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Development of additive manufacturing biofilaments from recycled high-density polyethylene and rice husks: optimization of extrusion parameters

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Abstract

The success of additive manufacturing technology continues to depend heavily on new material development. Recycled bioplastic materials have been found to offer an alternative choice with promising results due to their economic and environmental benefits. In fused filament fabrication technology, the unsatisfactory performance including poor adhesion between layers for many bioplastic materials derived from recycled plastics has, however, made researchers continue exploring new ways of enhancing their performance. This study presents findings on the fabrication and optimization of extrusion parameters of biofilaments from recycled high-density polyethylene and rice husks, an agricultural by-product. The process parameters identified in the filament fabrication were heater temperatures, screw speed, and the rate of fan cooling of the filament-making machine, the Composer 450, by 3Devo. Using the Taguchi design of experiment and analysis, the optimal process parameters identified for the filament fabrication were heater temperature of 220°C, screw speed of 10 rpm, and fan cooling of 30% to produce a filament of 1.63 mm thickness with a circular cross-section.

Keywords: Additive manufacturing, Fused Filament Fabrication (FFF), Taguchi method, Design of experiment.

1.0 Introduction

Plastics are the most widely used and adaptable material in the world today, due to their performance and superior characteristics compared to other materials such as wood and metals. These characteristics include chemical resistance, dimensional stability, lightweight, flexibility, and noise-proof capabilities (Agumba et al., 2023; Gall et al., 2020; Mohanty et al., 2002; Sardon and Dove, 2018; Zhu et al., 2021). The material is employed in many aspects of daily life, including

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consumer goods, packaging, transportation, construction, and electronics. Due to the increased demand for the material, plastic manufacturing and consumption worldwide is on the rise leading to a depletion of petroleum resources from which it is derived (Herianto et al., 2020). However, most plastic applications are single-use, after which the item is disposed of, leading to environmental pollution (Mwanzia et al., 2023). As such, a lot of research and development in the recycling and recovery of plastic is taking place worldwide as one of the promising solutions in dealing with the plastic menace and towards achieving a circular economy (Al-Maaded et al., 2012; Andanje N. Maurine et al., 2022; Garcia and Robertson, 2017; Milios et al., 2018; Shen and Worrell, 2014; Singh et al., 2017).

High-density Polyethylene (HDPE), is one of the most commonly used plastics finding applications in plastic bottles, toys, chemical containers, and piping systems. Its popularity as a raw material is due to properties such as high stiffness, high strength-to-weight ratio, chemical resistance, durability, and affordability. This makes HDPE among the highly recyclable plastics (Mungathia et al., 2021.). The plastic, however, requires a lot of space for landfills, is not biodegradable, and depletes natural resources such as fossil fuel (Sangwan and Bhakar, 2017). In terms of processing HDPE, conventional machining technologies such as extrusion and molding processes have been dominant. The development of advanced manufacturing technologies such as additive manufacturing brings a new level of ease to polymer processing, in terms of sustainable lightweight construction and the creation of intricate multifunctional material systems.

In Additive Manufacturing (AM), also referred to as three-dimensional (3D) printing, digitally created virtual 3D objects are turned into actual 3D items using digital slicing where layers of the printing material are deposited on top of one another (Gardan, 2016). The products are constructed in a single step, as opposed to subtractive or formative production, and without the need for molds or machining. Fused filament fabrication (FFF), also known as fused deposition modeling (FDM), is the most widely used additive manufacturing (AM) technology, being employed in the industry (Dhinakaran et al., 2020). Commonly used polymers in FFF include acrylonitrile-butadiene-styrene (ABS), polyethylene terephthalate (PET), polylactic acid (PLA), thermoplastic polyurethane (TPU), poly-ether-ketone (PEEK) and polycarbonate (PC). HDPE, though very popular, has not been widely explored in FFF due to challenges such as difficulty in adhering to the build plate during printing, as well as shrinkage of the material upon solidification, particularly when it crystallizes (Schirmeister et al., 2019).

As a result of the unsatisfactory performance of the polymer materials currently used, the majority of additively manufactured polymer items are still employed as conceptual models rather than functional components owing to the lack of strength, inability to bear loads as well as lack of industrially approved standards. These shortcomings constrain the widespread industrial application of additively manufactured polymers. To improve the performance of AM materials,

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various studies on combining plastics with various reinforcements have been conducted (Haibo et al., 2019; Le Duigou et al., 2016; Nguyen et al., 2018; Sasse et al., 2022; Wang et al., 2017).

