Fabrication of Self-reinforced PPS Composites via A Novel Injection Over-molding Method

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Abstract

Polyphenylene sulfide (PPS) composites have demonstrated enhanced mechanical properties through reinforcement with fibers such as glass, carbon, and aramid. However, exploring the performance of self-reinforced PPS composites, particularly utilizing PPS fibers within a PPS resin matrix, remains less investigated. Self-reinforced PPS composites cannot be fabricated using traditional methods such as compression molding due to the similar melting temperatures of the PPS resin and fibers. Here, we present a study establishing a robust fabrication protocol for self-reinforced PPS composites through injection over-molding, embedding PPS fibers within a PPS resin matrix. After extensive trials using both manual and automatic injection molding, we achieved consistent results, helping overcome initial fabrication challenges. Our method focuses on optimizing critical injection molding parameters, including temperature profiles, injection pressures, and holding times, to maximize mechanical performance. We utilized uniaxial tensile testing to quantify the tensile strength the resulting samples, benchmarking these against pure-PPS specimens. Digital microscopy allowed us to evaluate the quality of the composite production by assessing fiber separation during the injection process. Our results show notable improvements in the mechanical properties of self-reinforced PPS composites compared to their pure-PPS counterparts, with the self-reinforced composites with only 9% fiber volume fraction demonstrating an average increase in both ultimate tensile strength and the elastic modulus of 15.66% and 33%, respectively. This research underscores the viability of self-reinforced PPS composites as high-performance materials for various engineering applications, adding a significant dimension to the existing body of knowledge on PPS composites. By demonstrating that consistent and superior mechanical properties can be achieved using both manual and automatic methods, this work contributes to advancing the practical application and manufacturing accessibility of PPS-based composites. These findings have important implications for industries seeking lightweight, durable, and efficient materials, underscoring the potential for self-reinforced PPS composites in addressing contemporary engineering challenges and expanding options in the materials science domain.

Introduction

Existing research has demonstrated the enhanced mechanical properties of PPS materials reinforced with fibers such as glass, carbon, and aramid. Studies have shown that these reinforcements improve tensile strength and fracture toughness, and that the effects are strongly influenced by factors like cooling rates, fiber orientation, and interfacial interactions [2, 4]. For instance, PPS composites with higher crystallinity, influenced by optimal cooling rates, show improved mechanical strength due to better stress transfer from the matrix to the fibers [1]. The orientation of fibers and strong interfacial bonding are critical factors that contribute to these enhanced properties [3].

Despite the promising results from traditional fiber-reinforced PPS composites, the performance of self-reinforced PPS composites using PPS fibers within a PPS resin matrix has been largely unexplored. Self-reinforced composites, where the matrix and fibers are chemically identical, offer unique advantages such as enhanced recyclability and potentially lower cost due to simpler material systems. Furthermore, these composites could reduce issues of thermal expansion mismatch found in heterogeneous composites, leading to better dimensional stability. Our work aims to fill this gap by developing a fabrication protocol for self-reinforced PPS composites through injection over-molding.

Our technical objectives included optimizing injection molding parameters—temperature profiles, injection pressures, and holding times—to maximize the mechanical performance of the composites. We employed uniaxial tensile testing to quantify the tensile of the resulting specimens, while using digital microscopy to analyze the degree of fiber separation during the injection process, ensuring composite quality. These objectives were partially met; after initial challenges with achieving consistency in manual and automatic injection molding methods, consistent results were achieved, allowing us to move forward with relevant testing. Although complete optimization of all parameters and full validation across a broader range of conditions and applications remain ongoing, our progress demonstrates key advancements in composite manufacturing processes, providing new avenues for the utilization of PPS composites in engineering applications.

Progress

In the initial injection over-molding trials (Trials #1-#8), we used the Galomb S-100 injection molder to conduct manual injection molding with the goal of establishing preliminary injection parameters. To prevent the unnecessary use of PPS before determining these parameters, we opted for high-density polyethylene (HDPE) as a substitute matrix. Sheets of HDPE were cut to fit the dimensions of the injection molder's hopper (approximately 16 mm in diameter). For each trial, up to 3.88 g of HDPE was placed inside the hopper, melted, and then dispensed into a circular mold (2.7 mm thick and 37.7 mm in diameter), which held a sheet of PPS fabric intended as the reinforcement material. With literature indicating a melting point of 140 ℃ for HDPE, the material in the hopper was heated to an average temperature of 193.33 ℃ for approximately four minutes in these trials to ensure complete melting. We then injected the HDPE into the circular mold and assessed the extent to which the fibers within the mold bonded with the injected HDPE to form the composite. During Trials #1-2, although the melted HDPE flowed into the mold successfully, our inability to maintain consistent injection speed and pressure led to premature solidification of the HDPE, resulting in incomplete molds. For Trials #3-#8, the mold containing the PPS fabric was pre-heated to temperatures as high as 190 ℃. This was done to mitigate the temperature gradient between the hopper and the mold and to diminish the anticipated impact of inconsistent injection pressure and speed. By Trial #8, these

adjustments resulted in a fully bonded HDPE/PPS composite of adequate quality, allowing us to proceed with trials for producing self-reinforced PPS composites.

