Solid Freeform Fabrication 2019: Proceedings of the 30th Annual International
Solid Freeform Fabrication Symposium – An Additive Manufacturing Conference
Reviewed Paper

PROCESSING AND CHARACTERIZATION OF 3D-PRINTED POLYMER MATRIX COMPOSITES REINFORCED WITH DISCONTINUOUS FIBERS

Ankit Gupta*, Seymur Hasanov*, Ismail Fidan†

*Department of Mechanical Engineering,
†Department of Manufacturing and Engineering Technology
Tennessee Technological University,
Cookeville, TN 38505

Abstract

The objective of this study is to fabricate discontinuous fiber (short fiber) reinforced polymer matrix composite material (CM) by additive manufacturing (AM) technology using single extruder 3D printer. For this study, short carbon fibers (diameter = 7.2µm, length = 150µm) reinforced filaments were extruded with fiber concentrations of 3% - 7.5% in volume. Input process parameters used for 3D printing to obtain good quality short carbon fiber (SCF) reinforced polymer specimens are reinforcement percentage and printing speed by fixing nozzle temperature, layer thickness, bed temperature and print orientation. It was analyzed that the surface characteristics and mechanical performance of 3D printed samples are greatly influenced by varying input process parameters. Scanning electron microscopy was performed to observe microstructural behavior of 3D printed samples. Tensile strength, ductility, and toughness were examined to validate the adhesiveness of the matrix and reinforcement. From the microhardness test, it was observed that the hardness properties are significantly affected by increasing the reinforcement percentage. The results obtained in this study could be quite useful in fabricating polymer matrix composites (PMCs) with improved overall characteristics for applications in automotive industry and medical field.

Keywords: Additive Manufacturing, PMC, SF, Tensile strength, 3D printing, microhardness

Introduction

Additive Manufacturing (AM) is becoming an advance technology that is shaping the future of manufacturing processes. AM, also referred as 3D printing that utilizes computerized 3D model data to build a geometry by depositing material layer-by-layer [1]. AM now has far reaching applications, including manufacturing end use products and currently has diverse application areas such as in biomedicine, building construction, rapid tooling [2], automotive and aerospace industries [3].

Fused Filament Fabrication (FFF) is one of the popular AM technologies used in the fabrication of strong, durable, and dimensionally stable parts with the best accuracy. FFF has attracted much attention among researchers and hobbyist, due to its simplicity, the possibility to use various types of materials, low cost process and also the advantage of recycling printed parts. In FFF, components are built layer by layer, so it is possible to produce complex geometries like internal features that would be otherwise difficult and time consuming to fabricate with conventional manufacturing processes [4].

Currently, there has been limited number of studies on developing new materials especially concerning the fiber reinforced composites (FRC) for the FFF process. The need for low-cost, design flexibility and automated process have triggered the development of AM for FRC [1]. The same technology is used to fabricate the SCF reinforced PMC materials with improved mechanical performance, thermal stability, and less weight. So, the goal of this research study is to generate a number of original mechanical findings of the SCF-PMC production processes in FFF. Fig.1 shows the schematic view of the process that how the FFF technology deposits SCF reinforced material layer by layer on the polymer matrix.

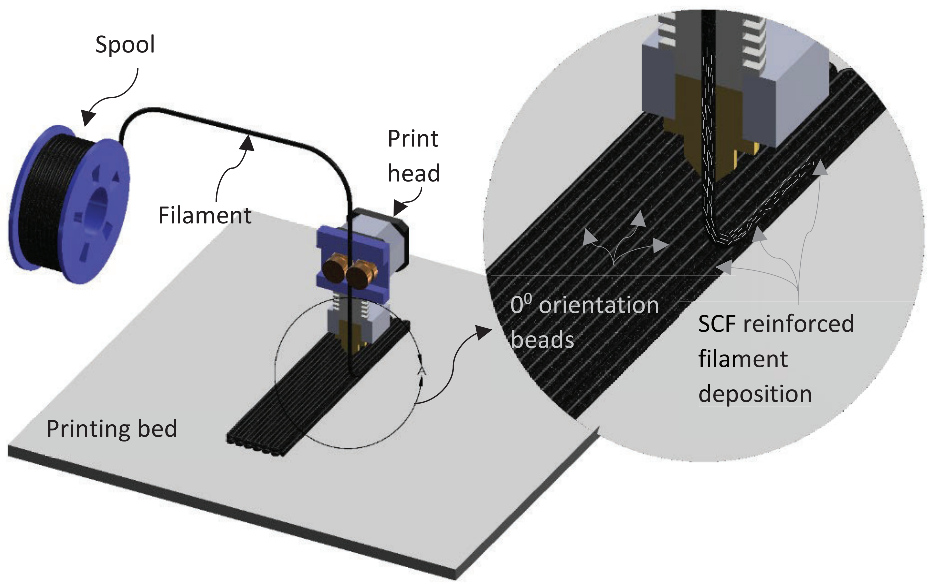


Fig.1. Schematic view of FFF process with incorporated short fibers

SCF-PMC have a wide range of applications due to fiber reinforcement properties and are able to withstand high tensile load and have elastic modulus. Although, SCF-PMC have progressed significantly over the years but further improvement in the fabrication and part properties allow these materials to widen its applications. Mechanical properties of common additively manufactured parts have been evaluated in several studies. However, a few studies have been published to report the findings in detail on the plethora of a new advanced materials with desired surface characteristics and mechanical properties. In this regard, the use of SCF reinforced PMC 3D printed parts is promising and limited work has been done in the literature [5]. Thermoplastic materials like Acrylonitrile butadiene styrene (ABS), Polylactic acid (PLA) and Polyethylene terephthalate glycol (PETG) are embedded with SCF using FFF process and are carefully investigated in the literature because of their common usage in AM based industries.

