Developing Recycled Carbon Fibre Reinforced Material for Sustainable Additive Manufacturing

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ABSTRACT

This paper explores the compounding of recycled polyethylene terephthalate glycol (rPETG) with recycled carbon fibres (rCF) for use in Sustainable Additive Manufacturing. Different forms of rCFs and weight fractions were added to rPETG to assess the dispersion and thermal properties of the formulations achieved when using a two-stage compounding process. The polymer composite mixture was blended in the chamber at 260°C for 5 mins, cooled down to 230°C for 1 minute prior to extrusion, this process was repeated with the produced filament. Cross-section and in-plane samples were potted, grinded and polished to facilitate optical microscopy of the samples. The resultant micrographs were analysed to evaluate the fibre orientation and fibre area fraction. Differential scanning calorimetry of the samples was also undertaken to investigate the effect of adding the fibres on the glass transition temperature of the corresponding polymer composite.

The primary objective of the work was to create and evaluate polymer composite from recycled materials that could be used with 3D printing as an alternative to composite filament made of virgin materials at a fraction of the material cost without compromising its properties. The results show a high degree of directional conformity between the filament extrusion direction and the fibre orientation. The micrographs also indicate a homogeneous distribution of fibres after the two-stage compounding process. The addition of recycled carbon fibres did not appear to have a significant effect on the glass transition temperature of the polymer composites.

1 INTRODUCTION

Carbon Fiber Reinforced Plastics (CFRPs) are becoming increasingly popular for use in high-performance applications due to their impressive lightweight and superior mechanical properties. These features make them an ideal replacement for more traditional materials. The demand for CFRP increased from 128,500 tons in 2018 to 141,500 tons in 2019 and is foreseen to be 197,000 tons in 2023 [1]. However, lacking commercial solutions for composites waste and a narrow market for recycled carbon fibre (rCF) products have become the main challenges to achieving sustainable composites manufacturing. It has been reported the production of virgin Carbon Fibre (vCF) is extremely energy-consuming (183–286 MJ/kg), with a minimum price of 30–40€/kg [1]. Hence, extending the carbon fibre's life cycle, converting the recycled carbon fibre (rCF) to the high value intermediate materials and rolling them back to the design & manufacturing loop is an urgent enquire from the composites industry to achieve NetZero – Resource Efficient Manufacturing.

Similarly for polymeric materials such as thermoplastic, their recycling rate is still relatively low, currently estimated to be around 14% on a global scale. In Europe, despite taking the lead in the global fight against climate change and environmental degradation, its recycling rate is only around 30%, with the rest either being incinerated or ending up in landfills [2]. The largest contributor to the production of plastic waste is the packaging industry. To reduce the impact of plastic on the environment, the EU Commission has targeted that, by 2030, all plastic packaging placed on the EU market is either reusable or can be recycled in a cost-effective manner [3].

Polymer materials are one of the most extensively used materials for additive manufacturing, (also known as 3D printing) making this manufacturing technology a viable option for utilising recycled polymers in order to reduce cost and carbon footprint. It is anticipated that the use of 3D-printed polymers will continue to expand due to their technological advancements and increased production capabilities. The material extrusion additive manufacturing favours low melting point thermoplastic such as polycarbonate (PC), poly-lactic acid (PLA) and Acrylonitrile butadiene styrene (ABS) [4, 5] which are commercially available at a relatively low price. However, these polymers are not that suitable for structural load bearing applications. Recent advances have seen high performance engineering thermoplastics such as Poly-ether-ether-ketone (PEEK), Polyamide (PA), polyvinylidene fluoride (PVDF) have been developed by several companies to be used with commercial industrial and desktop 3D printers [4]. However, these polymers are often expensive, and they require special hot temperature nozzles, heated chambers, and relatively high bed temperatures, thereby increasing manufacturing cost. In the last few years, Polyethylene terephthalate glycol-modified (PETG) has gained considerable attention in the 3D printing sector due to its properties resembling those of ABS, but with a less viscous consistency and less shrinkage [5]. In addition, PETG also has impressive tensile properties with high processability and excellent chemical resistance [4]

Fibres are added into thermoplastics to create polymer composites filament with enhanced properties. It has been reported that the incorporation of either short or powdered carbon fiber into polymeric samples demonstrated a substantial improvement in the tensile properties of PLA, nylon and ABS [6]. Nonetheless, these improvements are largely influenced by several factors such as fibre-matrix compatibility, fibre dispersion and homogeneity, fibre length and fibre content which needs to be optimised during blending and compounding of these materials.

