

Optimization of Extrusion Process Parameters of Recycled High-Density Polyethylene-Thermoplastic Starch Composite for Fused Filament Fabrication

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Abstract

High-density poly-ethylene (HDPE) is a nonbiodegradable recyclable plastic which is widely utilized in single use packaging applications. Consequently, it constitutes a significant amount of plastic waste found in landfills. From literature, it has been shown that parts produced using composites of HDPE with carbohydrate-based polymers, such as thermoplastic starch (TPS), experience mechanical degradation through hydrolytic degradation process. The possible utilization of recycled-HDPE (rHDPE) and TPS composite in nonconventional manufacturing processes such as Fused filament fabrication (FFF) has however not been explored. This study explores the potential application of rHDPE and TPS composites in FFF and optimizes the extrusion process parameters used in rHDPE-TPS filament production process. Taguchi method was utilized to analyze the extrusion process. The extrusion process parameters studied were the spooling speed, extrusion speed and the extrusion temperatures. The response variable studied was the filament diameter. In this research, the maximum TPS content achieved during filament production was 40 wt%. This filament was however challenging to use in FFF printers due to frequent nozzle clogging. Printing was therefore done with filaments that contained 0 - 30 wt% TPS. The experimental results showed that the most significant parameter in extrusion process was the spooling speed, followed by extrusion speed. Extrusion temperature had the least significant influence on the filament diameter. It was observed that increase in TPS content resulted in reduced warping and increased rate of hydrolytic degradation. Mechanical properties of printed parts were investigated and the results showed that increasing TPS content resulted in reduction in tensile strength, reduction in compression strength and increase in stiffness. The findings of this research provide valuable insights to plastic recycling industries and researchers regarding the utilization of recycled HDPE and TPS composites as substitute materials in FFF.

Keywords

Additive Manufacturing (AM), Fused Filament Fabrication (FFF), High Density Polyethylene (HDPE), Thermoplastic Starch (TPS), Bio-Composite

1. Introduction

Plastics have a wide range of applications across various industries including construction, consumer goods, electronics, agriculture, healthcare and automotives. The increase in popularity of plastics is due to its durability, design flexibility, light weight and cost-effectiveness. One of the emerging technologies that relies on plastic as its primary material is Fused Filament Fabrication (FFF). In FFF, thermoplastic filaments are heated and deposited layer by layer to create a near-net-shape 3D product. The most commonly used thermoplastics filaments are acrylonitrile butadiene styrene (ABS) and polylactic acid (PLA) [1]. Others include Nylon, Polyethylene Terephthalate Glycol (PETG), Polyether ether ketone (PEEK), Polyetherimide (PEI), High Impact Polystyrene (HIPS) and Polycarbonate (PC) [2].

Majority of products made from plastic are utilized for a brief period before being disposed of, creating a pervasive global pollution problem [3]. Consequently, there has been a growing interest in developing alternative materials for FFF such as the use of recycled plastics. Examples include the research done by Herianto *et al.* [4] who conducted an optimization study on the extrusion process parameters for producing recycled polypropylene (PP) filament. Filament with average diameter of 1.6 mm was successfully produced using the optimized parameters. Sotohou *et al.* [5] evaluated the use recycled polypropylene as a base matrix and bean pod powder as natural reinforcing filler to make 3D printing filaments. In this research, it was found that mechanical properties of the resulting composite were dependent on the quantity of bean pod powder present. Mwambe *et al.* [6] successfully produced filaments using recycled polypropylene with 14 wt% carbon fiber reinforcement and also investigated the microstructure and mechanical properties of the filaments and 3D printed specimens.

A different strategy proposed for minimizing plastic waste accumulation involves adding degradable materials to plastics to form composites so as to promote degradation of manufactured parts after the end of their life cycle [7]. Examples include addition of biodegradable agricultural products such as starch or the use of derivatives of agricultural products such as thermoplastic starch (TPS).