In the subsequent injection over-molding trials (Trials #9-#17), we followed the same initial setup as outlined previously. However, due to the absence of PPS pellets that could be readily melted and used as a matrix material, we opted to cut sheets of available PPS fabric to serve as both the matrix and reinforcement material for the resulting composites across all trials. According to the literature, PPS has a melting point of 280 °C, so the PPS placed in the hopper was initially heated to 289 °C, while the mold containing the PPS fabric was pre-heated separately to 250 °C for four minutes.

We experienced immediate difficulties in dispensing the melted PPS fibers into the mold after this period, suspecting that the material cooled and hardened too quickly after ejection. To address this, we gradually increased the temperature in the heating element over subsequent trials, eventually reaching 312 °C by Trial #17 and extending the melting times to a maximum of 12 minutes. We also attempted to raise the pre-heating temperature of the mold and PPS fabric. However, temperatures above 250 °C caused the PPS fabric to deform inside the mold during continued exposure, making it unusable. By Trial #17, we successfully produced a fully bonded, self-reinforced PPS composite using a melting temperature of 312 °C and a mold-heating temperature of 210 °C.

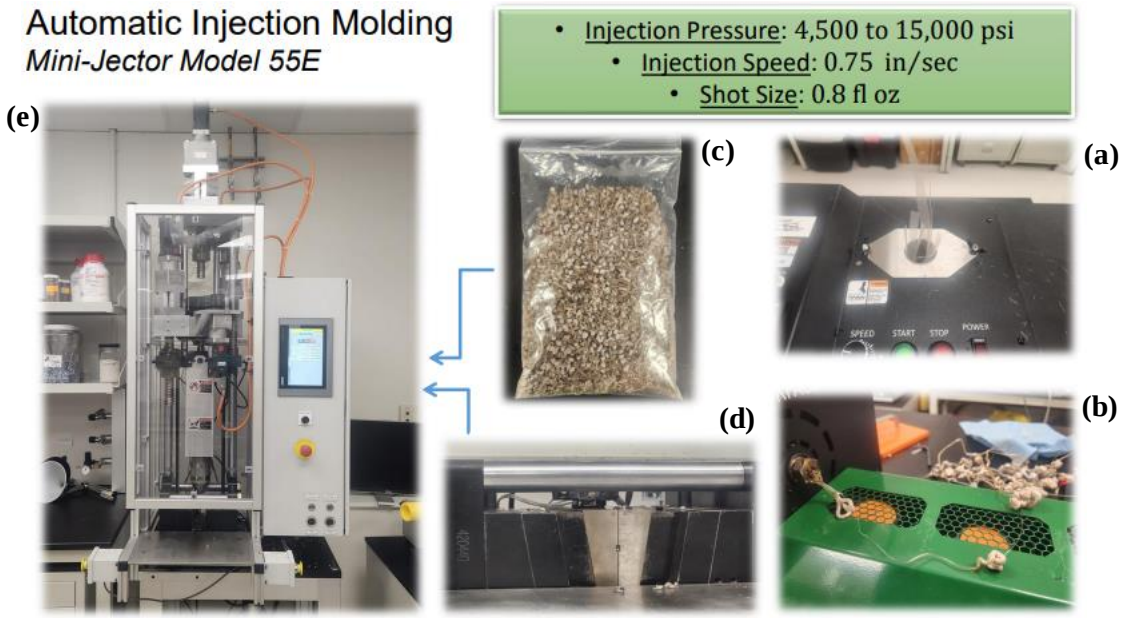


Fig 1. Overview of the final process used to produce self-reinforced PPS composites via automatic injection molding consisting of the (a-b) extruding process, (c) the resulting PPS pellets, (d) the PPS fabric-containing mold and (e) the automatic Model 55E Mini-Jector.

