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A comparative analysis of additive manufacturing filaments developed from recycled high-density polyethylene and recycled polypropylene: Extrusion process optimization

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[1]***Abstract*** — Plastic solid waste continue to present opportunities. The use of recycled plastic material in additive manufacturing, also known as 3D printing, is expected to make the process more sustainable and help to address the global problem of plastic waste. However, there are still limitations to using recycled plastics as filament material such as getting the right quality of filaments that are well characterized. There is growing interest among researchers on the development of filaments from recycled plastics towards the realization of a circular economy in the additive manufacturing technology.

In this paper, we present outcomes about filaments fabricated from recycled high density polyethylene and recycled polypropylene using the extrusion method. The extrusion process parameters considered in the fabrication of the filaments included extrusion temperature, screw speed and fan cooling. These parameters were analyzed and optimized using the Taguchi design of experiments technique. The response variable was the filament diameter which was desired to be 2.85 ± 0.05 mm with a circular cross section.

The fabricated filaments were characterized and compared to establish their sustainability for fused filament fabrication. The results from this study are very significant in the development of printable filaments that meet the standards for 3D printing.

***Keywords* —** Additive Manufacturing (AM), High Density Polyethylene (HDPE), Polypropylene (PP), Fused Filament Fabrication (FFF), Taguchi Design of Experiments, ANOVA.

I. INTRODUCTION

With the continual depletion of petroleum resource to produce plastic materials and the environmental pollution caused by plastic disposal, there's been a growing concern to develop sustainable industrial materials. Efforts have been made globally to deal with the increasing amounts of plastic waste. Recycling of plastic waste is one of the main efforts that has shown promising results in dealing with this menace [1].

Additive manufacturing (AM), or 3D printing, provides many benefits over the conventional manufacturing method, including flexible designing and production, rapid prototyping, cost effectiveness, waste reduction, and environmental friendliness [2]. Additive manufacturing materials have gained much attention as a way to address the demands of various industries, with plastics being regarded as an important material. Polylactic acid (PLA), acrylonitrile butadiene styrene (ABS), high-impact polystyrene and nylon are the most commonly used plastics in the fused filament fabrication (FFF), the most popular additive manufacturing technology applied in industry [3].

High-density polyethylene (HDPE) is among the most popular recyclable polymers which can be utilized as a feedstock for filament extrusion. Due to its excellent mechanical properties, flexibility, chemical stability and high strength-to-weight ratio, HDPE finds use in industrial, medical and biomedical applications [4]. However fused filament fabrication (FFF) of HDPE has proven difficult due to the material's significant shrinkage, voiding, and warpage issues, as well as its poor adhesion to printers build plates and extruded strands [5]. Proper identification of extrusion parameters during filament fabrication and optimization of printing process addresses the challenges of working with HDPE as a feedstock material.

Polypropylene (PP) has been extensively utilized in human life, including in electrical appliances, automotive components and home appliances. Just like HDPE, PP is highly recyclable and can be a potential FFF feedstock material. Herianto et al. [6] and Pickering et al. [7] researched on the feasibility of using PP as a feedstock material in FFF. Herianto et al. reported that optimization of extrusion parameters during filament development was key in ensuring successful production of filaments. Pickering et al. on the other hand reported the need to minimize shrinkage of the fabricated PP filaments to achieve good printability and desired mechanical properties of the printed parts.

In this study, we report on a successful approach to develop printable filaments from rHDPE and rPP using Taguchi optimization technique. The most relevant process parameters in the filament development process were identified as extrusion temperature, screw speed and fan speed. The

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developed filaments were characterized and printability of the materials tested.

II. MATERIALS AND METHODS

The materials used for the production of the 3D printing filaments were recycled high density polyethylene (rHDPE) and recycled polypropylene (rPP) pellets. The rHDPE pellets had a melt flow rate of 1.6 g/10 min and were yellow in color. The recycled polypropylene pellets had a melt flow rate of 21 g/10min and were white in color. The pellets were supplied by Mr. Green Africa. The melt flow rate is a significant thermoplastic property that enables the determination of the polymer's flowability at its melting point under a standard weight.

i. Filament Fabrication

a. b.