1.1. Process Parameters in Filament Extrusion

Production of high-quality filaments is one of the greatest challenges faced when non-conventional materials are to be used in FFF. Filaments should have smooth surface and good diameter consistency. FFF printers use filaments of diameter 1.75 mm or 2.85 mm with a tolerance \pm 0.05 mm. Parameters that can affect the diameter of filaments during extrusion are heater temperature, speed of the extrusion screw, filament spooling speed and filament cooling rate. Wenjie *et al.* [1] investigated the effect of factors such as extrusion temperature and screw speed on the diameter of PLA filaments. Parameters were varied one factor at a time while the rest were kept constant. It was observed that when extrusion temperature is kept constant, filament diameter increased with increase in the speed of the extrusion screw. This was due to increase in the rate of flow of the melted plastic. When the screw speed was kept constant, increasing the extrusion temperature resulted in reduced filament diameter due to reduction in melt viscosity.

Several researchers have looked into optimizing filament production process in order to produce good quality filaments for FFF. Herianto et al. [4] optimized extrusion process parameters for the production of recycled polypropylene (PP) filament. The most significant parameter was found to be the extrusion temperature followed by the spooler speed and lastly the extrusion speed. The filament produced in this research had a curved surface. While the reason for this was not investigated, possible reasons suggested from literature are internal stress caused by uneven cooling of extruded plastic or irregular geometry of the nozzle which can affect the flow behaviour of the plastic. Haq et al. [8] studied the die temperature, roller puller speed, spindle speed, and inlet temperature as parameters for PCL-PLA composite materials. In this research, the results showed that the roller puller speed and spindle speed have a strong interaction as a factor in making filaments. Adenje et al. [9] performed extrusion process optimization of filaments developed from recycled high-density polyethylene (HDPE) and recycled polypropylene (PP). The results showed that recycled HDPE is a comparatively better material for filament production than recycled PP as recycled HDPE exhibited a higher filament diameter consistency and lower melt flow rate. It was however also noted that recycled HDPE had poor printability properties such as warpage and poor adhesion to build plate. The most significant parameter in filament production was found to be screw speed followed by fan cooling and lastly the heater temperature.

As can be seen, most of the research in filament production has been in the area of using virgin or pure recycled plastics. There are few researches that investigate filament production process through extrusion involving blending plastics which exhibit similar properties but optimization is rarely carried out. There is currently very limited knowledge in the possibility of utilizing composites consisting of materials that exhibit dissimilar properties such as recycled HDPE and TPS in additive manufacturing.

1.2. Polymer Degradation

Polymer degradation refers to the change in tensile strength, color, molecular weight, or other properties of the polymer when it is exposed to environmental factors such as heat, light, chemicals, or other external forces [10].

Andrew *et al.* [11] conducted a study to investigate the properties of HDPE:TPS composites made by adding deep eutectic solvent (DES) to promote homogeneity. Both DES and TPS are soluble in water. They found that HDPE:TPS blends remained stable in water at room temperature for weeks. However, boiling the plastic causes the leaching of starch and DES components, which facilitated its mechanical degradation.

Kormin *et al.* [12] developed degradable sago starch-low density polyethylene (LDPE) composites through extrusion and injection molding process and the mechanical properties were investigated. The incorporation of native starch into LDPE was observed to lead to reduction in the mechanical properties. Addition of plasticizer enhanced the interfacial adhesion between the starch and LDPE matrix which consequently resulted in improved performance. Rodriguez-Gonzalez *et al.* [13] employed a different approach of using thermoplastic starch in place of native starch. Polyethylene and TPS blends were processed using a novel one-step process without the use of an interfacial modifier. The PE/TPS blends exhibited comparable levels of ductility and modulus to virgin polyethylene even at high amounts of TPS.

While various researches have been conducted using polyethylene and starch, the possibility of using these blends and composites for FFF are yet to be investigated. This research explores the possibility of utilizing recycled HDPE and TPS in FFF by establishing the optimum range of extrusion process parameters for producing the rHDPE:TPS filaments using Taguchi optimization technique and examining the mechanical properties of the resulting printed objects.

2. Material and Methods

The overview of the materials and steps followed are as shown in **Figure 1**. The extrusion process parameters studied were extrusion temperature, extrusion speed and filament spooling speed. Filament diameter was chosen as the response variable as it showed significant variation when extrusion process parameters were varied unlike other process responses like diameter tolerance and ovality. Tests conducted include tensile test, compression test, 3-point bending test and water absorption test.

