“Implementing Composite Materials In Additive Manufacturing.”
Preface and Legal Notice

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Kongens Lyngby, Denmark
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Abstract (English)

The presented research work contains studies concerning fiber-reinforcement in additive manufacturing. The overall process chain was considered and investigated during the project, including materials, design, production, quality control, and simulation.

A series of experimental and numerical investigations were carried out to determine the influence of short fibers on the additive manufacturing process chain. These investigations were conducted on multiple additive manufacturing machines, injection molding machines, and scientific testing equipment. Methods were developed to optimize and enhance the required properties of the material and the final parts in terms of thermal properties, mechanical properties, lifetime, life cycle, and implementation in a digital production environment.

Descriptive research was performed on the manufacturing process of parts with fiber-reinforcement focusing on fiber-matrix interface, fiber orientation, and fiber distribution within the part. A specialization was considered for vat photopolymerization technologies. Conclusions were drawn from the investigations and included in the development of injection molding inserts as an industrial application.

The digital manufacturing process chains of injection molding machines with a special focus on injection molding inserts were investigated further and challenges were identified. Among them were a reduced lifetime, increased molding cycle time, and thermal management of the insert. Those challenges were tackled in further investigations resulting in a significantly enhanced lifetime and a cycle time reduction.

Further aspects of digital manufacturing were considered in conceptional investigations regarding advanced cooling of the inserts and opening the possibility for automated injection molding prototyping and design phases. The later will play a significant role in an industry 4.0 manufacturing environment.

In comparison to the vat photopolymerization process, investigations were performed on extrusion-based methods elaborating on the differences and enhanced challenges as compared to resin-based manufacturing technologies.

**Keywords:** additive manufacturing technologies, fiber-reinforced polymers, injection molding, numerical simulations.
Dette Ph.D. projekt indeholder undersøgelser vedrørende fiberforstærkning af additiv fremstilling. Under projektet blev den overordnede proceskæde overvejet og undersøgt, herunder materialer, design, produktion, kvalitetskontrol og stimulering.

En række eksperimentelle og numeriske undersøgelser blev udført for at bestemme indflydelsen af fibre på additiv fremstillingsproceskæden. Disse undersøgelser blev udført på flere additiv fremstillingsmaskiner, sprøjtestøbemaskiner og videnskabeligt testudstyr. Metoder blev udviklet for at optimere og forbedre materialets påkrævede egenskaber og de endelige dele med henblik på termiske egenskaber, mekaniske egenskaber, levetid, livscyklus og implementering i et digitalt produktionsmiljø.

Beskrivende forskning blev udført på fremstillsingsprocessen af dele med fiberforstærkning med fokus på fiber-matrixinterface, fiberorientoering og fiberfordeling inden for delen. En specialisering blev overvejet for vatfotopolymeriseringsteknologier. Konklusionerne fra undersøgelserne blev inkluderet i udviklingen af sprøjtestøbningsindsatser som industriel anvendelse.


Yderligere aspekter af digital fremstilling blev overvejet i konceptuelle undersøgelser vedrørende fiberforstærkning vedrørende avanceret afkøling af indsatsen og en åbning af muligheden for automatiserede sprøjtestøbnings prototyping og designfaser. Det sidstnævnte vil spille en væsentlig rolle i et industri 4.0 fremstillingsmiljø.

I sammenligning med vatfotopolymerisationen blev undersøgelser udført på ekstruderingsbaserede metoder, der uddybede forskellene og udvidede udfordringer sammenlignet med harpiksbaserede fremstillingsteknologier.

**Nøgleordene:** additive fremstilling teknologier, fiber-forstærket kunststoffer, sprøde teknologi, numerik simuleringer.
Zusammenfassung (German)


Die digitalen Fertigungsprozessketten von Spritzgussmaschinen mit besonderem Fokus auf Spritzgusseinsätze wurden weiter untersucht und Herausforderungen identifiziert. Darunter waren eine verkürzte Lebensdauer, eine erhöhte Formzykluszeit und ein thermisches Management des Einsatzes. Diese Herausforderungen wurden in weiteren Untersuchungen angegangen, was zu einer deutlich verbesserten Lebensdauer und einer Verkürzung der Zykluszeit führte.

Weitere Aspekte der digitalen Fertigung wurden in konzeptionellen Untersuchungen zur erweiterten Kühlung der Einsätze und zur Ermöglichung automatisierter Spritzguss-Prototyping- und Design-Phasen betrachtet. Letzteres wird in einer Produktionsumgebung der Industrie 4.0 eine wichtige Rolle spielen.

Im Vergleich zur Vat-Photopolymerisation wurden Untersuchungen an extrusionsbasierten Verfahren durchgeführt, die Unterschiede und weitere Herausforderungen im Vergleich zu harzbasierten Fertigungstechnologien wurden erläutert.

Stichwörter: Additive Fertigungstechnologie, Faserverstärkte Kunststoffe, Spritzguss, Numerische Simulation.
I State of the Art

1 Introduction
   1.1 Background ........................................ 6
   1.2 Project Objectives and Scientific Methods ............. 8
   1.3 Project Contents and Research Questions ................ 10

2 Implementation of Fiber-Reinforcement in Additive Manufacturing 13
   2.1 Materials for the Implementation of Fiber-reinforcement ... 13
      2.1.1 Fiber Material ................................... 13
      2.1.2 Matrix Material ................................. 15
      2.1.3 Thermoset Photopolymer Composition ............... 16
   2.2 Technologies for the Implementation of Fiber-reinforcement 18
      2.2.1 Fiber-reinforcement in ME Manufacturing .......... 18
      2.2.2 Fiber-reinforcement in VPP Manufacturing ........... 23
      2.2.3 Fiber-reinforcement in other AM Technologies ......... 26
      2.2.4 Development of Mechanical Properties ............ 28
   2.3 Applications of Fiber-reinforcement in AM ............... 33
      2.3.1 Big Area Additive Manufacturing ................... 33
      2.3.2 Applications in Biomedical Engineering ............. 37
III Industrial Application: Fiber-Reinforced Injection Molding Inserts

6 Lifetime and Surface Topography Development
   6.1 Introduction .................................. 88
   6.1.1 Concept ...................................... 88
   6.1.2 Insert Generation .............................. 89
   6.1.3 Molding and Industrial Application ................. 90
   6.2 Methods ......................................... 90
   6.3 Results .......................................... 92
   6.3.1 Lifetime and Cracks ............................ 92
   6.3.2 Surface Texture and Roughness ..................... 96
   6.4 Summary ......................................... 99

7 Process Simulations of Thermal Mold Behavior
   7.1 Introduction .................................. 102
   7.2 Methods ......................................... 102
   7.3 Results .......................................... 104
   7.4 Case Study Example: Industrial and Topology Optimization Mold . 109
   7.5 Summary ......................................... 111

8 Internal Fiber Structure and Crack Propagation
   8.1 Introduction .................................. 113
   8.2 Methods ......................................... 114
   8.3 Results .......................................... 116
   8.3.1 Bottom-up Printing ............................. 116
   8.3.2 Uniform Fiber Orientation ....................... 121
   8.4 Summary ......................................... 124

9 Life Cycle Analysis
   9.1 Introduction .................................. 128
   9.2 Methods ......................................... 131
   9.2.1 Life Cycle Assessment .......................... 132
   9.2.2 Material Inventory ............................. 133
   9.3 Results .......................................... 134
   9.3.1 Environmental Performance ...................... 138
   9.3.2 Potential Break-even Between the Alternatives ......... 141
   9.3.3 Relevance in Production Context .................. 143
   9.4 Summary ......................................... 143
Contents

IV Conclusion 145

10 Summary and Outlook 147
  10.1 Result Summary ........................................... 147
  10.2 Personal Reflection ........................................ 150
  10.3 Recommendations for Further Development ............... 151

List of Publications 153

Bibliography 157

Acknowledgments 183

V Appendix 185

A Supplementary information to chapter 2 187

B Supplementary information to section 2.2 191

C Supplementary information to chapter 3 193

D Supplementary information to chapter 4 197
  D.1 Domain Drawings of the Moving Mesh Domains ............... 197
  D.2 Modified Build Plate Design ............................... 200

E Supplementary information to chapter 5 203
  E.1 Construction Drawings for the Tape Placement Assembly .... 203
  E.2 COTS Materials for the Tape Placement Assembly .......... 212
  E.3 Construction Drawings for the Joule Heating Extruder ...... 216

F Supplementary information to chapter 7 231
  F.1 Construction Drawings of Insert Designs .................... 231
  F.2 Construction Drawings for Standard Cooling ................. 240
  F.3 Construction Drawings for Para-Conformal Cooling .......... 245
  F.4 Construction Drawings for Multi-Scale, Para-Conformal Cooling . . 250
  F.5 Construction Drawings for Multi-Scale, Para-Conformal Enhanced Cooling .................................. 256
List of Figures

1.1 Distribution of the use of AM systems extracted from data provided in [24] in 2018. Numbers are given in percent of the total field. 7

1.2 Design, process, and material represent the three aspects of the AM triangle that cannot be separated. 8

1.3 Structure of the thesis. 11

2.1 Tensile strength increase in relation to the fiber weight content [2, 15, 16, 39, 48, 52, 56, 62, 63, 69, 70, 74, 77, 85, 95, 98, 99, 102, 113, 122, 128, 129, 139, 161–163]. This figure was published earlier in modified form in [1, 3, 164]. 29

2.2 Young’s modulus increase in relation to the fiber weight content [2, 15, 16, 39, 52, 56, 62, 69, 70, 74, 77, 85, 95, 98, 102, 110, 122, 128, 161–163]. This figure was published earlier in modified form in [1, 3, 164]. 30

2.3 Comparison of photopolymers with and without CFs to injection molded ABS of the same form. (1) ABS injected at 250 bar, (2) 300 bar, (3) 350 bar, (4) 400 bar, (5) 450 bar, (6) HTM 140v2 5%_wet, (7) HTM 140v2 10%_wet, (8) HTM 140v2 2000 flashes, (9) E-Tool 2000 flashes, (10) Nature resin, (11) RCP 30 8000 flashes. This figure was published earlier in modified form in [3]. 31

3.1 Process scheme for bottom-up and top-down VPP. The build plate is moved according to the arrows and the photopolymer is exposed from the direction of the arrow by a digital projection unit or a laser in a rastered manner. 47

3.2 Front (left) and back (right) side of the molding cast made from PTFE and Al plate. 48

3.3 Fiber distribution at top view (left) within the photopolymer and fibers at the border (right) for 5%_wet fiber content. This figure was published earlier in [108]. 49

3.4 Rough cut through the layered structure under the SEM for 5%_wet fiber content. This figure was published earlier in [108]. 50

3.5 Fiber distribution and orientation in the middle of the printed part (left) and at an inner corner (right) showing an even fiber distribution among the layers for 5%_wet fiber content. This figure was published earlier in [108]. 51
List of Figures

3.6 Detailed view of clustered fibers (left) and single fiber (right) with gap and crack for 5\%_wt fiber content. This figure was published earlier in [1, 108]. .................................................. 52

3.7 Examples of fiber-matrix interface defects and cracks around fibers in a thermoset photopolymer matrix cured in an unordered manner (top). Improved fiber-matrix interface of the same manufacturing configuration at a different curing temperature (bottom). ........ 52

3.8 Tensile strength at break (top) and Young’s modulus (bottom) determined according to DIN EN ISO 527-2BB at different curing temperatures. Errorbars are calculated from the standard deviation among 7 specimen per sample. This figure was published earlier in modified form in [4, 232]. ........................................ 54

4.1 Design of experiment (DOE) and design thinking of the numerical simulations in order to predict and improve the fiber orientation within the final product (left). Modified build plate design in order to allow a vertical fluid flow pattern (right). This figure was published earlier in modified form in [4, 242]. ........................................ 60

4.2 Domain of liquid resin for a rectangular build plate (left) and circular build plate (right). Bottom-up and top-down printing processes were considered by flipping the domain by 180°. This figure was published earlier in modified form in [4, 242]. ................... 61

4.3 Velocity in mm/s of a bottom-up domain (top) and velocity arrows in the three dimensional case (bottom). Note the irregular arrow directions around the corners of the build plate. Both images were taken after 2 s of simulation when the flow was fully developed. This figure was published earlier in modified form in [4, 242]. ........ 63

4.4 Stream lines colored according to the fluid velocity of a rectangular (top) and circular (bottom) build plate in bottom-up printing. Note the vortices around the corner of the domain at the bottom of the build plate. This figure was published earlier in modified form in [4, 242]. .................. 64

4.5 Stream lines colored according to the fluid velocity of a rectangular (top) and circular (bottom) build plate in top-down printing. Note the vortices around the corner of the domain at the bottom of the build plate. This figure was published earlier in modified form in [4, 242]. .................. 66

4.6 Stream lines colored according to the fluid velocity of a modified, generally rectangular build plate showing a significant improvement in the mixture of the fiber orientation in the building area (top). Velocity volume arrows of a modified, generally rectangular build plate (bottom). This figure was published earlier in modified form in [4, 232, 242]. ........ 67
4.7 Vertical fluid velocity fraction over the build plate velocity. First row: rectangular build plate top-down on top of the resin (left) and 2 mm over the build plate (right). Second row: modified build plate top-down on top of the resin (left) and 2 mm over the build plate (right). Third row: modified build plate top-down on top of the build plate (left) and on the bottom of the build plate (right). Forth row: 2.5 mm radius holes in modified build plate top-down on top of the resin (left) and 1 mm radius holes in modified build plate top-down on top of the resin (right). This figure was published earlier in modified form in [4, 232]. ........................................ 69

5.1 First (left) and second (right) layer geometry of the printed dog bone. This figure was published earlier in [164]. .................. 73

5.2 Fiber orientation and interface to the PLA in a polished cut (left). Detail view of the interface between the fibers and the PLA matrix in a polished cut (right). This figure was published earlier in [164]. ...... 74

5.3 Holes in the PLA matrix in a polished cut; variations in amount and size of holes at the different layers are visible (left). Details of holes in the PLA matrix in a polished cut in a macro scale (right). This figure was published earlier in [164]. ................................. 75

5.4 Fiber orientation according to the print direction on a fracture surface after a tensile test; orientation of the fibers in the direction of the extrusion. This figure was published earlier in [164]. ............... 76

5.5 Holes in the PLA matrix on a fracture surface after a tensile test; variations in amount and size of holes at the different layers are visible (left). Detail of holes in the PLA matrix on a fracture surface after a tensile test; variations in orientation and size of holes at the different layers (right). This figure was published earlier in [3, 164]. .... 76

5.6 GF in a nylon matrix after a pull test experiment (left). Weak interface between printed layers at 45° printing orientation (right). This figure was published earlier in [3]. .......................... 77

5.7 Compiled tensile test data according to ASTM D638 IV with Young’s modulus (EMod) and tensile strength (TS) with 12%wt and 30%wt in 0°, 45°, and 90° printing orientation. If not noted differently, the experiments were performed on nylon material. .................. 77

5.8 Representative tensile test examples according to ASTM D638 IV with 12%wt and 30%wt in 0°, 45°, and 90° printing orientation. Further experiments were performed but are not shown individually. ........ 78

5.9 Freely extruded filament (left), transversal (middle), and medial lateral (right) cut under CT investigation. This figure was published earlier in [2, 164]. ........................................ 79

5.10 Principle sketch of the tape placement roller system. .................. 81

5.11 Prototype and full assembly of the tape placement roller system. ... 81

xix
5.12 Prototype (left) and simulation (right) of the full extruder utilizing both Joule pre-heating as well as conventional heating on the bottom. 83

6.1 Development phases of IM inserts. a) and b) represent sub-phases and refinements of the development phase. This figure was published earlier in modified form in [4, 232, 259, 260]. 90

6.2 Graphical representation of the molding cycle. The cooling times before and after ejection of the part can be considered the largest potential for cycle time reduction (left). Final part shot at too low injection pressure in the configuration shown in Figure 6.3 (right). This figure was published earlier in [259, 260]. 91

6.3 Single insert in the IM machine before the first shot (left) and PE-LD parts arranged in the IM machine (right). This figure was published earlier in modified form in [2, 137, 178, 259, 260, 266, 267] as well as by other authors with and without citation. 91

6.4 Fibers standing out of the produced layers under SEM investigation confirming the findings presented in subsection 6.1.2. The image was taken before the first shot. The raster pattern on the surface results from the VPP process and the projector resolution. This figure was published earlier in [178]. 93

6.5 Crack propagation velocity over the number of shots (averaged). This figure was published earlier in modified form in [178]. 94

6.6 Three exemplary cracks in the insert after shot 50, 200, 500, 1000, 1500 and 2000. This figure was published earlier in modified form in [1, 178]. 95

6.7 Surface cracks in the insert after 2658 shots under an SEM, the debris result from PE-LD stuck in the cracks (left). Insert surface after 2658 shots showing crack origin at edges as well as parts of the protected surface without cracks (right). This figure was published earlier in modified form in [3, 178]. 96

6.8 Part surface and rounded edges after 2658 shots (left). Detail of surface degradation with PE-LD material stuck in the crack (right). This figure was published earlier in modified form in [178]. 97

6.9 Graphical representation of the surface topography after 10 shots showing the profile under a focal variation 3D-microscope in the middle of the insert. This figure was published earlier in modified form in [178]. 97

6.10 Averaged roughness propagation during the first 1000 shots neglecting bigger cracks in the surface. Errorbars are calculated from the standard deviation among 7 specimen per sample. This figure was published earlier in modified form in [1, 3, 178]. 98

7.1 Thermal material properties of the simulated materials in comparison. 104
7.2 Temperature of one mold side. The right figure shows transparent features in order to see the cooling system. This figure was published earlier in [232, 259, 260]. ................................................. 105

7.3 Temperature of conventional (left) and para-conformal (right) cooling channels at a throughput of 5 L/min. Due to the improved flowability of the fluid in the channels with rounded corners, a higher throughput of up to 10 L/min is possible. This figure was published earlier in [4, 232, 260]. .................................................. 106

7.4 Cooling of the surface of the insert with standard cooling. Ejection of the molded part appeared after 8.5 s (top). Cooling of the surface of the insert with para-conformal cooling. Ejection of the molded part appeared after 8.5 s. The improved cooling allows for a shorter molding cycle as the temperature of the PhP insert reaches lower values more quickly (bottom). This figure was published earlier in [4, 259]. .......................................................... 107

7.5 Difference to standard cooling of PhP inserts (top). Difference to standard cooling of brass inserts (bottom). This figure was published earlier in [4, 259]. .................................................. 108

7.6 Graphical representation of the insert with active liquid cooling inside the insert. .......................................................... 110

7.7 Back and front of the industrial application insert with temperature sensor mounted in the back of the insert. .................................................. 110

7.8 Concept drawing of a topology optimized insert with cavity. Blue represents the cooling liquid, grey represents the shell and topology optimized insert material, white represents the cut through cavity. ... 111

8.1 IM insert after twice the usual lifetime (left). Measurement positions for the CT analysis (right). This figure was published earlier in [137]. 115

8.2 Fiber orientation analysis for position 1, 2, and 3. As suggested in [2, 272], fibers are oriented along the printed layers. This figure was published earlier in modified form in [4, 137, 232]. 117

8.3 Cut through position 2. Cracks are mainly oriented in between the printed layers. This figure was published earlier in [137]. 118

8.4 Overview on deeper cracks in terms of orientation and dimensions through position 2, which align in general with the printing layer orientation. This figure was published earlier in [137]. 119

8.5 Projected view with surface crack orientation (left). Surface cracks originate from the steep temperature gradients during the IM process. Cracks are mainly oriented within the printed layers. Detailed view on deeper cracks through position 1 of the middle picture (right), which align in general with the printing layer orientation. This figure was published earlier in [4, 137, 232]. 120
8.6 Fiber extrapolation (left) and exemplary crack orientation and dimension measurements (right) in position 1 for smaller surface cracks. Projection view of deeper cracks in position 1 (right). This figure was published earlier in [137].

8.7 Sub volume 1 (left) and sub volume 2 (right) with numerically extrapolated pores. This figure was published earlier in [137].

8.8 Sub volume and fiber extraction with more isotropic fiber orientation.

8.9 Fiber orientation analysis for the middle and top position. The parts were printed with the modified build plate design. Orientation within the printing plane is shown on the left. Orientation orthogonal to the printing plane is shown on the right. This figure was published earlier in modified form in [4].

9.1 Combined flow chart of the two assessed systems of inserts; AM and CNC. All inputs and outputs around the insert production and insert EoL are included all the way from/back to earth. Note: Indicated by grey text and a dashed line, the actual IM process is excluded from the direct comparisons as it is the same for both systems. This figure was published earlier in [232].

9.2 Cumulative GWP impact neglecting the polymer waste. This figure was published earlier in modified form in [266, 267].

9.3 Cumulative HH impact neglecting the polymer waste. This figure was published earlier in modified form in [266, 267].

9.4 Ratio of GWP impact of ABS waste material as compared to the total impact on the LCA. This figure was published earlier in modified form in [266, 267].

9.5 Ratio of GWP impact of PE-LD waste material as compared to the total impact on the LCA. This figure was published earlier in modified form in [266, 267].

9.6 Ratio of HH impact of ABS waste material as compared to the total impact on the LCA. This figure was published earlier in modified form in [266, 267].

9.7 Ratio of HH impact of PE-LD waste material as compared to the total impact on the LCA. This figure was published earlier in modified form in [266, 267].

9.8 GWP of AM inserts and brass inserts in an application with up to 4500 shots, requiring 1 AM insert and 1 brass insert. Contributing life cycle processes are indicated by color. (The top-four processes in the nomenclature relate to the brass insert, all below relate to AM insert.)

9.9 HH impact of AM inserts and brass inserts in an application with up to 4500 shots, requiring 1 AM insert and 1 brass insert. Contributing life cycle processes are indicated by color. (The top-four processes in the nomenclature relate to the brass insert, all below relate to AM insert.)
9.10 GWPs (top) and HH impact (bottom) of inserts over number of shots, for the range of 1000 to 100,000 shots. ..................142

10.1 Summary of the investigated areas and development steps during the project. This figure was published earlier in modified form in [4]. . . . 148
List of Tables

2.1 General formulation of the photopolymer used in [88] whereas the percentages can be varied slightly in order to change the material properties accordingly. .......................... 17
2.2 Chemicals used in the formulation of the photopolymer used in [89], Percentages were not given. Materials marked with * were used as resin. 17
2.3 Chemicals used in the formulation of the photopolymer used in [90], Percentages were not given. .......................... 17
2.4 Matrix and fiber materials from various scientific contributions. . . . . 18
2.5 Overview of available FRP technologies in AM. .......................... 32
4.1 Simulation domain parameters. ......................... 61
7.1 Thermal material properties of the simulated materials in comparison. 103
8.1 Measurement descriptions for positions according to Figure 8.1 (right). 115
9.1 Weight of 1 insert of the different kinds. .................. 131
9.2 Assumed lifetime of 1 insert of the different kinds for injection of PE-LD. 131
9.3 Weight of 4 produced parts and residual material of the IM process that was not used in the final parts for 1 shot. .................. 132
9.4 Four different maximum numbers of shots and the related numbers of required inserts, in AM (VPP 10\%) and brass respectively. 133
9.5 LCA data for 1 kg of the part materials. .................. 134
9.6 LCA data for 1 kg of the insert materials. .................. 134
9.7 Ratios of waste material as compared to the total of GWP and HH impacts for 1 shot. .......................... 134
Nomenclature

List of Acronyms

3D      three dimensional
3DP     polyjet 3D printing
ABS     acrylonitrile butadiene styrene
AM      additive manufacturing
ASTM    American Society for Testing and Materials
BAAM    big area additive manufacturing
CAD     computer aided design
CF      carbon fiber
CFD     computational fluid dynamics
CFRP    carbon fiber-reinforced polymer
CMC     ceramic matrix composites
CNC     computer numerical control
CNF     carbon nano fibers
CNT     carbon nanotube
COTS    commercial off-the-shelf (products)
CT      computed tomography
DALY    disability adjusted life year
DB      dichlorobenzene
DCM     dichloromethane
Nomenclature

DGEBA  bisphenol A diglycidyl ether
DICY  dicyandiamide
DIN  Deutsches Institut für Normung
DIW  direct ink writing
DLP  digital light processing
DMAc  N,N-dimethylacetamide
DMLS  direct metal laser sintering
DOE  design of experiment
DOF  degree of freedom
DTU  Technical University of Denmark (Danmarks Teknisk Universitet)
ELCD  European Life Cycle Data
EMod  Young’s modulus
EN  European Norm
EoL  end-of-life
F1  Formula 1
FDM  fused deposition modeling
FEAM  fiber encapsulation additive manufacturing
FLM  fused layer manufacturing
FRP  fiber-reinforced polymer
GF  glass fiber
GO  graphene oxide
GWP  global warming potential
HH  human health
IM  injection molding
Nomenclature

IP    intellectual property
IR    infrared
ISO   International Organization for Standardization
LCA   life cycle assessment
LDM   liquid deposition manufacturing
LiCl  lithium chloride
LOM   laminated object manufacturing
MCC   microcrystalline cellulose
ME    material extrusion
MIT   Massachusetts Institute of Technology
MMA   methylmethacrylate
MRI   magnetic resonance imaging
MSDS  material safety data sheets
mSLA  mask stereolithography
MWCNT multi-walled CNT
NASA  National Aeronautic and Space Administration
NMR   nuclear magnetic resonance
ORNL  Oak Ridge National Laboratories
PA    polyamide
PA12  polyamide-12
PA6   polyamide-6
PAN   polyacrylonitrile fiber fillers
PAN-sc polyacrylonitrile shortcut
PC    polycarbonate
Nomenclature

PCL polycaprolactone
PEEK polyetheretherketone
PEI polyethylenimine
PEKK polyetherkethonekethone
PhP photopolymer
PLA polylactide
PMMA polymethyl methacrylate
PP polypropylene
PPS polyphenylene sulfide
PPSF polyphenylsulfone
PPSU polyphenylsulfone
PS polystyrene
PU polyurethane
RF radio frequency
ROMM randomly-oriented multi-material
RP rapid prototyping
RTM resin transfer molding
SEM scanning electron microscope
SI Système International d’Unités
SiC silicon carbide
SLA stereolithography
SLM selective laser melting
SLS selective laser sintering
SME small and medium enterprise
Nomenclature

SMP  shape memory polymer
SOP  standard operating procedure
SOTA state of the art
TEOS tetraethyl orthosilicate
TS   tensile strength
UV   ultraviolet
UV-3D ultraviolet-assisted 3D
VARTM vacuum assisted resin transfer molding
VGCF vapor-grown carbon fiber
VPP  vat photopolymerization
WHAM wide and high additive manufacturing
ZnO  zinc oxide

List of Mathematical Symbols

\%_{wt} \quad \text{percentage referring to the weight fraction} \\
\chi_1 \quad \text{orientation-efficiency factor} \\
\chi_2 \quad \text{fiber-length correction factor} \\
F \quad \text{external forces} \quad \text{N} \\
g \quad \text{gravitation} \quad \text{m/s}^2 \\
I \quad \text{identity matrix} \\
u \quad \text{velocity vector} \quad \text{m/s} \\
\mu \quad \text{viscosity} \quad \text{m}^2/\text{s} \\
\nu \quad \text{Poisson number} \\
\Phi \quad \text{azimuth angle of fiber orientation} \\
\Phi_f \quad \text{volume fraction of the fiber}
Nomenclature

\( \Phi_m \) volume fraction of the matrix

\( \rho \) density \( \text{kg/m}^3 \)

\( \Theta \) elevation of fiber orientation

\( a \) dimensionless length

\( buildX \) build plate length/diameter \( \text{mm} \)

\( buildZ \) build plate height \( \text{mm} \)

\( c_P \) heat capacity \( \text{J/kg}^\circ\text{C} \)

\( d \) diameter of the fiber \( \text{mm} \)

\( E_c \) tensile modulus of the composite \( \text{MPa} \)

\( E_f \) tensile modulus of the fiber \( \text{MPa} \)

\( E_m \) tensile modulus of the matrix \( \text{MPa} \)

\( k \) thermal conductivity \( \text{W/m}^\circ\text{C} \)

\( l \) length of the fiber \( \text{mm} \)

\( n \) dimensionless length

\( p \) pressure \( \text{N/m}^2 \)

\( q_V \) heat source \( \text{W} \)

\( R \) dimensionless length

\( Sa \) area surface roughness \( \mu\text{m} \)

\( T \) temperature \( ^\circ\text{C} \)

\( t \) time \( \text{s} \)

\( T_g \) glass-transition temperature \( ^\circ\text{C} \)

\( vatX \) resin vat length/diameter \( \text{mm} \)

\( vatZ \) resin vat height \( \text{mm} \)
Scientific Contributors

The following people contributed scientifically to this thesis. The list stands in alphabetic order sorted by surname.

Iñaki Arrieta Azanza contributed to the research on printing of carbon nano tubes as a master thesis student.

Sina Baier contributed to the research on crack propagation by computed tomography measurements.

Niki Bey contributed to the research on life cycle analysis.

Gregory Dreifus contributed to the research on fast fused filament fabrication during the external stay at MIT.

Jamison Go contributed to the research on fast fused filament fabrication during the external stay at MIT.

Carsten Gundlach contributed to the research on crack propagation by computed tomography measurements.

Ingomar Gutmann contributed to the research on fiber-reinforced thermoplastic material fiber orientation.

Hans Nørgaard Hansen contributed to all topics serving as a supervisor.

A. John Hart contributed to the research on fiber-matrix interface and fast fused filament fabrication serving as a supervisor during the external stay at MIT.

Ralf Jagenteufel contributed to the research on printing of polypropylene.

Florian Kamleitner contributed to the research on printing of polypropylene.

Thomas Koch contributed to the research on fiber orientation in thermoplastic material extrusion.

Andreas Lunzer contributed to the research of thermal mold simulations by generating parts of the mold geometry as a bachelor thesis student.

Michael Mischkot contributed to the research on in-situ testing of additively manufactured injection molding inserts and life cycle analysis.

David Bue Pedersen contributed to all topics serving as a supervisor.
Abhinav Rao contributed to the research on fiber-matrix interface during the external stay at MIT.

Jon Spangenberg contributed to the research on fiber orientation by supervising fluid flow simulations.

Adam Stevens contributed to the research on fast fused filament fabrication during the external stay at MIT.

Philippe Maurice Stotz contributed to the research on life cycle analysis.

Guido Tosello contributed to all topics serving as a supervisor.

Camilla Himmelstrup Trinderup contributed to the research on fiber orientation with a data analyzing tool.

This thesis has been evaluated by an assessment committee consisting of the following professors.

Leonardo De Chiffre served as chairman. He is professor at the Department of Mechanical Engineering of the Technical University of Denmark.

Jürgen Fleischer is professor at the wbk Institute of Production Science of the Karlsruhe Institute of Technology and Director of Machines, Equipment and Process Automation.

Henning Zeidler is professor at the Institute for Machine Elements, Engineering Design and Manufacturing of the TU Bergakademie Freiberg.

