CHALLENGES IN DEVELOPING NYLON COMPOSITES COMMINGLED WITH DISCONTINUOUS RECYCLED CARBON FIBRE

Kok H. Wong*, Thomas A. Turner and Stephen J. Pickering

Polymer Composites Group, Division of Materials, Mechanics & Structures, Faculty of Engineering University of Nottingham, Nottingham, NG7 2RD, UK.
**kok.wong@nottingham.ac.uk*

Keywords: Recycling, carbon fibre, nylon, compression moulding

Abstract

This paper investigates the challenges associated with the development of a composite material containing commingled nylon fibres and recycled carbon fibres. Carbon fibre recycled from end-of-life composites is generally discontinuous and randomly orientated. A conversion process is required to improve its physical form to make it more readily applicable for use in the composites industry. Various conversion processes have been investigated in the past, including milling, papermaking type processes, carding and spinning. In this study, a papermaking process is used to produce random non-woven aligned commingled mats. The mats were compression moulded and their mechanical properties were measured. The interfacial adhesion between the fibre and matrix was assessed by analysis of fracture surfaces under scanning electron microscopy. The impact of fibre alignment on composite mechanical performance will be reported.

1 Introduction

Awareness of the need for carbon fibre recycling has become more widespread over the last decade and this has led to the establishment of several commercial scale carbon fibre recycling operations in the Europe and the North America [1]. Besides having comparable mechanical and adhesion properties to virgin fibre, the recycled carbon fibre can also provide potential cost saving for composite manufacture and thus to end users due to reduced energy consumption in the recycling process. However, unlike the virgin fibre, which is continuous and supplied in bobbins, fibre recycled from end-of-life products is discontinuous, filamentised and fluffy and this imposes difficulties in handling and processing. This has limited its usage to low-grade filler for thermal [2] and electrical conductivity [3]. An intermediate process to convert the sizing-free, discontinuous recycled fibre into a form that can be processed more easily is thus necessary. The approach proposed in this study is commingling recycled carbon fibre with thermoplastic fibre in a wet papermaking process to form a non-woven mat. Commingling brings both reinforcement and polymer fibres close together which will reduce the melt flow distance and should promote more complete resin impregnation and minimal void formation. In this study, a study into the effect of fibre alignment was also included with the aim of increasing the packing of the fibres so that the fibre reinforcing potential can be fully realised.

2 Experimentation

2.1 *Materials*

The thermoplastic matrix used in this study was a bi-component Grilon BA 3100 polyamide (PA) staple fibre of 6mm long, which was supplied by EMS-Chemie AG, Switzerland. The fibre sheath was made of PA6 and its core was PA66 and their weight ratio was 1:1. Melting temperature for the sheath and core was 220°C and 260°C respectively. Virgin carbon fibre was sourced from Toho Tenax Europe GmbH and of a grade of Tenax-A HTC124. The fibre was coated with water soluble sizing and had a length of 12mm, the fibre manufacturers properties are given in Table 1. Recycled carbon fibre was recovered from out-of-date prepreg rolls supplied by Cytec, UK, which comprised Toray T600SC carbon fibre in MTM28-2 epoxy resin. A fluidised bed process, developed at the University of Nottingham, was used to recycle the carbon composite [4]. Tensile properties of the recycled carbon fibre, which are shown in Table 1, were measured according to BS/ISO 11566 standard using a 5N load cell on 4mm gauge length samples. Table 1 contains also tensile properties of virgin T600SC fibre. The effect of fluidized bed process on the change in resulting fibre length preservation was analysed by measuring the dimension of the shredded prepregs and recycled fibre length using a digital camera for image capturing and an ImageJ software for image data processing. The difference in fibre length distribution is shown in Figure 1.

Fibre type	Diameter (μm)	Tensile modulus (GPa)	Tensile strength (MPa)	Number average length (mm)
Virgin HTC 124	7	225	4275	12
Virgin T600SC fibre	7.01±1.2	232±44	4098±873	4.12
Recycled T600SC fibre	7.11±1.0	223±36	2732±636	1.43

Table 1: Mechanical properties of the virgin and recycled carbon fires

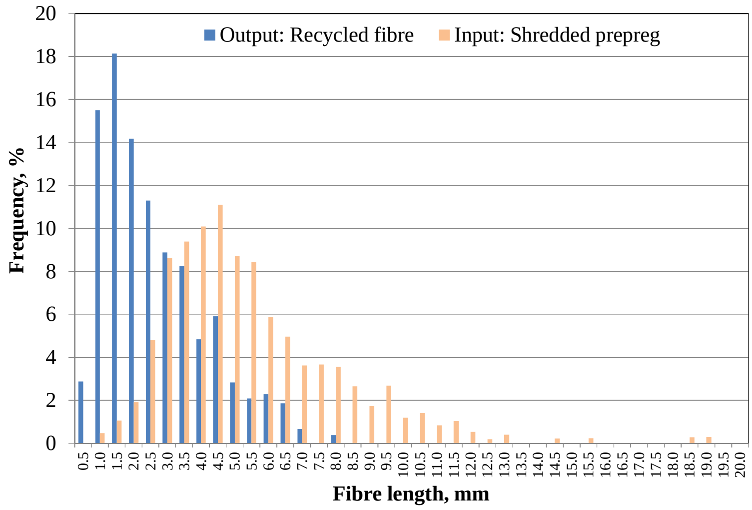


Figure 1: Fibre length distribution of the shredded composite waste and resulting recycled carbon fibre