2.1. Material Selection

HDPE is a commonly used and widely recyclable plastic, which makes it an



Figure 1. Process Overview.

important material to consider when addressing plastic waste reduction and recycling efforts. When HDPE is combined with carbohydrate-based polymers like thermoplastic starch (TPS), the composite formed undergoes a mechanical degradation caused by hydrolytic degradation. Recycled HDPE (rHDPE) and TPS were therefore selected for FFF filament production process. Pellets of rHDPE with tensile strength of 23.5 MPa and melt flow index of 1.6 g/10min were obtained from Mr. Green Africa.

Pellets of TPS with tensile strength of 10 MPa [14] and melt flow index of 5 g/10min were obtained from Shijiazhuang Tuya Technology Co.

2.2. Experimental Design and Optimization of Extrusion Process Parameters

Pellets of rHDPE and TPS were shredded to reduce particle size to less than 3.5 mm. The shredded materials were then dried in an oven at 70°C for 24 hours to reduce moisture content of TPS to less than 1% for TPS and less than 0.5% for rHDPE as high moisture could result in formation of voids in the filament during extrusion process. The dried samples were then weighed and then mixed in different ratios ranging from 0 - 40 wt% TPS at intervals of 10 wt%. The mixtures were then melted and extruded using SJ 35 single screw extruder machine. The extruder has two heating chambers. In the setup illustrated in **Figure 2**, the water-cooling unit was bypassed and air-cooling method was used as water cooling is not suitable when dealing with hydrophilic materials such as TPS.

1) Preliminary Experiment

Filament production was carried out by varying temperature, extrusion speed and spooling speed. One of the aims of the preliminary experiment was to identify the range of process parameters that resulted in the continuous flow of material at the extruder nozzle without formation of smoke or change in color of the filament due to burning of the thermoplastics. The boundary conditions selected were 165°C - 220°C for extrusion temperature, 5 - 6 rpm for extruder screw speed and 1.5 - 2 rpm for spooling speed. Filament produced using TPS content above 40 wt% had inconsistent flow due to increased tendency of TPS to agglomerate at the infeed section of the screw. Changing the extrusion process parameters when using 40 wt% did not result in improvements in the quality of the filament that was produced. The highest level of TPS content was therefore selected as 40 wt%. The other aim was to determine the response valuables that should be studied. Possible response variables were filament diameter, tolerance and ovality. It was noted during the preliminary experiment face that tolerance and ovality did not vary significantly with change in extrusion process parameters while there was noticeable change in filament diameter. Filament diameter was therefore selected as the response variable in the optimization study.

2) Taguchi Analysis

Taguchi analysis was conducted using Minitab software. The levels of process parameters shown in Table 1 were selected based on preliminary experiment.



Figure 2. SJ35 single screw extruder machine.

	Temperature Profile (°C)						Extrusion speed (rpm)			Spooling Speed (rpm)		
TPS Content	Level 1		Level 2		Level 3		Level 1	Level 2	Level 3	Level 1	Level 2	Level 3
	Heater 1	Heater 2	Heater 1	Heater 2	Heater 1	Heater 2						
0%	220	215	215	210	210	200	5	5.5	6	1.5	2	2.5
10%	185	180	180	175	175	170	5	5.5	6	1.5	2	2.5
20%	180	175	175	170	170	165	5	5.5	6	1.5	2	2.5
30%	180	175	175	170	170	165	4.5	5	5.5	1	1.5	2
40%	170	165	165	160	160	155	4	4.5	5	1	1.5	2

Temperature profile parameter setting involved selecting the temperature setting of 2 heaters. It is recommended that the primary heater (Heater 1) be at a higher temperature than the secondary heater (Heater 2). The response variable selected was filament diameter. Taguchi L9 orthogonal array was selected as the design matrix to be used in the experiment. The optimum process parameters were determined after conducting the experiment which involved filament extrusion and obtaining measurements of the resulting filament diameter. Extrusion was then carried out using the optimum parameters to verify the results. Spools of filaments were then produced using the optimized and verified parameter and these filaments were then used in the printing stages.

