

Mechanical Behaviour and Compatibility Analysis of Thermoplastic Polyurethane/ Polycaprolactone-Based New Fused Deposition Modelling Filament Composite

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ABSTRACT

The investigation focuses on the development of a Thermoplastic Poly Urethane (TPU), and Polycaprolactone (PCL) based new flexible polymer composite fused deposition modelling (FDM) filament feedstock. In this research study, the mechanical behaviour of the new polymer composite material is fabricated by injection moulding and tested. The mechanical behaviour of injection moulded TPU/PCL composite samples with various blend formulations was investigated experimentally using several mechanical testings. Several combinations of the blend formulations for the new TPU/PCL flexible feedstock was done by volume percentage (vol. %). Based on the experimental data obtained from the mechanical testing done which is the hardness and tensile of the new polymer composite of TPU/PCL has a high potential to be fabricated as the flexible filament feedstock. The blend ratio of 80:20 which as a medium hardness and a higher tensile strength proved to be a highly potential choice to be fabricated as the flexible filament feedstock. The research resulted in the success of extrusion of 1.75 mm of flexible filament for all three ratios of composites and testing it in FDM machine.

Keywords: *Additive Manufacturing, Fused Deposition Modelling, Flexible Filament Feedstock, Thermoplastic Polyurethane, Polycaprolactone*

Introduction

Additive manufacturing (AM) is an advanced manufacturing technique which uses a layer by layer manufacturing to fabricate a solid physical model of a part using three-dimensional computer-aided-design (CAD) file [1]. The standard AM techniques are stereolithography (SLA), laminated object manufacturing (LOM), selective laser sintering (SLS), three-dimensional printing advancement of new materials and enabling technology; these AM techniques emerged out and had been practised in rapid tooling and moulding, direct formed usable part, and bio-manufacturing [2]. (3DP), and fused deposition modelling (FDM). Decades ago, FDM had started to be vastly used additive manufacturing technology for various applications in engineering such as design verification, medical applications, functional testing, and patterns for casting processes [3], [4].

The FDM technique is considered as unique where it is more cost-efficient for fabricating small to medium size parts in the shortest lead time [5]; this is due to the delay in SLA and SLS techniques to build models where the both technique uses resin and laser which deteriorates over time that causes difficulty in ensuring repeatability. Fused Deposition Modelling (FDM) has been most commonly among all other rapid prototyping (RP) processes where it is the automated construction of the physical objects. The FDM process is developed by Stratasys Inc., uses a heated extrusion system with plastic based materials in layer by a layer process using the additive manufacturing technology. The FDM fabricated parts are mainly used for functional testing, design verification, medical applications and patterns for casting processes.

The conventional FDM systems are capable of fabricating parts only using waxes and thermoplastics, and the modern FDM systems are able to process parts in a range of plastics such as acrylonitrile butadiene styrene (ABS), polycarbonate (PC) and blends of ABS and PC. The FDM process includes layer-by-layer deposition of melted material extruded through a nozzle using feedstock filaments from a spool. The basic fundamental of the FDM process, as shown in Figure. 1 provides highly potential for a range of other materials incorporates polymers, and polymer based composites to be researched and developed to be used in the FDM process as long as the newly fabricated material is in a feedstock filament form of which satisfies the required size and mechanical properties[6].

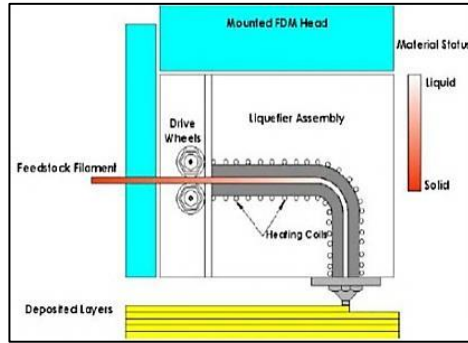


Figure 1: Schematic diagram of the FDM Filament through heated liquefied nozzle

There are needs for new materials for FDM process to increase its application in rapid manufacturing and rapid tooling. Over the years, the amount of research conducted to develop new materials for the FDM process is limited. They had created such components on the FDM system using the mixture of metal/ceramics powders and organic binder system. The mechanical and thermal behaviour of the blended feedstock filament meets the flexibility, stiffness, and viscosity required for successful FDM processing. The vital objective of this investigation is to develop a flexible filament feedstock using TPU and PCL composite for FDM process.