Additive manufacturing using polymer composites produces parts that have enhanced structural or functional features that cannot be attained by unreinforced polymers. The creation of polymer matrix composites, which have great mechanical performance and outstanding functionality is made possible by the incorporation of reinforcements made of particles, fibers, or nanomaterials (Wang et al., 2017). This study incorporates rice husk microparticles into the matrix of recycled HDPE to produce biocomposite filaments for AM. The heater temperature, screw speed, and fan speed which are the main extrusion parameters in the filament fabrication process are optimized using Taguchi design of experiments.

2.0 Materials and methods

The materials used for the production of the 3D printing biofilaments were recycled high-density polyethylene (rHDPE) pellets and rice husks (RH). The pellets were supplied by Mr. Green Africa, a Kenyan company that supplies various recycled plastics to plastic manufacturers. The rHDPE pellets had an average diameter of 4.46 mm and a melt flow rate of 1.6 g/10 min (with 5 kg weight at 190 ℃). The melting temperature was between 120 – 150 ℃. The melting enthalpy was 176 J/g, and the tensile modulus was 715 MPa. The rice husks, a by-product from rice milling were supplied by Mwea Rice Mills, Kenya.

2.1 Preparation of the material

The recycled HDPE was dried in an oven for 24 hrs. at a set temperature of 80 °C to achieve a humidity level of less than 0.05 % as per the recommendation in the material data sheet. The rice husks were dried for 2 hrs. at a temperature of 60 °C. The rice husks were milled first using the hammer mill and then further milled in a ball mill to get finer particle sizes. The milled rice husks were then classified using the sieving machine to achieve a particle size of 150 microns. The mixing ratio of recycled HDPE to rice husks was 9:1. This is a ratio that has been considered by other researchers such as (Morales et al., 2021).

2.2 Filament Development

A filament-making machine, the Composer 450, by 3Devo B.V., was used to produce filaments from the recycled HDPE pellets and rice husks mixture. The specifications of the Composer 450 are listed in Table 1. The filament-making machine has an optical sensor and puller wheel mechanism for consistent diameter control and a live data monitoring system.

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Table 1: Specifications of the Composer 450

Parameter	Value
Temperature capacity	Up to 450 °C
Filament diameter	0.5 – 3.0 mm
Screw rotation speed	2 – 15 rpm
Fan cooling	0 – 100 %
Nozzle extruder diameter	4 mm
Voltage	110 – 230 V
Frequency	50 – 60 Hz
Maximum power consumption	1300 W
Heating zones	4

2.3 Taguchi design of experiment

Using the Taguchi approach, an experimental design was carried out in the Minitab 18 program. The three key processing variables for the filament-making process were identified to be heater temperature, screw speed, and fan speed. These are the parameters that were used to optimize the extrusion process.

Table 2: Process parameters and respective levels

Process parameter	Level 1	level 2	Level 3
a. Heater temperature °C	210	220	230
b. Screw speed (rpm)	8	10	12
c. Fan cooling (%)	20	30	40

This level range was informed by preliminary experiments done to identify the range of each process parameter. For heater temperatures below 210 °C, unmelted polymer particles were inherent in the filament from visual inspection. Temperatures above 230 °C resulted in the burning of the polymer as some fumes could be seen being released from the filament maker. A screw speed below 8 rpm was too slow in expelling the extruded filament whereas a screw speed of above 12 rpm was too fast to allow for spooling of the extruded filament. A fan cooling of less than 20 % was insufficient in cooling the filament as it got squished at the puller wheels whereas a fan cooling above 40% made the filament too stiff to be adjusted by the puller wheel mechanism. The L_9 (3^3) orthogonal array was chosen for this experimental design since three factors were controlled at 3 different levels, Table 2. Therefore, nine experimental runs were conducted with parameters set as shown in Table 3 for the different experimental runs.

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Table 3: Design of experiment for 90% rHDPE + 10% RH biocomposite

Experiment No.	Heater Temperature (°C)	Screw speed (rpm)	Fan Cooling (%)
1	210	8	20
2	210	10	30
3	210	12	40
4	220	8	30
5	220	10	40
6	220	12	20
7	230	8	40
8	230	10	20
9	230	12	30

From the optimal parameters identified, validity testing was performed experimentally.

2.4 Fused filament fabrication of the developed biofilament

Fused filament fabrication (FFF) was carried out on the Pruser i3 MK3S+. The Pruser Slicer 2.5.0 was used to generate the G-codes for 3D printing from CAD test samples. The test sample used was the tensile test specimen according to ASTM D638 type IV. Unlike other common polymers used in FFF, HDPE experiences poor adhesion on the build plate of the printers. To ensure that there was good adhesion on the steel build plate, a slight modification had to be done whereby a print plate made from polypropylene was mounted. To deal with the challenge of warping, an outer brim was included during printing. With this modification, good adhesion of the extruded material was achieved on the print bed. The parameters identified for printing the developed biocomposite material are shown in Table 4.