With both preliminary injection and mold heating parameters established, we utilized the Model 55 Mini-Jector to conduct automatic injection molding for Trials #18 through #28, aiming to achieve more consistent injection temperatures and pressures compared to the manual injection molding process. In Trials #18 through #20, we followed the same preparation procedure for the matrix materials as before, filtering the PPS matrix material into the automatic injector and heating it to dispense at 329 °C into the molds. However, the preparation of the fabric-containing mold differed. The molds in the automatic injector featured a trapezoidal

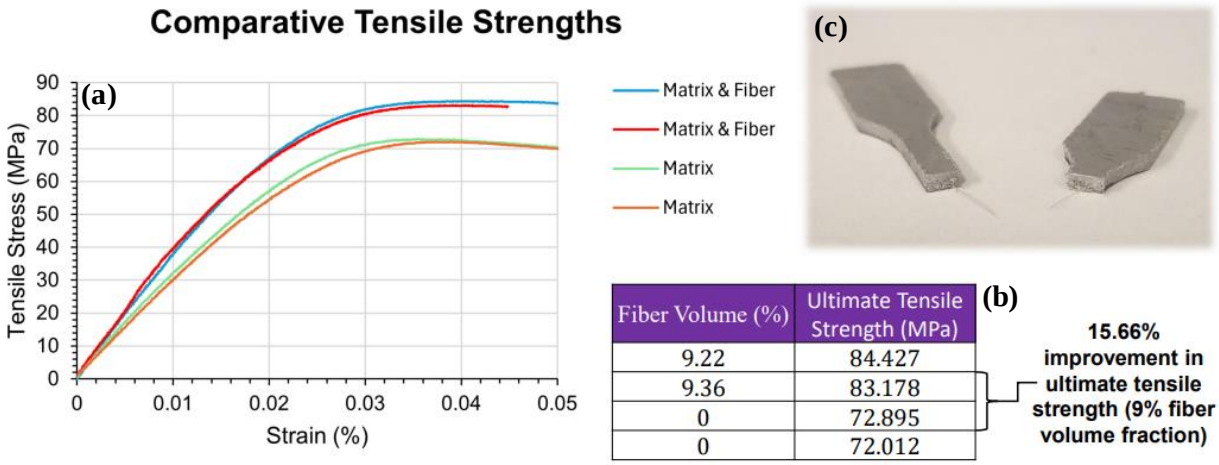
geometry and were not flush with the top plate of the pressing machine used to heat the mold. As a result, all trials with the automatic injector involved heating the bottom and top surfaces of the mold to 200 °C and 250 °C, respectively, to maintain an elevated and relatively even temperature profile throughout the mold.

We encountered similar issues in our initial trials with the automatic injection molder as we did with the manual injection molder. Due to the low density of the PPS fiber that we cut up and inserted, it was challenging to fully compress, melt, and integrate the material once it was inside the chamber, leading to incomplete injection molds in all initial trials. To enhance matrix consistency and mold fabrication, we used Trials #21 through #23 to both: [1] injection mold pure PPS dogbone specimens for later use as benchmarks and [2] to explore further pre-processing methods for PPS fabric before injection. To achieve this, we extruded strips of PPS fabric to produce a continuous plastic, which we then granulated to create small PPS pellets for feeding into the injection molder. This method of matrix preparation allowed for a more complete injection of the matrix material into the mold in all subsequent trials.

In Trials #24 through #28, we continued utilizing this method to produce five self-reinforced PPS composites, making a single modification by lowering the dispensing temperature of the matrix material to 304 °C to ensure material integrity. For Trials #24 and #25, we used a single-sided rectangular mold to hold the PPS fabric, with dimensions of 1.45 mm in thickness, 50.59 mm in width, and 75.99 mm in height. In Trials #26 through #28, we employed a double-sided rectangular mold, with dimensions of 2.88 mm in thickness, 50.59 mm in width, and 75.99 mm in height. A water jet was used to cut each resulting self-reinforced PPS composite into four dogbone specimens for tensile testing.

Fig 2. Overview of the process used for cutting the (a) self-reinforced PPS composites using a water jet to produce (b) tensile specimens which were then subjected to (c) tensile testing to gather experimental data.

We conducted tensile testing on specimens from Trials #23-#28, resulting in 10 pure-PPS dogbone specimens (without reinforcing material) and 12 self-reinforced PPS dogbone specimens. Specimens from Trial #23 exhibited an average ultimate tensile strength (UTS) of 69.301 ± 5.032 MPa, while those from Trial #24 showed an average UTS of 69.189 ± 2.005 MPa. Most self-reinforced PPS tensile specimens had lower UTS values, typically in the 40-50 MPa range. This was largely due to one of two issues: significant or numerous voids in the PPS matrix, which became apparent upon water jet cutting and suggested incomplete extrusion during fabrication, or dislocation of the PPS fabric within the matrix material, leading to an asymmetrical distribution that hindered effective tensile stress redistribution. Both issues were particularly evident in tensile specimens from the double-sided mold in the self-reinforced PPS composites. In contrast, two of the four tensile specimens from the self-reinforced PPS composite of Trial #25 (9% fiber volume fraction), which used a single-sided mold, exhibited a UTS exceeding that of any pure PPS tensile specimens.



