



Development of a melt-impregnation and co-extrusion system for additive manufacturing of continuous carbon fiber-reinforced polyetherimide composites

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ABSTRACT

Impregnating continuous carbon fiber with high-performance thermoplastics such as polyetherimide is often limited by insufficient fiber wetting and low fiber content, which reduces the mechanical performance of composite parts. This article presents a modified melt-impregnation process that incorporates a fiber-spreading mechanism before the heating zone, followed by material co-extrusion of unidirectional composite samples. Four tows were manufactured: Tow A (no fiber spreading, 3 rpm impregnation speed), Tow B (fiber spreading, 2 rpm impregnation speed), Tow C (no fiber spreading, 2 rpm impregnation speed), and Tow D (fiber spreading, 3 rpm impregnation speed). The results showed that changing a single parameter did not significantly increase tensile force. Combining spreading with 2 rpm speed (Tow B) increased mean tow peak force by about 21% and was statistically significant. The composite samples reinforced with Tow B showed a statistically significant 25% increase in tensile strength, consistent with the higher printed fiber volume fraction and improved fiber-matrix wetting observed by scanning electron microscopy. Combining fiber spreading and reduced impregnation speed produced a fiber-rich (high fiber mass fraction), better-wetted, and more consolidated impregnated tow, which enhanced the mechanical strength of the tows and composite parts. The second impregnation in co-extrusion printing enables polymer coating around the fiber-rich tow, improving printability.

1. Introduction

The Material Extrusion (MEX) based additive manufacturing technique is widely used to manufacture composite parts reinforced with short or continuous fibers. However, printing quality is strongly influenced by defects such as weak interfacial bonding, improper fiber wetting, and air voids in the composite structure [1]. Polylactic acid (PLA) is extensively used as a matrix material in composites, but its low toughness, melting point, and inherent brittleness limit its suitability for high-performance applications. To overcome these limitations, high-performance thermoplastics like polyether ether ketone (PEEK), polyetherimide (PEI), and polyphenylene sulfide (PPS) are utilized to manufacture parts with high glass transition temperature (T_g) and melting points.

Raw continuous carbon fiber (CCF) tow cannot be easily mixed with the polymer because of poor adhesion between the polymer and the fibers. Therefore, an impregnation procedure is needed to improve fiber

wetting and increase interfacial adhesion between the polymer and the fiber. In melt impregnation, tow impregnation quality is improved by controlling processing parameters such as impregnation speed, temperature, number, geometry, and configuration of fiber-spreading pins to ensure proper wetting of CCF by the molten polymer [2–5]. Researchers have developed various methods to impregnate CCF tow because the pre-impregnated CCF tow is utilized to fabricate composite structures using the MEX technique [6]. Luo et al. developed an in-situ impregnation procedure for CCF tow [7]. The dry CCF tow was passed through an elevated-temperature extruder, mixed with molten polymer, and then pulled through the nozzle as impregnated tow. This impregnation process had a short contact time between the tow and the polymer, resulting in low interfacial adhesion and insufficient tow wetting.

To overcome low adhesion, Nergün et al. developed a complex melt impregnation line [8]. The CCF was passed through the spreading zone, polymer mixture zone, molding zone, and winding zone to produce good-quality impregnated tow. The fiber-spreading mechanism

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increased fiber surface area, improving fiber wetting [9]. The stepper motor's speed directly affects impregnation quality. Higher traction speed leads to low impregnation and void formation [5]. A drawback of this process is its complexity and the large external setup required for impregnation.

Rimašauskas et al. impregnated the continuous tow using a solution impregnation procedure. The PLA pellets were dissolved in methylene chloride at room temperature; the CCF tow was passed through it, exited the solution through a nozzle, and was wound onto the spool. The use of hazardous chemicals is a limitation of this process; however, the impregnated tow had good mechanical properties and improved adhesion [10].

Pandelidi et al. manufactured composite CCF/PEI specimens using an in-situ impregnation technique. Their research resulted in fiber damage, reduced tow consolidation, void formation, and low adhesion between the fiber and the matrix material. Polyetherimide is a high-melt-viscosity thermoplastic, and the short contact time between the fiber and the polymer caused these issues, resulting in poor impregnation [11]. The MEX-based additive manufacturing of PEI/CCF composites revealed that printing parameters and fiber wetting strongly affect microstructure and processability, indicating that impregnation and printing parameters should be controlled together [12]. Krishnanand and Taufik developed an in-situ filament and pellet co-extrusion system that improved the surface finish in MEX-printed parts, making it suitable for industries requiring high precision. Void formation and poor surface finish adversely affect printed-part strength [13]. Maqsood and Rimašauskas reported that increasing fiber content improved the tensile strength and Young's modulus of CCF/PLA composites [14].

While melt impregnation, fiber spreading, and MEX-based additive manufacturing of CCF composites have been previously reported, most research focuses on either tow impregnation or additive manufacturing. Fiber spreading and impregnation speed are established parameters, but it is unclear whether they work independently or in combination to improve tow impregnation. This study tested the hypothesis that fiber spreading and impregnation speed act together. It was expected that spreading opens the CCF tow, but a lower impregnation speed was expected to provide more time to wet the inner fibers inside the tow to produce fiber-rich impregnated tow. If either parameter is changed independently, surplus polymer will remain on the surface and will not wet the inner fibers, which will decrease the fiber mass fraction and tow consolidation, affecting the tensile force. Four groups of CCF/PEI tows were manufactured: no fiber spreading at 3 rpm (Tow A), fiber spreading at 2 rpm (Tow B), no fiber spreading at 2 rpm (Tow C), and fiber spreading at 3 rpm (Tow D) to test this hypothesis. The material co-extrusion technique was used to fabricate PEI/CCF composites reinforced with Tow A and Tow B. Printed samples were assessed for fiber volume fraction, tensile properties, and scanning electron microscopy (SEM) analysis to understand the relationship between process, structure, and composite properties. This study combined tow and co-extrusion printing performance based on tow impregnation conditions. PEI composite structures have potential for aerospace applications, including unmanned aerial vehicle airframes, landing gear, and wing spars [15–18].