When examining the pricing of 3D printing filament, it is evident that fibre-reinforced thermoplastic filament is nearly twice the price of the neat polymer filament [1]. In addition, the neat virgin polymer filament is two or three times the price of pellets [7]. Thus, if a substitution of these virgin materials were to be made, an obvious financial benefit would be seen in the reduction of the cost of composite filaments. This step-changing material combined with an improved recycling process for both polymers and fibres, will not only make additive manufacturing an affordable option for SMEs to manufacture lightweight sustainable 3d printed products but also make the zero-waste manufacturing concept a reality.

2 EXPERIMENTAL WORK

2.1 Materials

Recycled PETG (rPETG) was provided by EVO-3D, Exeter, UK in the form of transparent resin pellets. These pellets were produced through the mechanical recycling of medical tray waste. The recycled carbon fibres were purchased from Procotex, Belgium. The densities of these materials are listed in Table 1.

Table 1: Raw Material Properties

Material	Density (g/m3)
Recycled PETG, rPETG	1.27
Recycled CF - short fibres, rCF	2
Recycled CF - granules, gCF	2

2.2 Preparation of Polymer Composite Filaments

A compounding process was used to combine recycled PETG (rPETG) and recycled carbon fibres (rCF). The Xplore Micro Compounder twin screwdriver (Xplore Instruments BV, Sittard, The Netherlands) with a mixing chamber volume of 15 mL was used to blend materials and provide mixing dispersion. Initial processing took place at 260°C, with a screw speed of 100 rpm which produced up

to 40 Nm of torque. The rPETG was fed into the micro compounder by an auto feeder, where the polymer was initially processed individually in this environment for two minutes to ensure it was fully melted prior to the addition of fibre reinforcement. The rCFs was then manually fed into the compounding chamber to be dispersed in the polymer matrix for 5 minutes. The chamber was then cooled to 230°C for 1 minute before being extruded, producing an approximately 1.75mm diameter filament. To further improve the fibre dispersion, the filament was pelletized and reprocessed using the same parameters. Different material compositions as displayed in Table 2 have been explored by altering the weight fraction and fibre length of rCFs. These alternatives were tested to evaluate processability and material performance.

Table 2 : Formulation of polymer composites filament

Sample ID	Formulation	rPETG, wt%	rCF, wt%	rCF, Vf %	Fibre length, mm	Forms
Sample 1	rPETG/gCF5	95	5	3.23	0.3	Granuled
Sample 2	rPETG/gCF15	85	15	10.08	0.3	Granuled
Sample 3	rPETG/rCF5	95	5	3.23	3	Short fibre
Sample 4	rPETG/rCF15	85	15	10.08	3	Short fibre

2.3 Differential Scanning Calorimetry (DSC)

DSC analysis was conducted using a Netzsch DSC 214 Polyma. Measurements were performed on samples of 8-16 mg, placed in aluminium concavus crucible pans. The specimens were subjected to a heating and cooling temperature cycle of 25 °C to 300 with a heating and cooling rate of 20 °C/min under a nitrogen stream of 40ml/min. The DSC thermogram of the 2nd heating and cooling cycle was examined.

2.4 Microscopy Analysis

Optical microscopy analysis was carried out on polished specimens using the Leica DM12000 M. Micrographs were taken of the cross-section and in-plane segments of the samples to provide fibre volume area and fibre directionality respectively. The micrographs also allowed for the fibre dispersion to be visually inspected.

3 RESULTS AND DISSCUSSION

A combination of thermal and microscopy analysis techniques was used to ascertain compound properties and were a means for comparison.

3.1 Thermal property -Glass transition temperature

The thermal properties of rPETG and their corresponding polymer composites are shown in Figure 1and Table 3. The DSC analysis was performed to evaluate the effect of adding recycled carbon fibre reinforcement on the glass transition temperature (T_g). As shown by the results in Table 3, the T_g for neat rPETG was measured as 80°C which is consistent with the datasheet [8]. Because PETG is 100 % amorphous polymer, no melting and crystallisation peak were observed. The addition of 5 wt% and 15 wt% granuled rCF did not have a significant effect as the T_g was only slightly reduced. The lowest T_g was observed with the addition of 15 wt% recycled short fibres. The enthalpy relaxation of PETG indicated by the small endotherm seen after the glass transition, is associated with the physical aging process due to rapid cooling of the polymer below its T_g during the extrusion process [4].