Fig. 1: a. Pellets of rHDPE and b. rPP

A Composer 450 filament making machine, by 3Devo B.V., was used to produce the filaments from the rPP and rHDPE pellets. This machine has a set of four heaters inside the barrel that heats the material after which the molten material is pushed through a nozzle by a rotating screw. Once the material exits the nozzle, it is cooled by a twin-fan arrangement. The cooled filament is then passed through an optical sensor system which measures the filament and a set of puller wheels rotate to adjust the thickness of the filament according to the desired set diameter.

Fig. 2: The Composer 450 Filament maker

Heater temperature, screw speed and fan cooling speed were identified as the three main processing parameters for the filament making process. Other parameters to be set included the spooling speed, when the machine was set on manual mode and size of the spooler wheels.

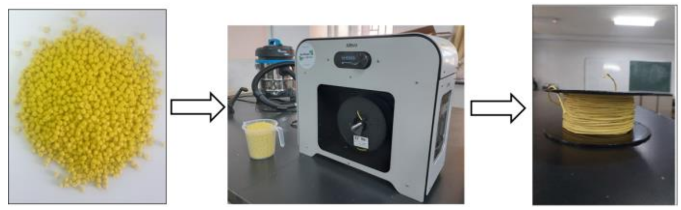


Fig. 3: Pellets of rHDPE fabricated to filaments

Figure 3 shows the pellets of rHDPE which are fed into the filament making machine. The pellets are melted, extruded and the cooled filament is spooled around a wheel.

ii. Experimental design and optimization

A design of experiment was conducted in Minitab 18 software using Taguchi technique [8] for the rPP and rHDPE. Tables 1 and 2 show the different levels of process parameters for rHDPE and rPP respectively that were identified for filament fabrication from the pellets based on literature and experimental data.

Table 1: Process parameters and respective levels for rHDPE

Process parameter	**Level 1**	**level 2**	**Level 3**
a. Heater temperature °C	180	200	220
b. Screw speed (rpm)	4	5	6
c. Fan speed (%)	50	60	70

Based on the parameters identified, the experiment was designed using L_9 (3^3) orthogonal array. The Taguchi design orthogonal array comprised of 3 factors with each factor at 3 levels. Therefore 9 experimental runs were conducted. The response variable used for this study was filament diameter of 2.85 ± 0.05 mm, which ideally should be constant throughout the spool.

Table 2: Process parameters and respective levels for rPP

Process parameter	**Level 1**	**level 2**	**Level 3**
a. Heater temperature °C	190	200	210
b. Screw speed (rpm)	2	4	6
c. Fan speed (%)	20	30	40

Inconsistencies in the filament diameter would lead to low material extrusion rate during printing in the case of a low diameter or an excess of the material in the case of a higher diameter which leads to nozzle clogging [6]. The resultant experimental designs for the two material are presented in Tables 3 and 4.

TABLE 3. : L_9 TAGUCHI EXPERIMENTAL DESIGN FOR RHDPE

Experiment No.	Heater Temperature (°C)	Screw speed (rpm)	Fan Cooling (%)
1	180	4	50
2	180	5	60
3	180	6	70
4	200	4	60
5	200	5	70
6	200	6	50
7	220	4	70
8	220	5	50
9	220	6	60

TABLE 4. : L_9 TAGUCHI EXPERIMENTAL DESIGN FOR RPP

Experiment No.	Heater Temperature (°C)	Screw speed (rpm)	Fan Cooling (%)
1	190	2	20
2	190	4	30
3	190	6	40
4	200	2	30
5	200	4	40
6	200	6	20
7	210	2	40
8	210	4	20
9	210	6	30

a. Characterization techniques

i. Filament diameter consistency

The filament diameter was measured automatically by the Composer 450 filament maker. The data was acquired automatically by connecting the machine to a laptop installed with the DevoVision application, an application which is specific to this filament maker that enables recording of the data logs of the experimental results. From the DevoVision application real-time data of the filament diameter, heater temperatures, extruder speed in RPM, puller speed, current, filament length and other settings could be displayed. The filament diameter was measured every second when the machine status was at the 'running mode' meaning all four heaters had attained their set temperatures. Measurement was done for five minutes and the average diameter recorded.

