OPTIMIZING MECHANICAL PROPERTIES OF ADDITIVELY MANUFACTURED FRPC

Matthias Domm¹, Jens Schlimbach² and Peter Mitschang³

¹ Institut für Verbundwerkstoffe GmbH, Erwin-Schrödinger-Straße, Gebäude 58, 67663 Kaiserslautern (Germany), matthias.domm@ivw.uni-kl.de, http://www.ivw.uni-kl.de
² Institut für Verbundwerkstoffe GmbH, Erwin-Schrödinger-Straße, Gebäude 58, 67663 Kaiserslautern (Germany), jens.schlimbach@ivw.uni-kl.de, http://www.ivw.uni-kl.de
³ Institut für Verbundwerkstoffe GmbH, Erwin-Schrödinger-Straße, Gebäude 58, 67663 Kaiserslautern (Germany), peter.mitschang@ivw.uni-kl.de, http://www.ivw.uni-kl.de

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ABSTRACT

Additive manufacturing methods enable production of very complex structures without the need of a specific tool. However, poor strength and stiffness of neat polymers prevent additively manufactured polymer parts from being used for structural applications. Therefore, a novel additive manufacturing process for FRPC was developed, based on Fused Deposition Modeling (FDM) process. Special features of the so called Fiber Integrated Fused Deposition Modeling (FIFDM) process are the processing of continuous reinforcing fibers, the use of fully impregnated semi-finished products and the placing in all spatial directions without the need for supporting materials. To characterize the FIFDM process and optimize process output and stability as well as quality of the printed parts, a theoretical process evaluation was executed to identify relevant influencing parameters. Furthermore, a test methodology was developed to allow a quantification and comparison of process and print qualities for different parameter settings. Tensile and 3-point bending tests of printed laminates were carried out and compared to autoclave laminates to get an initial classification of mechanical properties of such printed structures. A parameter study shows the influence of conveying speed, nozzle offset and nozzle temperature on quality of printed laminates.

1 INTRODUCTION AND MOTIVATION

Additive manufacturing methods enable production of very complex structures without the need for a specific tool. This leads to several production advantages such as high degree of part individualization, high material efficiency, as well as minimal tooling costs. In recent years, additive manufacturing is no longer only considered as a gadget for fast and flexible production of prototypes and decoration elements, but is becoming increasingly important for industrial applications. According to a recent study published by Ernst & Young [1], already 24% of the overall interviewed companies are using additive manufacturing in different applications. While 38% aim to implement additive manufacturing in their serial production within the next few years, only 5% of those have started so far. This predicts a great growth of this still young technology but also confirms the need for extensive research in this field. Besides optimization of process technology, especially development of new material groups is crucial to exploit the full industrial potential.

Additive manufacturing processes are mostly used for metallic, ceramic, and polymer materials. Due to their low density, polymers are particularly well-suited for lightweight applications. Furthermore, thermoplastic polymers can be melted and solidified reversibly which allows very fast processes with high material output. However, poor strength and stiffness of neat polymers prevent additively manufactured polymer parts from being used for structural applications. In order to overcome this drawback, polymers can be reinforced, e.g. with glass, carbon, or aramid fibers. While discontinuous reinforcement fibers have been implemented in additive manufacturing processes and initial efforts for the integration of continuous fibers in-plane have been done so far, only continuous fiber reinforcement in load direction guarantees the maximum effect aiming at high mechanical and
lightweight properties like specific strength and stiffness of Fiber Reinforced Polymer Composites (FRPC). Hence, the objective of this work is to overcome challenges regarding the integration of continuous fiber reinforcements into polymer additive manufacturing processes.

2 APPROACH AND RESEARCH OBJECTIVES

There are several additive manufacturing processes for polymers. All processes can be divided in radiation-curing processes (e.g. Stereolithography), sinter processes (e.g. Selective Laser Sintering), and extrusion-based processes (e.g. Fused Deposition Modeling) [2]. Extrusion-based processes are most suitable for the integration of continuous fibers, since the raw material is already a continuous polymer filament being placed in a semi-continuous process. Therefore, efforts were made regarding the additive manufacturing of continuous reinforced thermoplastics, based on the Fused Deposition Modeling (FDM) method [3]. Within the FDM process, a polymer filament is pressed through a heated nozzle where it is melted. The extruded material is then placed on a print bed, building up the part geometry layer by layer. For the integration of continuous fibers into the new Fiber Integrated Fused Deposition Modeling (FIFDM) process, an impregnated continuous Fiber Reinforced Polymer Strand (FRPS) were identified to be most suitable aiming at fast process speeds while maintaining high fiber volume content and high impregnation quality. [3]

