the moving fibers, $\eta$ denotes the resin viscosity, $V_{\text{pul}}$ is the fiber pull speed, and $A_3$ is the surface interested by viscous effects, whose length is determined by the gel-point. Several approaches for the estimation of $\lambda$ have been adopted in the literature, mainly based on the fiber packing, the radius $r_f$, the volume fraction $V_f$, or permeability considerations [16, 17]. In the present investigation, the following relation has been employed [17]:

$$\lambda = r_f \left( 1 - \frac{1}{2} \sqrt{\frac{3\pi V_f}{2}} \right).$$

The rheological behavior is herein modeled following the well-recognized three parameters correlation model [12, 13, 16, 17], which is expressed as follows:

$$\eta = \eta_{\infty} \exp \left( \frac{\Delta E_{\eta}}{RT} + K \alpha \right),$$

where $R$ is the gas constant, $T$ is the absolute temperature, $\eta_{\infty}$, $\Delta E_{\eta}$, and $K$ are material parameters provided by experimental data fitting.

After the gel point, the resin flow and the viscous effects are obviously inhibited and the composite is mechanically pulled through the die. Consequently, the interaction between the processing material and the die surface is mainly characterized by frictional effects. Generally, the entity of the frictional force can be inferred by considering the friction coefficient $\mu$ and the contact pressure $\sigma$, according to the following equation:

$$F_{\text{fric}} = \int_{A_3} \mu \cdot \sigma \, dA,$$

being $A_3$ the die surface from the gel-point to the detachment point. It should be noted that the value of the friction coefficient depends on the DOC during the resin gelation and further varies at the glass transition. However, due to the lack of thorough experimental data, generally the averaged values are utilized [14–21]. Regarding the magnitude of the contact pressure, $\sigma$ is considered to be affected by two contrasting conditions: the transverse thermal expansion of the composite due to the increase in temperature and pressure and the resin chemical shrinkage related to crosslinking reaction. The latter phenomenon leads to a progressive reduction in the size of the composite cross section until it shrinks away from the die internal wall (detachment point).

It is worth noting that the separation of the processing material from the die cavity induces the formation of a thin (thermally insulating) air layer. As a consequence, a thermal contact resistance (TCR) is interposed between the heated die and the processing material. In the present investigation, each contribution has been computed using the numerical and the semianalytical models, as explained in detail in the following.

### 2.2. Impregnation Analysis

In a conventional pultrusion process, reinforcing fibers are wetted out inside the resin bath before entering the heating die. After the impregnation, the wetted fibers typically show an excess of resin with respect to the amount needed for the final product. As a consequence, in the tapered zone of the die (inlet) the processing material is compacted resulting in a pressure increase with respect to the atmospheric value. Material compaction is affected by several factors, such as the volume fraction and the permeability of the reinforcement, the resin viscosity, and the geometrical features of the die-material system [12]. The impregnation model describes the pressure distribution and the resin flow in the first part of the die, including the tapered or rounded zone and a portion of the straight die (Figure 4). Velocity and pressure in the reinforcement-free zones of the domain are inferred by means of the conjunct solution of the well-known mass and momentum equations. In particular, since the early part of the die is not heated in order to avoid premature resin gelation, it is assumed that the temperature and the DOC variations are negligible, and therefore the resin viscosity remains constant. Furthermore, under the hypothesis of incompressibility of the liquid resin and neglecting body forces, the equilibrium equations can be written as follows:

$$\begin{align*}
\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} &= 0, \\
\eta \left( \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2} \right) - \frac{\partial p}{\partial x} &= 0,
\end{align*}$$
\[
\eta \left( \frac{\partial^2 V}{\partial x^2} + \frac{\partial^2 V}{\partial y^2} + \frac{\partial^2 V}{\partial z^2} \right) - \frac{\partial p}{\partial y} = 0, \\
\eta \left( \frac{\partial^2 w}{\partial x^2} + \frac{\partial^2 w}{\partial y^2} + \frac{\partial^2 w}{\partial z^2} \right) - \frac{\partial p}{\partial z} = 0, 
\]

where \( u, v, \) and \( w \) are the velocity components of the resin along the \( x, y, \) and \( z \) directions, respectively, and \( p \) is the liquid pressure. The reinforcing fibers have been treated as a moving porous media, in which the porosity and the permeability vary according to geometrical considerations, ensuring always the final fiber volume. The following modified Darcy model has been solved in the porous region:

\[
\begin{align*}
u &= U - K_{xx} \frac{\partial P}{\eta \Phi} \\
v &= V - K_{yy} \frac{\partial P}{\eta \Phi} \\
w &= W - K_{zz} \frac{\partial P}{\eta \Phi},
\end{align*}
\]

where \( U, V, \) and \( W \) represent the velocity components of the porous media along the \( x, y, \) and \( z \) directions, respectively. It should be noted that, assuming that \( z \)-direction is the pull direction, the component \( W \) is constant and it is the only nonzero term in the straight portions of the domain, while other components should be locally modified considering the geometric configuration of the tapered zone [12]. Tow permeability has been defined according to the Gebart model as follows:

\[
K_{xx} = K_{yy} = C_1 \left( \frac{V_{f,\text{max}}}{V_f} - 1 \right) r_f^2, \\
K_{zz} = \frac{8 \sigma_f^2 V_f^2}{c} \left( 1 - V_f \right)^3,
\]

where \( r_f \) is the fiber radius, \( V_{f,\text{max}} \) the maximum achievable fiber volume fraction, \( C_1 \) and \( c \) are constants equal to 0.231 and 53, respectively [13]. The impregnation model has been implemented and solved using a FV scheme. The commercial software ANSYS-CFX has been employed for this purpose. The pressure distribution provided by the impregnation model is then used in (2) to evaluate \( f_{\text{bulk}} \).

### 2.3. Thermochemical Analysis

In this section, theoretical backgrounds of the implemented continuous and porous models are presented.

#### 2.3.1. Continuous Model

The basic assumption of the continuous (homogenized) model is that in each location of the processing composite material, all the constituents experience the same temperature. As a consequence, the whole temperature field is established solving a unique nonlinear equation using the lumped material properties [4–11, 16–18], which can be written as follows:

\[
\rho_v C_{\text{pc}} \left( \frac{\partial T}{\partial t} + V_{\text{pul}} \frac{\partial T}{\partial z} \right) = k_{xx} \frac{\partial^2 T}{\partial x^2} + k_{yy} \frac{\partial^2 T}{\partial y^2} + k_{zz} \frac{\partial^2 T}{\partial z^2} + V_s q,
\]

where \( T \) is the temperature, \( t \) is the time, \( \rho_v \) is the density, \( C_{\text{pc}} \) is the specific heat, \( k_{xx}, k_{yy}, \) and \( k_{zz} \) are the thermal conductivities of the composite material along \( x, y, \) and \( z \) directions, respectively, and \( V_s \) is the resin volume fraction. Material properties are assumed to be constant throughout the process. The source term \( q \) in (10) is related to the internal heat generation due to the exothermic resin reaction and is expressed as follows:

\[
q = \rho_v H_{tr} R_r,
\]

where \( R_r \) is the reaction rate, \( H_{tr} \) is the total heat of reaction, and \( \rho_v \) is the density.

Several kinetic models have been proposed and discussed in the inherent literature to describe the evolution of the cure reaction. In the present investigation the well-established nth-order model has been adopted, assuming an Arrhenius type dependence on the absolute temperature:

\[
R_r (\alpha, T) = \frac{\partial \alpha}{\partial t} = \frac{1}{H_{tr}} \frac{dH(t)}{dt} = K_0 \exp \left( -\frac{\Delta E}{RT} \right) (1 - \alpha)^n,
\]

where \( \alpha \) is the degree of cure and \( H(t) \) is the heat generated during cure. The above equations have been solved in a 3D domain using a FE approach. The evaluation of the DOC and the reaction rate has been obtained by means of an iterative inhouse developed routine implemented into the commercial software package ABAQUS [28], until the matching of temperature and DOC tolerances to reach the steady state. The DOC is obtained by using the following discretization [7, 25]:

\[
\left( \frac{\partial \alpha}{\partial t} + V_{\text{pul}} \frac{\partial \alpha}{\partial z} \right) = R_r (\alpha, T).
\]

#### 2.3.2. Porous Model

Differently from the continuous model, the porous model treats the pultrusion process as a reactive liquid (resin) flow through a moving porous media (reinforcement) inside a defined rigid boundary (die cavity). In other words, it is a CFD based nonthermal equilibrium model considering each component as a different entity on macroscale; therefore a finite difference between the reinforcement and the matrix temperatures is admitted. As a consequence, besides the continuity and the momentum equations for the fluid phase, one energy balance equation for each component is needed. This allows heat to be transferred between contiguous phases. Assumption that the processing composite is only composed by the reacting resin and the fibrous reinforcement, that is, neglecting voids and porosity effects,
the temperature field can be obtained by solving the following equations:

\[
\varphi \rho_f C_p, f \frac{\partial T_f}{\partial t} + \rho_f C_p, f \dot{v}_p = \frac{\partial^2 T_f}{\partial x^2} + k_{x,f} \frac{\partial^2 T_f}{\partial y^2} + k_{z,f} \frac{\partial^2 T_f}{\partial z^2} + Q_f, \tag{14}
\]

\[
\varphi \rho_r C_p, r \frac{\partial T_r}{\partial t} + \rho_r C_p, r \dot{v}_r (\frac{\partial T_r}{\partial x} + \frac{\partial T_r}{\partial y} + \frac{\partial T_r}{\partial z}) = \frac{\partial^2 T_r}{\partial x^2} + k_r \frac{\partial^2 T_r}{\partial y^2} + k_r \frac{\partial^2 T_r}{\partial z^2} + \varphi q + Q_{fr}, \tag{15}
\]

where the subscripts \(r\) and \(f\) refer to the resin and fiber, respectively. In the above equations, \(\varphi = 1 - \varphi_f\) represents the volume porosity of the medium (ratio between the volume available for fluid flow and the total volume). Assuming the absence of voids, \(\varphi\) coincides with the resin volume fraction \(V_r = 1 - V_f \cdot \varphi_f = -Q_{fr}\), is the interfacial heat transfer between the fluid and the solid depending on the temperature difference, the interfacial area density, and the physical properties of the two phases. It should be borne in mind that in the porous model the DOC is treated as an additional scalar variable with transport properties existing only in the fluid phase and varying according to a source term generated by the reaction rate previously defined in (12). Similarly, the heat generation term \(q\) in (11) is restricted to the reactive resin and the exothermic reaction affects the fiber temperature by means of conductive heat transfer. As for the impregnation model, the software ANSYS-CFX [29] has been used to solve the porous thermal model employing a FV numerical scheme. The temperature and the DOC distributions are utilized to compute the resin viscosity and the viscous drag, according to (5) and (3), respectively.

2.4. Mechanical Analysis. As mentioned above, the process induced stress and distortions, including also the die-composite contact pressure, are predicted using the two different procedures. The former approach is based on the solution of a 2D quasi-static FE mechanical model, sequentially coupled with the 3D continuous thermochemical FE model. The latter is a semianalytical approach based on the applications of the well-established principles of the linear elasticity to the results provided by the above described porous model.

2.4.1. FE Model. In this model, the 2D cross section of the part is assumed to be moved along the pulling direction while tracking the corresponding temperature and DOC profiles provided by the FE model. A detailed description of this procedure, that is, the mapping of the predicted fields \(T, \alpha\) to the 2D mechanical plain-strain model, is shown in Figure 6. The implemented mechanical FE model assumes that the longitudinal strains, that is, parallel to the pulling direction, are negligible with respect to the transverse components of the strain tensor. This approximation is well justified considering the remarkable difference, for pultruded products, between in plane (cross sectional) and out of plane (product length) dimensions, being the former of few square millimeters and the latter of several meters before the cutout. As a consequence, the problem can be reduced to a two dimensional plane strain analysis, as discussed in [25]. The corresponding transient distortions and the evolution of the process induced stresses and strains are calculated considering the temperature and the current distributions, assuming the following contributions to the incremental total strain \((\Delta \varepsilon_{\text{tot}})\):

\[
\Delta \varepsilon_{\text{tot}} = \Delta \varepsilon_{\text{mech}} + \Delta \varepsilon_{\text{th}} + \Delta \varepsilon_{\text{ch}}, \tag{16}
\]

where \(\Delta \varepsilon_{\text{mech}}\) is the incremental mechanical strain, \(\Delta \varepsilon_{\text{th}}\) is the incremental thermal strain, and \(\Delta \varepsilon_{\text{ch}}\) is the incremental chemical strain due to the volumetric shrinkage of the resin. The details of the relations between the stress and strain tensors used in the present FE approach can be found in [25].

The CHILE approach [25, 27] has been implemented by means of user-routines in the commercial package ABAQUS to derive the instantaneous local resin elastic modulus \((E_r)\), assuming a linear relation of the stress and strain tensors. The expression for the elastic modulus, assuming secondary effects of temperature as negligible, is given as follows:

\[
E_r = \begin{cases} E_r^0 \quad & T^* < T_{C1} \\ E_r^0 + \frac{T^* - T_{C1}}{T_{C2} - T_{C1}} (E_r^{\infty} - E_r^0) \quad & T_{C1} \leq T^* \leq T_{C2} \\ E_r^{\infty} \quad & T^* > T_{C2}. \end{cases} \tag{17}
\]

The fictitious temperature \(T^*\) is defined as the difference between the \(T^0\) and the actual resin temperature \(T\) and expressed as follows:

\[
T^* = T^0 - T = \left( T_g^0 + \alpha_{tg} \alpha - T \right), \tag{18}
\]
homogenized material is linear elastic, the solved boundary value problem is significantly nonlinear, due to the space and time variations of all physical and mechanical properties involved.