2. Materials and methods

2.1. Materials

This study used high-performance thermoplastic polyetherimide ULTEM 1010, provided by INTAMSYS, as the matrix material; it has a melting temperature of 370°C to 390°C, a Tg of 217°C, and tensile strength and Young's modulus of 90 MPa and 3.4 GPa, respectively. TORAY Japan supplied the CCF (3K, T300B-3000, untwisted, 7 μm diameter) as the reinforcement. The manufacturer listed a tensile strength of 3530 MPa, Young's modulus of 230 GPa, and a density of 1.76 g/cm³ [19].

2.2. Impregnation procedure

A custom-made melt impregnation line was used to impregnate CCF with PEI. Before impregnation, the PEI filament was dried in a universal oven at 200°C for 2 h to remove moisture, then sealed in an enclosed container.

Fig. 1(a) shows the melt impregnation line impregnating the continuous carbon fiber tow. The dry tow entered the heating block through the input nozzle, while the PEI filament was fed and melted at 400°C. The high melt temperature improved PEI melt fluidity, allowing it to enter the CCF tow more easily, as it is near the upper end of the 350–410°C processing window [20]. Tow A and C were impregnated without fiber spreading, with molten PEI filament fed at 25 mm/min and 15 mm/min, respectively, and exited through a 0.8 mm diameter output nozzle, where excess polymer was removed, and the tow was shaped into a circular cross-section. The cooling fan helped to solidify the polymer immediately before the impregnated tow was wound onto the spool. A stepper motor connected to the output spool, controlled by a microcontroller, manages the entire impregnation process. Higher motor speeds reduce tow duration in the melt zone, reducing impregnation quality, whereas lower speeds promote deeper polymer wetting into the tow.

Fig. 1(b) shows the modified impregnation process for Tow B and Tow D. The CCF tows also follow the melt impregnation line, but a series of three convex rollers introduced the fiber-spreading mechanism. The CCF tow width was 2 mm and increased to 4 mm; the stationary rollers' tension spread it laterally, increasing surface area and opening the inner fiber strands, enabling more uniform polymer penetration before it entered the heating block containing molten thermoplastic [21–25]. Three rollers were sufficient to spread the fibers; in this setup, increasing the number of rollers would cause resin agglomeration and fiber breakage, as reported by Ref. [26]. The heating block temperature was kept constant to ensure that molten PEI wetted the spread tow. The PEI feed rate for Tow B was reduced to 15 mm/min, and for Tow D was maintained at 25 mm/min to match the impregnation speed and prevent melted polymer from overflowing the heating block input channel. The fibers exited the nozzle, and the cooling fan solidified them before they were wound onto the spool. The impregnation speed was selected as a processing variable to study the effect of tow duration in the heating block. A lower processing speed gave the fiber more time in the heating block, while a higher impregnation speed reduced the contact time between the fiber and the polymer in the heating zone. The combined effect of fiber spreading and impregnation speed was assessed for the tensile force of the impregnated tow. Zoutsos and Kechagias [27] also justified using low motor speeds. To study the effects of fiber spreading and impregnation speed, four tow groups (Tow A, Tow B, Tow C, and Tow D) were manufactured by adjusting fiber spreading and impregnation speed. Table 1 presents melt impregnation parameters for the tows.

The resin and fiber content were determined according to ASTM D4018 [28], Section 12.3.1, using the mass-per-unit-length method on 1 m dry CCF and impregnated tows. The samples were conditioned at 160°C for 5 h to remove moisture, then weighed on a Kern ADJ 200-4 analytical balance with a readability of 0.0001 g. The resin content was calculated as

$$RC = 100 \times \left(1 - \frac{MUL}{MUL_i} \right)$$

where MUL is the mass per unit length for dry tow, and MUL_i is the mass per unit length of impregnated tow. Meanwhile, fiber content (wt%) was calculated as 100 - RC. Four samples were evaluated for every tow type.

2.3. Additive manufacturing of composite samples using co-extrusion

Fig. 2(a) illustrates the schematic of composite sample

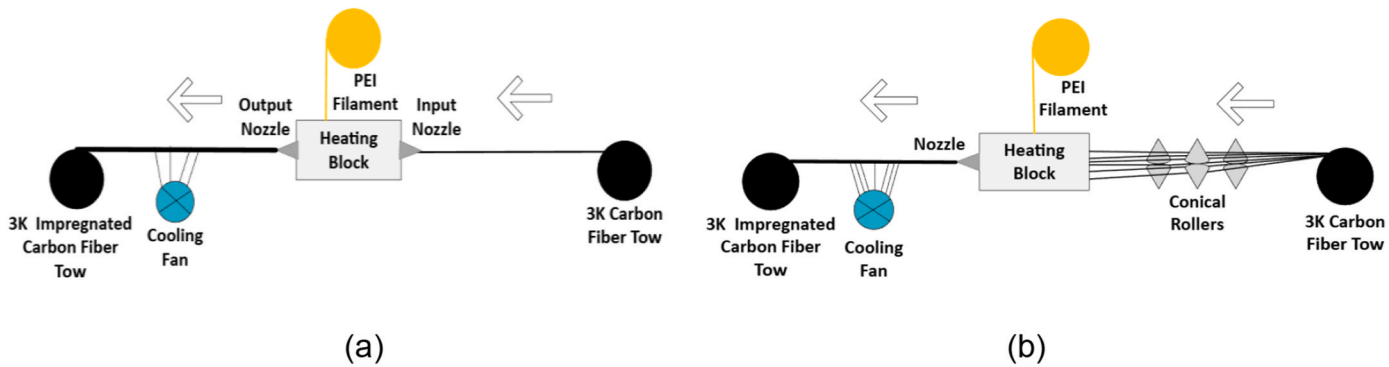


Fig. 1. Schematic diagram of the melt impregnation process; (a) Tow A and C, (b) Tow B and D.