I would like to express my sincere gratitude to all contributors for their valuable input to this project and would like to thank them for their time and collaboration.
Part I

State of the Art
Additive manufacturing technologies have received both academic and industrial attention in recent years for their use in multiple materials such as metals, ceramics, and polymers. This part represents a review to analyze the technology of fiber-reinforced polymers and its implementation with additive manufacturing. Recent developments, ideas, and state-of-the-art technologies in this field are reviewed and an overview of the materials currently available for fiber-reinforced material technology is given.

The content of this part was taken and in parts modified from the following publications:


1 Introduction

Fiber-reinforced polymers (FRPs) find applications in multiple industries such as aerospace, automotive, wind energy, production industry, and biomedical engineering. They are based on highly strain-resistant fibers embedded into a softer polymer matrix. Recently, they were also utilized in additive manufacturing (AM).

This can, on the one hand, take place in an ordered manner using directional fiber placement within the matrix, and on the other hand, in a non-ordered manner. The aim of the fibers is to strengthen the material in terms of load, whereas the aim of the polymer matrix is to protect the fibers and to distribute the load among them [5–7]. It is concluded in [6] that functional applications are limited if the methodical implementation is not performed carefully. In [7], the authors conclude a shift from traditional materials in composites towards functionalized nano-structures.

A comparison of the different technologies and their mechanical potential is given in [8, 9] showing a similar modulus to strength ratio as compared to conventionally manufactured FRPs, but at lower moduli and strength rates. Characteristics are the high potential of complex geometries at low performance as compared to conventional manufacturing. This leaves the potential for high performance carbon printing. In contrast, it was claimed in [9] that FRPs from AM performed better than conventionally manufactured parts.

The directional fiber placement will result in anisotropic mechanical properties whether placed in an ordered or non-ordered manner, whereas the stress and strain resistance will increase only in the direction of the fibers.

The materials need to interact at their interface to distribute stress. Moreover, they need to be chemically compatible. Microcracks close to the interface between fibers and matrix lead to a significant loss of stiffness and strength.\footnote{This aspect has been researched and discussed in detail and is presented in subsection 3.3.3.} For this reason, it is mentioned in [10] that a focus is currently put on non safety-critical applications. In [11, 12], self-healing functionality is discussed eliminating damages automatically. Recent developments in composite manufacturing, including AM systems have been discussed in [13] concluding that some processes are not developed to an extent to make it interesting for industrial applications and they are too costly to replace conventional materials in current process chains.
Advantages of fiber-reinforcement were summarized in [14] as:

- better out-of-plane properties for multi-directional preforms and for printing spatially oriented fibers,
- broad structural designability,
- improved structural integrity and damage tolerance, and
- cost-effectiveness.

It was pointed out in [15–17] that fiber-reinforcement comes with advantages in terms of the distortion of the object during the printing process. It was concluded that fiber-reinforcement can achieve increased strength, stiffness, thermal conductivity, and can reduce part distortion when using milled fibers. Similar effects were detected for nano composites in [16]. This allows larger objects to be printed.

Nano composites in AM were reviewed in [18, 19] concluding a specialized applicability of nano composites for flexible microelectronics, sensing devices, micro antennas and tissue engineering. Material properties in terms of newly developed composites made manufacturable by AM with focus on design were reviewed in [20] concluding new possibilities also in products and production paradigms currently developed from the top down and the bottom up. Another review was performed in [21] concluding a need for optimization methods in extrusion-based AM. Mechanical properties and manufacturers were reviewed in [13, 22, 23], which provide an overview over different technologies for fiber composites. The general availability of fibers in AM was as usual also reviewed by the annual Wohler’s report [24], which provides a reference for general purposes.

Embedding fibers in AM not only challenges in terms of fiber-matrix interface, chemical composition, and manufacturing, but also the design phase as pointed out in [25]. It was reviewed in [26] that fibers make a part not only stronger, but also come with difficulties connected with the manufacturing process in all areas. Some of the challenges and opportunities of modern manufacturing techniques in the range of AM were reviewed in [27] with a focus on automated manufacturing concluding the ability to reduce cycle times and waste production. Nevertheless, interfacial properties need to be understood better, which is also researched in this thesis in particular in subsection 3.3.3.

1.1 Background

According to [28], already in 2014, industrial companies using AM were utilizing it for either prototyping (24 %) or product development (16 %) followed by innovation (11 %), increased efficiency (10 %), and cost reduction (9 %).
1.1 Background

Recent investigations presented in the annual Wohler’s report 2018 [24] confirmed a distribution of the use of AM systems as shown in Figure 1.1 reporting furthermore a significant annual increase of system sales valued at more than 5000 $. This thesis contributes to the field of research, patterns for prototyping tooling, and tooling components and therefore covers 25.5% of the range of AM systems though fiber composites represent a much smaller niche.

The strategic aspects were analyzed in [29] and summarized as speed and implementation of new technical concepts in the manufacturing process, whereas indirect influences in product development in fields such as molding and castings were analyzed in contrast to direct manufacturing technologies. Further advantages were named as freedom of design, production of traditionally not manufacturable products, variation of mass products, personalization of mass products, realization of new manufacturing strategies, and realization of new materials.

In order to ensure the functionality and reliability of manufactured parts, they can be adapted by manufacturing a combination of a soft polymer matrix with a high-performance fiber.

**Figure 1.1:** Distribution of the use of AM systems extracted from data provided in [24] in 2018. Numbers are given in percent of the total field.
Fiber-reinforcement of polymers leads to modified mechanical properties and opens the possibility for a range of applications in the field of biomedical engineering, automotive, aerospace, wind energy and others ranging from small and high-precision applications to large-scale precision applications. Given the large number of conventional applications, the improvement potential in terms of AM technologies in this field is high.

1.2 Project Objectives and Scientific Methods

It was the ambition of this project to develop systems and applications for FRPs and polymer composites based on AM technologies considering mechanical properties as well as issues of surface quality to ensure proper functionality. In this context, three aspects are crucial to incorporate in any research and development activity: design, process, and material shown in Figure 1.2. Each factor is dependent on the other and can therefore not be changed without changing the other aspects as well. While the focus of research in fiber-reinforcement of AM technologies might rise the preliminary impression of work based on the material aspect, the topic must be seen in a broader perspective in order to understand the full potential of the topic.

An additional focus has been put on applications in two categories:
1. Applications with small dimensions (in the μm- and mm-range) and the need for high-precision mechanical and surface properties (in the range of 10 to 100 μm tolerance and surface roughness $S_a$ in the range of 0.1 to 1.0 μm).

2. Applications with larger dimensions (in the mm- and cm-range) facing multiple mechanical challenges such as moments, pressure and tension.

The research focused on the establishment of novel technologies for the generation and deposition of FRPs via AM technologies.

The new possibilities have led to a range of novel applications in the fields mentioned above. The applications and manufacturing mechanisms developed during the Ph.D. project have been tested and validated under industrial conditions targeting the current and future challenges of the application.

Investigations have been performed in terms of applications and possibilities emerging from the newly developed manufacturing technologies that could not be performed before.

The first efforts were made to develop fiber-reinforced injection molding (IM) inserts produced by vat photopolymerization (VPP) showing an increased lifetime over non-fiber-reinforced inserts produced with the same technology. The inserts were among others evaluated in terms of surface roughness, fiber distribution and lifetime.

Investigations on the fiber orientation in a part produced by material extrusion (ME) were performed using scanning electron microscopy as well as computer tomography.

It is the objective to improve upon the partly existing process chain of fiber-reinforced AM starting at the part conceptualization, manufacturing characteristics, material properties, up to the final industrial application. Challenges regarding materials for both fiber and matrix are faced especially in the interface between the two materials making it difficult to manufacture.

New concepts are presented to enhance the strength of the part, modify it according to the mechanical and thermal requirements in the application, and enhance the crucial parameters for the final industrial application. In the particular case of this thesis, lifetime, surface accuracy, and thermal behavior have been identified as critical factors that were improved upon.

The research is limited to VPP and ME but the content presented in this thesis does not cover large applications in the m-range. The terms according to ISO/ASTM 52900:2015 [30] are further described in [31] and were used throughout the thesis.
1 Introduction

1.3 Project Contents and Research Questions

The project was structured according to Figure 1.3 and divided into the following parts:

**State-of-the-art of fiber-reinforced polymers**  The technology and methods for the fabrication with FRPs in the context of AM require an understanding of both technology groups: composite manufacturing, and additive manufacturing. The combination provides solutions that can be expected to be placed in the range between typically cheap polymer AM, and rather expensive metal AM allowing to close this gap and combine the advantages of both technology groups.

Research on this combination is limited and incremental material properties such as mechanical strength, stiffness, and thermal properties such as conductivity and heat capacity are defined by the materials used in the composite manufacturing. Furthermore, materials are strongly connected to the manufacturing processes and the design of the final part. Hence, they define the possible manufacturing processes and applications.

A fundamental understanding of all included technologies and their combinations is key to improve upon the state of the art. Investigations regarding these topics can be found in Part I. The “material” aspect of the AM triangle (see Figure 1.2) is covered in this part.

**Guiding Question:** To which extent are fiber composite materials exploited within the state-of-the-art AM methods?

**Identification and manufacturing of polymers for matrix and fiber content**  The manufacturing process as one of the core parts in the product chain can significantly influence the final part performance. While this is a known and well-investigated factor for non-reinforced AM technologies, further challenges arise when composite materials are added to the process. Factors such as defects, fiber orientation, rheology, and chemical properties have to be taken into account in both a descriptive and predictive manner.

Theoretical and practical research has shown the advantages and disadvantages of both short and continuous fiber-reinforcement in polymer matrices. In the case of polymer AM, thermoplastic and thermoset materials are identified with different manufacturing technologies. Factors such as fiber-matrix interface need to be taken into account especially in an industrial context.

Understanding and development of existing AM processes in both a descriptive and predictive manner is required in order to improve upon existing technologies.
1.3 Project Contents and Research Questions

Figure 1.3: Structure of the thesis.
1 Introduction

Investigations regarding these topics can be found in Part II. The “process” aspect of the AM triangle (see Figure 1.2) is covered in this part.

Guiding Question: To which extent do commercially available FRP-based AM systems exist, and how can they be mapped?

Applications of fiber-reinforced AM in an industrial context The digital technologies and flexible production environment that are present in the current industrial development require a change in paradigm from expensive and time-consuming prototyping and design processes to flexible and cost-effective development cycles. AM has filled the gaps in a number of current manufacturing technologies and is under constant development. An example of this are IM inserts, which adapt and in parts replace expensive steel molds.

The disadvantages of AM inserts such as low lifetimes are the subject of ongoing investigations in both academic and industrial contexts. Fiber-reinforcement has shown improvements of the manufactured parts in terms of mechanical properties and can therefore be considered for the underlying applications followed by further improvement of the applications in terms of size, production cycle, and environmental impact.

The implementation of fiber-reinforcement in an existing industrial application opens new challenges and tackles existing ones. It is strongly connected to the materials and technologies in action. Investigations regarding these topics can be found in Part III. The “design” aspect of the AM triangle (see Figure 1.2) is covered in this part.

Guiding Question: To which extend can the design of the part be influenced in order to achieve improved results?

System validation, application testing and demonstration Validation and improvement are part of every prototyping cycle and are therefore also considered. Introduced technologies and applications require a benchmark in terms of process robustness and quality assurance capability of the developed system in comparison with the state-of-the-art industry standards.

Conclusions have to be drawn following the results of the investigations in order to improve upon them and further develop the underlying application.

Investigations regarding these topics can be found in Part III and are summarized in Part IV.

Guiding Question: So what? To which extend can the thesis contribute to solving scientific and industrial challenges?
2 Implementation of Fiber-Reinforcement in Additive Manufacturing

2.1 Materials for the Implementation of Fiber-reinforcement

Composites built up from a matrix and a fiber material are well-established in industrial processes and applications. While the fiber fraction varies depending on the technology and the desired properties of the final part, it is relatively constrained in additive manufacturing applications. The following chapter gives an overview on the available materials and their implementation in fiber-reinforced additive manufacturing.

2.1.1 Fiber Material

Two groups of sources for fibers can be identified:

- natural fibers, and
- synthetically fabricated fibers.\(^1\)

The first group, natural fibers, can be further separated into fibers from plants and fibers from animals. Cellulose fibers from plants, Kenaf, Sisal, jute, and silk are a widely used example [32–35], as well as harakeke (flax) and hemp used in [36]. However, it was concluded in [32] that the mechanical properties of AM composites are below traditional manufacturing technologies such as compression processes and further optimization of the biocomposite formulation is required for further improvement. Synthetic fibers are fabricated under dry or wet conditions. After production, they are spun into filaments, ropes, or strings as described in [37].

Commonly used fibers for carbon fiber-reinforced polymers (CFRP) are continuous carbon fibers (CFs) described in [38]. They are widely used, e.g. in the Airbus

\[^1\]Mineral fibers such as silica and fibrous dust have been omitted in this thesis.
A350 and Boeing 787 aircrafts, and as components in the automotive industry, wind turbine blades, and endoscopic surgery [8, 39–44].

Carbon nanotube (CNT) composites in a polymer matrix are reviewed in [7] concluding new technologies to manufacture composites with CNTs including mass production technologies elaborating on functional materials including e.g. self-healing mechanisms. CNTs were used in [45–48] for micro structure analysis. It was shown in [45] that CNT-based materials can be used to manufacture strain-sensing platforms at high degrees of flexibility. However, the strength increase presented in [46] does not fully represent the theoretical increase that could be reached in optimal conditions. The reasons for the low performance are described in [47] as porosity due to increased printing speed, specimen shrinkage, raster angle, and increased void size. The influence of the printing conditions was further studied in [48] including the dispersion of CNTs concluding significant challenges with brittleness and realization of the theoretical mechanical performance.

CNTs reach a tensile strength of 100 to 600 GPa and Young’s moduli of 1 to 5 TPa. These properties make it necessary to invest in a good interface between the CNTs and the surrounding matrix. It was shown in [48] that the porosity of the final part is not increased by including 1% and 5% CNTs. The melting temperature, however, was increased.

Research has also been performed on the usage of glass fibers (GF) in combination with AM processes ([10, 49–53]). They will be described further in section 2.2.1. Long GFs allow the transportation of information through the material and therefore also allow communication through the matrix material [54]. Aramid fibers were used in [55] and are further described in section 2.2.3. In comparison to GFs, the Young’s modulus was not increased to the intended level.

Vapor-grown carbon fibers (VGCF) were used in [56] mixed by shear processing and focused on an alignment of the fibers and avoiding fiber breakage. VGCFs come with high stiffness and strength as well as low weight. Other advantages are the electrical and thermal properties of 50 μΩ cm and 2000 W/mK. Experiments performed in [57] on a CF filled material extrusion printer (as will be described in subsection 2.2.1) concluded a resistivity of $(0.09 \pm 0.01) \, \Omega/m$ within the printed layer and $(0.12 \pm 0.01) \, \Omega/m$ orthogonal to the printed layers. The measurements were taken out on a 5 mm cube of a commercial formulation of polycaprolactone (PCL) matrix. Young’s modulus is located at high values of 360 to 600 GPa [58–61].

However, an increase of electrical resistivity after the printing process was concluded in [62] using CNTs in an acrylonitrile butadiene styrene (ABS) matrix. The electrical conductivity strongly depends on the fiber material varying also between CFs as concluded in [63] reporting furthermore the usability of the printed parts in sensors.

A range of natural fibers is described in [64]. A characterization of silicon carbide (SiC) fibers for applications influenced by high temperatures is given in [65]. PCL is a fiber material used in melt electrospinning writing in hydrogel and is useful for
higher weight fractions of filler material as reviewed in [19, 66, 67]. A conclusion
drawn in [19] is also based on the availability of printers in terms of manufacturing
parameters, compatibility of printer and resin formulations, and printer resolutions.
Parameter studies concerning e.g. temperature and other ambient parameters as
concluded in [67] together with the machine parameters according to [66] can enable
multiple applications in microfluidics, robotics, and bionics.

Thermal conductivity was enhanced in [68] using copper fibers in an extrusion-based
AM system. The aim was to produce complicated shapes for heat exchangers saving a
significant amount of energy during production and during run time. It was also noted
that 20\% in volume of copper fibers were equipped with twice the through-plane
conductivity of spherical fillers.

2.1.2 Matrix Material

A wide range of polymer matrix materials is available in conventional manufacturing
processes, including polypropylene (PP), polystyrene (PS), and polyurethane (PU).
Currently, three technologies are available in literature to produce raw composite
material. They are solution casting, in-situ polymerization, and melt compounding
[36, 69, 70]. Other technologies used in industry to manufacture composite parts
are hand lay-up, spray lay-up, filament winding, resin transfer molding (RTM), and
pultrusion, which are summarized in [71] and in Appendix A.

Also commonly used as matrix material are epoxy, polyester, and acrylate blends
featuring a high surface quality in terms of roughness and a good connection to GF
material [10].

In terms of AM, the range of employed polymers is currently limited. It includes but is
not exclusive to examples of the application of ABS, polycarbonate (PC), polylactide
(PLA), polyamide (PA), polyamide-6 (PA6), tetraethyl orthosilicate (TEOS), nylon,
polyethylenimine (PEI), polyphenylene sulfide (PPS), and polyphenylsulfone (PPSU)
[39, 40, 56, 72–79]. High performance polymers are not very common in AM. In
[48, 80] PEI of the polyetheretherkethone (PEEK) and polyetherketoneketone
(PEKK) family was used as a matrix material. A main advantage is the high glass-
transition temperature of over 200°C. It was proposed in [80] to use fiber coatings
to optimize the fiber-matrix interface. A model for temperature ranges and flow
parameters in the manufacturing process of PEEK was reported in [81]. A model
with polymethyl methacrylate (PMMA) powder, which was later melted around the
fibers, was implemented in [82], whereas a low density of the parts was concluded.

In [37], PP was discussed as matrix material in detail, naming the main advantages of
PP such as high heat-distortion temperature, flame resistance, dimensional stability,
and suitability for filling, reinforcing, and blending. A recycled product of PP was
used in [36] for embedding natural fibers resulting in increased tensile strength of
30\% and Young’s modulus of 143\% using 30\%wt hemp and harakeke (flax) fibers.
PLA was used as a matrix material for microcrystalline cellulose (MCC) whiskers in [69, 83, 84]. It was concluded in [69] that MCC can be used as starting material. The remaining crystallites of the cellulose whiskers after swelling and separation was confirmed. The thermally stable region was concluded to be between 25 to 220°C according to [83]. After a swelling process involving N,N-dimethylacetamide (DMAc) containing lithium chloride (LiCl), the whiskers were mixed with a PLA melt during the extrusion process forming an all biodegradable composite material with increased strength by 20% and Young’s modulus by 10% at a fiber content of 5%\textsubscript{wt}. Wood flour was used in a PLA matrix in [85] in order to generate an extrudable filament. Thermal degradation was slightly decreased while the melting temperature stayed constant before and after adding the filler material.

Although PLA is widely used in AM, especially in fused layer manufacturing (FLM)/ME, few applications of this composite in actual components are available in literature. ME is also known under the commercial trademark fused deposition modeling (FDM), which is commonly known but slowly replaced by standardized terms used throughout this thesis. The production of the feedstock itself was named as key challenge in [86].

Bioactive glass, calcium phosphate, hydrogel, calcium phosphate, paper-based substrate, and glass ceramics are other rarely used matrix materials for specific applications reviewed in [19].

### 2.1.3 Thermoset Photopolymer Composition

The literature shows a dominance of acrylic-based photopolymers such as urethane acrylic [52] as well as epoxy-based materials (see also [71, 87] and Appendix A).

Most technologies use ultraviolet (UV) radiation or blue light (with higher wavelength than UV) as curing mechanism. Light intensity as well as curing time per layer height in technologies like vat photopolymerization (VPP) such as digital light processing (DLP) or stereolithography (SLA) are significantly influenced by the polymer composition. The reason is light absorption by the additives and fibers. A reduction of maximum cured thickness of 52% at 15 s UV exposure by adding 2% of CFs was found in [88, 89]. In [88], the composition presented in Table 2.1 was used where 2%\textsubscript{wt} CFs were added during the curing process.

The photopolymerization system of methylmethacrylate (MMA) forming a thermoset material combined to a composite with GFs was extensively described in [89], whereas the materials noted in Table 2.2 were used. Another formulation was given in [90] with a configuration shown in Table 2.3.

GF and CF are the most used fiber materials for AM composite manufacturing exemplary shown in Table 2.4 whereas it can be noted that the chemical interaction between GFs and photopolymer materials is significantly stronger than with CFs.
2.1 Materials for the Implementation of Fiber-reinforcement

<table>
<thead>
<tr>
<th>Chemical</th>
<th>Producer</th>
<th>%wt</th>
</tr>
</thead>
<tbody>
<tr>
<td>Genomer2252/GP25</td>
<td>Rahn</td>
<td>35</td>
</tr>
<tr>
<td>Genomer1117</td>
<td>Rahn</td>
<td>55</td>
</tr>
<tr>
<td>Miramer M300</td>
<td>Rahn</td>
<td>10</td>
</tr>
<tr>
<td>Irgacure 819</td>
<td>BASF</td>
<td>1.5</td>
</tr>
<tr>
<td>ITX</td>
<td>SIGMA</td>
<td>0.5</td>
</tr>
<tr>
<td>4-methoxyphenol</td>
<td>SIGMA</td>
<td>0.05</td>
</tr>
</tbody>
</table>

**Table 2.1:** General formulation of the photopolymer used in [88] whereas the percentages can be varied slightly in order to change the material properties accordingly.

<table>
<thead>
<tr>
<th>Chemical</th>
<th>Distributor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isopropylthioxanthone (ITX)</td>
<td>Sigma-Aldrich</td>
</tr>
<tr>
<td>Ethyl 4-(dimethylamino)benzoate (EDB)</td>
<td>Sigma-Aldrich</td>
</tr>
<tr>
<td>Tert-Butylhydroperoxide (THB)</td>
<td>Sigma-Aldrich</td>
</tr>
<tr>
<td>Cobalt(II)-2-Ethylhexanoate (CO\textsuperscript{II})</td>
<td>Sigma-Aldrich</td>
</tr>
<tr>
<td>2-(4-Methoxyphenyl)-4,6-bis(trichloromethyl)-1,3,5-triazine (TA)</td>
<td>PCAS</td>
</tr>
<tr>
<td>Cobalt(II) 2-ethylhexanoate in solvent mixture (NL51P)</td>
<td>AkzoNobel</td>
</tr>
<tr>
<td>Diphenyl(2,4,6-trimethylbenzoyl)-phosphine oxide (TPO)</td>
<td>BASF</td>
</tr>
<tr>
<td>Peroxan ME 60-L</td>
<td>Pergan</td>
</tr>
<tr>
<td>Methylmethacrylate (MMA)*</td>
<td>Sigma-Aldrich</td>
</tr>
<tr>
<td>Ebecryl 270 (Eb270)*</td>
<td>Crytec</td>
</tr>
<tr>
<td>Tripylene glycol diacrylate (TPGDA)*</td>
<td>Sartomer</td>
</tr>
<tr>
<td>Crestapol 1250*</td>
<td>Scott-Bader</td>
</tr>
</tbody>
</table>

**Table 2.2:** Chemicals used in the formulation of the photopolymer used in [89]. Percentages were not given. Materials marked with * were used as resin.

<table>
<thead>
<tr>
<th>Chemical</th>
</tr>
</thead>
<tbody>
<tr>
<td>1,2,4,5-benzenetetracarboxylic dianhydride (PMDA)</td>
</tr>
<tr>
<td>2-phenyl-(4,4'-diaminodiphenyl ether) (p-ODA)</td>
</tr>
<tr>
<td>4-phenylethynylphthalic anhydride</td>
</tr>
</tbody>
</table>

**Table 2.3:** Chemicals used in the formulation of the photopolymer used in [90]. Percentages were not given.
### Table 2.4: Matrix and fiber materials from various scientific contributions.

<table>
<thead>
<tr>
<th>Matrix</th>
<th>Fiber</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>(multiple)</td>
<td>carbon black</td>
<td>[91] (Review)</td>
</tr>
<tr>
<td>De Solite SCR310 (urethane acrylic)</td>
<td>glass</td>
<td>[52]</td>
</tr>
<tr>
<td>CibaTool SL5170 (epoxy-based)</td>
<td>glass</td>
<td>[87]</td>
</tr>
<tr>
<td>RIG including Irgacure 819</td>
<td>carbon</td>
<td>[88]</td>
</tr>
<tr>
<td>Somos 3100</td>
<td>glass</td>
<td>[92]</td>
</tr>
<tr>
<td>Ciba Geigy XB5081</td>
<td>glass</td>
<td>[92]</td>
</tr>
<tr>
<td>Methylmethacrylate (MMA)</td>
<td>glass</td>
<td>[89]</td>
</tr>
<tr>
<td>Ebecryl 270 (Eb270)</td>
<td>glass</td>
<td>[89]</td>
</tr>
<tr>
<td>Tripylene glycol diacrylate (TPGDA)</td>
<td>glass</td>
<td>[89]</td>
</tr>
<tr>
<td>Crestapol 1250</td>
<td>glass</td>
<td>[89]</td>
</tr>
<tr>
<td>Methylmethacrylate (MMA)</td>
<td>vinylon</td>
<td>[93]</td>
</tr>
<tr>
<td>Assymmetric Imide oligomer</td>
<td>carbon</td>
<td>[90]</td>
</tr>
</tbody>
</table>

### 2.2 Technologies for the Implementation of Fiber-reinforcement

The term “additive manufacturing” is used for a number of technologies such as material extrusion where the part is build up by layers, which are manufactured by extrusion of filaments placed in a certain manner to generate a layer. Additive manufacturing, however, is the umbrella term for a number of technologies comprising multiple materials from both the matrix and the fiber part. Not all of the technologies use polymers, but rather metals or ceramics. This chapter focuses entirely on the technologies to implement fiber-reinforcement.

Technologies and materials significantly influence properties such as mechanical strength, stiffness, thermal conductivity, ... and are synthesized at the end of the chapter.

#### 2.2.1 Fiber-reinforcement in ME Manufacturing

**Short Fibers** ME manufacturing using CF in ABS is widely available and can result in increased tensile strength of 42 MPa at 5\%_w_t fiber content as shown in [39]. At a fiber content of 10\%_w_t, the tensile strength was 34 MPa, which is close to the tensile strength of pure ABS (depending on the composition of the ABS). It was shown in [77] that especially with short CFs in the sub-mm length range, the fiber length has a significant influence on the tensile strength of the final part – as well has the fiber diameter, cross-section shape, and orientation according to a review in [94]. Glass-transition temperature $T_g$ and melting temperature of the ABS matrix were only effected of single-digit percentages by the increased fiber content.
Young’s modulus was largest at 7.5\%_{wt} fiber content with a value of 2.5 GPa. Toughness and yield strength decreased when adding fibers to pure ABS. The same is given for ductility. Experiments were performed with fiber lengths of 150 μm resulting in a higher Young’s modulus than fibers with 100 μm length. It was concluded in [39, 40] that a pure plastic specimen compared to added CF into plastic materials could lead to an increased tensile strength and Young’s modulus, but also to a decreased toughness, yield strength, and ductility.

An orientation of 91.5\% of the embedded CFs was concluded to reach the optimal mechanical properties in [95], using fibers with the length of 200 to 400 μm. Experiments concluded an increase of tensile strength by approximately 115\% and Young’s modulus by approximately 700\%. This value is the highest among the reviewed literature.

A similar experiment was performed earlier in [56] using VGCF in an ABS matrix. After Banbury mixing, extrusion, and ME manufacturing, limited porosity of the composite and an average tensile strength increasing of 15\% up to a value of 24.4 MPa at the highest level was observed. The absolute values strongly depended on the composition of the ABS polymer.

ME of ABS with short GFs that were showing the possibility of up to a 30\%_{wt} GF content in an ABS matrix were performed in [49, 50]. Higher values require chemical treatment of the ABS using plasticizers and compatibilizers to provide a uniform distribution of the fibers. This allows uniform and determined material parameters throughout the final part.

ME was used in combination with FRPs and achieved an increase of the modulus by 40\% with a weight content of chopped carbon AS4 fibers of 10\%_{wt} in [80]. Moreover, the moisture content was reduced by this fabrication step, which reduced the porosity of the printed objects.

Other experiments using ME in combination with FRPs are presented in [15, 16, 96], including natural wood fibers in [32] investigating the fiber orientation after the ME print. It was pointed out that ME prints include a high porosity of around 20\% leading to damage mechanisms and water absorption, including swelling. An increasing porosity with increasing nozzle width was detected. In [16], one of the earlier publications in this field of research, zero porosity in the parts after processing with Banbury mixing was reached. Similar results about the microstructural characteristic of ME-produced parts were presented in [97] investigating CFs in an ABS matrix. For the sake of uniform reinforcement, the prints were not performed in a layered manner, but with multi-axe, 3D-extrusion technology.

Short jute fibers were investigated in [98, 99]. Compared to [33], the strengthening of the ABS matrix material was lower and located at 9\% for tensile strength in unidirectional tensile testing with 5\%_{wt} fiber content. It was concluded that the fibers located themselves in the middle of the printed layer. This interface was investigated and improved by infrared preheating moderately over glass-transition temperature
in [100]. A method investigated in [101] used single layers of FRP material with CF and a PLA matrix. In tensile tests, the performance of the reinforced specimen was 66% better than the equivalent non-reinforced material.

A similar system using ABS as matrix material for big area additive manufacturing (BAAM) was established in [100, 102, 103] using CFs at 20% wt resulting in a 100% increase in strength and a 380% increase in stiffness and, at 40% wt, resulting in a 380% increase in strength and a 470% increase in stiffness.²

Computational fluid dynamics (CFD) simulations modeling the fiber distribution within an ME nozzle and final product were performed in [104]. Thermal simulations with a focus on continuous CF were performed in [105] concluding a significant influence of the nozzle geometry and an essential steep temperature gradient between the feeding part and the nozzle with heater block as earlier investigated in [106].

A conclusion was drawn in terms of fiber orientation during the final part in strong relation to the printing orientation in the ME process. Due to the two cycles of extrusion of the material

1. when generating the filament from granule; and
2. during the AM process,

the fibers were oriented longitudinally to the printing path. This is supported by simulations conducted in [107] showing a fiber orientation after the exit of the nozzle with consideration of swelling mechanisms.

Ellipsoidal holes with the major axis in the nozzle orientation occurred in the PLA matrix with average sizes of 50 to 100 μm possibly resulting in a reduction of tensile strength. Compared to FRP in VPP, the size of the holes was significantly increased [108]. A similar discovery was made in [109] showing droplets spreading along the fibers. The interface between fiber and matrix material has significant influences on the tensile strength. The distribution of fibers was hence also influenced, whereas the fibers were generally oriented in the middle of the extrusion line [108, 110].³

The interface between the layers was indistinct due to the remelting of material during the extrusion of the next layer, allowing for a continuous matrix material. This also resulted in an interconnected orientation of longitudinally-oriented and orthogonally-oriented fibers.

