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Development of a Novel Hybrid Manufacturing Technology for Continuous Fiber-Reinforced Thermo-Plastic Composites

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Abstract

In this study, we present a novel approach for the production of continuous fiber-reinforced thermoplastic composites by combining injection molding and additive manufacturing. After exploring the design requirements, we manufactured inserts via continuous fiber-reinforced 3D printing, then we used them as reinforcement for injection-molded samples. Improper fiber placement can cause warpage as the continuous fibers prevent shrinking; however, warpage can be compensated with the insert geometry. The reinforcement resulted in an increase of about 30 % in the properties tested.

Keywords: *additive manufacturing, injection molding, fiber-reinforced polymer composite.*

1. Introduction

The additive manufacturing of continuous fiber-reinforced composites is one of the fastest growing polymer manufacturing technologies today. 3D-printed composites have the advantage of designable fiber orientation (even along several degrees of freedom [1], but the technology has not yet reached its potential in terms of mechanical properties, and cycle times are still relatively high [2, 3].

On the other hand, injection molding is one of the fastest ways of producing thermoplastic composites. The common practice in the industry is still the use of short fibers, and to increase the length several methods have been reported lately [4, 5]. However, besides the fiber length, the orientation is also important to achieve the desired increase in mechanical properties. The orientation is determined by the melt flow direction, which does not necessarily coincide with the stress directions [6, 7]. The need therefore arises for tailorable fiber properties in injection-molded composites.

Technologies for continuous fiber-reinforced injection-molded products have also emerged [8, 9]. Usually, the fiber-reinforced preform or sheet is placed in the mold and then the thermoplastic polymer matrix is injected onto it [10]) Usually, the fiber-reinforced preform or sheet is placed in the mold and then the thermoplastic polymer matrix is injected onto it. The procedure is often referred to as overmolding as well [11, 12]. The sheets are mostly reinforced thermoplastic laminates with a specific fabric orientation and layup sequence, which provides the strength and stiffness comparable to that of thermosetting composites, however, this type of reinforcement is restricted in terms of design freedom. Fiber placement and orientation in the product is strictly determined and the fiber content can only be varied by the number of laminas.

Extrusion-based additive manufacturing of continuous fiber-reinforced composites might be a promising alternative. 3D printing can be used to create customized reinforcing structures directly where stresses apply, thus reducing material and the costs as well [13].

In this research, we investigate the applicability of 3D-printed composites for overmolding. We present the design requirements of a 3D-printed composite insert and the property modifying effects of the continuous fibers in the injection-molded product.

2. Materials and methods

2.1. Materials

For the 3D-printed inserts, Markforged "Tough Nylon" (later called "End of life Nylon") filament was used as matrix material. The filament is stored in a drybox. Markforged Continuous Carbon Fiber filament was used as reinforcement [14], According to the literature, the filament contains 1000 single fibers [15].

For injection molding (overmolding) Alphalon 27 type PA6 was used from Grupa Azoty ATT Polymers [16]. Before molding the granulate was dried at 80 °C for 4 hours based on the manufacturer's recommendations.

2.2. Methods

The inserts were prepared with a Mark Two (Markforged, USA) type FFF-based composite 3D printer. For slicing, the company's cloud-based software (Eiger) was used. Main printing parameters are shown in Table 1 where the values fixed in the slicer are marked. Parameters regarding the matrix material (Fill pattern and Fill density) were set to produce a solid structure with as few voids as possible. The fiber reinforcement was placed along the outer walls. We aimed to achieve the maximum fiber content at minimum wall thickness applicable, thus creating strong and lightweight inserts. The different insert geometries allowed for different amounts of fiber to be placed, therefore fiber volume fractions also varied. The fiber volume fractions were determined with Eq. (1) and the volumes were calculated within the slicer.

$$v_f = \frac{V_f}{V_c} \quad (1)$$

where V_f (cm^3) is the fiber volume content and V_c (cm^3) is the composite volume.

Before injection molding, simulations were run for the "U" and "K" type inserts using Moldflow (Autodesk, 2016). The aim of the simulations was to visualize the melt flow around the inserts and to determine the mold temperature that could theoretically ensure the conditions for polymer bonding. CAD models were prepared dusing Autodesk Inventor Professional (Autodesk, 2020) and the meshes were prepared in Hypermesh (Altair Hyperworks, 2017.1).