ii. Morphology

The surface morphology of the filaments developed was investigated using an Olympus Optical Microscope SC50 by Olympus Corporation. The internal and external structures of the filaments were studied. Preparation of specimen for internal structure analysis entailed cutting across the cross-sections of the filaments with a sharp razor, then sticking the specimen on some plasticine for proper attachment. This was then mounted on the stage of the microscope. For external structure analysis, the specimen was cut to about 3mm length then this was mounted on the stage along the length using plasticine. The specimen was then placed under the microscope for observation.

iii. Surface Roughness, Ra (µm)

The surface roughness, Ra, was determined using the Surface Roughness Tester SRG – 4000 according to ISO 4287:1997.

The surface roughness, Ra, was determined for both the rHDPE and rPP. The Surface Roughness Tester SRG – 4000 has a probe which moves along the length of the filament and determines the Ra value. This testing equipment is able to give values with a sensitivity of up to 0.001 µm.

iv. Tensile testing

Tensile testing was carried out on the developed filaments using the Universal Testing Machine (UTM) Shimadzu UMH – 30. The testing was done according to the ISO 6892-1:2016 with the gauge length set at 80 mm. The strain rate for determination of upper yield strength was 0.00025 s^{-1} with a relative tolerance of ± 20 %.

The clamping of the filament for loading on the UTM proved to be a challenge. Two options were explored to deal with this challenge. The first option was drilling a hole of 3mm in mild steel tensile specimen, inserting 80 mm of the filament end in the drilled hole and then bonding the metal to the filament using super glue. This option was unsuccessful as the filament kept slipping under loading conditions. The glue was not strong enough to hold the filament to the metal gripper.

The second option was using a serrated gripper to hold the filament during loading as opposed to the super glue bonding on the mild steel. The use of a mechanical file proved to be effective in forming a gripping end for the filament on the UTM.

v. Printability of filament

To investigate the printable performance of the developed filaments, the Fused Filament Fabrication (FFF) was carried out on the Ultimaker S3 printer. The ASTM D638 Tensile Specimen, Type I, was designed as shown in Fig. 4 and its printability tested.

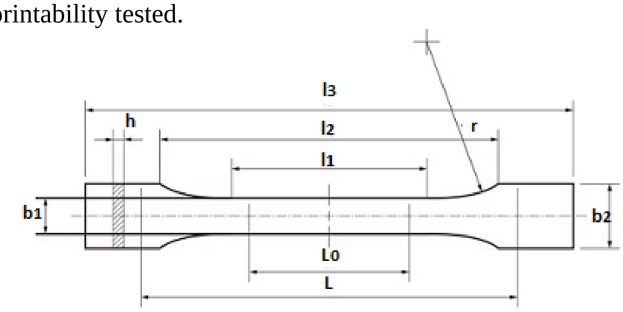


Fig. 4: ASTM D638 Tensile Specimen, Type 1

The sizes of the specimen were as shown in Table 5.

TABLE 5. SPECIFICATIONS OF THE TENSILE SPECIMEN

Parameter	**Value**
Full length, l_3	165 mm
Length of narrow section, l_1	57 mm
Gauge length, L_0	50 mm
Width of narrow section, b_1	13 mm
Thickness, *h*	3.5 mm
Overall width, b_2	19 mm
Distance between grips, *L*	115 mm
Radius of fillet, *r*	76 mm

The G-Codes required for the printing were generated from the Ultimaker Cura 4.9 software. The typical printing parameters are shown in Table 6.

TABLE 6. : PRINTING PARAMETERS OF RHDPE BY FFF.

Parameters	**0.4mm nozzle**
nozzle temperature	260 °C
build plate temperature	60 °C
layer thickness	0.27 mm
line width	0.2 mm
filling degree	100 %
material flow	100 %
filling pattern	triangles
printing speed cooling speed	25 mm / s 40%

III. Results and Discussions

i. Taguchi optimization of extrusion parameters

The filament diameter was the variable response in this study. A Taguchi analysis was conducted to study the response at different levels of the selected parameters based on the experimental design. Since a response of 2.85 ± 0.05 mm was desired, the "nominal is best" was the preferred condition for analysis. A response table for means was used to analyze the average response at different parameter levels.