There are already possibilities to reinforce additively manufactured polymer parts with short fibers and fillers [4, 5]. Furthermore, initial efforts regarding the integration of continuous fiber reinforcement in the FDM process were made [6-10]. Unique characteristic of the FIFDM process presented in this paper is the processing of fully impregnated continuous fiber reinforced polymer strands, so that the impregnation is sourced out of the printing process. In addition, the screened processes with except of [10] are only able to place the extruded material in-plane.

To exploit full potential of the continuous fiber reinforcement, two key challenges arise:

- Load-specific orientation of the fibers
- High laminate quality

The first challenge has been addressed in a previous study [3]. The reinforcing effect of continuous fibers is strongly dependent on the fiber orientation, i.e., the mechanical properties differ by a factor of 10 between fiber direction and perpendicular to fiber direction [11]. Therefore, the process was further developed to print not only layer by layer but also out-of-plane in all spatial direction without the need for supporting materials. This enables a load-specific three-dimensional orientation of the fibers in the final part. The findings of the study were implemented into new manufacturing devices like extruder unit, cooling system, and cutting unit for an X400 FDM-printer by German RepRap.

Based on the preliminary developments, aim of the research work presented in this paper is to contribute to optimization of process output, stability, as well as quality of the printed structure for the new FIFDM technology. Subsequently, a test methodology is developed to allow a quantification and comparison of process and print qualities for different parameter settings. Tensile and bending parameters are determined and classified by autoclave reference. Finally, the influence of different process parameters on the mechanical properties and therefore on the laminate quality of printed structures is investigated.

3 MATERIALS AND SETUP

Specimens for the required tests were manufactured by a modified X400 FDM-printer with new extrusion unit for processing of continuous fibers. The printer with an assembly space of 400 x 400 x 400 mm³ is shown in Figure 1, left side.

On the right side of Figure 1, the semi-finished product used as FRPS is presented. It is a continuous glass-fiber reinforced Polypropylene (GF-PP) strand. The FRPS has an oval shape with a diameter of approximately 1.9 mm, a fiber volume content of approximately 30%, and a void content of 6.44 ± 1.4 vol%.
Figure 1: German RepRap x400 FDM-printer (left side), GF-PP strand (right side).

4 RESULTS

The following chapter describes the results of theoretical process evaluation, development of test methodology, and mechanical characteristic of printed laminates.

4.1 Theoretical Process Evaluation

To optimize the FIFDM process regarding output, stability, and print quality, a theoretical process evaluation was carried out to identify relevant process variables and influencing factors. Figure 2 shows the schematic assembly of the FIFDM process. The process is divided into the sub process steps: pre-heating, conveying, extrusion, and placing. Within the pre-heating process, the FRPS is heated up to a temperature close to melting point ($T_m$) in order to reduce the temperature gradient which has to be applied in the print nozzle. Since heating of the FRPS is the limiting factor, the pre-heating unit enables higher process speeds. The conveying process applies the force to pull off the FRPS and press it through the printing nozzle. Thus, the conveying unit defines the process speed. In the print nozzle, the actual extrusion process takes place. The FRPS is heated up to process temperature ($T_{process} > T_m$). Due to discrepancy of FRPS shape regarding print nozzle hole, a pressure is applied on the FRPS changing its outer contour and inner structure. After extrusion, the material is placed during the placement process either in free space or on top of printing bed respectively, on already placed structures. By using the cooling device an intelligent temperature management can help to control the outer contour, the placing deviation and the warping of the extruded FRPS, especially for placing into free space.

All process steps and, of course, the used semi-finished product have several variables that influence process output and stability as well as printing and part quality. All relevant influencing factors are given in Figure 2.

Figure 2: Process steps and influencing parameters.
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In the present paper, a test methodology for the investigation of extrusion and placing process is developed. Based on that, the influence of following parameters on laminate quality were investigated:

- Conveying speed: determined by conveying rollers
- Nozzle temperature: controlled temperature of print nozzle
- Nozzle offset: distance from print nozzle to printing bed respectively already placed structures

The parameters were chosen because time, pressure and temperature play a major role for consolidation and therefore quality of laminate structures.

