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Reviewed Paper

Static Mixing Nozzles for Long and Short Fiber Additive Extrusion Processes

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Abstract

Additive manufacturing is conventionally used to create structures by extruding plastic or metal layer by layer. In the case of polymer processes, fibers are typically added to increase stiffness and reduce warping during building. The length of the fiber exiting the nozzle can impact the overall mechanical properties of the structure. Using long fiber pellets can increase the starting length of the pellets to help increase the average fiber length coming out from the extruder. However, extruded long fiber materials tend to have low fiber alignment and high porosity leading to poor mechanical properties. By blending long fiber and short fiber resins using a static mixing nozzle, consolidated beads can be created to produce more stable and solid structures while adding a fixed amount of long fiber into the extruded bead to increase mechanical performance.

Introduction

Additive manufacturing (AM) for thermoplastics comprises the conversion of a polymer feedstock (filament, powder, thermoset, etc.) to a structure in space, created layer by layer until a near-net shape component is complete [1]. During this process, toolpaths are created in specific widths per pass (bead width) and height (layer height). Machines on the market for this space range from a desktop scale of around 0.3 m × 0.3 m × 0.3 m (X, Y, and Z directions) up to large scale, such as Big Area Additive Manufacturing (BAAM, Cincinnati, Inc.) with a print scale of 6 m × 2.4 m × 1.8 m (X, Y, and Z directions) [2]. While small-scale systems are often used for rapid prototyping, fine resolution printing, and new material trials, larger systems are used to create custom tooling components, complex core designs, and traditional rapid prototyping [3]. During large-scale printing, shrinkages occur from the heating and cooling of layers at different rates and between beads, which can lead to cracking and warping [3]. To add stiffness to the structure, fillers such as carbon, glass, and wood fibers are added to help prevent these distortions [4]. While adding these fillers increases the stiffness and helps mitigate the fundamental issues leading to warping, they also introduce anisotropy to the structure. Fibers tend to align along the print direction due to the shear introduced during extrusion. The fiber alignment can help in high stiffness and strength along the print direction but can cause significantly lower stiffness and strength along the layer-layer direction (Y and Z-Direction). Differences in fiber alignment can create anisotropy in the composite's mechanical properties

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and affect its thermal expansion coefficient (CTE). CTE anisotropy becomes a severe issue during larger prints. In addition, the extrusion process creates defects within the bead, such as micro-porosity. These porosities decrease the mechanical properties of the components. The surface porosities are also undesirable in mold/die applications, where high vacuum surface integrity is required [5,6].

One way to counter the problem of porosity created inherently during AM process is to compression mold the part after printing either by post-heating a printed preform or applying compression rapidly just after printing while the structure is still hot enough to deform [7,8]. This process is known as Additive Manufacturing Compression Molding or AM-CM. AM-CM is being developed for creating preforms with the AM process and then compression molding them (CM) to remove porosities and other defects to produce a cohesive end-use product [9]. A significant advantage to using AM-CM is that fibers can be aligned along a particular direction before compression molding using an automated system, reinforcing specific directions and locations of a preform by maintaining fiber alignment after compression molding is complete [10]. While fiber alignment can be attained with compression molding by doing layups, the angles and curvatures that can be achieved are limited. Using a standard AM system, any curvature desired can be obtained with tailored fiber alignment using the discontinuous fiber-filled polymers. While most standard materials used on AM are short-chopped fibers and have relatively consolidated extrudates, long fiber (LF) pellets can also be extruded to increase the average fiber length of the composite [11]. Increased fiber length in composites can increase the part stiffness and strength when the fibers are aligned during deposition. However, printing with LF materials often introduces challenges for AM, such as severe porosity, fiber breakage, and metering uniformity. Severe porosity and metering issues create problems for mechanical performance and deposition and make it difficult to produce large structures spanning multiple layers. Despite these issues, preforms only require 1-2 layers of printing, and the porosity issue and variations in extrusion can be removed/altered during CM to create a composite that starts with high amounts of defects and results in a final structure that is void-free, fiber aligned, and uniform. In the present work, short and long fibers are used to prepare composites using the AM-CM process, and their mechanical properties are discussed in correlation to the fiber orientation and porosity values. In addition, the challenges in printing long fiber through extrusion were resolved by utilizing custom-made mixing nozzles. The mixing nozzle can be used to increase dispersion of material blends within the print bead as well as reduce the porosity of the printed structure.

