ADVANCING CONTINUOUS FIBER FUSED FILAMENT FABRICATION FOR HIGH PERFORMANCE APPLICATIONS

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ABSTRACT

Continuous fiber fused filament fabrication (CF-FFF) is a promising technology for manufacturing complex, ultralightweight composite components. Despite its potential, there is currently a lack of commercially available preimpregnated thermoplastic feedstock selection, especially in terms of high fiber volume contents (i.e., >50%) and advanced engineering polymer matrix systems (e.g., PEEK, PEI, PPS). The ability to manufacture composite parts with these material properties via the CF-FFF process would potentially be of value for high performance applications such as aerospace. This study demonstrates the use of an adjustable multi-die pultrusion system for converting a high fiber volume content (54%) polyetherimide-carbon fiber commingled yarn into a preimpregnated CF-FFF feedstock material. In an attempt to increase manufacturing throughput, higher than typical processing speeds (200-800 mm/min) and processing temperatures (400-490 °C) are tested. A full factorial experiment is performed in which preheater temperature, pultrusion die temperature, and pultrusion speed are varied in order to determine optimal processing conditions for fiber impregnation. Unfortunately, due to large, concentrated bundles of reinforcing fibers in the commingled yarn, impregnation distances were large and prevented full consolidation from being achieved.

1 INTRODUCTION

Fused filament fabrication (FFF) is a simple, robust, and highly cost-efficient method of additive manufacturing (AM) which has led to it becoming the most widespread 3D printing technology in present times. This manufacturing process involves the extrusion of a melted thermoplastic polymer feedstock through a heated nozzle, which is deposited layer-by-layer to form an output geometry. This technology has proven to be exceptionally useful for fabricating prototypes and non-structural end-use parts; however, thermoplastics have limited mechanical properties which prevents use in applications requiring high strength and stiffness characteristics. Due to its method of depositing material in continuous paths, FFF is well suited for the integration of continuous fiber reinforcements which can greatly increase mechanical performance. The past decade has seen rapid development of continuous fiber fused filament fabrication (CF-FFF) technology with the release of several commercial manufacturing systems and a multitude of scientific research studies. During this time, two separate approaches for supplying the fiber and thermoplastic feedstock components to the CF-FFF process have become predominant: 1) Directly extruding preimpregnated material [1]. 2) Impregnating dry fibers with melted thermoplastic within the nozzle, termed in-nozzle impregnation [2], [3].

The use of preimpregnated thermoplastic feedstocks is the simpler of the two methods as it avoids altogether the difficult challenges associated with impregnating tightly packed fiber bundles with highly viscous molten thermoplastic during the printing process. In-nozzle impregnation necessitates the use of high pressures and temperatures, along with slow processing rates in order to achieve reasonable levels of fiber impregnation [4], [5].

In a thorough search of in-nozzle impregnation scientific literature, the authors were unable to find an example of complete fiber wet-out being achieved with this process.

There are three main mechanisms involved in the consolidation of thermoplastic matrix composites: intimate contact development, autohesion, and fiber impregnation. Phillips et al. [6] demonstrated that for a PEI matrix carbon fiber composite, the processes of intimate contact formation and autohesion can take on the order of 1% of the overall time needed for fiber impregnation. As such, for effective CF-FFF processing, it should be highly beneficial for the feedstock material to already be completely preimpregnated to significantly reduce the processing time necessary for manufacturing a well consolidated composite.

Currently there is very little selection of commercially available preimpregnated feedstock materials for the CF-FFF process. By far the most accessible and widely tested preimpregnated feedstock materials for CF-FFF in research are those manufactured by the company Markforged and sold for use with their continuous fiber 3D printing systems. Their current offerings include carbon, glass, and Kevlar reinforcements [7] in what is likely a polyamide matrix system [8]. These feedstocks have fiber volume contents on the order of 35-40% [8]. In order to advance the state of CF-FFF for use in high performance applications such as aerospace, it is highly desirable to begin testing the use of materials with high fiber volume contents (i.e., >50%) and advanced engineering thermoplastic matrix systems, e.g., polyether ether ketone (PEEK), polyetherimide (PEI), or polyphenylene sulfide (PPS). As commercially available preimpregnated CF-FFF feedstocks are not currently available with these properties, methods for producing them are of interest. One method of producing such feedstocks is through the pultrusion of commingled yarns.

