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A Comparative Study in Fused Filament Fabrication Using Commercial and Recycled High-Density Polyethylene

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ABSTRACT: The recycled high-density polyethylene (rHDPE) has the potential to be used as a sustainable printing material for fused filament fabrication (FFF). The main challenges producing 3D printable filament from recycled plastic include maintaining a consistent filament-like diameter and ovality, ensuring the printability of the material and achieving adequate mechanical strength in the printed part. This study compares the printability, filament geometry, melt flow index (MFI) and tensile properties of printed specimens using rHDPE and commercial HDPE filaments. The research successfully produced 3D printing filament from rHDPE at 220°C, which was compatible with FFF printing. However, rHDPE filament exhibited significantly higher diameter deviations and filament ovality compared to commercial HDPE filament. During printing, both rHDPE and commercial HDPE specimens experienced warping at the bottom layers, a common challenge in 3D printing high-crystalline thermoplastic materials. rHDPE had a lower MFI value than commercial HDPE, allowing it to be printed at a lower temperature. For optimal tensile strength and modulus, both materials performed best when printed at 230°C. However, printed specimens made from rHDPE showed lower tensile strength and modulus than those made from commercial HDPE filament. This study highlights the feasibility of using rHDPE as a feedstock for FFF printing while addressing the challenges related to filament geometry, warpage and mechanical properties.

Keywords: recycled HDPE, commercial HDPE, fuse filament fabrication, 3D printing

1. INTRODUCTION

Additive manufacturing, also generally known as 3D printing, is a revolutionary technology that allows the creation of 3D object from a digital file by depositing the material layer over layer.[1–3] This method differs from traditional subtractive manufacturing methods, in which material is removed from a solid block to create a finished product.[4] Additive manufacturing method has many advantages over subtractive manufacturing method, such as generating less waste material, using up to 80% lesser material for building an object, consuming lesser energy and significantly lesser in tooling cost. Due to these advantages, usage of additive manufacturing has increased nowadays and mainly used for rapid tooling, rapid prototyping and rapid manufacturing.[5]

Among many types of additive manufacturing technology, FFF is popular choice for 3D printing due several advantages, including affordable equipment costs, easy handling and wide range of printable materials. The industrial applications for FFF have vastly increased for rapid tooling and manufacturing. In rapid manufacturing, FFF allows for fabricating customised and complex parts with relatively low costs, as this manufacturing approach does not involve high tooling costs.[6] For an example, Philip offers user for customising pendant and lamp covers via FFF printing. For rapid tooling, FFF can now be used for printing patterns for casting, and these 3D printed patterns can be effectively burning out, leaving a cavity for the casting mould.[7] This process not only reduce the lead time of fabricating casting mould, but also reduce the costs associated with traditional mould-making techniques.

Thermoplastic filaments made from polylactic acid (PLA), polyethylene terephthalate glycol (PETG) and acrylonitrile butadiene styrene (ABS) are commonly used in FFF printing.[8] In the market, most 3D printing filaments are made from virgin plastic synthesised from petroleum. Filaments made from recycled plastic are limited. To promote sustainable 3D printing development, using recycled plastic as a 3D printable material offers several benefits as compared to virgin plastic filament, such as lowering carbon dioxide (CO_2) emission and reducing plastic waste which is abundantly available. Thereby indirectly reducing the environmental impact of plastic waste.[9] A notable company named Fishy Filament has successfully produced recycled nylon filament from fishing net waste. This filament had utilised by Philips for printing their lighting applications. The use of this material had resulted in 76% lower CO_2 emissions during the manufacture process.[10]

In the market, 3D printing filament made from recycled high-density polyethylene (rHDPE) is currently unavailable. There is still limited research on the production of 3D printing filament from rHDPE, particularly involving the use of the 3DEVO filament maker. Furthermore, a study assessing the 3D printability of rHDPE through the temperature tower test to determine printing temperature has yet to be found in open-source literature, highlighting a significant gap in current literature. The present study focuses on producing filament from rHDPE and comparing the printability and filament characteristic between rHDPE and commercial HDPE. In addition, the mechanical properties of 3D printed parts from rHDPE and commercial HDPE were evaluated.