Table 1
Melt impregnation parameters for Tow A, Tow B, Tow C, and Tow D.

Parameters	Tow A	Tow B	Tow C	Tow D
Fiber Spreading	No	Yes, 3 fixed convex rollers	No	Yes, 3 fixed convex rollers
Winding Speed (rpm)	3	2	2	3
Temperature (°C)	400	400	400	400
PEI feed rate (mm/min)	25	15	15	25
Output Nozzle (mm)	0.8	0.8	0.8	0.8

manufacturing with impregnated CCF and a PEI matrix. Fig. 2(b) shows a custom-designed printing head with dual input channels for processing PEI-impregnated CCF tows and PEI filament. The system utilized a co-extrusion approach, permitting simultaneous feeding of the impregnated tow and an additional PEI filament into a dual-input high-temperature printing head. In the first channel, a gear-based feeding mechanism fed the PEI filament into the heating chamber. The second channel is dedicated to feeding the impregnated CCF tow (Tow A and Tow B) directly into the same heating chamber. Inside the chamber, the molten PEI and impregnated tow combined to form a second impregnation, ensuring printability for fiber-rich tows by covering the tow

surface with polymer to print composite structures. This design enabled precise control over the polymer content during printing and improved matrix continuity around the fiber bundle. The fiber orientation was 0° unidirectional, with four layers for each printed sample.

The printing parameters were selected from the literature for CCF composites and refined through printing trials to achieve the best available conditions to print the composite structures [27,29,30]. A 1.5 mm nozzle diameter was chosen to ensure uniform flow of the impregnated tow and PEI filament without risking nozzle clogging. Layer height was set to 0.5 mm, and a printing speed of 180 mm/min was selected to ensure uniform deposition of the co-extruded tow and sufficient consolidation relative to the nozzle diameter. The extrusion multiplier was adjusted to 0.5 based on preliminary printing trials to achieve continuous deposition without excessive PEI extrusion and accumulation around the fiber tow [14]. The high nozzle temperature was required to melt the PEI filament and ensure adequate flow during printing, while the bed was kept at 90°C to improve first-layer adhesion and reduce warping. These parameters were kept constant for every printed sample. Table 2 presents all printing parameters.

The resin and fiber volume fractions for the printed samples were determined from the continuous fiber length and printed sample mass. Fiber length was measured from the printed sample dimensions. The mass of the printed sample was measured on the same analytical scale. Fiber mass was calculated from the linear density of carbon fiber (0.198 g/m) and fiber length. The mass of PEI was obtained by subtracting the

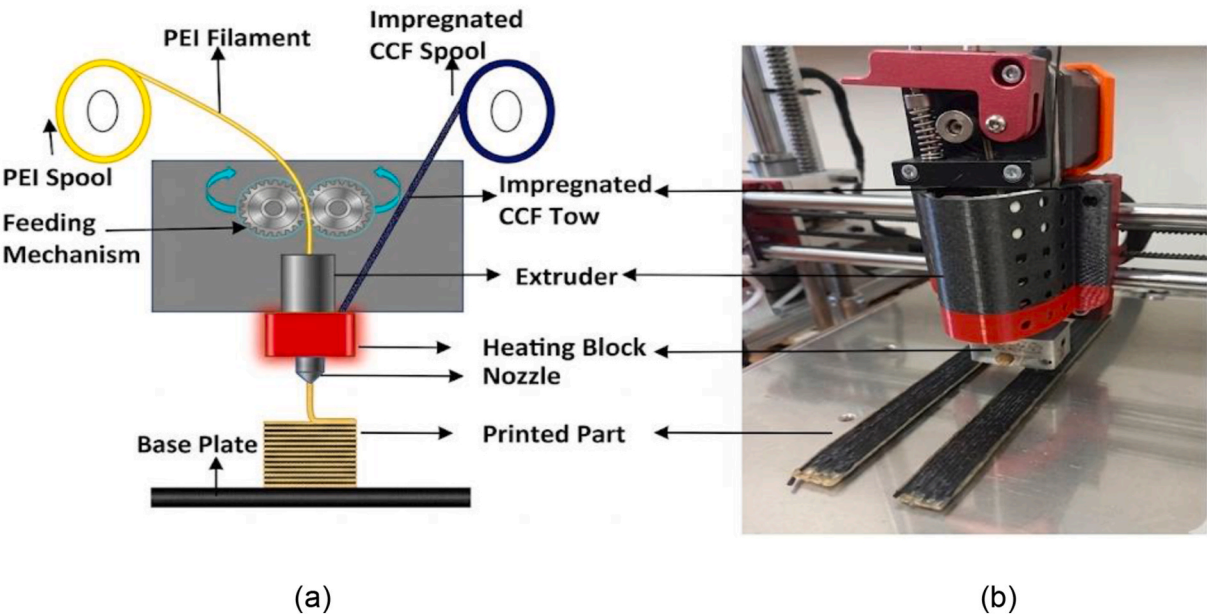


Fig. 2. (a) Schematic diagram of composite printing using co-extrusion technique, (b) Photo of dual channel extruder for co-extrusion printing.

Table 2
Printing parameters for co-extrusion MEX printing.

Printing settings	Value
Nozzle Diameter	1.5 mm
Extrusion multiplier	0.5
Layer Height	0.5 mm
Printing Speed	180 mm/min
Extrusion Temp	400°C
Bed Temp	90°C

mass of the fiber from the total mass of the printed sample. Fiber and resin volume were obtained by dividing the CCF and PEI masses by their corresponding densities (CCF 1.76 g/cm³, PEI 1.27 g/cm³). A total of 10 samples were prepared: five samples printed with Tow A and five samples printed with Tow B.

A rule-of-mixtures (ROM) estimation was used to compare tensile properties for printed samples, using the equation:

$$\sigma_{c,ROM} = V_f \sigma_f + (1 - V_f) \sigma_m$$

where $\sigma_{c,ROM}$ is the ideal tensile strength, V_f is the fiber volume fraction, σ_f is the tensile strength of CCF, $1 - V_f$ is the PEI volume fraction, and σ_m is the tensile strength of PEI. The manufacturer's datasheet provided the CCF and PEI strengths.