2.2 *Manufacture of commingled mats*

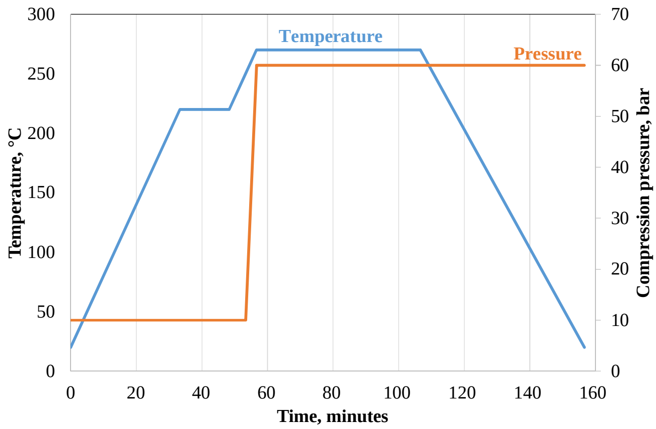
Commingled mats were produced via a wet papermaking process undertaken in Technical Fibre Products Ltd., UK. Carbon fibre and PA fibre were metered and dispersed in a white water to form a fibre suspension. The volume percentage ratio of carbon fibre to PA fibre in the suspension was 25:75. The liquid was filtered out during deposition of the suspension onto a moving mesh to form a wet mat. The mat was later subjected to binder application, drying and winding into a 600mm width roll. By altering the papermaking process parameters, orientation of the depositing fibres can be controlled to align them along the direction of mesh motion, which is generally known as the machine direction (MD). On the other hand, the term cross direction (CD) refers to a direction along the width of the roll. Four types of commingled mats were produced for this study, their details are given in Table 2.

Mat	Carbon fibre type	Fibre structure	Areal density, gsm	Designation
1	Virgin	Random	100	VR100
2	Virgin	Aligned	20	VA20
3	Recycled	Random	100	RR100
4	Recycled	Aligned	20	RA20

Table 2: Material designation for the produced commingled mats

2.3 Manufacture of composite

Composite was manufactured via a compression moulding process. Commingled mats were cut to fit inside a picture frame with 5mm clearance along the edges. The number of mats required for the 20gsm and 100gsm samples were typically 110 and 15 layers respectively to fill up the mould cavity. Mats were stacked together along the machine direction and were sandwiched between the two steel tools. The tools were then processed according to the temperature and



pressure profile shown Figure 2.

Figure 2: Temperature and moulding profiles for compression moulding

2.4 Composite mechanical properties

Composite tensile strength and stiffness were measured using an Instron 5969 universal testing machine according to BS EN ISO 527-4 (1997). Tensile force was applied along the machine direction of the composite specimens. Strain developed during the test was recorded using an extensometer with a gauge length of 25mm, which was attached to the centre of the test specimen. A 50kN load cell was adopted, and the crosshead speed was set to 2mm/min. At least 5 specimens were tested for each material variant.

Composite flexural properties were measured according to BS EN ISO 178 (2003) using a Hounsfield HTE S-series H25KS/05 universal testing machine. Composites were tested along and normal to the machine direction for anisotropic properties determination. A 1kN load cell was attached to the crosshead, and a test speed of 1mm/min was applied. 6 specimens were tested for each sample.

2.5 Composite void content measurement

Composite was casted with a catalysed polyester resin in a 40mm diameter pot. Upon complete curing, the cast sample was sectioned, polished and analysed using an optical microscope. A frame grabber was used to take digital images of the sample, which were later stored in a PC for void content measurement. The measurement was performed using ImageJ software.

2.6 Interfacial adhesion property

Morphology of the composite fracture surface after tensile test was studied to analyse the interfacial adhesion property between the carbon fibres and nylon matrix. This was performed using a JSM6400 scanning electron microscope with silver-coated surfaces.