2. METHODOLOGY

2.1 Preparation of Filaments

The rHDPE pellets were provided by Everlantern Plastic Industry Sdn. Bhd. The rHDPE pellets were dried at 60°C for at least 4 h using a Thermo-Line Drying Oven (model: SOV240B) for removing moisture. The rHDPE filament was prepared by using 3DEVO filament composer. This experiment used extrusion temperature ranging from 200°C to 220°C. The extruded filament showed better finishing and diameter consistence at 220°C. Additionally, the screw rotational speed was set at 4.5 rpm. the spooling speed was auto calibrated in order to obtain the filament diameter of 1.75 mm. For the commercial HDPE filament was purchased from Etilong High-tech Material Ltd. Changsha. Figure 1 shows the overall preparation process from filament to 3D printed specimens.

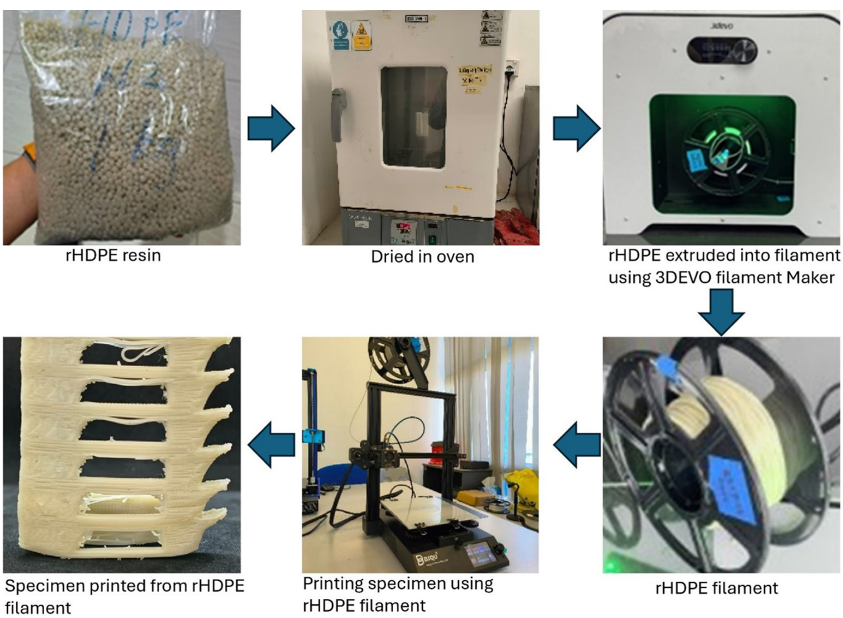

Figure 1: Preparation of rHDPE filament and specimens.

2.2 Printing of Specimens

All G-code files used for printing were prepared using PrusaSlicer (Ver 2.7.1). The nozzle size used for printing was 0.8 mm. A temperature tower was used in this experiment to determine the best printing temperature for rHDPE and commercial HDPE filaments. The temperature towers were printed at temperatures ranging from 200°C to 240°C. For tensile specimens, the printing temperatures were set at 220°C, 230°C and 240°C, as these temperature ranges showed good printing output during the printing of the temperature tower. The tensile specimens were printed with an infill direction of 90° to ensure the printed lines were parallel with the direction of tensile load. All specimens were printed using 3D printer (Brand: BIQU, model: B1). There was a necessary for applying biaxially oriented polypropylene (BOPP) tape to the printing bed to enhance the adhesion of printed part. The configuration for the printing was listed in Table 1.

Table 1: 3D printing parameters

Parameters	Value
Nozzle diameter	0.8 mm
Bed temperature	60°C
Bed thickness	3.0 mm
Layer height	0.4 mm
Line width	0.8 mm
Filling width	0.8 mm
Infill density	100%
Brim	5 mm
Filling pattern	0°
Flow	100%
Speed	50%
First layer speed	25 mm/s

2.3 Testing and Characterisation

The extruded filaments at temperatures ranging from 200°C to 220°C were captured using an Eakins' digital microscope with magnification of ×150. The cross-section of the extruded filament was also captured to analyse its ovality. A 200 cm length of the extruded filament was measured for its maximum and minimum diameter, with measurements taken at 10 cm intervals. The filament's ovality was calculated by Equation 1.

$$Ovality\ (\%) = \left[\frac{2 \times (Max_{OD} - Min_{OD})}{(Max_{OD} + Min_{OD})} \right] \times 100 \tag{1}$$

Melt flow index (MFI) measurements were conducted using the Dynisco LMI4000 Melt Flow Indexer referring to ASTM D1238 standard. The rHDPE and commercial HDPE filaments were first pelletised and dried in oven at 60°C prior to the test. The preheat time for the test was 5 min and temperature at 220°C, 230°C and 240°C. The load used in this experiment was 2.16 kg.