4.2 Test Methodology for Investigation of Extrusion and Placement Process

To receive optimal comparability of the examined print parameter settings, the test methodology consists of microscopic and macroscopic analysis as well as mechanical experiments. A distinction is drawn between analysis of the extrusion process, the placing of single FRPS, and the placing of coherent structures, like laminates. Table 1 gives an overview of the determined characteristics and the considered test methods.

<table>
<thead>
<tr>
<th>Influence on</th>
<th>Quantification</th>
<th>Test Method</th>
</tr>
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<tbody>
<tr>
<td>Print quality</td>
<td>Quality of Single Extruded FRPS</td>
<td>Micrograph: grayscale analysis</td>
</tr>
</tbody>
</table>
| Extrusion process | • Void content |}
| Placing of single extruded filaments | • Fiber orientation | Computer tomography |
| Placing of laminates | • Fiber distribution | Micrograph: center of fibers |
| • Outer Shape | • Micrograph: roundness |

| Part quality | Outer Shape | Caliper |
| Placing of laminates | • Dimensions, Warping | White-light interferometer |
| • Surface | | |

**Table 1: Test methodology for investigation of extrusion and placement process**

Hereafter, the focus will be on analysis of mechanical characterization, supported by microscopic test methods like light microscopy and micro computed tomography. The different test methods are discussed regarding the applicability as instrument for the analysis of the extrusion and placement process of the FIFDM process. Furthermore, first mechanical characteristics are determined and compared to autoclave reference.

4.3 Determination of Tensile and Bending Characteristics

Tensile and bending characteristics are determined by following the common standards for FRPC DIN EN ISO 527-5 (specimen type A) and DIN EN ISO 14125 (specimen type IV). The specimen geometry was adjusted to height restriction for the FIFDM process, resulting from the dimension of the FRPS. Tests were carried out with a universal testing machine by Zwick (type 1474, max. 10 kN) and 5 repetitions each. The print parameter settings (nozzle temperature: 235 °C; nozzle offset: 1.2 mm; conveying speed: 60 mm/min) were determined in pre-studies [3], ensuring a stable process also for large specimens. To enable a top benchmark, the mechanical characteristics are compared to an almost idealized laminate. Therefore, a plate was produced in an autoclave using the same FRPS. Temperature for autoclave manufacturing was 190 °C, pressure 24 bar, heating time 20 min, and holding period 60 min. Tensile and bending specimens were cut out of the plate following dimension specifications of above listed common standards.
Printed tensile specimens are built up out of one layer with six paths of parallel placed extruded FRPS. The average dimensions of printed tensile specimens are 250.00 x 14.56 x 1.54 mm³, and 250.00 x 14.47 x 1.50 mm³ for autoclave specimens. The specimens for 3-point bending are built up out of two layers with six paths of parallel placed extruded FRPS each. The average dimensions are 60 x 14.94 x 2.88 mm³ for printed and 60 x 14.93 x 2.30 mm³ for autoclave specimens.

The diagram in Figure 3 compares the results from the tensile experiments for printed (FIFDM) and autoclave samples showing maximum tensile strength and tensile modulus. In addition, representative pictures of printed and autoclave specimens before and after testing are presented.

**Figure 3: Comparison of tensile tests between FIFDM and autoclave specimens**

The tested specimens in Figure 3 show a mechanical failure in the center, representing a valid tensile test. Both printed and autoclave specimens have a separate FRPS failure. In the pictures of the tested specimens the initial FRPS can be detected. It is also noticeable that the tensile strength of the printed sample is 34% and stiffness 39% lower than for the autoclave samples. The result is surprising due to the fact, that the fibers in either case are orientated in load direction and bonding of the single extruded FRPS respectively consolidation quality should play a minor role.

The diagram in Figure 4 compares the results from the 3-point bending experiments for printed (FIFDM) and autoclave specimens showing maximum bending strength and flexural modulus. In addition, representative pictures of printed and autoclave bending specimens before and after testing are presented.