TABLE 7: RESPONSE TABLE FOR MEANS OF RHDPE

Level	**Heater Temperature**	**Screw speed**	**Fan cooling**
1	2.73	2.74	2.76
2	2.76	2.71	2.70
3	2.75	2.79	2.77
Delta	0.03	0.08	0.07
Rank	3	1	2

It is observed in Table 7 that the delta value (the difference between the highest and lowest average response) was highest in screw speed and lowest in the heater temperature. This therefore ranked screw speed as the parameter with the greatest effect on the filament diameter, followed by fan cooling and lastly the heater temperature for the case of rHDPE.

Table 8 shows the response table for means of rPP where it is observed that the heater temperature has the highest rank, followed by screw speed and lastly fan cooling according to the L_9 orthogonal experimental design.

TABLE 8: RESPONSE TABLE FOR MEANS OF rPP

Level	**Heater Temperature**	**Screw speed**	**Fan cooling**
1	2.72	1.52	2.08
2	1.73	2.44	2.09
3	1.25	1.73	1.53
Delta	1.47	0.92	0.56
Rank	1	2	3

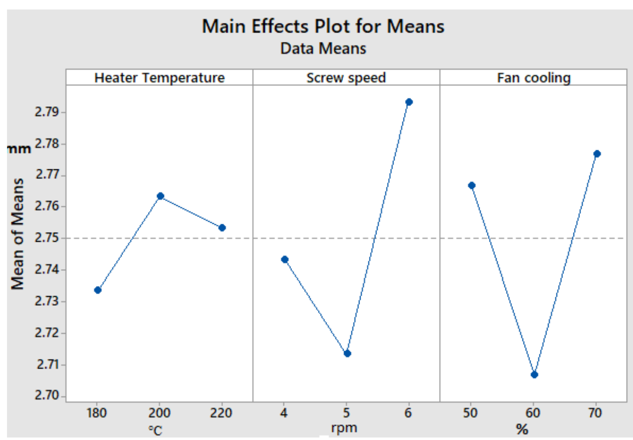


FIG. 5: MAIN EFFECTS PLOT FOR MEANS FOR rHDPE

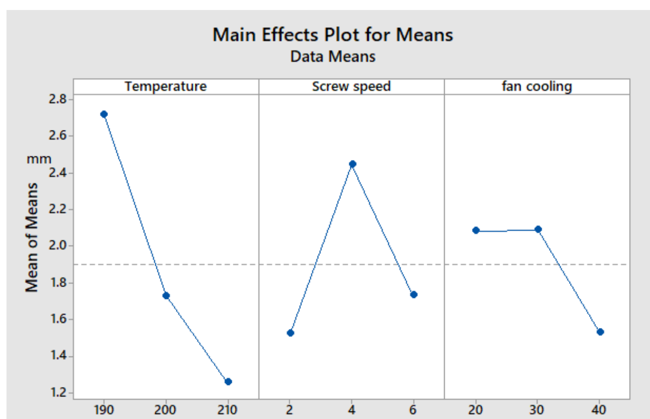


FIG. 6: MAIN EFFECTS PLOT FOR MEANS FOR rPP

Figures 5 and 6 show the main effects plots for the means of the filament diameter where the effect of each parameter on the response is observed. From these plots the optimal process parameters for the filament making process were obtained.

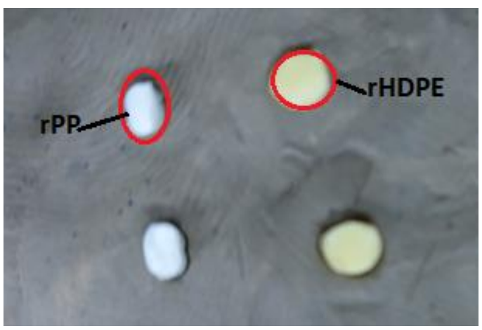
TABLE 9: OPTIMAL PROCESS PARAMETERS FOR rHDPE

Heater Temperature	**Screw speed**	**Fan cooling**	**Mean diameter**
200 °C	6 rpm	70%	2.83