The interface between the fibers and the PLA matrix was destroyed during a tensile-test. This was fatal at the layers with orthogonal fiber orientation towards the tensile strength. 40% of all fibers in a longitudinal orientation towards the tensile strength were ripped out of the matrix at the fracture surface.

²A detailed review of FRPs in BAAM is presented in subsection 2.3.1.
³This aspect has been discussed further in chapter 5.
A device to align fibers in the ME nozzle was recently patented [111] in 2016, claiming reinforcement of the polymer due to the directional fiber distribution. A modified nozzle design using multiple matrix materials was described in [112]. The nozzle provided the ability to include fibers and increase mechanical properties. The design is claimed to reduce the staircase effect, warp, and gaps as well as selective reinforcement and load-based infill. Gaps and cracks were reported in a number of research contributions between especially CFs and thermoplastic materials such as in [113]. Recent approaches to align fibers in ME manufacturing were reviewed in [114]. The evaluation of available manufacturing systems and binders was concluded to be a crucial step before part manufacturing.

Pseudo-alignment was discussed in [115] using particles which were electrically aligned in order to form fiber-like structures. The system was successfully used with metals, ceramics, glass, CNTs, and aluminum micro-particles in a resin. The particles were aligned using two electrodes on both sides of the curing UV source creating a field aligned in parallel to the surface of the resin.

A different subject was discussed in [116] investigating the effect of printing parameters as well as composite materials such as PLA filled with multi-walled CNTs (MWCNTs) as well as nylon on the adhesion of the print on PLA fabric surfaces. It was concluded that the printing parameters have an increased influence on the adhesion force.

Adhesion between the different printed layers was investigated in [117] reporting findings of an improved layer interface by radio frequency (RF) welding. The interface is molten by the introduced heat transported through the embedded CNTs and results in a strength increase of 275%.

**Continuous/Long Fibers** While the above-mentioned research concerns short fibers, an investigation in [33, 118] concerning continuous, fiber-reinforced, ME technologies using PLA with short CFs or continuous twisted yarns of natural jute fibers are available. It was shown that unidirectional CF-reinforced plastic are equipped with mechanical properties superior to jute-reinforced and unreinforced thermoplastics. An improvement of continuous fiber-reinforcement above conventional AM polymer-based composites in terms of tensile strength was presented.

The jute fibers did not significantly increase the tensile strength, while the CFs increased the tensile strength by 435 to 599%. Investigations using PLA with an embedded continuous fiber were presented in [119] concluding a weak interface between the fiber and the matrix. Flexural strength increased from 13.8 to 164% and storage moduli from 166 to 351% compared to conventional parts. A general overview on the use of continuous fiber in a PLA matrix is given in [120].

Yarn was also used in [121] in a newly developed mechanism to combine the fiber material and the matrix material directly at the extruder for continuous fiber material. This technology allows for high fiber fractions in the fiber material.
Other investigations concerning continuous fiber were presented in [122] with a focus on fiber orientation which is controlled by the fabrication mechanism. Concentric, isotropic, as well as a stacking sequence of alternating nylon and FRP were compared among each other in the context of mechanical properties in tensile tests. The strength increase of the nylon matrix with CF, fiberglass and Kevlar up to a factor of 6.3 was found exceeding the mechanical properties of aluminum. The increased strength was highest for CF-reinforcement and lowest for Kevlar-reinforcement.

Similar to the findings presented in [113, 123], it was found that the fiber-matrix-interface was weakened by an increased fraction of fibers. This can be considered a common problem, which is also known for photopolymers and has been highlighted in [13]. An algorithm-based approach for improved mechanical properties and reduced porosity with continuous fibers was introduced in [124] based on process planning algorithms.

Continuous fused manufacturing was developed by the company MarkForged [125] offering ABS and other matrix materials with Nylon-micro-carbon, carbon, nylon, fiberglass, Kevlar and high-temperature fiberglass as fiber materials. The technology was also presented in [13, 23] producing tensile test specimens with fibers embedded in the central part of the specimen.

This technology has been further developed in [23, 126] adding the continuous CFs with the help of a heated hypodermic needle after the extrusion process. This is hence the third possible implementation of continuous fibers in an extrusion-based system besides fibers embedded in the filament or merged during the extrusion process, which was also developed and presented in [127]. However, the fibers are located in strands embedded in a thermoplastic polymer matrix instead of a distribution of single fibers.

A similar experiment was performed in [128] embedding CNTs in an Ultem® matrix, an amorphous, thermoplastic PEI. Not only mechanical strengthening was shown, but an electrical conductivity was established allowing the final part to function as a sensor.

The research in [129] showed an increase of tensile strength by nearly a factor of 5 when adding 10% wet CF into an ABS matrix. This represents the highest value available in literature. The downside is the low withstanding of shear stress. Strain around holes was in general reduced in [130] when using woven CFs in a part produced by ME technology in comparison to die-punched holes.

Investigations on continuous GF materials with laser-assisted melting of the thermoplastic matrix material was published in [127] reaching increased tensile strength even with bi-directional layer placement. The placement process was conducted in two steps: laser bonding and laser cutting of the final part applying pressure over a roller as well as heat over a laser setup.
Kevlar fiber with a multi-orientation infill in tensile test dog bones were used in [131] for tensile measurements. While fibers aligned with the stress result in higher strength and modulus, the multi-orientation infill provides nearly isotropic mechanical properties.

FRPs in AM were furthermore reviewed in [13] with a focus on extrusion-based manufacturing. The authors rated material utilization and flexibility in part design as increased possibilities of FRP in AM, whereas quality, investment costs, surface quality, and market maturity were rated negatively. Mechanical properties were rated neutral among the other composite manufacturing technologies.

Another thorough review can be found in [31] pointing at potential issues during manufacturing and following with the conclusion of a poorer performance of short fibers in comparison to continuous fibers regardless of the technology.

It can be synthesized that ME materials for both fibers as well as matrix is relatively wide comprising industrially used materials such as ABS as well as high-performance materials such as PEEK. Most research is performed for CF and GF. Manufacturing can be concluded as a challenge to be tackled investigating the wetting of the fibers in order to allow a functioning fiber-matrix interface and therefore reaching the theoretical improvement of mechanical and thermal properties.

An increase of strength and stiffness has proven more efficient for continuous fibers, which requires different manufacturing technologies. Flexibility in terms of geometrical possibilities is higher for short fibers whereas the fiber orientation control can be concluded to be a critical factor and has therefore been investigated in chapter 4.

### 2.2.2 Fiber-reinforcement in VPP Manufacturing

The possibilities of manufacturing three-dimensional printed objects with additives using mask stereolithography (mSLA), also known as DLP, but standardized as VPP, were investigated in [51]. GFs were chosen for the process with a diameter of 10 to 15 μm. A weight concentration of 10%\text{wt} was selected to reach optimal mechanical parameters, but still keep viscosity of the resin in an applicable regime. The experiments described in [51] were unsuccessful, even with a weight concentration of 3 to 5%\text{wt}. It was not possible to mix the resin with the fibers and keep the concentration in an isotropic distribution. From the experiments, it was concluded that the concentration of the fibers needed to meet specific levels to gain proper results.

Experiments using CNTs were performed investigating a UV-curable resin in a mixture with carbon nanotubes of 0.1 to 5%\text{wt} [132]. The experiments were performed using molds filled with the curable resin, which were then cured under UV light for 30 to 120 min. Other results of FRPs in UV-curable resins were presented in [133].
Earlier research was undertaken in [134] using Somos® 7110 epoxy-based resin and layered, loosely-woven, long, fiber material. It demonstrated the possibility of increasing the elastic modulus along the direction of the fibers due to the fact that they created a layered fiber structure.

Short GFs have also been used in literature e.g. in [87]. The fibers were mixed into the resin and were therefore randomly oriented in space, resulting in a uniform reinforcement. Adding 15\% \text{wt}, the authors calculated an increase of the tensile modulus from 1.5 GPa to 2.5 GPa [52, 87] and calculated the modulus by the equation

\[ E_c = \chi_1 \chi_2 \Phi_f E_f + \Phi_m E_m \] (2.1)

where

\begin{align*}
E_c & = \text{tensile modulus of the composite} \\
E_f & = \text{tensile modulus of the fiber} \\
E_m & = \text{tensile modulus of the matrix} \\
\Phi_f & = \text{volume fraction of the fiber} \\
\Phi_m & = \text{volume fraction of the matrix} \\
\chi_1 & = \text{orientation-efficiency factor} \\
\chi_2 & = \text{fiber-length correction factor}
\end{align*}

and

\begin{align*}
\chi_2 & = \frac{l - \tanh (na)}{na} \quad (2.2) \\
n & = \sqrt{\frac{2E_m}{E_f \ln \left( \frac{2R}{d} \right)}} \quad (2.3) \\
\frac{2R}{d} & = \sqrt{\frac{\pi}{4\Phi_f}} \\
a & = \frac{l}{d} \quad (2.4)
\end{align*}

where

\begin{align*}
l & = \text{length of the fiber} \\
d & = \text{diameter of the fiber} \\
a & = \text{dimensionless length} \\
n & = \text{dimensionless length} \\
R & = \text{dimensionless length} \\
\nu & = \text{Poisson number}
\end{align*}
The factor $\chi_1$ depends on the fiber orientation (0.2 for three-dimensionally, randomly oriented and 1 for unidirectional orientation). Experiments using molding technology showed an increase of tensile strength by 64%. Experiments using rapid prototyping (RP) VPP showed an increase of tensile strength by 60%, whereas the level of tensile strength is half the value of the material used for the experiments manufactured by molding [52, 87].

The effect of fibers in the matrix can be signified by a control mechanism for the fibers. This was achieved in [135] using magnetic printing to orient the fibers in the liquid photopolymer resin.

The manufacturing machine used in [87] implemented a combination of ME and VPP feeding the material from a mixing device directly onto the surface where it was cured using UV light. Interlayer rivets were introduced in order to enhance the layer-to-layer bonding.

A different study also using short GFs in an urethane acrylic-based resin cured with a laser system was performed in [52, 53]. It showed an increase of the mechanical properties in dependence on the angle of the fiber. The shrinkage of usually 1 to 6% was reduced by the short GFs. The mechanical properties were directly connected to the fiber concentration, increasing the strength when increasing the fiber content. Moreover, the laser power and lower layer pitches were found to increase the mechanical strength. Shrinkage decreased with increasing fiber content.

Building on this research, other filler material of which carbon can be considered the most similar to FRPs was added in [91]. The authors were able to significantly increase the mechanical properties by adding so-called “carbon black” with particle sizes of 30 nm.

In terms of conventional manufacturing, a review of the flow characteristics of a resin filled with fibers is given in [136]. A special focus was set on the characteristic flow on the corners of objects when coating parts with the resin. It was claimed that residual stresses inducing defects such as cracking and delamination are inevitable. The research according to [136] should go into the direction of microbuckling of fibers, void formation, as well as global layer buckling and resin percolation at corners.

As the VPP process is based on a layer-wise build-up of the manufactured object, fiber orientation within the object is limited by the fiber characteristics and the layer height. Orientation and distribution of fibers were investigated for the first time during this project and presented in Part II as well as in [108, 137]. Other experiments regarding short fibers in VPP, including optical as well as thermal curing, were recently performed at the North Dakota State University of Agriculture and Applied Science using a dual cure system and are presented in [138].
2 Implementation of Fiber-Reinforcement in Additive Manufacturing

2.2.3 Fiber-reinforcement in other AM Technologies

Fiber-reinforcement in Binder Jetting Manufacturing While most references are discussing the implementation of FRP on ME (or a modification of ME) and VPP, in [139] fibers were added to a matrix of cellulose-modified gypsum powder. The authors were using a ZPrinter® 300 by ZCorporation with a layer thickness of 0.1 mm. As fibers, polyacrylonitrile (PAN) fiber fillers, polyacrylonitrile shortcut (PAN-sc) fiber, polyamide (PA) fiber fillers, and alkali resistant zirconium silicate glass shortcut fibers were discussed. Fibers at a content of 1%\textsubscript{wt} and the matrix were mixed before the manufacturing process was started. This led to an anisotropic direction of the fibers within the matrix, which stands in contrast to the experiments using ME. The bending strength was increased by up to 180% while flexural strength was increased by up to 400%. A fiber weight content of over 1.5%\textsubscript{wt} led to a slight decrease. The printing was performed with a fiber content of up to 2.5%\textsubscript{wt}.

A review of available technology was published in [140] suggesting to use a multi-process technology to overcome present challenges such as precision, materials, and functional constraints.

Fiber Encapsulation Additive Manufacturing (FEAM) As a new technology, FEAM, was introduced in [141]. It combines the advantages of ME technology with the reinforcement by long fibers. The fiber is placed onto a surface and covered with a mantle of melted polymer from an ME extruder. The technology allows the equipping of the manufactured object with certain properties of mechanical parameters as well as magnetic properties. It is furthermore possible to generate adaptive surface properties described in [142].

Randomly-oriented Multi-Material (ROMM) using a Polyjet 3D printing (3DP) machine was introduced in [6]. Nevertheless, there have not yet been any known attempts to implement a technology on additively manufacturing FRPs. An experiment was conducted using 3DP with fiberglass or an aramid fiber and a layer thickness of 0.028 mm [55]. It could be shown that Young’s modulus with fiberglass was significantly increased from 2100 MPa of the non-reinforced material to 3700 MPa. While the strength of the non-reinforced object was relatively constant, the strength of the reinforced object varied strongly.

Fiber-reinforcement in Sheet Lamination Manufacturing An attempt to embed fibers into a sheet lamination (previously known as laminated object manufacturing (LOM)) process using continuous fiber ceramic matrix composites (CMCs), and short fiber CMCs was performed in [143]. The layers finally had a thickness of 230 to 260 μm and a relatively high fiber content compared to the ME process as described.
above of approximately 50%\textsubscript{wt}. Other examples were given in [144] concluding the increased strength and reduced weight.

Research on highly-ordered fiber composites was conducted in [73] aiming at the development of military applications in the field of aerospace using biologically-inspired composites with short fibers of around 3 mm in length. GFs with zinc oxide (ZnO) coated by tetraethyl orthosilicate (TEOS) were used resulting in self-healing of the composite material in case of defects. The system was inspired by the fiber-based structures of tendons and their interaction among each other. The synthetic fibers were aligned acoustically with ultrasonic waves. Wool fibers were partly reviewed in [145] as part of a review on material issues in AM.

**Fiber-reinforcement in Powder Bed Fusion Manufacturing** The implementation of composites in a powder bed fusion (previously known as selective laser sintering (SLS)) process generating a metal matrix using graphene oxide (GO) as filling material, enhancing the mechanical properties was presented in [146, 147]. Experiments with CF were performed in [148]. They were sintering carbon nano fibers (CNF) from PA12 with 3%\textsubscript{wt} filler content. It was possible to increase the strength of the object as well as the storage module by 22%. A significantly increased porosity especially at the layer interfaces was reported in [149] using a PA12/CF composite powder bed configuration.

**Further Technologies** Another approach was performed in [82] where carbon and metallic fibers were embedded into a PMMA bed. Tensile strength was thereby increased by 27%.

A novel method of extrusion-based fiber-reinforced printing of epoxy resin was presented in [150] concluding a highly ordered fiber orientation by direct ink writing (DIW). The material is optically (UV) cured after the extrusion and optionally also thermally cured [151]. The technique also allows for multiple filler materials to be printed in an oriented and non-oriented manner.

Direct writing was presented in [152] using nylon fibers in a soft, UV-curable, matrix material. The fibers were electrically aligned before the application on the surface.

Introduced in [153], material jetting (also known as liquid deposition manufacturing (LDM)), combines a polymer matrix material (in this case PLA) with a conductive filler material (in this case CNTs) as well as a solvent (in this case dichloromethane (DCM)). The material is then extruded in an analog way as ME. By this method, also freeform out-of-plane printing becomes possible.

Concerning ultraviolet-assisted 3D (UV-3D) printing, an investigation was performed in [110] of short CFs with a co-formulation of a photocurable acrylic resin with a thermocurable epoxy resin with the raw materials Bisphenol A diglycidyl ether
(DGEBA), 1,1-dimethyl, 3-(3’,4’-dichlorophenyl) urea (Diuron®), dicyandiamide (DICY), and fumed silica.

A similar experiment was performed earlier in [154] furthermore discussing alignment mechanisms for CFs claiming an ultimate strength increase by 44.1% and sample modulus by 42.6%, when the CFs were aligned along the stress orientation.

AM or 3D printing was recently further developed adding time as fourth dimension. Materials for this printing technology were reviewed in [155] and are mainly elastic polymers and gels, but also embed fibers such as CF, wood, cellulose, coconut, and shape memory polymer (SMP) fibers. Research was also performed in [32, 126, 156–160].

2.2.4 Development of Mechanical Properties

As a major aim of FRPs in AM, improved mechanical properties are stated frequently. A literature survey among various papers showed an increase of Young’s modulus as well as tensile strength coming with an increased porosity of the material. A graphical representation of the change in tensile strength and Young’s modulus is given in Figure 2.1 and Figure 2.2. Specific data gained using a Zwick Roell Z005 pull test machine is given in Figure 2.3 comparing different materials that were molded with different pressures, as well as additively manufactured with different fiber contents and different matrix materials. Post-curing was performed using stroboscope flashing. The underlying experiment aimed to understand the behavior of different technologies in terms of mechanical properties.

While the mechanical properties of FRPs are usually improved as compared to the same matrix material without fibers, it was argued in the case of PP matrix material with GF filler in [165] that “simple processing of the polyolefins is compromised, and second, high filler loadings inevitably lead to heavier products”. The authors proposed to use nano clay materials as filler in order to reduce the temperature. The number of flashes in the figure represents the post-processing method utilized in order to cure yet uncured material and strengthen the existing reactions.

A tabular overview on the FRPs already produced by AM is given in Table 2.5. All fiber percentage values are stated in weight content.
Figure 2.1: Tensile strength increase in relation to the fiber weight content [2, 15, 16, 39, 48, 52, 56, 62, 63, 69, 70, 74, 77, 85, 95, 98, 99, 102, 113, 122, 128, 129, 139, 161–163]. This figure was published earlier in modified form in [1, 3, 164].
Figure 2.2: Young’s modulus increase in relation to the fiber weight content [2, 15, 16, 39, 52, 56, 62, 69, 70, 74, 77, 85, 95, 98, 102, 110, 122, 128, 161–163]. This figure was published earlier in modified form in [1, 3, 164].
Figure 2.3: Comparison of photopolymers with and without CFs to injection molded ABS of the same form. (1) ABS injected at 250 bar, (2) 300 bar, (3) 350 bar, (4) 400 bar, (5) 450 bar, (6) HTM 140v2 5\%\text{wt}, (7) HTM 140v2 10\%\text{wt}, (8) HTM 140v2 2000 flashes, (9) E-Tool 2000 flashes, (10) Nature resin, (11) RCP 30 8000 flashes. This figure was published earlier in modified form in [3].
Implementation of Fiber-Reinforcement in Additive Manufacturing

Fibers
- ME
- carbon, CNT/MWCNT, glass, copper, Kevlar, wood, jute, yarn, flax, harakeke, wollastonite, hemp

Matrix
- ABS (and modifications), PLA, PP, PA6, nylon, PC, PEI, Ultem®, PEEK, PEKK, PPS, PPSF, PC, PET, Ultem, ABS (and modifications), PLA

Sources
- increased tensile strength and Young's modulus by 27% at 3% fiber content
- increased tensile strength by 22 to 27% at 3% fiber content
- increased tensile strength up to 50% at up to 15% fiber content

Table 2.5: Overview of available FRP technologies in AM.

<table>
<thead>
<tr>
<th>Technology</th>
<th>Fiber</th>
<th>Matrix</th>
<th>Added value properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>VPP</td>
<td>CNT, glass</td>
<td>photopolymer, epoxy, blends</td>
<td>tensile modulus of 2.5 GPa at 15 % fiber content</td>
</tr>
<tr>
<td>binder jetting</td>
<td>cellulose, PAN, PA</td>
<td>gypsum</td>
<td>tensile strength increased by 180 % at up to 2.5% fiber content</td>
</tr>
<tr>
<td>sheet lamination</td>
<td>carbon, glass</td>
<td>ceramic, ZnO</td>
<td>higher fiber content up to 50 %</td>
</tr>
<tr>
<td>UC</td>
<td>SiC, NiTi</td>
<td>Al</td>
<td>54, 179–185</td>
</tr>
<tr>
<td>powder bed fusion</td>
<td>metal, graphene oxide, carbon</td>
<td>metal, PA12, PNA</td>
<td>tensile strength increased by 22 to 27 % at 3 % fiber content</td>
</tr>
<tr>
<td>direct writing</td>
<td>nylon</td>
<td>uv-curable soft polymer</td>
<td>increased tensile strength and Young's modulus</td>
</tr>
<tr>
<td>LDM</td>
<td>PLA</td>
<td>MWCNT</td>
<td>significante increase in electrical conductivity, integration of conductive features as small as 100 μm</td>
</tr>
</tbody>
</table>

Notes
- increased tensile strength at higher nozzle temperature
- the increase at 7.5% of the control
- Young's modulus is increased

References
2.3 Applications of Fiber-reinforcement in AM

Applications of fiber-reinforced polymers in conventional manufacturing are widely known and often used. Applications for additive manufacturing are currently rare and mostly focus on material extrusion. Advantages such as lightweight and increased strength for multiple applications were pointed out in [144]. Examples are given in the following sections.

2.3.1 Big Area Additive Manufacturing

Introduction An AM technology used for large-scale applications such as wind turbines is known as big area additive manufacturing (BAAM) and is promoted as future manufacturing technology or assisting technology for renewable energy production [186] in the biggest research facility at Oak Ridge National Laboratory (ORNL) where the PEI matrix material, ABS, the PEI-like material Ultem®, polyphenylsulfone (PPSU), polyphenylene sulfide (PPS) with CF as well as GF filler material are used.

The approach towards wind energy applications comes indirectly via mold design, which was later combined with conventional manufacturing processes in order to generate the final part [76, 78, 162]. According to [94], there was a significant influence involved in the diameter, cross section shape and orientation of the fibers, whereas the fiber diameter can also range in the mm regime for BAAM systems. The challenges and opportunities were also discussed in [76, 78, 162, 175, 176, 187–189] and partly reviewed in [94].

In [187], the authors stated the requirement for development for “large-scale and high-productivity AM with low-cost material feedstocks and minimal environmental controls”. The use of high performance computing methods was recommended. In [78, 188], the first successfully installed system was described in detail, which uses pelletized feedstock described further in [175] and reached part parameters similar to injection molding as concluded in [162]. The influence of rheology was discussed in [76] concluding the need for stability of the parameters throughout the print. The rheology of both ABS and PPSU was concluded to be dependent on shear rates, CF loading, and frequency at different temperatures. Wrapping and cracking was identified as major challenge in [176] and described mathematically as function of the top layer temperature.

PPSU was used for the good thermal stability also at higher temperatures. It was shown that the screw speed as well as viscosity properties are the major influencing factors for production speed as well as mechanical properties of the final part. It was concluded that the viscosity increases significantly at higher weight fractions of fibers. An increase of the CF weight content from 20 to 35 % wt increased the viscosity of ABS by 60 % and of PPSU by 300 %.

33
The wide and high additive manufacturing (WHAM) project was also performed at ORNL in order to support the transportation and energy industries in the United States with rapid manufacturing technologies exceeding the dimensions of 6 m supplying 454 kg/h of extruded material. A machine from the company Ingersoll® capable of additive, subtractive, pick and place as well as tape placement was used and developed further by applying an extruder from the company Strangpresse® allowing the possibility to upscale accordingly.

High-performance applications were manufactured in [81] using PEEK, which requires higher processing temperatures but in return provides better mechanical properties. Thermal properties of the final parts are also dependent on the filler material as shown in [175] characterizing CF and GF filler materials.

The thermal behavior of the freshly extruded material was recently analyzed in [176]. An approach towards the optimization of data sets for BAAM was performed in [190] aiming to improve the print quality by optimizing the particular design parameters, tool paths, and printer settings in order to e.g. reduce the number of stops that the printer performs due to print path changes. The research was performed using an infrared (IR) camera for direct feedback from the BAAM system.

The internationally working wind turbine manufacturer Vestas A/S is also working on composite and metal materials in AM for the lightweight design as stated in [191, 192]. The idea, however, to use AM for large-scale airfoil applications as well as molding has already been discussed in 1996 in [193] using fiber-reinforcement.

**Direct Additive Manufacturing of Airfoils** While AM of molds allows to apply conventional technologies for airfoil manufacturing, direct AM of airfoils requires additional investigations especially in terms of structural mechanics. Patents were filed in [194–196] claiming the invention of a high temperature AM device to protect the airfoil’s leading edge.

Research on the BAAM system at ORNL was furthermore presented in [78, 162] investigating the mechanical properties of BAAM structures further with and without fiber-reinforcement in the form of CF and GF in ABS and PPS matrix material. The authors claimed a high strength of more than 60 MPa and stiffness of 12 GPa, which is an increase of some compositions of 95% and 420% as compared to the plain material. Given the improved mechanical properties of the materials, direct manufacturing would be possible to pursue.

**Generator Technology** Advancements in permanent magnets by using AM technology were reported in [197, 198] saving resources and energy in the production. The magnets were manufactured using isotropic, near-net-shape, neodymium-iron-boron
2.3 Applications of Fiber-reinforcement in AM

(NdFeB) bonded magnets produced by a BAAM machine at ORNL. The newly developed manufacturing techniques allow for up to 50% of waste reduction as compared to sintered magnets as well as better magnetic and mechanical properties.

Further research was published in [199, 200] for extrusion-based 70% in volume NdFeB in a nylon matrix. A comparison was performed between sintered, IM and AM magnets. Porosity has the potential for improvement while the electric properties are comparable to conventionally sintered materials. The ultimate tensile strength of sintered magnets was significantly higher than the compared materials. IM magnets and AM magnets are comparable in their properties except for the ultimate strain, which is higher for AM magnets.

The grain size of the same material processed by powder bed fusion in [201, 202] was located in the range of 1μm demanding a thorough consideration of speed, focus, power, and layer thickness. On the other hand, post-processing was not necessary. Internal texture and structure growth patterns were detected allowing the possibility of anisotropic AM magnets.

Model and In-situ Building One possible application of AM in wind energy is model building discussed in [203, 204]. The aim was to rapidly manufacture models of blades, wind turbines, or entire wind parks in order to test them e.g. in wind tunnels where the airflow can be visualized. The relation between the dimensional expansion and the wind speed in the model and in reality can be calculated using Reynold’s number.

Other scientific contributions used AM in order to manufacture models for wind tunnel experiments at a significantly scaled model allowing for conventional AM technologies using both extrusion-based printing of polymers as well as powder bed fusion for metal models investigated among others in [205–209]. In most cases, the internal structures of the model were neglected as different parameters were considered in the overall experiment. The models not only represented airfoils but also models of other components of wind energy systems. Different models were presented showing applications in wind tunnel testing as scientific research method.

Another possibility presented in [203] is in-situ printing of wind turbines, which would essentially benefit rural and hardly accessible regions, which on the one hand would grant easier access to energy to these regions and on the other hand reduce the transportation costs and efforts to transport entire wind turbine blades into those regions.

A similar situation applies to wind turbines e.g. manufactured off-shore for more efficient energy production using the more suitable wind conditions off-shore. Production time is moreover significantly reduced as compared to conventional manufacturing. It was concluded in [43, 44] that extrusion-based AM of the wind turbine blades is possible within 5 to 7 days, but they are only suitable for wind speeds of up to 9 m/s.
A large number of extrusion-based models for smaller wind turbines is available online mostly provided by amateur printers. In terms of construction, AM can increase the functionality beyond the currently known capabilities as concluded in [210]. Given that the construction industry tends towards modular solutions, AM can play an incremental role as in single material structures and fully autonomous constructions.

**Energy, Resource, and Cost Efficiency**  While authors of some of the above-cited papers argued that the newly developed technologies especially in terms of generator technology require less energy and resources than conventional manufacturing technology, it was pointed out in [211] that AM technologies are still missing or incomplete in available databases for life cycle assessment (LCA), which are further described in chapter 9.

It was, however, argued in [212] that manufacturing process efficiency can be significantly increased by tools and molds produced by AM. On the other hand, direct manufacturing can skip these parts as well and increase the production speed. The aim of Siemens Power Generation Services is to improve maintenance, support and repair by providing spare parts. Therefore, direct metal laser sintering (DMLS) was acquired.

Research should also go into the direction of manufacturing parts for multi-purpose and multi-time use as the material costs make up 27.8% and processing costs make up 21.4% of the entire part costs of a BAAM part manufacturing according to [75]. This was still an optimization as compared to an industrial extrusion-based machine with material cost fraction of 38.3% and processing cost fraction of 58.7%. Costs of BAAM systems were shifted to post-processing, which made up for 45.5% of the costs. The processing cost fraction of powder bed fusion was still significantly higher than extrusion-based printing and BAAM.

Cost-efficiency can be increased as pointed out in [213] arguing that commonly-based peer production is able to manufacture a modular helix wind turbine. In this case, all elements of the turbine were printed on an extrusion-based system except for the motor. The advantage in this approach is the modularity of the wind turbine allowed by the helix design. This avoids BAAM manufacturing techniques.

While several extrusion-based models of AM airfoils for smaller wind turbines are available online for download and self-printing, smaller models are also used in research laboratories and especially wind tunnel experiments. Most models neglect the mechanical properties of the turbine blades, but focus on the airflow around them in the model size. AM in these cases is chosen for cost-efficiency, production speed, and flexibility in design.
Larger applications of airfoils are not manufactured directly in most cases, but rather a form is manufactured from polymer or metal material and used for further airfoil production as an open forming mold. This also involves post-processing of the surfaces and is chosen for cost-efficiency, production speed, flexibility in design, and reduced waste production as compared to conventional manufacturing.

Energy production in general also profits from AM magnets used in generators. While the properties of AM magnets are similar to sintered and other conventionally manufactured magnets, waste production is significantly reduced.

The most intensive research on BAAM as well as wind energy related research of AM is conducted at ORNL and sponsored by the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy as well as several other companies engaging in cooperation with the laboratory. Research is also performed directly at wind turbine manufacturers, but usually underlies non-disclosure regulations. Advances in wind turbine manufacturing can benefit renewable energy production not only in places where conventionally manufactured turbines are currently in use, but also in rural areas due to in-situ printing where transportation and assembly is usually impossible or uneconomical.

### 2.3.2 Applications in Biomedical Engineering

Strategies to induce self-healing behavior in FRP-based composites were introduced in [10]. The aim was to induce a self-healing functionality in the polymers by filling the fiber tubes with a healing agent. It was concluded that it was difficult to exactly locate the fibers in the epoxy polymer which increased the production costs considerably. Recent research was conducted with non-ordered fibers [11, 214].