1. Experiment Design and Materials

Samples were created on the Big Area Additive Manufacturing (BAAM) system using a standard 0.4-inch nozzle and a mixing 0.4-inch nozzle. Two materials of interest were used, long glass fiber thermoplastic polyurethane (LGF TPU, colored black) and short glass fiber acrylonitrile butadiene (SGF ABS, colored orange) from Techmer PM. Both materials contained 40% glass fiber by weight and were mixed using the pellet mixer for BAAM and processed through the BAAM extruder.

The LGF TPU started as 12 mm pellets, while the SGF ABS is a 4 mm short-chopped fiber pellet. The 12 mm LGF pellets were cut into approximately 6 mm by hand for two samples to observe any potential differences between initial fiber lengths. Five composite formulations were studied with varying ratios of ABS and TPU and LGF pellet sizes: 100% ABS (SGF), 95% ABS/5% TPU (12mm), 95% ABS/5% TPU (6mm), 90% ABS/10% TPU (12mm), and 80% ABS/20% TPU (12mm). Two different nozzle geometries were used during the prints, a standard 0.4 in nozzle diameter and the second being a custom static mixing nozzle with a 0.4 in nozzle diameter. Each formulation with printed with a standard nozzle, and the mixing nozzle was additionally used to print the 95/5 ABS/TPU (12mm) and 90/10 ABS/TPU (12mm) materials. Prints completed with both nozzles had a layer height of 3.81 mm (0.15 in), and a bead width of 12.7 mm

(0.5 in). Single bead (1 perimeter) boxes were manufactured with a joining wall between the two boxes with a double bead width to minimize start and stops between layers with a layer time of 1.5 min.

To create compression molded samples, charges were cut from the printed boxes, as shown in Figure 1. Charges cut measured to approximately 215-230 g to allow for extra material to ensure the plaques are solid for sample collection. The final compression molded plaque was found to be approximately 200 g. Charges cut from the as printed walls were placed in a square aluminum mold. The mold contained a 152 x 152 mm square cutout with a thickness of 5mm (final compressed plaque thickness), such that the print direction was aligned in one direction of the plaque with the Z-direction the perpendicular position [Figure 3]. The charge is heated and applied with pressure to form the material into a solid thinner plaque to obtain tensile samples.

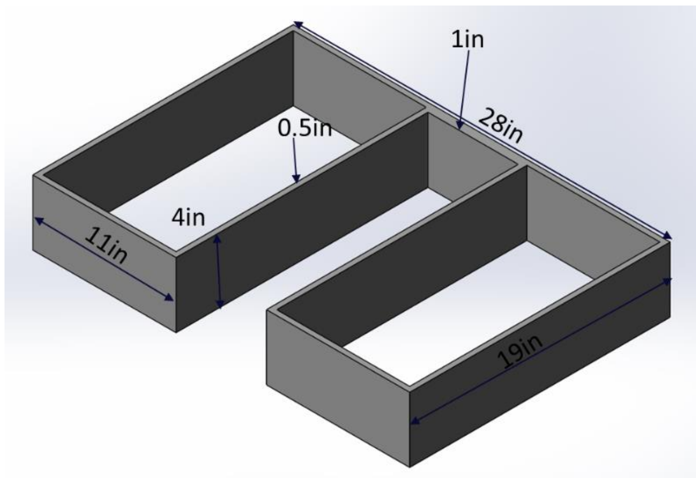

Figure 1: Print Geometry on BAAM

Figure 2. Compression molding of 3D printed preforms was completed using heated hydraulic press.

