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Applications of Fiber-Reinforced Polymers in Additive Manufacturing

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Abstract

Additive manufacturing technologies are these years entering the market of functional final parts. Initial research has been performed targeting the integration of fibers into additive manufactured plastic composites. Major advantages, among others, are for example increased tensile strength and Young’s modulus. Key challenges in the field, as of now, are proper fiber placement, fiber seizing, an increased knowledge in the used materials and how they are applied into engineering solutions through proper control of the additive manufacturing process. The aim of this research is the improved understanding of fiber-reinforcement in additive manufacturing in terms of production and application. Vat polymerization and material extrusion techniques for composite additive manufacturing were investigated with respect of increasing adhesion between the matrix material and the fibers. Process optimization was performed in order to avoid matrix cracks and delamination.

Keywords: Additive Manufacturing, Plastic Composites, Fiber-reinforcement, Fiber placement

1. Introduction

This paper describes two promising additive manufacturing technologies using Fiber-Reinforced Polymers (FRPs): Vat Polymerization (VP) and Material Extrusion (ME).

VP uses a projection unit in order to expose the photopolymer to crosslink it in a layer-wise manner.

ME extrudes a fiber filled filament through a heated nozzle using the low viscosity of the material at a temperature higher than its melting temperature ($T_m$). The filament is then deformed and deposited for the production of a layer-wise build object.

1.1. Vat Polymerization

Previously performed experiments with Vat Polymerization (VP, also known as Digital Light Processing, DLP) showed the possibility of Additive Manufacturing (AM) as cost-efficient, quick and sustainable alternative for injection molding (IM) inserts [1,2]. IM requires the cavity to withstand pressure and heat cycles during the injection process.

Under the aspect of pilot production, high relative costs for machining tools evolve given a low number of produced parts. Additively manufactured IM inserts were introduced by [3] on a modular basis of standardized cavities. Further advantages are the smaller voxel size which is achievable by VP compared to subtractive manufacturing technologies as pointed out by [1]. This applies especially for corners and sharp edges as well as micro-features.

Nevertheless, the lack of adequate and compatible lifetime made it necessary to develop the technology in a way of increasing the lifetime and decreasing the crack propagation velocity.

VP or similar technologies such as stereolithography (SLA) with FRP have been performed showing the possibility of manufacturing parts using these processes [4–15].

The effect of fibers on the lifetime and surface deterioration of the IM inserts was first investigated by [15] based on the conclusion of [1,2] that significant advantages at the cost, environmental and efficiency levels can evolve. The lifetime of AM inserts made from photopolymer using VP is shorter compared to inserts made from brass or steel and, therefore, lifetime in terms of surface quality was the subject of the research presented in this paper. It was indicated in [2] that photopolymer is advantageous over inserts made from brass and steel in terms of climate change and human toxicity indicators until around 100 shots (against steel) and around 300 shots (against brass) based on an assumed brass and steel insert lifetime of 10 000 shots [2].

The newly performed research contributes to the overall 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 by [16] that composite materials for IM inserts made from poly-
mer and copper particles improved the heat conductivity of the inserts. This investigation was developed further using short carbon fibers placed during the AM process of an insert with dimensions of 3 mm x 19 mm x 19 mm equipped with micro features such as sharp corners, cylindrical cavities and edges as shown in Fig. 1.

1.2. Material Extrusion

Quantitative investigations published by [17] based on the research of [18–28] concluded that a relation between fiber content and tensile strength as well as Young’s modulus exists at reasonable weight fractions of the fiber content up to 30 %. The investigations were based on Material Extrusion (ME) prints with line orientation in the stress direction of the associated tensile tests.

The aim of this research was to investigate in a smaller scale evaluating the exact position and orientation of fibers in a freely extruded filament with carbon fiber and PolyLactic Acid (PLA) matrix material.

Fiber orientation has a significant influence on the material strength and Young’s modulus. The ability of orienting fibers in a polymer matrix allows manufacturing processes to efficiently adapt material properties according to the technical requirements.

2. Methods

2.1. Injection Molding Inserts

A fiber-reinforcement of the IM insert with a mixture of 5 % and 10 % in weight of short virgin carbon fibers was produced with fibers of 7.2 μm diameter and 100 μm average length [29].

The HTM 140 photopolymer was chosen with respect to the thermal and mechanical properties to support the fiber structure as a matrix material and to allow a comparison to [3] for the VP machining.