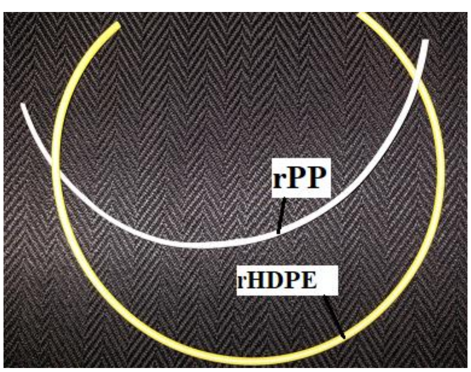
TABLE 10: OPTIMAL PROCESS PARAMETERS FOR rPP

Heater Temperature	**Screw speed**	**Fan cooling**	**Mean diameter**
190 °C	4 rpm	40%	2.52

Taguchi optimization technique is able to give a prediction of the response based on the optimal parameters established. The predicted results for rHDPE was 2.83 mm whereas that of rPP was 2.52 mm. It is important to note that rHDPE was able to give a near perfect circular cross section whereas achieving a circular cross-section with rPP was a challenge. This is shown in Fig. 7 below.



(a)



(b)

Fig. 7: Photo showing the sections of the developed filaments (a) cross-section, (b) filaments side by side

ii. Filament consistency

One of the main goals in filament fabrication is to achieve filament consistency. Variation in filament diameter is a great challenge to be overcome when extruding filament for FFF. If this variation is too large, it may affect the quality of the print since the material input may not be consistent enough or the intake mechanism may have problems.

The filament diameter consistency was observed from the Filament Maker DevoVision application. A lot of inconsistencies was observed during filament fabrication of rPP compared to rHDPE. This could be attributed to the high melt flow rate of rPP (21g/10 min) which causes higher flow of material compared to that of rHDPE (1.6g/min). A high melt flow rate depicts a low viscosity hence high output rate from the extruder that would be difficult for the puller mechanism to adjust to the desired diameter, hence the inconsistency in rPP.

Furthermore, PP has a high degree of polymer chain branching making it to be highly tactile compared to HDPE. This increases its flow properties which have to be well monitored during filament fabrication [9].

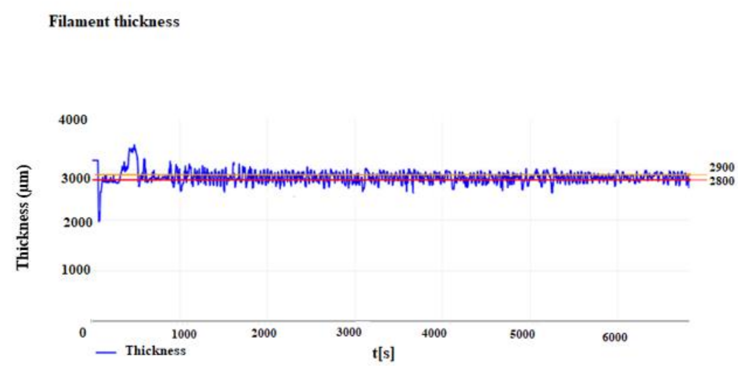


Fig. 8: Filament diameter consistency of developed rHDPE

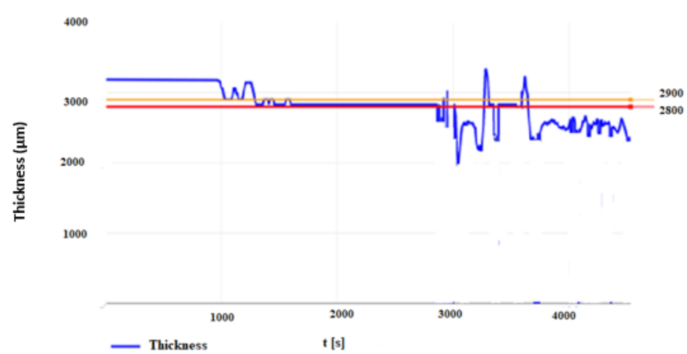


Fig. 9: Filament diameter consistency of developed rPP

iii. Morphology analysis

Figures 10 and 11 show the internal structures of the rHDPE and rPP under the optical microscope where it was observed that the rPP has presence of air bubbles whereas rHDPE has a smooth internal surface. This difference could be attributed to the humidity levels of the pellets as provided in the certificates of analysis which showed that the humidity level of rHDPE was 0.05% whereas that of rPP was 0.14%. During the elevated extrusion temperatures, presence of moisture in the material led to water vapor pockets in the material on evaporation of the moisture.