So far, there are no approved applications of AM of FRP in the field of medicine-related technologies. A summary of the possibilities of AM in biomedical engineering focusing on tissue and scaffold generation is given in [215]. It was concluded that the main advantage of AM is located in the reproduction of hierarchical structures.

In [216], FRPs in biomedical engineering were discussed with a focus on biologically-inspired materials using direct printing. It could be concluded that the storage module and the shear stress increased significantly when the polymer was filled with SiC fibers. This makes AM FRP suitable for bone-like structures [217].

Fibroplast cells were in part reviewed and investigated in [66, 218–220] as potential ingredient for fiber-reinforced organic structures for biomedical applications. In this method, collagen-based photocurable hydrogel as well as endothelial cells would be used in order to place them onto a biopaper or other 3D objects. For example, an artificial meniscus cartilage was printed. The objects were manufactured in a droplet-like deposition order. It is planned to use this technology further in such a
way to allow multiple filler materials. However, it was concluded in [218] that the printing of organs is not to be expected within this decade.

2.3.3 Applications in Aerospace

Research performed by the National Aeronautic and Space Administration (NASA) and published in [80] aimed to reduce emissions, fuel burn, and weight of the turbofan engines of business jets. For this aim, FRPs were used in combination with the ME printing of chopped CF in PEI for a valve on the first stage of the compressor blade.

Another example of the commercial use of FRP and AM in aerospace is the Airbus A350-XWB as well as the Airbus A380 [39–41, 75, 221, 222], where weight reduction at similar or higher strengths are key issues. At ORNL, PPS with 50\% wt CF was used for part migration from Al to AM polymer composites for firms like Boeing, Ford, Techmer, and BSF [75].

The properties of FRP in AM, comparing CF-reinforced parts produced by AM with aerospace-quality aluminum were discussed in [15, 223]. It was concluded that production rates are extremely low and that the physical size of the parts is generally small. Moreover, the mechanical properties of the polymer parts are generally poor, which limits the potential for direct part replacement and functional use of the polymer components.

The reduction of part count and assembly steps using AM in terms of aircraft makeup was discussed specifically in [221–223]. Moreover, it was discovered that deformations during curing were reduced at a layup of continuous fibers with a resin matrix material. In this specific case, 40.6\% of weight savings was achieved at a part count reduction of 50\%.

Satellite applications with a focus on design and functionality were mentioned in [221]. Fundamental research for general aerospace applications was conducted in [172] testing the commercially available nylon/CF composite material Ultem® under different thermal and mechanical conditions.

CF filler material was among other materials tested in an ABS matrix material for cube satellites in [113] in terms of mechanical, thermal and optical properties concluding a significantly increased porosity as well as the already reported gaps and cracks around CFs in a thermoplastic matrix due to the weak chemical interactions.

2.3.4 Applications in Molding

Small and Multi-Scale Molding (Injection Molding) The aim of this technology is to replace conventional injection molding (IM) cavities by inserts manufactured by
VPP at a cost of reduced lifetime and with the advantages of reduced costs, production time, and environmental impact. Additively-manufactured, fiber-reinforced inserts can be considered suitable for pilot production with low part numbers, and are therefore an effective alternative to more expensive inserts made from brass or steel.

Over the past years, IM with AM inserts has become state of the art and can be purchased as a service by a number of both national and international suppliers. However, the introduction of fiber-reinforced composite materials in this technology represents a novelty described further in Part III.

Large-Scale Molding

Large-scale molds with FRPs were investigated in [174] using epoxy coatings and machining to improve the quality of the surface finish. The quality of the mold does not entirely compare to metal molds, but is rather only suitable for low-volume production. Large-scale molds consisting of CF filled ABS were investigated in [224] at relatively low costs of 10$/kg as compared to 20 to 100$/kg for high performance polymers. The surface was thereafter milled using a CNC mill. Research was performed at ORNL and described among others in [163] formulating the process steps as follows:

1. CAD-based 3D model,
2. tool path generation,
3. mold manufacturing using BAAM system,
4. mold surface finish/coating,
5. dimensional analysis using laser scanning,
6. vacuum assisted resin transfer molding (VARTM),
7. second laser scanning to obtain dimensional changes, and
8. dimensional/deviation analysis.

Mold technologies for large-scale AM are proposed in [81, 225] for metal BAAM at ORNL for metal printing of 45 kg/h, and the current maximum print envelope of (6 × 2.4 × 1.8) m³. The maximum volume depends on the size of the print chamber similar to conventional powder bed fusion technologies.

This form of manufacturing eliminates the oven that is usually necessary to manufacture sintered parts. By using SLS, the energy consumption of the manufacturing process can be significantly reduced as compared to a conventional manufacturing process according to [75]. BAAM for metal as well as general design principles were extensively discussed in [226].

Despite metal molds, foam molds were reported in [227] for a refrigeration foaming process. The mold was equipped with integrated fluid passageways and significantly
2 Implementation of Fiber-Reinforcement in Additive Manufacturing

reduced the production time. Polymer molds with additional fiber-reinforcement allow to increase the lifetime of molds under thermal stresses significantly, which has extensively been investigated in this thesis. This technology can not only be used for wind energy applications, but was also used for car applications presented at the Detroit Auto Show in January 2015 [102].

Research on large-scale AM technologies at ORNL is among others funded by the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy in cooperation with companies like Lockheed Martin [228] and Cincinnati [190].

2.3.5 Applications in Racing

Combining the need for high individualization, high strength, lightweight, and a low production number, Formula 1 (F1) racing cars use fiber-reinforced powder bed fusion parts with CFs in their wheel suspension [147].

ME technology has been used to build an oil sump with increased mechanical properties, combined with the advantage of a more free design of the layered structure of the ME part in comparison to conventional manufacturing [229, 230].

2.3.6 Applications in Train Technologies

Modern technologies such as powertrains are critical in structure weight. BAAM for prototyping was used in [231].

2.3.7 Applications in Electrical Components

The potential use of especially carbon-based fibers in ME parts for sensors was discussed in [62, 63]. The possibility of producing flexible electronic components as well as microelectronics was reviewed in parts in [18, 218]. In [63], the mechanical strength of the components was increased by 70% for tensile and 18.7% for bending strength. It was furthermore concluded that the fiber-matrix interface needs to be significantly improved. Electrical properties as well as thermal properties were significantly increased according to [62] at CNT content between 1 to 6%wt.

MWCNTs were used in a thermoplastic PU matrix in [45] and ABS in [62] achieving piezoresistive behavior as well as a Joule heating effect. The composite was embedded by a multi-nozzle ME extrusion system into a polymer encapsulation and provides a flexible electronic component. A steady-state behavior was achieved after a single-digit number of cycles. By using DLM, conductive features down to sizes of 100 μm were manufactured in [153] using PLA matrix material and MWCNT filler. This allows for sensors as well as multi-dimensional electrical components.
Several solutions for FRP in AM have been recently investigated and are currently available. Experiments using ME, VPP, UC, sheet lamination, and binder jetting in combination with short fibers were successful. Sheet lamination and ME were also performed with long fibers.

Current applications for additively manufactured FRPs are rare, although there are multiple conventional applications using short and long fibers embedded in a polymer matrix such as aerospace, automotive, wind energy, and biomedical engineering. The technologies were mostly used in order to reduce costs, weight, increase manufacturing flexibility, and reduce production waste. In biomedical engineering, potential applications were pointed out where AM might be able to intelligently deposit fibers into the polymer to customize mechanical properties.

FRPs, in any case, lead to new mechanical, thermal, and chemical properties of the polymer, metal and ceramic matrix. The alignment and proportion of the fibers compared to the matrix have a critical influence on these properties. While ME can be concluded to be relatively flexible towards an increased fiber content, technologies such as VPP are significantly influenced, especially when working with CFs. This behavior results from the optical curing mechanism that is influenced by the optical properties of CFs. This effect has been investigated and is discussed in chapter 3 where the light input has been significantly increased in order to allow a solid layer manufacturing.

Fiber orientation depends strongly on the fiber material and manufacturing technology. While continuous fibers are usually aligned, the alignment of short fibers is a difficult task. Short fibers are usually placed randomly in the original material (e.g. the filament, resin, or powder) and the orientation is in a later step influenced by the manufacturing technology. Efforts and investigations to align fibers in a polymer matrix by the use of AM have been undertaken in industry, but are relatively rare and most parts are comprised with random or non-controlled, but influenced fiber orientation. Investigations of fiber orientation in a VPP process have been performed and are presented in chapter 4. Material extrusion methods are usually aligned along the extruded filament and are also further investigated in chapter 5. Furthermore, fiber orientation has a significant influence on the propagation of crack through parts in application. This aspect is investigated in chapter 6 and chapter 8.

Moreover, the fiber-matrix interface requires critical investigations in the chemical reactions between the surfaces of matrix and filler material. Thermal conductivity of specific polymers matrix material can be influenced by adding fibers with higher thermal conductivity and performed better than spherical fillers of the same material. Strength and stiffness are naturally desired properties to increase, which is usually the case for fiber-reinforcement of AM. However, the full, theoretical potential of fiber-reinforced AM is not yet explored due to a lack of technologies and methods to
ensure a fully developed and strong fiber-matrix interface. Investigations have been performed throughout this thesis in order to ensure a better quality especially for GFs using the chemical properties of thermoset photopolymers. The results can be found in subsection 3.3.3.

It can be concluded that the potential for FRPs in AM is very promising for future applications especially in terms of material parameter customization, which can be used to adapt the material properties to the desired applications. This thesis in particular presents fiber-reinforced IM inserts as potential application in Part III showing improved values in the final application. Most technologies for embedding FRPs in additively manufactured products are currently not fully developed. This aspect will need particularly dedicated research efforts in order to increase the applicability of FRPs in AMs. There are e.g. no safety-critical applications of additively manufactured FRPs.

In most cases, the aim of the specific application is to reduce the weight of the application and not suffer from the weaker mechanical properties of simple polymers without composite material. Besides this observation, the freedom of design that comes with AM application has been utilized as stronger mechanical parameters. FRPs in AM therefore prove to close the gap between cheap, but weaker polymer AM, and significantly more expensive metal AM.
Part II

Additive Manufacturing with Short Fiber-Reinforcement
3 Challenges in Manufacturing with Vat Photopolymerization

Literature research shows multiple applications of fiber-reinforced polymers respectively in material extrusion and binder jetting influencing the quality of the products in terms of stress and strain resistance as well as flexibility. So far, applications of fiber-reinforced polymers in vat photopolymerization are limited.

Fiber-reinforced polymer composites were manufactured into test objects using vat photopolymerization as well as vat casting. Short fibers were used in an unordered manner. An anisotropic property due to fiber orientation within the material was observed. The importance of fiber length and shape compared to layer thickness has been investigated including concepts to circumvent clustering of the fibers.

This research contributes to the implementation of fiber-reinforced polymers in additive manufacturing technologies. Vat photopolymerization allows generation of miniaturized objects with relatively high surface quality compared to other additive manufacturing technologies. This chapter aims to move fiber-reinforced resin parts one step closer towards mechanically strong production-quality components.

The content of this chapter was taken and in parts modified from the following publications:


The research on fiber-matrix interface was performed during a research stay at the Mechanosynthesis Group of the Massachusetts Institute of Technology (MIT) in collaboration with A. John Hart and Abhinav Rao.

### 3.1 Introduction

In this chapter, VPP was used to test a photopolymer resin combined with short CFs with dimensions of 7.2 μm diameter and 100 μm average length. The fibers were contributed by the ZOLTEK Corporation under the name “PX 35”. A technical data sheet is provided in Appendix C and [233]. The density was 1.75 g/cm³ with a carbon content of 99%. A bottom-up style VPP platform was used. The interest in the printed objects among others was the distribution of the fibers within the object.

For the resin without fibers, two products that are freely available on the market were chosen. The FunToDo [234] resin has a density of 1.016 g/cm³ and is cured at wavelengths between 350 to 450 nm. The VenusCreator [235] resin can be cured at a range of 340 nm and 410 nm.

Further research was performed in order to improve the fiber-matrix interface. A customized thermoset photopolymer material composed from 49.9% trimethylol-propane triacrylate, 49.9% 2-hydroxyethyl methacrylate, and 0.2% irgacure 819 (Bis(2,4,6-trimethylbenzoyl)-phenylphosphineoxide) was utilized with short GFs with dimensions of 7.2 μm diameter and 100 μm average length. Up to 5% wt GFs were utilized since (1) the chemical interaction between a thermoset photopolymer and GFs usually performs better than with CFs, and (2) since a casting technique was used, which only allowed for GFs since CFs absorb the applied curing UV light.

Research presented in [236] and more recently in [138, 237] suggests a gelation region for thermosetting materials, which can be achieved by increased temperature during the curing process. The material behavior is hence depending on both optical as well as thermal curing parameters. This knowledge has been utilized in order to improve the fiber-matrix interface of the presented thermoset photopolymer with GFs and as a result improve the mechanical properties of the investigated material. Bad wetting of the fibers is also mentioned in [13] as main favor of non-optimal material parameters in ME, but can be extrapolated as a general problem in fiber composite manufacturing, and especially AM.

### 3.2 Methods

A general differentiation of VPP technology can be established by printing orientation where bottom-up and top-down VPP can be isolated as described in Figure 3.1. The
3.2 Methods

Bottom-up printing

Top-down printing

Figure 3.1: Process scheme for bottom-up and top-down VPP. The build plate is moved according to the arrows and the photopolymer is exposed from the direction of the arrow by a digital projection unit or a laser in a rastered manner.

orientation of the UV and light projection has a significant influence on the process parameters and challenges connected to the printing process, fiber orientation, and printing speed as further described in chapter 4. While historically, VPP machines used bottom-up technology, recently top-down has become more common with certain characteristics, especially connected to light transmission and surface tension.

In these specific investigations, fibers were mixed into the resin at ratios of 5\%_{wt} and 10\%_{wt}. The composite photopolymer resin was cured from the bottom in a VPP machine modified from a design developed in [238]. The light passed from the projector through the bottom of the vat and the resin onto the build platform. After the exposure of one layer, it was necessary to lift the build platform by one layer height plus 5 mm allowing the resin to flow beneath the build platform and provide a uniform distribution of the fibers. The axis resolution was 0.625 μm. For the entire printing process, the machine was configured to expose a fiber-filled resin.

The manufactured parts were then inspected under a Zeiss Stemi 2000-C optical microscope with a TV2/3”C adapter for the view from top and bottom as well as under a JSM-5910 scanning electron microscope (SEM) using cuts orthogonally to the layered structure cutting in the middle of the object. SEM images with frame were taken on a ZEISS Merlin high-resolution SEM. The probes were molded, polished for 1 μm resolution and gold-plated with a 10 nm thick layer.

A cubic brick with attached cylinders was chosen for investigation for its thin walls and combination of round and flat geometries. Moreover, multiple exposure parameters were chosen for the two different photopolymer resins. Layer thickness was set to
3 Challenges in Manufacturing with Vat Photopolymerization

35 μm physically forcing the fibers to be placed aligned with the layers, but not orthogonally to the layers.¹

Other experiments were performed in a casting environment and further tested in a pull test on an Instron® 5984 machine under conditions in compliance with DIN EN ISO 527-2BB [239]. The parts were casted in a dog bone shaped vat shown in Figure 3.2 and Appendix C where the vat was manufactured from PTFE and cut by water jetting in order to achieve an optimal surface roughness. The bottom of the vat was manufactured from aluminum in order to achieve a good heat transfer from the heating bed placed below the casting vat. The entire vat was exposed to UV light from the top for 30 s.

The light penetrated from the top through the transparent resin and therefore caused the final curing. Initial curing was triggered by the heat propagating from the bottom where the vat was placed on the heat bed until a uniform temperature distribution was achieved. The temperature was set from room temperature upwards, whereas the gelling of the resin increased with elevated temperatures. The parts were not post-cured e.g. by flashing, but stored in a light protected container in order to avoid a generation of brittleness as is a usual effect of polymers under continuous UV radiation.

¹The orientation of fibers within the object, and therefore in the first instance during the manufacturing process, can have a significant influence on the crack propagation, lifetime, and strength of the final part and is therefore investigated further in chapter 8.
3.3 Results

The build plate had to be lifted after the exposure of every layer in order to allow the resin to flow beneath the build plate and create a uniform distribution of fibers. This resulted in a longer manufacturing time of the part. Since bottom-up technology was utilized in the described experiments and the resin vat was fully wetted, the surface tension of the photopolymer resin had no influence on the layer geometry or part quality.

3.3.1 Optical Microscopy

Due to the nearly transparent photopolymer, the part could be investigated in a view from the top (orthogonal to the layered structure) shown in Figure 3.3 (left). The figure shows a random distribution of fibers within the object. The fiber direction in the view layer is random within the layer, but not orthogonally to it. It can also be seen that the standard deviation of the fiber length is high compared to the average fiber length of 100 μm.

Investigation of the borders shown in Figure 3.3 (right) concluded that the fibers did not stay within the cured photopolymer but were standing out of the object. This makes it necessary to post-process the object in order to receive the favored surface quality.²

²Further investigations in AM IM inserts showed a similar effect of fiber sticking out of the part that were broken during the first shot of thermoplastic material and stuck to the injected part leaving the insert surface with not noticeable defects. The results of these investigations are presented in Part III.
Figure 3.4: Rough cut through the layered structure under the SEM for 5\%_wt fiber content. This figure was published earlier in [108].

No clustering around edges was detected, allowing the conclusion of a more even distribution of fibers standing in contrast to IM where clustering around edges is a common phenomenon. Post-processing was necessary due to the fact that the CFs were not affected by the visual curing in the VPP machine. Therefore, the fibers were standing out of the boundary of the object. This resulted in an uneven surface, which needed to be further processed.

3.3.2 Scanning Electron Microscopy

A rough cut through a sample orthogonal to the layers is plotted in Figure 3.4 and shows the layered structure of the object as well as the fibers. Except for some fibers, the direction of the fibers lies within the manufactured layers. Note again the different lengths of the CFs.

Fibers were oriented along the layers as can be seen in Figure 3.5 (left) allowing a reinforcement of the printed part by placement of it during the printing process. An uneven distribution of the fibers as discussed in [240] for conventional manufacturing
3.3 Results

Figure 3.5: Fiber distribution and orientation in the middle of the printed part (left) and at an inner corner (right) showing an even fiber distribution among the layers for $5\%_{\text{wt}}$ fiber content. This figure was published earlier in [108].

could not be concluded from the results of the SEM observations. Figure 3.5 (right) shows an inner corner of the part with evenly distributed fibers among the layers. The upper right corner or the figure shows molding material.

Clustering of the fibers was recognized in 30\% of all detected fibers under the SEM resulting in holes within the photopolymer as shown in Figure 3.6 (left). The fibers themselves were surrounded by gaps between the fibers and the photopolymer resulting from the shrinkage of the photopolymer during and after exposure. Figure 3.6 (right) also shows a crack in the photopolymer which will result in decreased mechanical strength and durability. The fiber diameter is shown to be consistent with the product specifications of around $7.2\,\mu\text{m}$ given in [233].

The gaps between the fibers and the polymer with the diameter of 1 to $2\,\mu\text{m}$ resulted in cracks of the polymer shown in Figure 3.6 (right) with an average length of 3 to $4\,\mu\text{m}$, which may affect the strength of the final part. Another attempt to prevent crack propagation was performed in [135] controlling the direction of the fibers and therefore allowing for design optimized printing. A further investigation on this field of research can be found in chapter 4.

3.3.3 Fiber-Matrix Interface

Figure 3.7 shows examples of fiber-matrix interfaces before and after an increase of the curing temperature in order to reach a gel state during the curing. While Figure 3.7 (top) shows a similar behavior as the examples presented with a commercial photopolymer in Figure 3.6, the interface of the samples cured at elevated temperatures shows different characteristics. All pictures were taken under SEM investigations after pull tests have been performed on the parts.
Figure 3.6: Detailed view of clustered fibers (left) and single fiber (right) with gap and crack for 5\%_w fiber content. This figure was published earlier in [1, 108].

Figure 3.7: Examples of fiber-matrix interface defects and cracks around fibers in a thermoset photopolymer matrix cured in an unordered manner (top). Improved fiber-matrix interface of the same manufacturing configuration at a different curing temperature (bottom).
The samples in Figure 3.7 were pulled, but not polished. They show a characteristic behavior of fiber cracking along the major crack line. Over the course of several investigations, a number of fibers were detected that were pulled out of the matrix material, especially in the non-improved configuration. Otherwise, the brittleness of the fiber material caused sharp breaks of the fiber after the matrix has been pulled apart.

Experiments performed at elevated temperatures showed two effects:

**Fitting** of fiber and matrix shown in Figure 3.7 (bottom left) where the matrix ends immediately at the boundary of the fiber and also penetrates the rough surface of the fiber, which has the effect of a stronger fitting as compared to a smooth surface.

**Connecting Layer** of the matrix in the surrounding of the fiber as shown in Figure 3.7 (bottom right) where the matrix material changes in form and crack behavior around the fibers. It is though unknown whether this happens due to re-melting of the matrix material filling up the gaps around the fiber and matrix; or whether the temperature of the fiber was elevated to a level that caused the matrix material to change its properties around the fiber.

In fact, the elevated temperature had positive effects on the mechanical properties of the specimen as shown in Figure 3.8. In total, a factor of 3 was established in tensile strength increase as well as a factor of 1.6 in modulus increase. However, the increase is not guaranteed since the standard deviation shown in the graphs of the sample size of 7 specimen per sample was significant. Moreover, the increase of strength and modulus is not equally distributed at the elevated curing temperatures. While tensile strength at break increases significantly at lower elevated temperatures, the modulus is massively increased at 100°C. It was also noted that the brittleness of the specimen increased massively not only by elevated temperatures during the curing process, but also by the increased amount of GFs in the polymer matrix.

Depending on the application and the therefore desired mechanical properties, the curing profile can be chosen accordingly. It has, however, shown very difficult to cure the underlying thermoset resin at higher elevated temperatures since the uniform temperature profile cannot be guaranteed and furthermore the practicability is questionable.

Another system using elevated temperatures in order to print thermoset photopolymers in a hot lithography process has been introduced by the company cubicure [241] addressing furthermore problems of viscosity of resins at different configurations. The system runs in the UV spectrum from 375 to 405 nm using temperature control systems in order to gain up to 120°C.
3 Challenges in Manufacturing with Vat Photopolymerization

Figure 3.8: Tensile strength at break (top) and Young’s modulus (bottom) determined according to DIN EN ISO 527-2BB at different curing temperatures. Errorbars are calculated from the standard deviation among 7 specimen per sample. This figure was published earlier in modified form in [4, 232].
3.4 Summary

Manufacturing comes as a step in the process chain following the material selection. It moreover has to be considered as one of the crucial factors for the further performance of the manufactured part. This consideration has to take place in the beginning before deciding which technology (bottom-up or top-down) to utilize. This decision is moreover crucial for the fiber orientation, but can be utilized as control mechanism as is shown in chapter 4. In the final part, the fiber orientation can significantly influence the lifetime of the part as well as the crack propagation shown in chapter 8 since cracks due to thermal or mechanical stresses propagate through the matrix and slide along the fibers whereas they are blocked when penetrating a fiber from the side.

The choice of fibers is furthermore crucial for the later manufacturing process since e.g. CFs come with different optical properties than GFs, and the light intensity during the curing process needs to be elevated at higher levels of CF fiber content, layer height, or curing speed. The manufacturing process is therefore a function of multiple parameters that need to be considered for the different applications.

Fiber segregation, however, can be prevented by mechanisms in the resin vat generating e.g. a vortex to stir the resin. A number of mechanisms are already in use, especially for bottom-up printers with the additional purpose to clean the resin vat from additionally cured material on the vat surface.

In the final part, fibers tend to stick out on the surface of the part where the resin remains uncured but the fiber sticks to the part due to the high length/diameter ratio of the fibers. The very same ratio becomes a limiting factor in terms of clustering of fibers as well as the resulting gaps within the fiber clusters. This also results from the mixing process of the fibers into the resin where air intrusions can result from both fiber bulks as well as during the shaking process. The number of air intrusions that significantly weaken the mechanical properties of the final part have been investigated further in chapter 8.

Curing mechanisms of thermoset photopolymers were utilized furthermore in the prevention of cracks around fibers resulting from the different expansion coefficients of the matrix and fiber material. The resin was chemically activated by increasing the temperature of the resin bringing it into a gel state and therefore utilizing both thermal and later optical curing mechanisms. This method resulted in a significant increase of both tensile stress and Young’s modulus and can be utilized as additional curing mechanism when working with CFs currently limited in layer thickness and fiber content due to the light absorption of the black fibers.
4 Orientation of Short Fibers During Vat Photopolymerization

While fiber-reinforced additive manufacturing combines the advantages of generative manufacturing technologies with the knowledge and abilities of composite materials, there has not been a suitable prediction of fiber orientation during the vat photopolymerization process. Descriptive research shows that material is strengthened along the fiber orientation, while cracks can propagate along the fibers. Cracks are slowed or stopped when targeting fibers from the side, which makes fiber orientation a crucial factor for both mechanical properties as well as crack propagation.

A numerical model was used to predict the fiber orientation for multiple use cases of vat photopolymerization of thermoset photopolymers. Traditional bottom-up processes as well as top-down printing are investigated and a modified build plate design is proposed in order to reach a more isotropic fiber orientation. Predictions on fiber clustering and overall fiber orientation are given as well as an outlook into potential fiber orientation control.

The content of this chapter was taken and in parts modified from the following publications:


4 Orientation of Short Fibers During Vat Photopolymerization

4.1 Introduction

Fibers are usually oriented along the printed layers as described further in chapter 3 and chapter 8. This specification is especially predominant in bottom-up printing where the printing space is confined between two solid plates and the usual ambition is to reduce the gap between the plates to a minimum in order to improve the accuracy of the print. The challenges are moreover comprised of clustered fibers and an ill-conditioned fiber-matrix interface due to the weak chemical interaction of CFs with the photopolymer matrix as discussed in chapter 3.

Predicting the fiber orientation in ME processes was intensively investigated e.g. in [107, 243] concluding a strong influence of swelling mechanisms on the fiber orientation, whereas the convergence length of the nozzle was negligible for the fiber orientation, but geometric changes in the nozzle could modify the fiber orientation in the extruded material [107]. Findings regarding the influence of the die swell on fiber orientation were not reported in [243]. Instead, other factors such as an interaction coefficient and a slowness factor were considered as important influences on the fiber orientation in the thermoplastic material.

However, the ME technology is subject to a number of limitations constraining the range of applications. For this reason, VPP processes have gained attention e.g. in IM inserts due to their flexibility in production, freedom of design, surface features, and product design cycle time [114]. In this process, fibers are subject to 6 DOFs (degrees of freedom; 3 spacial, 3 rotational), while the rotational DOF along the axis of the fiber is not considered, as it has no influence on the mechanical, thermal, electrical, and other properties of the final part, when assuming fibers with a round cross section and isotropic topology.

These considerations were combined with existing knowledge on fiber orientation in moving liquids, which were investigated analytically as well as numerically for several decades in [244–246]. The improved understanding was utilized in [247] allowing for the conclusion that fibers orient along streamlines under a set of conditions summarized in [247] as

1. the fiber may be represented by an ellipsoid of revolution,
2. no-slip conditions prevail at the surface of the fiber,
3. the velocity field is only locally perturbed by the motion of the fiber,
4. there is no interaction between fibers,
5. the flow far from the fiber is steady and homogeneous on a length scale that is large compared to the fiber dimensions,
6. the motion is sufficiently slow that inertia forces are negligible, and
7. the fiber translates with the fluid velocity.
Challenges have been faced regarding the lifetime of polymer inserts described in Part III in connection with the involvement of thermal stresses, crack propagation, and molding cycle time. While the thermal effusivity of polymer inserts significantly exceeds those of metal inserts, the importance of temperature control and cooling of the inserts has increased. Fiber-reinforcement has improved the lifetime by a factor of 10, but further increase of lifetime is limited by fiber orientation as is experimentally shown in chapter 8, which has proven to influence the lifetime at a similar level as a defect fiber-matrix interface, and significantly influences tensile strength and stiffness. It was hence inevitable to investigate methods to predict and control fiber orientation in VPP parts.

The model simulates both bottom-up and top-down printing with a circular, rectangular, and a customized build plate. The structure of this chapter is as follows: initially, the model is presented together with its boundary conditions. Subsequently, the results of the model are presented and discussed and finally remarks of the investigation are presented.

4.2 Methods

Numerical flow models have previously been employed to analyze different processes with great success e.g. casting of sand molds [248–252].

The development of the numerical model in this investigation was conducted using the fluid flow module of Comsol Multiphysics® version 5.3a. The Navier Stokes equations were solved on a moving mesh domain:

\[
\rho (\mathbf{u} \cdot \nabla) = \nabla \cdot \left[ -pI + \mu (\nabla \mathbf{u} + (\nabla \cdot \mathbf{u})^T) - \frac{2}{3} \mu (\nabla \cdot \mathbf{u}) I \right] + \mathbf{F} + \rho g \tag{4.1}
\]

\[
\nabla \cdot (\rho \mathbf{u}) = 0 \tag{4.2}
\]

where

- \( \mathbf{u} \) = velocity vector
- \( \rho \) = density
- \( p \) = pressure
- \( I \) = identity matrix
- \( \mu \) = viscosity
- \( \mathbf{F} \) = external forces
- \( g \) = gravitation

Flat rectangular and circular build plates were considered in bottom-up and top-down processes. The process scheme has already been described in Figure 3.1. The
**Figure 4.1:** Design of experiment (DOE) and design thinking of the numerical simulations in order to predict and improve the fiber orientation within the final product (left). Modified build plate design in order to allow a vertical fluid flow pattern (right). This figure was published earlier in modified form in [4, 242].

underlying geometries were chosen as examples for reasonable build plate designs in commonly available VPP printers combining sharp corners, round geometries, different flow scenarios, and corner effects. The build plate is in the most cases manufactured from metal in order to allow for easier post-processing, but is also available as glass plates.

Information about the three scenarios that were studied is presented in Figure 4.1 referring to movements presented in Figure 3.1.

**Bottom-up Process** A rectangular or circular build plate is moved vertically within a photopolymer resin. The resin is exposed from the bottom and the part is built up layer-wise. Continuous printing according to the current state of the art is only possible when allowing oxygen to flow through the bottom of the resin vat as established in [253].

**Top-down Process** A rectangular or circular build plate is moved vertically within a photopolymer resin. The resin is exposed from the top and the part can be built up in a layer-wise or continuous manner. Viscosity and surface tension of the resin have a significant influence on the manufacturing process.

**Modified Design** In order to allow faster printing as well as improve the fiber orientation, holes were modeled in the build plate (as seen in Figure 4.1 (right)) allowing the resin to flow through the build plate. Viscosity and surface tension are still crucial factors in the manufacturing. Their influence is reduced and the flow pattern of the resin changes.
4.2 Methods

Figure 4.2: Domain of liquid resin for a rectangular build plate (left) and circular build plate (right). Bottom-up and top-down printing processes were considered by flipping the domain by 180°. This figure was published earlier in modified form in [4, 242].