The consolidation process for commingled yarns has been modeled by Bernet et al. [9], in which consolidation time at a given pressure and temperature is determined to be predominantly driven by the size and permeability of dry fiber bundles through which the thermoplastic matrix must flow, and can be predicted using Darcy's law. Higher temperatures are shown to reduce consolidation time due to the reduction in viscosity of the matrix material.

In a paper by Eichenhofer et al. [10], the use of a multi-die pultrusion system was shown to significantly reduce the degree of pultrudate deconsolidation over single die setups, by cyclically reforming the composite and thereby reducing the residual stresses. This study did not make use of a cooling die and was able to produce a high quality pultrudate from a 52% carbon fiber-polyamide 12 commingled yarns at rates of up to 315 mm/min. The reduction in deconsolidation achieved with multi-die pultrusion systems may prove to be beneficial for the CF-FFF process, as deconsolidation of the material during the deposition process could result in the need for slower processing speeds in order to reconsolidate the material. Furthermore, if significant deconsolidation were to occur in the hot end of the deposition tool, it could lead to jamming issues.

In a study by Ghaedsharaf et al. [11], braided 52-53% carbon fiber-PEI commingled yarns (manufactured by Concordia Manufacturing LLC) were pultruded in a multi-die pultrusion system. The material was preheated to a temperature of 300 °C and pulled through four dies set to 400 °C at rates of 50 and 150 mm/min. The pultrudate manufactured at 50 mm/min was reported to have a lower void content than the one produced at 150 mm/min.

The study presented herein seeks to determine if higher than typical pultrusion speeds (200-800 mm/min) can result in fully impregnated pultrudates for CF-FFF feedstock materials when using processing temperatures near or exceeding the thermal degradation point of the matrix material. By doing so, the viscosity of the thermoplastic matrix should be reduced, potentially improving the rate of consolidation. Higher speeds would result in larger manufacturing throughput, making this process more appealing for commercial usage. A 54% carbon fiber-PEI commingled yarn is processed using a custom developed adjustable multi-die pultrusion system. The effects of the temperature of a contactless preheater are also examined.

2 METHODOLOGY

2.1 Material

For this study, a 54% fiber volume fraction commingled yarn manufactured by Concordia Manufacturing LLC was used. This yarn consists of a 12K tow of Hexcel HexTow IM2A intermediate modulus carbon fiber commingled with SABIC ULTEM 9011 polyetherimide (PEI) fibers. A microscope cross section of this yarn is shown in Figure 1. The yarn dimensions exhibit a high aspect ratio, with a width to thickness ratio on the order of 20:1 (the complete yarn cross section is not shown in the figure). The yarn is supplied in this form, perhaps as the result of a fiber spreading operation used during processing. As can be seen, the commingling process has resulted in a reasonably good distribution of carbon and PEI fibers; however, there are still distinct regions showing large concentrations of each material. This poses a challenge as long impregnation distances though tightly packed fiber bundles will be encountered. The nominal total cross-sectional area of PEI and carbon fibers for this yarn is 0.46 mm^2. The glass transition temperature of the PEI matrix was determined to be 217 °C through the use of a Mettler Toledo DSC 3 differential scanning calorimeter.