The platens of the compression system were heated to 415 F and allowed to soak for 2 min. The mold and charge were placed and then compressed such that each end of the mold was in contact with the platens, with slight pressure applied to allow for heat transfer into the mold and charge. After a soak time of 5 min, the material was compressed at a pressure of 1-2 tons for a time of 5 min. The pressure was then increased to 8 tons for an additional 5 min. The platens were then cooled, and the pressure and mold were released once the overall temperature of the system reached 160 F. The compression-molded plaque was the removed for water jet cutting [Figure 3].

Figure 3: Compression molded GF ABS sample

3. Results and Discussion

As more long fibers are introduced into the extruder, the porosity in extrudates increases significantly, and it becomes more challenging to manufacture 3D printed structures. Long fiber pellets tend to capture more air during feeding, creating voids that expand once the extrudate exits through the nozzle. CT scans of these samples indicate the presence of fiber pockets consisting of highly misaligned fibers that swirl around the bead [Figure 4].

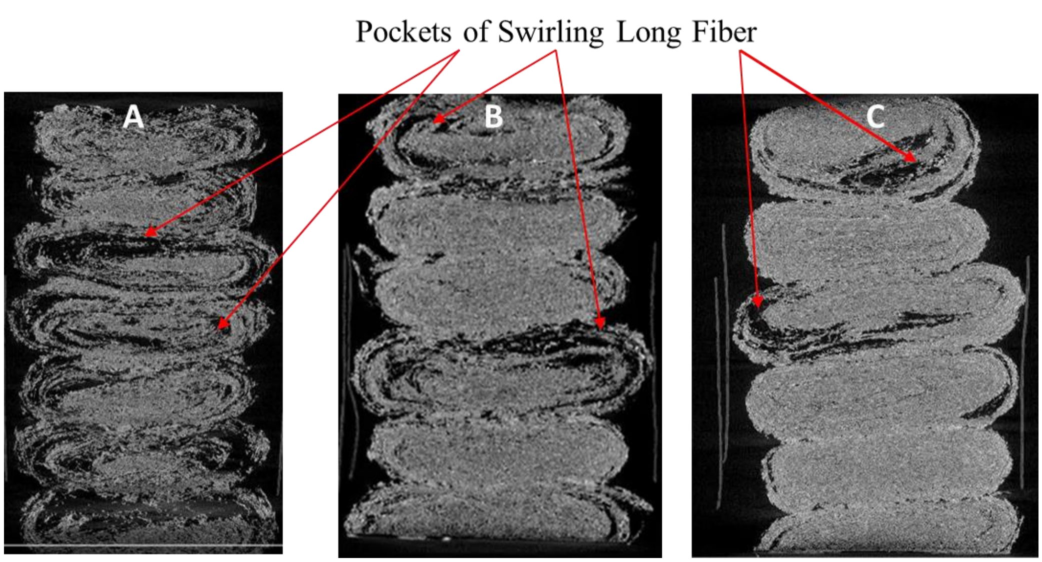

Figure 4: CT images of cross section of as printed 12mm LGF blends using standard 0.4 in nozzle A) 80% GF ABS-20% LGF TPU, B) 90% GF ABS-10% LGF TPU, C) 95% GF ABS-5% LGF TPU

Another potential reason is that the materials (fibers and matrix) were not mixing thoroughly within the extruder, resulting in resin- and fiber-rich regions. Therefore, a static mixing nozzle was introduced to aid material mixing throughout the melt stream. Samples created using the mixing nozzle had a more uniform color, visually indicating that the fibers mixed throughout the polymer more successfully. In addition, the static mixing nozzle was capable of not only mixing the materials but also helped reduce large pores in the beads creating more uniform and solid cross sections within the structure [Figure 5].