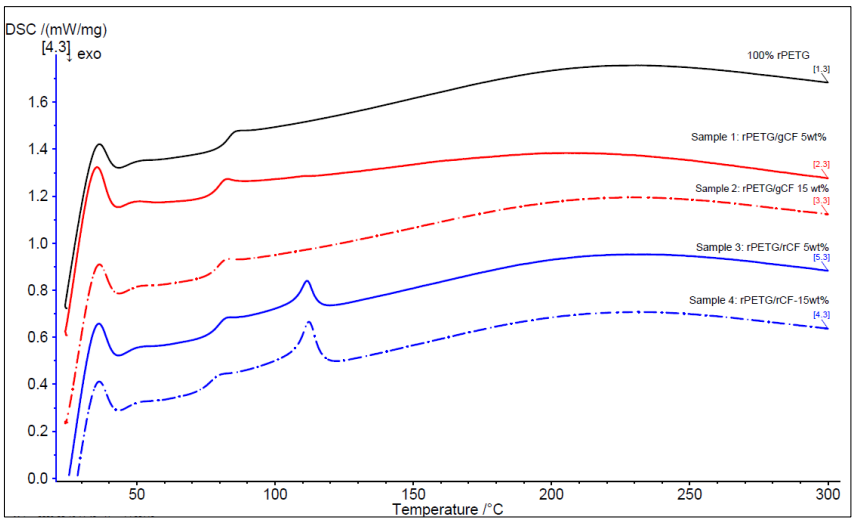

Figure 1: DSC curves of polymer composites

Table 3: Glass transition temperature

Sample ID	Formulation	T_g (°C)
Neat polymer	100% rPETG	79.6
Sample 1	rPETG/gCF5	76.4
Sample 2	rPETG/gCF15	76.0
Sample 3	rPETG/rCF5	76.0
Sample 4	rPETG/rCF15	73.2

3.2 Microscopy Analysis

In-plane micrographs were taken in parallel with the filament extrusion direction. The filament boundary was included in the micrographs to provide a datum from which the fibre directionality was established, see Figure 2. The images were captured at 5x magnification at three locations along the filament length. Subsequently, these images were processed using Fiji ImageJ's 'directionality' [9] feature which provided data on the average direction and dispersion (standard deviation) of the fibres. Table 4 below shows the directionality results from the four samples analysed. Figure 3, shows an exemplar histogram from Sample 4.