Injection-molded specimens were prepared with an Arburg Allrounder Advance 270S 400-170 injection molding machine at zone temperatures of 255 °C, 260 °C, 265 °C, 270 °C, 270 °C, a mold temperature of 80 °C and an injection pressure of 1500 bar. The injection molding parameters were determined based on the simulation results and the manufacturer's recommendations [16].

Quasi-static flexural tests were performed on a Zwick Z005 type machine according to the MSZ EN ISO 178 standard [17] on at least 5 samples. Support distance was 64 mm and the test speed was 5 mm/s.

Table 1. *Printing parameters*

Parameter	value
Fill pattern	Solid
Fill density	100 %
*Fill orientation	45°
Fiber infill type	concentric
*Nozzle diameter	0.4 mm
*Nozzle temperature	270 °C

3. Design of the composite inserts

3.1. Geometry

Continuous fiber-reinforced inserts were designed for injection-molded specimens with the purpose of increasing the flexural properties. Three main groups of requirements were defined for the insert geometries. First, the design must meet the printability requirements determined by the equipment and the slicing software. Second, the geometry must be applicable for overmolding which imposes constraints on geometry (simply put, it should fit the mold) and material use (the inserts must withstand the shear, pressure, and temperature of the polymer melt). Finally, the end product must have adequate bonding between the matrix and the reinforcement that lasts until failure, and the inserts must enhance one or more chosen mechanical properties.

First, two types of inserts were designed as shown in Figures 1a–b. Then, based on the experiences of the first overmolding, a third geometry was also designed (Figure 1c). For each type, the fibers are placed parallel with the longitudinal axis, so that the reinforcement is in the part of the product most exposed to stress. The placement of an insert in the mold can be seen in Figure 2. The tolerance of the external dimensions is designed for a tight fit so that the inserts do not fall out during mold closing. Fiber volume fractions of the inserts are shown in Table 2.

Table 2. *Fiber volume fractions of the inserts*

Insert type	vf (%)
U	26,9
K	8,3
W	10,5

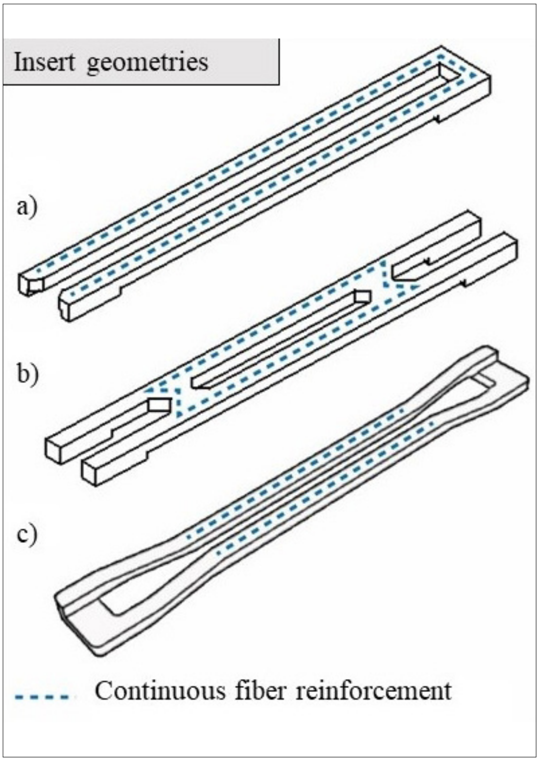

Figure 1. *Schematics of the insert geometries: a) type "U" b) type "K" c) type "W". The dashed line indicates the planned placement of the continuous fiber reinforcement*