Tekinalp et al. [6] investigated SCF reinforced ABS composite printing at different fiber loadings to fabricate composite test specimens utilizing FFF technology. Test results showed that there is a significant improvement in both strength and elastic modulus due to 40 wt% of fiber loadings. Ning et al.[1] studied the effects of FFF process parameters on tensile properties of SCF reinforced ABS composites. It has been concluded that tested specimen with 5 wt% carbon fiber content had the largest tensile strength and specimen with carbon fiber content of 7.5 wt% had the largest Young's modulus. Recently, Ferreira et al. [7] have investigated thermo-mechanical characterization of SCF reinforced PETG printed parts using Thermal gravimetric analysis (TGA), Differential scanning calorimetry (DSC) and tensile testing methods. Moreover, El Magri et al. [8] studied the influence of nozzle temperature and infill line orientations for parts made with SCF reinforced PLA composite. The results showed that the combination of 0^0, 15^0, -15^0 orientations relative to the long axis of the test bar yielded the highest levels of tensile properties for both PLA and reinforced PLA.

Studies mentioned above dealt with different types of materials and reinforcements to improve the mechanical performance of the printed objects. This work intends to test and characterize the mechanical properties of polycarbonate (PC) reinforced with SCF composite material. PC is a high performance, tough, amorphous and transparent thermoplastic polymer material that offers a unique combination of properties such as high impact strength, high dimensional stability, toughness, heat and chemical resistance [9]. Domingo-Espin et al. [10] investigated mechanical characterization and simulation of PC parts made by FFF process, aiming to find a model to simulate printed parts and correlate with a finite element analysis (FEA) simulation. In addition, Smith at al. [11] studied the structural characteristics of FFF printed PC material with different orientations. The results showed that there was an approximate 45% decrease in elastic modulus and ultimate tensile strength between 30% to 60% in comparison with a bulk PC material.

The present study used the FFF process to fabricate SF reinforced PC composite material using a single extruder 3D printer and aims to investigate surface characteristics and mechanical properties such as tensile behavior and microhardness of the 3D printed specimens. From the literature survey, it has been concluded that SCF reinforced with PC has not been produced with 3D printing technology and no investigation has been done to observe the effect of printing speed and reinforcement percentage. The specimens produced potentially have a wide range of applications in automobile, aerospace and medical industries.

Materials and Methods

Experimental setup

For the fabrication of 3D printed high temperature PC reinforced with SCF, Ultimaker 3 (3D Universe) was used with nozzle temperature 270 °C and bed temperature 107 °C. 3D printing parameters are clearly shown in Table 1. The setup for this composite material 3D printing includes the single extruder 3D printer, furnace, filament extruder, air path, spooler and dryer as shown in Fig. 2. Low temperature furnace was also used to dry the pellets and SCF before extruding the filament. Single screw filament extruder was used to convert the PC pellets and SCF into the filament form, Air path was used to lower the temperature of the extruded filament before spooling occurs in order to maintain the constant diameter of the filament. 2.85 mm diameter filament was extruded to perform the 3D printing. This filament was used in 3D printer to print the specimens according to required dimensions.

Table 1. 3D printing parameters

Properties	*Values*
Nozzle temperature (°C)	270
Bed temperature (°C)	107
Layer thickness (mm)	0.2
Infill density (%)	100
Infill pattern	Linear
Cooling fan speed (m/sec)	50
Infill pattern direction (°)	0

Material Selection

For the 3D printing process, the PC pellets (3DXTECH) were taken as the matrix material and SCF's (ZOLTEK) were taken as the reinforcement. PC is commonly used in medical devices, automobile industries, digital disks, lighting fixtures, lenses in eyewear, etc. because of its favorable mechanical and thermal properties. In order to widen its applications, PC is incorporated with reinforcements. Reinforcement could possibly enhance its performance in terms of stiffness, strength and hardness by creating strong bonds with PC. Typical properties of the PC (as received) and SCF are shown in Table 2.

Table 2. Matrix properties (Polycarbonate)

Properties	*Values*
Extrusion temperature (°C)	270
Density (g/cm^3)	1.21
Toughness (J/m^3)	0.619
Tensile strength (MPa)	S_x = 54.5 S_y = 58.24 S_z = 58
Tensile modulus (GPa)	E_x = 2.18 E_y = 2.57 E_z = 2.49
Hardness (MPa)	76.59

Experimental Procedure

For the fabrication of SCF reinforced polymer filament, SCF with diameter 7.2 µm and length 150 µm was used as the reinforcement and high temperature PC polymer was used as the matrix material. Three mixtures were prepared with these materials,

1. 97% PC + 3% SCF (by vol.),
2. 95% PC + 5% SCF (by vol.),
3. 92.5% PC + 7.5% SCF (by vol.)