The research concentrates on developing a proper formulation and mixture procedure of TPU and PCL materials to acquire a homogeneous condition to produce the flexible filament feedstock. The primary outcome of this study is to fabricate a flexible, strong, and smooth FDM filament feedstock to be produced by the extrusion machine.

The samples to determine the ideal composition of blending ratio of constituent material was fabricated by an injection moulding process for the mechanical analysis. In this study, there are two dominant polymer materials which were used to develop the new composite material. The main material, used for the polymer matrix composite, is the Thermoplastic Polyurethane (TPU) thermoplastic resin with initial melting temperature approximately 85 to 90 °C, flexible and suitable material for the extrusion process.

Polycaprolactone (PCL) was blended with the TPU thermoplastic material as a binder content to form the flexible polymer matrix composite in Brabender mixer. The additive material is needed to control the melt flow behaviour during an extrusion process. The results presented for mechanical properties.

Experimental

Material

The fabrication of a new polymer matrix composite of FDM flexible feedstock required two types of material, which is a TPU and PCL. The TPU material used is manufactured by The Lubrizol Corporation, United States of America. The material has a specific gravity of 1.13 g/cm^3 and a melting temperature around $85 \text{ }^\circ\text{C}$. Figure 2 shows the TPU resin for blending process.

The PCL utilised in this research is in a resin form supplied by Shenzhen Bright China Industrial Co, China. PCL has a low melting point in the range of 55°C to 60°C and glass transition temperature of -60°C .



Figure 2: TPU Resins



Figure 3: PCL Resins

The blend ratios of the TPU resins and PCL resins are 100% to 80% TPU, 10% to 40% PCL by volume percentage. Firstly, the TPU materials were then loaded in the Brabender mixer, type W50 at $100 \text{ }^\circ\text{C}$ for compounding process in constant speed around 50 Rpm in order to achieve a homogeneous mixture, the compounding procedure was followed. The volume of the mixer is a maximum of 40 cm^3 per mix approximately and 45 minutes to one hour mixing time.

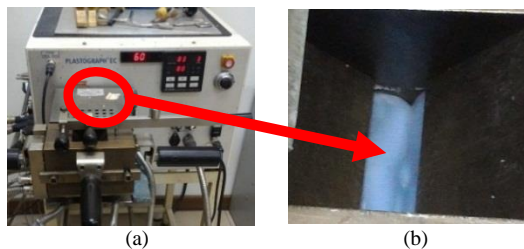


Figure 4: Shows the (a) Brabender mixer, (b) Compounding process of TPU and PCL resins.

All the polymer blends which were mixed has been analysed by using scanning electron microscope (SEM) images to confirm a strong homogeneous compound that is suitable for the FDM filament feedstock preparation. The blended materials were scissor manually to form tiny pellets with 2 mm – 4 mm in size approximately. All the blended composites were rubber-like and elastic and didn't have a breakpoint to be crushed using the crushing machine. The samples for the mechanical analysis testing done by injection moulding the blended TPU/PCL materials on a horizontal NP7-1F moulding machine. Table I shows the composition of constituents of PCL filled TPU composite material for five samples of PMC compounded with the different density value [7].

Table 1: Constituents of PCL filled TPU composites material

Sample	By Percentage Volume of TPU and PCL (Vol %)	
	% TPU	% PCL
1	100	0
2	90	10
3	80	20

Injection Moulding Machine

The specimen was fabricated using injection moulding process. The injection moulding machine specifications are the screw diameter 19 mm, injection capacity 14 cm³, injection rate 50 cm³/second and injection pressure 161 MPa[7]. Standard test specimens of mechanical properties analysis were prepared based on ISO 37 and ISO 178. The temperature zones consist of five areas, where the nozzle temperature was 150 °C, front and middle were set to 150 °C and 150 °C, while for rear 2 and rear 1 was 140 °C and 130 °C. Figure. 5 shows, the injection moulding machine and the zone temperature in injection moulding machine.

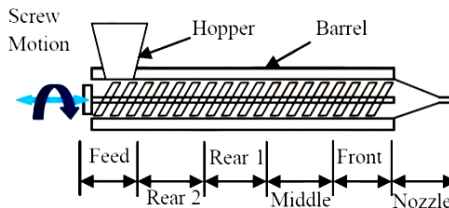


Figure 5: Zone temperatures in injection moulding machine [8].