2.4. SEM imaging

SEM imaging was used to characterize and observe the cross-sectional impregnation of the CCF-reinforced surface and the polymer matrix surface [31]. The SEM analysis was performed using a Hitachi SU 3500 microscope. Samples for Tow A and Tow B were cut to 50 mm in length using cutter pliers. Similarly, a section from the gauge section of the printed samples was cut to (15 × 12 × 2) mm for SEM analysis. Polymer penetration, dry areas, and dark cavities were compared for Tow A, Tow B, and the samples made from these tows. The printed samples were compared for fiber alignment and wetting. The cavity areas for Tow A and Tow B cross-sections were measured using Fiji (ImageJ).

2.5. Tensile testing of impregnated tow and composite samples

First, tensile testing was performed on four CCF tow types (Tow A, Tow B, Tow C, and Tow D) to assess differences in impregnation and tensile force. The impregnated tow was cut to 200 mm in length, with a 150 mm gauge length. Additionally, 25 mm sandpaper tabs were glued to the ends of the samples to improve clamping reliability. Five samples for each tow type were taken for the tensile testing. The tensile test was performed using a Tinius Olsen H25 KT universal testing machine at a crosshead speed of 2 mm/min. Fig. 3(a) shows the testing samples.

After testing the impregnated CCF tows, composite samples were printed to examine the effect of impregnation on the mechanical performance of the composite structure reinforced with Tow A and Tow B. The unidirectional samples shown in Fig. 3(b, c) were designed according to the ASTM D3039 recommendations. The dimensions of the

samples were (150 × 12 × 2) mm. The 2 mm thickness corresponds to four layers of 0.5 mm height. In total, five samples were printed in each group. These dimensions were chosen due to the build volume limitations of the MEX system. Tensile testing was performed in accordance with the same standard recommendations at room temperature. Additionally, PLA tabs (50 × 12 × 2) mm were printed and glued to minimize stress concentration and better transfer of load to the composite sample. The gauge section was marked with six points to determine longitudinal and lateral strains. Strains were determined using a video extensometer (Tinius Olsen VEM 300). The samples were fixed in manual wedge-action grips and loaded on the same universal testing machine at a crosshead speed of 2 mm/min. Horizon software was used for data acquisition and primary analysis.

Statistical comparison between two independent experimental groups was assessed using Welch's *t*-test with a two-tailed test, assuming unequal variances. A significance level of $p < 0.05$ was used for all statistical comparisons. Statistical analyses were performed in Microsoft Excel.

3. Results and discussion

3.1. Resin and fiber content for impregnated tows

Table 3 exhibits the resin and fiber content wt% for Tow A, Tow B, Tow C, and Tow D. Fiber spreading did not increase the carbon fiber in the tow; instead, it changed the polymer content, which in turn changed the fiber mass fraction. In contrast, combined fiber spreading and lower impregnation speed produced a fiber-rich tow than the non-spreading fibers.

Table 3 shows that fiber spreading and a lower impregnation speed together increased fiber content. For Tow A, the fibers were densely packed, and the high impregnation speed did not allow the molten PEI to penetrate inside the tow and wet the inner fiber strands. Most of the polymer deposited on the top surface, increasing resin mass fraction (wt %), while fiber mass fraction (wt%) decreased. For Tow B, fiber spreading opened the tow, making the inner fiber strands visible; combined with low impregnation speed, this wetted the inner fibers, increasing fiber mass fraction while reducing resin mass fraction.

Similarly, for Tow C, the fibers were not spread, but the impregnation speed was lowered to 2 rpm. This low speed allowed the polymer to remain on the tightly packed tow, increasing resin (wt%) while reducing fiber (wt%) because the polymer did not penetrate the inner fibers. For Tow D, the fibers were spread the same way as in Tow B; the impregnation speed was higher, and the inner fibers were not wetted sufficiently. Most of the polymer deposited on the tow surface, making it a

Table 3
Resin and fiber content for impregnated tows.

Impregnated Tow	Resin Content %	Fiber Content %
Tow A (no spreading, 3 rpm)	53.6 ± 0.6	46.4 ± 0.6
Tow B (fiber spreading, 2 rpm)	39.4 ± 2.9	60.5 ± 2.9
Tow C (no spreading, 2 rpm)	70.0 ± 0.5	29.9 ± 0.5
Tow D (fiber spreading, 3 rpm)	69.3 ± 0.4	30.6 ± 0.4

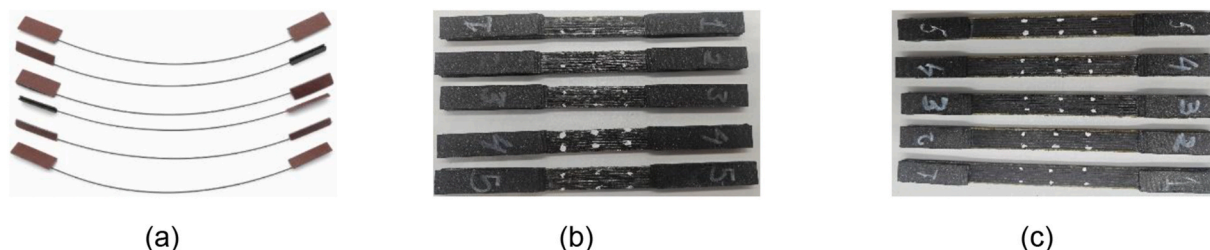


Fig. 3. Tensile testing samples: (a) impregnated (Tow A, Tow B, Tow C, and Tow D), (b) printed samples with Tow B, (c) printed samples with Tow A.

resin-rich tow and lowering the fiber mass fraction (wt%). This condition can also be applied to other high-viscosity matrices where the polymer does not penetrate the fiber due to poor spreading; when the tow is impregnated at higher speed, it deposits polymer on the surface and reduces the tow's fiber content. Resin and fiber mass fraction (wt%) alone cannot determine improved fiber wetting and better impregnation. Visual inspection is also required to determine whether fiber tows are properly wetted by polymer or whether fibers are only covered on the surface.