**Figure 4: Comparison of 3-point bending tests between FIFDM and autoclave specimens**
Also the bending specimens in Figure 4 show a mechanical failure in the center, representing a valid 3-point bending test. While printed specimens failed by pressure load on the top side of the specimen, autoclave specimens show a combined failure of pressure and tensile load on top and bottom side. Beyond that, for several of the printed specimens cracks occur between the two layers (Figure 4 side view).

The influence on the mechanical properties turns out stronger for bending than for tensile experiments, considering the difference between printed and autoclave structures. Maximum bending strength of the printed structure is 79% and flexural modulus 80% lower than the comparative autoclave values. For the printed specimens, lower bending values were expected, because of the anticipated worse consolidation of the FRPS. The cracks between the layers of printed specimens indicate this assumption. Even though, the extent of discrepancy is significant.

**Analysis of the Laminate**

To explore the big difference between the tensile and bending results of printed and autoclave specimens, the specimens are analyzed by optical test methods. Especially for bending characteristics, the difference is significant. Therefore, bending specimens are further investigated by microscopic test methods to determine the cause for the lower performance. Figure 5 shows the micrographs of an untested autoclave and printed 3-point bending specimen.

![Cross Section of 3-Point Bending Specimen (Autoclave)](image)

![Cross Section of 3-Point Bending Specimen (FIFDM)](image)

**Figure 5: Micrographs of 3-point bending specimens manufactured by autoclave and FIFDM**

The cross section of the autoclave specimen shows an almost homogenous and consolidated structure with a minor proportion of small voids. Only on closer inspection, the single FRPS become apparent by the distribution of fibers. This is an indication for the separated FRPS failure of the autoclave tensile specimens, which was detected for the tensile tests in Figure 3.

However, in the cross section of the FIFDM printed specimen, all FRPS still exist as separate unit. Therefore, the surface is significantly rougher compared to autoclave specimen. Warping perpendicular to fiber direction can be detected on the left side of the micrograph. In addition, the void content about 9.6 vol% is also much higher, consisting of voids inside the FRPS, which could not be closed during the extrusion process, and also voids and gaps between the FRPS resulting from the round shape of the FRPS. The observations indicate a worse bonding between the placed FRPS and therefore also a worse consolidation of the laminate. This strongly affects the mechanical properties of a printed product and lead to lower strength and stiffness values.
Beyond that, it can be detected that the fibers in most of the extruded FRPS are shifted to the top of the particular FRPS. The reason for this effect is assumed in the placement process. Between extrusion and placing on the print bed, the FRPS is temporarily bended by an angle of 90°. Because all fibers have the same length and are not stretchable, fibers following a smaller radius are crimped while fibers following a wider radius are shifted to smaller radii. Crimped fibers are straightened again when the FRPS is aligned on the print bed. Fibers shifted to the top of the extruded FRPS stay in their position because of a missing restoring force.

To get a better impression of the fiber orientation and distribution Figure 6 presents views out of the investigation of a printed but untested 3-point bending specimen by computer tomography.

![Micro computer tomography images of 3-point bending specimens](image)

**Figure 6: Micro computer tomography images of 3-point bending specimens**

The images following view direction 1 demonstrate that the round shape of the GF-PP FRPS leads to an inhomogeneous fiber distribution. In addition, the image following view direction 2 clearly shows that a considerable amount of fibers is not orientated straight in FRPS direction but following undulated paths. As an analysis of the FRPS before printing showed that only 3% of the fibers have a deviation of more than 5° to FRPS direction, the printing process induces the fiber deviation.

Considering all mentioned observations by optical test methods, it becomes obvious that the loss in tensile and bending characteristics compared to autoclave structures can be explained by a worse bonding of the FRPS, lower consolidation quality of the laminate, and disorientation of the fibers. The main reason for the difference in laminate quality is suggested in the lower pressure and pressure time that can be applied in the FIFDM process. A possibility to compensate this drawback could be the increase of process temperature, as in a further tape placement study a decline of pressure influence for higher tape temperatures on consolidation quality has been found [13]. Additional solutions like consolidation devices or squared FRPS result in a limitation of process and geometry flexibility.