In order to calculate the weights in grams, the volume percentages were converted to weight percentages. The corresponding weights are found out to be 47.74g/2.26g, 46.275g/3.725g, 44.485g/5.515g for composition mixtures of 1, 2 and 3 in 50g mixture. The mixture of materials was dried for 2 hours at 90 °C in low temperature furnace and then the mixture was poured inside the hopper of extruder. Single screw extruder was allowed to rotate at constant speed which forced the material to come out from the 2.85 mm diameter nozzle. Table 3 shows the parameters employed during filament extruding process. Diameter of the filament was precisely checked with the Vernier caliper simultaneously during the filament extruding process.

Table 3. Filament extruding input process parameters

Properties	***Values***
Extruding temperature	270 ^{0}C
Extrusion speed	25 mm/sec
Filament diameter	2.85 mm
Air path fan speed	30 m/sec

Then this extruded filament was used to fabricate the samples according to its dimensions. The entire process was repeated for all the samples by considering the experimental design parameters listed in Table 4. The flow chart representation of fabrication of 3D printed SCF reinforced CM is shown in Fig. 2

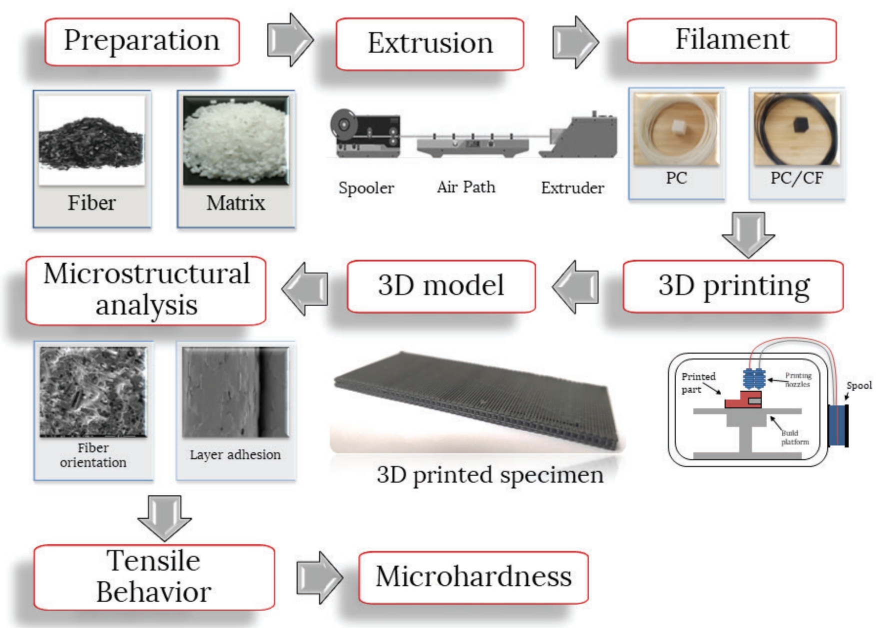


Fig. 2. Flow chart representation of fabrication of 3D printed SCF reinforced composite material

Table 4. Different experimental parameters

S.No.	***Reinforcement % (v/v)***	***Printing Speed (mm/sec)***
1		25
2	0	50
3		75

4		25
5	3	50
6		75
7		25
8	5	50
9		75
10		25
11	7.5	50
12		75

Results and Discussion

Field Emission Scanning Electron Microscopy (FESEM) was performed to observe the microstructural images of the fiber-matrix interface on PC sample reinforced with 5% SCF. Fig. 3 clearly shows that there are insignificant gaps at the interface of the matrix and reinforcement. Interface analysis is important because it strongly influence the tensile properties, microhardness and thermal stability of the final CM [12].