FDM Filament Fabrication

The FDM filament wire fabrication process is done after all the thermal properties of the new polymers composites are identified. All the sliced polymer composite pellets are fed into the customised single screw hopper extruder which has been arranged and set with a water bath cooling system and also a roller winder. It appears that these new polymer composites have a slow cooling and solidifying characteristic, as an appropriate cooling agent is in need to increase the cooling rate of the new polymer composites.



Figure 6: a) Temperature and screw speed control panel b) Heating barrel and extrusion die c) Hopper



Figure 7: Extrusion Process

Method and Testing

The tensile test is a mechanical test for the evaluation and analysis of the ultimate strength, elongation, modulus, yield strength, and the toughness of material. ISO 527-5a is normally used standard testing for polymer materials.

Tensile stress-strain properties of a polymer and thermoplastic material are tested and identified using the ISO 527-5a testing method. All the injection moulded samples were in a dumb bell shape associated with the ISO 527-5a standard. The tensile testing is done in a Universal testing machine in compliance with ISO 5893. The UTM machine has the capability of performing the tensile test at the rate of traverse of a maximum of 500 mm/min. The crosshead speed used was 5 mm/min about the standard used. This type of UTM machine has dual crossheads where one is adjusted for the specimen length, and another is driven to apply tension to the test specimen. The machine must acquire proper capabilities to test specimen.

The general method to measure the hardness of rubber and soft materials by shore hardness method using a durometer. The penetration of a needle in durometer into the material under specified conditions of force and time was used to measure the hardness. The test method is used to determine the specific hardness of polymers and elastomers or as a quality control to measure on a bunch of materials. The hardness numbers are taken from a scale of Shore D in and converted to Shore A, with the shore A scale being utilised for softer materials and the shore D scale utilised for harder materials. The penetration durometer indenter into the material recorded the value of the hardness.

The FDM filament compatibility test consist of melt flow test and extrusion temperature test which was conducted using the single axis DIY FDM machine and by using the Pronterface software. The TPU/PCL composite results in the melt flow test where it is FDM compatible and can be extruded.

Results and Discussion

The findings of the present study revealed that the influence of different blending ratio of TPU/PCL on the mechanical properties. The tensile and Shore hardness result clarifies the difference in each and every blending ratio.

Tensile Test Results

The pure TPU which is tested without PCL concentration showed the lowest tensile strength compared to all other TPU composites. It was learned by the experimentation and analysis that there is an increment on the tensile strength of TPU polymer after the addition of PCL polymer in it. The incorporation of TPU into the PCL polymer eventually improved the tensile strength up to 10%. As it can clearly be observed that the tensile strength of TPU/PCL composite has increased from 20.85 MPa for the pure TPU to 23.58 MPa for TPU with 10% of PCL concentration but is lower when compared with the TPU with 20% of PCL concentration which has a tensile strength as high as 24.18 MPa.

However, incorporating PCL into the TPU polymer eventually showed a slight increment in the maximum and break strain where the TPU/PCL 80:20 ratio has a higher strain than TPU/PCL 90:10 and pure TPU. The possibility of the PCL addition which could affect the distribution of particles inside the TPU matrix.