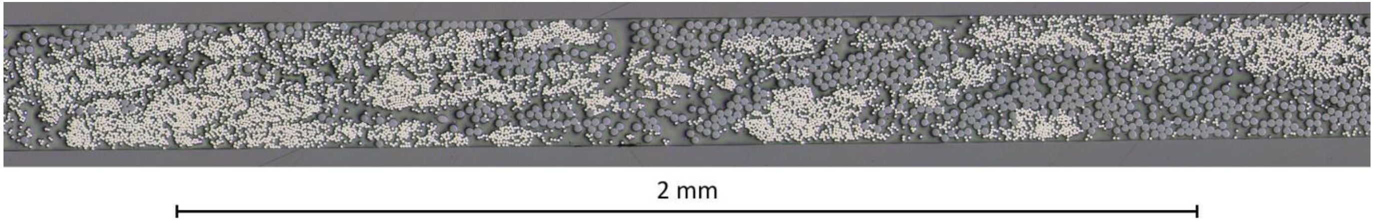

Figure 1. Microscope cross section of the commingled yarn showing carbon (white) and PEI (grey) fibers.

2.2 Pultrusion System

A multi-die pultrusion system has been constructed to convert commingled yarns into consolidated CF-FFF feedstocks (see Figure 2). The commingled yarn is first fed through a friction-based tensioning system made up of staggered polyethylene spacers. The friction against the spacers increases the force needed to pull the yarn into the dies, thus placing the fibers under tension and pulling them together into a tight bundle. This has proven to be especially important as without tensioning, the yarn will swell up in the preheater leading to a jam at the entrance of the first die.

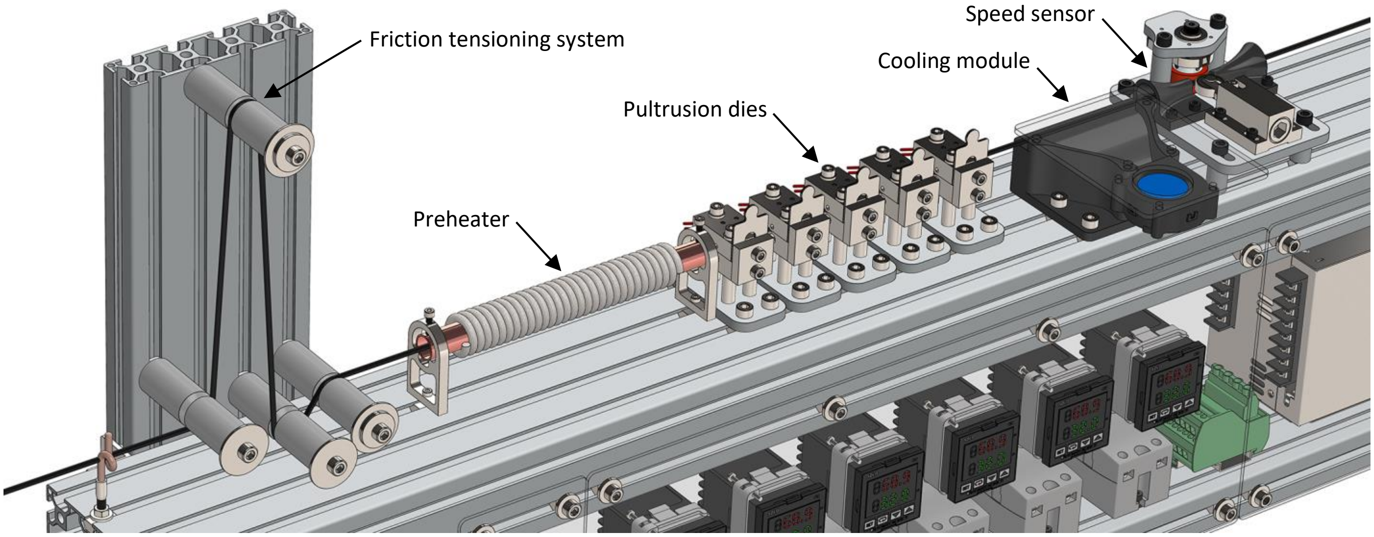

Figure 2. Components of the pultrusion system.