The printed specimens were tested for their tensile properties with aid of Instron universal testing machine (model: AGS-X, from Shimadzu). The test was performed by referring to standard ASTM D638. The crosshead speed of the machine was set to 10 mm/min and load cell of 15 kN was used.

3. RESULT AND DISCUSSION

3.1 Filament Characterisation

Figure 2 illustrates the effect of extrusion temperature on the extruded rHDPE filaments. At a temperature of 200°C, the filament exhibited a rough surface, and the extruded diameter was visibility inconsistent. When the temperature was increased to 220°C, the extruded rHDPE filament showed a noticeably smoother surface, and the extrusion process produced filaments with more consistent diameters. All rHDPE filaments displayed visible contamination; however, no large particles were observed within the filaments, which is expected when using recycled plastic materials. During the extrusion process, shear stress from the screw and tensile stress from the spooling unit caused the molecular chains of the melted plastic to stretch before relaxing as they exited the die. Figure 3 provides a schematic representation of the molecular chain orientations of the melted plastic at different processing temperatures. Semi-crystalline materials like HDPE typically exhibit die swelling during the extrusion process.[11] For recycled HDPE (rHDPE), die swelling was more pronounced at 200°C due to the reduced flexibility and stretchability of the molecular chains at this temperature. As a result, upon exiting the die and cooling, part of the molecular chains underwent elastic recovery, leading to an unsteady flow and a rough surface on the extruded filament. Conversely, at 220°C, the die swelling effect was significantly reduced. The higher temperature enhanced the flexibility of the molecular chains, allowing them to align more effectively under the applied shear and tensile stresses. This resulted in a steadier flow and a smoother surface finish for the filament. Based on these findings, an extrusion temperature of 220°C was selected for rHDPE in this experiment.

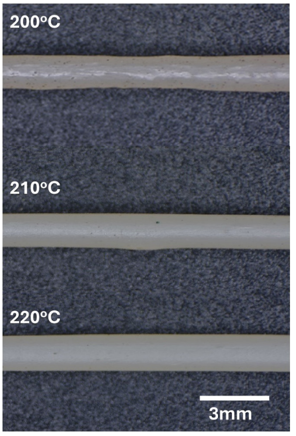

Figure 2: Extruded rHDPE filaments at different temperatures.

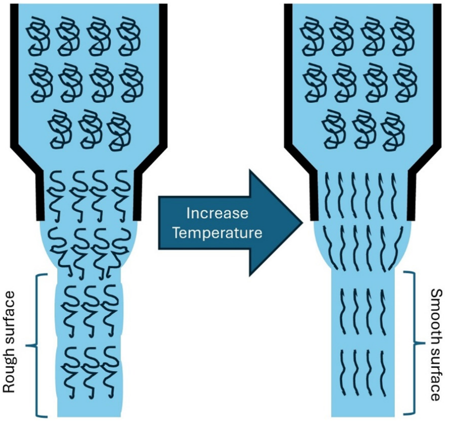

Figure 3: Schematic explanation on filament roughness due to temperature differences.

The standard diameter for a 3D printing filament is 1.75 mm with a tolerance of ± 0.05 mm. However, many manufacturers can control the tolerance within ± 0.03 mm. Figure 4(a) shows that the average measured diameter of commercial HDPE filament was 1.72 mm with a tolerance of ± 0.03 mm. In contrast, the filament made from rHDPE had an average measured diameter of 1.75 mm with a larger tolerance of ± 0.1 mm, meaning the diameter ranged from 1.65 mm to 1.85 mm. Generally, filament manufacturers produce filaments with slightly smaller diameters than 1.75 mm to avoid oversizing issues in extruders, which could lead to clogging at Teflon tube. In this study, the laboratory-scale filament extruder used for producing rHDPE filaments has limitations in controlling diameter tolerances. Additionally, the rHDPE resin comprises various grades of HDPE, including injection-grade and extrusion-grade resins, which poses a challenge in maintaining a consistent melt flow rate and achieving uniform extrusion geometry. Shiferaw et al. also produced 3D printing filament from rHDPE using their own fabricated filament extruder.[12] They reported that at a melting temperature of 200°C and below, the filament appeared rough, and its diameter was noticeably inconsistent. Their findings also indicated that a temperature of 220°C was suitable for producing rHDPE filament with a diameter of 1.75 mm within a low tolerance value.