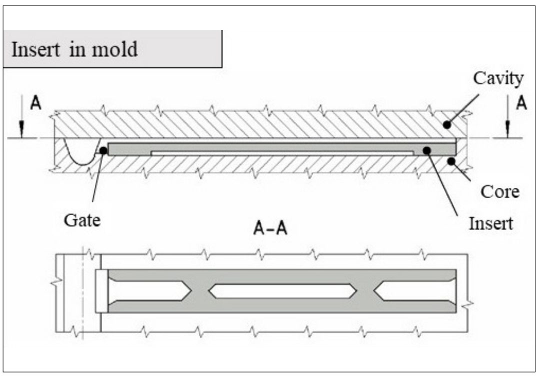

Figure 2. *Schematics of the type "K" insert in the mold.*

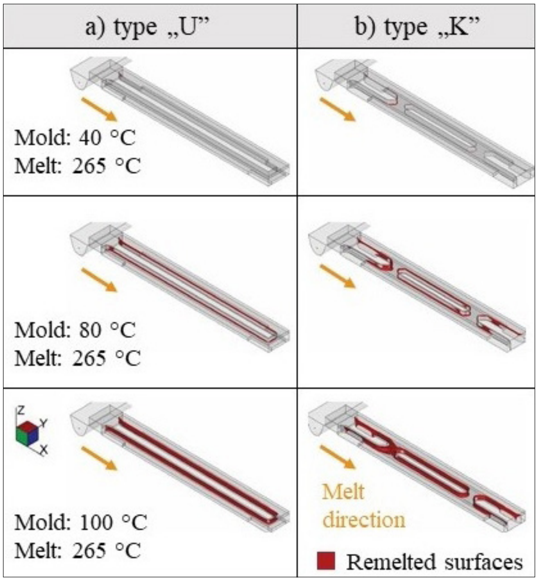

Figure 3. *Remelted surface areas as a function of mold and melt temperature.*

3.2. Injection molding simulations

Injection molding simulations were run to gain information about the melt flow and the expected bonding between the inserts and the injection-molded matrix.

The remelted surface areas of the inserts were examined as a function of mold temperature. Results can be seen in **Figure 3.** As expected, the amount of remelted surface areas will increase with higher temperature, therefore – assuming the use of compatible materials – better bonding can be expected. These results can provide the basis for the selection of the injection molding parameters. It can also be seen that the injected melt is expected to fill the gaps around and within the inserts. This is of great importance as voids would serve as failure locations.

4. Insert preparation and injection molding

The inserts were 3D printed, then placed in the mold manually. Then the matrix material was injected around the inserts. The first overmolding experiments showed a significant warping of the specimens. This is because after ejection the polymer shrinks, but the continuous carbon fibers do not, therefore the fiber reinforcement blocks the deformation where it is placed. Thus, in case of asymmetric fiber placement, the degree of shrinkage differs at the sides of the sample which