A contactless preheater was added to the system to increase the temperature of the polymer fibers before they enter the first pultrusion die. This both lowers the polymer viscosity and aids in ensuring that polymer fibers enter the narrow die opening by melting them onto the reinforcing fibers. The preheater was constructed from a 200 mm long, 14 mm inner diameter copper tube with a 400 W pipe heater coiled around it. A type K thermocouple is used to monitor the surface temperature of the copper tube and an off-the-shelf proportional–integral–derivative (PID) temperature controller regulates the system. The preheater was wrapped with ~20 mm thick stone wool insulation to minimize thermal losses (not shown in figure).

This pultrusion system has been designed to produce CF-FFF feedstocks with varying types of reinforcing fibers and polymer matrices, as well as varying fiber volume fractions. As such, input yarns may exhibit a range of cross-sectional areas and thereby require different pultrusion die outlet areas. The developed pultrusion dies allow the outlet width to be varied while maintaining a constant outlet height through the use of a two-part design (see Figure 3). As commingled yarns are typically made-to-order with the ability to control the approximate quantity of thermoplastic and reinforcing fibers used, similar cross-sectional areas can be achieved for different material systems. The pultrusion die design gives enough adjustment freedom to make up for the small differences in areas while still producing a pultrudate with an approximately square cross section. The dimensions of the channel formed by the two die halves is shown on the right in Figure 3. Die opening widths can be varied with the use of shims with a minimum thickness increment of 0.001 inch (0.025 mm). The dies were machined from H13 tool steel and subsequently heat treated to achieve an average Rockwell C hardness of 49 so as to minimize abrasive wear. Two 70 W heater cartridges are used per die set along with one type K thermocouple. Temperature of the dies is controlled through the use of an off-the-shelf PID temperature control. It should be noted that no cooling die is used in this system as the pultrudate will undergo remelting during the subsequent CF-FFF process.

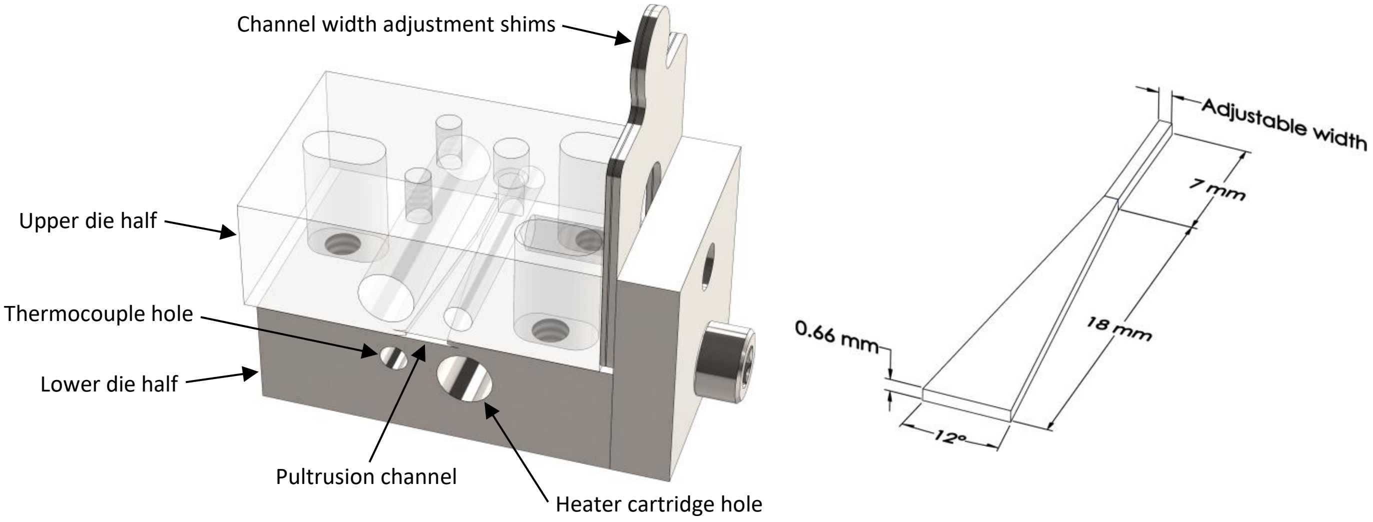

Figure 3. Left: Components of the pultrusion die. Right: Dimensions of the channel formed by the pultrusion die halves.