Table 4: Printing parameters for 90% rHDPE + 10% RH biocomposite

Parameter	Value
Nozzle diameter	0.4 mm
Nozzle temperature	230 °C
build plate temperature 1st layer	70 °C
build plate temperature	65 °C
layer thickness	0.2 mm

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filling degree density	100 %
filling pattern	Rectilinear
Printing speed	45 mm/s
Fan speed	100 %

3.0 Results and discussions

3.1 Taguchi analysis

The desired response in this study was the filament diameter of 1.75 ±0.05 mm. The filament-maker could display the diameter of the produced filament as measured by the optical sensor at every second during the extrusion process. The values were then stored in a log when the filament maker was connected to a laptop. For each experimental run, the average diameter of the extruded filament was determined for an extrusion period of 10 minutes from the stored data logs.

Table 5: Experimental results of the average filament diameter for 90% rHDPE + 10% RH biocomposite

Experiment No.	Heater Temperature (°C)	Screw speed (rpm)	Fan Cooling (%)	Average filament diameter
1	210	8	20	1.59
2	210	10	30	0.98
3	210	12	40	1.53
4	220	8	30	1.60
5	220	10	40	1.71
6	220	12	20	1.92
7	230	8	40	2.19
8	230	10	20	2.56
9	230	12	30	2.71

Table 5 shows the experimental results of the produced filament diameter from the nine experimental runs that were conducted. The "nominal is best" condition in Taguchi was the one recommended for this analysis since a desired response of 1.75 ± 0.05 mm was sought. The average response at the different parameter levels was examined using a response table for means shown in Table 6. It was also observed that the delta value, which is the difference between the largest and the smallest average response, was highest in heater temperature followed by the screw speed and lastly the fan cooling. This depicted the ranks of the different parameters. It therefore implies that the heater temperature was the parameter with the most effect on the

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response value during the filament extrusion process and the parameter with the least effect was the fan cooling.

Table 6: Response Table for Means of the developed biocomposite

Level	Heater Temperature	Screw speed	Fan cooling
1	1.37	1.79	2.02
2	1.74	1.75	1.76
3	2.49	2.05	1.81
Delta	1.12	0.30	0.26
Rank	1	2	3

For polymer filaments, the heater temperature is the most significant parameter since it determines the fluidity of the material. For the plastic pellets to be extruded into filaments, they must be heated above their melting temperature. This continually softens the material until it's melted and reshaped into filaments. Thermoplastics, such as HDPE, soften when heated and harden when cooled. As a result, they can be recovered from waste, reshaped, and reused through the recycling process.

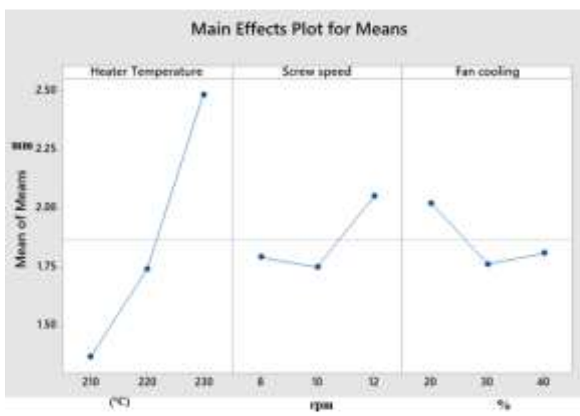

Figure 1: Main Effects Plot for Means for the biofilament

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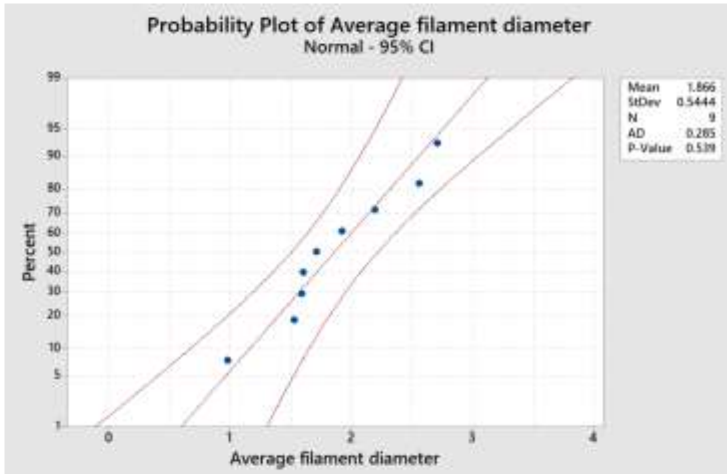