3.2. Fiber and resin volume fraction for printed samples

Table 4 shows higher fiber weight and volume fractions for samples printed with Tow B than for Tow A. This increase is attributed to the tow impregnation process and the higher fiber content present in Tow B. For Tow A, the dry fibers inside the tow remain tightly packed; hence, the gap is filled by extra polymer residing on the tow surface. During MEX printing, Tow A had a higher PEI percentage in the printed coupon, resulting in lower fiber wt% and vol%. For Tow B, spreading opened the fibers and the polymer penetrated deeper, so the resin amount decreased at the surface; instead, it wetted the inner fibers. The higher tensile strength is attributed to the higher fiber content and improved wetting.

3.3. Tensile properties of tows and printed samples

The tensile testing of Tow A and Tow B was performed; force–displacement curves are depicted in Fig. 4(a) and (b). Five representative samples from each group are presented in the graph, which shows the behavior observed during tensile testing.

The tow results show peak force (N), not tensile strength (MPa). For the 1 m impregnated tow, the diameter was measured at 30 points, and it ranged from 0.39 to 0.58 mm, with an average of 0.457 ± 0.046 mm and a variation of 10%. The impregnated tows did not have a uniform cross-sectional area, and defining a single tow diameter without introducing error is difficult. The maximum tensile force for Tow A ranged from 70 N to 95 N, with a mean tensile force of 87.8 ± 9.1 N. Improper impregnation also caused fiber slippage, as most fibers in the tow did not receive adequate polymer wetting because of insufficient fiber spreading and higher impregnation speed. Moreover, Tow A was not properly consolidated, and the load was not properly transmitted from the matrix to the fiber; therefore, the tensile force was lower. In contrast, Tow B showed higher load resistance and greater displacement before failure. The load ranged from 85 N to 122 N, with a mean tensile force of 106.1 ± 14.3 N. In the elastic region, the samples exhibited a linear response, and a steeper slope indicated greater stiffness and improved load transmission from the matrix to the fiber due to improved fiber-polymer wetting from fiber spreading and reduced impregnation speed. Tow B showed a 21% increase in tensile force compared with Tow A. The higher tensile force in Tow B resulted from better-consolidated fibers and improved impregnation.

To check the impact of fiber spreading and impregnation speed individually, Tow C and Tow D tows were manufactured. The tensile force of five samples for Tow C revealed a mean tensile force of 90.2 ± 14.3 N, while for Tow D, a mean tensile force of 95.0 ± 6.4 N was observed. The results showed that impregnation speed alone did not significantly increase tensile force. When compared with Tow A, the

observed increase for Tow C was 3%. Similarly, for Tow D, fiber spreading and higher impregnation speed resulted in an 8% increase in tensile force compared to Tow A. These two values are not statistically significant (Tow C: $p = 0.77$ and Tow D: $p = 0.19$). Combining fiber spreading with a lower impregnation speed (Tow B) provided a significant 21% increase in tensile force ($p = 0.047$). Thus, to achieve high force, fiber spreading and impregnation speed should be adjusted together.

Because Tow C and Tow D showed low statistical significance and a low increase in force, their curves were not depicted. Only the curves for Tow A and Tow B, which displayed a good increase in tensile force, were reported.

To evaluate the effect of fiber spreading and improved fiber wetting, the tensile testing of the composite samples reinforced with Tow A and Tow B was carried out, and results are depicted in Fig. 5(a) and (b). A total of five samples were printed using Tow A, and five samples were printed using Tow B. The tensile strength for Tow A ranges from 152.8 MPa to 178.2 MPa, with a mean of 162.5 ± 11 MPa. The samples displayed a linear response in the elastic region, followed by a sharp drop after failure. Strain variation can be attributed to non-uniform polymer distribution, poor impregnation, misaligned fibers, and printing defects. Researchers [32,33] also observed low tensile strength due to improper impregnation and low fiber volume fraction.

On the other hand, composite samples reinforced with Tow B exhibited higher tensile strength, ranging from 185.5 MPa to 224.6 MPa, with a mean tensile strength of 203 ± 13 MPa. In the elastic region, the samples displayed a linear response, and high slopes indicate good fiber-matrix bonding. The average Young's modulus of the samples reinforced with Tow A and Tow B is 22.6 ± 1.5 GPa and 25.8 ± 1.4 GPa, respectively, indicating an increase in stiffness due to the effective load transfer between the fibers and the PEI matrix. This increase in tensile strength and stiffness is attributed to a higher fiber volume fraction, better fiber wetting, and improved impregnation. Co-extrusion added a second polymer layer to the impregnated tow, which filled gaps between layers during printing, but it cannot wet the inner fibers inside the tow; this is why Tow A shows lower strength values than samples printed with Tow B under the same printing parameters. Only Tows A and B were printed using co-extrusion: Tow B was fiber-rich and statistically stronger than Tow A; Tows C and D remained resin-rich with no significant increase in tensile force.

Table 5(a) presents the mean tensile force and standard deviation for five samples from each tow in terms of max force and position. The mean peak force increased from 87.8 ± 9.1 N for Tow A to 106.1 ± 14.3 N for Tow B, a rise of about 21%. Tow B's higher maximum force is consistent with its higher CCF content and improved impregnation (60.5 ± 2.9 wt %) than Tow A (46.4 ± 0.6 wt %), resulting in more load-bearing carbon fiber within the tow. Fiber spreading and reduced impregnation speed together promote polymer penetration, fiber wetting, and consolidation, permitting effective load transfer between the PEI and CCF. Compared with Tow A, Tow C increased by 3% to 90.2 ± 14.3 N, while Tow D increased by 8% to 95.0 ± 6.4 N. These results suggest that the increase in tensile force is not only due to fiber content, but that the tow's mechanical response is also affected by the degree of fiber wetting, consolidation, fiber alignment, and distribution.