<table>
<thead>
<tr>
<th>Domain Parameter</th>
<th>rectangular</th>
<th>circular</th>
</tr>
</thead>
<tbody>
<tr>
<td>vatX</td>
<td>150 mm</td>
<td>300 mm</td>
</tr>
<tr>
<td>vatZ</td>
<td>20 mm</td>
<td>20 mm</td>
</tr>
<tr>
<td>buildX</td>
<td>100 mm</td>
<td>200 mm</td>
</tr>
<tr>
<td>buildZ</td>
<td>10 mm</td>
<td>10 mm</td>
</tr>
<tr>
<td>velocity</td>
<td>0.25 mm/s, 0.5 mm/s, 1 mm/s</td>
<td></td>
</tr>
</tbody>
</table>

Table 4.1: Simulation domain parameters.

The modified build plate is designed in order to facilitate a vertical fluid flow pattern. The design is inspired by existing build plates focusing on accompanying the higher viscosity of the commercially available photopolymers. However, the build plate requires part modifications as the bottom layers placed over the drilled holes will be deformed into the holes.

Boundary conditions shown in Figure 4.2 were applied to a liquid domain. Moving mesh techniques were used to induce a flow in the domain using a hyperelastic mesh deformation, which allowed the mesh to partly compensate for the shift in mesh size during the deformation. Starting conditions were set to leave a two-digit μm gap between the build plate and the resin vat or resin surface in order to avoid singularities. Velocities of 0.25 mm/s, 0.5 mm/s, 1 mm/s were considered for the build plates. The later was considered for result evaluation in order to build a dimensionless number of the fluid movement. The full set of domain parameters can be found in Table 4.1.

While the described technologies often use step-wise movements into the printing directions, continuous movement of the build plate is also possible and used by some applications. The model mimics both a step-wise and continuous movement of the build plate. The fiber orientation is dependent on the velocity tensor, not
necessarily the absolute velocity. Moreover, top-down printing is already available in a continuous printing process, which has been modeled.

A significant difference between the bottom-up and top-down processes is given in the open boundary surface in the resin vat. While the other surfaces have been assumed to have slip conditions, there is no free surface affecting the printing position in a bottom-up printing. A traditional bottom-up machine has been assumed without any diffusion through the bottom of the resin vat as proposed in [253].

Streamlines were calculated using the Comsol Multiphysics® feature at a uniform density at an integration tolerance of 0.001 with 5000 integration steps and both stationary point and loop tolerance of 0.01. The streamlines were smoothed inside the material domain and applied with a color expression applying the absolute velocity. The fibers are assumed to follow the streamlines in their orientation.

**4.3 Results**

**4.3.1 Bottom-up Printing**

Clogging of the build plate seems unlikely due to higher velocities around the corners as seen in Figure 4.3 (top), which allows the argumentation that the moving build plate generates a suction effect that drags fibers towards the center of the build plate. This decreases at higher z-levels of the build plate as does the risk of clogging. A segregation of the fibers on the bottom due to the difference in density between fibers and resin has been accounted for in the simulation by a volume force. At a later printing stage with a larger gap between the resin vat and the build plate, velocities will moreover counteract the gravitational forces due to the vortex generated around the build plate.

Figure 4.3 (bottom) shows the general movement in the three dimensional case, whereas a symmetry effect around the vertical center axis is visible. Nevertheless, irregular fluid movement is visible around the corners of the domain around the build plate. It should therefore be considered to round the edges or use circular build plates, especially for bottom-up printing. However, considering a VPP mechanism, most commercially available technologies base on rectangular projection areas where fiber content will need to be considered around the corners of the projected area.

As mentioned earlier, streamlines give an approximation of the fiber orientation. As shown in Figure 4.4, fibers will orient preferably parallel to the produced layers and therefore parallel to the build plate. Not only the fluid flow, but also the desired layer height of the final part (and therefore during the manufacturing process) results in a forced orientation. Since most available milled CFs are equipped with lengths in the range of 100 μm, whereas usual VPP layer heights are in the range of lower two-digit μm–range, which does not physically allow the fibers to orient perpendicular to the
Figure 4.3: Velocity in mm/s of a bottom-up domain (top) and velocity arrows in the three dimensional case (bottom). Note the irregular arrow directions around the corners of the build plate. Both images were taken after 2s of simulation when the flow was fully developed. This figure was published earlier in modified form in [4, 242].
Figure 4.4: Stream lines colored according to the fluid velocity of a rectangular (top) and circular (bottom) build plate in bottom-up printing. Note the vortices around the corner of the domain at the bottom of the build plate. This figure was published earlier in modified form in [4, 242].
printed layer in the first layers. At larger gap sizes, a vertical orientation is also not likely since the liquid resin flows along the resin vat in a horizontal flow.

### 4.3.2 Top-down Printing

A top-down printing process can provide potential orientation to the fibers. As shown in Figure 4.5, a resin without surface tension can provide additional vertical orientation both for a rectangular as well as a circular build plate.

While the geometric domain represents a rotated copy of the bottom-up geometry, a larger fraction of the domain boundaries is represented by an open boundary and therefore regulated by surface tension. The moving build plate generates a downwards pull-effect that orients the fibers accordingly.

### 4.3.3 Modified Geometry

A further development, however, has been accomplished with a transmittable build plate, which in this particular case was equipped with circular holes in order to allow the liquid resin to flow through the building area and allow a flow of liquid and fibers through the build plate in a vertical direction. Figure 4.6 visualizes an additional mixing of the fibers through the specific geometry of the build plate, which can benefit the fiber orientation in contrast to results presented for conventional build plates in Figure 4.3, Figure 4.4, and Figure 4.5. However, some additional modifications need to be considered for the part as deformations might occur for the first printed layers. A printed build plate below the object cannot be inserted, as it would counteract the achieved effect.

Since the vertical velocity of the fluid provides an indication of the vertical alignment of the fiber material, it is investigated further in Figure 4.7 and normalized on the vertical build plate velocity. Once again, the clogging around the rectangular build plate can be seen to be unrealistic due to the higher velocities (top row).

More importantly, the vertical fluid velocity is structured significantly different from the normal top-down scenario when adding the modified build plate. Vertical velocities are visible where the holes in the build plate are located. Nevertheless, the velocity decreases at larger distances to the center of the build plate. It can be expected that a removal of the holes in the center (e.g. by design or due to a printed object placed on this location) will change the fluid patterns above and below the other holes.

Figure 4.7 shows velocities above the build plate (middle row) as well as immediately on the plane on the surface of the build plate (bottom row). It can clearly be seen that the influence of the build plate is still valid on top of the resin at an even increased level as compared to the velocity directly on the top and on the bottom of
Figure 4.5: Stream lines colored according to the fluid velocity of a rectangular (top) and circular (bottom) build plate in top-down printing. Note the vortices around the corner of the domain at the bottom of the build plate. This figure was published earlier in modified form in [4, 242].
4.3 Results

Figure 4.6: Stream lines colored according to the fluid velocity of a modified, generally rectangular build plate showing a significant improvement in the mixture of the fiber orientation in the building area (top). Velocity volume arrows of a modified, generally rectangular build plate (bottom).

This figure was published earlier in modified form in [4, 232, 242].
the build plate. This is also visible at the same geometry with a different radius of 2.5 mm shown in Figure 4.7 (forth row, left) whereas a reduction of the radius of the holes down to 1 mm reduces the vertical velocity significantly. It can be expected that an increased viscosity will enhance this trend.

4.4 Summary

Both bottom-up and top-down printing processes come with characteristic properties such as available layer height, surface accuracy, and printing speed. Whereas most commercially available bottom-up machines utilize a step-wise printing process, which allows to interrupt the print in order to e.g. stir the resin, continuous processes are available as well.

Top-down machines do not require additional hardware to utilize a continuous printing process, but are limited e.g. by surface tension and viscosity of the resin, which is also determined by the fiber content as described in chapter 3.

As VPP is a process determined by a large number of parameters, the development of simulation and prediction tools has become necessary to describe fiber orientation as an additional parameter effecting both the printing process as well as the properties of the final part.

A simulation technique for VPP processes has been presented allowing for the first time to predict the fiber orientation and partly distribution of both bottom-up as well as top-down processes. Additionally, a design modification has been suggested in order to enhance the mixing effect in the build area.

In terms of generative design, stress distribution and adaptive material composition, the orientation of fibers in thermoset photopolymer matrices spares a significant potential in the prevention of e.g. crack propagation along the printed layers as shown in several publications.

The presented method allows a predictive fiber orientation within the resin during the printing process before a final part has been generated and can be included in a dynamic adaptation of part design in a digital product development process. This effect is further discussed in chapter 8, where it is shown that the fiber orientation can be seen as more isotropic throughout the part when using the suggested modified build plate design.

Future developments can also include the possibility to specifically align or increase the concentration of fibers at specific positions and hence allow for a reduced material volume or diameter. This method can therefore bring technologies such as topology optimization to a new level saving additional weight while not decreasing mechanical stability.
4.4 Summary

Figure 4.7: Vertical fluid velocity fraction over the build plate velocity. 
First row: rectangular build plate top-down on top of the resin (left) and 2 mm over the build plate (right).
Second row: modified build plate top-down on top of the resin (left) and 2 mm over the build plate (right).
Third row: modified build plate top-down on top of the build plate (left) and on the bottom of the build plate (right).
Forth row: 2.5 mm radius holes in modified build plate top-down on top of the resin (left) and 1 mm radius holes in modified build plate top-down on top of the resin (right).
This figure was published earlier in modified form in [4, 232].
5 Orientation of Fibers During Thermoplastic Extrusion

The aim of this chapter is the understanding of the fiber orientation by investigations in respect to the inner configuration of a thermoplastic matrix reinforced with short CFs after a material extrusion process. The final parts were analyzed by X-ray, tomography, and magnetic resonance imaging allowing to resolve the orientation of the fibers and distribution within the part. The research contributes to the understanding of the fiber orientation and fiber reinforcement of material extrusion parts in additive manufacturing.

The content of this chapter was taken and in parts modified from the following publication:


The research on Joule pre-heating was performed during a research stay at the Mechanoynthesis Group of MIT in collaboration with A. John Hart, Jamison Go, and Adam Stevens.

5.1 Introduction

Extrusion-based AM is known to be the oldest form of AM, though the availability and common knowledge about manufacturing technologies such as VPP and metal AM is increasing. Based on the less-sensitive set of parameters for the printing process as well as the blending mechanisms for fiber and matrix material, the availability of fiber-filled filaments is higher and the range of materials is increased as has already been shown in several examples presented in Part I.

In connection with the investigations in chapter 4 and chapter 8, similar experiments were performed to investigate the behavior and impact of fibers on extruded parts. Experiments were performed on freely extruded PLA filament as well as nylon-based materials.
5 Orientation of Fibers During Thermoplastic Extrusion

Investigations of the final part were performed using a JSM-5910 SEM. SEM images with frame were taken on a ZEISS Merlin high-resolution SEM. Possibilities for the transmissive investigation of PLA are supported by [254] stating a continuous transmission of PLA of over 90% at wavelengths of 250 nm and higher.

To investigate the spatial alignment, orientation and distribution density of the fibers within the printed part as well as the conformity of the printed results with the intended shape both on mesoscopic and macroscopic level, radiological modalities like computed 2-dimensional X-ray radiography on photostimulated luminescence image storage plates, CT with human-grade multi-detector 128-line CT and research-grade micro-CT Scanners were performed. Finally, the material properties were explored with quantitative magnetic resonance imaging (MRI) methods at 3 T.

5.2 Methods

The parts were produced using PLA filament with 15%\textsubscript{wt} virgin, short CF content with average diameters of 7.2 μm and an average length of 100 μm as well as nylon with 12%\textsubscript{wt} GF content from the company Stratasys® as well as 30%\textsubscript{wt} GF content. A material data sheet can be found in Appendix E. During the manufacturing process, the fiber-reinforced filament was heated over the material glass-transition temperature and fed through a nozzle with a diameter of 400 μm. This resulted in an orientation of the carbon fibers along the path of the filament. This property could be used when aiming to reinforce the product in a certain main direction.

An orthogonal cut through the layers of the ME print of the objects was performed, followed by a polishing finish in order to visualize features of 1 μm voxel size before the SEM investigation. Due to the layer-wise orthogonal alignment of the infill, the cut showed one layer along the line produced by the ME nozzle and one cut orthogonal to the line allowing an elaboration on the orientation in three dimensions as well as the interface between the different extrusion directions.

Moreover, SEM investigations of the fracture surface were performed after a pull test on an Instron® 5984 machine conducted according to ASTM D638 IV [255] until the failure of the part. The dog bones used for this investigation were produced by the above-mentioned ME printer under layer-wise alternation of the printing direction in order to avoid globally anisotropic material properties in the final dog bone.

The printing paths are graphically represented in Figure 5.1. The outer paths were tracked around the object to provide a higher surface accuracy and a cohesion of the entire dog bone. By investigation of a single surface, cuts in two directions through the extruded filament as well as an investigation of the interface between the filament lines are possible.
Figure 5.1: First (left) and second (right) layer geometry of the printed dog bone. This figure was published earlier in [164].

This production method can be justified with the above-mentioned investigation aims of a detection of fiber orientation. Investigations aiming to find the increase of tensile strength or Young’s modulus have concluded a fiber orientation entirely in longitudinal orientation.

This research contributes to the understanding of ME processing with FRPs by observing the fiber orientation after the manufacturing and within the final part. The orientation of the fibers allows a directional reinforcement of the final part that may be manipulated on a mesoscopic level.

This material may offer susceptibility matching properties of greatest interest for nuclear magnetic resonance (NMR) / nuclear MRI applications in technology as well as in medicine. In [256] the benefits of using pyrolytic graphite foam as a passive magnetic susceptibility matching material were shown. This material seems to have similar desirable properties and at the same time offers improved mechanical strength while retaining the property of being able to be additively manufactured into desired shapes with readily available AM machines.

One possible further application to be pointed out is the individualised AM of potential medical implants. Conventional implants manufactured from non-ferromagnetic metals still retain a certain degree of resultant image artifacts in MRI and hence may make forthcoming examinations difficult to impossible.

Notwithstanding the tentative nature of the experience with the material and the clear implication of the strongest need for more research, the material may provide an easy way to manufacture, lightweight and economically attractive solution.
5 Orientation of Fibers During Thermoplastic Extrusion

Figure 5.2: Fiber orientation and interface to the PLA in a polished cut (left). Detail view of the interface between the fibers and the PLA matrix in a polished cut (right). This figure was published earlier in [164].

5.3 Results

5.3.1 Scanning Electron Microscopy

The polished cut through an extrusion line shows the interface between the carbon fibers and the PLA matrix in Figure 5.2 (left). The interface neither exhibits gaps between the fiber and the matrix nor cracks in the matrix. The detailed view in Figure 5.2 (right), nevertheless shows deformed and destroyed fibers. The bottom-right corner of the image shows a part of the matrix where the viscosity of the PLA melt exceeded the possibilities of filling the sharp concave corners.

Significantly stronger influence can be stated for holes resulting from the manufacturing process in the ME nozzle. The holes shown in Figure 5.3 (left) result from the thermal setting of the nozzle.

The holes show a characteristic oval shape when cut longitudinally to the fiber orientation and a round shape when cut orthogonally to the fiber orientation allowing the interpretation of an ellipsoidal shape with the major axis in the extrusion orientation. The same observation can be made in Figure 5.3 (right).

The characteristic interface between the produced extrusion vanishes due to the remelting of the matrix polymer during the extrusion and deposition process. Intersection of differently-oriented fibers between the layers is visible in the fracture surfaces of Figure 5.4 which, together with Figure 5.5, confirms again the orientation and size of the holes in the matrix material. Therefore, the tensile strength is reduced by the disturbed matrix material.

Figure 5.4 and Figure 5.5 show the characteristic fiber orientation longitudinally to the extrusion path. Confirming the common theory of the effect of fiber orientation
on tensile strength in FRP, the influence of the orientation on the fracture surfaces is shown by the high number of orthogonally oriented fibers missing in the fracture surface, whereas only 40% of the fibers in a longitudinal orientation towards the tensile strength were pulled out of the matrix at the fracture surface.

Fiber-matrix interface is significantly better for nylon and GF due to the better chemical interaction and different rheological parameters as can be seen in Figure 5.6 (left). Significant improvement can be gained by adapting the interface between the printed layers seen in Figure 5.6 (right), which is especially important for extruded materials with higher melting or glass-transition temperatures since the previous layer is required to melt or soften before another layer can be added.

5.3.2 Pull Test According to ASTM D638 IV

It can be seen from Figure 5.7 that the printing orientation of 45° gives the best performing output in terms of Young’s modulus as well as tensile strength, whereas the standard deviation of the modulus is significantly lower than of the tensile strength at a sample size of 7 specimen.

Comparison tests on untreated filaments showed a different behavior in terms of moduli, whereas interestingly the mechanical performance of nylon was turned out to be weaker than the one of PLA, which leads to the assumption that the manufacturing method is of significant importance for the mechanical performance of the final specimen.

For visual example, a sample was chosen in Figure 5.8 showing a stress-strain diagram of different specimen configurations. It can be concluded from the diagram that
Figure 5.4: Fiber orientation according to the print direction on a fracture surface after a tensile test; orientation of the fibers in the direction of the extrusion. This figure was published earlier in [164].

Figure 5.5: Holes in the PLA matrix on a fracture surface after a tensile test; variations in amount and size of holes at the different layers are visible (left). Detail of holes in the PLA matrix on a fracture surface after a tensile test; variations in orientation and size of holes at the different layers (right). This figure was published earlier in [3, 164].
5.3 Results

Figure 5.6: GF in a nylon matrix after a pull test experiment (left). Weak interface between printed layers at 45° printing orientation (right). This figure was published earlier in [3].

Figure 5.7: Compiled tensile test data according to ASTM D638 IV with Young’s modulus (EMod) and tensile strength (TS) with 12% wt and 30% wt in 0°, 45°, and 90° printing orientation. If not noted differently, the experiments were performed on nylon material.
fiber orientation (and therefore printing orientation) plays a significant role in the stress at break of the tested specimen as all tested mechanical parameters performed weakest in comparison to the other specimen.

5.3.3 Radiological Imaging

In spite of all efforts and using a state of the art high end human multi-detector CT, the resolution limit for resolving fibers or even distribution densities remained too high to draw any conclusions that concur with the high professional level we desire. At the present moment, no apparent potential for using human grade multi-detector CT in determining the mesoscopic structure of materials is in our grasp. However, this may change with the rise of multi-energy and multi-beam technologies. Cone Beam CT may provide a solution, as some manufacturers claim to provide devices with resolutions down to the order of about 10 μm on human (i.e. easily available) scanners.

Research grade micro-CT has the potential to provide deeper insight into the structure as shown in Figure 5.9. Clearly visible are fiber bundles, individual fibers and air inclusions in an extruded filament.
5.3 Results

Figure 5.9: Freely extruded filament (left), transversal (middle), and medial lateral (right) cut under CT investigation. This figure was published earlier in [2, 164].

CR is proved to be most effective for this application, mostly due to its sufficient resolution, simple setup and uncomplicated availability.

Notwithstanding being at the edge of resolvability and certainly with some degree of spatial aliasing, it was possible, after optimization using a carefully prepared honeycomb shaped geometry for the print with illumination lateral relative to the main axis of the honeycomb rhombic geometry, to work out details of the fiber bundle distribution. Further investigations using a different CT machine were conducted on VPP processed fiber-reinforced materials allowing for a much higher resolution and therefore more detailed information on fiber- and defect orientation. Results of these investigations can be found in chapter 8.
5.4 Case Study Example: Tape Placement System for Continuous Fibers

The connection of different layers has been discussed throughout this document and are necessary to enforce the peel-off resistance of the final part perpendicular to the printed layers. This is in particular important in the tape placement industry e.g. for aerospace or wind energy systems requiring a large amount of material and finalizing on the throughput of the system.

The manufacturing of fiber composite components today is limited to the manual layout of fiber mats or by automated fiber placement machines that require a pre-wet (pre-preg or pre-impregnated) fiber for the layout.

Figure 5.10 shows the construction of the system. A grated wheel is used to affix fibers (carbon, glass, or similar) through the wheel on the underlying substrate. The grates of the wheel allow for the matrix material dispensing system in the middle of the wheel to dispense (by droplets or by a spray) material out through the gratings and directly onto the fiber material. Hereby the fibers and matrix material is consolidated and laid out in one simple continuous motion as the wheel traverse the substrate.

Any surplus matrix material that may build up on the grated wheel is being collected in a drip-tray above the dispensing system in order to ensure a controllable process. This surplus material may be forced off the wheel and onto the drip tray by either an air-knife or by a mechanical wiper blade. A static mixer allows for a variation of the chemical composition during the printing process and is built in as a disposable part.

Chemicals are introduced on the side of the assembly as shown in Figure 5.11 and residual/waste material will be transported outwards by the bridge-like structure. The mounting plate on the top allows for additional cleaning devices such as brushes (with or without dissolving chemicals). The curing initiator and the oligomer are mixed together right in the static mixer.

Short fibers can additionally be introduced via the nozzle holder in the center of the wheel and provide additional strength perpendicularly to the orientation of the continuous feedstock. Due to the small surface applying the pressure on the surface, the fiber orientation will not be significantly changed.

Further advantages of the system can be comprised as follows:

- Self-cleaning as a cleaning system can be included on the top. A drain system above the nozzle transports the waste material (including a potential chemical solvent) out of the system into a drainage.
- Spring loaded grates in the harvesting wheel can allow for a better sticking onto the ground substrate.
5.4 Case Study Example: Tape Placement System for Continuous Fibers

Figure 5.10: Principle sketch of the tape placement roller system.

Figure 5.11: Prototype and full assembly of the tape placement roller system.
5 Orientation of Fibers During Thermoplastic Extrusion

- The chemical components are separately introduced via the center of the wheel and mixed in a static mixer, which is designed to be replaced after use.
- Automatic fiber retention allows for pre-stretching of the fiber from the pull-off spool.
- Since the fibers are not pre-wetted, it is possible to locally modulate the chemistry of the matrix material.
- The system is compatible with conventional roller systems (which is especially relevant for the feeding system of the fiber feedstock).
- Additionally, pre-wetted fibers can be added as thermal and UV curing devices can be added on the mounting plate.

The full set of drawings can be found in section E.1 and section E.2.

5.5 Case Study Example: Joule Pre-Heating During Material Extrusion

In advance to the pure application of fibers in an AM application, fibers can add functionality to the part in the sense of e.g. mechanical, thermal, or magnetic properties as described in Part I. These properties relate to the final part and are in usual ME or VPP applications not used during the manufacturing process. Investigations performed earlier and presented in [257, 258] concluded an increase of printing speed by a factor of 10 in an extrusion-based system by using pre-heating of the introduced filament.

A further project was performed in order to use the electrical conductivity of CFs in an ABS filament in order to pre-heat the filament before introducing it into a standard heater block in an ME nozzle. The electrical conductivity is given by the entanglement of the fibers in contact with each other, which is more likely for fibers rather than e.g. spherical filler materials.

The fact that the filament leaves the nozzle in a molten state allows to place the ground on the (conductive) nozzle, whereas the other pole of the current is placed on the filament via a brush normally used in electric motors. The electrical connection is only allowed via the filament, which requires the full insulation within the pre-heater. The feed as well as the moving system can be used similarly to conventional ME machines. The entire device was screwed together using ceramic screws.

A prototype shown in Figure 5.12 was build using Macor as well as Fish Paper as insulating material. The electrical conductivity of the material has proven to have significant influence on the temperature of the filament. Furthermore, additional challenges arise in the guiding of the pre-heated filament since the stiffness of the
5.6 Summary

Figure 5.12: Prototype (left) and simulation (right) of the full extruder utilizing both Joule pre-heating as well as conventional heating on the bottom.

Material is influenced by the temperature. For this reason, a guiding rod was utilized.

The relatively long heating phase of the filament has the potential to harm the properties of the matrix material and therefore should be avoided in future revisions of the design, as it additionally increases the required momentum transferred from the stepper motor (not visible in Figure 5.12) since the friction needs to be overcome.

The full set of drawings can be found in section E.3.

5.6 Summary

A conclusion can be drawn in terms of fiber orientation during the final part in strong relation to the printing orientation in the ME process. Due to the two cycles of extrusion of the material (1) when generating the filament from granule; and (2) during the AM process, fibers were oriented longitudinally to the printing path. The orientation of fibers therefore proves easier and more straightforward as compared to VPP technologies presented in chapter 4.

Ellipsoidal holes with the major axis in the nozzle orientation occurred in the PLA matrix with average sizes of 50 to 100 μm possibly resulting in a reduction of tensile strength. Compared to FRP in VPP, the size of the holes was significantly increased as is further discussed in chapter 8. The distribution of fibers was therefore also influenced whereas the fibers were generally oriented in the middle of the extrusion line.

The interface between the layers was indistinct due to the remelting of material during the extrusion of the next layer allowing for a continuous matrix material. This also resulted in an interconnected orientation of longitudinally-oriented and orthogonally-oriented fibers.
The interface between the fibers and the PLA matrix was destroyed during the tensile-test. This was fatal at the layers with orthogonal fiber orientation towards the tensile strength. 40% of all fibers in a longitudinal orientation towards the tensile strength were ripped out of the matrix at the fracture surface.

A further line of investigation, would be to proceed in investigating radiography of thin cut slices in order to minimize projective geometry and partial volume artifacts.

Tensile tests of nylon matrix material with GF filler material have proven slight increases in the mechanical properties of the final parts (dogbones) regarding the printing paths. It was shown that the tensile strength of a raster 45° printing orientation had the highest levels.

Furthermore, a tape placement device was introduced combining the advantages of traditional automated tape placement with AM technologies allowing to formulate the resin in-situ and therefore also introduce short fibers into the material to strengthen the inter-layer connections and therefore reduce the risk of peel-off effects at failure. This effect can prove necessary in critical applications such as wind turbines and aerospace wings.

Another case study was presented elaborating on the Joule heating effect during ME processes where the electrical conductivity of certain filament compositions (e.g. filament with CF filler material) allow a pre-heating of the filament before entering the nozzle with conventional heating mechanisms. The technique can allow for faster ME printing.
Part III

Industrial Application:
Fiber-Reinforced Injection Molding
Inserts
6 Lifetime and Surface Topography Development

This chapter investigates the lifetime and surface deterioration of additively manufactured, injection molding inserts. The inserts were produced using digital light processing and were reinforced with oriented short carbon fibers. The inserts were used during injection molding of low-density polyethylene until their failure. The molded products were used to analyze the development of the surface topography and wear. By enhancing the lifetime of injection molding inserts, this work contributes to the establishment of additively manufactured inserts in pilot production.

The content of this chapter was taken and in parts modified from the following publications:


6 Lifetime and Surface Topography Development

6.1 Introduction

The possibilities of AM for IM were already pointed out in a number of both academic and industrial publications and their engagement towards the public has been increased in recent years. However, the effect of fibers for the lifetime and surface deterioration of the IM inserts was not yet extensively investigated, although significant advantages at the cost, environmental, and efficiency levels can evolve.

FRPs in AM show a directional placement of the fibers within the manufactured layers allowing the reinforcement of the IM insert in two directions further elaborated in chapter 4. The lifetime of additively manufactured inserts made from photopolymer using VPP is located below those of inserts made from brass or steel and, therefore, the lifetime in terms of surface quality was the subject of this investigation.

This research contributes to the development of new technologies for IM inserts reducing production costs as well as the environmental impact of prototyping and proof-of-concept manufacturing. It was pointed out in [261] that composite materials for IM inserts made from polymer and copper particles improved the heat conductivity of the inserts thereby increasing the lifetime of the inserts. This investigation was developed further using short CFs.

6.1.1 Concept

AM as a technology for digital manufacturing in an industry 4.0 context has been introduced earlier in this thesis and bears a significant potential for advanced IM production and prototyping (see section 2.3.4). A number of examples are available in scientific publications as well as offered by small and medium enterprises (SMEs) as a service. The initial research presented in this context is in parts extending the research performed in [262] by Michael Mischkot. Further concepts have been developed and fiber-reinforcement has been added to the parts.

The interest of developing AM inserts for the purpose of IM is based on the following factors:

**Lead time** AM and RP allow for a fast production of the printed part in a matter of hours until a print is finished. Printing times are constantly reduced as new technologies are developed (e.g. [263]). Not taking this latest development into account, it was concluded in [264] that the lead time of IM based on AM technology can be reduced by 60 to 70% allowing for faster developments in a flexible market. This accounts especially for complex forms with small features requiring a more intense tooling process, while VPP is simply constrained by the height of the printed part. Moreover, multiple inserts can be printed at the same time in the same machine reducing the time per insert significantly.
6.1 Introduction

**Costs** Reducing a number of required steps that are usually necessary for conventional manufacturing by the use of tooling, cost factors such as human labor as well as material costs are reduced resulting in a cost reduction of 80 to 90% according to [264].

**Freedom of design** This aspect separates in two sub-factors: (1) Especially concave corners are limited in their accuracy during tooling by the diameter of the tool, while VPP is limited by either the laser diameter or the voxel size of the projection unit. In either case, the complexity of the part does not play a significant role during the AM process. This aspect is further described in [265]. (2) With regards to cooling, an important aspect that is described further in this chapter, the freedom of design in AM allows for additional cooling of the inserts and therefore also allows for more complex designs as has been elaborated in chapter 7. Dissolvable molds have been used by the company Addifab® allowing for novel geometries that were not moldable beforehand.

**Environmental aspects** Since the use of metal is reduced, but the use of chemical substances is increased in the production of inserts, aspects of environmental impact need to be considered carefully and are further evaluated in chapter 9. Especially the aspect of recycling of metal and chemical waste has become an issue since standard operating procedures (SOPs) are missing.

**Protection of IP** While digital manufacturing allows for sharing of e.g. CAD data, this might not be an option due to intellectual property (IP) right protection. The disposing of molds that were removed from production with the aspect of IP protection is a significant issue and requires special precautions such as destroying the mold in order to avoid reverse engineering. Given that IM inserts produced by AM are manufactured from polymers, this aspect has become easier but is not discussed further in this thesis.

Fiber-reinforcement has enabled a larger number of shots per insert and therefore has also enabled a further development of the technology. Initial tests were performed on geometries introduced in [262]. Thereafter, a new geometry suitable for micro-IM feature observation has been developed as described in Figure 6.1. The development phases are further described in section 7.1 followed by a thermal process simulation. The geometries are described in Appendix F.

### 6.1.2 Insert Generation

The IM inserts were produced using VPP from a photopolymer resin with 0% wt, 5% wt, and 10% wt short CF content equipped with an average diameter of 7.2 μm and an average length of 100 μm. The layers were placed perpendicular to the expected pressure tensor from the polymer melt from the IM process resulting in a fiber placement in the manufacturing layers. The back and sides of the inserts were milled to reduce warpage of the inserts and increase the accuracy of the mold assembly.
6 Lifetime and Surface Topography Development

Figure 6.1: Development phases of IM inserts. a) and b) represent sub-phases and refinements of the development phase. This figure was published earlier in modified form in [4, 232, 259, 260].