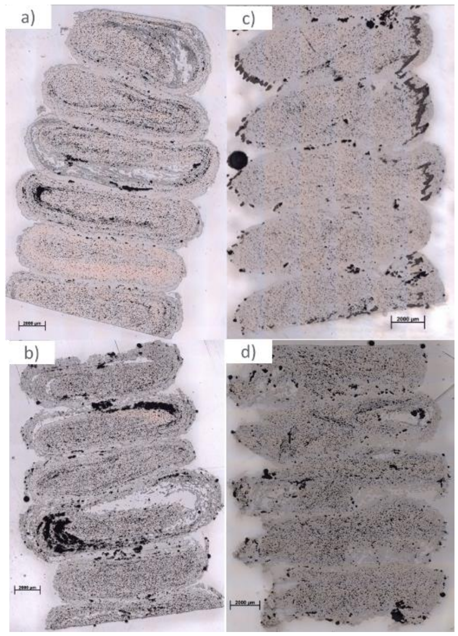

Figure 5: Cross section comparison between mixing nozzle and standard nozzle. a) 95% ABS 5% TPU 12mm fiber standard nozzle. b) 90% ABS 10% TPU 12mm fiber standard nozzle. c) 95% ABS 5% TPU 12mm mixing nozzle. d) 90% ABS 10% TPU 12mm mixing nozzle

ImageJ was utilized on a single bead of material from each of the images in Figure 5 to extract the average porosity as well as pore size [Figure 6]. A smaller cross section was selected such that only a single printed bead was extracted for the image. A smaller cross section was used to minimize the number of background pixels which would decrease the porosity reading. Cross section locations were selected for areas with visually large defects in attempt to capture worst case porosity values.

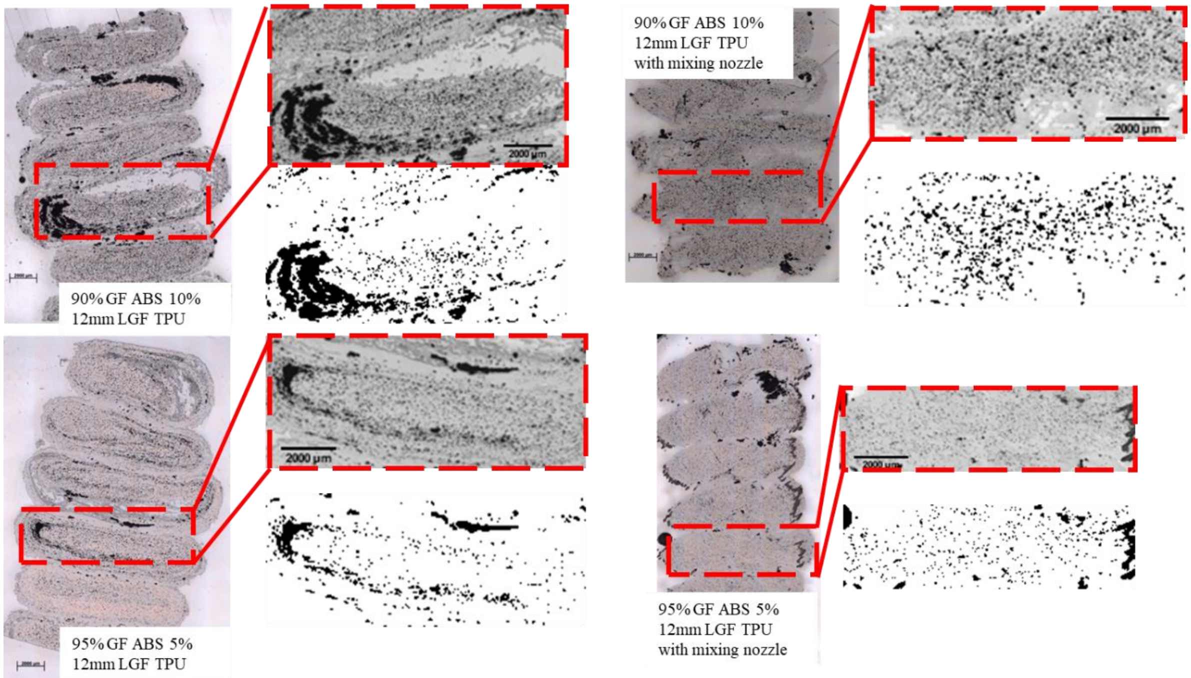

Figure 6: Image J cross sections of mixing and non-mixing fiber blends. Image shows image section selection and porosity observed by Image J

When a mixing nozzle is not used, porosity content is observed as 7.2% and 14.3% with an average pore size of 154um and 164um (5% LGF TPU and 10% LGF TPU) respectively. When a mixing nozzle is introduced, porosity of 5% LGF TPU increases to 8.5% with an average pore size of 112 um and the 10% LGF TPU decreases to 9.5% with an average pore size of 138um. Addition of a mixing nozzle effectively decreased the average pore size for both samples as well as decreased the overall porosity in the sample as the long fiber content increases. While the porosity increases slightly when low long fiber contents are used, a visually more consistent bead with less visibly large voids is still obtained. Higher bead uniformity allows for more consistent printing patterns which is critical for additive manufacturing.