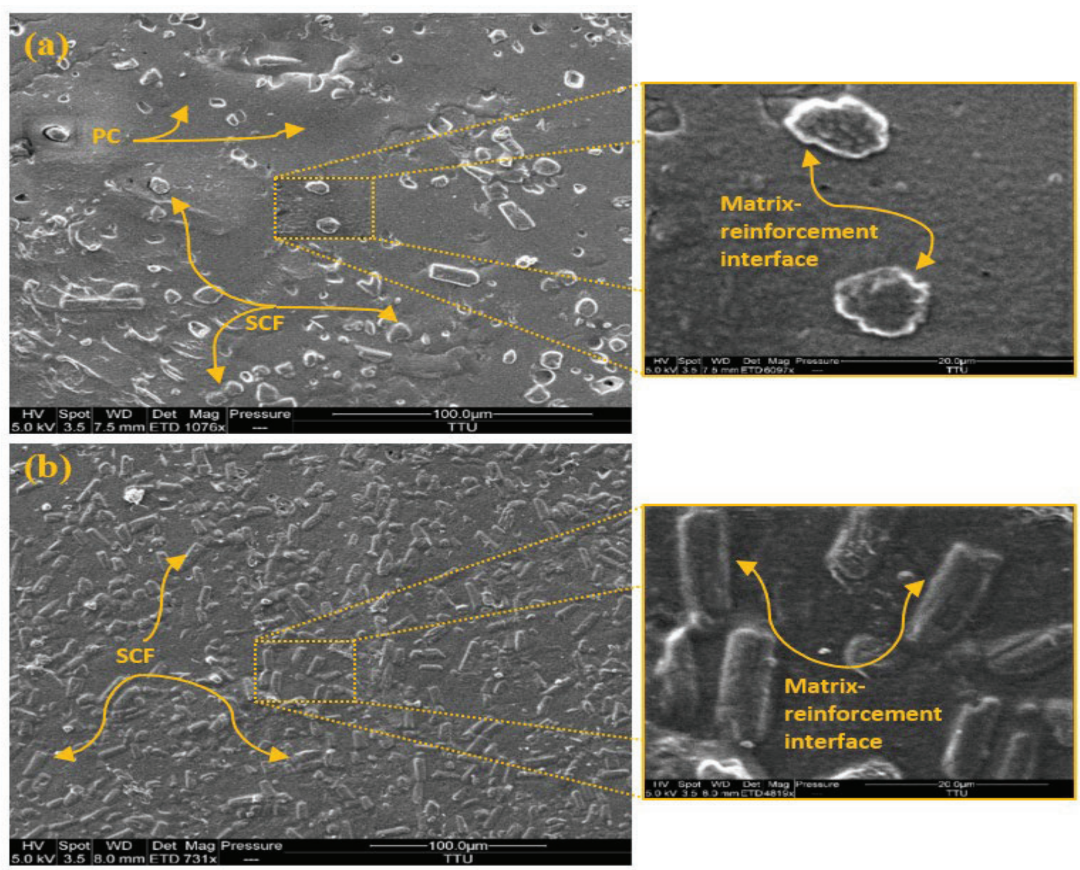


Fig. 3. FESEM images showing no gaps at the interface of fibers and the matrix **(a)** cross section **(b)** side view

Microstructure

After the 3D printing process, FESEM was performed. For the microstructural analysis, a specimen with dimensions 10mm×10mm×10mm was prepared and cut from the middle with the help of a diamond saw cutter. The cut samples were etched with permanganic etching solutions to make the SCF more visible inside the PC. The detailed microstructure characterization of 3D printed PC reinforced with 5% SCF is shown in Fig. 4. From the microstructural analysis, we can conclude that SCF are uniformly distributed inside the matrix material and eventually create the strong influence in enhancing the mechanical properties of the as received polymer material.

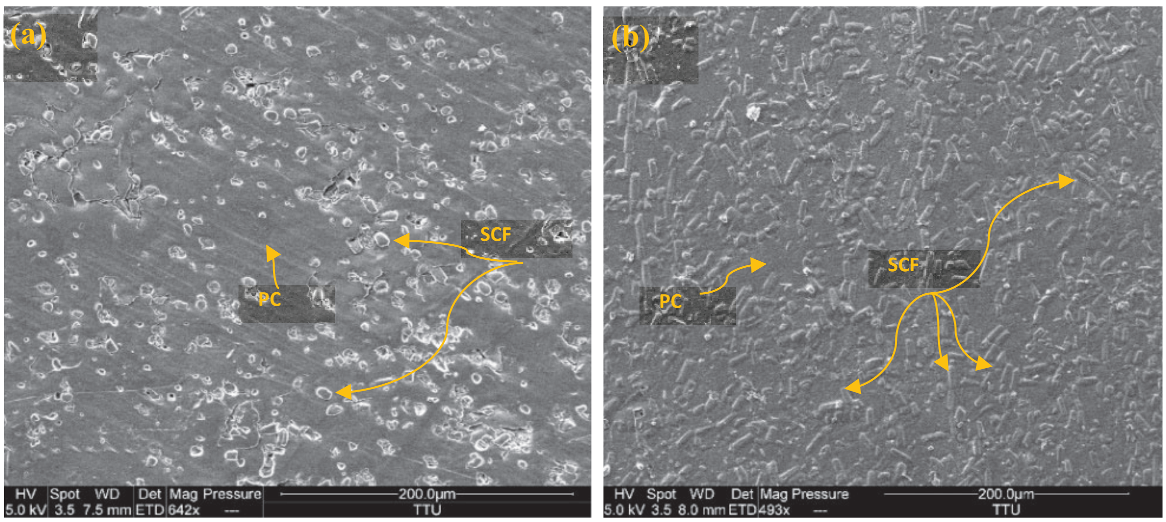
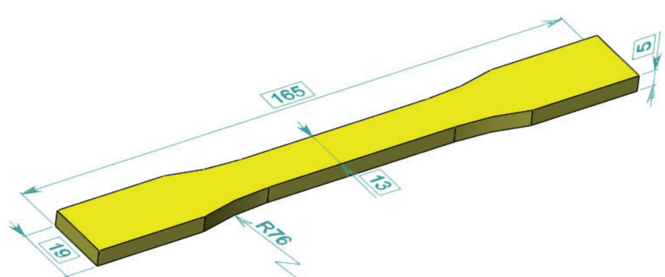


Fig. 4. FESEM images to show the uniform distribution of fibers in 3D printed sample (PC/SCF-5%) **(a)** cross-section **(b)** side view

Tensile strength and Young's modulus

After microstructural analysis, tensile testing was performed with the help of universal testing machine (INSTRON 5582) with ASTM D638 [13] standard as shown in Fig. 5. The change in the tensile strength (TS) and young's modulus (YM) was examined by varying the printing speed and fiber concentrations. There is significant increase in the tensile strength and young's modulus values due to SCF reinforcement. Fig. 6 indicates that the tensile strength increases as we increase the reinforcement of SCF inside the PC. This happened because SCF are uniformly distributed inside the PC matrix material. The uniform distribution of SCF allows the load to be uniformly distributed or transferred from matrix to fibers in the entire 3D printed specimen.