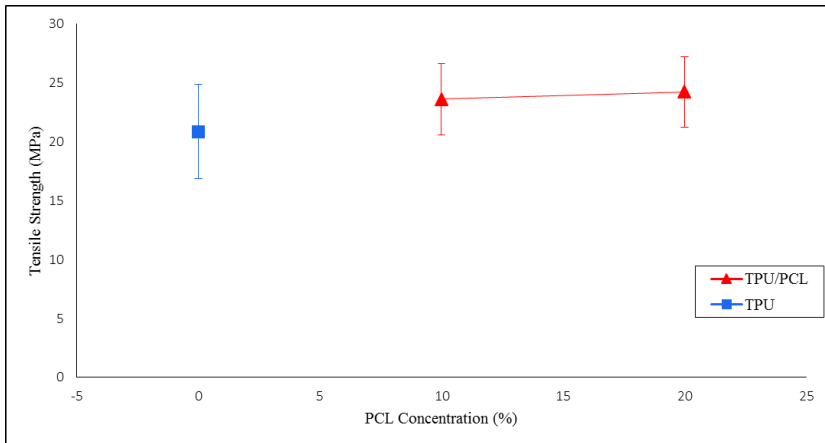


Figure 8: Tensile strength of TPU/PCL composites

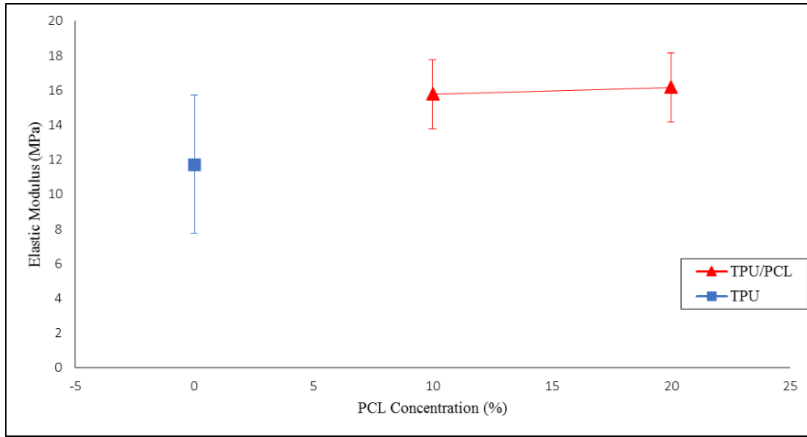


Figure 9: Elastic Modulus of TPU/PCL composites

Based on the graph above in figure 9, it is noted that the elastic modulus of TPU/PCL composites has a sharp increment by adding PCL into the TPU matrix. The elastic modulus of the TPU/PCL composite which has been mixed 20% of PCL was higher than the other TPU/PCL composite which has only 10% PCL concentration. The stiffness and toughness are lowest for the pure TPU where the elastic modulus is lower, and the rubber-like properties should be higher in the pure TPU. The observation on the TPU/PCL composite has an increment on the elastic modulus of where the recorded elastic modulus for TPU with 20% concentration is 24.18 MPa, and it drops as the PCL concentration was constantly decreased by 10 % by volume in the other TPU/PCL composite which also recorded a value at 23.4 MPa. The lowest elastic modulus was detected in the pure TPU composite where the value recorded for the elastic modulus is 20.85 MPa. The pure TPU has been shown to be less stiff than both TPU/PCL composites which show that it has a lower tensile strength than the composites. The trend of the elastic modulus of TPU/PCL composites indicates that the addition of PCL concentration into the TPU composite do make it stiffer when the concentration is increased more than 20% by volume. The elastic modulus data has identified that PCL concentration has reinforced the tensile strength and the elastic modulus of the TPU/PCL composites but has lowered the flexibility of the composites. The highest elastic modulus and tensile strength are found in the TPU/PCL composite which has 20% of PCL concentration in it.

Shore Hardness Results

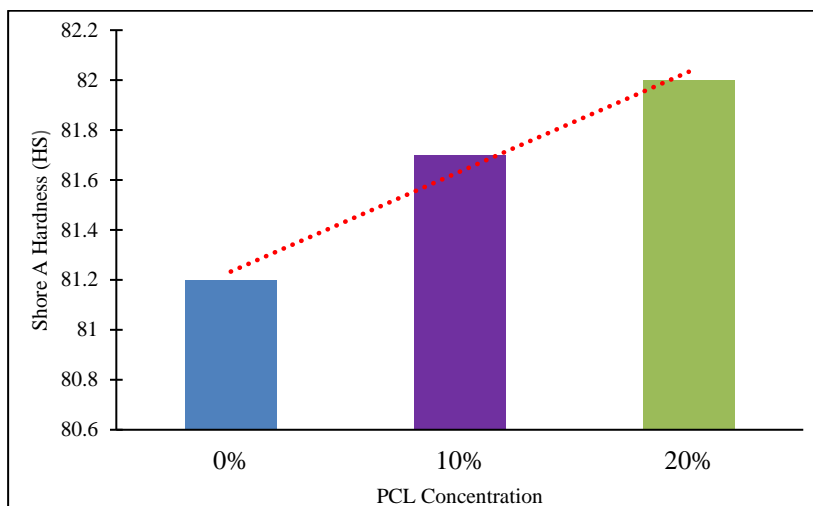


Figure 10: Shore Hardness Type A value of TPU/PCL composites

From the hardness results the characterizations of the injection moulded sample it is clearly shown that the TPU/PCL composite samples are harder than pure TPU. It is because of the effect of the PCL concentration towards the characterization of the materials.