2.3. Characterization of Filament

Properties of filaments produced using the optimum parameter were analyzed based of the consistency of their diameter as well as their surface roughness. Filament diameter was measured using a digital vernier calipers. Measurements were taken at intervals of 10 cm for filament lengths of 5 m and the mean value of the diameter calculated. On the other hand, Surface Roughness analysis was conducted using Marsurf CM explorer. The arithmetic mean roughness (Ra), and the difference between the highest peak and lowest valley (Rz) were measured as they are the most commonly used parameters when measuring surface finish.

2.4. Characterization of Printed Samples

Printing of test samples was carried out on the Prusa iMKS3+ printer. Printing parameters that were used are shown in **Table 2**. Printing was done on P-surface 141 by PPprint Gmbh sheet which was laid on the printer bed to prevent warpage and promote part adherence to the printing surface during printing process. The P-surface 141 sheet is a sheet that is used to prevent warping when printing with polypropylene but it was noted that it can also work with other

Table	2.	Printing	parameters.
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Parameters	Value		
Nozzle size	0.4 mm		
Build plate temperature	first layer: 70°C other layers: 30°C		
Layer thickness	0.2 mm		
Infill	100 %		
Fan cooling	10%		
Printing Temperature, Speed	0 wt% TPS: 220°C, 50 mm/s		
	10 wt% TPS: 220°C, 40 mm/s		
	20 wt% TPS: 215°C, 30 mm/s		
	30 wt% TPS: 215°C, 30 mm/s		
	40 wt% TPS: 210°C, 20 mm/s		

filaments that experience warping and poor adhesion when printing. Additionally, when printing with filaments that had 0 wt% and 10 wt% TPS content, brims had to be used to further prevent warping. Brims were not necessary when using filaments with higher TPS content.

Pasco Material Testing Machine ME-8236 was used to perform tensile test, compression test and 3-point bending test. The dimensions (in mm) of samples used for the tests are shown in **Figure 3**.

Rate of water absorption was also studied by immersing test sample ASTM D 570 (Figure 3(d)) in water at room temperature. The samples that had been submerged in water were subsequently dried to determine the quantity of mass reduction caused by TPS dissolving in the water. Equation (1) was used to calculate the percentage mass reduction (M_s).

 $M_{s}(\%) = (M_{o} - M_{i})/M_{o} \times 100\%$ (1)

where (M_i) is the initial mass of the sample and (M_o) is the residual mass.

3. Results and Discussion

3.1. Taguchi Analysis

The factors were heater temperature, extruder screw speed and spooling speed. They were all varied at 3 levels. Filament diameter was used as the response variable. L9 orthogonal array was used to reduce the number of experimental runs needed for the analysis. The desired response was 1.75 ± 0.05 mm and the "nominal is best" was selected as the condition for the analysis. From the results tabulated in **Table 3**, it can be seen that largest variation in diameter occurred when extruding composite with high TPS content. This could be due to partial clogging of the extrusion nozzle from time to time and greater resistance of flow of rHDPE due to the low extrusion temperatures used.

The main effects plots for the means of the filament diameter are shown in



Figure 3. (a) Tensile Test specimen, ASTM D638 Type IV; (b) Compression Test specimen, ASTM D695, (c) 3 point bending test specimen, ASTM D790; (d) Hydrolytic Degradation Test specimen, ASTM D 570.