After exiting the last pultrusion die, the material enters a simple cooling module in which a fan blows ambient temperature air across the filament over a distance of 80 mm. Next, the filament passes through a line speed sensor which tracks the rate of pultrusion via the rotation of a urethane roller mounted to an encoder.

A spooling system serves to both apply the pulling force as well as to collect the processed material. The spooling system consists of a 222 mm diameter spool mounted on a large gear which is pinion driven by a DC brushed gear motor. The pultrudate is taped to the spool to begin the pulling process. A graphic user interface has been developed, running on a Raspberry Pi, in which the pulling speed and the length of material to be processed can be specified. A simple PID program references the signal from the line speed sensor to control the rotational speed of the motor.

2.3 Processing Parameters

For this study, a full factorial experiment was performed in which preheater temperature, die temperature, and pulling speed were varied as shown in Table 1. The combination of all three processing parameter variations led to a total of 80 variants. A TGA analysis of SABIC ULTEM 9085 performed by Padovano et al. [12] showed a degradation temperature of 447.5 °C when analyzed in air, which is likely similar for the ULTEM 9011 used in this study.

Table 1. Range of values chosen for the full factorial processing parameter study.

Parameter	Values
Preheater temperature (°C)	300, 350, 400, 450
Die temperature (°C)	400, 425, 450, 475, 490
Pulling speed (mm/min)	200, 400, 600, 800

Preheater temperature was varied in increments of 50 °C in the range of 300-450 °C. This range was chosen as it starts at a fairly low processing temperature for PEI and ranges to around the thermal degradation temperature and near the upper limit of the preheater's capabilities. As this is a contactless system, the material will not be able to fully reach these temperatures due to thermal losses and the limited time spent in the preheater.

Die temperatures started at 400 °C, which was a typical temperature used in literature, and increased in increments of 25 °C up to 475 °C. The initial intention was to also test at 500 °C; however, the pultrusion dies were not able to maintain this temperature consistently which led to the use of a reduced value of 490 °C. The higher temperatures used here are likely greater than the degradation temperature of the PEI matrix; however, it is possible that the limited time spent in the pultrusion dies will prevent degradation reactions from occurring to a significant degree.

Pulling speeds were varied by increments of 200 mm/min in the range of 200-800 mm/min. The maximum speed was chosen as it was near the limit of the spooling system's capabilities.

The pultrusion die outlet areas were linearly varied from 0.50-0.44 mm^2 (see Table 2) which should result in a filament of square cross section.

Table 2. Pultrusion die outlet widths and areas used for this study.

Die	Outlet Width (mm)	Outlet Area (mm^2)
1	0.76	0.50
2	0.74	0.49
3	0.71	0.47
4	0.69	0.45
5	0.66	0.44

For each variant, two meters of material was processed with the first meter discarded as it had spent time idling in the preheater and pultrusion dies, which would significantly impact processing consistency. Longer pultrusion lengths may have resulted in higher confidence that steady state conditions had been reached and would have led to better averaging out of potential inconsistencies in the commingled yarn; however, due to the potential for the accumulation of broken fibers and degraded polymer in the die channels to drastically alter results, the usage of additional material did not seem justified. Furthermore, as this study is the first to be performed on the newly built pultrusion system, it was difficult to determine if the correct processing window had been chosen.

2.4 Three-Point Bend Testing

Three-point bend testing was used as a simple method to quantify the pultrudate quality for each processing parameter variant. An increase in three-point bending failure load should be correlated to higher levels of impregnation, as more fibers are able to contribute to the overall strength of the composite material. Five 20 mm long samples from each variant were cut using a bench shear. The samples were taken a minimum of 100 mm apart from each other to help identify any variance that might have occurred during processing. The 3-point bending fixture used a 6 mm diameter loading nose and two 3 mm diameter supports. A span of 12 mm was used to give an approximate span-to-thickness ratio of 16:1 (see results section for final pultrudate dimensions). A Zwick/Roell Z030 universal testing machine was used with a 30 kN Zwick/Roell Xforce K load cell. Failure loads were on the order of 20 N which is below the recommended usable range of this load cell; however, the results showed good repeatability and worked acceptably for basic sample comparison. The rate of loading was set at 1 mm/min and the ultimate force for each sample was recorded for analysis.