6.1.3 Molding and Industrial Application

The inserts were used during manufacturing with an IM machine e-motion 110 by the company ENGEL injecting PE-LD as well as ABS at 350 bar maximum injection pressure during a 1.5 s filling time followed by a 7 s packing time, 1.5 s opening and ejection, and 10 s cooling phase for the insert in order for the insert to cool down to a temperature of 50 °C. The insert dimensions were (20 × 20 × 2.7) mm³ for the small geometry and (80 × 60 × 10) mm³ for the multiscale geometry. The total cycle time of the molding was 20 s and is graphically represented in Figure 6.2.

The additively manufactured insert was built into a multi-functional frame in the IM machine as shown in Figure 6.3 (left). The liquid polymer was injected from the reverse side of the insert and through channels guiding the polymer to the mold. The final part before ejection can be seen in Figure 6.3 (right). Other geometries were used during case studies and are described in the corresponding chapters.

6.2 Methods

A SEM JEOL JSM-5900 was used showing the deterioration of the surface during the molding process. A Dino-Lite Pro AM 4000 digital microscope was used to inspect the surface of the inserts and the parts after the molding process.
6.2 Methods

Figure 6.2: Graphical representation of the molding cycle. The cooling times before and after ejection of the part can be considered the largest potential for cycle time reduction (left). Final part shot at too low injection pressure in the configuration shown in Figure 6.3 (right). This figure was published earlier in [259, 260].

Figure 6.3: Single insert in the IM machine before the first shot (left) and PE-LD parts arranged in the IM machine (right). This figure was published earlier in modified form in [2, 137, 178, 259, 260, 266, 267] as well as by other authors with and without citation.
Filling experiments in the IM machine were performed in order to achieve the best possible output of parts. The temperature at ejection was configured to 80 °C, whereas the machine was put on hold afterwards in order to let the insert cool down to 50 °C before another injection was started as elaborated in Figure 6.2. The parts were collected, sealed from further contamination or damages to the surface, and investigated without further processing e.g. regarding surface or heat treatment.

Tests of the surface structure in terms of roughness were performed using an Alicona® focus variation 3D microscope system with a vertical resolution of 500 nm and a lateral resolution of 3 μm. The scanned surface point was placed in the middle of the insert in order to achieve comparable measurements over the entire insert lifetime. The insert itself was not tested further in the focus variation microscope.

The data of the scanned surface was thereafter aligned using global leveling for the inspected areas taking the surface average as leveling base. It can be expected that no errors in the surface roughness analysis were caused by this procedure since the roughness is strongly determined by the peek-to-peek level of the surface rather than the angle of the surface profile walls. The mean surface roughness was thereafter determined neglecting already existing cracks on the surface. Measurements were performed on the final parts since the main interest in industrial processes is placed at the part level rather than the process level.

6.3 Results

6.3.1 Lifetime and Cracks

During this investigation, the observations presented in subsection 3.3.1 concerning outstanding CFs at the border of the object were confirmed as the first manufactured parts showed residual CF material in the upper layer of the part. The outstanding fibers broke off after the first shot and stuck to the produced part. A similar effect of outstanding fibers was detected during tensile tests of thermoset matrix material in combination with GFs and is further described in subsection 3.3.1 and subsection 3.3.3. The effect of outstanding fibers, however, is caused by the curing mechanism since the matrix is cured by light (or heat in other examples) and therefore only effects the photopolymer in certain regions, while fibers are randomly oriented in the entire resin.

No residuals of broken fibers were found at the second shot and later. The outstanding fibers in the original IM insert are shown in Figure 6.4. It shall be noted that no fibers were standing perpendicular to the top surface layer since the part was post processed in order to remove warpage that might affect the lifetime due to thermal or mechanical stresses in the part.
Figure 6.4: Fibers standing out of the produced layers under SEM investigation confirming the findings presented in subsection 6.1.2. The image was taken before the first shot. The raster pattern on the surface results from the VPP process and the projector resolution. This figure was published earlier in [178].
Surface investigations of produced parts showed cracks of the insert in the μm regime after about 300 shots leading to a change in roughness of the surface of the manufactured part. Compared to other tests on photopolymer inserts without fiber-reinforcement, the enlargement and lengthening of the cracks were significantly reduced allowing a continued production up to 4500 parts.

The crack propagation had an average velocity of $(0.145 \pm 0.034)$ mm/100 shots after the first sign of the crack until the failure of the part. Figure 6.5 shows the averaged crack propagation velocity over time showing an increase of the velocity up to 0.19 mm/100 shots followed by a decrease until failure of the insert. The crack propagation is visualized in Figure 6.6 showing the first cracks at shot 500. Crack 2 is blocked by crack 1 after shot 1300 and therefore does not propagate any further. Note also the degradation of the edges, as can especially be seen in the round parts on the left side of the figures. Crack 3 also propagates on another level on the lower part after shot 1300.

Although the surface of the insert was cleaned with isopropyl alcohol after the post processing of the print, further changes in the reflection of the surface appeared over the course of the first 300 shots e.g. penetrating from the right in Figure 6.6. It can be concluded that these changes happened due to either thermal or abrasive effects caused by the injected polymer. Investigations presented in chapter 8 furthermore showed small surface cracks connected to the injection direction of the polymer in the machine causing changes in the surface over the course of the entire lifetime.

**Figure 6.5:** Crack propagation velocity over the number of shots (averaged). This figure was published earlier in modified form in [178].
Figure 6.6: Three exemplary cracks in the insert after shot 50, 200, 500, 1000, 1500 and 2000. This figure was published earlier in modified form in [1, 178].
Cracks of the insert were reduced to surface cracks as shown in Figure 6.7 after 2658 shots with low propagation speed and gap size in the range of 1 to 5 μm. Most cracks originated from the edges in the surface of the insert as shown in Figure 6.7. Those were degrading in such a way that the edges became round as can be seen in Figure 6.8.

Figure 6.7 includes parts of the surface protected against thermal and mechanical strain. No cracks evolved in the protected part of the surface and no cracks originated at its border. The cracks were reproduced in the molded part in the form of flashes for low gap sizes. At larger shot numbers, PE-LD got stuck in the gap as shown in Figure 6.8 (right) as a detail of Figure 6.7 (left) and therefore caused severe damage to the produced part. The insert was then characterized as destroyed.

An average number of shots of 2580 was gained using the fiber-reinforced inserts with 5%wt and 4500 shots with 10%wt. Compared to the non-fiber-reinforced inserts, the experiments resulted in an increase of the number of shots by a factor of 10. Crack propagation was reduced to 1.25% of the velocity in the plain insert.

### 6.3.2 Surface Texture and Roughness

A graphical representation of the surface of an insert in Figure 6.9 shows a central surface area with respect to height variations and roughness of the surface. The figure allows conclusions about scratches that were produced during the milling and polishing of the insert before the first shot. These features were not visible at other states over the lifetime of the insert.
6.3 Results

**Figure 6.8:** Part surface and rounded edges after 2658 shots (left). Detail of surface degradation with PE-LD material stuck in the crack (right). This figure was published earlier in modified form in [178].

**Figure 6.9:** Graphical representation of the surface topography after 10 shots showing the profile under a focal variation 3D-microscope in the middle of the insert. This figure was published earlier in modified form in [178].
Surface roughness over multiple shots allows the conclusion that the changes in the general surface roughness can be neglected. The average surface roughness was calculated over 7 inserts with respect to the first 1000 shots (see Figure 6.10). The figure does not show a significant increase in surface roughness. Given the relatively large standard deviation, the conclusion can be drawn that the influence of surface wear can be neglected.

The analysis of the surface degradation showed the impact of the fibers within the polymer by reducing the speed of crack propagation within the insert allowing for the running of shots producing parts with a smooth surface appearance although cracks were already present in the insert. The lifetime was investigated by the consideration of cracks on the surface or throughout the entire insert. Cracks producing a flash that was clearly visible without magnification were considered a failure of the insert.

It could be shown that the crack propagation was reduced by inserting the fibers in a layer standing orthogonal to the crack orientation. In this way, the fibers contributed to better mechanical properties in terms of strength and durability. The cracks were not influenced by the surface structure that was applied by the post processing after the printing process, however the change in topology mentioned earlier in connection to the injection of molten polymer did not affect the surface roughness.
6.4 Summary

Figure 6.8 (right) allows for the conclusion that the surface topography is strongly influenced by the heat introduced onto it, since there are no sharp features except for the crack lines (which has not been investigated further). While not proven, a valid argumentation can be the melting of smaller parts of the surface of the insert on the top surface effecting the topology in a smoothing way. The features are in the range of 5 to 20 μm in diameter and therefore do not affect the purpose of prototyping in an industrial environment.

It can, however, be argued that optical effects on the surface such as e.g. roughness, fluorescence, or reflecting effects will be influenced at a later stage of the insert lifetime. The effect of reduced molding cycle time as described further in chapter 7 on the above described effects of surface melting has not been investigated further.

In that sense, the surface degeneration is placed in an acceptable range for prototyping applications, but the lifetime of the insert is far from acceptable for production environments.

6.4 Summary

The lifetime of AM inserts was initially a significantly influencing factor of the process chain and has determined the usability of the technology as prototyping assistance in the IM industry. Adding 10 %\textsubscript{wt} of CFs to the thermoset photopolymer matrix has the effect of a reduced crack propagation. While non-reinforced inserts had to be discarded a few shots after the initial cracks appeared on the surface, crack propagation was reduced to 1.25 %. The nature of crack propagation within the IM inserts was investigated further and is described in chapter 8. The cracks slide along the fibers, but are blocked when penetrating the fiber from the side.

It can be concluded that the lifetime of fiber-reinforced IM inserts could be extended compared to plain IM inserts. The surface wear in terms of mean surface roughness was negligible when inspecting the surface without crack-like features. This development opens up the possibility to increase the size of the inserts in order to allow for other applications to be fulfilled. While an increased insert volume has a higher risk of failure due to thermal stresses and cracks, the thermal management of the insert has to be determined as well as the cycle time of the part production has to be considered. An example of an industrial application is given in chapter 7 showing the challenges and possibilities of thermal management of the insert.

Minor cracks on the surface appeared early from 300 shots but did not propagate to major cracks, and therefore did not result in fatal failure of the entire insert. Propagation of major cracks through the entire insert were found to spread slower compared to plain inserts. Thus, it can be concluded that the lifetime of the insert was increased by generating a composite using short CFs.
Additively manufactured, fiber-reinforced inserts can be considered suitable for pilot production with low part numbers, and therefore are an alternative to more expensive inserts made from brass or steel. Furthermore, advantages in terms of lead time, costs, freedom of design, environmental aspects, and protection of IP have to be considered. The environmental aspects of additively manufactured IM inserts have been investigated further in chapter 9 showing that the lifetime has a significant influence on the impact of the insert material on factors such as climate change and toxicity.
7 Process Simulations of Thermal Mold Behavior

While the lifetime compared to conventional materials such as brass, steel, and Al, is reduced, the prototyping and design phase can be shortened significantly by using flexible and cost-effective additive manufacturing technologies. Higher manufacturing volumes still exceed the capability of additively manufactured inserts, which are overruled by the stronger performance of less-flexible but mechanically stronger materials.

This chapter discusses the heat transportation within the inserts made from a thermoset material, brass, steel, and ceramic material. It therefore elaborates on the possibilities of injection molding as well as the thermal challenges connected with the use of polymer inserts. They are an essential part for further calibrations of the injection molding process.

The content of this chapter was taken and in parts modified from the following publications:

7 Process Simulations of Thermal Mold Behavior

7.1 Introduction

Over the past years, several scientific investigations have been conducted in order to improve the lifetime of AM IM inserts starting from the elementary manufacturing without reinforcement based on VPP processes of photopolymers (PhPs) [268]. The proposed development phases are illustrated in Figure 6.1, whereas phase 1 and phase 2a for inserts with small dimensions were already experimentally investigated and showed a statistically reproducible lifetime improvement by a factor of 10 (see chapter 6).

Further developments shown in phase 3 and phase 4 include the advancement of the IM inserts into multiscale in the cm-range using the advantage of VPP of producing micro-features without further processing complexity. Phase 4 circumvents the heat transfer challenges between non-welded solid materials by a cooling fluid flow.

All of the development phases were covered in numerical simulations for multiple IM cycles predicting challenges and possibilities of further development phases.

7.2 Methods

The numerical simulations were conducted using the Comsol Multiphysics® version 5.3 modules for laminar fluid flow and heat transfer as well as the multiphysics model for non-isothermal flow. The model was driven by the Navier Stokes equations and heat transfer:

\[ \rho (u \cdot \nabla) = \nabla \cdot \left[ -pI + \mu (\nabla u + (\nabla \cdot u)^T) - \frac{2}{3} \mu (\nabla \cdot u) I \right] + F \]  
\[ \nabla \cdot (\rho u) = 0 \]  
\[ \dot{q}_V = \rho c_p \frac{\partial T}{\partial t} - \nabla \cdot (k \nabla T) \]
7.2 Methods

<table>
<thead>
<tr>
<th>Part</th>
<th>Material</th>
<th>phases</th>
<th>Ti with ABS in °C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conventional Mold</td>
<td>Steel</td>
<td>1-4b</td>
<td>32.2</td>
</tr>
<tr>
<td>PhP</td>
<td>1-4b</td>
<td>121.0</td>
<td></td>
</tr>
<tr>
<td>Brass</td>
<td>1-4b</td>
<td>30.0</td>
<td></td>
</tr>
<tr>
<td>Steel</td>
<td>1-2b</td>
<td>32.2</td>
<td></td>
</tr>
<tr>
<td>SiC</td>
<td>1-2b</td>
<td>27.4</td>
<td></td>
</tr>
<tr>
<td>Injected Material</td>
<td>ABS</td>
<td>1-4b</td>
<td>n/a</td>
</tr>
<tr>
<td>Cooling Fluid</td>
<td>Water</td>
<td>2a-4b</td>
<td>n/a</td>
</tr>
</tbody>
</table>

Table 7.1: Thermal material properties of the simulated materials in comparison.

where

\[
T_i = \frac{b_1 T_1 + b_2 T_2}{b_1 + b_2} \quad (7.4)
\]

\[
b_i = \sqrt{k_i \rho_i c_{P_i}} \quad (7.5)
\]

Materials were chosen according to Table 7.1: PhP, brass, steel, silicon carbide (SiC). The thermal material properties vary significantly among each other and therefore are compared in Figure 7.1.

The interface between the flowing and cooling injected material ABS was approximated by applying effusivity between the materials according to the following equations (all units were considered according to SI): [269]

\[
T_i = \frac{b_1 T_1 + b_2 T_2}{b_1 + b_2} \quad (7.4)
\]

\[
b_i = \sqrt{k_i \rho_i c_{P_i}} \quad (7.5)
\]

where

\[
T = \text{temperature at interface}
\]

\[
k = \text{thermal conductivity}
\]

\[
\rho = \text{density}
\]

\[
c_{P_i} = \text{heat capacity}
\]
The cycle was considered as 1.5 s injecting, 7 s packing, 11.5 s mold opening and natural cooling adding up to a cycle of 20 s. The fluid flow was approximated as constant during the entire cycle with 5 L/min (development phases 2a, 2b, 3a, 4a) and 10 L/min (development phases 3b, 4b). The ambient temperature was approximated with 25 °C in infinity. The fluid cooling was considered with a constant inlet temperature of 15 °C over the entire cycle. Surfaces were cooled by natural convection when exposed to the surrounding.

### 7.3 Results

The high interface temperature due to the effusivity of the PhP cooling required a higher heat transfer into the mold and surroundings visualized in Figure 7.2. The conventional mold showed only a slight temperature increase, however it needs to be considered that the overall heat capacity of the steel mold is significantly greater than the inserts. An influence of the inserts on each other was therefore not detected, which allows for further prototyping techniques e.g. a reference insert while the other inserts are changed during the prototyping development.

Investigations confirmed the crucial importance of advanced cooling mechanisms such as para-conformal cooling, where the standardized mold geometry is modified by techniques available for conformal cooling providing a uniform surface temperature and therefore reducing thermal stresses. The amount of heat removed from the
system by the cooling liquid shown in Figure 7.3 has been increased from 2.9 kW to 5.7 kW after the change from conventional cooling to para-conformal cooling. This factor reaches significance as insert and part volumes increase while at the same time utilizing the freedom of design that comes with AM techniques.

In all development phases, the PhP insert showed significantly higher temperatures (Figure 7.4), which can be explained by the higher interface temperature as well as the smaller conductivity of the polymer. Moreover, the interface between the inserts and the conventional mold allows for a higher heat transfer when pairing two metals, as they are not welded.

Figure 7.5 elaborates on the potential of development phases 2b–4b. It is also shown that the cooling mechanism in the higher development phases provides a stronger heat transfer. Para-conformal cooling molds applying the advantages of AM in terms of pressure reduction, e.g. by rounded edges, allow higher throughput of the cooling fluid, which leads to a further temperature decrease. This is however not necessary for the metal and ceramic insert materials as their possible cycle time is decreased as compared to PhP.

It is moreover shown that an increased absolute heat capacity consequently increases the insert temperature whereas the temperature of the cooled inserts is significantly lower than the temperature for simple para-conformal cooling. The higher absolute heat capacity can therefore be compensated.

Concerning the development phase 4 inserts, it will be required to consider thermal stresses in the layers between the cooling channels and the injected ABS material.

Figure 7.2: Temperature of one mold side. The right figure shows transparent features in order to see the cooling system. This figure was published earlier in [232, 259, 260].
Figure 7.3: Temperature of conventional (left) and para-conformal (right) cooling channels at a throughput of 5 L/min. Due to the improved flowability of the fluid in the channels with rounded corners, a higher throughput of up to 10 L/min is possible. This figure was published earlier in [4, 232, 260].

By the use of enhanced cooling, the overall temperature of the insert was reduced (as shown in Figure 7.4 and Figure 7.5), which allows a deduction of the natural cooling time by 6s while at the same time start the molding cycle with the same insert temperature. The use of larger insert geometries corresponds to longer cooling times, which can be reduced by an advanced cooling of the standardized steel mold in the background.
Figure 7.4: Cooling of the surface of the insert with standard cooling. Ejection of the molded part appeared after 8.5 s (top). Cooling of the surface of the insert with para-conformal cooling. Ejection of the molded part appeared after 8.5 s. The improved cooling allows for a shorter molding cycle as the temperature of the PhP insert reaches lower values more quickly (bottom). This figure was published earlier in [4, 259].
Figure 7.5: Difference to standard cooling of PhP inserts (top). Difference to standard cooling of brass inserts (bottom). This figure was published earlier in [4, 259].
7.4 Case Study Example: Industrial and Topology Optimization Mold

A case study example of a specific industrial application was performed utilizing active cooling of the additively manufactured insert. This research was conducted in cooperation with an industry partner and all non-disclosure agreements were waived for this section. Any geometrical or other data that might reveal proprietary data was changed in the experimental setup in such a way that the output of the experiment was not affected, but no conclusions towards trade secrets can be drawn.

The insert shown in Figure 7.6 and Figure 7.7 was used in an industrial setup transporting water as cooling liquid through the insert at a rate of 8 L/min. Thermal measurements using a thermistor were performed inside the insert with and without an active cooling flow.

The measurements showed an average temperature reduction of 5 °C whereas the peak-to-peak difference was not changed significantly. The technology is therefore capable to remove large material volumes and therefore reduce the total overall heat capacity.

Methods such as conformal cooling, which are already state of the art for a number of years in metal AM, are a natural reaction to the problems that process engineering is confronted with in polymer AM inserts. A further improvement concept is presented below where digital technologies are used in order to improve the cooling mechanisms further.

Topology optimization as such can be used in order to remove the density of the back of the insert generating room for freely flowing cooling water as shown in Figure 7.8. The soft tooling mold consists of a shell equipped with a cavity. The back of the insert is in a first step completely removed in the CAD file and then built by e.g. topology optimization in order to withstand and compensate for the injection pressure. The topology optimized back can then be flooded with cooling water. As the effective diameter is bigger, the flow rate can be increased, and therefore the cooling rate is as well. Warpage can furthermore be reduced by this approach as has been shown in similar setups and advanced cooling is discussed in [270].

The proposed advantages of the underlying system are as follows:

- Actively cooled inserts cool faster. However, stresses are introduced as some parts of the insert are cooled and others are not cooled to the same extent. With this approach, the part is constantly cooled over the entire surface.
- Induced stresses due to the ABS injection are equal everywhere since this can be accounted for by the topology optimization.
- No bigger thermoset photopolymer parts are present that heat up the inside.
Figure 7.6: Graphical representation of the insert with active liquid cooling inside the insert.

Figure 7.7: Back and front of the industrial application insert with temperature sensor mounted in the back of the insert.
• Photopolymer is expensive and charged not only by the z-height of the part, but also by volume.

• The mold can be generated individually and digitally, and the process can be automated.

• The molding cycle can be significantly reduced as the mold cools faster. Therefore the molded part cools faster and can be ejected earlier.

• Better cooling allows for bigger geometries of the IM inserts.

7.5 Summary

Mechanical properties play an important role in parameters for further industrial production when directly influencing the molding cycle time. While the lifetime of the inserts has been significantly improved, other factors such as molding cycle time need to be considered. For further information on the performed investigations on lifetime improvement of AM IM inserts, chapter 6 can be referred to. Conventional metal materials are equipped with a significantly higher thermal conductivity and therefore propagate the heat into the back of the mold where it is mostly dissipated through the cooling liquid.

With the absence of this cooling mechanism, heat radiation on the surface of the insert becomes more important. Nevertheless, cooling can be improved by increasing the cooling flow. This can e.g. happen by a higher performance cooling in the form of para-conformal cooling channels, which requires the conventional back mold to be built by AM as well.

AM inserts, however, allow for additional cooling by the use of a cooling flow running through the insert as such. This effect has also been tested in an industrial case.
study. Since the natural cooling time has been reduced by over 50\%, they can be considered a sustainable method for future production in a smart, digital fabrication environment.

While the effusivity of the polymer insert material increases the cooling time, additional cooling reduces the molding cycle time significantly. In conclusion, this allows for an increased production volume. At the same time, the surface roughness of the insert does not essentially react upon the molding pressure and temperature cycle, which gives another advantage over the conventionally used materials. Other advantages of FRP IM inserts manufactured by AM are given in chapter 6 where a surface roughness analysis is also presented.

The findings presented in this chapter allow for the further development in phase 3 where (1) geometrical scales are significantly larger, (2) cavities and therefore the volume of injected polymer are bigger, (3) the contact surface between the injected polymer and the insert is increased, and (4) warping due to the VPP process is (a) increased in the process and (b) more significant due to the larger geometries.

All factors result in higher stresses due to thermally introduced internal stresses, and stresses due to limitations of space in the standardized mold, which need to be accounted for by the manufacturing and post-processing of the insert. The experiments used a polishing post-processing to remove the warpage on the back of the insert, which increased the lifetime significantly.

Furthermore, an industrial case study and further development possibilities of digitally manufactured inserts was presented allowing for improvements in the combination of numerical methods such as topology optimization as well as optimized production environments. The innovation requires a different understanding of cooling mechanisms and the use of cooling liquid without constraining walls of channels.
8 Internal Fiber Structure and Crack Propagation

In this contribution, the internal structure of a high-performing, fiber-reinforced injection molding insert was analyzed. The insert reached a statistically proven and reproducible lifetime of 4500 shots. Computer tomography, and tensile tests were performed in order to provide an understanding of the internal structure of the fiber-reinforced, additively manufactured injection molding inserts.

The content of this chapter was taken and in parts modified from the following publications:


8.1 Introduction

A significant challenge mentioned in several investigations in this thesis was introduced by the layer-wise orientation of CFs due to process parameters including build plate movement and print orientation, as well as flow patterns. While a theoretical approach was conducted in chapter 4, this contribution investigates the end-of-life (EoL) parameters of a used IM insert, which was kept in the production cycle twice as long as the usual lifetime. Data analysis was conducted using X-ray computed tomography (CT) in order to gain fiber orientation, fiber volume fraction, crack orientation, crack length, width, depth, and air intrusions.
This study therefore contributes to a further understanding of IM inserts produced by additive manufacturing, which comes with major advances in flexibility, cost efficiency, cycle time, and micro-features. The enhanced lifetime reduces the production cost share as well as the workforce required to change defect inserts. Moreover, the suitability for thermally more pretentious materials is increased (see chapter 9).

### 8.2 Methods

The AM inserts have been manufactured according to subsection 6.1.2 using the commercially available photopolymer resin HTM-140v2 and a bottom-up machine in a “hanging” configuration meaning that the printed layers were oriented in parallel to the longer edge of the rectangular feature at a layer height of 20 μm. The light exposure was increased by 20% in order to compensate for the light extinguishing of the 10%wt of CFs. This value changes with different configurations of fiber ratio and layer height.

After production, the inserts were then used in an IM machine running cycles until reaching twice the average lifetime of comparable inserts. One insert was selected for further investigations.

X-ray tomograms of the sample were obtained using a ZEISS XRadia 410 Versa device. The instrument was operated at 40 kV and 10 W using the low energy 1 (LE1) filter belonging to the system, and either a macro objective (LFOV) or a 4X objective. Three different configurations were determined according to the description in Table 8.1 based on chosen positions shown in Figure 8.1. Image reconstruction and beam hardening correction were performed using the built-in acquisition and reconstruction software package provided by ZEISS, while for further analysis and visualization the software “Avizo9.4.0” (FEI) was used.

The software was used to determine the length of cracks, to obtain pore size distributions in different sub volumes and to determine the volumetric amount of CFs. The following procedure was used to examine the volumetric amount of fibers: 1) definition of a sub volume inside the material not covering regions affected by artefacts; 2) interactive thresholding using a threshold of 42 000 cts (unit of intensity) for fibers, 3) removing of spots smaller than 10 px; 4) using the “material statistics” module to obtain the volume of the thresholded material. To obtain the full volume, the threshold in step 2) was set to 1 cts and step 3) was skipped. In order to obtain the pore size distribution, the “porosities analysis wizard” provided by the software was used. A mask threshold of 38 120 cts was used for the material, while the initial value to detect strong voids was set to 38 385 cts. Neither a threshold for weak voids, nor areas of material without volumes were defined, and no separation of voids was performed. The pore radius was calculated from the pore volumes. The fiber orientation was determined using Matlab scripts based on [271].
### Position 1
For overview measurements, image acquisition was performed using the LFOV objective and 1601 projections with 2 s exposure time, which resulted in a total measurement time of 1.56 h min. A pixel size of 23 μm was obtained using a 2x2 binning.

### Position 2
Using a 4X objective, a pixel size of 4.0 μm was obtained with 2x2 binning and tomograms were recorded with 3201 projections and 10 s exposure time resulting in 10.27 h min total measurement time.

### Position 3
The highest resolution measurements were acquired using the 4X objective and modifying the sample-to-detector distance, which resulted in a pixel size of 2.0 μm. The exposure time was adapted to 45 s and the total acquisition time was 42.44 h min.

**Table 8.1:** Measurement descriptions for positions according to Figure 8.1 (right).

---

**Figure 8.1:** IM insert after twice the usual lifetime (left). Measurement positions for the CT analysis (right). This figure was published earlier in [137].
8 Internal Fiber Structure and Crack Propagation

8.3 Results

8.3.1 Bottom-up Printing

The fibers are described as vectors oriented in space and $\Theta$ describes the elevation and $\Phi$ the azimuth angle. The fibers are predominantly oriented perpendicular to the azimuth plane (along 90° elevation ($\Theta$)), and less oriented in $\Phi$. This finding supports the assumption that the fibers are oriented along the printing direction, which is vertically to the surface containing the depletions in Figure 8.1 (left) meaning that the insert has been printed in a “hanging” orientation in a bottom-up printing process. This printing technique allows for a higher part per machine ratio but requires a layered printing (unless e.g. an oxygen permeable membrane is used). The orientation was sampled in a spherical coordinate system whereas $\Phi$ represents the orientation within the printing plane (Figure 8.2, left) and $\Theta$ represents the orientation outside the printing layer (Figure 8.2, right).

Crack orientations and dimensions have been analyzed and are shown in Figure 8.3 and Figure 8.4. The surface cracks have a tendency to orient randomly, but do not penetrate deeper than a few μm.

Figure 8.5 (left) furthermore shows a concentration of small surface cracks on the left of the insert. With consideration to investigations presented in chapter 6, the crack concentration is most likely to origin from the injected polymer and thereafter resulting temperature cycles. Since the polymer was injected from the right side of the insert, peek-to-peek values of the different temperatures were higher and furthermore the velocity of the injected polymer was higher resulting in higher surface wear as compared to other surface regions in the opposite part of the insert.

Deeper cracks up to 1.76 mm length are present in between the printed layers. These cracks propagate deeper and longer through the insert. The reason for this behavior is the missing connection between the layers reducing the crack propagation velocity as investigated during the IM process described in chapter 6. As an example, measured surface cracks are shown in Figure 8.5 (left) showing a smaller penetration of the insert than aligned cracks shown in Figure 8.4.

Solutions to prevent this behavior are discussed in chapter 4 and include a change from bottom-up printing technology to top-down printing technology with advanced build plates, which are permeable for fiber-filled resins. This can e.g. be achieved by equipping the build plate with holes which significantly change the fluid orientation and subsequently the fiber orientation within the resin, which is later represented in the final printed part.

Adhesion between the printed layers poses a major problem for both VPP as well as ME processes and are enforced by the use of fibers that are oriented along the printed layers, since they strengthen the part within these directions. Enforcing the
Figure 8.2: Fiber orientation analysis for position 1, 2, and 3. As suggested in [2, 272], fibers are oriented along the printed layers. This figure was published earlier in modified form in [4, 137, 232].
Figure 8.3: Cut through position 2. Cracks are mainly oriented in between the printed layers. This figure was published earlier in [137].
8.3 Results

Figure 8.4: Overview on deeper cracks in terms of orientation and dimensions through position 2, which align in general with the printing layer orientation. This figure was published earlier in [137].
8 Internal Fiber Structure and Crack Propagation

Figure 8.5: Projected view with surface crack orientation (left). Surface cracks originate from the steep temperature gradients during the IM process. Cracks are mainly oriented within the printed layers. Detailed view on deeper cracks through position 1 of the middle picture (right), which align in general with the printing layer orientation. This figure was published earlier in [4, 137, 232].

part in the direction perpendicular to the printed layers requires the following further elaborations:

**Fibers perpendicular to the printed layers** This topic is discussed further in this chapter as well as in chapter 4. Fibers moreover need to be placed throughout the entire layer, which poses challenges in dealing with surface tension effects.

**Fibers sticking out of the last layer and into the upcoming one** This challenge is in particular interesting since it requires objects with high aspect ratios (e.g. fibers) to stick out of a surface with typically a high surface tension. This can be generated by either applying a force to pull out the fibers while keeping them perpendicular to the printed layers, as this does not affect the remaining fibers that are ideally randomly oriented within the layer, or by placing fibers on top of the layer and forcing them to penetrate the previous layer.