However, tensile strength of reinforced PC is marginally less than pure PC in all the concentrations. Only 3.3% decrease in tensile strength is noticed from the pure PC. The reason for this decrease is that, the SCF inside the matrix material are not perfectly aligned in one direction or to the loading direction. So, if we stretch the sample, the SCF inside the matrix materials acts like a stress concentrator which eventually reduces the tensile strength of the 3D composite material. But in case of young's modulus, CM reinforced with SCF has higher stiffness than the pure PC as shown in Fig. 7, because SCF restrain the polymer chain movement in the vicinity of the other chains. 183% increase in young's modulus is noticed after the SCF reinforcement. Improper melting in the melting zone and slipping of filament in the extruder assembly are some of the reasons of decrease in TS and YM as we performed the printing on high speed [13]. Fig. 8 shows the microstructural images of 3D printed sample with 5% SCF reinforcment after tensile testing. Figure clearly indicates that fiber pull out occurs after the application of tension load which is the other reason of decrease in tensile strength of PC/SCF CM in comparison to pure PC.

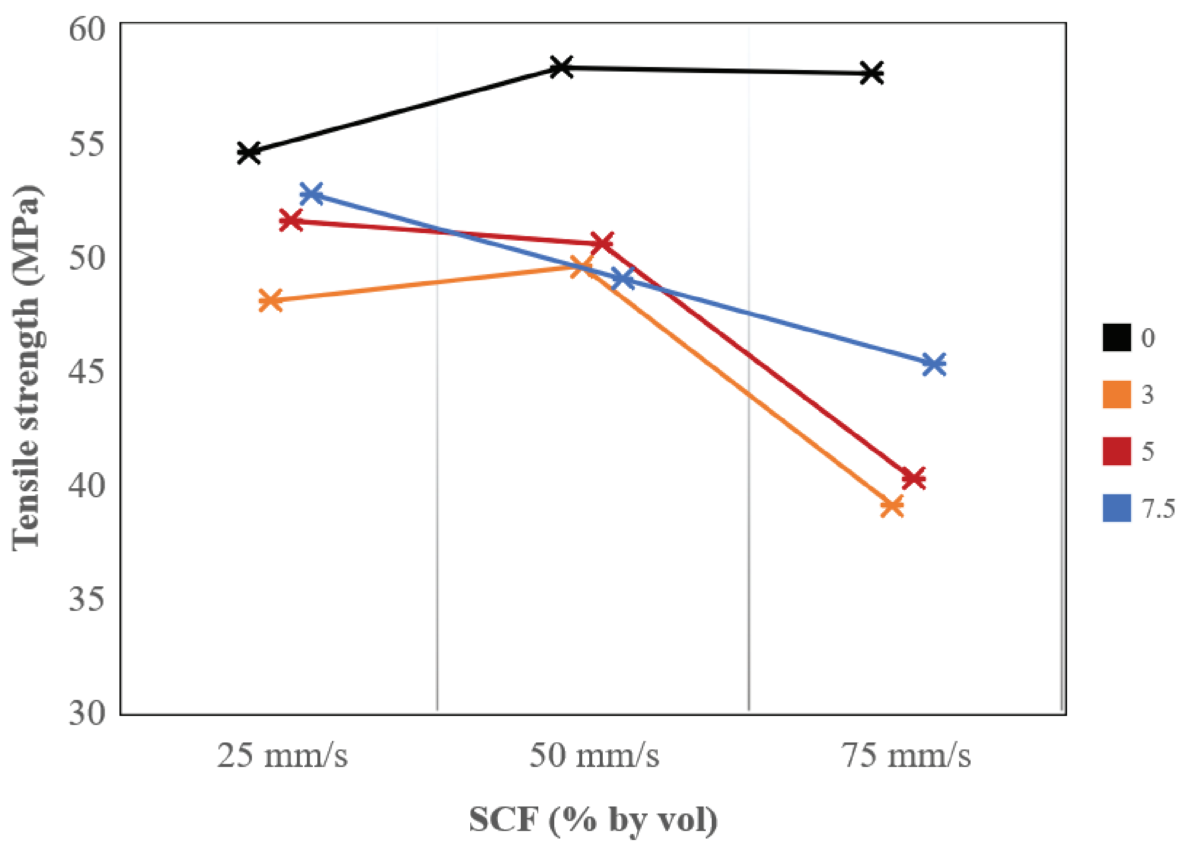


Fig. 6. Effect of printing speed SCF concentration on Tensile strength

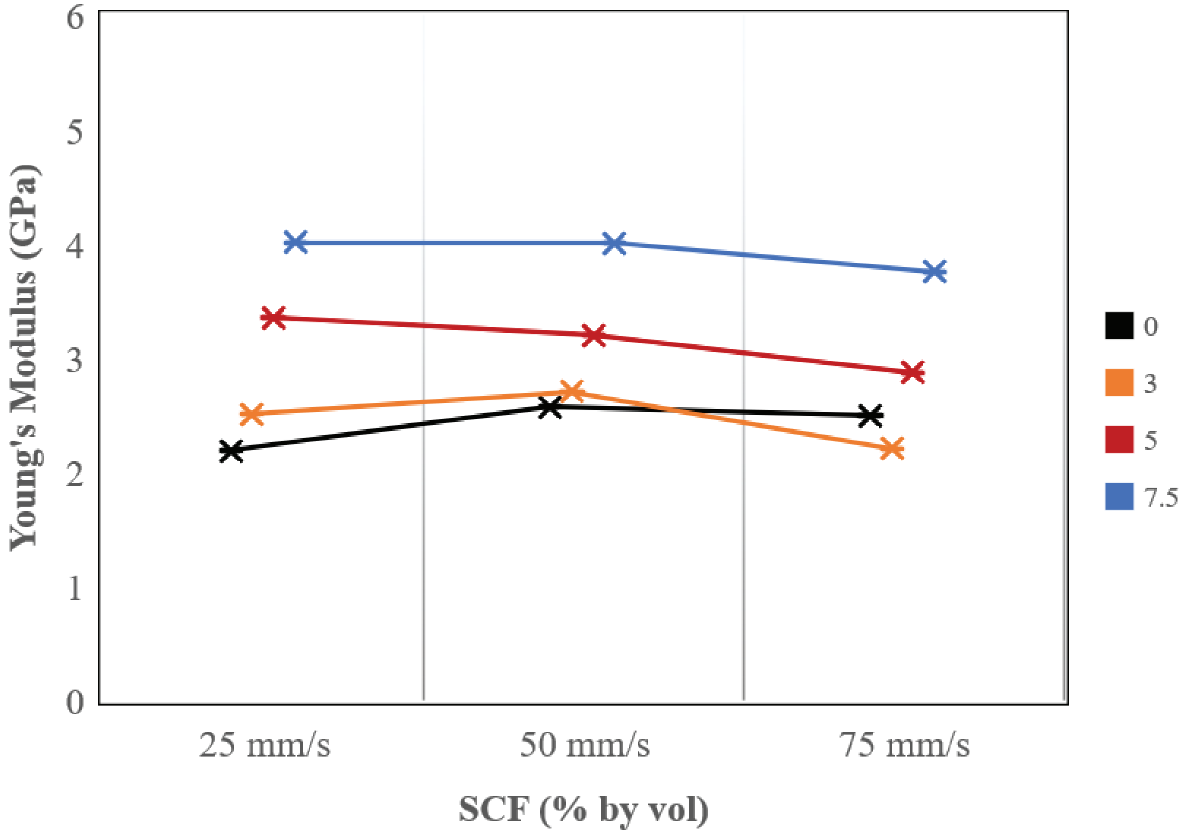


Fig. 7. Effect of printing speed and SCF concentration on Young's Modulus

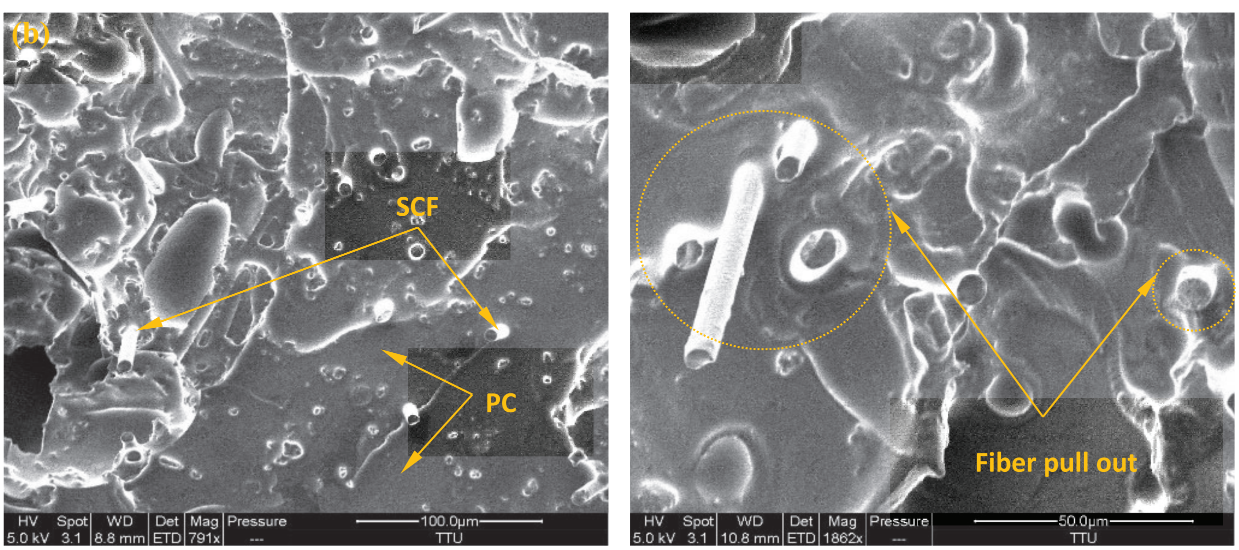


Fig. 8. (a) Dog bone samples of PC/SCF (5%) after tensile testing **(b)** FESEM image showing fiber pull out after tensile testing

Toughness and Ductility

Effect of SCF reinforcement on toughness and ductility was investigated and shown in Fig. 9. As we increase the SCF content inside the PC, the percentage elongation of the 3D printed composite material decreases due to increase in brittle behavior. This eventually results in decrease in toughness and ductility values of the final 3D printed specimen. The lowest mean values of toughness and ductility were observed in PC matrix CM reinforced with 7.5 % SCF [14].