Fiber Volume (%)	Ultimate Tensile Strength (MPa)
9.22	84.427
9.36	83.178
0	72.895
0	72.012

Fig 3. (a) Graph comparing the ultimate tensile strengths of two self-reinforced PPS composites with those of non-reinforced PPS tensile specimens. (b) Table comparing the ultimate tensile strengths of different tensile specimens based on fiber volume percentage. (c) A picture of a fractured self-reinforced PPS tensile specimen.

Future Work

Preliminary results have shown the effectiveness of the injection over-molding method in creating mechanically enhanced self-reinforced PPS composites with small (<10%) volume fractions of PPS fabric. Despite this, the number of successful tensile tests remains limited. Future injection molding will utilize larger volume fractions (>30%) of PPS fabric integrated into the PPS resin matrix to explore how increasing fiber composition impacts the tensile properties of PPS composites.

However, the high failure rate among tensile specimens indicates a need to refine the molding process to ensure both matrix and composite integrity. This involves optimizing the pre-heating of the mold and securing the placement of the PPS fabric within it to prevent the fiber dislocation observed in most tensile specimens. Additionally, since processed PPS fabric hasn't consistently facilitated matrix fabrication, PPS pellets will be used in future injection molding to improve the fabrication process.

Impact on Laboratory or National Missions

The development of self-reinforced PPS composites aligns with DOE's goals of promoting energy efficiency and sustainability by offering a lighter alternative to carbon or glass-fiber composites, which can lead to significant fuel savings and reduced emissions in automotive and numerous other engineering applications. The project's innovative approach to materials synthesis using a novel injection over-molding method further supports DOE's focus on advancing sustainable material technologies.

At PNNL, the project contributes to advancing energy resilience by developing cutting-edge composite materials that enhance performance across various sectors, including automotive, aerospace, and defense. This research could augment existing projects at PNNL by integrating new reinforcement methodologies, stimulating new research avenues, and potentially necessitating infrastructure development for testing and manufacturing these innovative materials.

Conclusions

A novel injection over-molding method has been developed to fabricate self-reinforced PPS composites using only PPS fabric. This approach has demonstrated significant improvements in the mechanical properties of the resulting composites by incorporating a 9.29% volume fraction of PPS fibers, on average, into the PPS matrix. Notably, this integration produced an average increase in both ultimate tensile strength and the elastic modulus of 15.66% and 33%, respectively, highlighting the ability of the chosen injection over-molding method to enhance the material's tensile performance effectively. These findings underscore the potential of the new fabrication technique to produce superior PPS composites with enhanced strength and stiffness, suitable for demanding engineering applications.

Acknowledgements

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Appendix

Not counted toward the six-page maximum. Comes after the report's body. The following are additional pieces of information that are required (if applicable—talk to your mentor) in the appendix.

Participants

Name	Institution	Project Role
Ashton Petersen	Santa Rosa Junior College	DOE CCI Intern; assisted in establishing testing parameters; conducted injection over-molding trials and data analysis.
Dounia Boushab	Pacific Northwest National Laboratory	Postdoctoral researcher; coordinated intern activities, tasks, and development; supervised injection over-molding trials.
Yao Qiao	Pacific Northwest National Laboratory	Materials Scientist; served as intern mentor; provided instrumentation training and technical assistance.
Yelin Ni	Pacific Northwest National Laboratory	Materials Scientist; provided technical assistance.
Kevin L Simmons	Pacific Northwest National Laboratory	Research Line Manager; provided technical assistance.

Scientific Facilities

Most of the experiment procedure detailed herein was conducted within PSL, the Physical Sciences Laboratory, a U.S. DOE National Scientific User facility located at the Pacific Northwest National Laboratory (PNNL). However, mechanical testing and analysis of all self-reinforced PPS composites was conducted within MSTL, the Materials Science and Technology Laboratory, a U.S. DOE National Scientific User facility located at the Pacific Northwest National Laboratory (PNNL).

Notable Outcomes

The research efforts detailed in this report will be presented at PNNL's 2025 Research Symposium on April 23, 2025, held at the Energy Sciences Center on the PNNL campus in Richland, Washington. Thereafter, further research incorporating the goals and adjustments in procedure laid out in this report will be used to produce a journal article detailing final research achievements. Owing to such research, a patent may be filed relating to the novel injection over-molding fabrication method used during this research.