2.4.2. Semianalytical Analysis of Distortions and Pressure. The proposed semianalytical approach is based on the computation of a virtual unconstrained cross section of the processing material. It is assumed that during the process the position of the center of mass (barycenter) of the cross section is always preserved [11]. The composite distortions are related to the thermal expansion of each component and the chemical shrinkage of the reactive resin. As a consequence, each virtual dimension of the \( i \)th control volume can be computed multiplying its initial value by the correction factor as follows:

\[
\delta_{c,i} = V_r \delta_{r,i} + V_f \delta_{f,i},
\]

(19)

where \( \delta_{r,i} \) and \( \delta_{f,i} \) are the variations of a unit dimension of the \( i \)th volume entirely filled with resin and fiber, respectively. Defining the CTEs of the resin as \( \alpha_r \) and of the fibers in the transverse direction as \( \alpha_{f,t} \), and the percentage volumetric shrinkage of the fully cured resin as \( \gamma_r \), it follows

\[
\delta_{r,i} = (1 + \alpha_r (T_{r,i} - T_0)) \cdot \left(1 - \frac{V_f \alpha_f}{100}\right)^{1/3},
\]

\[
\delta_{f,i} = (1 + \alpha_{f,t} (T_{f,i} - T_0)),
\]

(20)

where the subscripts \( r \) and \( f \) refer to resin and fiber, respectively. Here, the utilized temperature and the DOC values are the volume averaged values calculated by considering the results of the porous model described in Section 2.3.2.

With reference to the circular cross section investigated, the dimensional variation \( \Delta_{r,j} \) of the \( i \)th volume, along the radial direction, is given by

\[
\Delta_{r,j} = r_j \left(\delta_{c,j} - 1\right).
\]

(21)

The total displacement \( \Delta_r = \sum \Delta_{r,j} \) and the virtual radius \( r_v \) can be evaluated by extending equation (21) to the whole radius. In particular, from the die inlet until the detachment point, due to the prevalence of the thermal expansion on the chemical shrinkage, the virtual section of the processing composite results reasonably greater than the die cavity. Consequently, the pultruded part is compressed by the die internal walls. In this case, the contact pressure is evaluated following the well-known principles of materials science for thick walled cylinders, schematizing the virtual section as a series of concentric and contiguous annulus (delamination phenomena are not included) and assuming plane strain hypothesis. As for the FE model described in Section 2.4.1, material elastic properties are evaluated according to local temperature and DOC, using the CHILE approach and the SCFM relationships. Taking into account this, the continuity of the material imposes the congruence of the circumferential strains \( \varepsilon_\theta \) and the radial stress \( \sigma_r \) at the boundaries between adjacent layers, using the subscript \( j \) to identify each annulus (increasing with the radial position) and the subscripts int and ext to localize the strain at the inner or outer radius of the annulus, respectively, which results in the following:

\[
\varepsilon_{\theta,j,int} = \varepsilon_{\theta,j+1,int}
\]

\[
\sigma_{r,j,int} = \sigma_{r,j+1,int}
\]

(22)
Furthermore, considering that the enlargement of the real cross section is prevented by the rigid die walls (the unconstrained section previously computed is a purely virtual one), the circumferential strain on the external radius results in the following:

\[ \varepsilon_{\theta} = -\frac{\Delta r}{r_0}, \]  

providing the closure to the considered problem. A schematic representation of the calculation procedure is depicted in Figure 7. It is trivial to outline that, in correspondence with the external radius, the radial solicitation \( \sigma_r \) equals to the opposite of the pressure \( \sigma \) acting on the die internal wall, allowing one to derive the frictional contribution using (6).

Frictional resistance vanishes when the shrinkage effect prevails, inducing the detachment of the material from the die. In this case, an additional TCR is induced between the die and the composite. TCR values are computed in the corresponding locations assuming that the empty space between the die surface and the processing composite is fulfilled by air. Since radial displacements and TCR values along the die length are not known as a priori, an iterative procedure, connecting the thermochemical model with the dimensional change model, has been implemented, until reaching the convergence of a temperature criterion.

3. Results and Discussion

3.1. Case Study. The pultrusion process of a UD graphite/epoxy composite rod with circular cross section is simulated to compare the numerical outcomes provided by the proposed models as well as with results discussed in the literature [6, 13]. The radius of the processing rod is 4.75 mm, while the length \( L_{\text{die}} \) of heating die is 914 mm, which are adopted for the numerical and experimental analysis detailed in [6]. It should be noted that, in the performed simulations, the temperature distribution on the internal die surface is used to provide the required closure of the above described thermochemical problem; that is, the die is not included in the calculation domain, as also done in [6]. Despite the implemented thermochemical models that allow one to define more complex boundary conditions, this relatively simpler case has been reproduced in order to compare numerical results with data reported in [6]. The inlet temperature is assumed to be equal to the resin bath temperature (38°C), while the matrix material is assumed to be totally uncured (\( \alpha = 0 \)) at the same cross section. Only a quarter of the 3D model has been considered due to the symmetry and in order to reduce the computational effort. A schematic view of the simulation domain is depicted in Figure 8.

The variation of the internal section in the tapered inlet is not taken into account in the thermochemical model as well as for the stress and distortions calculations in the mechanical model. The reason is that the size of the tapered section is relatively small and there is almost no heat transfer, curing, and stress development observed in that region.

In addition, considering that the composite material in the die exit section is still at elevated temperature, it is reasonable to suppose that the cure reaction proceeds also in the postdie region, leading to a certain amount of DOC increase, as already discussed in [6, 25]. This aspect has been included in the model extending the length of the pultruded composite to the postdie region. The postdie is characterized by a total length \( L_{\text{post-die}} \) equal to 1370 mm, ensuring that no further reaction will take place in the material. In the postdie region, convective cooling in the room temperature (27°C) is imposed as a boundary condition on the external surface of the pultruded product. The dependence of the convective cooling coefficient on the surface temperature is defined using the well-known principle of heat transfer for horizontal cylinder. The pull speed \( v_p \) has been defined as 5 mm/s [6].

The pultruded composite rod consists of Shell Epon 9420/9470/537 resin and Graphite Hercules AS4-12K fibers \( (r_f = 13 \mu m) \). The properties of components and the resin kinetic parameters are listed in Tables 1 and 2, respectively. The parameters used in the CHILE approach are given in Table 3.

3.2. Impregnation Analysis. The impregnation model is considered for the first 30 mm of the die, assuming that after this length flow perturbations induced by the convergent section of the inlet vanish. The tapered inlet has been modeled assuming a rounded shape with length \( L_t \) and radius \( R_t \) being equal to 6 and 6.35 mm, respectively [13]. The preform ratio, defined as the ratio between the cross sectional area of the impregnated material before and after the compaction due to the tapered inlet, is assumed to be 1.44, neglecting shape variations of the pulled material. As a consequence, the wetted fibers approaching to the inlet have been modeled as a cylindrical porous medium with radius being equal to 5.7 mm.

As aforementioned, a constant viscosity assumption is adopted, taking into account that generally in the very early part of the die no significant reaction is observed. The reference viscosity value has been obtained according to (5), considering the resin as fully uncured (\( \alpha = 0 \)) at a temperature equal to 38°C, as for the thermochemical models. It should be noted, however, that the catalyzed resin, before the impregnation and entering of the die, lays into the open bath for some time. During this period, a small amount of reaction cannot be excluded a priori. Even if the degree of cross-linking in the resin bath does not significantly affect the evolution of the solidification process, it can influence the
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Figure 8: Schematic view of the pultrusion domain for the composite rod. All dimensions are in mm.

Table 1: Material physical properties and concentration [6, 9–11].

<table>
<thead>
<tr>
<th>Property</th>
<th>Graphite</th>
<th>Epoxy</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\rho$ [kg m$^{-3}$]</td>
<td>1790</td>
<td>1260</td>
</tr>
<tr>
<td>$c_p$ [J kg$^{-1}$ K$^{-1}$]</td>
<td>712</td>
<td>1255</td>
</tr>
<tr>
<td>$k_x$ [W m$^{-1}$ K$^{-1}$]</td>
<td>11.6</td>
<td>0.2</td>
</tr>
<tr>
<td>$k_y$ [W m$^{-1}$ K$^{-1}$]</td>
<td>11.6</td>
<td>0.2</td>
</tr>
<tr>
<td>$k_z$ [W m$^{-1}$ K$^{-1}$]</td>
<td>66</td>
<td>0.2</td>
</tr>
<tr>
<td>$E_x$ [GPa]</td>
<td>$2.068E+1$</td>
<td>—</td>
</tr>
<tr>
<td>$E_y$ [GPa]</td>
<td>$2.068E+1$</td>
<td>—</td>
</tr>
<tr>
<td>$E_z$ [GPa]</td>
<td>$2.068E+2$</td>
<td>—</td>
</tr>
<tr>
<td>$v_{xx}$</td>
<td>0.2</td>
<td>0.35</td>
</tr>
<tr>
<td>$v_{yy}$</td>
<td>0.2</td>
<td>0.35</td>
</tr>
<tr>
<td>$v_{zz}$</td>
<td>0.5</td>
<td>0.35</td>
</tr>
<tr>
<td>$G_{xx}$ [GPa]</td>
<td>$2.758E+1$</td>
<td>—</td>
</tr>
<tr>
<td>$G_{yy}$ [GPa]</td>
<td>$2.758E+1$</td>
<td>—</td>
</tr>
<tr>
<td>$G_{xy}$ [GPa]</td>
<td>$6.894E+0$</td>
<td>—</td>
</tr>
<tr>
<td>$\alpha_x$ (1/°C)</td>
<td>$7.2E-6$</td>
<td>$4.5E-5$</td>
</tr>
<tr>
<td>$\alpha_y$ (1/°C)</td>
<td>$7.2E-6$</td>
<td>$4.5E-5$</td>
</tr>
<tr>
<td>$\alpha_z$ (1/°C)</td>
<td>$-9.0E-7$</td>
<td>$4.5E-5$</td>
</tr>
<tr>
<td>$\gamma_v$ (%)</td>
<td>—</td>
<td>4</td>
</tr>
<tr>
<td>Volume fraction</td>
<td>0.6</td>
<td>0.4</td>
</tr>
</tbody>
</table>

internal viscosity for the impregnation and compaction analysis. This situation is investigated in the present work by simulating the compaction process using three different viscosity values: 1.05 Pa·s ($\alpha = 0$), 1.5 Pa·s ($\alpha = 0.008$) [13], and 2.60 Pa·s ($\alpha = 0.02$). In the impregnation model, the die surfaces are modeled as rigid walls, defined with a no-slip condition. An inlet condition is imposed to the inlet surface corresponding to the preform, while an opening condition allowing the creation of the resin backflow is applied on the surrounding surface. In both cases, a zero relative pressure is defined. The velocity of the processing material crossing the outlet section has been assumed to be equal to the pull speed.

In Figures 9–11 the results provided by the impregnation model are reported which show the pressure profiles at the centerline of the processing material (Figure 9), a streamline plot of the resin flow in the tapered region (Figure 10), and the calculated bulk compaction force (Figure 11). For all the simulated conditions, an increase in the pressure has been predicted before the intersection point, which is identified by the contact between the reinforced preform and the die internal surface and is depicted in Figure 9 by the vertical dashed line. This pressure variation is due to the effect of the resin backflow (well highlighted by the streamlines opposite to the pull speed in Figure 10), which prevents the free flow of the resin inside the preform towards the nonreinforced zones. The same figure also highlights the excellent agreement between the obtained pressure profiles and the data reported in [13], confirming the validity of the implemented numerical model. Furthermore, as already indicated in [13], for the considered configuration more than half of the total pressure increase has already developed at the intersection point. It is also worth noting in Figure 10 that, at the very beginning of the straight portion of the die, the resin velocity converges on the pull speed imposed to the reinforcing fibers.

Obtained outcomes also show that the activation of the cure reaction inside the resin bath is quite undesirable, even if the degree of crosslinking achieved before entering the die is reduced. Indeed, the premature crosslinking of the catalyzed resin increases its viscosity and, as a consequence, higher pressures are needed to squeeze the excess resin out of the preform. This results also in a proportional increase of the pulling force contribution due to material compaction (Figure 11).

3.3. Thermochemical Analysis. The calculated centerline temperature and DOC profiles are shown in Figure 12 together with the temperature profile imposed on the die wall. It is seen
Table 2: Epoxy resin rheological parameters \cite{6,9–12}.

<table>
<thead>
<tr>
<th>(K_0) [s(^{-1})]</th>
<th>(\Delta E) [J mol(^{-1})]</th>
<th>(n)</th>
<th>(H_0) [kg(^{-1})]</th>
<th>(\eta_{\infty}) [Pa s]</th>
<th>(\Delta E_0) [J mol(^{-1})]</th>
<th>(K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>19.14E + 4</td>
<td>60.5E + 3</td>
<td>1.69</td>
<td>323.7E + 3</td>
<td>5.12E – 7</td>
<td>3.76E + 4</td>
<td>45.0</td>
</tr>
</tbody>
</table>

Table 3: Resin properties for modulus calculation (CHILE and glass transition) \cite{25,27}.