Filament Composition	Run	Temperature (°C) H1; H2	Extrusion Speed (rpm)	Spooling Speed (rpm)	Avegrage Diameter (mm)	Standard Deviation (mm)
	1	220; 215	5	1.5	1.92	0.091
	2	220; 215	5.5	2	1.72	0.016
	3	220; 215	6	2.5	1.25	0.026
	4	215; 210	5	2	1.7	0.036
100% HDPE 0% TPS	5	215; 210	5.5	2.5	1.69	0.014
110	6	215; 210	6	1.5	1.93	0.079
	7	210; 200	5	2.5	1.43	0.065
	8	210; 200	5.5	1.5	1.59	0.049
	9	210; 200	6	2	1.81	0.070
	1	185; 180	4	1.5	1.86	0.139
	2	185; 180	4.5	2	1.64	0.048
	3	185; 180	5	2.5	1.56	0.241
	4	180; 175	4	2	1.61	0.117
	5	180; 175	4.5	2.5	1.7	0.151
	6	180; 175	5	1.5	1.66	0.036
90% HDPE 10%	7	175; 170	4	2.5	1.76	0.143
115	8	175; 170	4.5	1.5	1.79	0.161
	9	175; 170	5	2	1.54	0.098
	1	180; 175	5	1.5	1.97	0.227
	2	180; 175	5.5	2	1.86	0.021
	3	180; 175	6	2.5	1.69	0.136
	4	175; 170	5	2	1.99	0.015
	5	175; 170	5.5	2.5	1.57	0.068
	6	175; 170	6	1.5	1.92	0.060
80% HDPE 20%	7	170; 165	5	2.5	1.65	0.133
115	8	170; 165	5.5	1.5	1.77	0.132
	9	170; 165	6	2	1.88	0.016
	1	180; 175	4.5	1	1.81	0.227
	2	180; 175	5	1.5	1.8	0.042
70% HDPE 30% TPS	3	180; 175	5.5	2	1.82	0.066
11.5	4	175; 170	4.5	1.5	1.74	0.035
	5	175; 170	5	2	1.7	0.117

 Table 3. Experimental Results-Taguchi L9 orthogonal array.

Continued						
	6	175; 170	5.5	1	1.78	0.166
	7	170; 165	4.5	2	1.74	0.030
	8	170; 165	5	1	1.99	0.136
	9	170; 165	5.5	1.5	1.6	0.120
	1	170; 165	4.5	1	1.81	0.230
	2	170; 165	5	1.5	1.8	0.192
	3	170; 165	5.5	2	1.82	0.216
	4	165; 160	4.5	1.5	1.74	0.187
60% HDPE 40% TPS	5	165; 160	5	2	1.7	0.131
	6	165; 160	5.5	1	1.78	0.169
	7	160; 155	4.5	2	1.74	0.163
	8	160;155	5	1	1.99	0.140
	9	160; 155	5.5	1.5	1.6	0.137

Figure 4. The effect of each parameter to the diameter of the filament can be seen. Optimal parameters are obtained from these plots and the results were verified and tabulated in **Table 4**. The results obtained showed that the spooling speed was identified as the most crucial parameter in the extrusion process followed by extrusion speed. Extrusion temperature had the least impact on the filament diameter. This trend was the same for all variations of TPS content.

The response for means is shown in **Figure 5**. The response that had the least significant influence on filament diameter was temperature. These findings were similar to the ones reported by Adanje *et al.* [9] who conducted an optimization of extrusion process parameters on rHDPE using a different extruder. It is possible that for extrusion using rHDPE-TPS composites, extrusion temperature had the least influence because it was varied within a narrow range to avoid TPS burning. Spooling speed had the highest influence on filament diameter. The material flow rate of the composite was not very high even when high extrusion speed and high extrusion temperatures were used due to the melt flow index of the materials. This could be the reason why even the slightest change in the spooling speed or hauling speed drastically affected the diameter of the filament.

Optimum parameters were determined and verification was performed by extruding filaments using optimum parameters. The final optimum parameters are as tabulated in **Table 4**.

3.2. Filament Characterization

Diameter measurement and Surface roughness measurements were conducted on samples of filaments obtained through extrusion process using the optimized parameters.



Figure 4. Main effects plot for means for TPS HPDE ratios/mixtures. (a) 100%HDPE, 0% TPS; (b) 90% HDPE, 10% TPS; (c) 80% HDPE, 20% TPS; (d) 70% HDPE, 30% TPS; (e) 60% HDPE, 40% TPS.