For the printed samples, mean tensile strength increased from 162.5 ± 11 MPa to 203 ± 13 MPa, a ~25% increase ($p = 0.001$), as presented in Table 5 (b). The mean Young's modulus also increased from 22.6 ± 1.5 GPa to 25.8 ± 1.4 GPa, and the mean tensile elongation at break % increased from $0.7 \pm 0.1\%$ to $0.81 \pm 0.07\%$. These enhancements align with the fiber content after impregnation, the higher fiber volume fraction after printing the samples using MEX, together with improved fiber wetting, better fiber alignment, and consolidation obtained during tow manufacturing. Because fiber spreading and impregnation speed changed simultaneously, these improvements were attributed to the combined impregnation process (fiber spreading and reduced impregnation speed). Increase in tensile strength with increasing fiber volume

Table 4
Resin and fiber volume fractions for samples printed with Tow A and Tow B.

Samples	CCF wt%	PEI wt%	CCF vol%	PEI vol %
Printed using Tow A (no spreading, 3 rpm)	18.8 ± 0.6	81.1 ± 0.60	14.3 ± 0.4	85.6 ± 0.7
Printed using Tow B (fiber spreading, 2 rpm)	22.4 ± 0.9	77.5 ± 0.9	17.3 ± 0.7	82.7 ± 0.7

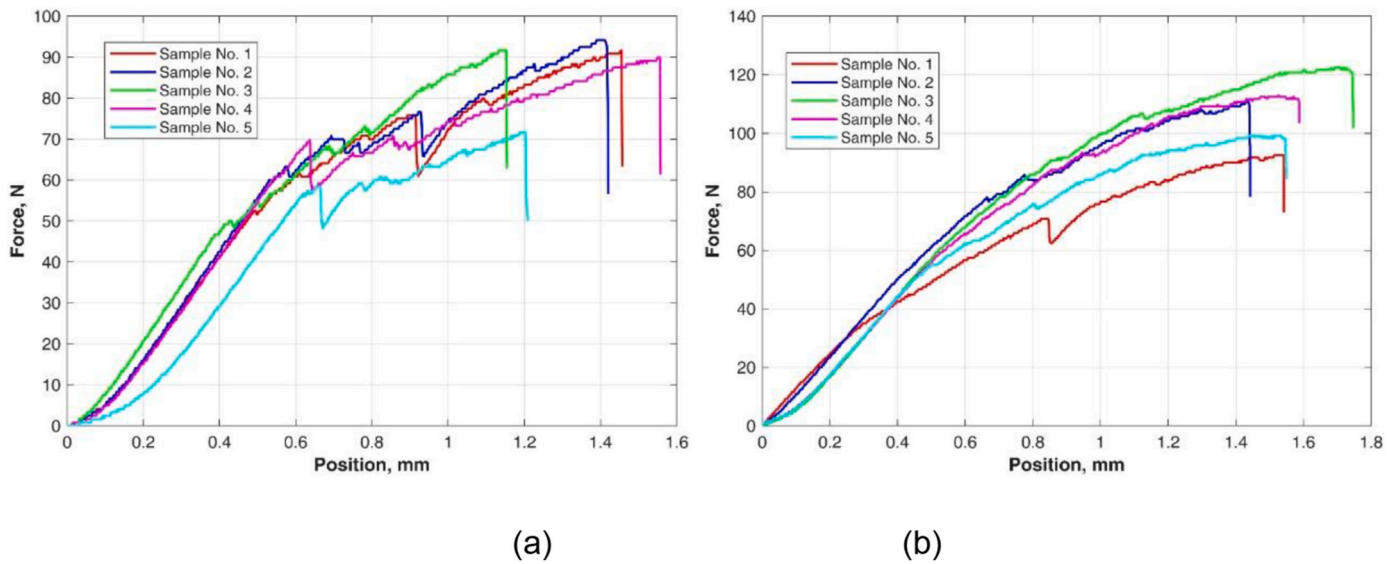


Fig. 4. Tensile testing results: (a) Tow A, (b) Tow B.

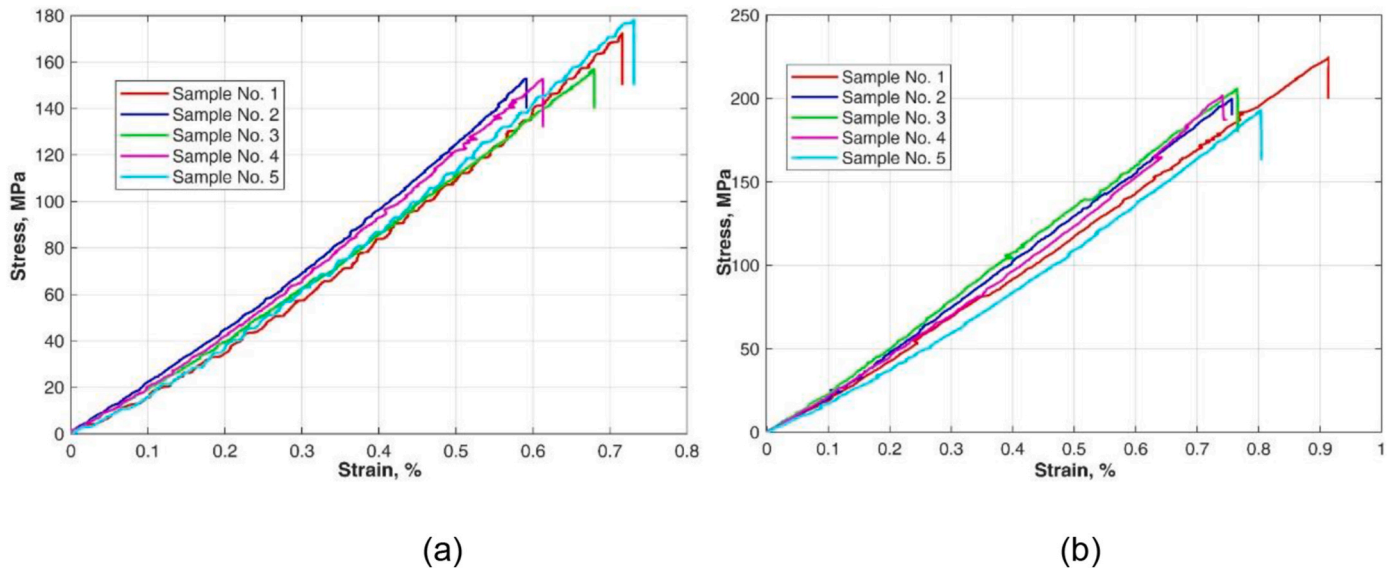


Fig. 5. Tensile testing results of composite samples, (a) reinforced with Tow A, (b) reinforced with Tow B.