<table>
<thead>
<tr>
<th>(T_{C1}) [(^\circ)C]</th>
<th>(T_{C2}) [(^\circ)C]</th>
<th>(T_0) [(^\circ)C]</th>
<th>(\alpha_{fP}) [(^\circ)C]</th>
<th>(E_0) [MPa]</th>
<th>(E_{\infty}) [MPa]</th>
</tr>
</thead>
<tbody>
<tr>
<td>-45</td>
<td>12</td>
<td>0</td>
<td>195</td>
<td>3.447</td>
<td>3447</td>
</tr>
</tbody>
</table>

Figure 10: Streamline of the resin flow in the tapered region of the die.

Figure 11: Influence of the resin viscosity (initial degree of cure) on the compaction term of the pulling force.

Figure 12: Temperature and DOC profiles: comparison of the present outcomes with the reference data \cite{6}.

that the predicted results match quite well with the available experimental data in \cite{6}. This evidences that the numerical schemes adopted for the continuous homogeneous FEM model (denoted as “CM” in Figure 12) and the porous nonhomogeneous FV model (denoted as “PM” in Figure 12) are stable and converged to a reliable solution. The temperature in the center of the composite rod becomes higher than the die wall temperature after approximately 390 mm from the die inlet due to the internal heat generation of the epoxy resin. At that point a peak of the reaction rate is obtained, inducing a sharp increase of the DOC. The maximum composite temperature is calculated approximately as 208\(^\circ\)C. What is more, at the postdie region, the DOC is increased slightly which indicates that the curing still takes place after the die exit, as also observed in \cite{6}. The centerline DOC is increased from 0.84 (at the die exit) to 0.87 (at the end of the process), while, at the surface, it varies from 0.80 to 0.83, indicating a global percentage increase of approximately 3.6%.

The depicted DOC profiles in Figure 12 show an earlier activation of the cure reaction at the composite surface due to the rapid temperature increase related to conductive heat transfer from the die wall. As a consequence, the DOC at the external radius initially results higher than at the center. This trend varies after the activation of the reaction in the core of the material; indeed, the relatively low thermal conductivity of the resin prevents the heat generated at the center to flow towards the external zones, inducing a significant and localized temperature increase at the center, which strongly promotes monomers crosslinking. It is worth noting that the cure crossover (intersection between the DOC profiles at the center and at the top) is reached approximately at \(\alpha = 0.5\), that is, well above the gel point (\(\alpha = 0.265\)) of the considered resin system, indicating a delay in the establishment of the desired in-out solidification direction. Indeed, as evidenced by the viscosity profiles depicted in Figure 13, the activation of the cure reaction implies a sharp viscosity increase at gelation, occurring earlier at the top surface, at a distance approximately equal to 360 mm from die entrance and separating the liquid zone (where viscous drag acts) from the gel.
zone (dominated by frictional resistance). The same viscosity trend is observed at the center of the composite rod after approximately 405 mm from the die entrance. It should be also noted that, in the first 200 mm from the inlet, the temperature increase leads to a slight viscosity reduction before the beginning of crosslinking phenomena, as also highlighted in Figure 13.

In the same figure (Figure 13) the work piece radius, as a function of the axial distance, is reported. As highlighted by numerical outcomes, in the liquid zone the materials thermal expansion prevails on chemical shrinkage, leading to a virtual radius of the work piece greater than the die internal radius. As a consequence a further pressure increase (shown in what follows) is to be expected. Even if this pressure increase does not theoretically implies further contributions to the total pulling force (being the wall surface parallel to $F_{pull}$), from a practical point of view it is very interesting since, in conjunction with the aforementioned viscosity reduction, it promotes the reduction of voids in the final product. As can be seen, both models fairly agree with the individuation of the detachment point, which is the intersection point between the virtual radius and the die internal radius during shrinkage. Please note that the zero radial displacement provided by the FE model (CM) is due, in agreement with reality, to the nonpenetrating condition applied at the mechanical contact between the composite and the rigid die surface [25]. The detachment point for the outer surface of the composite rod is found to be approximately at 540 mm from the inlet (more precisely 535 mm and 545 mm for the FEM and SAM); as a consequence the die length interested by the frictional effect (gel zone) is estimated to be approximately 180 mm. The delayed position of the detachment point predicted by the SAM with respect to the FEM suggests also that a relatively major computation of the virtual radius (or radial displacement of the cross section). This aspect can be related to the assumption of the lumped CTE employed in the FE model in contrast with the usage of a different CTE for each constituent (when the resin is in liquid phase) adopted by the SAM. After the detachment point, TCRs are induced between the work piece and the die. Nevertheless, very negligible differences (less than 0.5°C) in the temperature distributions have been found with and without the TCR inclusion in the calculations. The work piece radius in the exit section as provided by the analytical calculation coupled to the finite volume model, results 4.742 mm, in good agreement with the value (4.739 mm) reported in [11]. A slight difference (∼0.003 mm = 3 μm) regarding the work piece radius at the exit calculated using the FE model and the semianalytical procedure has been found. As can be seen in Figure 13, after the detachment point, the evolution of the radial distortion differs between the aforementioned approaches. The reason for this deviation could be found in the oversimplification of the semianalytical model (SAM), in which the displacements are calculated only in the radial direction without taking the effect of the mechanical behaviors in the longitudinal direction into account.

3.4. Mechanical Analysis. The evolution of the process induced transverse normal stresses in the $x$-direction ($S_{11}$) is shown in Figure 14(a). It is seen that at the end of the process, tensile stresses prevail at the inner region (center) and compression stresses occur at the outer region (top) while upholding the self-static equilibrium in which there is no applied external load. This observation resembles with the one presented in [25]. The stress levels are found to be relatively small (<1 MPa). The main reason is that there are an almost uniform temperature and DOC developments over the cross section of the composite rod which provides relatively lower through-thickness gradients promoting almost no residual stresses at the end of the process. The variation in $S_{11}$ is due to the internal competition between expansion and contraction of the part. The effective longitudinal and the transverse moduli (calculated by the SCFM) of the composite rod at the end of the process are found to be 130.2 GPa and 9.7 GPa, respectively, which agrees well with typical values given in [30] for T300 carbon/epoxy with a fiber volume fraction of 60%. In Figure 14(b) the resin modulus development due to monomers crosslinking is depicted. It is seen that almost same evolution pattern is obtained using the CHILE model in FEM model and SAM model.

Undeformed contour plots of the stresses $S_{11}$ (in $x$-direction) and $S_{22}$ (in the $y$-direction) are shown in Figure 15. As expected, the $S_{11}$ distribution is almost symmetric with the $S_{22}$ distribution with respect to the diagonal of the composite rod, since all the mechanical boundary conditions are the same. The maximum normal tensile and compression stresses are found to be approximately 0.26 MPa and −0.82 MPa, respectively, for $S_{11}$ and $S_{22}$.

In Figure 16, the contact pressure profiles (between internal die surface and the outer surface of the part, that is, top point indicated in figure) provided by the implemented models (FEM, SAM) are shown. Both models highlighted a progressive pressure increase (up to approximately 0.2 MPa for the FEM and 0.27 for the SAM) from the die inlet to the strong activation of the resin reaction since the composite part tries to expand because of the temperature increase; however the internal die surface restricts this expansion. The difference
Figure 14: In-plane stresses ($S_{11}$) evolution at the center and at the top of the composite rod (a) and resin modulus development at the same locations as a function of the axial distance (b).

Figure 15: The undeformed contour plots of the inplane stresses: $S_{11}$ (a) and $S_{22}$ (b).

between these two predictions is due to the aforementioned considerations in virtual section calculations, which relies on the specific assumptions in the FEM and in the SAM. Afterwards, due to resin chemical shrinkage, a continuous pressure reduction is observed until the detachment occurs.

According to the calculated viscosity and pressure profiles, in the thermochemical analysis the total pulling force together with its components is predicted (Figure 17). For the calculation of the frictional resistance, the friction coefficient $\mu$ has been assumed to be 0.25, as also used in [19]. Numerical outcomes show that, for the simulated process, the viscous force represents the principal amount of the total resistance, being $F_{\text{bulk}} = 4.9$ N, $F_{\text{visc}} = 313.7$ N, and $F_{\text{fric}} = 184.1$ N, as predicted by the semianalytical procedure. A relatively smaller frictional resistance (112.9 N) is predicted by the FE mechanical model, due to the lower contact pressure profile in Figure 16. The key role played by the viscous drag with respect to the frictional force can be related to the reduced die length affected by the frictional phenomena and to the delayed development of the resin (and the composite) modulus. The contribution due to the material compaction is found to be not significant as compared to other amounts, being less than 1% of the total load.

4. Conclusions

In the present work, different approaches for modeling and simulations of several physical aspects, such as fluid flow, heat transfer, chemical reaction, and solid mechanics, involved in a conventional pultrusion process are proposed and compared. The proposed models are based on different numerical techniques (FEM, FVM) as well as the analytical
Taking into account the discussed outcomes, it can be concluded that

(i) the resin pressure increases at the tapered die inlet, promoting the backflow of the excess resin; however, as soon as the straight portion of the die begins, a flat velocity profile is enforced. It is found that the compaction force increases with the viscosity (or degree of cure) of the resin in the impregnation bath;

(ii) adopted numerical schemes (FEM, FVM) are found to be accurate and converged to a reliable solution, since the predicted values match well with the reference data [6]. Moreover, both models fairly agree in the evaluation of viscosity profiles, dimensional variations and extension of the three zones (i.e., liquid, gel, and solid). The inclusion of the thermal contact resistance due to material contraction inside the die in pultrusion modeling does not affect the simulation results significantly;

(iii) in the initial portion of the curing die, the thermal expansion of the processing materials dominates the resin shrinkage, which induces a progressive contact pressure increase and, consequently, frictional resistance after gelation. However, as the cure reaction proceeds, the chemical contraction of the reactive resin prevails causing the detachment of the work piece from the die internal surface and the vanishing of the contact pressure as well as the frictional force;

(iv) in the residual stress model, relatively small residual stress values were predicted at the end of the process due to the uniform distribution of the temperature and degree of cure over the cross section of the composite part having a relatively small diameter (9.5 mm). The thickness of the composite part together with the total volumetric shrinkage of the resin has an important effect on the residual stress evolution [25]. At the end of the process, it is found that tension stresses prevail for the center of the part since it cured later and faster as compared to the outer regions where compression stresses were obtained while upholding the self-static equilibrium;

(v) the viscous drag is found to be the main contribution as the frictional force to the overall pulling force, while the contribution due to material compaction at the inlet is found to be negligible.

Investigating the several aforementioned processing physics simultaneously provides a better understanding of the entire pultrusion dynamics at a glance and therefore this study would be very much of interest to the composite manufacturing processing community and especially to scientists and engineers in the field of manufacturing process modeling.

Acknowledgment

This work is a part of DeepWind project which has been granted by the European Commission (EC) under FP7 Program Platform Future Emerging Technology.

References


“Reliability Estimation of the Pultrusion Process Using the First-Order Reliability Method (FORM)”

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Reliability Estimation of the Pultrusion Process Using the First-Order Reliability Method (FORM)

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Received: 15 August 2012 / Accepted: 5 September 2012 / Published online: 16 September 2012 © Springer Science+Business Media B.V. 2012

Abstract In the present study the reliability estimation of the pultrusion process of a flat plate is analyzed by using the first order reliability method (FORM). The implementation of the numerical process model is validated by comparing the deterministic temperature and cure degree profiles with corresponding analyses in the literature. The centerline degree of cure at the exit (CDOCE) being less than a critical value and the maximum composite temperature ($T_{\text{max}}$) during the process being greater than a critical temperature are selected as the limit state functions (LSFs) for the FORM. The cumulative distribution functions of the CDOCE and $T_{\text{max}}$ as well as the correlation coefficients are obtained by using the FORM and the results are compared with corresponding Monte-Carlo simulations (MCS). According to the results obtained from the FORM, an increase in the pulling speed yields an increase in the probability of $T_{\text{max}}$ being greater than the resin degradation temperature. A similar trend is also seen for the probability of the CDOCE being less than 0.8.

Keywords First order reliability method (FORM) · Pultrusion process · Reliability analysis · Curing · Computational modeling

1 Introduction

The pultrusion is a continuous manufacturing process for the production of composite structures with constant cross-sections. A schematic representation of the process with and without having a resin injection chamber can be seen in Fig. 1 (top) and (bottom). The fibers/mats and resin matrix are pulled together in the pultrusion direction by the pullers through the heating die and then the cured component is cut by a saw system.

There have been several studies related with the thermo-chemical simulation of the thermosetting pultrusion process [2–7]. Since pultrusion is a continuous process, the steady state temperature and the cure degree profiles inside the heating die play an important role for the characterization of the process quality. In [2–7], the overall goal was to obtain these
profiles and provide a better understanding of the process for the composite manufacturing industry. Beside the numerical studies, experimental studies of various composite profiles have been carried out in [8–14]. However, all these studies [1–14] are limited by the deterministic prescription of the process and its material parameters.

