Mechanical Properties of PETG Fibres and Their Usage in Carbon Fibres/Epoxy Composite Laminates

Abstract
This paper reports on a melt-spinning process of glycol-modified poly(ethylene terephthalate) PETG and regranulate of PETG. The effect of the processing temperature and winding reel velocity on the diameters of fibres was examined. It was observed with a scanning electron microscope that the surface of fibres produced from recycled PETG are thicker but smoother than fibres made of fresh PETG. Applying a higher drawing velocity helped to decrease the diameters, which were between 75-150 μm. Under static deformation, fibres showed different behaviour, with higher flexibility and lower strength observed for fibres made of PETG regranulate. Both types of fibre were chopped and added to carbon fibre reinforced polymers as interlayers to investigate their effect on mechanical properties. It was found that the flexural strength decreased in the presence of PETG fibres, while interlaminar shear strength improved, but only in the case of fresh PETG fibres.

Key words: PETG, fibres, CFRP, mechanical properties, SEM, recycling.

Materials and methods
Two types of material were used to produce fibres: PETG with the trade name Eastar™ Copolyester 6763 (Eastman Chemical Company, USA), and pellets of regranulate PETG (rPETG) produced by mechanical recycling of post-consumer printed foil. Both types of fibre were obtained directly from pellets using a laboratory twin-screw extruder (HAAKE Minilab, Thermo Scientific, Germany), a round shape nozzle with a diameter of 0.75 mm, and a winding reel. Two temperatures were applied (200 °C and 210 °C) and two winding reel velocities (600 rpm and 1000rpm), while the screw’s rotation was kept constant at 40 rpm. An example designation of fibres produced is PETG/200/600, meaning that they were made from fresh PETG at 200 °C and a winding reel velocity of 600 rpm. In total, 8 types of fibres were produced.

Three types of carbon fibre and epoxy resin laminate were produced. A mould consisting of a glass plate was coated with perforated foils instead of a release agent. 16 layers of carbon-epoxy UD prepreg MTM46/HTS5631 (Cytec Industrial Materials, UK) were cut to a size of 300 mm x 150 mm with a lay-up of [0]₁₀. The prepreg had an areal weight of 150 g/m², containing 35% epoxy resin and 12,000 carbon fibres. Again on the top layer of the prepreg, perforate foil was placed, along with a polyester mat to drain the rest of the epoxy resin. Finally the laminate was covered with nylon foil as a vacuum bag. The mould was placed...
in a vacuum oven, heated to 130 °C, and cured for 90 minutes. This laminate is referred to as the reference panel and has a thickness of 2.2 mm. Composite laminates with PETG fibres and rPETG fibres were manufactured in the same conditions as the reference panel. In these cases, the fibres were cut to a length of 7 cm and then distributed randomly by hand between each layer of the prepreg. The amount of PETG and rPETG fibres in each layer corresponds to an areal weight of 12 g/m² and 5 wt% of the total matrix content of the laminate. The volume fraction of PETG/rPETG fibres in the laminate was 0.5 and the thickness was 2.4 mm.

For microscopic observations, fibres were first coated with a thin layer of gold. The diameters of single fibres were measured using a scanning electron microscope (TM 3000 Hitachi, Japan) at a voltage of 5 kV. The average diameters were calculated from at least 100 measured fibres. The quality of composite laminates produced was also analysed by SEM. For this, a small piece of laminate was cut and finished using a polishing machine. The porosity of each laminate was determined from minimum 8 SEM images using ImageJ software. SEM was also applied to analyse the laminates after the ILSS test.

Mechanical properties of the fibres were characterised using a standard tensile machine (EZ-SX Shimadzu, Japan). A single fibre was fixed to a special thick paper frame equipped with metal disks using an epoxy glue. After solidification, the frame with the fibre was clamped to the machine’s holder with glue and cut directly before the test. Of each type of fibre, 10 samples were measured at a constant cross-head displacement rate of 10 mm/minute at room temperature. Taking into account the diameter of the fibres, the average tensile strength, elongation at break and Young’s modulus were calculated.

Mechanical properties of the laminates were characterised in a three-point bending test (ASTM D790-17) and short beam shear test (ASTM D2344) using an MTS Q/Test 10 universal testing machine. From each test, 8 specimens were cut from different places in the laminates in the direction of the carbon fibres. For the short beam shear, the dimensions of the specimen were 25 mm x 10 mm x 2.2 mm, and for the three-point bending, the dimensions were 42 mm x 12.7 mm x 2.2 mm for the reference panel and 46 mm x 12.7 mm x 2.4 mm for the laminates with inserted fibres. All samples were tested at room temperature at a speed of 1 mm/min.