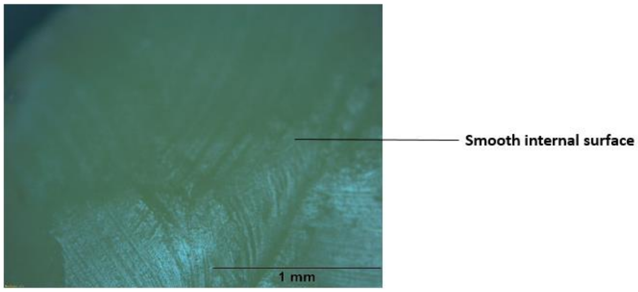


Fig. 10: Internal structure of rHDPE under the optical microscope

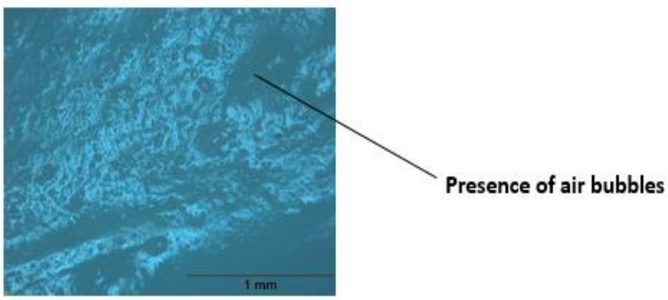


Fig. 11: Internal structure of rPP under the optical microscope

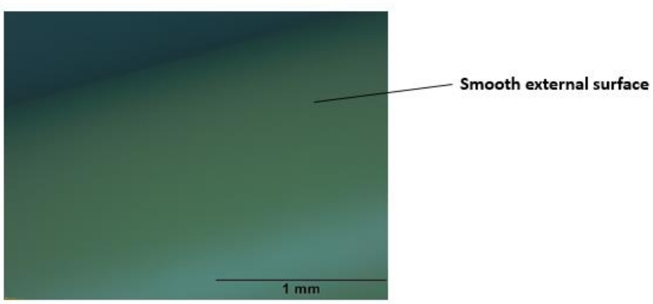


Fig. 12: External surface of rHDPE under the optical microscope

Similarly, the external surface of rPP, Fig.13, was observed to have air bubbles owing to the evaporation of moisture present in the material. Though HDPE and PP are reported to be hydrophobic materials, prior drying of the materials in an oven would have helped eliminate the high moisture content in rPP.

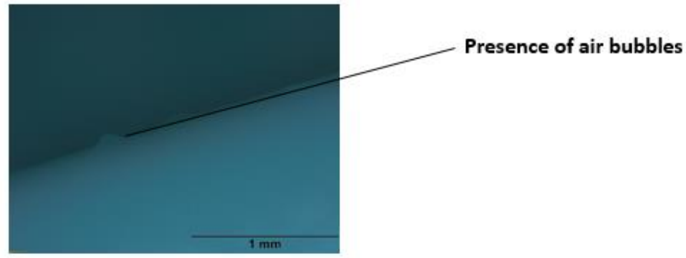


Fig. 13: External surface of rPP under the optical microscope

iv. Surface Roughness, Ra (μm)

The variation of a surface topography from an ideal level is described by various parameters, one of which is the arithmetic mean surface roughness, Ra, which is established according to the ISO 4287:1997 When evaluating the characteristics of the finished product as well as during production, the surface roughness is considered as having a significant impact on the product's functionality and reliability [10].

The average values obtained for surface roughness were as shown in Table 11. It was observed that rHDPE had a lower surface roughness of 1.3008 μm than rPP which had 2.337 μm. The high surface roughness affects the printability of the filaments since the inconsistencies and porosities cause feeding issues on the 3D printer nozzle.