Composite materials have large statistical variations in their mechanical properties [15] which may be due to the uncertainties in volume fraction of the resin content, degree of cure and process induced residual stresses during the composite manufacturing, probability of the defects and void formations inside the composites, etc. Hence, there is a need for a probabilistic or reliability analysis of the composite failure or the manufacturing process conditions as well as the product quality. Such an analysis plays a vital role in the strength evaluation of the composite structures. In contrast to the deterministic analysis of the composite materials, the probabilistic analysis gives a better understanding of the effect of the variations inherently involved in the geometry, material properties or manufacturing process. This makes it easier or more practical to predict how sensitive the scatter of the output parameters (e.g. performance of the composite, failure criterion, degree of cure etc.) with respect to scatter in the input design parameters is. In other words, this provides a way for an evaluation of the robustness of the process.

In the literature several studies have been investigated related with the reliability of composite materials [16–23]. The reliability analysis of buried pipelines which are made of corrugated polyethylene was performed in [16] by using the first order reliability method.

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**Fig. 1** A schematic view of the pultrusion process with resin injection chamber (top) [1] and without resin injection chamber (bottom)
Three different limit state functions (LSFs) were defined based on the failure criterion of the deflection, bending stress and bending strain of the corrugated pipes. For the three design criteria, it was found that the bending stress design criterion yields the lowest probability of failure (PF); the deflection design criterion yields the greatest PF and the bending stress design criterion turns out to be a highly conservative design procedure. A numerical study related with the reliability analysis of a composite wind turbine blade section was investigated in [17]. The FORM was used for the reliability analysis of the numerical model and two LSFs were defined as the maximum deflection and the first-ply failure. The FORM gave accurate results as compared to the importance sampling based on the failure probabilities of the LSFs. A reliability based optimization was performed in [18] by using a genetic algorithm (GA) and two types of Artificial neural networks (ANN). The objective was to minimize the thickness of the composite plate while satisfying that the probability of the composite failure should not exceed a critical value. It was concluded that the FORM gave very similar results compared to Monte Carlo simulations with importance sampling in the optimization routines. However, the relative computational time for the FORM was much less than the time for the Monte Carlo simulations. In [19] the reliability of inserts in sandwich composite panels was analyzed by using different reliability/probabilistic methods such as the Monte-Carlo simulations (MCS), the FORM, line sampling method and the subset simulation method. It was concluded that the FORM did not give a true PF of the composite while the MCS and the subset simulation method did. The reason for this observation was stated as the FORM failed to converge at high loads because the optimization algorithm failed to deal with a ridge in the performance function (i.e. LSF) due to the transition from one failure mode to the other. This can also be explained that the FORM, most probably, found the local minimum rather than the global one while searching for the minimum distance from the origin to the LSF.

Probabilistic analyses of composite manufacturing processes, particularly resin transfer molding (RTM), have been carried out by several researchers [20–23]. In [22], the probability of the process-induced deformations of a composite part exceeding a specified allowable tolerance was calculated by using the FORM. In [23], the reliability assessment of the process temperature and the cure degree were investigated based on the random variables by using the FORM.

In the present work, a reliability assessment of the pultrusion process is investigated by using the FORM. Pultrusion of a flat plate is considered and the process set-up is taken from [4, 9]. The control volume based finite difference (CV/FD) method is used and the estimated steady state temperature and cure degree profiles of the composite match well those in similar analyses from literature [4, 9, 24, 31]. The cumulative distribution functions (CDFs) of the centreline degree of cure at the exit (CDOCE) and the maximum composite temperature ($T_{\text{max}}$) during the process are calculated by using the FORM and the results are compared with the MCS. Following this, the probability of the CDOCE being less than 0.8 and the probability of the $T_{\text{max}}$ being greater than 240 °C (degradation temperature of the epoxy resin, [3]) are investigated by using the FORM with respect to different coefficients of variations (COVs) of the random variables, pulling speeds and temperature increase in the heaters.

2 Governing Equations

The steady state energy equations in a Cartesian coordinate system for the composite and the die block are given in Eqs. 1 and 2, respectively. Here, $x$ is the pulling (axial or longitudinal)
direction; y and z are the transverse directions in height and width, respectively. In the energy equation for the composite part, the convective \((u \partial T/\partial x)\) and the source \((q)\) terms are present due to the advection or pulling of the composite material and the internal heat generation of the resin, respectively.

\[
\rho_c C_p \left( u \frac{\partial T}{\partial x} \right) = k_{x,c} \frac{\partial^2 T}{\partial x^2} + k_{y,c} \frac{\partial^2 T}{\partial y^2} + k_{z,c} \frac{\partial^2 T}{\partial z^2} + q
\]

(1)

\[
0 = k_{x,d} \frac{\partial^2 T}{\partial x^2} + k_{y,d} \frac{\partial^2 T}{\partial y^2} + k_{z,d} \frac{\partial^2 T}{\partial z^2}
\]

(2)

where \(T\) is temperature, \(u\) is the pulling speed, \(\rho\) is the density, \(C_p\) is the specific heat and \(k_x, k_y,\) and \(k_z\) are the thermal conductivities. The subscriptions \(c\) and \(d\) correspond to composite and die, respectively. Lumped material properties are used and assumed to be constant. The internal heat generation \((q)\) \([\text{W/m}^3]\) due to the exothermic reaction of the epoxy resin can be expressed as

\[
q = (1 - V_f) \rho_r Q
\]

(3)

where \(V_f\) is the fiber volume ratio and \(Q\) is the specific heat generation rate \([\text{W/kg}]\) due to the resin exothermic cure reaction.

The expression of the degree of cure \((\alpha)\) can be written as the ratio of the amount of heat generated \((H(t))\) during curing, to the total heat of reaction \(H_{tr}\), i.e. \(\alpha = H(t)/ H_{tr}\). The rate of the cure degree, \(R_r\), can be written as an Arrhenius equation \([9]\),

\[
R_r(\alpha) = \frac{d\alpha}{dt} = \frac{1}{H_{tr}} \frac{dH(t)}{dt} = K_o \exp \left( - \frac{E}{RT} \right) (1 - \alpha)^n
\]

(4)

where \(K_o\) is the pre-exponential constant, \(E\) is the activation energy, \(R\) is the universal gas constant and \(n\) is the order of reaction (kinetic exponent). \(K_o, E, H_{tr}\) and \(n\) can be obtained by a curve fitting procedure applied to the experimental data evaluated using differential scanning calorimetry DSC \([9]\). As a result, \(Q\) (in Eq. 3) can be expressed as

\[
Q = \frac{dH(t)}{dt} = H_{tr} R_r(\alpha)
\]

(5)

The transient time integration scheme for the rate of cure degree can be derived by using the chain rule. From this, the rate of cure degree (Eq. 4) can be expressed as:

\[
\frac{d\alpha}{dt} = \frac{\partial \alpha}{\partial t} + \frac{\partial \alpha}{\partial x} \frac{dx}{dt} = \frac{\partial \alpha}{\partial t} + u \frac{\partial \alpha}{\partial x}
\]

(6)

and from Eq. 6, the steady state relation of the resin kinetics equation can be expressed as

\[
\frac{\partial \alpha}{\partial t} = R_r(\alpha) - u \frac{\partial \alpha}{\partial x} = 0
\]

(7)

where it is the expression in Eq. 7 which is used in the numerical model.
3 Numerical Implementation

3.1 The CV/FD Method

A similar approach as in [24] using the CV/FD has been implemented for the discretization of the energy equations. The concept of the total thermal resistances, \( R \) (K/W), is applied to calculate the temperature as well as the degree of cure at the CVs [25]. In order to solve the matrix system for the temperature values in a stable and fast way, the internal heat generation, i.e. the source term \( q \) in Eq. 1, is coupled with the energy equation in an explicit manner during the iteration procedure. Therefore the degree of cure (in Eq. 8) is updated explicitly based on the calculated temperature. This iterative procedure can be seen as a flowchart in Fig. 2. In order to obtain accurate results and overcome the oscillatory behavior in the numerical implementation, the upwind scheme is used for the convective term in the energy equation \((u\partial T/\partial x)\) and for the discretization of the term \((u\partial \alpha/\partial x)\) in the resin kinetics equation. The built-in equation solver in MATLAB [26] is used to solve the equation system arising from the utilized numerical schemes.

3.2 The First Order Reliability Method (FORM)

The FORM solves the probability of failure integral, \( P(Y \leq 0) \), given in Eq. 8 by approximating the LSF, i.e. \( g(X) = 0 \), using the first order Taylor series expansion at the most probable point (MPP). The LSF represents a condition beyond which the relevant design criterion is no longer being fulfilled [27, 28].

\[
P(Y \leq 0) = \int_{g(x) \leq 0} f_X(x) dx
\]  

Fig. 2 Flowchart of the iteration procedure to solve the equation system for the temperature and the degree of cure
where \( f_X(x) \) is the joint probability density function of random variables \( X = [X_1, X_2, \ldots, X_n] \). These random variables are assumed to be mutually independent.

The FORM analysis involves three main steps:

1. Transformation of the random variables,
2. Search for the MPP,
3. Calculation of the failure probability.

The details of these steps are explained in the following:

1. This step requires transformation of the original random variables \( X = [X_1, X_2, \ldots, X_n]^T \) (in \( X \)-space) into standard normal variables \( U = [U_1, U_2, \ldots, U_n]^T \) (standard normal space, i.e. \( U \)-space) by using the Rosenblatt transformation [29] (Fig. 3):

\[
    u_i = \Phi^{-1}[F_{X_i}(x_i)], \quad i = 1, 2, \ldots, n 
\]

where \( \Phi^{-1} \) is the inverse CDF of the standard normal distribution, and \( F_{X_i}(x_i) \) is the CDF of \( X_i \). After the transformation, the LSF \( g(X) = 0 \) in \( X \)-space is expressed as \( g(U) = 0 \) in \( U \)-space.

2. Next, search for the MPP (the point on the integration boundary, i.e. \( g(U) = 0 \), with the minimum distance to the origin in \( U \)-space, see Fig. 3) is performed. This step needs an iterative optimization process to search for the minimum value of \( \beta \) in Eq. 10, where \( \beta \) is the distance between the MPP and the origin in \( U \)-space defined in Eq. 10 and is called the reliability index. For this purpose, a gradient based optimization algorithm is implemented in the present study where the search direction and the corresponding normalized magnitude vector are calculated.

\[
    \begin{cases}
        \min_u & \beta = ||u|| \\
        \text{s.t.} & g(u) = 0
    \end{cases} \tag{10}
\]

3. Finally, the probability of failure is calculated. At the MPP, the joint probability density function of \( U \) has the highest value on the limit state in \( U \)-space. The LSF is linearized

![Fig. 3 An overview of the FORM procedure for two random variables [30]](image-url)
at the MPP in U-space to ensure the minimum accuracy loss. The probability in Eq. 8 is then calculated analytically by the following equation,

$$F_Y(y) = P[Y \leq 0] \simeq \Phi(-\beta)$$

(11)

where $\Phi$ is the CDF of the standard normal distribution.

4 Validation Case

4.1 Problem Description

A three dimensional steady state simulation of a flat plate is performed as a validation case. A fiberglass/Shell EPON 9420 epoxy based composite and a steel die is used for the flat plate and the die block, respectively [9]. The material properties and the resin kinetic parameters are given in Tables 1 and 2, respectively. A schematic view and the dimensions of the model are seen in Fig. 4. Instead of modeling the heaters, insulators and metal platens as in [9], three heating zones having set temperatures of 171–188–188 °C (in the order of the pulling direction) are defined. The spacing between the heaters is 15 mm.

Only the quarter of the pultrusion domain is considered due to symmetry. A schematic view of the CVs and the nodes of the composite plate and the die block are given in Fig. 5 including the dimensions of the cross section. 18 × 18 and 18 × 18 × 122 nodes are used for the cross section and the whole model, respectively. Perfect thermal contact is assumed at the die-part interface as in [4]. Initially the temperature of all the nodes are assigned to ambient temperature (27 °C) and the cure degree of all composite nodes are assigned to 0. The temperature and cure degree of all composite nodes at the die inlet are set to the resin bath temperature (30 °C) and 0, respectively. At the symmetry surfaces, adiabatic boundaries are defined on which no heat is transmitted across the boundaries. The remaining exterior surfaces of the die are exposed to ambient temperature with a convective heat transfer coefficient of 10 W/m²K except for those located at the heating regions. At the die exit, an adiabatic boundary condition is applied to the composite surface. The cooling channels are located at the first 100 mm under the first heating region. Hence, all nodes at the layers A-A and B-B in Fig. 5 are set to cooling temperature (50 °C) during the whole process. The convergence limits of temperature and degree of cure for the iterative procedure (Fig. 2) are set to 0.001 °C and 0.0001, respectively.