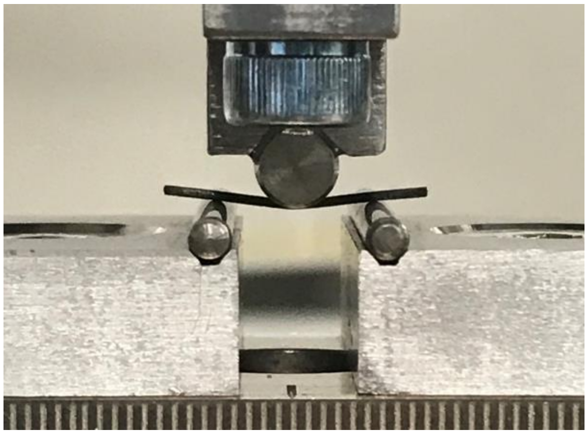

Figure 4. Sample undergoing three-point bend testing.

2.5 Optical Microscopy

To examine the level of impregnation of the pultrudate at different processing parameters, samples from each variant were embedded in epoxy (Struers EpoFix), ground (Struers MD-Piano 120, 220, 500, and MD-Largo with 9 µm diamond suspension), and polished (Struers MD-Dac with DiaPro Dac 3 µm and MD-Nap with DiaPro Nap R 1 µm) using a LECO GPX200 grinder/polisher. A Leica DM6000 M microscope with a Leica MC190 HD camera was used to photograph the specimens.

3 RESULTS AND DISCUSSION

The entrances of the pultrusion dies all showed a small amount of polymer backflow at the beginning of the study, indicating the development of consolidation pressure within the die channels. Unfortunately, several meters into the study the backflow in the middle three dies tapered off and did not return. The first and last die continued to exhibit backflow until the completion of the study which processed approximately 160 m of material. An image of the built-up polymer at the last die entrance upon completion of the study is shown below in Figure 5. Despite the outlet area of the first die being larger than the nominal cross-sectional area of the commingled yarn, a similar level of backflow developed here as to the last die.

Figure 5: Polymer build-up at the entrance of the last die at the end of the study. Note the absence of such build-up on the second last die.

Three-point bend test results exhibited a wide range of average failure loads, from 11.6 N (600 mm/min, 490 °C die temperature, 450 °C preheater temperature) up to 25.6 N (400 mm/min, 475 °C die temperature, 300 °C preheater temperature). There was significant data scatter in the results, potentially due to material inconsistencies along the length of the commingled yarn or fluctuations in the buildup of broken fibers/degraded polymer within the dies during processing; however, some trends may be deduced.

Samples processed at speeds of 200 and 400 mm/min often exhibited higher strength than those processed at 600 and 800 mm/min (see Figure 6). This could be attributed to the lower amount of time that the material would spend at temperature and pressure, resulting in lower levels of fiber impregnation. Interestingly, samples processed at 400 mm/min often showed comparable or better performance than those processed at 200 mm/min. This perhaps indicates that the PEI matrix had undergone a higher level of shear thinning (or thermal degradation) which lowered the viscosity enough to enable faster impregnation rates.