Results and discussion

PETG and rPETG fibres

The melt-spinning process of PETG and rPETG was uncomplicated and fibres had a rather low tendency to break during winding. The noticeable difference between the colour of the fibres produced was presented in Figure 1. Those made from PETG were white, while those from rPETG were slightly pink. Moreover fibres made from rPETG were significantly smoother and flatter on the surface, as is visible from the SEM images presented in Figure 1.

The effect of the processing conditions on fibre thickness was observed by measuring their diameters using SEM (Table 1). For PETG, the melt-spinning process was continuous when the extrusion temperature was 210 °C. The fibres obtained at 200 °C had smaller diameters. In the case of rPETG fibres, the application of a higher extrusion temperature of 210 °C resulted in a much more difficult process and thicker fibres with diameters over 200 μm. This was caused by a strong Baer effect, enhanced by remaining traces of solvent [9]. Except for these fibres, the rest had diameters around 150 μm. Applying a higher winding reel velocity re-

### Table 1. Mechanical properties and average diameters of fibres produced.

<table>
<thead>
<tr>
<th>Fibre</th>
<th>Average diameter, μm</th>
<th>Tensile strength, MPa</th>
<th>Elongation at break, %</th>
<th>Young’s modulus, MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>PETG/200/600</td>
<td>138 ± 26.9</td>
<td>122 ± 22.9</td>
<td>136 ± 21.0</td>
<td>2437 ± 294.4</td>
</tr>
<tr>
<td>PETG/200/1000</td>
<td>126 ± 41.5</td>
<td>157 ± 20.8</td>
<td>111 ± 21.1</td>
<td>3364 ± 696.1</td>
</tr>
<tr>
<td>PETG/210/600</td>
<td>151 ± 24.9</td>
<td>92.7 ± 27.5</td>
<td>142 ± 35.9</td>
<td>1555 ± 451.2</td>
</tr>
<tr>
<td>PETG/210/1000</td>
<td>75.1 ± 3.87</td>
<td>205 ± 26.5</td>
<td>48.4 ± 11.8</td>
<td>4678 ± 656.9</td>
</tr>
<tr>
<td>rPETG/200/600</td>
<td>157 ± 33.9</td>
<td>24.5 ± 4.37</td>
<td>195 ± 56.2</td>
<td>1656 ± 284.2</td>
</tr>
<tr>
<td>rPETG/200/1000</td>
<td>139 ± 22.9</td>
<td>25.1 ± 4.05</td>
<td>137 ± 59.7</td>
<td>1467 ± 250.0</td>
</tr>
<tr>
<td>rPETG/210/600</td>
<td>272 ± 47.9</td>
<td>8.26 ± 1.45</td>
<td>171 ± 41.6</td>
<td>532.2 ± 96.01</td>
</tr>
<tr>
<td>rPETG/210/1000</td>
<td>238 ± 58.7</td>
<td>7.77 ± 2.66</td>
<td>136 ± 21.2</td>
<td>669.2 ± 237.7</td>
</tr>
</tbody>
</table>
sulted in thinner fibres, as was especially observed for PETG processed at 210 °C and 1000 rpm. Decreasing the diameters of fibres is an important step in their further application in CFRP. Using fibres that are too thick decreases the volume fraction of carbon fibres and, consequently, the mechanical properties of the laminates.

Representative strain–stress curves for both types of fibre are presented in Figure 2. The first section of the curve is rectilinear and indicates the elastic deformation of the fibres. From this part, Young’s modulus was determined. After the yield point was reached, the fibres started to deform more plastically, which is associated with the orientation of the amorphous phase and the formation of a neck in the sample. Interestingly, in PETG, this resulted in strain hardening and increased stiffness, while rPETG fibres deform and become softer, without hardening. This difference in mechanical deformation may be related to the shorter polymer chains in regranulate and the effect of plasticisers from the solvent used to remove inks from the PETG foils.