4.2 Results and Discussion of the Validation Case

The predicted centerline temperature and cure degree profiles for the composite flat plate are seen in Fig. 6 (top) and (bottom), respectively for a pulling speed of 20 cm/min. It is seen

<table>
<thead>
<tr>
<th>Material</th>
<th>$\rho$ (kg/m³)</th>
<th>$C_p$ (J/kg K)</th>
<th>$k_x$ (W/m K)</th>
<th>$k_y, k_z$ (W/m K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Epoxy resin</td>
<td>1260</td>
<td>1255</td>
<td>0.21</td>
<td>0.21</td>
</tr>
<tr>
<td>Fiberglass</td>
<td>2560</td>
<td>670</td>
<td>11.4</td>
<td>1.04</td>
</tr>
<tr>
<td>Lumped ($V_f=63.9%$)</td>
<td>2090.7</td>
<td>797.27</td>
<td>0.9053</td>
<td>0.5592</td>
</tr>
<tr>
<td>Steel die</td>
<td>7833</td>
<td>460</td>
<td>40</td>
<td>40</td>
</tr>
</tbody>
</table>
that the results agree well with those calculated by using a transient analysis for the similar set-up in [4, 9]. It is seen from Fig. 6 (top) that the maximum centerline temperature is predicted to be approximately 190 °C which is higher than the heater temperature (i.e. 188 °C). This is due to the fact that while curing is progressing, after some point in time, an exothermic internal heat generation is taking place inside the thermosetting resin. The centerline degree of cure at the exit (CDOCE) is calculated as approximately 0.91.

5 Reliability Estimation

5.1 Problem Description

In the present study, the random variables listed in Table 3 have been utilized to predict and assess the reliability of the pultrusion process. A normal distribution with a mean ($\mu$) and a standard deviation ($\sigma$) is used where $\sigma = \mu \times \text{COV}$. In general, these statistical characteristics are obtained from an extensive data collection and data analysis. In the present study, the mean values of the random variables are taken from the deterministic analysis given in Section 4 and the standard deviation values are defined based on the studies carried out in [23, 31]. Here, the 4th random variable, namely the heater temperature multiplier (“$\text{cons}$”), is defined as a multiplier for all three heater temperatures. For instance, the temperature of the first heater becomes a random variable by multiplying it with a random parameter, i.e. $\text{cons}$. Hence, it has a normal distribution with a mean ($\mu$) of 171 °C and a standard deviation ($\sigma$) of $171 \times 0.022 \approx 3.4 \degree C$. The same approach is also valid for the other two heater temperatures.

In a pultrusion process, the CDOCE is desired to be sufficiently high while the maximum composite temperature ($T_{\text{max}}$) is expected not to be higher than the degradation temperature of the epoxy resin. Hence, the CDOCE being less than a certain value (i.e. critical cure

Table 2  Resin kinetic parameters

<table>
<thead>
<tr>
<th>$H_n$ (J/kg)</th>
<th>$K_0$ (1/s)</th>
<th>$E$ (J/mol)</th>
<th>$n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>324000</td>
<td>192000</td>
<td>60000</td>
<td>1.69</td>
</tr>
</tbody>
</table>

Fig. 4  Quarter of the pultrusion domain for the composite flat plate. All dimensions are in mm.
degree, $\alpha_{\text{crit}}$) and $T_{\text{max}}$ being greater than another value (i.e. critical temperature, $T_{\text{crit}}$) are selected as the LSFs and given in Eqs. 12 and 13, respectively.

$$LSF_1 : \quad CDOCE \leq \alpha_{\text{crit}}$$

$$LSF_2 : \quad T_{\text{max}} \geq T_{\text{crit}}$$

5.2 Results and Discussion of the Reliability Analysis

The CDF of the CDOCE and $T_{\text{max}}$ are calculated by using the FORM based on the random variables given in Table 3. The CDFs can be expressed as the probability of the CDOCE being less than $\alpha_{\text{crit}}, P(CDOCE \leq \alpha_{\text{crit}})$ and the probability of $T_{\text{max}}$ being greater than $T_{\text{crit}}, P(T_{\text{max}} \geq T_{\text{crit}})$. In order to validate the implementation of the FORM, the same CDFs are also predicted by using the MCS with the LHS. The MCS is one of the most common techniques used for uncertainty analyses. The LHS technique is selected for the sampling method since it has a sample memory which avoids the repetition of the samples. However, its accuracy depends on the sample size and computational expense is much higher as compared with that of FORM for a certain number of random variables.

A total of 1,000 samples (function evaluations) are used in the MCS analysis. On the other hand, it requires approximately 50 function evaluations for the FORM to calculate the probability of failure for this problem which has 14 design variables (Table 3). The FORM is performed for 14 different $\alpha_{\text{crit}}$ and 8 different $T_{\text{crit}}$ values used in the LSF$_1$ (Eq. 12) and LSF$_2$ (Eq. 13), respectively based on the equivalent division of the CDOCE and $T_{\text{max}}$ intervals. The results are given in Fig. 7 (left) and (right) for the CDF of the CDOCE and $T_{\text{max}}$. It is seen that the FORM gives accurate results as compared with the ones obtained from the MCS. For instance, the probability of the CDOCE being less than 0.89, i.e. $P(CDOCE \leq 0.89)$ is approximately 20% (i.e. indicated with a solid line in Fig. 7, left) and the
probability of $T_{\text{max}}$ being greater than 195 °C, i.e. $P(T_{\text{max}} \geq 195 \degree \text{C})$, is approximately 7 % (100-93=7 %) (i.e. similarly in Fig. 7, right).

Regarding the sensitivities of the random variables with respect to the CDOCE and the $T_{\text{max}}$, similar results are obtained from the FORM and the MCS. The sensitivities are taken from the gradient information of the random variables for the FORM, i.e. the normalized magnitude vector calculated during the optimization procedure to search for the minimum $\beta$.
The normalized magnitude vector is called the sensitivity indicator. On the other hand, the sensitivities are obtained by calculating the linear correlation coefficients for the MCS. The sensitivities of the random input variables are given in Figs. 8 and 9 as a pie chart for the CDOCE and the $T_{\text{max}}$, respectively. It is seen from Fig. 8 that the FORM and the MCS results are very close to each other based on the most sensitive four random variables, namely the activation energy ($E$), the heater temperature multiplier ($\text{cons}$), the order of reaction ($n$) and the pulling speed which cover almost 90% of the sensitivity distribution in Fig. 8. According to the sensitivity indicator for the FORM as a bar plot in Fig. 8 (left) and the linear correlation coefficients for the MCS in Fig. 8 (right), the statistical variation in the activation energy ($E$) and the heater temperature multiplier ($\text{cons}$) has the highest effect on the variation in the CDOCE. Similar observation has also been given in [31] using the MCS with three different sampling procedures. The linear correlation coefficient or the sensitivity indicator value for the $E$ and the $\text{cons}$ are calculated as approximately ($-0.78$) and ($0.61$), respectively.

### Table 3: The random variables and their statistical characterizations for the pultrusion process

<table>
<thead>
<tr>
<th>NR.</th>
<th>Property</th>
<th>Symbol</th>
<th>Unit</th>
<th>$\mu$</th>
<th>COV</th>
<th>Distribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Pulling speed</td>
<td>$u$</td>
<td>cm/min</td>
<td>20</td>
<td>0.02</td>
<td>Normal</td>
</tr>
<tr>
<td>2</td>
<td>Fiber volume ratio</td>
<td>$V_f$</td>
<td>–</td>
<td>0.639</td>
<td>0.02</td>
<td>Normal</td>
</tr>
<tr>
<td>3</td>
<td>Inlet temperature</td>
<td>$T_{\text{left}}$</td>
<td>°C</td>
<td>30</td>
<td>0.02</td>
<td>Normal</td>
</tr>
<tr>
<td>4</td>
<td>Heater temperature multiplier</td>
<td>$\text{cons}$</td>
<td>–</td>
<td>1</td>
<td>0.02</td>
<td>Normal</td>
</tr>
<tr>
<td>5</td>
<td>Density of resin</td>
<td>$\rho_r$</td>
<td>kg/m$^3$</td>
<td>1260</td>
<td>0.05</td>
<td>Normal</td>
</tr>
<tr>
<td>6</td>
<td>Density of fiber</td>
<td>$\rho_f$</td>
<td>kg/m$^3$</td>
<td>2560</td>
<td>0.05</td>
<td>Normal</td>
</tr>
<tr>
<td>7</td>
<td>Specific heat of resin</td>
<td>$C_{p_r}$</td>
<td>J/kg K</td>
<td>1255</td>
<td>0.05</td>
<td>Normal</td>
</tr>
<tr>
<td>8</td>
<td>Specific heat of fiber</td>
<td>$C_{p_f}$</td>
<td>J/kg K</td>
<td>670</td>
<td>0.05</td>
<td>Normal</td>
</tr>
<tr>
<td>9</td>
<td>Thermal conductivity of resin</td>
<td>$k_r$</td>
<td>W/m K</td>
<td>0.21</td>
<td>0.05</td>
<td>Normal</td>
</tr>
<tr>
<td>10</td>
<td>Thermal conductivity of fiber in $y$-axis</td>
<td>$(k_{y_f})$</td>
<td>W/m K</td>
<td>1.04</td>
<td>0.05</td>
<td>Normal</td>
</tr>
<tr>
<td>11</td>
<td>Thermal conductivity of fiber in $x$-axis</td>
<td>$(k_{z_f})$</td>
<td>W/m K</td>
<td>11.4</td>
<td>0.05</td>
<td>Normal</td>
</tr>
<tr>
<td>12</td>
<td>Total heat of reaction</td>
<td>$H_{tr}$</td>
<td>J/kg</td>
<td>324000</td>
<td>0.01</td>
<td>Normal</td>
</tr>
<tr>
<td>13</td>
<td>Pre-exponential constant</td>
<td>$K_0$</td>
<td>1/s</td>
<td>192000</td>
<td>0.01</td>
<td>Normal</td>
</tr>
<tr>
<td>14</td>
<td>Activation energy</td>
<td>$E$</td>
<td>J/mol</td>
<td>60000</td>
<td>0.01</td>
<td>Normal</td>
</tr>
<tr>
<td>15</td>
<td>Order of reaction</td>
<td>$n$</td>
<td>–</td>
<td>1.69</td>
<td>0.01</td>
<td>Normal</td>
</tr>
</tbody>
</table>

Fig. 7 The CDF of the CDOCE (left) and $T_{\text{max}}$ (right) predicted by using the FORM and MCS with LHS.
respectively. Here, a positive correlation indicates that an increase in the input parameter provides an increase in the output parameter and vice versa. According to Fig. 9, cons has the highest sensitivity indicator value or correlation coefficient (positive) and the magnitude is close to 1 (0.98) which indicates that the variation in the heater temperatures is strongly correlated with the $T_{\text{max}}$ (maximum composite temperature). The corresponding sensitivities are given in a pie chart in Fig. 9 (left) and (right) for the FORM and the MCS, respectively. The difference between the sensitivities obtained from the FORM and the MCS results can be referred to the accuracy of the FORM which is based on the availability of the gradient information, however for the MCS the accuracy depends on the sampling size. The random variable cons covers approximately 80% of all the sensitivity distribution.

The relationship between the probability of the CDOCE being less than 0.8, i.e. $P(\text{CDOCE} \leq 0.8)$, and the COV of the most sensitive random variables taken from Fig. 4 are shown in Fig. 10. The FORM is used for only one failure probability, i.e. $P(\text{CDOCE} \leq 0.8)$, to exemplify the potential use of the technique. In general, the uncertainty or the scatter in the random variables increases with a larger COV. This also provides an increase in the probability of failure, therefore $P(\text{CDOCE} \leq 0.8)$ is expected to increase. The scatter in the $E$ has a more significant effect on $P(\text{CDOCE} \leq 0.8)$ than the scatter in cons according to the

**Fig. 8** The sensitivity indicators and the linear correlation coefficients calculated by using the FORM (left) and the MCS (right), respectively, for the CDOCE

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**Fig. 9** The sensitivity indicators and the linear correlation coefficients calculated by using the FORM (left) and the MCS (right), respectively, for $T_{\text{max}}$
Fig. 10. On the other hand, the scatter in the pulling speed \( u \) and the order of reaction \( n \) have a much smaller effect on \( P(CDOCE \leq 0.8) \) as compared to the \( E \) and \( \text{cons} \). Here, it should be noted that these random variables are varied independently at the corresponding COV values in Fig. 10.

The probability of the maximum composite temperature \( T_{\text{max}} \) being greater than the degradation temperature of the resin (240 °C, [3]), i.e. \( P(T_{\text{max}} \geq 240 \, ^\circ\text{C}) \), and \( P(CDOCE \leq 0.8) \) are estimated by using the FORM with respect to different pulling speeds \( u \) and the temperature increase (\( \Delta T \)) in the set temperature of the heaters. The random variables listed in Table 3 are used including \( u \) and it should be noted that an increase in \( \mu \) results in an increase in the corresponding \( \sigma \) value. For instance, if the \( \Delta T \) is 10 °C, the set temperatures of the heaters become \((171–188–188 \, ^\circ\text{C}) + \Delta T = (181–198–198 \, ^\circ\text{C})\) in the order of pulling direction. The results based on the statistical variation defined in Table 3 are given in Fig. 11 (left) and (right) for \( P(T_{\text{max}} \geq 240 \, ^\circ\text{C}) \) and \( P(CDOCE \leq 0.8) \), respectively. It is seen from Fig. 11 (left) that \( P(T_{\text{max}} \geq 240 \, ^\circ\text{C}) \) increases with the increase in \( \Delta T \) as expected. In addition to that \( P(T_{\text{max}} \geq 240 \, ^\circ\text{C}) \) also increases with \( u \) for \( \Delta T \) values greater than 30–40 °C. The reason is that the position of the peak exothermic reaction of the epoxy resin is shifted towards the die exit as \( u \) increases and the reaction is occurring under the 2nd and 3rd instead of the 1st heater which a has lower set temperature. In addition to that, the corresponding

Fig. 11 Probability of the maximum composite temperature being greater than 240 °C \( (P(T_{\text{max}} \geq 240 \, ^\circ\text{C}) \) \( (\text{left}) \) and the probability of the CDOCE being less than 0.8 \( (P(CDOCE \leq 0.8)) \) \( (\text{right}) \) with respect to different pulling speeds and the temperature increase (\( \Delta T \)) in the set temperature of the heaters
increase in $\sigma$ for higher $u$ values also promotes an increase in $P(T_{\text{max}} \geq 240 \, ^\circ\text{C})$. Hence, higher temperatures are observed for $T_{\text{max}}$ which also increases $P(T_{\text{max}} \geq 240 \, ^\circ\text{C})$. Moreover, the positive linear coefficient of $u$ in Fig. 9 also indicates that there is a positive correlation between $u$ and $T_{\text{max}}$. Although the heater temperatures are 206–223–223 °C for $\Delta T=35$ °C, $P(T_{\text{max}} \geq 240 \, ^\circ\text{C})$ is estimated to be around 11 % with a pulling speed of 100 cm/min due to the exothermic internal heat generation of the epoxy resin.