The ovality of the filament is another significant factor. Figure 4(b) shows that the commercial HDPE filament had an ovality of 3.2%, while the rHDPE filament exhibited a higher ovality of 8.3%. Figure 5 highlights that the cross-sections of both filaments were noticeably oval in shape. For 3D printing filaments, the ovality should typically remain below 5%. HDPE is inherently a high-crystallinity material, which poses challenges in maintaining the filament's geometric consistency. Schirmeister et al. also extruded filament from virgin HDPE. They reported that the cross-section of the extruded filament was slightly oval, but it was still printable.[13] The 3DEVO filament extruder used in this experiment has two air blowers for cooling the extruded filament to solidify it. However, the cooling was uneven, with faster cooling occurring at the left and right sides compared to the front and rear sides. This uneven cooling led to differences in the crystallisation rate within the same cross-section of the filament, resulting in a high-ovality cross-section for the rHDPE filament. Industrial-scale equipment, with more advanced cooling systems, ensures more even cooling of the extruded material, allowing commercial HDPE filaments to achieve lower ovality compared to rHDPE. Despite the high tolerance and ovality observed in the rHDPE filament produced in this experiment, no printing issues were encountered. This is because the filament diameter remained below 1.9 mm, which is the inner diameter of the polytetrafluoroethylene (PTFE) tube used in the 3D printer. As long as the filament diameter is smaller than the tube's inner diameter, clogging at the tube will not occur.

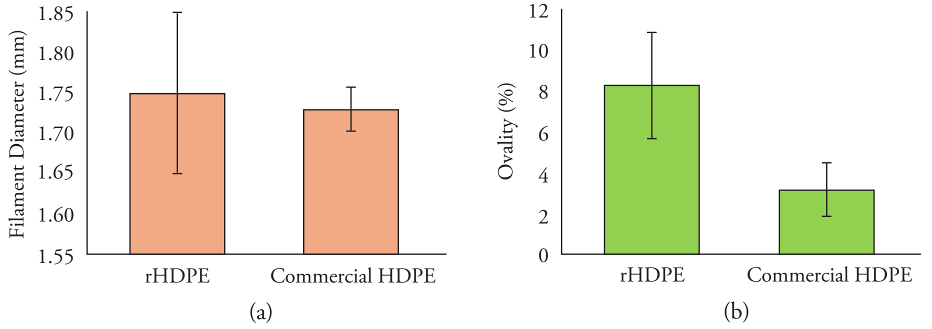

Figure 4: (a) Filament diameter and (b) ovality of rHDPE and commercial HDPE filaments.

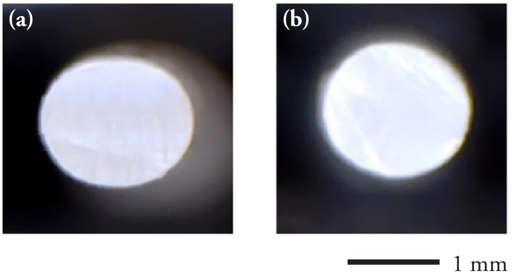

Figure 5: Photomicrograph of cross-section of (a) rHDPE and (b) commercial HDPE filaments.