The insert production was performed by a VP machine exposing a resin vat filled with the above mentioned photopolymer from the bottom. Light intensity was increased by 10 % above the normal exposure value in order to overthrow the obscuring of the resin by the carbon fibers. The layer thickness of 35 μm was set to support the increased density of the photopolymer and carbon fibers. In order to provide an even fiber distribution, the build plate was moved up by 5 mm before increasing the layer height for the next exposure level. This step was performed to allow a fiber flow below the build plate and to avoid a clustering of the fibers at the corners of the build plate.

A flexible self-peeling vat introduced by [30] was used during the process reducing the forces induced into the part during the lifting process.

Warping induced during the AM process due to contraction while the photopolymerization was machined using milling technique for subtracting parts of the rear surface to generate parallel surfaces. This procedure was aiming to prevent deformation and therefore stress within the part during the IM process.

In order to support the hypothesis of increased mechanical strength, tensile tests were performed at a Zwick Roell Z005 machine with a test geometry according to DIN EN ISO 527-2 [31] shown in Fig. 2. 9 samples were tested for each material under the conditions of 10 mm/min, 100 mm/min and 1000 mm/min.

Injections in the IM machine took place using Low-Density PolyEthylens (LD-PE) at the conventional maximum injection pressure of 210 bar for 3 s followed by 10 s packing time and 10 s cooling phase. The inserts were arranged according to Fig. 3 where the LD-PE was injected from the center back and transported into the cavities resulting in a constellation shown in Fig. 4.

2.2. Computed X-ray Tomography

The ME parts were produced using PLA filament with 15 %virgin short carbon-fiber as described in section 2.1.

A 400 μm extruder nozzle was used to extrude the PLA filament slightly over melting temperature at 180 °C. The diameter of the nozzle served as a key factor in the orientation of
the fibers within the matrix. As concluded from the results of numerical simulations by [32], the polymer flow in a polymer extrusion nozzle is subject to a parabolic velocity distribution with strong tendencies of the center velocities. Given the average fiber length of 100 μm, the influence of the orientation of the fibers is influenced to align parallel to the polymer flow.

The extrusion was performed at 30 mm/s in a free environment without deposition on a build plate in order to prevent fiber movement during the change of directions at the point of contact between the polymer strand and the build plate.

The alignment, orientation and density distribution as well as the fiber–matrix interaction within the newly extruded filament was investigated on mesoscopic and macroscopic level: Radiological modalities likeComputed 2-dimensional X-ray Radiography on photostimulated luminescence image storage plates (CR), Computed X-ray Tomography (CT) with human-grade multi-detector 128-line CT and research-grade micro-CT Scanners was performed based on research of [17]. Finally, the material properties were investigated with quantitative Magnetic Resonance Imaging (MRI) methods at 3 T.

3. Results

3.1. Manufacturing

A major difficulty concerning the manufacturing process in VP using photopolymers was detected in terms of the segregation of the carbon fibers with a density of 1.75 g/cm³ [29] in the photopolymer of a density of 1.016 g/cm³ [33].

This segregation made it necessary to stir the mixture after some layers. Given the current development of VP machines, a wiping system should be introduced for a better mixture of the fibers within the resin. This is a phenomenon that has also been described by [12,17].

Clustering of fibers around the corners of the build plate could be eliminated by the above mentioned technique of lifting up the build plate by 5 mm after every layer. A similar effect was already used by [30] introducing a self-peeling vat with a flexible membrane in order to reduce the forces on the part and therefore allow a better sticking to the surface of the build plate. The increased exposure time connected with a higher grade of sticking to the surface made it seem natural to reduce them using the self-peeling vat.

3.2. Tensile Tests

Tensile tests of multiple materials from AM as well as IM production were conducted resulting in an increased Young’s modulus (EMod) for FRP whereas the standard deviation was significantly widened and break stress as well as tensile strength was decreased. (Table 1) The number of flashes indicates the post-curing method in order to reduce any residual solvent from the part. E-TOOLS, Nature resin and RCP 30 are commercial resins which are comparable to the HTM resins and were manufactured in the same manner.