According to Fig. 11 (right), $P(CDOCE \leq 0.8)$ decreases with an increase in $\Delta T$ since the cure degree increases with $\Delta T$. This information can also be obtained from Fig. 8 where there is a positive correlation between the heater temperature multiplier ($\text{cons}$) and the CDOCE which results in a decrease for $P(CDOCE \leq 0.8)$. On the other hand, an increase in $u$ yields an increase in $P(CDOCE \leq 0.8)$ or a decrease in CDOCE. This is also expected from the negative correlation coefficient between $u$ and the CDOCE in Fig. 8. To illustrate, for $\Delta T=0$ and $u=100$ cm/min, $P(CDOCE \leq 0.8)$ is 1 (100 %) in Fig. 11 (right) which shows that the CDOCE is less than 0.8 for sure. However, for the original case, i.e. $\Delta T=0$ and $u=20$ cm/min, $P(CDOCE \leq 0.8)$ is almost 0 as found in Fig. 3 (left).

6 Conclusions

In the present study the application of the FORM for the reliability estimation of the pultrusion process was investigated. The reliability analyses of a flat plate were performed based on the deterministic model taken from the literature [4, 9]. The implementation of the CV/FD method was validated by comparing the centerline temperature and cure degree profiles for the composite flat plate with those in [4, 9]. The CDOCE being less than a value, i.e. $CDOCE \leq \alpha_{\text{crit}}$ and $T_{\text{max}}$ being greater than a value, i.e. $T_{\text{max}} \geq T_{\text{crit}}$ were selected as the limit state functions (LSFs). The CDFs of the CDOCE and $T_{\text{max}}$ as well as the corresponding sensitivity indicator values were calculated using the FORM. It was seen that the FORM gave accurate results and took much lower computational time as compared to the corresponding analysis with the MCS. The statistical variation in the activation energy ($E$) and the heater temperature multiplier ($\text{cons}$) were found to have the highest effect on the variation in the CDOCE. It was also concluded that the $\text{cons}$ had the highest correlation coefficient (positive) which indicated that the variation in the heater temperatures was strongly correlated with the $T_{\text{max}}$. According to the COV analysis, the scatter in the $E$ had more significant effect on the probability of the CDOCE being less than 0.8, i.e. $P(CDOCE \leq 0.8)$, than the scatter in $\text{cons}$. It was also found that an increase in the pulling speed ($u$) yielded an increase in the probability of $T_{\text{max}}$ being greater than the resin degradation temperature, i.e. $P(T_{\text{max}} \geq 240 \, ^\circ\text{C})$. A similar trend was also seen for $P(CDOCE \leq 0.8)$.

Acknowledgment This work is a part of DeepWind project which has been granted by the European Commission (EC) under FP7 program platform Future Emerging Technology.

References


“Probabilistic Analysis of a Thermosetting Pultrusion Process”

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*Science and Engineering of Composite Materials (SECM)*
2014. (*Accepted*).
Probabilistic Analysis of a Thermosetting Pultrusion Process

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Abstract

In the present study, the effect of the uncertainties in the material properties of the processing composite material and the resin kinetic parameters, as well as the process parameters such as pulling speed and inlet temperature on the product quality (exit degree of cure) are investigated for a pultrusion process. A new application for the probabilistic analysis of the pultrusion process is introduced using the Response Surface Method (RSM). The results obtained from the RSM are validated by employing the Monte Carlo simulation (MCS) with Latin Hypercube Sampling (LHS) technique. According to the results obtained from both methods, the variations in the activation energy as well as the density of the resin are found to have a relatively stronger influence on the centerline degree of cure at the exit. Moreover, different execution strategies are examined for the MCS to investigate their effects on the accuracy of the random output parameter.

Keywords: Pultrusion, Monte Carlo Simulation, Response Surface Method, Probability, Computational analysis.
1. Introduction

Pultrusion is a continuous process of manufacturing composite profiles having constant cross sectional area. It has been widely used for producing high strength and lightweight composite structures such as wind turbine blades, ballistic resistance panels, spars of ship hulls, thin wall panel joiners, door/window frames and drive shaft of the vehicles. A schematic representation of the process can be seen in Fig. 1. The reinforcing material (Rovings/mats) and the resin matrix are pulled together via a pulling mechanism through the preforming guiders. The wetted reinforcements advance through a heating die and then the cured product is cut into desired final length by a saw mechanism.

Virtual manufacturing, in essence applying a numerical process simulation, is an important step towards designing enduring and better-functioning products in the pultrusion process as well as in other manufacturing processes. There have been several numerical modelling studies specific to the thermo-chemical simulation of the pultrusion process in literature. Generally, the numerical techniques such as the finite difference method (FDM) and the finite element method (FEM) with control volume (CV) technique or the nodal control volume (NCV) technique have been used for the simulation of the process [1–7]. According to the results obtained in [1–7], similar temperature and cure degree behaviours have been found, e.g. the temperature of the composite is initially lagging behind the die temperature, however the composite temperature exceeds the die temperature during curing.
due to the internal heat generation of the resin. As a means of supporting the numerical thermo-chemical modelling of the pultrusion process, experimental studies of various composite profiles have been carried out in [8–13]. However, the studies [1–14] are limited by the deterministic prescription of the process and its material parameters. The uncertainties in the simulation of composite manufacturing processes, particularly the resin transfer molding (RTM), have been studied by several researchers [15–18]. In [15], the effect of the variation in the isothermal curing temperature and the resin kinetic parameters on the cure time was investigated for the RTM process using the Latin Hypercube Sampling (LHS) technique. It was concluded that the statistical variation in the curing temperature was found to have a greater impact on the curing time as compared to the change in the resin kinetic parameters. The same sampling technique was used in [16] in order to explore the uncertainties in the preform permeability, the resin viscosity and the kinetic parameters for the RTM process. The random output variables were determined as the fill time and the maximum cure inside the composite at the end of the non-isothermal filling process during RTM. In [17], the probability of the process-induced deformations of a composite part exceeding a specified allowable tolerance was calculated by using the first-order reliability method (FORM) integrated with a deterministic modelling tool which simulates the various physical phenomena during processing of composite structures. In [18], the sensitivities and the probabilities of the maximum and the minimum process temperature and the cure degree were investigated for the prescribed random input variables by implementing a gradient based reliability analysis, i.e. FORM. Following the sensitivity analysis, an efficient two-level reliability based design optimization (RBDO) approach was performed in order to minimize the cure cycle time and associated manufacturing cost for a RTM kind process. Apart from the composite manufacturing processes, probabilistic analyses of various engineering applications [19–23] have been investigated by using the probabilistic design system (PDS) toolbox of ANSYS [24].

In the present paper a new application for the probabilistic analysis of the pultrusion process is presented. The effects of the uncertainties in the pultrusion process parameters on the degree of cure at the die exit are investigated. The deterministic analysis is based on a thermo-chemical model of the pultruded AS4/Epon 9420/9470/537 carbon fiber/epoxy composite rod. It is performed for the validation purpose using the finite element with nodal control volume (FE/NCV) method. For the probabilistic analysis, Monte Carlo simulation (MCS) and the response surface method (RSM), which is the first contribution of its kind in the numerical modelling of pultrusion process, have been performed. The LHS technique is used in the MSC and the RSM for generating the sample data. For this purpose, the PDS toolbox of ANSYS is utilized incorporating with a parametric deterministic model developed by the authors using its own scripting language APDL (ANSYS Parametric Design Language). This approach gives a better understanding of the effect of the variations or uncertainties inherently being present in the process and makes it easier or more practical to predict how large and sensitive the scatter of the output parameters (e.g. exit degree of cure) with respect to scatter in the input design parameters is. In other words, this study provides practical information about the robustness of the process model.
2. Governing Equations

The steady state heat transfer equation in a two dimensional (2D) cylindrical coordinate system for the composite rod can be written as [4]:

\[
\rho C_p u \frac{\partial T}{\partial z} = k_z \frac{\partial^2 T}{\partial z^2} + \frac{k_r}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right) + q
\]  

(1)

where \( T \) is the temperature, \( u \) is the pulling speed, \( \rho \) is the density, \( C_p \) is the specific heat, \( k_z \) and \( k_r \) are the thermal conductivities in the axial or pulling direction (z) and in the radial direction (r), respectively. Lumped material properties are used for the pultruded composite rod and assumed to be constant. The internal heat generation \( q \) [W/m\(^3\)] due to the exothermic reaction of the epoxy resin can be expressed as [8]:

\[
q = (1 - V_f) \rho_r Q
\]  

(2)

where \( V_f \) is the fiber volume fraction, \( \rho_r \) is the density of the epoxy resin and \( Q \) is the specific heat generation [W/kg] due to the exothermic cure reaction of the resin.

The expression of the degree of cure (\( \alpha \)) can be written as the ratio of the amount of heat generated \( (H(t)) \) during curing, to the total heat of reaction \( H_{tr} \), i.e. \( \alpha = H(t)/H_{tr} \). The rate of the degree of cure, \( R_r \), can be written as an Arrhenius type of equation [8]:

\[
R_r = \frac{d\alpha}{dt} = \frac{1}{H_{tr}} \frac{dH(t)}{dt} = K_o \exp\left(-\frac{E}{RT}(1 - \alpha)^n\right)
\]  

(3)

where \( K_o \) is the pre-exponential constant, \( E \) is the activation energy, \( R \) is the universal gas constant and \( n \) is the order of reaction (kinetic exponent). \( K_o, E \) and \( n \) can be obtained by curve fitting procedure applied to the experimental data evaluated using the differential scanning calorimetry (DSC) tests [8]. As a result, \( Q \) (Eq. 2) can be expressed as:

\[
Q = \frac{dH(t)}{dt} = H_{tr} R_r
\]  

(4)

By using the chain rule, the rate of degree of cure \((d\alpha/dt)\) can be expressed as:

\[
\frac{d\alpha}{dt} = \frac{\partial \alpha}{\partial t} + \frac{\partial \alpha}{\partial z} \frac{dz}{dt} = \frac{\partial \alpha}{\partial t} + u \frac{\partial \alpha}{\partial z}
\]  

(5)

and from Eq. 5, the steady state resin kinetics equation is written as:

\[
\frac{\partial \alpha}{\partial t} = 0 = R_r - u \frac{\partial \alpha}{\partial z}
\]  

(6)

which is used in the thermo-chemical analysis of the pultrusion process.
3. Numerical Implementation

The FE/NCV method, which is well documented in the literature [2, 3] is implemented for the steady state simulation of the pultrusion of the composite rod using the APDL in ANSYS [24]. The internal heat generation (Eq. 2) together with the resin kinetics equation (Eq. 3) is coupled with the energy equation (Eq. 1) in an explicit manner in order to obtain a fast numerical procedure since the internal heat generation is highly non-linear [25]. The degree of cure is subsequently updated explicitly for each NCV using Eq. 6 in its discretized form. The upwind scheme is used for the space discretization of the cure degree \( \frac{\partial \alpha}{\partial z} \) in Eq. 6 in order to obtain a stable solution. The degree of cure as well as the internal heat generation is updated explicitly at each node, as mentioned before, according to the calculated steady state nodal temperature data in ANSYS.

4. Validation Case: Deterministic analysis

The deterministic thermo-chemical pultrusion simulation of a composite rod taken from the literature [8] is performed as a validation case. The temperature and the cure degree distributions are predicted with a given die wall temperature profile based on the experimental work of Valliappan et al. [8]. The model geometry and the boundary conditions are shown in Fig. 2 (left). An axi-symmetrical model is used since the die wall temperature profile \( T_w(z) \) in Fig. 2) is assumed to be constant throughout the periphery of the rod. The graphite fiber reinforcement (Hercules AS4-12K) and epoxy resin (SHELL EPON9420/9470/537) system are used for the composite. The thermo-physical properties of the composite are given in Fig. 2 (right). The resin kinetic parameters used in the simulations are [8]: \( H_tr = 323,700 \) [J/kg], \( K_o = 191,400 \) [1/s], \( E_o = 60,500 \) [J/mol] and \( n = 1.69 \). The preheating temperature of the composite rod, i.e. \( T_{left} \) in Fig. 2, is taken as the resin bath temperature 38\(^\circ\)C. The initial cure degree of the composite is assumed to be 0. In order to reach the steady state the convergence limits are defined as the maximum temperature and cure degree difference between the new time step and the old time step and these are set to 0.001\(^\circ\)C and 0.0001, respectively.