**Measurement Error Analysis**

Due to the big difference between manufacturing process and structure of printed and autoclave specimens, a measurement error analysis was carried out. As illustrated in Table 2, several measurement errors occur that lead to an underestimation of tensile and bending results of printed specimens compared to autoclave specimens. A rough surface can result in an error, measuring the dimensions of the cross section. In addition, the round shape of the FRPS as well as warping effects enlarging the measured cross section area for printed specimens. As autoclave specimens are cut out of a plate and have a significantly better consolidation than printed specimens, the overall material usage of autoclave specimens is higher for the same measured volume compared to printed specimens. Particularly the different amount of fibers lead to an underestimation of the tensile and bending results for printed specimens. To quantify the difference in material usage, the effective material usage both
for printed and autoclave specimens were determined. Calculations show 8% higher material usage for tensile autoclave specimens and 11% higher material usage for bending specimens compared to particular printed specimens. To avoid different material usage, autoclave specimens have to be manufactured with the same amount of FRPS. However, this leads to different dimensions of printed and autoclave specimens which also can affect the mechanical test results.

Beyond that, the rough surface leads to an uneven force application in the clamping area of the tensile test setup. As shown in Table 2, this results in an inhomogeneous force distribution, so that the FRPS are not charged equally at the same time or even slide in the clamps. For bending specimens the rough surface and also the warping perpendicular to FRPS direction lead to an unsteady placement on the 3-point bending supports and also to an inhomogeneous load application by the stamp. In both cases, this results in a reduction of mechanical values. Therefore, printed tensile specimens have to be reinforced at the ends with cap strips.

<table>
<thead>
<tr>
<th>Measurement Error Analysis</th>
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<tbody>
<tr>
<td>Tensile Test</td>
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<tr>
<td>Overestimation of cross section by measurement with caliper</td>
</tr>
<tr>
<td>Lower material usage of printed compared to autoclave specimens</td>
</tr>
<tr>
<td>Inhomogeneous load application and distribution</td>
</tr>
</tbody>
</table>

*Table 2: Measurement error analysis for tensile and bending experiments*
4.4 Comparison between Test Methods for Analysis of Process Parameters

Based on the knowledge gained by determination of tensile and bending characteristics of printed FIFDM laminates, a test method must be found to optimize mechanical properties of printed structures. Thereby, it is more important to enable a good comparability for the variation of print parameter settings than generating absolute values. Peel, short beam, 4-point and 3-point bending tests were compared to each other regarding to test effort and significance of test results. Experiments for every test method with three varying nozzle offset settings were carried out. The four different test methods with its advantages and disadvantages are listed in Table 3.

<table>
<thead>
<tr>
<th>Test Method</th>
<th>Setup</th>
<th>Advantages</th>
<th>Disadvantages</th>
</tr>
</thead>
</table>
| Short Beam Bending   | ![Short Beam Bending](image) | + Measure for the bonding of layers  
+ Small specimens  
+ Small testing effort | - No definite mode of failure |
| Peel Test            | ![Peel Test](image) | + Measure for the bonding of layers | - Test method for thin tapes  
- Wide variations  
- Large specimens |
| 4-Point Bending      | ![4-Point Bending](image) | + Reproducible results  
+ Bending load without shear stress | - No definite measure for layer bonding  
- High testing effort |
| 3-Point Bending      | ![3-Point Bending](image) | + Reproducible results  
+ Small testing effort | - No definite measure for layer bonding |

*Table 3: Comparison of test methods for analysis of process parameter influence on structures manufactured by FIFDM*

Short beam bending and peel test have in common, that they are a direct measure for the bonding of the single layers and therefore also for the consolidation of the whole laminate. Short beam bending is a bending test, trying to maximize the shear forces in the center of the specimen by minimizing the distance of the supports to measure the interlaminar shear strength. However, with short beam bending tests, no definite mode of failure could be achieved. As the test is designed for stiff polymer systems, it is assumed that PP is too soft to cause shear strength failure.

Within the peel test, a wedge is placed in a crack between the two layers of the specimen. The specimen is then pulled through the stationary wedge. The results of the peel test, which initial is designed for thin tape materials, showed wide variations causing a massive restriction of the significance of the results.

The results of the 3-point and 4-point bending test cannot directly be addressed to the bonding of the laminate layers. Nevertheless, they are a good indicator for laminate quality. 4-point bending test has the advantage that in the testing zone of the specimen between the two stamps, there is a shear tension free area. This allows bending characteristics without superimposed shear forces. However, the measuring effort is significant higher due to the fact that the traverse movement cannot be used as extent of deflection. Because only comparative values are required, 3-point bending test is chosen for further investigation of influencing parameters on printed laminates.