1) Diameter consistency

Results from preliminary experiments showed that high-speed extrusion resulted formation of smoke due to burning of the TPS. Moreover, the recommendation from extruder manufacturers and previous researches is for low extrusion speed to be used in order to facilitate proper and uniform melting of materials which will consequently provide a consistent flow of material at the nozzle. Design of experiment was therefore done with low extrusion speeds to ensure high consistency in filament diameter. Diameter of the filament produced after optimization was fairly consistent and had a circular cross-section. Results of diameter measurements shown in **Table 5** indicate that diameter inconsistency was highest in filament that had 40 wt% TPS. There was little difference in the level of diameter variability of filaments that had 10–30 wt% TPS. The filament with the most consistent diameter had 0 wt% TPS. High level of diameter inconsistency as that exhibited by filament with 40 wt% TPS would result in poor print quality due to inconsistent flow of material at the infeed section of the printer.

2) Surface roughness

Results of filament surface roughness tests in Table 6 show that surface



Figure 5. Summary of delta values.

Table	4.	Verification	of results.
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TPS content (wt%)	Temperature (°C)	Extrusion speed (rpm)	Spooler speed (rpm)	Actual Average diameter (mm)
0	215,210	5	2	1.73
10	175,170	5	1.5	1.76
20	170,165	5.5	1.5	1.80
30	175,170	4.5	2	1.79
40	170,165	5.5	2	1.68

Table 5. Average filament diameter.

TPS	Diameter (mm)	Standard Deviation (mm)
0%	1.73	0.014
10%	1.76	0.051
20%	1.80	0.049
30%	1.79	0.053
40%	1.68	0.072

TPS in the composite (wt%)	Ra (µm)	Rz (µm)
0	1.24	8.43
10	1.31	10.45
20	2.43	13.39
30	3.02	18.54
40	4.51	29.10

 Table 6. Surface roughness test.

roughness increased with increase in TPS content. This could have been due to difference in the properties of material used. Recycled HDPE is hydrophobic and has a lower surface energy than that exhibited by TPS which is hydrophilic. This difference in surface energies can cause phase separation, which can lead to the formation of surface irregularities, such as bumps or protrusions. Chemical modification of the properties of these polymers using interfacial modifiers or further mixing of the composite by subjecting it to a second extrusion process could potentially improve the surface quality of the filaments. However, the filaments obtained in this research with surface roughness shown in **Table 6** did not significantly affect the quality of the finished print as the printing process involved a second heating and extrusion process and was carried out at low speeds.

3.3. Printed Sample Characterization

Test samples were successfully printed with recycled HDPE filament and composite filaments with TPS content of 10 wt%, 20 wt% and 30 wt%. Printing with composite filament that had 40 wt% TPS proved to be a challenge due to frequent clogging of the nozzle. The high coefficient of thermal expansion of HDPE led to extreme contraction and deformation when cooling. This coupled together with poor bed adhesion led to warping during the printing process. To improve adhesion of part to the printing surface, P-surface 141 print sheet was used. The printed samples in **Figure 6** show how the use of P-surface 141 print sheet successfully reduced the amount of warping. Additionally, brims were required to further promote bed adhesion and prevent warping when using filaments with 0 wt% and 20 wt% TPS. Filaments with a higher TPS content experienced less warping, as the rate of contraction during cooling was lower, and therefore did not require brims.



Figure 6. (a) Test sample printed without P-surface 141; (b) Test sample printed on P-surface 141

3.3.1. Investigation of Mechanical Properties

Mechanical properties of printed samples produced were evaluated using tensile test, compression test and 3-point bending test.

1) Tensile test

Tensile test of printed samples was conducted as per the ASTM D638 (Type IV) standard. Figure 7 shows a tensile test sample. Figure 8 shows the Ultimate Tensile Strength and the elongation at break of the printed samples. Increase in TPS content caused a decrease in ultimate tensile strength and ductility. This result is attributed to the lower tensile strength and ductility of TPS present in the composite. It is also possible that the plasticizer present in TPS permeated into rHDPE which further led to disruption of the crystalline structure of rHDPE. This would mean that the more the TPS content present, the larger the amount of plasticizer that rHDPE was subjected and thus the lower the tensile strength of the resulting composite.