The characteristics of TPU/PCL samples also vary when the percentage of PCL is increased. Based on the results the TPU/PCL composites with 10% and 20% of PCL concentration are observed and identified to be slightly harder than the pure. The result taken from Durometer type D of hardness testing is calculated by its average and accurate value of hardness for each material. The reading of Durometer was quite consistent even when the reading was taking from several different places on the samples. The observation on the results proves that the hardness of TPU/PCL composites is hard to where the hardness of all three formulations of TPU is 80 Hs above. The graph above proves that the TPU/PCL has a similar trend when the addition of PCL in the composite.

FDM Compatibility Test

FDM Melt Flow Test Results

The TPU/PCL composite filaments initially started to melt around 200°C to a maximum of 230°C. The maximum temperature of 230°C was fixed as the extrusion temperature for this melt flow test. The timer was fixed around 10s and the output of the each ratio were calculated and weighed for each 10s. The average value of melt flow were calculated from 3 composition values. Among the three TPU/PCL composites, the pure TPU material shows a higher amount of extrusion output in 10 seconds. The table 2 and figure 11 below show the melt flow test.

Table 1: Melt flow test data

Blending Ratio TPU/PCL	FDM Heating Compatible	Melting point (°C)	Average Extrusion Output (g/10s)
100:0	YES	230	1.38
90:10	YES	230	1.18
80:20	YES	230	1.05



Figure 11: FDM Melt flow test

FDM Extrusion Temperature Test

Based on the FDM extrusion temperature test result shows positive results on the extrusion temperatures of TPU/PCL filaments. The TPU/PCL composite filaments has shown promising results in the extrusion temperature test where it is FDM compatible and can be extruded. The TPU/PCL composite filaments initially started to test at 205°C to a maximum of 230°C. Figure 12 shows the results for the temperature test done.

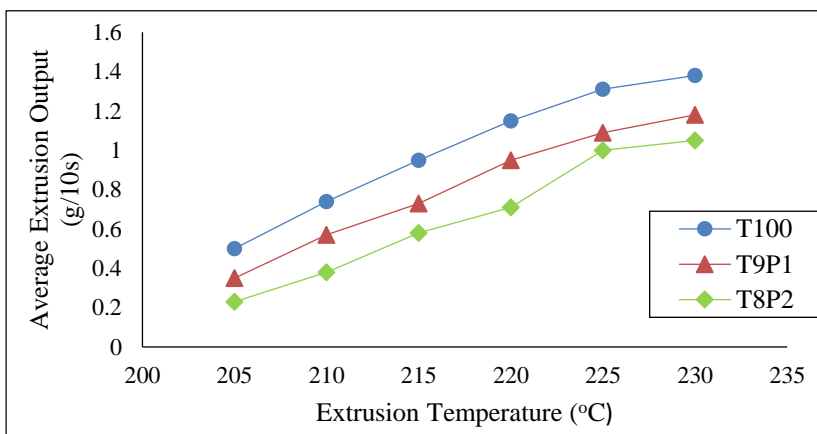


Figure 12: Extrusion temperature test data analysis

There was difference of 5°C between each testing extrusion temperature to get the difference of temperature. The timer was fixed around 10s and the output of the each ratio were calculated and weighed for each 10s. The average value of melt flow were calculated from 3 composition values. Among the six different temperature, it can be said that the temperature from 205°C till 220°C was giving a poor extrusion and output.

Most of the time the filament output is disconnected or slower than 220°C above. The higher temperature was producing more consistent filaments than lower temperature. The temperature around 230°C had excellent extrusion, and the output of pure TPU was better than TPU/PCL composites.

Conclusion

A new TPU/PCL composite material has been fabricated by extruding from the injection moulding machine and tested successfully on the hardness and tensile properties. From the result obtained, it has clearly shown that the mechanical properties of the polymer composite material were enhanced towards the tensile strength, elastic modulus and hardness with the addition of

PCL without effecting the flexibility of the materials. The suitable material with the mixing method and parameter setting on the melting temperature, compression pressure and the cooling time may provide a great potential area for the polymer matrix composite flexible filament feedstock fabrication in the wire filament extrusion through the filament extruder machine. The research resulted in the success of extrusion of 1.75 mm of flexible filament for all three ratios of composites and testing it in FDM machine. The FDM compatibility test has proved that the filaments could be used in FDM machine and the suitable temperatures to be used.

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