Another approach to reducing inhomogeneity in printed LGF structures was to reduce the overall pellet length of the LGF TPU by cutting the 12 mm pellets down to a 6 mm pellet length. This change created samples with more consistent feed and more homogeneous-looking beads [Figure 7]. Image J was used to calculate porosity content and average pore size. For the 6mm LGF TPU sample, a pore content of 6.7% average pore size of 98um was found. When shorter fibers are used, both lower porosity contents and as well as a smaller average pore size are observed indicating that shorter fiber lengths used can effectively increase extrusion quality as well as the mixing nozzle.

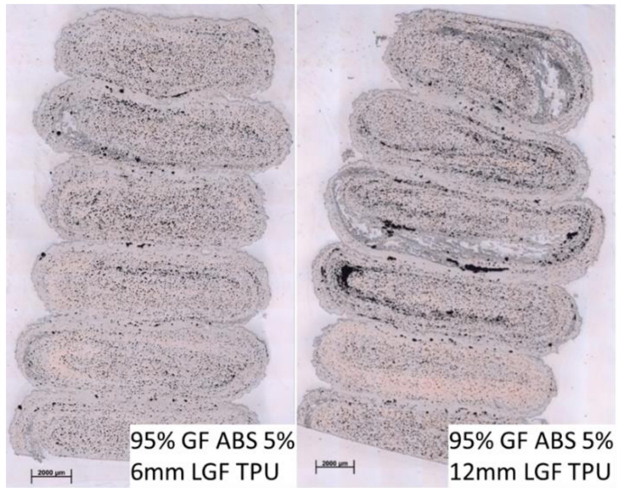

Figure 7: CT of differences between 6mm and 12mm LGF 95/5 ABS/TPU print cross sections