4.5 Parameter Study

Following the test methodology, 3-point bending experiments were carried out to investigate the influence of process parameters on quality of laminates printed by FIFDM. As time, pressure, and temperature play a major role for consolidation of laminates, in a first parameter study conveying speed, nozzle offset, and nozzle temperature are varied based on reference settings (conveying speed: 60 mm/min; nozzle offset: 1.2 mm; nozzle temperature: 235 °C). The 3-point bending characteristics were determined by following the common standards for DIN EN ISO 14125 (specimen type IV). The specimen geometry was adjusted to height restriction. The tests were carried out with a universal testing machine by Zwick (type 1474, max. 10 kN) and 5 repetitions each.

Figure 7 shows the results of the 3-point bending tests varying conveying speed. Besides the reference two faster speeds are tested at a distance of 30 mm/min each.
Figure 7: 3-point-bending characteristics for specimens printed by FIFDM with varying conveying speed

No definite trend can be detected with regard to the results given in Figure 7. While the strength decreases for higher conveying speeds the stiffness even increases for 120 mm/min. In addition, high standard deviation complicates to define a conclusion.

The influence of the nozzle offset on the mechanical properties of the printed structure is given in Figure 8. Due the fact that 1.2 mm already is the closest reasonable distance for the given FRPS diameter, two larger offsets were further investigated.

Figure 8: 3-point-bending characteristics for specimens printed by FIFDM with varying nozzle offset

As expected, the highest bending performance can be achieved with the lowest offset. Higher offset settings lead to a massive decline of the mechanical properties. The results indicate, that the higher nozzle offset induce a decrease of pressure which leads to a worse consolidation of the laminate. An increasing void content for higher offsets (9.6 vol% for 1.2 mm, 13.3 vol% for 1.7 mm, and 26.6 vol% for...
for 2.2 mm) supports this statement.

Bending strength and flexural modulus of 3-point bending tests varying nozzle temperature are presented in Figure 9. In addition to reference temperature, two higher temperatures (255 °C and 275 °C) are investigated.

![Influence of Nozzle Temperature on Bending Performance](image)

**Figure 9: 3-point-bending characteristics for specimens printed by FIFDM with varying nozzle offset**

The results in Figure 9 show a small dependency of higher bending strength for increasing nozzle temperatures, but a significant growth of flexural modulus. Hence, better consolidation of the laminate for higher temperatures can be assumed. One reason for this is the decreasing viscosity of the PP melt for increasing extrusion temperatures. Also, the already placed FRPS can be heated to higher temperatures, supporting a better bonding.

5 CONCLUSION

Based on a novel additive manufacturing process for the processing of continuous fiber reinforcing thermoplastics investigations are done to optimize process output and stability as well as quality of the printed parts.

In a first step, a detailed theoretical process evaluation was carried out to identify relevant influencing parameters. Furthermore, a test methodology is developed to allow a quantification and comparison of process and print qualities for different parameter settings. An initial classification of tensile and bending characteristics show a big gap compared to autoclave reference. While printed laminates are vulnerable to measurement errors, underestimating the mechanical characteristics, main reason is the worse consolidation of the printed structures due to lower process pressure and pressure time. Finally, a parameter study was carried out to investigate the influence of conveying speed, nozzle offset and nozzle temperature on laminate quality. Results show a dependency between lower mechanical properties and higher conveying speeds, significant lower properties and larger nozzle offset as well as, an increase of mechanical properties and higher nozzle temperatures. As one of the objectives is to increase process output a further decrease of conveying speed is not taken into account. Minor nozzle offset is unfeasible as well, due to the given diameter of the GF-PP FRPS. Greatest opportunity to increase mechanical properties is promised by investigation of the temperature profile of the FRPS during extrusion and placement process. Further improvement could also be achieved by applying the cooling unit that has not been used for the manufacturing of the laminates presented in this paper.

The acquired knowledge of the research work shall be used to improve output and stability of the process as well as quality of the printed parts. Furthermore, design guide-lines for the development of a robot-controlled end effector for the printing of continuous FRPC into free space will be determined.
REFERENCES