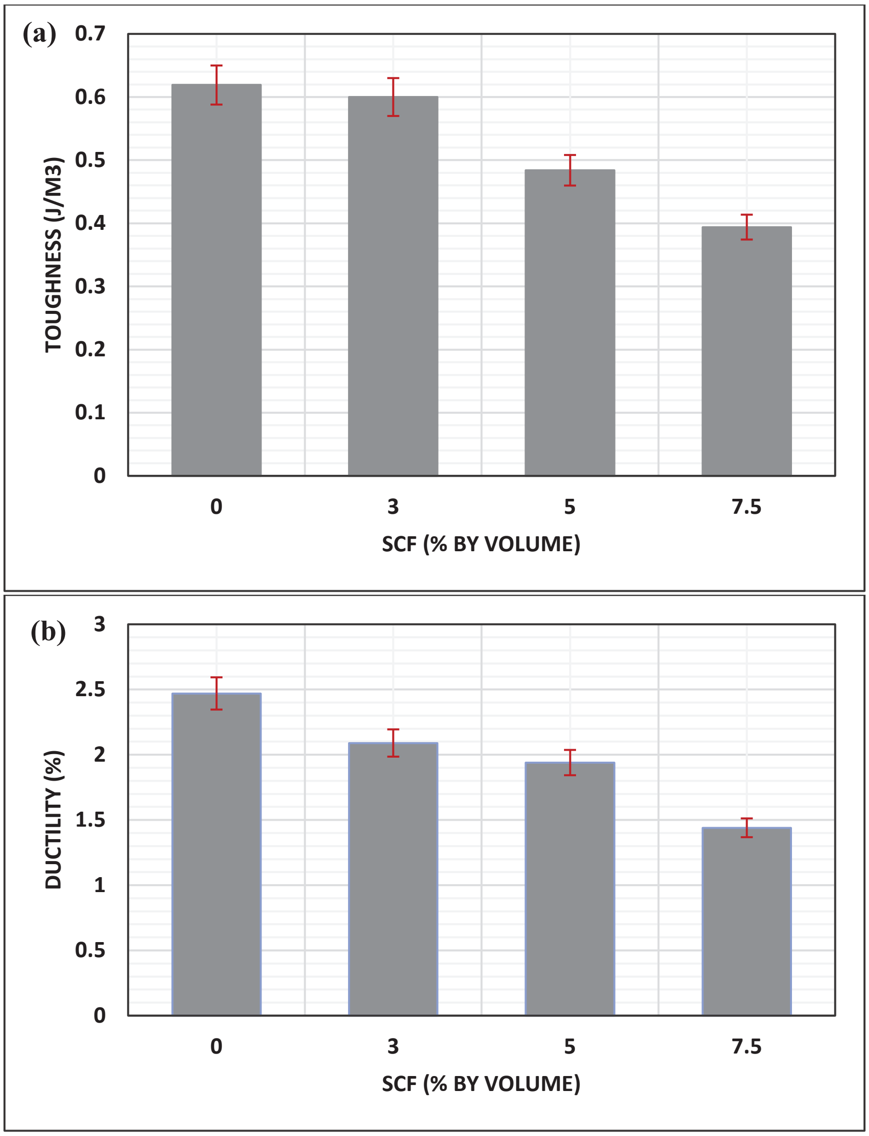


Fig. 9. Effects of SCF concentration on **(a)** toughness **(b)** ductility

Microhardness

Vickers micro-hardness indenter (Beuhler Digital Hardness tester) was used to characterize the micro-hardness properties of the SCF reinforced CM. The tests were performed on 3D printed samples having different concentrations with the load of 50 gf and dwell time of 30 sec. 10 indents were made and 10μm distance [15] within each indent was maintained per sample to find out the hardness values of the CM. From the results shown in Fig. 10., we concluded that the hardness values increase with the increase in SCF content [16] and maximum value (133.4 MPa) is observed in sample with 7.5% (by volume) SCF. This increase in hardness is observed due to uniform distribution of fibers inside the matrix material which decreases the indenter penetration on the surface of the 3D printed SCF reinforced CM.

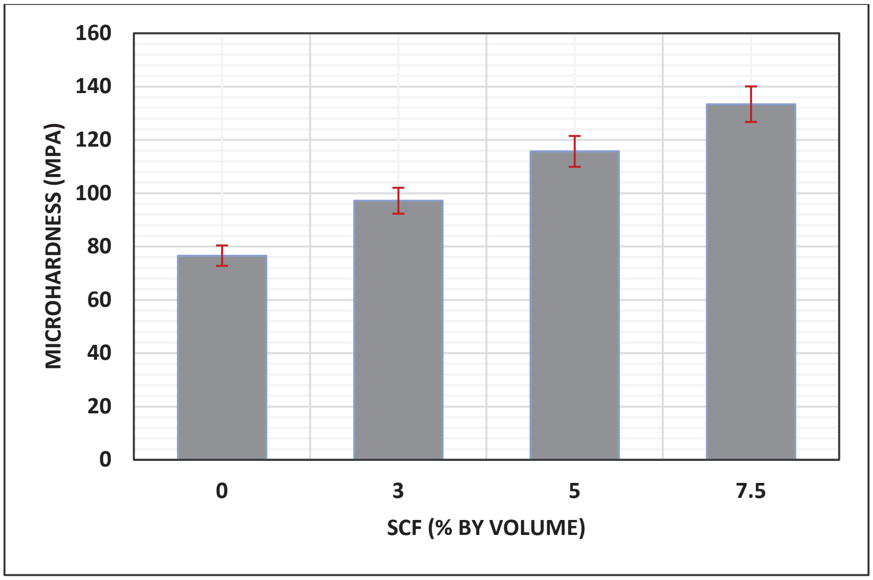


Fig. 10. Effect of SCF concentration on micro-hardness

Conclusion

FFF technology was used to fabricate the SCF reinforced PC matrix composite material. Mechanical properties and surface analyses were performed with respect to different parameter settings and reported in this article. On the basis of all the analysis, the following conclusions have been made.