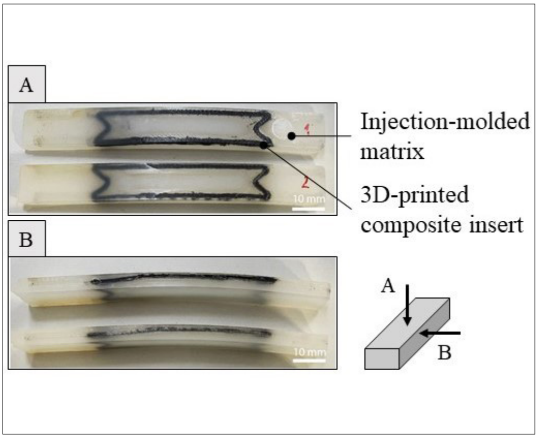

Figure 4. *Injection-molded specimen reinforced with type "K" insert*

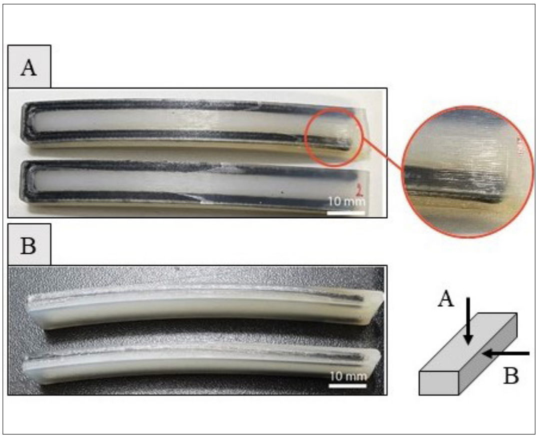

Figure 5. *Injection-molded specimen reinforced with type "U" insert*

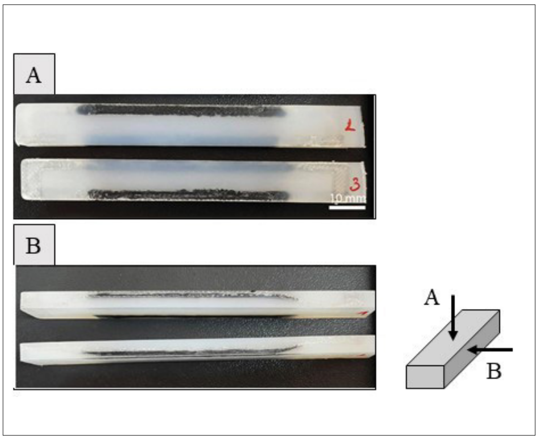

Figure 6. *Injection-molded specimen reinforced with type "W" insert*

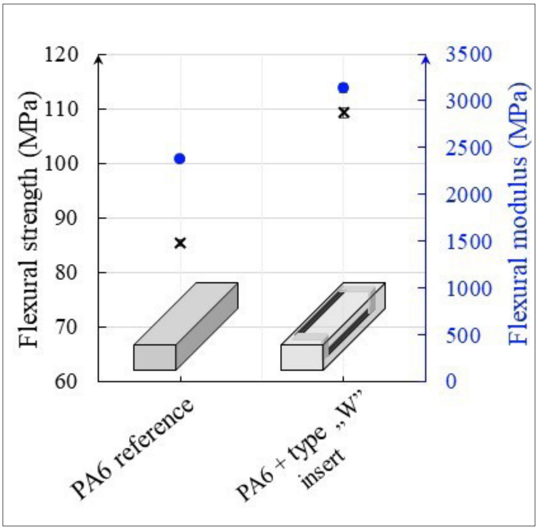

Figure 7. *Flexural strength and modulus of the unreinforced reference and the composite samples*

results in warpage. Figure 4 and 5 shows the injection-molded specimens reinforced with the type "K" and "U" inserts, respectively. Warpage is clearly visible in both cases.

A slight change in color can be seen along the walls of the inserts, similar to the burn marks of the diesel effect (Figure 5). There can be several reasons for this. The inserts may have prevented proper ventilation, and the entrapped gas caused ignition and therefore thermal degradation. The shear forces near the gate could also exceed the limit of the 3D-printed polymer. These results show that overmolding requires different parameters than traditional injection molding, and one must consider the material parameters of the inserts as well.

To compensate warping, a third insert geometry (type "W") was designed, in which equal amounts of reinforcing fibers are placed along the opposite sides. The symmetric fiber placement compensated shrinking and therefore warpage was visibly reduced (Figure 6).

5. Flexural mechanical properties

3-point bending was performed to investigate the effect of the reinforcement on the flexural mechanical properties. Results can be seen in Figure 7 Due to the warping of the type „U" and type „K" reinforced samples, only the type „W" specimens could be tested. It can be seen that the inserts increased the flexural stress and the modulus by approximately 30 %. The relatively small standard deviations suggest that the technology is well reproducible.

6. Conclusions

In this study, a novel hybrid technology is presented for the production of continuous fiber-reinforced thermoplastic composites. Carbon fiber-reinforced structures were produced with 3D printing, then the composites were placed in the mold and the polymer matrix was injected on them. Injection molding simulations were also run before sample preparation. It was found that improper fiber placement can cause warpage as the continuous fibers prevent shrinking, however, it was also presented that warpage can be compensated with the insert geometry. The composites produced with overmolding showed a 30 % increase in flexural strength and modulus compared to the unreinforced samples. Overall, the hybrid technology presented offers the potential for the productive manufacture of recyclable, continuous fibre-reinforced products with the possibility of customization as well.