The steady state temperature and cure degree distributions are predicted using the deterministic model with a pulling speed of 30 cm/min. The centerline temperature and degree of cure profiles are depicted in Fig. 3. It is seen that the results are found to agree well with the experimental data (for temperature) and the predicted data (for the degree of cure) provided in [8]. This shows that the employed deterministic thermo-chemical model gives converged results with a reliable solution. It is seen in Fig. 3 that the centerline temperature of the composite becomes higher than the die wall temperature in a distance of approximately 380 mm from the die inlet due to the internal heat generation. The peak temperature of the composite is found to be approximately 205\(^\circ\)C and the centerline degree of cure at exit is calculated as 0.844.
5. Probabilistic Analysis

5.1. Probabilistic Methods

In the present work, two different probabilistic approaches in particular the MCS and the RSM are implemented using the ANSYS PDS toolbox. The MCS is a “sampling” based method whereas the RSM is a “curve fitting” or “surrogate modelling” method which is used to approximate a function in terms of its variables. The details of these methods are explained in the following.

5.1.1. Monte Carlo Simulation (MCS)

The MCS is one of the most common “sampling” techniques used for the uncertainty or the sensitivity analyses. It is based on the random sampling of the input parameters for each simulation loop. The simulation loop or sample mentioned here indicates the iterative execution of a random parameter set. The LHS is selected for the sampling method since no overlap among the sample points is guaranteed which provides more random (in other words, distributed wide apart) sampling procedure than the direct sampling technique [20, 24]. Assuming the deterministic model is correct; the MCS theoretically converges to the correct probabilistic results with an increasing number of samples. This also forces the tails of a distribution to participate in the sampling process which is often not the case in real world applications having limited number of samples [19]. In order to capture the tails in a probability density function, an extremely large number of simulations is required for the MCS. For a given probability of failure ($p_f$), the minimum number of sample points $N_{min}$ required for the MCS is defined as [26, 27]:

$$N_{min} \propto \frac{1 - p_f}{p_f \delta^2}$$

(7)

where $\delta$ is the desired accuracy of the simulation result. It is seen that the $N_{min}$ is inversely proportional with the $p_f$ and $\delta$. For instance, for a given probability of 0.9999 ($p_f = 1 - 0.9999 = 0.0001$) in the tail, around 1 million sample points are needed in order to ensure a 10% accuracy of the estimated result from the MCS. It should be noted that the main focus of the present work is not on the evaluation of very low probabilities (tail probabilities). Despite the high computational cost, the MCS does not use any assumptions or simplifications on the input-output parameters which makes it easy to use.

The results of the MCS are based on the statistical procedures such as calculation of the mean, the standard deviation, the cumulative density functions and the correlations. The cumulative distribution function (CDF), here denoted as $F_i$, of a sampled data is derived from the cumulative binomial distribution function [19, 24, 28]:

$$\sum_{k=i}^{N} \binom{N}{k} F_i^k (1 - F_i)^{N-k} = 0.5$$

(8)
where the CDF of the $i^{th}$ sample data out of $N$, i.e. $F_i$, is solved numerically in ANSYS for the data sorted in ascending order.

In the present study, a linear correlation is used between the two random variables, e.g. the $i^{th}$ random input variable and the $i^{th}$ output variable. The correlation coefficients are calculated according to the following relation in ANSYS [24]:

$$r_p = \frac{\sum_{i}(x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_{i}(x_i - \bar{x})^2} \sqrt{\sum_{i}(y_i - \bar{y})^2}}$$

(9)

where $r_p$ is the correlation coefficient between the two random variables $x$ and $y$, $n$ is the sample size and $\bar{x}$ and $\bar{y}$ are the mean of the sample data $x$ and $y$, respectively.

5.1.2. Response Surface Method (RSM)

A true input-output relationship used in the MCS is replaced with an approximation function in the RSM. The response surfaces are generated with the use of the conventional Design of Experiments (DOEs). The accuracy of the results depend on the number of sample points or the DOEs employed in the RSM. There are two main steps in the RSM: The first one is the calculation of the random output parameters by performing the sufficient simulation loops based on the sample points, i.e. the generation of the response surfaces with the DOEs. The sample points or the DOEs are located in the space of the random input variables where the approximated mathematical function can be obtained most efficiently. The second step is the application of a linear regression analysis to determine the coefficients of the approximation function which is typically a quadratic polynomial defined as:

$$\hat{y} = c_0 + \sum_{i=1}^{n} c_i x_i + \sum_{i=1}^{n} \sum_{j=1}^{n} c_{ij} x_i x_j$$

(10)

where $\hat{y}$ is the approximation function, $n$ is the sample size, $x_i$'s are the random input variables, $c_0$ is the coefficient of the constant term, $c_i$'s are the coefficients of the linear terms and $c_{ij}$'s are the coefficients of the quadratic terms. In order to calculate these coefficients, a linear regression procedure [29] is utilized such that the sum of the squared differences between the true simulation results and the values of the approximation function, i.e. the magnitude of the residual, is minimized. Once the coefficients in Eq. 10 are estimated, then Eq. 10 can be used directly for the calculation of the output parameter instead of looping through the deterministic FE model. Hence, evaluating the quadratic polynomial thousands of times may require only a couple of seconds of computation time whereas the use of the original model may take minutes to hours. This approximation method is sufficient in many cases of engineering applications if the output or response parameter is a continuous or smooth function of the input variables.
A central composite design (CCD) is one of the commonly used and popular design for fitting the second-order response surfaces (quadratic polynomials). It is widely used in practice due to its efficiency in terms of the number of required function evaluations [29]. The total number of sample points are determined according to the following relation for the CCD design in ANSYS [24]:

\[ 2^{n-f} + 2n + 1 \]  

which is used to form the corresponding \( n \)-dimensional hypercube (response surface) for the number of input variables \( n \). Here, \( f \) is the fraction of the factorial points used in ANSYS based on \( n \) [24, 29]. In other words, a CCD is composed of 2 axis points per input variable (i.e. \( 2n \)), factorial points at the corner of the hypercube (i.e. \( 2^{n-f} \)) and one central point located at the center of the hypercube.

5.2. Description of the Probabilistic Model

For the probabilistic analysis of the pultrusion process, the pulling speed, fiber volume ratio, inlet temperature, all the characteristic material properties and the resin kinetic parameters are considered as the random input parameters (RIPs). Table 1 summarizes the total of 14 RIPs and their distributions. Here, ‘GAUSS’ denotes the Gaussian (Normal) distribution with a mean (\( \mu \)) and a standard deviation (\( \sigma \)) where \( \sigma = \mu \times COV \) and \( COV \) is the coefficient of variation. In general, the statistical characteristics are obtained from the extensive data collection and data analysis. In the present study, the mean values of the RIPs are taken from the deterministic analysis and the standard deviations are estimated based on the engineering intuition and common available data from the literature [18, 30]. The first three RIPs (process parameters) are more controllable than the material properties, i.e. RIPs between 4-10\(^{th}\) (Table 1), and hence the \( COVs \) of the first three RIPs are selected lower than the \( COVs \) of the RIPs between 4-10\(^{th}\). The last four RIPs are related with the resin kinetic parameters which are obtained from a curve fitting procedure of the DSC data where the deviation from the fitting may exist [31, 32] (i.e. 0.01 is used for \( COV \) in this work). The centreline degree of cure at the exit (CDOCE) is taken as the random output response since it directly affects the expected mechanical properties of the product as well as the possibility of the defects.

Two different probabilistic case studies (Case-1 and Case-2) with each having two sub-cases are performed and the summary of these case studies is given in Table 2. The total number of DOEs used in the RSM is calculated from the relation given in Eq. 11. In Eq. 11, \( f \) is specified as 3 and 6 for Case-1 \((n = 10)\) and Case-2 \((n = 14)\), respectively as reported in ANSYS [24]. As a result, 149 sample points for Case-1 and 285 sample points for Case-2 are obtained from the expression presented in Eq. 11 for the CCD design in the RSM. The details of the case studies are explained in the following.

a) Case-1: Only the first 10 RIPs are used for the MCS (Case-1.1) and the RSM (Case-1.2) in order to see the effect of the variation in the process parameters and the material properties on the variation of the output parameter.
a.1) **Case-1.1**: The MCS having a total of 1,000 simulations (samples) have been performed based on three different MCS options to investigate their effects on the accuracy of the output parameter:

- **Case-1.1a**: “Full MCS”. 1,000 simulations are divided into 1 repetition (cycle) such that all 1,000 samples are initially selected at once by using the LHS technique.
- **Case-1.1b**: “Incremental MCS”. 1,000 simulations are divided into 10 repetitions such that the 1,000 simulations are performed in 100 simulations with 10 repetitions. This adds more randomness to the LHS procedure.
- **Case-1.1c**: “Adaptive MCS”. 1,000 simulations are divided into 1 repetition with the adaptive stopping criterion option where the MCS is terminated before the total simulations are done if the convergence criterion is met. This states that the change in the mean and standard deviation of the random output parameter should be lower than or equal to 0.001 in the consecutive MCS iterations.

a.2) **Case-1.2**: The RSM is utilized where 149 DOEs (sample points) are required for generating the response surface (quadratic polynomial) for 10 RIPs by using the CCD design. In addition to the DOEs, 10,000 Monte-Carlo runs are performed for exploiting the response surface which take only about a couple of seconds.

b) **Case-2**: All the RIPs in Table 1 (total of 14) are used for the MCS (Case-2.1) and the RSM (Case-2.2) in order to investigate the effect of the variation in the resin kinetic parameters as well as the process parameters and material properties on the variation in the output parameter.

b.1) **Case-2.1**: The MCS with LHS having a total of 1,000 samples have been performed according to the results of Case-1.1 based on the best MCS option found so far.

b.2) **Case-2.2**: The RSM is utilized where 285 DOEs plus 10,000 MCS runs are performed for 14 RIPs using the CCD.

5.3. Results and Discussions

5.3.1. Case-1

In order to get a statistical significance (to have a more general conclusion), a total of 10 separate MCS runs, i.e. 10×1,000 simulations, are performed for Case-1.1 (i.e. for each option: Case-1.1a (Full MCS), Case-1.1b (Incremental MCS) and Case-1.1c (Adaptive MCS)). The 10,000 sequential runs require about 80 h using a 2.3 GHz quad-core processor. The results and the discussions of Case-1 are explained in the following:

i. Table 3 shows the mean values of the linear correlation coefficients between the RIPs and the corresponding output parameter (the CDOCE) for 10 separate runs. It is seen that the mean values of the correlation coefficients are very close to each other for the three options of Case-1.1 which indicates that the MCS converged for 1,000 samples.
ii. However, the standard deviations of the 10 runs for these three options differ as seen in Table 4 as a percentage (%) of the mean values given in Table 3. The “Incremental MCS” option has the lowest magnitude of standard deviation for the 6 RIPs out of 10 such as \(u, V_f, \rho_f, C_{pf} \) and \(C_{pr} \). This shows that the “Incremental MCS” is more accurate than the “Full MCS” and the “Adaptive MCS” since the LHS produces more random and diverse solutions in the “Incremental MCS”.

iii. The overall mean of the first three rows in Table 3 (correlation coefficients) is given as “MEAN” at the last row of Table 3. The same correlation coefficients, i.e. “MEAN”, are seen as a bar plot in Fig. 4 (right). As seen from this plot, \(\rho_r\) has the highest (and only) positive correlation coefficient \((r_p = 0.587)\) and the rest has a lower negative correlation with the CDOCE. Here, a positive correlation indicates that the input parameter is directly proportional with the output parameter and vice versa. The corresponding sensitivities are also given in the pie chart in Fig. 4 (right) based on the correlation coefficients. The percentage values indicate how sensitive the CDOCE is with respect to the statistical variations in the RIPs.

iv. The CDF of the CDOCE is shown in Fig. 4 (left) for 10 runs of each MCS option described in Case-1.1. It is seen that the CDFs for all runs \((3 \times 10)\) are found to be close to each other.

v. The sampling range of the CDOCE is found to be approximately between 0.823 and 0.869 (i.e. 0.844 in average with 0.7% standard deviation) as seen from the horizontal axis of Fig. 4 (left). The value of the CDF at each point indicates the probability of the CDOCE being less than a certain level. For instance, the probability of the CDOCE being less than 0.835 is approximately 5%, whereas the probability of the CDOCE being greater than 0.852 is around 100 - 90 = 10%.

vi. In Case-1.2, 10,000 Monte-Carlo simulations exploiting the response surface which require 149 DOEs are performed. The CDFs of Case-1.1 (1,000 MCS) and Case-1.2 (RSM - 10,000 MCS) are depicted in Fig. 5 by using the Gauss plot in order to visualize the tails of the distribution better. One of the results out of 10 taken from the “Incremental MCS” option is depicted in Fig. 5 for Case-1.1 (1,000 MCS). It is seen that the MCS results are found to agree with the RSM results in general, however at the tails of the CDFs for the MCS results diverged from the RSM results because of the limited sample size as aforementioned.