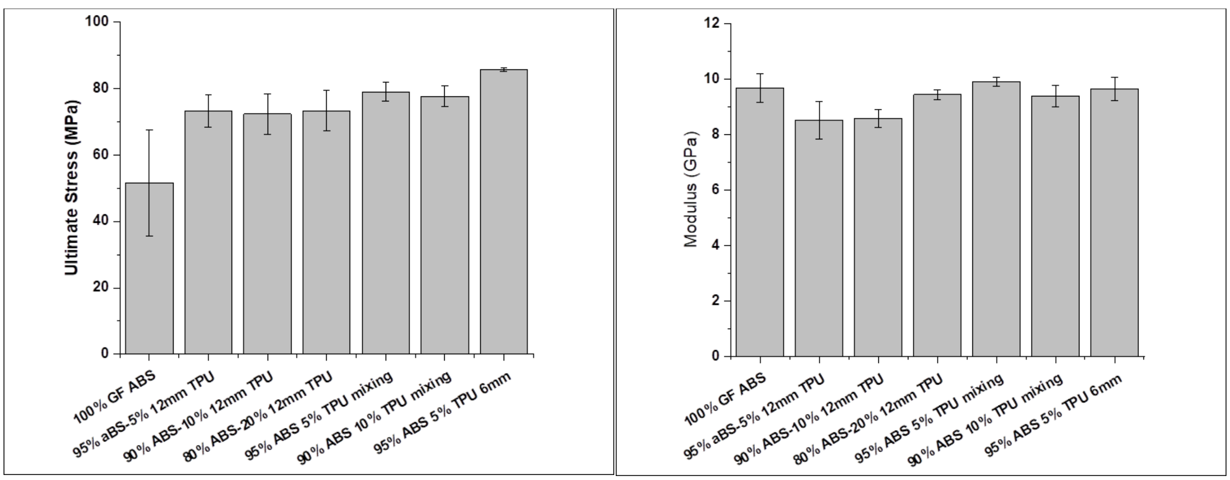

Figure 8: Stress and Modulus of LGF TPU and SGF ABS samples

The tensile testing results are given in Figure 8 to compare the impact of long fiber quantity, pellet length at the start, and nozzle type (mixing vs. standard) on the composite's strength and stiffness properties. Materials with LGF displayed a slight increase in fracture stress along with a drop in the overall modulus using the standard nozzle. With increasing LGF TPU content, the fracture stress and modulus remained nearly constant. A non-significant increase in modulus could be caused by fiber breakage in the screw or a lack of aligned fibers to allow for load transfer. As the quantity of long fiber increased, the porosity in the as-printed sample increased as well. During compression molding, porosity is removed from the material, but some air pockets remain trapped and become smaller pores. If the number of pores in the compression-molded samples increases as a function of long fiber quantity, it could drop mechanical performance. Likewise, if the long fiber added to the system undergoes significant breakage during extrusion due to shear and other harsh processing conditions, the long fiber's benefits can be significantly reduced. In addition, the alignment of the long fibers is key to maximizing their reinforcing potential since the as-printed samples contained large pockets of poorly aligned long fiber, which were more numerous as the long fiber content increased.

Mixing the material with a static mixing nozzle improved the stiffness and strength marginally while reducing the overall variance within the data. The static mixing nozzle was designed to mix two resins during extrusion. As such, material output from the system should be more uniform, removing islands of long fiber and making the overall structure more consistent. While all the 12 mm length pellets are similar in strength and modulus, the 6 mm length TPU pellets were found to increase the strength of the composite significantly. Smaller fibers are easier to process during extrusion, creating more consolidated beads when compared to 12mm fiber pellets extruded. While the overall porosity is only ~0.5% lower than the 12mm fiber version, the average pore size decreased from 154 um to 98 um. Fracture images of the samples are taken to observe differences in failure mode and find defects or poor mixing of material [Figure 9].