Table 5(a)
Summary of tensile force analysis of Tow A, Tow B, Tow C, and Tow D.

Samples	Max Force (N)	Position (mm)
Tow A	87.8 ± 9.1	1.36 ± 0.1
Tow B	106.1 ± 14.3	1.79 ± 0.3
Tow C	90.2 ± 14.3	1.61 ± 0.2
Tow D	95.0 ± 6.4	1.76 ± 0.13

Table 5 (b)
Summary of tensile properties for the samples printed with Tow A and Tow B.

Samples	Tensile Stress (MPa)	Elongation at break (%)	Young's Modulus (GPa)
Printed with Tow A	162.5 ± 11	0.7 ± 0.1	22.6 ± 1.5
Printed with Tow B	203 ± 13	0.81 ± 0.07	25.8 ± 1.4

fraction was reported by Refs. [34,35].

After using the Rule of Mixture analytical model, unidirectional strengths were approximately 582 MPa for Tow A prints and 685 MPa for Tow B prints. The experimental strengths (162.5 ± 11 MPa and 203 ± 13 MPa) corresponded to load-transfer efficiencies of about 28% and 30%, respectively. The difference between the ideal and calculated tensile strengths is due to improper impregnation and printing defects. Previous studies have reported that additively manufactured composite samples can exhibit a void volume of 15% to 20% [36].

The tensile strength of additively manufactured PEI samples ranged from 65 to 85 MPa [37]. In situ CCF/PEI printing reached a tensile strength of 240 MPa, as reported by Ref. [11]. Uşun [33] reported a tensile strength of 509 MPa for CCF/PEEK with a 25% fiber volume fraction. The present composite samples (162.5 ± 11 MPa and 203 ± 13 MPa at 14.3 and 17.3 vol% fiber) remain above neat PEI, and the literature also reports that increasing fiber volume fraction increases tensile strength. For CCF/PA composites, the difference between ROM and the experimentally measured value is attributed to improper impregnation and manufacturing defects [38].

3.4. Process–structure–property relation

The comparison between the four manufactured tows and the tow-printed groups uses a single sequence. Spreading establishes whether the tow is open, and winding speed affects how long the tow stays in the heating zone to mix with molten polymer at 400°C. If the tow is not spread with higher and lower impregnation speeds (Tow A, Tow C) or is spread but the impregnation speed is higher (Tow D), the PEI polymer stays on the surface, with low penetration into the tow. As a result, the fiber mass fraction decreases because the resin mass fraction increases. For Tow B, fiber spreading and impregnation speed changed together, wetting the inner fibers and increasing fiber mass fraction, improving fiber wetting and tow consolidation.

Co-extrusion adds an additional polymer layer that surrounds the existing impregnated tow. It forms a polymer coating that helps print the composite samples effectively by filling gaps in the fiber-rich and resin-rich impregnated tow. It does not further wet the inner fibers of the existing impregnated tow. Tow A and Tow B were coextruded with constant printing parameters, but composite samples printed with Tow A had lower tensile strength and were more resin-rich. This result shows that the second impregnation does not address defects caused by improper impregnation.

3.5. Fracture samples and SEM

The samples printed using Tow A in Fig. 6(a) exhibited failure in the tab section along with a noticeable clustered fiber pull-out as a result of inadequate impregnation because the matrix was not adequately wetting the inner fibers. However, the fibers in Fig. 6(b) remain largely intact within the matrix, demonstrating more effective resin infiltration throughout the tow and improved fiber–matrix load transfer. To support these statements, SEM analysis was performed to assess the impregnation quality of the CCF tow and identify failure modes in composite samples. SEM analysis was performed for the impregnated tow (Tow A and Tow B) and composite samples reinforced with Tow A and Tow B, as depicted in Fig. 7.

It is evident from the images in Fig. 7(a) that Tow A received less impregnation inside the fibers. Most of the matrix material is deposited on the top surface, and little penetrated inside the tow, leaving most inner fibers dry and unconsolidated. Because of improper impregnation, the SEM image showed dry fibers and fiber discontinuities. In Fig. 7(b), the cavities and fiber cross-section were manually marked in Fiji (ImageJ) software. The analyzed area was 402,165 μm^2 , and the total cavity area was 6618 μm^2 , giving a cavity area fraction of 1.65%. Fewer cavities do not mean the tow is well impregnated; rather, the unspread tow fibers are packed tightly, affecting the polymer wetting of the inner fiber strands. Similarly, the SEM analysis was carried out on composite samples reinforced with Tow A. The samples reinforced with Tow A exhibited misaligned fibers, and most fibers within the sample were less impregnated; fiber misalignment and fiber breakage are also visible in Fig. 7(c).

Fig. 7(d, e) depicts Tow B impregnation due to fiber spreading. The PEI matrix was evenly distributed on the surface and penetrated deep

into the fiber tow, wetting the inner fibers due to fiber spreading. The fibers are consolidated and aligned. The analyzed area was 278,889 μm^2 , and the total cavity area was 7499 μm^2 , resulting in a cavity area fraction of 2.69%. The visible cavities resulted from the spreading mechanism from opening the fibers. These SEM values are not a bulk void volume. Automatic thresholding could not separate dark cavities from fiber ends; the regions were manually drawn on one section per tow. The composite sample reinforced with Tow B shows properly aligned fibers with improved wetting and consolidation, as shown in Fig. 7(f).