5.3.2. Case-2

The MCS with the LHS is performed for 1,000 samples in Case-2.1 based on the “Incremental MCS” option defined in Case-1. On the other hand, in Case-2.2, 10,000 Monte-Carlo runs exploiting the response surfaces which require 285 DOEs are performed. The results and the discussion of Case-2 are explained in the following:

i. The correlation coefficients between the RIPs (total of 14) and the CDOCE are given as a bar plot in Fig. 6 for Case-2.1. It is seen that the \(E \) (activation energy) has the
highest correlation coefficient (negative) and the magnitude is close to 1 ($r_p = 0.964$) indicating that $E$ is strongly correlated (i.e. inversely proportional) with the CDOCE. This agrees with the similar observation obtained in [18] for the RTM process where the effect of the variation in $E$ on the maximum and minimum cure degree was found to be significant. The corresponding sensitivities are depicted as a pie chart in Fig. 6. It is seen that the parameter $E$ covers almost 57% of all the sensitivity distribution and the rest of the RIPs are sorted in a similar manner as in Case-1.

ii. The most sensitive 5 linear correlation coefficients among the RIPs and the CDOCE in Case-2 are given in Table 5. It is seen that the linear correlation coefficients are almost same for the two cases, i.e. Case-2.1 and Case-2.2.

iii. The Gauss plots of the CDFs in Case-2 together with Case-1 are shown in Fig. 7. It is seen that the overall MCS results agree with the RSM results. The sampling range of the CDOCE is found to be approximately between 0.725 and 0.915 in Case-2, which is wider than the range found in Case-1. This shows that the variation in the resin kinetic parameters (especially in $E$) has a more significant effect on the CDOCE as compared to the variation in the process parameters and the material properties.

iv. The mean and the standard deviation of the CDOCE are calculated approximately as 0.843 and 3.3% in Case-2.

6. Conclusions

In the present work a deterministic thermo-chemical simulation for the pultrusion of a composite rod was carried out and the results were found to agree well with the data from the literature [8]. After validating the deterministic model, a probabilistic analysis of the pultrusion process was performed in which two different probabilistic case studies were investigated. For this purpose, the MCS and the RSM which is the first contribution of its kind for the probabilistic modelling of pultrusion were employed. The LHS technique was utilized for the sampling procedure in ANSYS PDS toolbox. The outcomes of this study are summarized as follows:

i. The effects of the MCS options in ANSYS were investigated in Case-1.1 and it was found that the most accurate statistical results were obtained by dividing the total number of simulations (samples) into repetitions, i.e. total of 1,000 simulations were performed in 100 simulations with 10 repetitions using the LHS (here denoted as the “Incremental MCS”).

ii. In both cases, i.e. in Case-1 and Case-2, it was concluded that the overall MCS results (total of 1,000 samples) were almost converged to the results obtained from the RSM. Note that the tails of the probability density function is not considered in this work.

iii. The variation in the density of the resin was found to have the most significant influence (positive correlation) on the CDOCE out of 10 RIPs in Case-1.
iv. On the other hand, 14 RIPs were considered in Case-2 and it was found out that the variation in the activation energy (E) of the resin has a very strong correlation (negative) with the CDOCE. This shows that the variation in the resin kinetic parameters (especially in E) has a more significant effect on the CDOCE as compared to the variation in the process parameters and the material properties. Hence, it is very important to characterize the resin system correctly while using E together with the resin density.

7. Acknowledgements

This work is a part of DeepWind project which has been granted by the European Commission (EC) under the FP7 program platform Future Emerging Technology.

References

Figure 1: Schematic view of a pultrusion process.

Figure 2: Schematic view of the pultrusion model of the composite rod and the corresponding boundary conditions (left). Material properties of the composite rod (right).

<table>
<thead>
<tr>
<th>Material</th>
<th>$\rho$ (kg/m$^3$)</th>
<th>$C_p$ (J/kg K)</th>
<th>$k_z$ (W/m K)</th>
<th>$k_r$ (W/m K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Epoxy Resin</td>
<td>1260</td>
<td>1255</td>
<td>0.2</td>
<td>0.2</td>
</tr>
<tr>
<td>Graphite</td>
<td>1790</td>
<td>712</td>
<td>66.0</td>
<td>11.6</td>
</tr>
<tr>
<td>Lumped ($V_f = 62.2%$)</td>
<td>1589.66</td>
<td>874.69</td>
<td>0.6628</td>
<td>0.6416</td>
</tr>
</tbody>
</table>
Figure 3: Predicted temperature (top) and degree of cure profiles (bottom) at the centerline of the composite rod.
Figure 4: The CDFs of each option in Case-1 for 10 separate runs (left). The linear correlation coefficients (mean values of the three options in Case-1.1 for 10 runs) between the RIPs and the CDOCE in bar plot and corresponding sensitivities in pie chart (right).

Figure 5: The CDFs of Case-1.1 (1,000 MCS with LHS) and Case-1.2 (RSM + 10,000 MCS) in the form of a Gauss plot.
Figure 6: The linear correlation coefficients between the RIs and the CDOCE in bar plot and corresponding sensitivities in pie chart for Case-2.1.

Figure 7: The Gauss plots of the CDFs calculated in Case-1 and Case-2.
Table 1: Statistical characteristics of the random input parameters (RIPs) for the pultrusion process.

<table>
<thead>
<tr>
<th>Nr.</th>
<th>Parameter</th>
<th>Symbol</th>
<th>Unit</th>
<th>µ</th>
<th>COV</th>
<th>Distribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Pulling speed</td>
<td>( u )</td>
<td>cm/min</td>
<td>30</td>
<td>0.02</td>
<td>GAUSS</td>
</tr>
<tr>
<td>2</td>
<td>Fiber volume fraction</td>
<td>( V_f )</td>
<td>-</td>
<td>0.622</td>
<td>0.02</td>
<td>GAUSS</td>
</tr>
<tr>
<td>3</td>
<td>Inlet temperature</td>
<td>( T_{in} )</td>
<td>°C</td>
<td>38</td>
<td>0.02</td>
<td>GAUSS</td>
</tr>
<tr>
<td>4</td>
<td>Density of resin</td>
<td>( \rho_r )</td>
<td>kg/m^3</td>
<td>1260</td>
<td>0.05</td>
<td>GAUSS</td>
</tr>
<tr>
<td>5</td>
<td>Density of fiber</td>
<td>( \rho_f )</td>
<td>kg/m^3</td>
<td>1790</td>
<td>0.05</td>
<td>GAUSS</td>
</tr>
<tr>
<td>6</td>
<td>Specific heat of resin</td>
<td>( C_{pr} )</td>
<td>J/kg-K</td>
<td>1255</td>
<td>0.05</td>
<td>GAUSS</td>
</tr>
<tr>
<td>7</td>
<td>Specific heat of fiber</td>
<td>( C_{pf} )</td>
<td>J/kg-K</td>
<td>712</td>
<td>0.05</td>
<td>GAUSS</td>
</tr>
<tr>
<td>8</td>
<td>Thermal conductivity of resin</td>
<td>( k_r )</td>
<td>W/m-K</td>
<td>0.2</td>
<td>0.05</td>
<td>GAUSS</td>
</tr>
<tr>
<td>9</td>
<td>Thermal conductivity of fiber in ( r )-axis</td>
<td>( k_{r_f} )</td>
<td>W/m-K</td>
<td>11.6</td>
<td>0.05</td>
<td>GAUSS</td>
</tr>
<tr>
<td>10</td>
<td>Thermal conductivity of fiber in ( z )-axis</td>
<td>( k_{z_f} )</td>
<td>W/m-K</td>
<td>66.0</td>
<td>0.05</td>
<td>GAUSS</td>
</tr>
<tr>
<td>11</td>
<td>Total heat of reaction</td>
<td>( H_{tr} )</td>
<td>J/kg</td>
<td>323</td>
<td>0.01</td>
<td>GAUSS</td>
</tr>
<tr>
<td>12</td>
<td>Pre-exponential constant</td>
<td>( K_o )</td>
<td>1/s</td>
<td>191</td>
<td>0.01</td>
<td>GAUSS</td>
</tr>
<tr>
<td>13</td>
<td>Activation energy</td>
<td>( E )</td>
<td>J/mol</td>
<td>60</td>
<td>0.01</td>
<td>GAUSS</td>
</tr>
<tr>
<td>14</td>
<td>Order of reaction</td>
<td>( n )</td>
<td>-</td>
<td>1.69</td>
<td>0.01</td>
<td>GAUSS</td>
</tr>
</tbody>
</table>

Table 2: Summary of the probabilistic case studies.

<table>
<thead>
<tr>
<th>Analysis Type</th>
<th>Number of RIPs</th>
<th>Number of MCS loops</th>
<th>Number of DOEs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case-1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Case-1.1 (MCS)</td>
<td>10</td>
<td>1,000</td>
<td>-</td>
</tr>
<tr>
<td>Case-1.2 (RSM)</td>
<td>10</td>
<td>10,000</td>
<td>149</td>
</tr>
<tr>
<td>Case-2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Case-2.1 (MCS)</td>
<td>14</td>
<td>1,000</td>
<td>-</td>
</tr>
<tr>
<td>Case-2.2 (RSM)</td>
<td>14</td>
<td>10,000</td>
<td>285</td>
</tr>
</tbody>
</table>

Table 3: The mean values of the correlation coefficients between the RIPs and the random output parameter (centerline degree of cure at the exit (CDOCE)) for 10 runs based on each MCS options (Case-1.1a “Full MCS”, Case-1.1b “Incremental MCS” and Case-1.1c “Adaptive MCS”) in Case-1.

<table>
<thead>
<tr>
<th>( u )</th>
<th>( V_f )</th>
<th>( (k_z)_f )</th>
<th>( (k_{r_z})_f )</th>
<th>( k_r )</th>
<th>( \rho_f )</th>
<th>( \rho_r )</th>
<th>( C_{pf} )</th>
<th>( C_{pr} )</th>
<th>( T_{in} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case-1.1a</td>
<td>-0.470</td>
<td>-0.491</td>
<td>-0.018</td>
<td>-0.018</td>
<td>-0.283</td>
<td>-0.284</td>
<td>0.590</td>
<td>-0.102</td>
<td>-0.086</td>
</tr>
<tr>
<td>Case-1.1b</td>
<td>-0.470</td>
<td>-0.502</td>
<td>-0.009</td>
<td>-0.020</td>
<td>-0.273</td>
<td>-0.295</td>
<td>0.594</td>
<td>-0.108</td>
<td>-0.080</td>
</tr>
<tr>
<td>Case-1.1c</td>
<td>-0.468</td>
<td>-0.496</td>
<td>-0.008</td>
<td>-0.011</td>
<td>-0.260</td>
<td>-0.307</td>
<td>0.578</td>
<td>-0.105</td>
<td>-0.087</td>
</tr>
<tr>
<td>MEAN</td>
<td>-0.470</td>
<td>-0.496</td>
<td>-0.012</td>
<td>-0.016</td>
<td>-0.272</td>
<td>-0.296</td>
<td>0.587</td>
<td>-0.105</td>
<td>-0.085</td>
</tr>
</tbody>
</table>

Table 4: The standard deviations of the correlation coefficients between the RIPs and the random output parameter (centerline degree of cure at the exit (CDOCE)) for 10 runs based on each MCS options of Case-1. The standard deviation values are given in percentage (%) of the mean values given in Table 3.

<table>
<thead>
<tr>
<th>( u )</th>
<th>( V_f )</th>
<th>( (k_z)_f )</th>
<th>( (k_{r_z})_f )</th>
<th>( k_r )</th>
<th>( \rho_f )</th>
<th>( \rho_r )</th>
<th>( C_{pf} )</th>
<th>( C_{pr} )</th>
<th>( T_{in} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case-1.1a</td>
<td>4.34</td>
<td>4.46</td>
<td>151.81</td>
<td>234.90</td>
<td>-11.93</td>
<td>10.34</td>
<td>3.30</td>
<td>-28.24</td>
<td>-39.25</td>
</tr>
<tr>
<td>Case-1.1b</td>
<td>3.17</td>
<td>1.49</td>
<td>301.09</td>
<td>214.99</td>
<td>-11.74</td>
<td>3.48</td>
<td>1.83</td>
<td>-25.45</td>
<td>31.83</td>
</tr>
<tr>
<td>Case-1.1c</td>
<td>6.28</td>
<td>4.01</td>
<td>479.34</td>
<td>206.85</td>
<td>-9.18</td>
<td>6.79</td>
<td>3.19</td>
<td>-37.10</td>
<td>51.67</td>
</tr>
</tbody>
</table>

Table 5: The most sensitive 5 linear correlation coefficients between the RIPs and the CDOCE in Case-2.

<table>
<thead>
<tr>
<th>( E )</th>
<th>( \rho_r )</th>
<th>( n )</th>
<th>( V_f )</th>
<th>( u )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case-2.1 (MCS)</td>
<td>-0.964</td>
<td>0.107</td>
<td>-0.114</td>
<td>-0.087</td>
</tr>
<tr>
<td>Case-2.2 (RSM)</td>
<td>-0.962</td>
<td>0.130</td>
<td>-0.111</td>
<td>-0.097</td>
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</table>