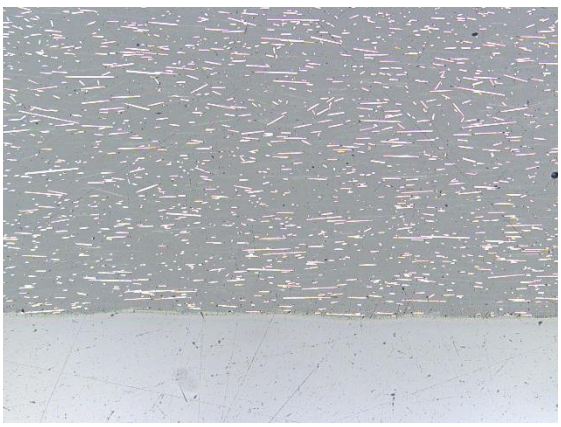

Figure 2: In-plane micrograph

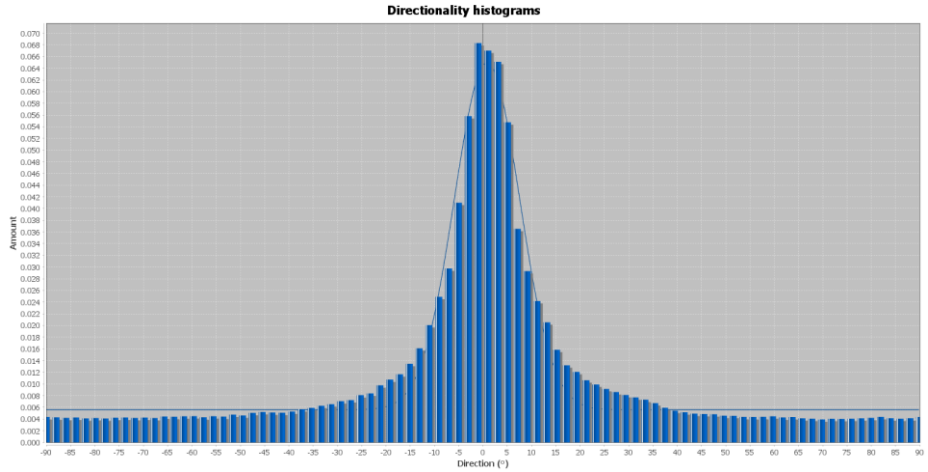

Figure 3 :Histogram

Table 4 : Direction and dispersion of fibres in polymer matrix

Sample ID	Formulation	Direction (°)	Dispersion (°)
Sample 1	rPETG/gCF5	0.167	6.083
Sample 2	rPETG/gCF15	1.113	7.313
Sample 3	rPETG/rCF5	-1.520	6.395
Sample 4	rPETG/rCF15	-1.060	7.193

The fibre directionality across all samples shows high degree of uniformity, within ±1.6° with an average dispersion of 6.746°, from the extrusion direction. As shown in Figure 4, the cross-sectional micrographs provided a good visual representation of the fibre distribution. At 5x magnification the whole fibre cross-section can be viewed, but for the fibre area fraction to be measured, 50x magnification was used at three areas of the fibre cross-section. The resultant micrographs, were analysed using Fiji ImageJ's 'analyse particles' [9] function which provided the fibre area fraction for each sample.

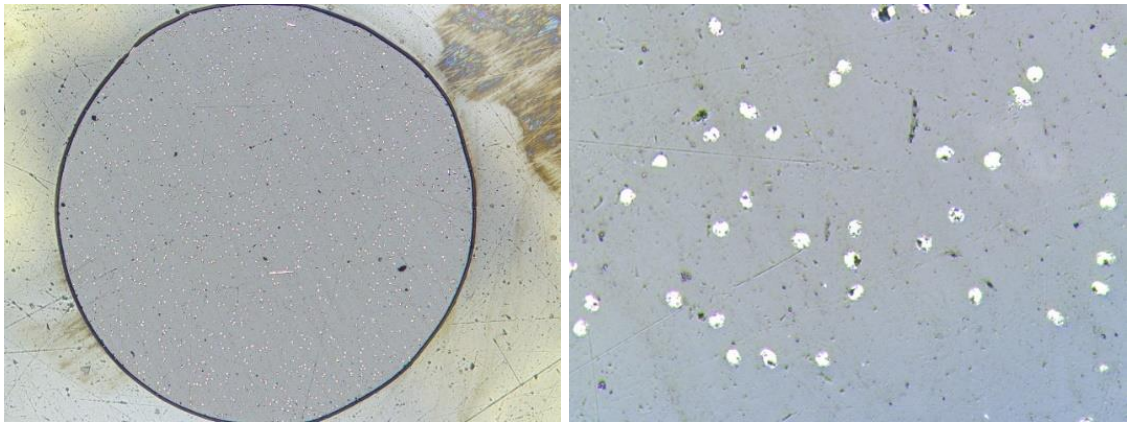

Figure 4 : Cross-sectional micrographs

From both the in-plane and cross-section micrographs, there is a homogeneous fibre distribution which validates the two-stage dispersion method when compounding the samples. Fibre directionality across all samples was strongly aligned with the filament extrusion direction. The fibre area fractions and resultantly calculated fibre weight fractions were lower than expected across all samples; this can be attributed to the manual feeding of rCF to the compounder and the small sample pool used for these initial findings.

4 CONCLUSIONS

This initial research has examined the compounding of rPETG with different fibre lengths and weight fractions of rCF. Initial results have shown that the two-stage compounding method was able to produce a homogeneous distribution of fibres through the matrix. Through fibre directionality analysis, a strong correlation between filament extrusion direction and fibre orientation has been observed. The fibre area fraction of the samples is lower than expected although this can be attributed to the manual addition of fibres during compounding process and the small sample pool – to this end, further work will be carried out in this area. DSC analysis has shown that the addition of recycled carbon fibres did not have significant effect on the glass transition temperature of the polymer composite.

5 FUTURE WORK

To have an indication of the carbon fibre content of the composite filament, and to validate the microscopy area fraction values, Thermogravimetric analysis (TGA), which measures material degradation as a function of time and temperature through changes in mass, will be performed. A further analysis on the mechanical properties of polymer composites will be conducted to ascertain the yield strength, ultimate tensile strength (UTS), breaking strength and Young's modulus of the different compositions. Processing parameters will also be optimised to improve yield efficiency and achieve consistent quality composite filament.

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