3.2 Visual Observation

Temperature tower is commonly used to determine the optimal printing temperature for a 3D printing filament. It allows for evaluating the material's printability by assessing the quality of printed features, such as bridges and overhanging regions, at various temperatures. Figure 6 shows the temperature towers printed using commercial HDPE and rHDPE filaments, with printing temperatures ranging from 200°C to 240°C. From the bottom edges of both temperature towers found an obvious edge crup up due to material shrinkage. The bottom edges of both temperature towers show noticeable curling, attributed to material warpage. As similar finding also found by Schirmeister et al. in printing virgin HDPE materials.[13] This behaviour is expected when printing with semi-crystalline materials.[9] During the cooling process, printed HDPE undergoes volumetric shrinkage due to crystallisation. The commercial HDPE was unable to complete the print at temperatures reaching 200°C, as it did not have sufficient heat to melt. In contrast, rHDPE was able to finish the print even at temperatures as low as 200°C. This may be due to the rHDPE containing different grades of HDPE resin, resulting in a lower melt viscosity compared to commercial HDPE. As a result, rHDPE can be printed at 200°C. If observed on printed bridge region of the temperature tower, tower printed with commercial HDPE temperature 215°C and below shows the printed layers were sagged. For tower printed with rHDPE, the bridge on the temperature tower were sagged at 225°C and below. This phenomenon is associated with the die swelling behaviour of plastic materials. As observed by Colon et al., plastic materials exhibit a high die swell ratio when printed at lower temperatures, while the die swell ratio decreases as the printing temperature increases.[14] At low printing temperatures, HDPE molecular chains experience increased die swelling. As the extruded HDPE exits the die, the chains undergo partial elastic recovery due to their reduced flexibility and stretchability. This behaviour makes it hardly to achieve a straight and stable extrusion line during printing. That is why the printed lines sagged when printing the bridge, it can also be observed in the printed overhanging region on the right side of the temperature tower that at lower printing temperatures, the edges of the overhanging region become noticeably distorted. This occurs because, at lower temperatures, the HDPE molecular chains are less flexible and stretchable, making it difficult for the extruded material to retain its shape during printing. The tower was well printed with commercial HDPE, starting at a printing temperature of 220°C, with no noticeable slagging at the bridge. In contrast, the tower printed with rHDPE required a higher printing temperature, starting at 230°C, to achieve better print quality. Based on these results, tensile test specimens were printed at temperatures ranging from 220°C to 240°C to further evaluate which printing temperature provides the optimum mechanical strength for the printed parts.

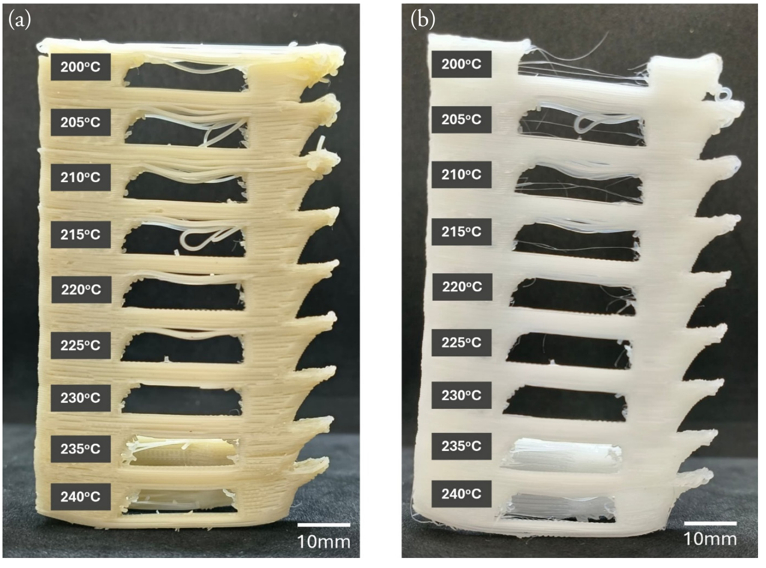

Figure 6: Temperature tower printed with (a) rHDPE and (b) commercial HDPE filaments.