The higher tensile strength of the samples printed with Tow B (203 ± 13 MPa) compared to the samples printed using Tow A (162.5 ± 11 MPa) correlates with deeper polymer penetration, improved wetting, and fiber alignment due to fiber spreading before impregnation, reduced impregnation speed, and increased fiber volume fraction, as visible for Tow B in Fig. 7(d, e, and f), not to a lower SEM cavity fraction. The quantitative image-analysis results for Tow A, Tow B are summarized in Table 6.

4. Conclusion

Four melt-impregnated PEI/CCF tows were compared—Tow A (no fiber spreading at 3 rpm impregnation speed) and Tow B (fiber spreading at 2 rpm impregnation speed), Tow C (no fiber spreading, 2 rpm impregnation speed), and Tow D (fiber spreading, 3 rpm impregnation speed), followed by MEX co-extrusion printing with Tow A and Tow B. Resin content analysis showed higher fiber content in Tow B, with a higher fiber volume fraction in printed samples made with this tow. Tow C and Tow D remained resin-rich and increased tensile force by 3% and 8% compared to Tow A. SEM analysis of Tow A and Tow B and the printed samples revealed that Tow B achieved improved tow wetting at a lower impregnation speed, increased tow surface area by fiber spreading, deeper resin penetration into the inner fiber strands, and better fiber alignment, indicating improved fiber–matrix wetting. As a result, the mean tensile force increased from 87.8 ± 9.1 N for Tow A to 106.1 ± 14.3 N for Tow B, showing a 21% increase, and the mean tensile strength of the co-extruded composites increased from 162.5 ± 11 MPa to 203 ± 13 MPa with a 25% increase, with Young's modulus increasing from 22.6 ± 1.5 GPa to 25.8 ± 1.4 GPa. Only Tow B vs. Tow A was statistically significant, and the samples were printed from these tows when fiber spreading and impregnation speed were changed together. At the tow level, fiber spreading and impregnation speed together provided good results. The printed samples used the same co-extrusion process; the improved mechanical properties can be attributed to the Tow B impregnation process conditions, improved wetting, and higher fiber volume fraction.

Data availability statement

Data will be made available upon request.



Fig. 6. Printed samples after tensile testing, (a) Tow A, (b) Tow B.

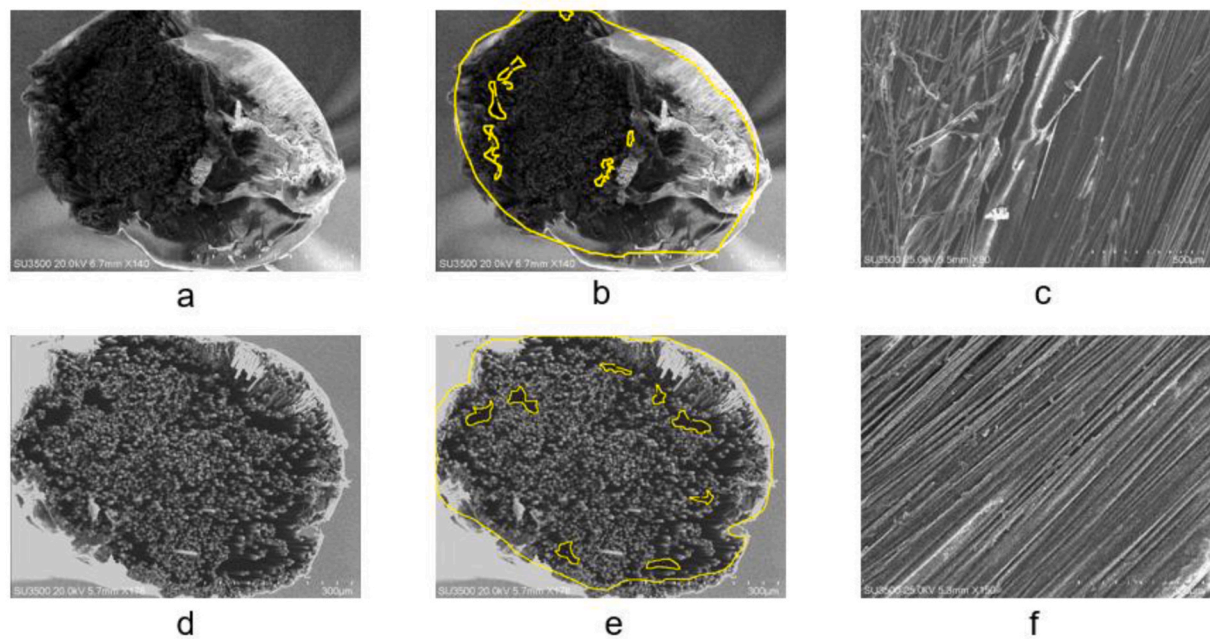


Fig. 7. SEM analysis. (a) Impregnated Tow A cross-section, (b) Tow A cavities marked, (c) Printed sample reinforced with Tow A, (d) Impregnated Tow B cross-section, (e) Tow B cavities marked, (f) Printed sample reinforced with Tow B.

Table 6
Cavity area fractions for Tow A and Tow B using ImageJ (Fiji) software.

Impregnated Tow	Area (µm ²)	Cavity Regions	Total Cavity Area (µm ²)	Cavity area fraction (%)
Tow A	402,165	8	6618	1.65
Tow B	278,889	8	7499	2.69

Authors’ contribution

Muhammad Abuzar Khan: Conceptualization, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft; Marius. Rimašauskas: Conceptualization, Formal analysis, Investigation, Methodology, Supervision, Validation, Writing, Review and Editing; Magdalena Mieloszyk: Formal analysis, Supervision, Investigation, Validation, Visualization, Writing, Review and Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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