It was noticed that rPETG fibres are more flexible than PETG fibres, as is confirmed by the higher elongation at break values obtained for rPETG in comparison to PETG fibres produced under the same processing conditions (Table 1). Moreover fibres which have high elongation also possess lower stiffness. For PETG fibres, the highest Young’s modulus achieved was 4.7 ± 0.6 GPa when the processing temperature was 210 °C and the winding velocity 1000 rpm. Increasing the fibre strength by applying higher rotational speed was observed for cotton yarns [10]. For fibres made from rPETG this is only 1.7 ± 0.3 GPa for a processing temperature of 200 °C and winding speed of 600 rpm. As expected, the tensile strength is higher for PETG fibres (205 ± 26.5 MPa) and much lower for rPETG fibres (25.1 ± 4.05 MPa). The reduced strength of fibres fabricated from PETG after recycling is related to shorter polymer chains and the lower molecular weight. However, the values of elongation at break obtained for rPETG fibres are much higher than for recycled PET fibres, for which the elongation was around 22% [11].

**Figure 2.** Examplary strain-stress curves for fibres made of neat PETG and rPETG.

**Figure 3.** SEM images of laminates reinforced with neat PETG fibres (a, b) and recycled PETG fibres (c, d).
Composite laminates

The incorporation of thermoplastic fibres into CFRP should not affect the quality of laminates, especially as it does not cause the formation of many pores. The value of porosity calculated was 0.14% for the reference panel, 0.01% for the panel with PETG fibres and 0.02% for that with rPETG fibres. It is confirmed by the cross-section of the laminates produced with layers of PETG and rPETG fibres, checked by SEM and presented in Figure 3. Generally neither laminates have pores or other defects, which means that the usage of thermoplastic fibres as additional layers does not disturb the fabrication process of laminates. The curing temperature and pressure applied resulted in the melting of both types of fibres and the formation of continuous layers between carbon fabrics (Figures 3.a, 3.c). Images created at a higher resolution (Figures 3.b, 3.d) also showed a lack of delamination and cracks in the composites, as well as satisfactory compatibility between PETG or rPETG, carbon fibres and epoxy resin.

The main goal of the introduction of thermoplastic polymer into CFRP is to improve the mechanical properties. Figure 4 shows that the flexural strength is highest for the reference panel, and is about 200 MPa and 300 MPa less in the case of laminates with PETG and rPETG, respectively. In the case of ILSS, there was a visible improvement of about 20%, but only when PETG fibres were inserted. For laminates reinforced with rPETG fibres, there was no improvement in interlaminar properties. The lack of a significant increase in the mechanical properties of CFRP by insertion of PETG and rPETG fibres could be related to the fact that the diameters of these fibres were too large, which strongly affects the effectiveness of strengthening due to insufficient wetting of carbon fabrics by the thermoplastic resin [12, 13]. The other possibility reported in the literature is inhomogeneous distribution of fibres in each layer, which leads to the formation of a local higher concentration of polymer [14].

SEM images of the laminate’s surface after the ILSS test is shown in Figure 5. In each case the laminate was not totally destroyed, but there is a visible difference in the broken area. The highest is for the reference panel (Figure 5.a), while the highest cohesion was exhibited by the
lamine with PETG fibres (Figure 5.c). Moreover in the reference panel the fracture is brittle, showing a lot of broken fibres in the destroyed area. Modification of CFRP with both thermoplastic fibres leads to less brittle fracture and better cohesion of the sample (Figures 5.d, 5.f) due to the reinforcement of weak interlaminar region of the lamine [15]. Interestingly the application of rPETG fibres as interlayers did not result in the same improvement in ILSS than for the reference CFRP. From the SEM images (Figure 5.f), much deeper and sharp fracture of fibres is seen, which could be related to a trash amount of solvent remaining in rPETG fibres, and consequently with lower adhesion of the polymer to the carbon fabric, or due to higher fibre brittleness.

Conclusions

In this study, a melt-spinning process was applied to produce fibres directly from fresh PETG and from regranulate of PETG obtained by washing out the printing from post-consumer foils. It was found that for fresh PETG, a suitable processing temperature is 210°C, which is too high for rPETG. The diameters of PETG fibres were lower than for rPETG, but they have a smoother surface and higher flexibility. Application of both types of fibres as interlayers in CFRP resulted in good quality lamines, but with lower flexural strength, while there was significantly improved ILSS in the case of fresh PETG fibres. SEM images of lamines after the ILSS test confirms that the application of thermoplastic polymer reduced the cracking area of CFRP.

Acknowledgements

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References