3.3 Tensile Properties

Figure 7 shows that the printed specimens using rHDPE filaments exhibited optimal tensile strength of 20 MPa and modulus of 880 MPa when printed at 230°C. In comparison, the printed specimens using commercial HDPE filament demonstrated optimal tensile strength of 23 MPa and modulus of 910 MPa. The tensile properties of the printed specimens were influenced by the adhesion between adjacent printed layers. For HDPE materials, optimal adhesion between the deposited material was achieved due to a decrease in the melt viscosity of the polymer as the printing temperature reached 230°C. From Figure 8, the printed specimens from both commercial HDPE and rHDPE exhibited ductile fracture without noticeable interlayer delamination. This phenomenon indicates that the specimens had good layer adhesion when printed at 230°C. In contrast, specimens printed at 220°C exhibited lower tensile strength and modulus. This was due to the lower printing temperature, which compromised adhesion between the printed layers. Figure 9 shows that both printed specimens experienced layer delamination under tensile loading, resulting in fracture at a lower strength. This evidence suggests that specimens printed at 220°C had weaker interlayer adhesion, contributing to their lower tensile strength and modulus. As the printing temperature increased to 240°C, the tensile strength and modulus of printed specimens using commercial HDPE and rHDPE decreased, likely due to thermal degradation, which compromised the mechanical properties of the materials. A similar observation has been reported by other researchers, who found that printed parts using both virgin and recycled plastic filaments exhibited optimal tensile strength and modulus at a specific printing temperature.[8,15] Beyond this temperature, the mechanical properties tended to decline. They also attributed this phenomenon to thermal degradation. Figure 7 shows that printed specimens using rHDPE filament exhibited lower tensile strength and modulus compared to those using commercial HDPE. This result was expected, as rHDPE typically contains a mixture of different HDPE grades and impurities. The incompatibility between these HDPE grades, along with the presence of contaminants, likely contributed to the deterioration of the material's mechanical properties. However, the difference in their optimum tensile strength was relatively small, at approximately 12%. This study also found that parts printed with recycled PLA exhibited significantly lower mechanical properties compared to those printed with virgin PLA.[16]

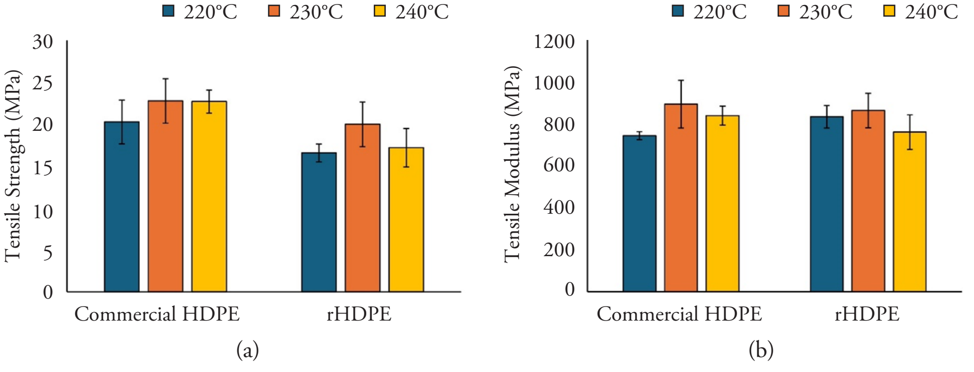

Figure 7: (a) Tensile strength and (b) tensile modulus of printed specimens using rHDPE and commercial HDPE at different temperature.

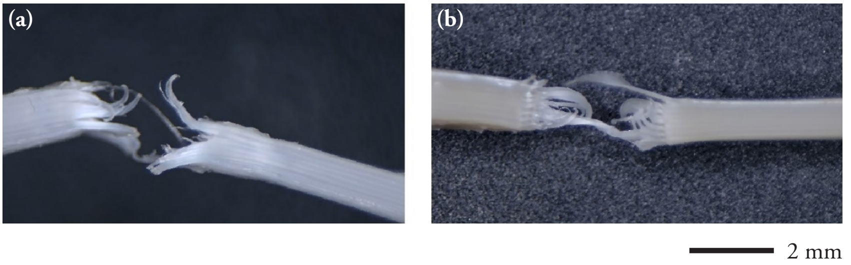

Figure 8: Fracture specimens printed with (a) commercial HDPE and (b) rHDPE at temperature of 230°C.

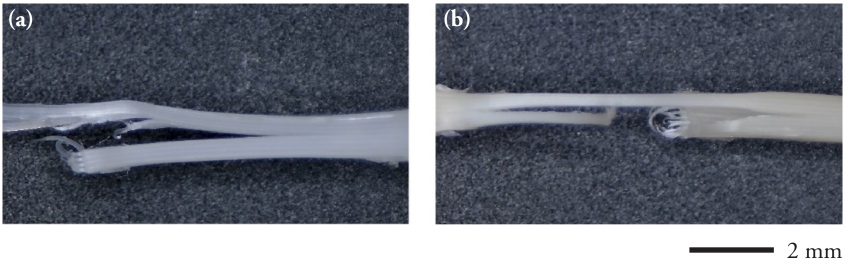

Figure 9: Fracture specimens printed with (a) commercial HDPE and (b) rHDPE at temperature of 220°C.