- SCF are uniformly embedded inside the PC matrix material with insignificant gaps at the interface.
- Tensile strength decreases marginally with the addition of CF inside the PC due to improper alignment of fibers. 3.3% decrease in tensile strength is noticed in the 3D printed CM from the pure PC.
- Tensile strength and young's modulus increase with increase in fiber concentration from 3% to 7.5% due to uniform distribution of load from matrix to fibers. 183% increase in young's modulus is noticed after reinforcing the fibers.
- Tensile strength and young's modulus decrease with increase in printing speed due to improper melting of composite filament. Better properties are achieved for the samples printed on 25 mm/sec.
- Toughness and ductility decrease with increase in SCF percentage inside the PC material.
- Microhardness increases with the increase in reinforcement concentration because of resistance to penetration on the surface of CM. Maximum hardness has been found to be 133.4 MPa with 7.5% SCF.

Acknowledgements

The technical and financial support provided by the Center for Manufacturing Research (CMR) is greatly appreciated.

References

[1] I. Fidan *et al.*, “The trends and challenges of fiber reinforced additive manufacturing,” *Int. J. Adv. Manuf. Technol.*, 2019.

[2] J. Gardan, “Additive manufacturing technologies: state of the art and trends,” *Int. J. Prod. Res.*, vol. 54, no. 10, pp. 3118–3132, May 2016.

[3] G. D. Goh, Y. L. Yap, S. Agarwala, and W. Y. Yeong, “Recent Progress in Additive Manufacturing of Fiber Reinforced Polymer Composite,” *Adv. Mater. Technol.*, vol. 4, no. 1, 2019.

[4] T. N. A. T. Rahim, A. M. Abdullah, and H. Md Akil, “Recent Developments in Fused Deposition Modeling-Based 3D Printing of Polymers and Their Composites,” *Polym. Rev.*, vol. 0, no. 0, pp. 1–36, 2019.

[5] M. Ivey, G. W. Melenka, J. P. Carey, and C. Ayranci, “Characterizing short-fiber-reinforced composites produced using additive manufacturing,” *Adv. Manuf. Polym. Compos. Sci.*, vol. 3, no. 3, pp. 81–91, 2017.

[6] H. L. Tekinalp *et al.*, “Highly oriented carbon fiber-polymer composites via additive manufacturing,” *Compos. Sci. Technol.*, vol. 105, pp. 144–150, 2014.

[7] I. Ferreira, D. Vale, M. Machado, and J. Lino, “Additive manufacturing of polyethylene terephthalate glycol /carbon fiber composites: An experimental study from filament to printed parts,” *Proc. Inst. Mech. Eng. Part L J. Mater. Des. Appl.*, vol. 0, no. 0, pp. 1–13, 2018.

[8] A. El Magri, K. El Mabrouk, S. Vaudreuil, and M. Ebn Touhami, “Mechanical properties of CF-reinforced PLA parts manufactured by fused deposition modeling,” *J. Thermoplast. Compos. Mater.*, p. 089270571984724, 2019.

[9] “Polycarbonate (PC) Plastic: Properties, Uses, & Structure - Guide.” [Online]. Available: https://omnexus.specialchem.com/selection-guide/polycarbonate-pc-plastic. [Accessed: 23-Jun-2019].

[10] M. Domingo-Espin, J. M. Puigoriol-Forcada, A. A. Garcia-Granada, J. Llumà, S. Borros, and G. Reyes, “Mechanical property characterization and simulation of fused deposition modeling Polycarbonate parts,” *Mater. Des.*, vol. 83, pp. 670–677, 2015.

[11] W. C. Smith and R. W. Dean, “Structural characteristics of fused deposition modeling polycarbonate material,” *Polym. Test.*, vol. 32, no. 8, pp. 1306–1312, 2013.

[12] H. Ku, H. Wang, N. Pattarachaiyakoop, and M. Trada, “A review on the tensile properties of natural fiber reinforced polymer composites,” *Compos. Part B Eng.*, vol. 42, no. 4, pp. 856–873, 2011.

[13] ASTM (D638-02a), “American Society for Testing and Materials. Standard test method for tensile properties of plastics (D 638 - 02a) - SCAN VERSION,” *Astm*, vol. 08, pp. 46–58, 2003.

[14] H. C. Wu and V. C. Li, “Trade-off between strength and ductility of random discontinuous fiber reinforced cementitious composites,” *Cem. Concr. Compos.*, vol. 16, no. 1, pp. 23–29, 1994.

[15] S. Misra, M. Hussain, A. Gupta, V. Kumar, S. Kumar, and A. K. Das, “Fabrication and characteristic evaluation of direct metal laser sintered SiC particulate reinforced Ti6Al4V

metal matrix composites," *J. Laser Appl.*, vol. 31, no. 1, p. 012005, 2019.
[16] A. Gupta, M. Hussain, S. Misra, A. K. Das, and A. Mandal, "Processing and characterization of laser sintered hybrid B 4 C/cBN reinforced Ti-based metal matrix composite," *Opt. Lasers Eng.*, vol. 105, no. January, pp. 159–172, 2018.