In both cases, further elaborations are necessary and the underlying results do not pose a full solution to the described challenges. Nevertheless, the CT investigations show a strong connection of the single layers in the orientation of the deeper cracks.

Figure 8.6 (left) shows the extracted fibers and densified parts of the surface, or remaining beam hardening artefacts. The orientation proposed in Figure 8.2 resulting in the crack propagation is shown in Figure 8.5. Fiber extraction resulted in volume fractions between 0.2 to 0.3%, which can be explained by not all fibers being resolved by the CT analysis, or having reached the threshold. This was connected to the voxel size of 4μm (position 1 and 2) and 2μm (position 3) per voxel, which is at
8.3 Results

Figure 8.6: Fiber extrapolation (left) and exemplary crack orientation and dimension measurements (right) in position 1 for smaller surface cracks. Projection view of deeper cracks in position 1 (right). This figure was published earlier in [137].

the limit for detecting the thin fibers with a diameter of 7.2 μm. Furthermore, the analysis of the fiber content was only performed locally at these positions, which might allow higher fiber contents in other areas of the sample.

Other detections concerned air intrusions within the insert, which have already been mentioned in subsection 6.1.2 as a result of irregularities during the manufacturing process weakening the overall strength of the part. Pore radii have been extrapolated numerically and range between 20 to 150 μm. The pores were studied in two sub-volumes shown in Figure 8.7.

8.3.2 Uniform Fiber Orientation

Investigations with the same configuration as described above for a part printed with the modified build plate configuration presented in chapter 4 have been performed resulting in a significantly different orientation shown in Figure 8.9. A fiber extraction is shown in Figure 8.8 where the fibers are more randomly/isotropically oriented in comparison to the bottom-up print. Sub volumes like these were extrapolated at different positions and numerically investigated.

In comparison to Figure 8.2, the fiber orientation is more uniform than with a conventional bottom-up printing technology. Nevertheless, the fiber orientation is not fully isotropic, which can result from surface tension effects, which are increased with composite materials.
8 Internal Fiber Structure and Crack Propagation

Figure 8.7: Sub volume 1 (left) and sub volume 2 (right) with numerically extrapolated pores. This figure was published earlier in [137].

Figure 8.8: Sub volume and fiber extraction with more isotropic fiber orientation.
Figure 8.9: Fiber orientation analysis for the middle and top position. The parts were printed with the modified build plate design. Orientation within the printing plane is shown on the left. Orientation orthogonal to the printing plane is shown on the right. This figure was published earlier in modified form in [4].
Underlying this result, fiber orientation can be predicted as well as controlled by the use of the build plate design. At the current state, a more isotropic fiber orientation has been established allowing the observation that the boundaries of the build plate as well as the resin vat have an influence on the fiber orientation throughout the entire build space, which could be used in future applications to gain control over the fiber orientation by an optimized printer design followed by the natural advantages of controlled fiber orientation.

8.4 Summary

Fiber orientation and defects within an IM insert have a significant impact on the lifetime. It is therefore necessary to understand the behavior of fibers as a result of the manufacturing process represented in the final insert. CT analysis has been used to track cracks evolving from the surface and the length, width, and depth of those cracks can be measured, as well as the fiber orientations and defects such as air intrusion. This part of the chapter concludes investigations presented earlier in subsection 6.1.2 and chapter 6 in a descriptive way, whereas prediction mechanisms are described further in chapter 4 and are valid for multiple applications.

The fibers show the expected orientation along the printing layers, which is also the preferred direction of cracks, since the cracks slide along the fibers and are stopped when penetrating from the side. Deeper cracks are shown to run through the entire insert and propagate along the printed layers respectively the fiber orientation. Furthermore, smaller surface cracks were shown to penetrate only in the sub-mm range and are not affected by the fiber orientation. These cracks can be concluded to be mainly thermal cracks, since they increasingly appear close to the position of the gate where the hot polymer is injected.

Since the IM process is very well established and understood, future manufacturing processes will need to tackle the challenge of isotropically orienting fibers e.g. by changing to a top-down printing process with a modified build plate as described earlier in chapter 4. The adaptive fiber concentration can furthermore improve upon the challenges of stress concentrations e.g. around corners of the insert where especially many larger cracks were detected.

Part printing with a modified build plate designed to allow for fiber movement through the build plate resulted in a better distribution of the fiber orientation outside the printed layer. The underlying conclusion is an increased potential for applications such as a further improvement of topology optimization, where the fiber orientation increases the possibility of weight reduction, heat transfer, and controlled unidirectional or isotropic properties.

Defects such as air intrusions could furthermore be investigated qualitatively and quantitatively using X-ray CT. A reduction of such defects needs to be addressed in

124
future manufacturing processes. Possible solutions might include a vacuum storage in order to remove intrusions from the liquid thermoset photopolymer resin.

CT investigations and additional analysis using numerical modeling can be concluded as a suitable method to investigate also AM composite structures. Structures such as topology optimized structures with adaptive fiber orientation presented in chapter 4 can furthermore be investigated on their effectiveness and safety bringing FRPs in AM to a level where they can be used for safety-related applications. So far, only few safety-critical applications of FRPs in AM have been seen and AM is predominantly used for weight reduction in non-critical components as described in section 2.3.
9 Life Cycle Analysis

Additive manufacturing technologies applied to an injection molding process chain have acquired an increasingly important role in the context of tool insert production, especially by vat photopolymerization. Despite the decreased lifetime during their use in the injection molding process, the inserts come with improvements in terms of production time, costs, flexibility, as well as potentially improved environmental performance as compared to conventional materials in a life cycle perspective.

However, representing a novel technology, inserts may also yield undesired characteristics e.g. in terms of potential environmental impact. This chapter provides a comparative life cycle assessment of conventional brass inserts in comparison to additively manufactured fiber-reinforced inserts used for injection molding and shows that while the comparison of the inserts is inconclusive, the contribution to the environmental impacts of the flow injection molding process is very modest and the choice of insert material should be based on other production performance parameters in a concrete application context.

This contribution supports the development of additively manufactured injection molding inserts with the use of fiber-reinforced vat photopolymerization technology. The life cycle assessment of the prototyping process chain for rapid prototyping with high flexibility provides a base for industrial applications in injection molding.

The content of this chapter was taken and in parts modified from the following publications:


9 Life Cycle Analysis

Additively Manufactured Injection Moulding Inserts for Rapid Prototyping”. In: 
*EUSPEN and ASPE Special Interest Group Meeting: Additive Manufacturing.* 
The European Society for Precision Engineering and Nanotechnology. 2017


9.1 Introduction

Investigations have shown a significant difference in potential environmental contributions such as climate change (global warming potential, GWP) [275] and human health (HH) [276] of IM inserts made from photopolymer exposed in a VPP process [178, 277] compared to conventional inserts made from brass and steel. Despite the described advantages, the lifetime (i.e. number of shots) of AM inserts is significantly lower than the lifetime of conventionally machined metal tools. A general review on environmental impact of AM compared to conventional manufacturing was performed in [278].

This investigation targets the life cycle of an advancement of IM inserts by FRPs [279, 280]. FRPs in IM inserts reduced the crack propagation velocity in the insert and increased the lifetime by a factor of 5 at 5\%_{wt} short CFs and 10 at 10\%_{wt} compared to plain inserts without fiber-reinforcement as well as inserts made conventionally from Al, brass and steel.

This chapter elaborates on previous research by comparing plain inserts without fibers to FRP inserts in the perspective of a life cycle assessment (LCA) according to ISO 14040/44 [281] on a screening level. It therefore contributes to the understanding of the effects of IM with AM inserts on environmental factors.

IM inserts can be advantageous e.g. in prototyping, since new product designs or shifting batches of products do not require the entire mold to be adapted or replaced but just the relatively small inserts. The typical material used today for mold inserts is metal (steel, aluminium, or brass) used in plates, which are CNC-machined into the desired insert shape.

With today’s AM process capabilities, such inserts can be produced by VPP offering application properties comparable to those of metals – but with less time requirement and much better detail and shape possibilities than with machined metal inserts. A
9.1 Introduction

drawback of polymer AM inserts is their limited mechanical properties (in particular strength and stiffness), making them useful mainly in prototyping applications with short lifetime requirements. In order to enhance insert lifetime, and thus also reduce the time and cost related to exchange of inserts, the use of FRPs as insert material is emerging, with a potential to increase insert lifetime by a factor of 10 as described in chapter 6.

Besides these desirable characteristics of AM inserts, an unknown, potentially undesirable one is the related environmental impact. In accordance with sustainable manufacturing frameworks in [282], a comparison of the environmental performance of AM inserts and conventional inserts should

1. consider the entire life cycle of the inserts,
2. analyze the environmental impacts in application context, i.e. not consider inserts as stand-alone objects, and
3. consider the relevant categories of environmental impact.

Existing research on VPP-based AM is scarce and compares such AM parts with conventionally manufactured parts based on related energy requirements [283]. However, for VPP in particular, direct impacts on HH and GWP are seen as an issue of concern [284, 285]. For FRP AM inserts in particular, no significant further reference was published before. This section contributes to closing this research gap, studying FRP AM inserts as an example. In support of decision-making on selection of molding insert technology, this section addresses three research questions:

1. How do fiber-reinforced AM inserts perform environmentally in a full life cycle perspective compared to conventional inserts in CNC-machined brass?
2. Are there break-even points in terms of environmental performance between the two alternatives? If yes, at which shot numbers do they occur?
3. How does the environmental impact of the inserts reflect in the general production context, i.e. what is environmentally preferable in the larger application context?

The research questions are addressed in a comparative environmental LCA of conventional IM inserts produced by CNC-machined brass, and additively manufactured FRP AM inserts. All inserts are applied in a conventional, standardized, and cooled metal mold. The LCA is focused on contributions to GWP and to HH impacts, which together represent the energy-related and the chemical and material related impacts of the compared systems shown in Figure 9.1.
Figure 9.1: Combined flow chart of the two assessed systems of inserts; AM and CNC. All inputs and outputs around the insert production and insert EoL are included all the way from/back to earth. Note: Indicated by grey text and a dashed line, the actual IM process is excluded from the direct comparisons as it is the same for both systems. This figure was published earlier in [232].
9.2 Methods

The LCA was performed on inserts produced with the proprietary photopolymer HTM-140v2 by the company Envisiontec®. The inserts were shaped in the described dimensions of \((20 \times 20 \times 2.7) \text{ mm}^3\) equipped with micro features such as sharp corners, cylindrical cavities and edges as shown in subsection 6.1.2 and Appendix F.

The FRP inserts were produced from a mixture of 5\%_{wt} and 10\%_{wt} of short virgin CFs with dimensions 7.2 \text{μm} \text{ diameter} and 100 \text{μm} \text{ average length} \cite{233}. They were modeled in the software SimaPro® according to \cite{286, 287} and will be referred to as VPP 0\%, VPP 5\%, and VPP 10\%.

The manufactured inserts were grouped in 4 inserts, since the tool used 4 inserts per shot as shown in Figure 6.3 whereas each insert of the compared materials had different specific weights shown in Table 9.1. The material is injected from the back of the machine from a granular primary material. The IM process parameters were not changed from conventional IM manufacturing. Therefore, the process was also not included in the LCA as the focus is on the comparison of different insert materials as well as the impact of waste generation on the IM process.

The lifetime was experimentally determined for PE-LD as part material and is listed in Table 9.2. It can be expected that the lifetime for ABS as part material is lower due to the higher process temperature.

In order to investigate the insert share of environmental impact of the entire IM process, the waste of injected polymer was additionally calculated and compared to the used inserts. ABS and PE-LD were chosen as injected materials. The weight characteristic of 1 shot is shown in Table 9.3. Polymer waste is responsible for approximately 60\% of the injected polymer weight in the process.

It was chosen to use GWP in \text{CO}_2 \text{ equivalent} as well as HH in kg1, 4 – DB equivalent (dichlorobenzene, DB), and later disability adjusted life year (DALY) as indicators for the life cycle impact of the investigated inserts and parts.

Environmental impacts were determined using environmental LCA. The objects of the LCA were inserts produced in two alternative materials: One option in

<table>
<thead>
<tr>
<th>VPP 0 %</th>
<th>VPP 5 %</th>
<th>VPP 10 %</th>
<th>Al</th>
<th>brass</th>
<th>steel</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.3 g</td>
<td>1.325 g</td>
<td>1.35 g</td>
<td>3.2 g</td>
<td>10.2 g</td>
<td>9.4 g</td>
</tr>
</tbody>
</table>

Table 9.1: Weight of 1 insert of the different kinds.

<table>
<thead>
<tr>
<th>VPP 0 %</th>
<th>VPP 5 %</th>
<th>VPP 10 %</th>
<th>Al</th>
<th>brass</th>
<th>steel</th>
</tr>
</thead>
<tbody>
<tr>
<td>500 shots</td>
<td>2600 shots</td>
<td>4500 shots</td>
<td>10 000 shots</td>
<td>10 000 shots</td>
<td>10 000 shots</td>
</tr>
</tbody>
</table>

Table 9.2: Assumed lifetime of 1 insert of the different kinds for injection of PE-LD.
9 Life Cycle Analysis

<table>
<thead>
<tr>
<th></th>
<th>ABS</th>
<th>PE-LD</th>
</tr>
</thead>
<tbody>
<tr>
<td>weight of 4 parts</td>
<td>2.00</td>
<td>1.60</td>
</tr>
<tr>
<td>weight of waste</td>
<td>2.72</td>
<td>2.28</td>
</tr>
<tr>
<td>total weight</td>
<td>4.72</td>
<td>3.88</td>
</tr>
</tbody>
</table>

Table 9.3: Weight of 4 produced parts and residual material of the IM process that was not used in the final parts for 1 shot.

CF-reinforced photopolymer compound produced by means of VPP and a second option in conventionally produced brass rods, from which slices are sawed off and subsequently CNC machined to give the desired cavity shape.

9.2.1 Life Cycle Assessment

The product system was modelled in the software tool SimaPro® (v8.4) with background data from the databases ecoinvent® 3.3, Industry data 2.0, and European Life Cycle Data (ELCD, v3.1). The two assessment objects and their related processes suggest potential relevance of not only energy-related impacts but also of chemicals-related impacts [284]. Therefore, two dedicated life cycle impact assessment methods were chosen to be applied: IPCC 2013 quantifying contributions to the impact category GWP expressed in CO$_2$ equivalent, and USEtox 2.0 [288] quantifying contributions to the impact category HH expressed in DALY (disability-adjusted life years). USEtox aggregates the weighted impact categories “human health, cancer” and “human health, non-cancer” [288]. The assessment covered all inputs and outputs from the modeled life cycle of the two systems apart from emissions occurring during handling e.g. in the AM pre/post-treatment chamber, or during use of the AM inserts. These may have impacts on HH, but no data was found to support their quantification. The relevance of this omission is addressed in the discussion of the results.

The functional unit of the study was defined as: “Provision of 4500 IM shots at the required part quality”. Underpinned by the earlier described tests, lifetimes were set to 4500 shots for the AM inserts and to 10 000 shots for the brass inserts during which the inserts provide the required part quality. Mechanically, the inserts last longer, but part quality would fall after the set number of lifetime shots. In order to determine potential break-even points between the options at higher shot numbers, analyses were performed for higher reference flows as presented in Table 9.4.

As shown in Figure 7.2, the mold contained four cavities for inserts, thus leading to 4 products per shot. This condition was taken into account in all assessments.

The geographical and temporal context of the study was production in Denmark under current conditions, thus assuming Danish electricity mix, Danish EoL treatment, transport scenario for a production site in Denmark, etc.
9.2 Methods

<table>
<thead>
<tr>
<th>No. of shots</th>
<th>Required no. of AM inserts</th>
<th>Required no. of brass inserts</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \leq 4500 )</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>( \leq 9000 )</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>( \leq 10000 )</td>
<td>3</td>
<td>1</td>
</tr>
<tr>
<td>( \leq 13500 )</td>
<td>3</td>
<td>2</td>
</tr>
</tbody>
</table>

Table 9.4: Four different maximum numbers of shots and the related numbers of required inserts, in AM (VPP 10%) and brass respectively.

9.2.2 Material Inventory

The composition of the AM polymer and central processes in its production had to be derived from patents and publicly available material-specific documentations e.g. material safety data sheets (MSDS). For CFs, a full life cycle inventory was found in literature, comprising material inputs, energy requirements, capital goods and services, and process emissions related to production of 1 kg of CF [286].

Wherever possible, model elements were based on or cross-checked against process data from a real, large-scale production setting, including information supplied by specialists from that production setting. Modeling principles of ecoinvent® were followed. Figure 9.1 shows the modeled production systems for the comparison of the two inserts. As illustrated in Figure 9.1, the two assessed systems included the entire life cycle of the respective type of molding insert:

1. Production of the raw materials, i.e. of brass, photopolymer, and CF.

2. Manufacturing of the inserts by CNC milling and VPP, respectively, including treatment of production waste. This also includes relevant aspects of the production of manufacturing equipment, i.e. of the AM machine and of the CNC mill. (The AM machine was not found in databases and was thus modeled along the modeling principle used in ecoinvent for the CNC mill found there).

3. The use stage comprises the number of inserts required to reach the relevant number of shots (see Table 9.4). All other use stage aspects e.g. the IM process itself are assumed to be the same for all inserts and are hence excluded from the comparison.

4. EoL of the inserts is reached after a pre-defined number of shots. The brass inserts go through metal recycling, while the polymer inserts are assumed to be incinerated. Composite recycling has e.g. been reviewed in [13] describing different processes that are in principle also applicable to fiber-reinforced AM parts.
9 Life Cycle Analysis

<table>
<thead>
<tr>
<th></th>
<th>ABS</th>
<th>PE-LD</th>
</tr>
</thead>
<tbody>
<tr>
<td>GWP in kg CO\textsubscript{2}eq</td>
<td>3.995</td>
<td>1.896</td>
</tr>
<tr>
<td>HH in kg 1, 4 – DBeq</td>
<td>7.057</td>
<td>2.486</td>
</tr>
</tbody>
</table>

**Table 9.5:** LCA data for 1 kg of the part materials.

<table>
<thead>
<tr>
<th></th>
<th>VPP 0 %</th>
<th>VPP 5 %</th>
<th>VPP 10 %</th>
<th>Al</th>
<th>brass</th>
<th>steel</th>
<th>CFs</th>
</tr>
</thead>
<tbody>
<tr>
<td>GWP</td>
<td>3.118</td>
<td>3.769</td>
<td>4.419</td>
<td>18.971</td>
<td>5.045</td>
<td>1.744</td>
<td>16.127</td>
</tr>
<tr>
<td>HH</td>
<td>621.790</td>
<td>596.313</td>
<td>570.835</td>
<td>269.738</td>
<td>4772.948</td>
<td>85.757</td>
<td>112.236</td>
</tr>
</tbody>
</table>

**Table 9.6:** LCA data for 1 kg of the insert materials.

### 9.3 Results

It was shown that the GWP increased at increasing CF content, whereas the contributions to HH decreased. An increase in GWP of approximately 5\% and a reduction of HH by less than 1\% is reached when adding 1\% CF to the AM insert compared to an insert without CF.

Brass leads to a significantly higher impact on HH as shown in Table 9.6 where influencing factors of 1 kg of the materials are listed. ABS has a higher impact on the LCA as compared to PE-LD with an accelerated impact due to its chemical composition and its production chain.

Waste is responsible for 60\% of the necessary injected polymer and makes up for up to 33\% of the contributions for 1 shot as shown in Table 9.7 for 1 shot. The factors were calculated as ratio of impact of waste to overall impact of necessary parts and waste. ABS can be considered more influential in both factors with a high impact on the LCA.

When neglecting the contributions of the polymer waste, CFs have a strong impact on the LCA of the IM process. Even at higher shot numbers, the GWP contributions to the 4 inserts VPP 10\% in the IM machine remain significantly below the GWP of the metal materials and VPP 5\% competes with steel at higher shot numbers as can be seen in Figure 9.2.

<table>
<thead>
<tr>
<th></th>
<th>VPP 0 %</th>
<th>VPP 5 %</th>
<th>VPP 10 %</th>
<th>Al</th>
<th>brass</th>
<th>steel</th>
</tr>
</thead>
<tbody>
<tr>
<td>GWP ratio ABS</td>
<td>0.3301</td>
<td>0.2857</td>
<td>0.2508</td>
<td>0.0159</td>
<td>0.0194</td>
<td>0.0569</td>
</tr>
<tr>
<td>HH ratio ABS</td>
<td>0.0043</td>
<td>0.0044</td>
<td>0.0046</td>
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**Table 9.7:** Ratios of waste material as compared to the total of GWP and HH impacts for 1 shot.
9.3 Results

Figure 9.2: Cumulative GWP impact neglecting the polymer waste. This figure was published earlier in modified form in [266, 267].

Due to the high effects of the photopolymer on HH, the AM inserts remain between Al and steel below and brass exceeding the impact. Still, smaller production numbers below 5000 shots favor the AM inserts as suitable alternatives for prototyping applications as shown in Figure 9.3.

It is moreover noticeable that the GWP of both ABS as well as LD-PE waste remains high compared to the insert materials. The contributions of the waste component of the IM process overrules the contributions of the insert material on the GWP as shown in Figure 9.4 and Figure 9.5. At higher shot numbers, the ratio converges to 1 meaning that the insert materials have negligible contribution to the global impact.

Due to the poor performance of brass in terms of HH, the contributions of waste ABS and PE-LD remains at 20% for brass in terms of ABS waste (Figure 9.6) and 10% for brass in terms of PE-LD waste (Figure 9.7). Production of VPP 0% inserts is influenced by up to 63% in terms of ABS waste and up to 40% in terms of PE-LD waste. Better performance of FRPs in the AM inserts in terms of lifetime results in a higher contribution of the waste material.
Figure 9.3: Cumulative HH impact neglecting the polymer waste. This figure was published earlier in modified form in [266, 267].

Figure 9.4: Ratio of GWP impact of ABS waste material as compared to the total impact on the LCA. This figure was published earlier in modified form in [266, 267].
9.3 Results

Figure 9.5: Ratio of GWP impact of PE-LD waste material as compared to the total impact on the LCA. This figure was published earlier in modified form in [266, 267].

Figure 9.6: Ratio of HH impact of ABS waste material as compared to the total impact on the LCA. This figure was published earlier in modified form in [266, 267].
9.3.1 Environmental Performance

The life cycle impacts on GWP and HH for the two insert alternatives are shown in Figure 9.8 and Figure 9.9 with distinction of their origins in the modeled life cycles. The major drivers for the GWP of the AM insert shown in Figure 9.8 are:

- the energy consumption during the curing process of the photopolymer leading to approximately 30% of total GWP,
- the production of the different ingredients required to produce the photopolymer liquid (e.g. PMMA beads, methyl acrylate, PAN fibers), collectively contributing with approximately 40% of total GWP, and
- the treatment of the toxic waste resulting from the uncured leftovers of the photopolymer, accounting for another approximately 11% of total GWP.

For the brass insert GWP profile, main contributors are:

- The energy-intensive steps of the raw material production as well as the re-melting during recycling (approximately 60% of total).
- The production of the insert itself (approximately 25% of total), primarily due to the energy consumption of the CNC machining process.
9.3 Results

Figure 9.8: GWP of AM inserts and brass inserts in an application with up to 4500 shots, requiring 1 AM insert and 1 brass insert. Contributing life cycle processes are indicated by color. (The top-four processes in the nomenclature relate to the brass insert, all below relate to AM insert.)
Figure 9.9: HH impact of AM inserts and brass inserts in an application with up to 4500 shots, requiring 1 AM insert and 1 brass insert. Contributing life cycle processes are indicated by color. (The top-four processes in the nomenclature relate to the brass insert, all below relate to AM insert.)
In Figure 9.9, the HH impact of the AM insert is primarily attributable to two drivers:

- the production of the AM machine itself (approximately 45% of total) and
- the energy consumed in the AM process (also approximately 45% of total).

The former has to be interpreted carefully as it is based on an approximation derived from a metal working machine used in the ecoinvent® database as a generic machine included e.g. in the CNC mill provided in ecoinvent®. Further it can be stated that the HH results for the AM insert are generally underestimated, as they depend on the individual handling of the chemicals, the local infrastructure such as ventilation, and potential impacts on HH, which is presently unknown. The latter relates especially to the finished AM part, which might still emit toxic components despite it being cured.

The HH impacts of the brass insert are dominated by the production of the brass itself. Copper is an essential alloying element of brass and known for the environmental burdens caused by its production process [286].

In terms of the global warming impacts, the brass option shows preferable over its life cycle, mainly due to the high recyclability of this classic metal. In terms of HH impacts, the comparison gives the opposite result with the AM option having a life cycle impact, which is an order of magnitude lower on than the brass option. While the uncertainties accompanying the modelling of HH impacts are considerably higher than for global warming [276], neither of the two alternatives can be concluded on this basis to be environmentally superior to the other.

### 9.3.2 Potential Break-even Between the Alternatives

Figure 9.8 and Figure 9.9 show results for applications that require up to 4500 shots, which means they show a 1-on-1 comparison of the two types of inserts. If the comparison would have to be done for an application requiring more than 4500 shots, 2 AM inserts would have to be compared to 1 brass insert, and the scores of the AM option would thus double (in one step, not gradually), while those for the brass option would remain the same. The required numbers of inserts for different numbers of shots in Table 9.4 are combined with the LCA results for the two insert types to investigate the performance of the two inserts at higher shot numbers in terms of climate change impacts and HH impacts. Figure 9.10 shows the results for global warming and HH impacts.

Key questions in the application context are: which option performs better at a given required number of shots and are there potential break-even points over the numbers of shots where one technology performs better than the other. Figure 9.10 supports this analysis showing the steps occurring at the shot numbers given in Table 9.4 and beyond, up to 100,000 shots. Since the lines representing the two insert options in
Figure 9.10: GWPs (top) and HH impact (bottom) of inserts over number of shots, for the range of 1000 to 100000 shots.
9.4 Summary

the figure do not cross at any shot number, there are no such break-even points in this case. In other words, irrespective of shot numbers, i.e. insert lifetimes, the brass option would always perform better than the FRP AM option regarding GWP. A very similar pattern is observed for the GWP but here with lowest impacts for the AM insert, and the results are hence not shown here.

However, as can be inferred from Figure 9.10, if the lifetime of the AM insert could be extended above 10,000 shots, it would be exactly on the same impact level as the brass insert, and in total thus preferable. Since ongoing technological developments indicate lifetimes of FRP AM inserts of about 15,000 shots, such a break-even in favor of AM seems achievable in time.

In summary, regarding global warming impacts, the brass option shows preferable irrespective of the number of shots, mainly due to the high recyclability of this classic metal. The AM option performs a factor 10 better on HH than the brass option, while the brass option performs a factor 2 better on GWP than the AM option irrespective of the number of shots.

However, since the related AM process is novel, inventory data are currently missing in established databases, such as ecoinvent®. While all ingredients and process steps were modeled based on specific data sheets using the same modeling principles as the ecoinvent® unit process database, especially the photopolymer composition and the effects of concrete handling conditions must be considered uncertain in the model.

9.3.3 Relevance in Production Context

If the GWP results are put into context of the IM machine, it becomes evident that the impacts arising from either insert are negligible: The electricity consumption of a standard IM machine thus results in 105 kg CO\textsubscript{2} for 4500 shots and the GWP of the AM-insert in its whole life cycle amounts to 0.121 kg CO\textsubscript{2}.

When deciding which of the inserts to choose, the difference between their environmental impacts are so modest compared to the overall impacts from the IM process that the environmental impact becomes of little interest compared to differences in important production technological characteristics of the two options. Further development might be possible using the AM-connected freedom of design to reduce the waste ratio of the part production.

9.4 Summary

CFs have a significant influence on the performance of AM inserts in the LCA. Concerning GWP contributions, AM inserts with a ratio of 10 %\textsubscript{wt} have a preferable performance in comparison to metal inserts.
AM inserts are most suitable for smaller production volumes in terms of their environmental impact, manufacturing costs and time as well as feature accuracy.

Due to the lower lifetime of AM inserts in the IM process, metal inserts are preferred for series production of high part volumes, whereas AM inserts are preferred in all key aspects for prototyping of low to medium volume production.

Judging from the performed assessment, the AM option turns out preferable regarding HH, but the brass insert is favored regarding GWP. Based on an AM insert lifetime of 4500 shots and brass insert lifetime of 10,000 shots, this outcome is valid for shot numbers from 1000 shots to 100,000 shots.

The question of what is environmentally preferable in a production context should be answered on a machine tool level rather than on an insert level. For both impact categories, the impacts from the life cycle of either insert type are very small compared to the impacts of the whole IM process. As the IM machine uses less energy per shot when producing many shots in a given time, the standard target of shorter cycle times and insert exchange times seems a more relevant metric to base the decision on than to look at the insert materials. Even the high uncertainties introduced by lack of inventory data etc. do not challenge this.

Future work should focus on improving the assessment basis in terms of life cycle inventory data for the entire VPP process, including the machine and emissions during insert manufacture, handling and use.

The total number of full-quality shots producible with fiber-reinforced AM insert is continuously increasing, as this technology becomes more and more interesting beyond prototype manufacturing and towards small series or even series production.
Part IV

Conclusion
10 Summary and Outlook

A number of experiments were performed with the aim of gaining a better understanding of FRPs in AM. The entire process chain was investigated by the use of examples starting from the raw materials to the final part in an industrial application. Improvements were achieved in all fields, whereas an overall understanding of the existing technologies has been gained at the start of the project in order to allow for a structuring of the fields and problems.

The different investigated areas of the thesis are described in Figure 10.1. The structure is concluded as state-of-the-art, technology, industrial application, and optimization and development. A further description is given in the final part of this chapter. Following the description given in section 1.2 and Figure 1.2, all three factors, design, material, and process, were investigated and improved. Challenges were faced on all levels providing the opportunity to improve upon them.

10.1 Result Summary

Fiber-Matrix Interface  Initial research showed an expected weak fiber-matrix interface resulting in gaps and cracks around the fiber weakening the mechanical properties of the printed part. Taking into account the chemical composition of thermoset photopolymers and their well-investigated curing mechanisms, it was possible to improve on the chemical interaction between the liquid polymer and the fibers and therefore improve the strength and modulus by up to a factor of 3.

Gaps and cracks were improved by a better chemical interaction between the fibers and the thermoset photopolymer matrix material. Stirring mechanisms in the original resin material as well as special mixing and post-mixing mechanisms were identified in order to reduce the amount of air intrusions in the resin and therefore provide a final part with less defects, which might lead to fatal material failure.

Following the presented investigations, it is the understanding that chemical interaction of fiber and matrix require further improvement, which can be provided by applying both optical and thermal cooling as is given in the nature of thermoset photopolymers. Furthermore, different chemical compositions allow for adjusted properties of the final part in terms of e.g. matrix strength, but can also account for an improved fiber-matrix interface.
Figure 10.1: Summary of the investigated areas and development steps during the project. This figure was published earlier in modified form in [4].