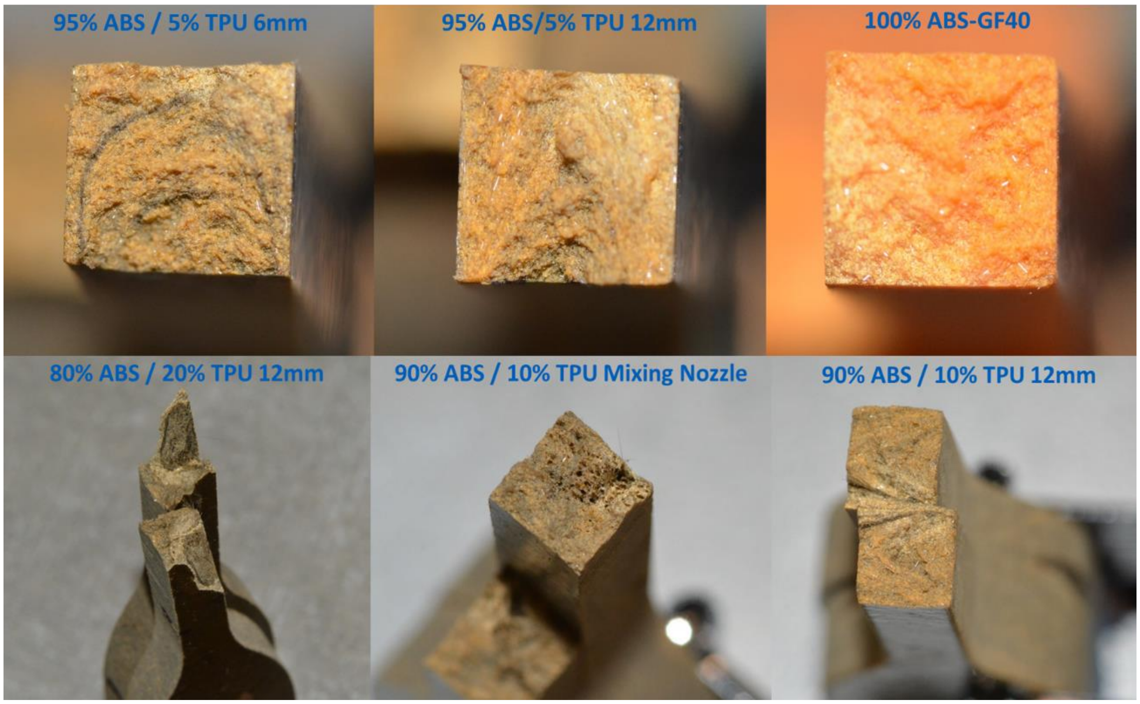

Figure 9: Fracture surfaces of tested samples

Fracture surface images of the broken samples indicate that porosity in the compression-molded samples increased with the mixing nozzle but had a more uniform texture and fewer gradients of material islands than the standard nozzle prints. In addition, the fracture surface became rougher with increasing long fiber content, especially in the case of the 20% LGF TPU sample in which the break travels down the gauge length of the sample. The mechanical data and fracture surfaces indicate that the LF being introduced into the matrix affects the overall fracture response of the structure, improving the composite quality. While the average pore size and pore density were decreased using a mixing nozzle, it was found that the design of the mixing nozzle could be further optimized. The design of the mixing nozzle was modified to add a compression zone at the end of the nozzle to further squeeze the material down from a 12.7mm output diameter down to a 5mm output to better compress the material to further increase bead quality [Figure 10].

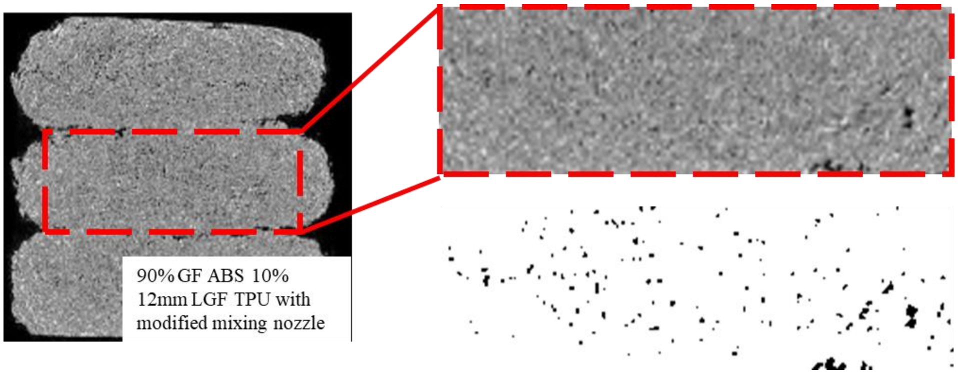

Figure 10: Modified mixing nozzle design with 10% LGF TPU porosity sample and section view

When the new nozzle design is used, samples that would normally have ~14.3% (no mixing) or 9.5% (mixing nozzle original design), are now decreased down to ~2.2% porosity. In addition, the average pore size was effectively decreased to 3um. The significantly decreased porosity and pore size can potentially enable the use of long fiber and short fiber blends for large scale polymer additive manufacturing.

4. Conclusion

Additive manufacturing is commonly used in industry to create molds, complex cores, and small-scale high-resolution geometries and prototypes. Fibers are added into large-scale polymer AM materials to increase stiffness and strength, preventing warping and cracking. The fibers extruded tend to align within the bead due to shear profiles imposed, creating highly anisotropic composites. Using AM, preforms can be created for subsequent compression molding to create unique products with highly aligned fibers that are challenging to create using traditional means. Fiber length has a critical influence on the overall composite performance, with longer aligned fibers outperforming standard short, chopped fiber composites. Printing structures with long-fiber pellets is difficult due to large porosities introduced, difficulties in feeding/pumping, lack of fiber alignment, and large amounts of fiber breakage. Combining long-fiber pellets with short/chopped fiber pellets can improve the processability of the material, allowing for larger structures such as preforms to be created. AM-CM can create higher performance composites by using a blend of long and short fiber pellets for preforms. As longer fibers are introduced to the system, bead quality decreases which may be caused by several feeding factors, fiber breakage mechanics, fiber clumps that form, etc. To increase the bead consistency and reduce fiber rich voids, a static mixing nozzle can be used. The mixing elements help promote not only mixing of the two resins but also aids in breaking up large voids. Further work can be done to increase the benefits of the mixing nozzle to enable this technology for standard additive manufacturing using these materials as well as the AM-CM process.