3.4 MFI Analysis

Figure 10 presents the MFI values of commercial HDPE and rHDPE at different temperatures. As the temperature increased, the MFI values of both materials also increased. This rise in temperature enhanced polymer chain mobility, leading to an increase in melt flow. Improved melt flow can enhance adhesion between printed layers. On average, commercial HDPE exhibited an MFI value 4.4% lower than that of rHDPE. Recycled plastic is expected to have a higher melt flow rate compared to virgin plastic due to several factors. The presence of unknown additives may have a lubricating effect, while the mixture of various plastic grades, especially a high proportion of injection-grade plastics which can lower melt viscosity. Additionally, the melting and reprocessing of recycled plastic can shorten polymer chains, further reducing viscosity and increasing the melt flow rate.[17] This finding supports the results reported in Section 3.2, where rHDPE demonstrated lower melt viscosity than commercial HDPE. This explains why rHDPE could be printed at 200°C, whereas commercial HDPE was not printable at this temperature. Due to its higher melt flow rate, rHDPE fuses more quickly, allowing for better coalescence between printed layers. In contrast, commercial HDPE, with its lower melt flow rate, requires a higher temperature to achieve proper fusion. Wang et al. also found that the PLA exhibited the similar observation, where the PLA with high MFI values can be properly printed at lower printing temperature.[18]

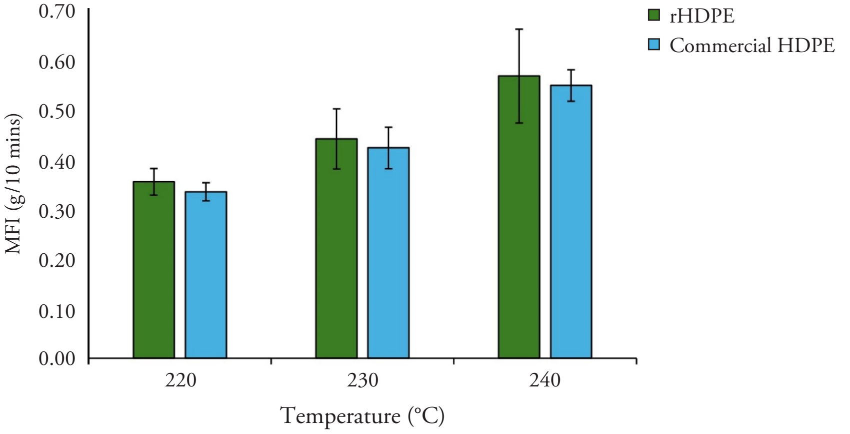

Figure 10: MFI of rHDPE and commercial HDPE at different temperature.

4. CONCLUSION

This study demonstrated that rHDPE can be successfully extruded into 3D printing filament at a temperature of 220°C. Below this temperature, the material experiences noticeable die swell, leading to challenges in controlling the filament diameter. Due to the presence of impurities, variations in HDPE grades within rHDPE, and differences in equipment, the filament can still be fabricated with an average diameter of 1.75 mm within an acceptable tolerance range for printing. However, its diameter deviation and filament ovality remain significant compared to commercial HDPE filament, which has a smaller tolerance value and lower ovality. Both printed parts made from rHDPE and commercial HDPE filaments exhibit noticeable warpage at the bottom layers of the print. This is the main limitation of using HDPE for 3D printing applications. The rHDPE filament can be printed starting from 200°C due to its lower melt viscosity and higher MFI value. In contrast, commercial HDPE, which has a lower MFI value, requires a printing temperature start from 205°C. Both printed specimens with rHDPE and commercial HDPE exhibited optimum tensile strength and modulus when printed at 230°C. However, printing beyond this temperature resulted in decreased tensile properties due to thermal degradation. Additionally, printed specimens with rHDPE showed lower tensile strength and modulus compared to those with commercial HDPE filament. It is important to know that the use of rHDPE as 3D printing material presents several constraints including inconsistent filament geometry, low mechanical strength and persistent warpage issues. These factors may limit application of rHDPE in functional or load-bearing 3D printed components. Therefore, future work should focus on enhancing filament diameter accuracy, reducing warpage, and improving the strength of printed parts.

5. ACKNOWLEDGEMENTS

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