Acknowledgements

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References

[1] van de Werken N., Tekinalp H., Khanbolouki P., Ozcan S.: *Additively manufactured carbon fiber-reinforced composites: State of the art and perspective.* Additive Manufacturing, 31 (2020) 100962.
https://doi.org/10.1016/j.addma.2019.100962
[2] Liu S., Li Y., Li N.: *A novel free-hanging 3D printing method for continuous carbon fiber reinforced thermoplastic lattice truss core structures*. Materials and Design, 137. (2018) 235–244.
https://doi.org/10.1016/j.matdes.2017.10.007
[3] Wang X., Jiang M., Zhou Z., Gou J., Hui D.: *3D printing of polymer matrix composites: A review and prospective.* Composites Part B: Engineering, 110. (2017) 442–458.
https://doi.org/10.1016/j.compositesb.2016.11.034
[4] Tábi T., Égerházi A. Z., Tamás-Bényei P., Czigány T., Kovács J. G.: I*nvestigation of injection moulded poly(lactic acid) reinforced with long basalt fibres.* Composites Part A: Applied Science and Manufacturing, 64. (2014) 99–106.
https://doi.org/10.1016/j.compositesa.2014.05.001
[5] Yan X., Cao S.: *Structure and interfacial shear strength of polypropylene-glass fiber/carbon fiber hybrid composites fabricated by direct fiber feeding injection molding*. Composite Structures, 185. (2018) 362–372.
https://doi.org/10.1016/j.compstruct.2017.11.037
[6] Yu S., Hwang J. Y., Hong H. S.: *3D microstructural characterization and mechanical properties determination of short basalt fiber-reinforced polyamide 6,6 composites*. Composites Part B: Engineering, 187. (2020) 107839.
https://doi.org/10.1016/j.compositesb.2020.107839
[7] Sun X., Lasecki J., Zeng D., Gan Y., Su X., Tao J.: *Measurement and quantitative analysis of fiber orientation distribution in long fiber reinforced part by injection molding*. Polymer Testing, 42. (2015) 168–174.
https://doi.org/10.1016/j.compositesb.2020.107839
[8] Valvedere M. A., Kupfer R., Wollmann T., Kawashita L. F., Gude M., Hallett S. R.: *Influence of component design on features and properties in thermoplastic overmoulded composites.* Composites Part A: Applied Science and Manufacturing, 132. (2020) 105823.
https://doi.org/10.1016/j.compositesa.2020.105823
[9] Schneider T.: *Lightweight construction: First composite gearbox housing with layer-optimized organo sheeting weighs 30 % less than a comparable aluminum component*. Reinforced Plastics, 63/1. (2019) 40–45.
https://doi.org/10.1016/j.repl.2017.11.018
[10] Karakaya N., Papila M., Özgoc G: *Overmolded hybrid composites of polyamide-6 on continuous carbon and glass fiber/epoxy composites: An assessment of the interface.* Composites Part A: Applied Science and Manufacturing, 131. (2020) 105771.
https://doi.org/10.1016/j.compositesa.2020.105771
[11] Fu L., Zhang M., Zhai Z., Jiang F.: The influence of preheating temperature on the mechanical properties of injection-overmolded hybrid glass fiber reinforced thermoplastic composites. Polymer Testing, 105. (2022) 107425.
https://doi.org/10.1016/j.polymertesting.2021.107425
[12] Andrzejewski J., Przyszczypkowski P., Szostak M.: *Development and characterization of poly(ethylene terephthalate) based injection molded self-reinforced composites. Direct reinforcement by overmolding the composite inserts.* Materials&Design, 153. (2018) 273–286.
https://doi.org/10.1016/j.matdes.2018.04.084
[13] Boros R., Rajamani P. K., Kovács J. G.: *Combination of 3D printing and injection molding: Overmolding and overprinting*. eXPRESS Polymer Letters, 13/10. (2019) 889–897.
https://doi.org/10.3144/expresspolymlett.2019.77
[14] www.markforged.com (elérés: 2022. 03. 10.).
[15] Dickson A. N., Barry J. N., McDonnell K. A., Dowling D. P.: *Fabrication of continuous carbon, glass and Kevlar fibre reinforced polymer composites using additive manufacturing*. Additive Manufacturing, 16. (2017) 146–152.
https://doi.org/10.1016/j.addma.2017.06.004

[16] https://grupaazoty.com/en/group-s-offer/plastics/alphalon-27 (elérés: 2022. 03. 10.).
[17] MSZ EN ISO 178: Műanyagok. A hajlítási tulajdonságok meghatározása, 2010.