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6. References

[1] M. Vaezi, S. Chianrabutra, B. Mellor, S. Yang, Multiple material additive manufacturing - Part 1: A review, Virtual Phys. Prototyp. 8 (2013) 19–50. doi:10.1080/17452759.2013.778175.

[2] A. Roschli, K.T. Gaul, A.M. Boulger, B.K. Post, P.C. Chesser, L.J. Love, F. Blue, M. Borish, Designing for Big Area Additive Manufacturing, Addit. Manuf. 25 (2019) 275–285. doi:10.1016/j.addma.2018.11.006.

[3] F. Adrian Rodriguez Lorenzana, Thermal characterization of abs/carbon fiber, abs/ glass fiber and petg/glass fiber reinforced composites used in large area additive manufacturing, The University of Texas at El Paso, 2019.

[4] L.J. Love, V. Kunc, O. Rios, C.E. Duty, A.M. Elliott, B.K. Post, R.J. Smith, C.A. Blue, The importance of carbon fiber to polymer additive manufacturing, J. Mater. Res. 29 (2014) 1893–1898. doi:10.1557/jmr.2014.212.

[5] P. Yeole, S. Kim, A.A. Hassen, V. Kumar, V. Kunc, U. Vaidya, Large-scale additive manufacturing tooling for extrusion-compression molds, Addit. Manuf. Lett. 1 (2021) 100007. doi:10.1016/J.ADDLET.2021.100007.

[6] K.M.M. Billah, J. Heineman, P. Mhatre, A. Roschli, B. Post, V. Kumar, S. Kim, G. Haye, J. Jackson, Z. Skelton, V. Kunc, A.A. Hassen, Large-scale additive manufacturing of self-heating molds, Addit. Manuf. 47 (2021) 102282. doi:10.1016/J.ADDMA.2021.102282.

[7] V. Kumar, S. Kim, V. Kishore, K. V Mungale, A. Nowlin, U. Vaidya, C. Blue, V. Kunc, A.A. Hassen, Hybrid manufacturing technique using large-scale additive manufacturing and compression molding for high performance composites, n.d. http://energy.gov/downloads/doe-public-access-plan.

[8] V. Kumar, S.P. Alwekar, V. Kunc, E. Cakmak, V. Kishore, T. Smith, J. Lindahl, U. Vaidya, C. Blue, M. Theodore, S. Kim, A.A. Hassen, High-performance molded composites using additively manufactured preforms with controlled fiber and pore morphology, Addit. Manuf. 37 (2021) 101733. doi:10.1016/j.addma.2020.101733.

[9] P.A. Kumar Jain, S. Sattar, D. Mulqueen, D. Pedrazzoli, S.G. Kravchenko, O.G. Kravchenko, Role of annealing and isostatic compaction on mechanical properties of 3D printed short glass fiber nylon composites, Addit. Manuf. 51 (2022) 102599. doi:10.1016/J.ADDMA.2022.102599.

[10] H.L. Tekinalp, V. Kunc, G.M. Velez-Garcia, C.E. Duty, L.J. Love, A.K. Naskar, C.A. Blue, S. Ozcan, Highly oriented carbon fiber-polymer composites via additive manufacturing, Compos. Sci. Technol. 105 (2014) 144–150. doi:10.1016/j.compscitech.2014.10.009.

[11] S.P. Alwekar, S.S. Kore, U. Vaidya, Comparison of long fiber thermoplastics produced via directional extrusion versus extrusion compression molding, in: CAMX - Compos. Adv. Mater. Expo, Dallas, TX, 2018.