Figure 2: Probability plot of the average filament diameter

The screw speed, the second important parameter, determines the cylindricity of the extruded filament. When the speed of the fluid polymer is very high it distorts the cylindricity of the filament due to high kinetic energy present in the fluid. This fluid polymer velocity is a function of kinetic energy. Very low speed might be undesirable as it would take the fluid very long to exit the extruder nozzle. Therefore a trade-off had to be done, taking into consideration the other two parameters. The last parameter affecting the filament extrusion was fan cooling. This was so because it was the parameter that interacted with the extruded filament at the nozzle point. At the exit point, crystallization starts taking place where the molecules in the material begin to align themselves in an orderly manner. If the cooling is very rapid, the alignment of the molecules would be distorted due to limited time.

To determine the optimal parameters of the extrusion process, Taguchi's design of experiments analyzed the experimental design and the respective responses. For this experiment, the main effects plot for means shown in Figure 1 was used to give the optimal process parameters. The values of the process parameters at the point where the desired response was 1.75 ± 0.05 mm, were the optimal process parameters. These are summarized in Table 7. The average filament diameter from the experimental runs was 1.866 mm, with a standard deviation, s, of 0.544 mm and a confidence level of 95%, Figure 2. This confidence level indicates reliability in the results obtained. The average filament diameter of 1.866 mm has a difference of 0.11 from the desired value. Since the filament printing range is 1.7 mm – 1.8 mm, the produced filament would achieve good printability. This was confirmed during testing of printability as explained in section C.

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Table 7: Optimal settings

Temperature	Screw speed	Fan cooling
220 ° C	10 rpm	30 %

Using the 'nominal is best' condition, the signal-to-noise (S/N) ratio was determined according to the formula:

$$\frac{S}{N} = -10 \times \log s^2$$

The signal-to-noise was therefore found to be:

$$\frac{S}{N} = -10 \times \log 0.544^2$$

$$\frac{S}{N} = 5.29$$

The signal-to-noise ratio of 5.29 indicates that the response is five times greater than the noise signal. This shows that the experimental design was well conducted and that the parameter levels worked well for the filament extrusion process.

Using the optimal process parameters, Taguchi's design of experiments predicted that the filament diameter would be 1.71 mm which was within the desired value of 1.75 ± 0.05 mm of the filament.

3.2 Validity testing

A validity test was performed using the identified optimal parameters in the filament extrusion process. The average filament diameter was determined experimentally to be 1.63 mm. From the Taguchi predicted average filament diameter of 1.71 mm, the validity test results had an error of 4.69 %. The test shows an accuracy rate of 95.31 % which can be further improved by improving the signal-to-noise ratio.

3.3. Additive manufacturing of the developed biofilament

The modification of the printing plate from a steel sheet to a polypropylene print plate made it possible for the adhesion of the developed filament. From the thermal analysis that was conducted using the Mettler Toledo differential scanning calorimetry, the addition of the rice husk filler resulted in a reduction of crystallinity from 54.91% of pure rHDPE to 45.98% which reduced the warpage of the printed part. A reduction in crystallinity implies a reduction in the regions

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where the polymer chains are well aligned, hence the reduction in warpage. Crystallinity is one of the factors that affect warpage in thermoplastic polymers.
The inclusion of an outer brim of 5 mm wide around the part improves the adhesion of the material to the print bed further reducing the warpage as seen in Figure 3.

Figure 3: Printed part with an outer brim

Figure 4: Image showing the difference in the prints when a brim is included

Figure 4 shows the difference in the prints when an outer brim was included to the part printed without an outer brim. The inclusion of a brim is seen to reduce the warping of the print.

4.0 Conclusions

The optimal process parameters for fabrication of recycled rHDPE and milled rice husks biocomposite were identified through the Taguchi DOE L_9 Orthogonal array. These were determined to be heater temperature of 220 ° C, screw speed of 10 rpm, and a fan cooling of 30 %. From the three parameters, it was established that heater temperature had the greatest effect

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on the filament diameter followed by screw speed and lastly the fan cooling. The predicted result from the Taguchi analysis was 1.71 mm. This was further validated experimentally to give an average filament diameter of 1.63 mm indicating an error of 4.69 %. The printability of the developed filament was tested using the FFF technique. Some modifications had to be done on the steel plate by fixing a polypropylene print surface to improve the adhesion. The inclusion of an outer brim was done to reduce the warping of the material. Further work should focus on improving the warping of the biocomposite material to match that of commercial filaments like PLA. Other mixing ratios of the recycled HDPE to the milled rice husks should be considered and the results compared.

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