The results provide an comparison of additively manufactured dog bones with IM whereas the influence of the injection pressure was taken into consideration. The high standard deviation compared to the averaged measurements can be explained by the above described defects in the polymer adding uncer-
tainty to the mechanical properties. Compared to IM ABS injected at 450, 400, 350, 300 and 250 bar, Young’s modulus is higher for HTM 140v2, but break strength and tensile strength are smaller. This allows for efficient usage of HTM 140v2 in lower stress levels compared to ABS material. Other photopolymers proved to having smaller Young’s moduli combined with higher break strength and tensile strength.

![Image](93x649 to 270x770)

**Table 1. Results of tensile measurements according to DIN EN ISO 527-2 [31].**

<table>
<thead>
<tr>
<th>Material</th>
<th>E_Mod N/mm²</th>
<th>Break N/mm²</th>
<th>Tensile Strength N/mm²</th>
</tr>
</thead>
<tbody>
<tr>
<td>ABS 250 bar</td>
<td>1972 ± 40</td>
<td>34.03 ± 3.35</td>
<td>45.8 ± 0.8</td>
</tr>
<tr>
<td>ABS 300 bar</td>
<td>2030 ± 47</td>
<td>35.35 ± 2.00</td>
<td>46.1 ± 1.6</td>
</tr>
<tr>
<td>ABS 350 bar</td>
<td>2030 ± 25</td>
<td>35.59 ± 2.36</td>
<td>48.2 ± 0.5</td>
</tr>
<tr>
<td>ABS 400 bar</td>
<td>2099 ± 28</td>
<td>35.41 ± 2.14</td>
<td>48.9 ± 0.6</td>
</tr>
<tr>
<td>ABS 450 bar</td>
<td>1989 ± 100</td>
<td>38.13 ± 3.96</td>
<td>46.9 ± 3.6</td>
</tr>
<tr>
<td>HTM 140v2 5%</td>
<td>2637 ± 154</td>
<td>11.47 ± 6.02</td>
<td>11.5 ± 6.0</td>
</tr>
<tr>
<td>HTM 140v2 10%</td>
<td>2642 ± 286</td>
<td>14.34 ± 7.00</td>
<td>14.6 ± 7.0</td>
</tr>
<tr>
<td>HTM 140v2 2000 flashes</td>
<td>2382 ± 108</td>
<td>15.16 ± 5.44</td>
<td>15.2 ± 5.5</td>
</tr>
<tr>
<td>E-TOOL 2000 flashes</td>
<td>416 ± 132</td>
<td>28.76 ± 3.85</td>
<td>29.9 ± 2.4</td>
</tr>
<tr>
<td>Nature resin</td>
<td>1807 ± 2</td>
<td>60.99 ± 2.08</td>
<td>62.3 ± 2.1</td>
</tr>
<tr>
<td>RCP 30 8000 flashes</td>
<td>1278 ± 79</td>
<td>33.56 ± 2.74</td>
<td>34.4 ± 3.0</td>
</tr>
</tbody>
</table>

### 3.3. micro-CT

The research grade micro-CT was able to determine fiber positions, orientation and size shown in Fig. 5. Fibers arrange in bundles similar to the investigations in VP printing of fiber-filled photopolymers [12]. Air intrusions are likely to occur in this configuration. Post-processing of the acquired data with focus on fiber extraction confirmed the clustering of fibers within the filament. It was moreover shown that the clusters contain fiber groups with a high grade of orientation shown in Fig. 6.

Colors shown in the figure indicate the general tendency of the fiber orientation. The fiber orientation characterizes the general flow of the polymer through the nozzle as well as the deformation after the nozzle exit. The research in this context therefore extends research performed by [32] discussing the velocity and pressure characteristics of an ME nozzle.

### 4. Conclusions

Additively manufactured IM inserts proved to be an environmentally friendly, cheap and fast method for flexible rapid prototyping and pilot production in IM technology despite the low lifetime. FRP in AM technology helped to improve life-time significantly and reduce crack propagation velocity to a minimum.

Short carbon fibers showed an increasing effect on Young’s modulus, but a decreasing one on break strength and tensile strength allowing to efficiently use the material in low-strength regimes where low deformation is needed during a stress-inducing process.

Despite the average increase of Young’s modulus, it was shown that the fiber-matrix interface requires improvement in order to reduce failures in the part and therefore also reducing standard deviation of the mechanical properties such as Young’s modulus, break strength and tensile strength.

Further investigations in CT analysis of fiber-reinforced ME printed parts will be necessary in order to improve the understanding of the grouping effect that not only occurs in resin-based printing, but also when printing thermoplastic materials.

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