**Fiber Orientation Prediction and Control**  It is the nature of composite materials to depend on the mechanical properties and on the orientation of the fibers within the matrix. This aspect was again proved in the experimental investigations following the ambition to improve upon fiber orientation and deposition. The orientation of fibers has been investigated in detail following a fiber orientation along the printing layers as a natural result of the printer geometry in a bottom-up setup where the resin is cured between two plates usually made from steel, or glass. This configuration leaves the fibers no physical DoF to orient perpendicular to the printed layer.

Top-down printing usually restricts the printing area by a steel or glass plate on the bottom and the surface tension on the top of the resin limiting the fiber orientation less whereas factors such as viscosity and strength of the surface tension play an important role.

In numerical simulations, it was for the first time possible to predict and describe the fiber orientation in both bottom-up and top-down configurations. A modified build plate design was furthermore introduced influencing the fluid flow in the resin vat and therefore determining the fiber orientation. Experimental investigations confirmed the numerical simulations showing a more isotropic fiber orientation in the final printed part. This represents the first approach towards controlling fiber orientation in a VPP process opening a massive potential for mechanically and thermally demanding applications. This includes a further degree of freedom for topology optimization with all its advantages.

Investigations using CT scanning as well as SEM have been used for post-production investigations extracting the fiber orientation, cracks, and internal defects such as air intrusions. The technology and post-processing algorithms have proven to be accurate enough to detect also smaller defects and therefore provide an option to describe the part quality and help to extend the potential applications towards safety-critical components.
Result Summary

Lifetime and Crack Propagation IM inserts were chosen as industrial application with the aim of improving both lifetime and cycle time during the process. By adding 10% wt of CFs, it was possible to improve the lifetime of the inserts by a factor of 10 as compared to the same configuration and geometry without reinforcement. This represents the highest number of shots in this configuration that have been statistically proven. The IM process is however still limited in terms of temperature and pressure of the injected polymer and therefore the comparability towards industrial production environment is not fully provided.

Crack propagation has been identified as a critical factor for the lifetime of inserts in the IM process. Due to the brittle nature of the thermoset photopolymer material, the initiation of a crack has been a strong indicator for a close failure of the insert when not using fiber-reinforcement. Investigations have shown that cracks propagate at 1.25% in speed in comparison to non-reinforced inserts. The final failure was therefore stretched in time. Despite the reduced surface quality of the final part, applications such as RP are still possible at affordable downsides.

Larger cracks align along the fibers where they can slide and are not blocked by other material. Taking into account the predicted stresses, which are well understood in the IM process, is a first step to prevent a quick failure of the insert. Further investigations have shown additional smaller cracks not significantly penetrating the surface of the insert and without specific alignment related to the fiber orientation, they are assumed to represent cracks resulting mainly from thermal stresses and are also concentrated in the regions of higher temperatures of the injected polymer.

Further development is given by an isotropic fiber orientation developed by the use of the modified build plate design. Advancement of the fiber orientation prediction and control can lead towards further applications in the environment of digital and optimized production. A case study example of topology optimized IM inserts was given as a future concept of adapted manufacturing paradigms.

The lifetime is limited by the thermal stability of the part. Given its polymer nature, thermal stresses can affect the mechanical properties of the part. However, surface roughness was not significantly affected over the lifetime of the insert despite changes in the surface profile caused by heat introduced by the injected polymer.

Injection Molding Cycle Time Thermal effusivity is the main reason for a higher average temperature of AM inserts during the IM process. This results in higher peak-to-peak temperatures of the insert as well as a reduced cycle time as the insert requires a longer packing and cooling time after the ejection of the part.

Numerical simulations have shown that the use of para-conformal cooling in the standardized metal mold is able to reduce the cooling time by 60% due to an increased heat absorption by the cooling liquid. Further development has been simulated using active cooling of the insert where the cooling liquid is led through
the insert itself removing heat at the source rather than after a propagation through the metal mold. This innovation has reduced the average temperature and therefore also cycle time at the same lifetime in an industrial case study.

Given the increase in lifetime, molding cycle time can be considered another major issue of soft tooling RP in the IM environment. Tackling these challenges combines multiple technologies presented in the industry 4.0 environment such as numerical simulations, optimization, as well as other automation technologies.

Life Cycle of Additively Manufactured Molds Several points have been shown during an LCA regarding environmental factors as well as EoL scenarios. The increased lifetime of fiber-reinforced inserts has significantly influenced the environmental impact in comparison to materials such as steel, brass, and aluminum. A severe issue is placed in the missing EoL solutions of cured photopolymer as well as material handling and waste during the part production.

These solutions exist for the injected polymer such as PE-LD and ABS. Nevertheless, the waste production during the IM process has a significant influence on the levels of GWP and HH. Despite these results, desirable EoL scenarios from an IP perspective in a competition-driven industry can give advantages.

10.2 Personal Reflection

While AM is often mentioned as the next industrial revolution, a deeper understanding of the underlying technology is a critical factor for future perspectives in this type of manufacturing and process engineering. After the first AM technologies were introduced in the 1980s, the initial hype has passed and the implementation of AM parts and systems have gone through a reconsideration phase.

While new systems for both polymer and metal AM are introduced on the market on a weekly basis, industry has developed an awareness of the advantages and disadvantages of AM as a novel technology. Not only has AM reached a level where it is used in actual applications and process chains, it has especially entered prototyping and rapid manufacturing.

Current and future leaders will not only have to consider how properties of AM systems and parts can be improved (e.g. in terms of mechanical, thermal, chemical, and electrical properties) in order to perform under the given considerations and demands, but they will also have to adapt to new process chains, design paradigms, applications, and value propositions. These presumptions fit the considerably quick developments in the framework of digital technologies summarized as industry 4.0. AM has to be considered as one of the future enabling technologies for further development of industrial processes and production.
The project has set the first boundaries for the implementation of fiber-reinforcement in AM where especially thermoset photopolymers have been considered. The recent scientific and industrial effort to further develop the required chemicals as well as the manufacturing processes emphasizes the further requirements of the technology to perform under elevated mechanical and thermal conditions.

This development can be seen as a representation of the current state in the technology life cycle. The technology has passed through stages of hype and disillusion before entering the stage of industrial adoption. The specific technology of FRPs in AM can serve as a gap closer between weaker non-composite polymer materials and expensive, heavy metal applications emphasizing the known advantages of AM in the industrial context.

While trade fairs such as the latest TCT Formnext, which was held in November 2018 in Frankfurt, Germany, reveal a large number of innovations, there seems not to be a common synthesis of the industry. Given the novelty of the field, a large number of both large, established, and start-up companies have entered the field and provide innovation. Nevertheless, only few applications have reached a level where they can compete against well-established production lines generating large numbers of parts.

Nevertheless, AM is in the process of leaving the niche region gaining an increased interest for other domains. It will be the challenge of engineers, business developers, and industry specialists to find and determine new values generated by AM. The progress in the field is enormous and the establishment in conventional industries will require further efforts.

10.3 Recommendations for Further Development

From the performed investigations, the application of fiber-reinforced IM inserts can be concluded as a potential application in an industry 4.0 digital manufacturing environment collecting the capabilities of AM systems with well-established IM technology. The methods to implement fiber-reinforcement at the moment do not allow automated printing as well as post-processing of the part. In the framework of automated, flexible, and individualized production, these points need to be considered and developed further in order to allow a reduction of human labor in an environment exposed to toxic chemicals and fiber materials.

Developments regarding simulations of part design, production, and e.g. fiber orientation will need to be improved further. Given the high specialization of this topic and the lack of educated talent to perform such simulations, it will be necessary to develop (semi-)automated simulation tools to perform geometric part optimization in both form and functionality. A special focus can hereby be put on cooling
mechanisms of the inserts, since this topic is a key issue in the manufacturing cycle time as pointed out throughout this thesis.

Further improvement efforts can be targeted towards a stronger fiber-matrix interface, better thermal conductivity by adaption of the fiber content, fiber material, fiber orientation, as well as part geometry. These factors cannot be generalized and therefore will require individual development for different applications. Since fiber-reinforcement of AM is a highly diverse topic, cooperation between multiple disciplines will be a key issue for further development.
List of Publications

Peer-reviewed Journal Papers


Conference Proceedings as First Author


List of Publications


• T. Hofstätter, D. B. Pedersen, G. Tosello, and H. N. Hansen. “Challenges and Opportunities of Fibre-reinforced Polymers in Additive Manufacturing with Focus on Industrial Applications”. In: *EUSPEN and ASPE Special Interest Group Meeting: Additive Manufacturing*. The European Society for Precision Engineering and Nanotechnology. 2017


• T. Hofstätter, D. B. Pedersen, G. Tosello, and H. N. Hansen. “Thermal Behaviour of Additively Manufactured Injection Moulding Inserts”. In: *EU-SPEN’s 18th International Conference & Exhibition*. The European Society for Precision Engineering and Nanotechnology. 2018
• T. Hofstätter, D. B. Pedersen, G. Tosello, and H. N. Hansen. “Evolution of Additively Manufactured Injection Molding Inserts Investigated by Thermal Simulations”. In: *34th Annual Meeting of the Polymer Processing Society (PPS34)*. 2018


### Conference Proceedings as Co-author


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158


Bibliography


160


Bibliography


Bibliography


Bibliography


Bibliography


Bibliography


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Part V

Appendix
A Supplementary information to chapter 2

The following document is reprinted with explicit permission by the company BASF as copyright holder. It gives a graphical overview on the available manufacturing methods for composite materials in an industrial context. The full document can be found in [71].
Manufacturing Processes for Thermoset Composites

Hand Lay-up

Hand lay-up consists of manual positioning of fiber reinforced mats or prepreg plies onto a prepared mold. Thermosetting resins are then applied by brushing, spraying or resin infusion. Entrapped air can be removed with the aid of rolling, and the laminate can be left to dry at room temperature. Curing can be accelerated in ovens or by vacuum. Hand lay-up is a simple and relatively low cost processing method for large components such as wind turbine parts and boats.

Spray Lay-up

Spray lay-up is a conventional open mold process. Gelcoat is applied as a first layer onto a previously waxed mold and then cured. Chopped reinforcements and thermoset resins are then sprayed onto the mold and left to cure at room temperature in ovens or autoclaves. Spray lay-up allows more shape complexity and a quicker production time than hand lay-up. This process is suitable for large components with less complex geometries such as boats and bath tubs.

Filament Winding

Filament winding refers to the winding of thermoset resin impregnated fiber rovings under tension around a rotating mandrel. These fabricated circular composite products with a hollow core are then cured at room temperature or in ovens and used in applications where weight, chemical resistance, pressure and temperature are an important concern. Pipelines, tanks and vessels are example applications of filament winding produced composite parts.
RTM (Resin Transfer Molding)

RTM is a vacuum-assisted closed mold process. Fiber reinforcements are positioned in a matched male and female mold, which is then closed and clamped. The matrix is injected under pressure until the mold is filled. The parts cured in the mold are normally heated by controller. RTM is a fast and clean process to produce composites with large surface areas, complex shapes and smooth finishes like boat hulls and wind turbine blades.

Pultrusion

Pultrusion is a high-volume production process for composite profiles. Reinforcement materials (long fibers, mats or fabrics) are pulled and guided through a bath of matrix for impregnation, passed through a heated die for curing and cut at the end of the line into various tubes and flat sheets with excellent mechanical and chemical properties.
B Supplementary information to section 2.2

The following dog bone design was utilized to investigate different polymer materials including IM parts as well as fiber-reinforced and plain thermoset photopolymer materials produced by VPP. The results are presented in Figure 2.3. The design was taken from ISO 527-1, design 1BB, which is predominantly suitable for polymer tensile tests.
B Supplementary information to section 2.2
C Supplementary information to chapter 3

The following document is reprinted with explicit permission by the company Zoltek as copyright holder. It represents the technical data sheet for the CFs used throughout this thesis whereas the category PX 35 was used as sponsored by the company.

The device presented thereafter was used for investigations regarding the fiber-matrix interface of thermoset photopolymers and GFs and represents a casting device with a PTFE cavity for better wall interaction as well as an aluminum plate for improved thermal conduction of the heat of the underlying heater to the photopolymer.

The device was manufactured in the workshop of the Department of Mechanical Engineering at the Massachusetts Institute of Technology.
Technical Datasheet

ZOLTEK™ PX35 Milled Carbon Fibers

DESCRIPTION

ZOLTEK PX35 Milled Fibers are specifically processed PAN (polyacrylonitrile) based fibers for lower costs applications. ZOLTEK PX30 MF High Purity Milled Fibers are 99+% pure carbon fibers derived from ZOLTEK’S high temperature batch graphitization process. All ZOLTEK milled products are free of any sizings. ZOLTEK’S in-house milling system ensures product quality and traceability from raw material through finished product.

APPLICATIONS

- Buoyancy
- RFI / EMI shielding of electronic devices
- Static dissipation
- Resin-rich surface reinforcement in composites
- Low-cost friction compounds

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The properties listed in this datasheet do not constitute any warranty or guarantee of values. This information should only be used for the purposes of material selection. Please contact us for more details.

CERTIFICATION

ZOLTEK PX35 & PX30 Milled Fibers are manufactured in accordance with ZOLTEK’S written and published data. A Certificate of Conformance is provided with each shipment.

SAFETY

Obtain, read, and understand the Material Safety Data Sheet (SDS) before use of this or any other ZOLTEK product.

ZOLTEK™ PX35

ZOLTEK PX35: each pallet contains 45 boxes and has a net weight of 1,600 lb (816 kg).

ZOLTEK PX30: each pallet contains 27 boxes and has a net weight of 1,080 lb (490 kg).

Each box is 14.5" x 14.5" x 14.5" (36.8 cm x 36.8 cm x 36.8 cm) and contains 40 lb (18.1 kg) of milled fiber in a polyethylene bag.
D Supplementary information to chapter 4

D.1 Domain Drawings of the Moving Mesh Domains

The following drawings represent the simulated domain used to predict fiber orientation during VPP printing as well as to establish a technique to control fiber orientation. The domains represent both bottom-up as well as top-down printing by changing the domain boundary conditions. The dimensions in the simulations were taken from real or realistic machine designs but are in principle scaleable to a certain extend.
Square Moving Mesh Domain
D.1 Domain Drawings of the Moving Mesh Domains
D.2 Modified Build Plate Design

The following drawing represents a modified build plate domain, which was able to achieve a more isotropic fiber orientation in the final part. The plate is equipped with 88 holes allowing the fiber-filled resin to flow through the build plate and therefore generate a different fiber orientation.

The device was manufactured in the workshop of the Department of Mechanical Engineering at the Technical University of Denmark.
D.2 Modified Build Plate Design

Modified Top Down Build Plate

- 50mm x 50mm
- 6mm center-to-center holes
- 11mm x 8mm
- Ø4
- A-A section view
E Supplementary information to chapter 5

E.1 Construction Drawings for the Tape Placement Assembly

The following drawings belong to a case study tape placement device aiming to place fibers onto a surface and at the same time impregnate the fibers with e.g. a thermoset photopolymer. The device comes with a number of advantages presented in the corresponding chapter.

The device was manufactured in the workshop of the Department of Mechanical Engineering at the Technical University of Denmark.
Cage Wheel

E.1 Construction Drawings for the Tape Placement Assembly
### Cage Wheel Side

<table>
<thead>
<tr>
<th>Section</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>A-A</td>
<td>Cross-section of the wheel side</td>
</tr>
</tbody>
</table>

![Diagram of cage wheel side](image-url)
E.1 Construction Drawings for the Tape Placement Assembly
Wheel Holder

E.1 Construction Drawings for the Tape Placement Assembly
Mixer Holder Bottom

Supplementary information to chapter 5
E.1 Construction Drawings for the Tape Placement Assembly

Mixer Holder Top

Diagram showing dimensions and labels.
E.2 COTS Materials for the Tape Placement Assembly

The following documents are reprinted with explicit permission by the companies SKF, Festo, and Siko BV as copyright holders. The parts were purchased from or sponsored by the manufacturers in order to assemble the tape placement device with the drawings presented in section E.1. The parts were not modified.
E.2 COTS Materials for the Tape Placement Assembly

61804-2RS1

**Dimensions**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>d</td>
<td>20</td>
<td>mm</td>
</tr>
<tr>
<td>D</td>
<td>32</td>
<td>mm</td>
</tr>
<tr>
<td>B</td>
<td>7</td>
<td>mm</td>
</tr>
<tr>
<td>d₁</td>
<td>23.85</td>
<td>mm</td>
</tr>
<tr>
<td>D₂</td>
<td>29.4</td>
<td>mm</td>
</tr>
<tr>
<td>r₁,₂</td>
<td>min. 0.6</td>
<td>mm</td>
</tr>
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</table>

**Abutment dimensions**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>dₐ</td>
<td>min. 22</td>
<td>mm</td>
</tr>
<tr>
<td>dₐ</td>
<td>max. 23.6</td>
<td>mm</td>
</tr>
<tr>
<td>Dₐ</td>
<td>max. 30</td>
<td>mm</td>
</tr>
<tr>
<td>rₐ</td>
<td>max. 0.3</td>
<td>mm</td>
</tr>
</tbody>
</table>

**Calculation data**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Basic dynamic load rating</td>
<td>C</td>
<td>4.03 kN</td>
</tr>
<tr>
<td>Basic static load rating</td>
<td>C₀</td>
<td>2.32 kN</td>
</tr>
<tr>
<td>Fatigue load limit</td>
<td>Pᵤ</td>
<td>0.104 kN</td>
</tr>
<tr>
<td>Limiting speed</td>
<td></td>
<td>13000 r/min</td>
</tr>
<tr>
<td>Calculation factor</td>
<td>kᵣ</td>
<td>0.015</td>
</tr>
<tr>
<td>Calculation factor</td>
<td>f₀</td>
<td>14.5</td>
</tr>
</tbody>
</table>

**Mass**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass bearing</td>
<td>0.018</td>
<td>kg</td>
</tr>
</tbody>
</table>
Supplementary information to chapter 5

Data sheet

<table>
<thead>
<tr>
<th>Feature</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Outside diameter</td>
<td>4 mm</td>
</tr>
<tr>
<td>Blending radius relevant for flow rate</td>
<td>18 mm</td>
</tr>
<tr>
<td>Inside diameter</td>
<td>2.6 mm</td>
</tr>
<tr>
<td>Min. bending radius</td>
<td>6 mm</td>
</tr>
<tr>
<td>Temperature dependent operating pressure</td>
<td>0.95 ... 10 bar</td>
</tr>
<tr>
<td>Operating medium</td>
<td>Compressed air in accordance with ISO8573-1-2010 [7-0-0] Water as per manufacturer’s declaration at <a href="http://www.festo.com">www.festo.com</a></td>
</tr>
<tr>
<td>Food-safe</td>
<td>See Supplementary material information</td>
</tr>
<tr>
<td>Ambient temperature</td>
<td>-35 ... 90 °C</td>
</tr>
<tr>
<td>Authorisation</td>
<td>TÜV</td>
</tr>
<tr>
<td>Product weight according to length</td>
<td>0.0085 kg/m</td>
</tr>
<tr>
<td>Pneumatic connection</td>
<td>For push-in connector outside diameter 4 mm for barbed connector internal diameter 3 mm with nut</td>
</tr>
<tr>
<td>Colour</td>
<td>neutral</td>
</tr>
<tr>
<td>Shore hardness</td>
<td>0 52 ±1/3</td>
</tr>
<tr>
<td>Materials note</td>
<td>Free of copper and PTFE Conforms to RoHS</td>
</tr>
<tr>
<td>Material tubing</td>
<td>TP6-EP(U)</td>
</tr>
</tbody>
</table>
### General Tolerances according to:

<table>
<thead>
<tr>
<th>Description</th>
<th>uncontrolled copy when printed</th>
<th>Designed</th>
<th>Changed</th>
<th>DKU</th>
<th>EDCOUST</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mixer</td>
<td></td>
<td>10.09.1994</td>
<td>21.08.2007</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mischer</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Element orientation to this axis**  
Elementausrichtung nach dieser Achse

- 7 Mixing Elements  
  - Material: POM (natural)  
  - Werkstoff: POM (natur)

- Mixer Housing  
  - Material: PP (natural, transparent)  
  - Werkstoff: PP (natur, transparent)

**Detail:** Scale 2:1  
Einzelheit: M 2:1

**Grat max. / Flash max.**  
0.3mm

---

**SULZER**

ISO "E"  
MA 3.0-07-S  
03
E.3 Construction Drawings for the Joule Heating Extruder

The following drawings belong to a Joule heating extruder aiming to pre-heat the filament in order to increase the heating speed for faster printing. The electrical conduction of the CFs is used to heat the filament from the inside whereas conventional extrusion-based printing heats the filament from the outside.

The device was manufactured in the workshops of the Department of Mechanical Engineering at the Massachusetts Institute of Technology.

The final document is reprinted with explicit permission by the company stratasys as copyright holder. The material was used for experiments in the Joule pre-heated nozzle as case study.
E.3 Construction Drawings for the Joule Heating Extruder

Imperial unit system.

The original dimensions were based on the...
E.3 Construction Drawings for the Joule Heating Extruder

![Construction Drawing Diagram]
Add Fish Paper in the same shape.

Joule Heater Insulating Layers

4

1 of 1
E.3 Construction Drawings for the Joule Heating Extruder

Joule Heater Bottom Connector

Dimensions:
- Length: 75
- Width: 30
- Height: 7
- Hole Diameter: 6
- Bolt Length: 25
- Bolt Diameter: 10
- Bolt Head Diameter: 17
E.3 Construction Drawings for the Joule Heating Extruder
E.3 Construction Drawings for the Joule Heating Extruder
FDM Nylon 12CF™ is a carbon-filled thermoplastic with excellent structural characteristics. The material is comprised of a blend of Nylon 12 resin and chopped carbon fiber, at a loading of 35% by weight. This combination produces one of the strongest thermoplastics in the FDM® material portfolio. It has the highest flexural strength of any FDM thermoplastic, resulting in the highest stiffness-to-weight ratio.

Appropriate uses include strong but lightweight tooling applications and functional prototypes in the aerospace, automotive, industrial and recreational manufacturing industries. FDM Nylon 12CF is available on the Fortus 450mc™ 3D Production System and is compatible with SR-110™ support material.
At the core:
Advanced FDM Technology
Fortus 3D Production Systems are powered by FDM (fused deposition modeling) technology. FDM is the industry’s leading additive manufacturing technology, and the only one that uses production-grade thermoplastics, enabling the most durable parts. Fortus® systems use a wide range of thermoplastics with advanced mechanical properties so your parts can endure high heat, caustic chemicals, sterilization and high-impact applications.

No special facilities needed
You can install a Fortus 3D Production System just about anywhere. No special venting is required because Fortus systems don’t produce noxious fumes, chemicals or waste.

No special skills needed
Fortus 3D Production Systems are easy to operate and maintain compared to other additive fabrication systems because there are no messy powders to handle and contain. They’re so simple, an operator can be trained to operate a Fortus system in less than 30 minutes.

Get your benchmark on the future of manufacturing
Fine details. Smooth surface finishes. Accuracy. Strength. The best way to see the advantages of a Fortus 3D Production System is to have your own part built on a Fortus system. Get your free part at stratasys.com.

### Mechanical Properties

<table>
<thead>
<tr>
<th>Property</th>
<th>Test Method</th>
<th>English</th>
<th>Metric</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tensile Strength, Yield (Type 1, 0.125&quot;, 0.2&quot;/min) PSI</td>
<td>ASTM D638</td>
<td>9,190 psi</td>
<td>63.4 MPa</td>
</tr>
<tr>
<td>Tensile Strength, Ultimate (Type 1, 0.125&quot;, 0.2&quot;/min) PSI</td>
<td>ASTM D638</td>
<td>10,960 psi</td>
<td>75.6 MPa</td>
</tr>
<tr>
<td>Tensile Modulus (Type 1, 0.125&quot;, 0.2&quot;/min) PSI</td>
<td>ASTM D638</td>
<td>1.1 Msi</td>
<td>7515 Mpa</td>
</tr>
<tr>
<td>Tensile Elongation at Break (Type 1, 0.125&quot;, 0.2&quot;/min) %</td>
<td>ASTM D638</td>
<td>1.9%</td>
<td>1.9%</td>
</tr>
<tr>
<td>Tensile Elongation at Yield (Type 1, 0.125&quot;, 0.2&quot;/min) %</td>
<td>ASTM D638</td>
<td>0.9%</td>
<td>0.9%</td>
</tr>
<tr>
<td>Flexural Strength (Method 1, 0.05&quot;/min) PSI</td>
<td>ASTM D790</td>
<td>23,650 psi</td>
<td>142 Mpa</td>
</tr>
<tr>
<td>Flexural Modulus (Method 1, 0.05&quot;/min) PSI</td>
<td>ASTM D790</td>
<td>1.5 Msi</td>
<td>10,620 Mpa</td>
</tr>
<tr>
<td>Flexural Strain at Break (Method 1, 0.05&quot;/min) PSI</td>
<td>ASTM D790</td>
<td>3%</td>
<td>3%</td>
</tr>
<tr>
<td>IZOD Impact, notched (Method A, 23 °C) ft-lb/in</td>
<td>ASTM D256</td>
<td>1.6 ft-lb/in</td>
<td>85 J/m</td>
</tr>
<tr>
<td>IZOD Impact, un-notched (Method A, 23 °C) ft-lb/in</td>
<td>ASTM D256</td>
<td>5.8 ft-lb/in</td>
<td>310 J/m</td>
</tr>
</tbody>
</table>

### Thermal Properties

<table>
<thead>
<tr>
<th>Property</th>
<th>Test Method</th>
<th>English</th>
<th>Metric</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heat Deflection (HDT) @ 264 psi</td>
<td>ASTM D648</td>
<td>289 °F</td>
<td>143 °C</td>
</tr>
<tr>
<td>Melting Temperature</td>
<td></td>
<td>433 °F</td>
<td>223 °C</td>
</tr>
</tbody>
</table>

### Electrical Properties

<table>
<thead>
<tr>
<th>Property</th>
<th>Test Method</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume Resistivity</td>
<td>ASTM D257</td>
<td>5.4E+03 - 3.8E+04</td>
</tr>
<tr>
<td>Surface Resistivity</td>
<td>ASTM D257</td>
<td>3.3E+03 - 6.9E+04</td>
</tr>
</tbody>
</table>
E.3 Construction Drawings for the Joule Heating Extruder

FDM Nylon 12CF DATA SHEET

<table>
<thead>
<tr>
<th>SYSTEM AVAILABILITY</th>
<th>LAYER THICKNESS Capability</th>
<th>SUPPORT STRUCTURE</th>
<th>AVAILABLE COLORS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fortus 450mc</td>
<td>0.010&quot;</td>
<td>Suitable</td>
<td>Black</td>
</tr>
</tbody>
</table>

The information presented are typical values intended for reference and comparison purposes only. They should not be used for design specifications or quality control purposes. End-use materials performance can be impacted (+/-) by, but not limited to, part design, end-use conditions, test conditions, etc. Actual values will vary with build conditions. Tested parts were built on Fortus 450mc @ 0.010" (0.254 mm) slice. Product specifications are subject to change without notice.

The performance characteristics of these materials may vary according to application, operating conditions or end use. Each user is responsible for determining the Stratasys material is safe, lawful and technically suitable for the intended application, as well as for identifying the proper disposal (or recycling) method consistent with applicable environmental laws and regulations. Stratasys makes no warranties of any kind, express or implied, including, but not limited to, the warranties of merchantability, fitness for a particular use or warranty against patent infringement.

1 Build orientation is on side long edge.
2 Literature value unless otherwise noted.
F Supplementary information to chapter 7

The following drawings belong to a simulation (and in parts experiment) series aiming to reduce the cycle time of the IM machine and therefore improve the productivity of the pilot production. The simulations were performed in different configurations including modifications regarding insert size as well as external and internal cooling.

F.1 Construction Drawings of Insert Designs

The first drawing is based on a geometry presented earlier in [262]. Drawings based on the industrial case study are only equipped with the most necessary dimensions in order to allow a scientific reproduction, but prevent the distribution of IP.

The small inserts were manufactured in the workshop of the Department of Mechanical Engineering at the Technical University of Denmark. The last inserts were manufactured by the company Wehl & Partner.
Insert with Small Features

1: Surface Exposed

to Injected Polymer
F.1 Construction Drawings of Insert Designs
Supplementary information to chapter 7

Multi-Scale Insert A

A-A

A

B

81

1

0.27

46x

15.0°
## F.1 Construction Drawings of Insert Designs

### Multi-Scale Insert B

**Dimensions:**
- Length: 60
- Width: 46
- Height: 96

**Notes:**
- Additional scaling and cooling features are included in the design.

---

**Table Layout:**

<table>
<thead>
<tr>
<th>Material</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>A4</td>
<td>Sheet format</td>
</tr>
<tr>
<td>V4</td>
<td>Version</td>
</tr>
</tbody>
</table>

---

**Dimensions:**
- Diameter: 0.5
- Thickness: 0.2

---

**Annotations:**
- A-A View
- B-B View
- 2D and 3D projections

---
Supplementary information to chapter 7
F.1 Construction Drawings of Insert Designs
F.2 Construction Drawings for Standard Cooling
F.2 Construction Drawings for Standard Cooling
F.2 Construction Drawings for Standard Cooling
F.3 Construction Drawings for Para-Conformal Cooling
F.3 Construction Drawings for Para-Conformal Cooling
F.3 Construction Drawings for Para-Conformal Cooling
F.4 Construction Drawings for Multi-Scale, Para-Conformal Cooling
Supplementary information to chapter 7
F.4 Construction Drawings for Multi-Scale, Para-Conformal Cooling
Supplementary information to chapter 7
F.4 Construction Drawings for Multi-Scale, Para-Conformal Cooling
F.5 Construction Drawings for Multi-Scale, Para-Conformal Enhanced Cooling
F.5 Construction Drawings for Multi-Scale, Para-Conformal Enhanced Cooling

![Diagram of Multi-Scale Molding Assembly with Additional Insert Cooling]
F.5 Construction Drawings for Multi-Scale, Para-Conformal Enhanced Cooling
The presented research work contains studies concerning fiber-reinforcement in additive manufacturing, including materials, design, production, quality control, and simulation. Methods were developed to optimize the required properties of the material and the final parts in terms of thermal properties, mechanical properties, lifetime, life cycle, and implementation in a digital production environment. Descriptive research was performed on fiber-matrix interface, fiber orientation, and fiber distribution within the produced part. A specialization was considered for vat photopolymerization technologies. The digital manufacturing process chains of injection molding machines with a special focus on inserts were investigated. Challenges included a reduced lifetime, increased molding cycle time, and thermal management of the insert. Further aspects of digital manufacturing were considered in conceptional investigations regarding advanced cooling of the inserts, automated injection molding prototyping, and design phases. The later will play a significant role in an industry 4.0 manufacturing.