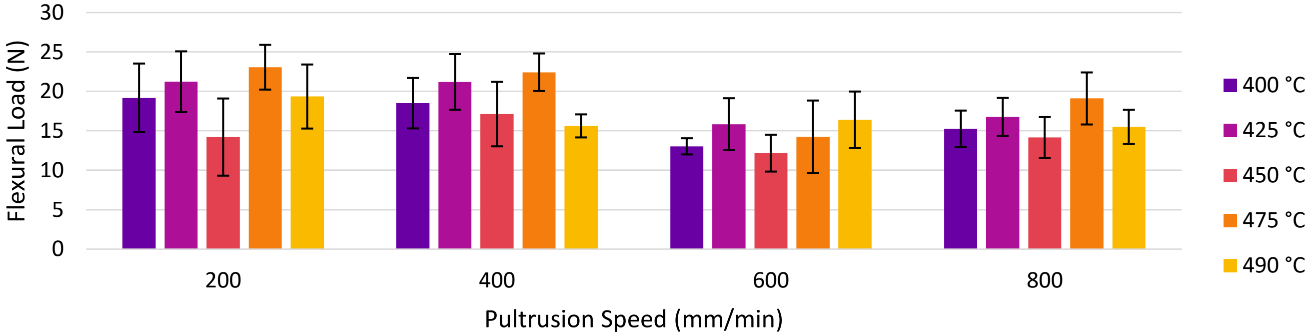

Figure 6. Variation in three-point bending failure load with respect to pultrusion speed and pultrusion die temperature when preheated at 350 °C.

For the processing speeds of 200 and 400 mm/min, an optimum die temperature appears to exist in the range of 425-475 °C. It seems quite likely that debris (e.g., degraded polymer/broken fibers) may have significantly built up in the dies while processing the variants with a die temperature of 450 °C. The results shown in Figure 6 and Figure 7 show a significant drop in performance at this temperature which does not correlate well with surrounding results. When heating the dies to 490 °C, strong smells developed which may have indicated degradation of the polymer. Furthermore, at this temperature the surface quality of the pultrudate began to degrade with small bits of material scattered on the exterior surface. Higher preheater temperatures appear to be more beneficial when using lower die temperatures, and more detrimental when using higher die temperatures.

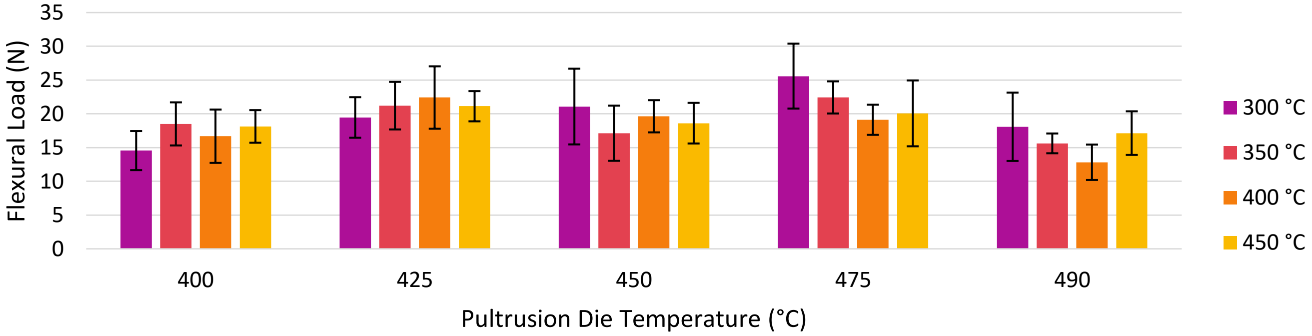

Figure 7. Variation in three-point bending failure load with respect to pultrusion die temperature and pultrusion preheater temperature when pulled at 400 mm/min.

Figure 8 shows microscope cross sections of the worst and best performing variants with poorly impregnated areas highlighted in purple. As can be seen, even the best performing variant is not fully impregnated, indicating that it may not be feasible to achieve the desired results while processing at the speeds used in this study. It is interesting to note that the final width and thickness values of the pultrudate are approximately 0.75 mm rather than the 0.66 mm of the last pultrusion die outlet. A potential reason for this could be relaxation of the polymer matrix upon exiting the pultrusion die, whereupon the consolidation pressure is removed while the polymer is still above its glass transition temperature.