The ultimate tensile strength obtained from the experiment was lower than the specification provided in the datasheet. This could be due to thermal degradation of the polymers as the process involved heating the polymers during filament fabrication process and during the printing process. Another possible cause is the manufacturing technique used. Tensile strength of parts produced through FFF is always lower than those produced using other techniques such as injection molding due to the layer-based structure of FFF printed parts. Moreover, inadequate layer bonding of the printed samples can further decrease the tensile strength of the parts. This problem can be addressed by using more suitable printing parameters such as increasing the nozzle temperature to promote proper melting and bonding between layers and reducing fan speed to prevent excessive cooling which can hinder bonding.



Figure 7. Tensile test sample printed from the rHDPE/TPS composite filament.



Figure 8. (a) Ultimate Tensile strength; (b) Elongation at break.

2) Compression tests

Compression test of printed samples were conducted as per the ASTM D695 standard. Test sample specimen shown in **Figure 9**. Compression test for brittle material is carried out up to point of fracture of specimen while test for ductile material is carried out up to appearance of first crack. Some materials however do not exhibit a defined yield point as deformation. This was the case with the printed samples that were tested as deformation in form of barreling continued throughout the loading process on the Pasco Material Testing Machine. In such a case, an arbitrary yield point is usually selected. Compression strength was therefore determined by measuring the loading force that resulted in 5% elongation. The results of the maximum compression strength are as shown in **Figure 10**. Increase in TPS content resulted in decrease in compressive strength. This is attributed to the lower compressive strength of TPS.

3) Bending tests

Bending test was conducted as per the ASTM D790 standard. A test sample for 3-point bending stress is shown in **Figure 11**. The results of the maximum stress for 3-point bending strength test are as shown in **Figure 12**. The bending or flexural strength reduced with increase in TPS content. The results of the test conducted on samples with low TPS content varied greatly. This was possibly due to inconsistent dimension as a result of bending and of the samples as can be seen in **Figure 11**, which introduced pre-existing stresses during testing.



Figure 9. 4 Samples of Compression test specimen printed from the rHDPE/TPS composite filaments.





3.3.2. Investigation of Water Absorption and Mass Loss

The aim of this test is to determine the amount and rate of water absorption by measuring sample weight before and after submerging it into water. It was however not possible to conclusively determine the amount of water absorbed by simply weighing the sample before and after submersion. This is because while absorption of water would lead to increase in weight, there would also be simultaneous weight reduction as TPS dissolves in water. For this reason, the test done focused on determining the weight lost due to loss of TPS. This was done by immersing the samples in room temperature water, then drying the samples for 24 hours at 100°C. These samples were then weighed. The results of % weight lost are as shown in **Figure 13**. The rate of weight loss increased with increase in TPS content.



Figure 11. Printed bending test sample.



Figure 12. Maximum Bending strength.





4. Conclusion

The study demonstrated the development of filaments using recycled HDPE and TPS blends. The temperature range could not be varied widely due to burning of TPS at temperature above 200°C and other challenges such as agglomeration. It was also clearly demonstrated that parts could be printed using FFF from the rHDPE-TPS filaments that were developed when P-surface 141 print sheet was used to prevent warping. It was also noted that increasing TPS content resulted in reduced warping. However, printing with filaments made from 40 wt% TPS presented great challenges due to frequent clogging of the nozzle. Results from the mechanical tests showed that increase in TPS resulted in reduced ductility and increased surface roughness. Increase in TPS content resulted in a linear decrease in tensile and compressive strength. This can be useful in predicting tensile strength of samples printed with various other ratios which are between 0 - 40 wt%. Decrease in bending strength with increase in TPS followed a less linear trend, Increase in TPS content also resulted in increase in rate of hydrolytic degradation. Possible areas of application of this plastic would be in single use products such as in tertiary packaging containers for powder detergents and cosmetics products.