TABLE 11: VALUES OF SURFACE ROUGHNESS

Experimental run	rHDPE	rPP
R1	1.228 μm	2.807 μm
R2	1.072 μm	1.819 μm
R3	1.337 μm	2.120 μm
R4	1.566 μm	2.602 μm
Average surface roughness, Ra	1.3008 μm	2.337 μm

v. Mechanical testing

From the Universal Testing Machine (UTM) Shimadzu UMH – 30, maximum loading force and engineering stress were determined.

TABLE 12: COMPARATIVE VALUES OF TENSILE STRENGTH AND STRAIN FOR rHDPE AND rPP

Filament	Maximum load, P	Original cross-section area, A_0	Tensile strength, $\sigma = P/A_0$
rHDPE	196.2 N	6.38×10^{-6} m^2	30.75 MPa
rPP	49.05 N	4.99×10^{-6} m^2	9.83 MPa

From table 12, it can be seen that rHDPE has high tensile strength than rPP. This high strength makes HDPE a preferred industrial material where high strength relative to weight is desired. The tensile strength of 30.75 MPa for rHDPE was within the range of commercial HDPE [11].

vi. Printability of the filaments

The FFF is considered the best additive manufacturing process for printing rHDPE although it is not an easy material to print with [5]. The two main issues encountered during printing of rHDPE was difficulty in adhering to the build plate of the 3D printer and warping of the printed part during printing, as can be seen in Fig. 14. The primary cause of this, is the crystallization the rHDPE, which is responsible for significant thermal shrinkage and warpage after cooling of the melt [12].

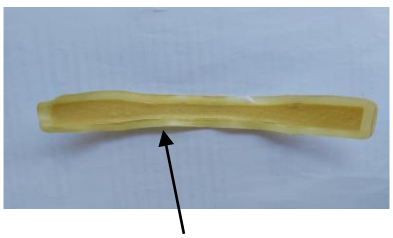


Fig. 14: Printed specimen

To address the issue of adhesion on the build plate, use was made of carton packing tape. The carton packing tape was spread on the build plate and by heating the build plate to 60 °C the rHDPE was able to adhere on the build plate. To counter the warping during printing, a brim of 5mm was included in the print parameters.

Some of the recommendations given to deal with the issues of adhesion and warping include:

- Use of packing tape on the build plate since HDPE adheres well to polypropylene [13]. This was one of the modifications done on the build plate.
- Use of a heated build plate thus keeping the material stable and warm during the printing process as it provides more adhesion to the part's foundation. For this study, a build plate temperature 60 °C proved to be sufficient as guided by [5].
- HDPE has a high coefficient of thermal expansion meaning its rate of shrinkage is approximately twice that of PLA/PETG on cooling [14]. Slow cooling during printing is one of the recommendations to get around this. A cooling speed of 40% proved sufficient in this study.
- Another recommendation is to include a wide brim around the printed part so that the brim is able to hold the part down when it starts warping. This was implemented in this study whereby a brim of 5mm was included in the part.
- Other recommendations to counteract warping include increasing the infill density, increase the flow per subsequent layer and incorporation of organic fillers into the HDPE matrix.
- For better morphological analysis, a scanning electron microscope (SEM) is recommended.

Due to the diameter inconsistences with rPP, printability of this filament was not tested. To further improve on the printability of rHDPE the printing parameters should be optimized to counteract the warping issues that make this material difficult to print and widen the selection of printable materials.

IV. CONCLUSION

The optimal process parameters for fabrication of AM printable filaments from rHDPE and rPP were identified through the Taguchi DOE L_9 Orthogonal array. Various characterization techniques were conducted to determine the developed filaments' printability using the FFF AM process. The study evaluated the feasibility of using rHDPE and rPP, common plastic materials, as feedstock for additive manufacturing. High filament consistency and low melt flow rate of rHDPE confirmed it to be a better material for AM than rPP due to smooth flow of the extrudate from the nozzles. Issues such as poor adhesion on the built platform and material warpage remain to be a hindrance on the adoption of HDPE as a possible 3D printing filament. Optimization of printing process parameters, inclusion of organic fillers in the matrix of HDPE, and modification of the printer's build plate have been suggested as possible solutions to these issues.

V. ACKNOWLEDGEMENT

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