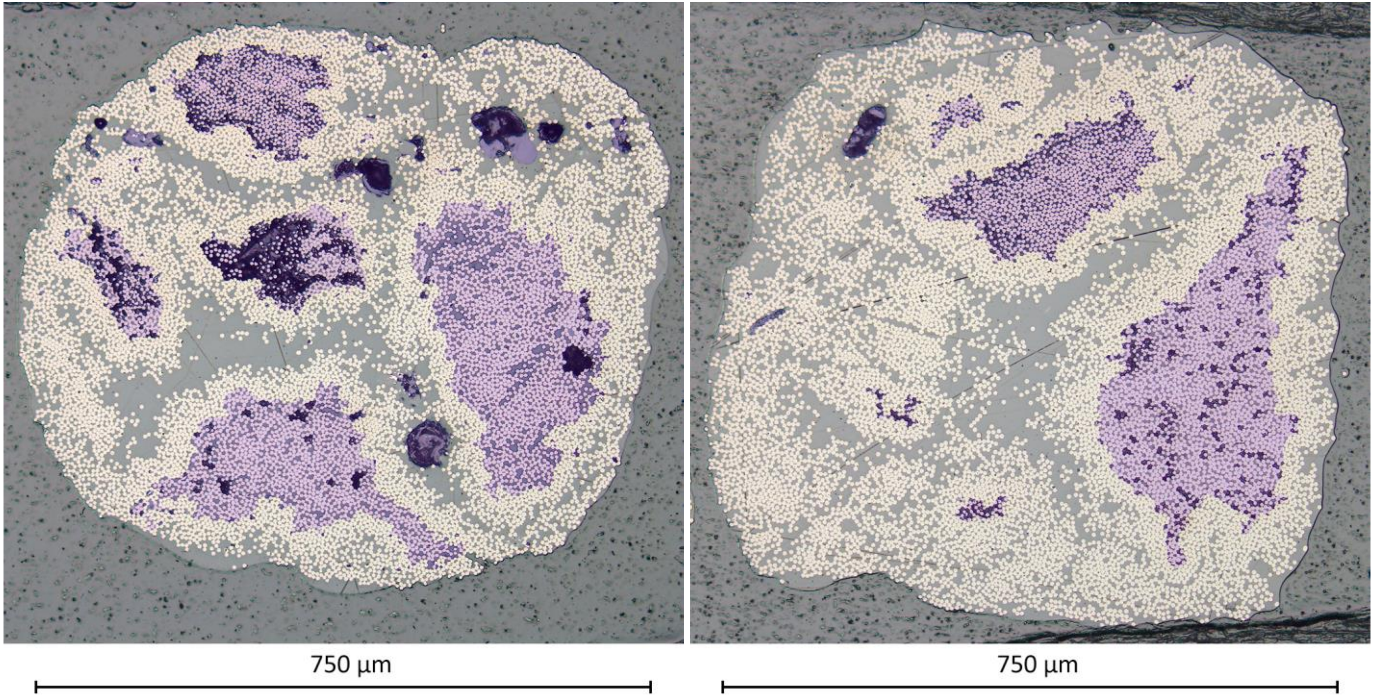

Figure 8. Microscope cross sections of pultruded filament with poorly impregnated regions highlighted in purple. Left: The study's worst performing variant (600 mm/min, 490 °C die temperature, 450 °C preheater temperature). Right: The study's best performing variant (400 mm/min, 475 °C die temperature, 300 °C preheater temperature).

4 CONCLUSION

This study examined the potential for pultruding CF-FFF feedstock materials from commingled yarns at higher than typical processing speeds using an adjustable multi-die system. In an effort to improve impregnation at these higher speeds, high processing temperatures were used so as to reduce polymer viscosity. Unfortunately, large impregnation distances through dense fiber bundles prevented complete impregnation from being achieved. It appears that there was not enough time for full impregnation to occur and that slower processing rates may be required. Future work will examine lower processing speeds along with the pultrusion of alternate matrix materials (e.g., PEEK and PPS).

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