5. Recommendation

Further work can be done to optimize printing parameters when using composites of TPS and rHDPE especially at low TPS content so as to reduce variation in dimensions, improve layer to layer adhesion and obtain consistent mechanical test results. Increasing the printer nozzle size to 0.6 mm could potentially minimize nozzle clogging and this approach could be utilized when working on composites with TPS content above 30 wt%. Chemical modification of the polymers such as interfacial modifier and other possible methods that can be used to improve homogeneity and consequently reduce the surface roughness of the filament can also be studied.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

[1] Liu, W., Zhou, J., Ma, Y., Wang, J. and Xu, J. (2017) Fabrication of PLA Filaments

and its Printable Performance. *IOP Conference Series: Materials Science and Engineering*, **275**, Article ID: 012033. <u>https://doi.org/10.1088/1757-899X/275/1/012033</u>

- [2] Dey, A., Eagle, I.N.R. and Yodo, N. (2021) A Review on Filament Materials for Fused Filament Fabrication. *Journal of Manufacturing and Materials Processing*, 2021, Article No. 69. <u>https://doi.org/10.3390/jmmp5030069</u>
- [3] Gourmelon, G. (2015) Global Plastic Production Rises, Recycling Lags. Vol. 22, Worldwatch Institute, Washington, D.C., 91-95.
- [4] Herianto, Atsani, S.I. and Mastrisiswad, H. (2020) Recycled Polypropylene Filament for 3D Printer: Extrusion Process Parameter Optimization. *IOP Conference Series: Materials Science and Engineering*, **722**, Article No. 012022. <u>https://doi.org/10.1088/1757-899X/722/1/012022</u>
- [5] Sotohou, F., Mwangi, J., Mutua, J. and Ronoh, E. (2023) Development and Evaluation of Recycled Polypropylene and Bean Pod Powder Composite Biomaterial for Fused Filament Fabrication. *Advances in Materials Physics and Chemistry*, 13, 31-48. <u>https://doi.org/10.4236/ampc.2023.133003</u>
- [6] Polline, M., Mutua, J., Mbuya, T. and Ernest, K. (2021) Recipe Development and Mechanical Characterization of Carbon Fibre Reinforced Recycled Polypropylene 3D Printing Filament. *Open Journal of Composite Materials*, **11**, 47-61. <u>https://doi.org/10.4236/ojcm.2021.113005</u>
- [7] Andanje, M.N., Mwangi, J.W., Mose, B.R. and Carrara, S. (2023) Biocompatible and Biodegradable 3D Printing from Bioplastics: A Review. *Polymers*, 15, Article No. 2355. <u>https://doi.org/10.3390/polym15102355</u>
- [8] Haq, R.H.R., Khairilhijra, K., Wahab, M.S., *et al.* (2017) PCL/PLA Polymer Composite Filament Fabrication using Full Factorial Design (DOE) for Fused Deposition Modelling. *Journal of Physics: Conference Series*, **914**, Article No. 012017. <u>https://doi.org/10.1088/1742-6596/914/1/012017</u>
- [9] Andanje, M.N., Mwangi, J.W., Mose, B.R. and Carrara, S. (2022) A Comparative Analysis of Additive Manufacturing Filaments Developed from Recycled High-Density Polyethylene and Recycled Polypropylene: Extrusion Process Optimization. *Proceedings of the* 2022 *Sustainable Research and Innovation Conference JKUAT Main Campus*, Kenya, 5-6 October 2022, 59-66.
- [10] Speight, J. (2019) Handbook of Industrial Hydrocarbon Processes. Gulf Professional Publishing, Houston.
- [11] Abbott, A.P., Abolibda, T.Z., Qu, W., Wise, W.R. and Wright, L.A. (2017) Thermoplastic Starch-Polyethylene Blends Homogenised Using Deep Eutectic Solvents. *Royal Society of Chemistry Advances*, 7, 7268-7573. https://doi.org/10.1039/C7RA00135E
- [12] Kormin, S., Kormin, F., and Beg, M.D.H. (2019) Effect of Plasticizer on Physical and Mechanical Properties of LDPE/SAGO Starch Blend. *Journal of Physics: Conference Series*, **1150**, Article No. 012032. <u>https://doi.org/10.1088/1742-6596/1150/1/012032</u>
- [13] Rodriguez-Gonzalez, F., Ramsay, B. and Favis, B. (2003) High Performance LDPE/ Thermoplastic Starch Blends: A Sustainable Alternative to Pure Polyethylene. *Polymer*, **44**, 1517-1526. <u>https://doi.org/10.1016/S0032-3861(02)00907-2</u>
- [14] Shijiazhuang Tuya Technology Co., Ltd. http://surl.li/gpsbl