3 Results and discussion

3.1 Moulding process optimization

The matrix used in this study was a bi-component polyamide fibre. The fibre has two distinct melting temperatures for its sheath and core structures, which is 220°C and 260°C respectively. A series of tests were performed on the VR100 mat in order to identify an optimised moulding condition for producing a composite with minimum void content. The first tests were performed at temperatures between 220°C and 290°C and with a moulding pressure of 5 bar being applied for the whole heating, holding and cooling periods. However, it was later found that the 290°C temperature was too high as significant thermal degradation was observed from the moulded part and thus it was not included in the subsequent study. Figure 3 shows that the composite void content stays at around 2.5% for temperature up to 250°C before which it increases rapidly to 20% at 270°C. An inspection to the moulding tools found that the amount of resin left on the edges of the moulding area increased with moulding temperature. As the sheath material was made of PA6 with a melting point of 220°C, the increase in moulding temperature would increase flow of the molten PA6 due to reduction in its viscosity, thus encouraging more polymer melt being squeezed out from the mats, which aggravated the problem of high void content. Although low void content was found from lower moulding temperatures, this imposed a problem in melting the PA66 core structure, as at 220°C, the core filaments can still be seen clearly in micrographs shown in Figure 4. At 230°C, fusion of adjacent core filaments commenced and only fully melted at 270°C. The presence of the PA66 core filaments was found to have a negative impact to the composite tensile properties as shown in Figure 5. Composite tensile strength was slightly promoted with increasing moulding temperature. A more evident improvement in tensile modulus, however, can be seen in Figure 5, with the composite moulded at 270°C showing the highest tensile performance. This suggested that void content had less detrimental effect on tensile properties compared to the presence of un-melted PA66 core filaments. As a result, a moulding temperature of 270°C was selected and a moulding profile, as given in Figure 2, was followed for the production of other commingled mats listed in Table 2. To ensure that sufficient moulding pressure was applied to achieve the required fibre volume content, the material stack was placed inside a picture frame of about 2mm thick and the moulding pressure was increased from 5 bar to 60bar. In addition, an extra of 5% by weight of mat was included to the material stack in order to compensate for potential resin lost during the moulding process.

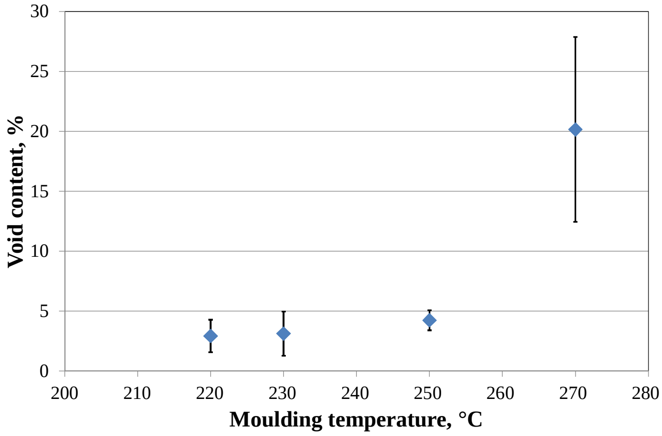


Figure 3: Void content of composites made of VR100 mat

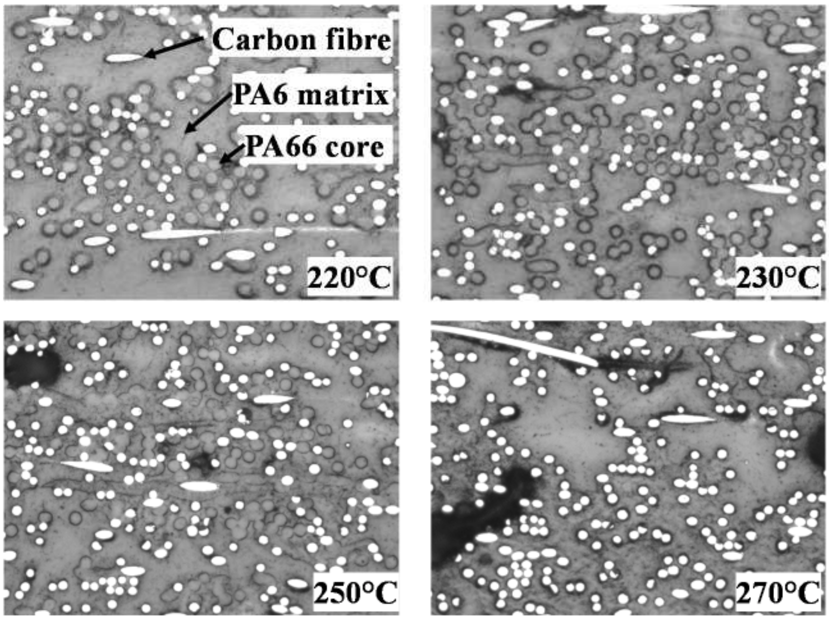


Figure 4: Micrograph of composite moulded at different temperatures (x5 magnification)

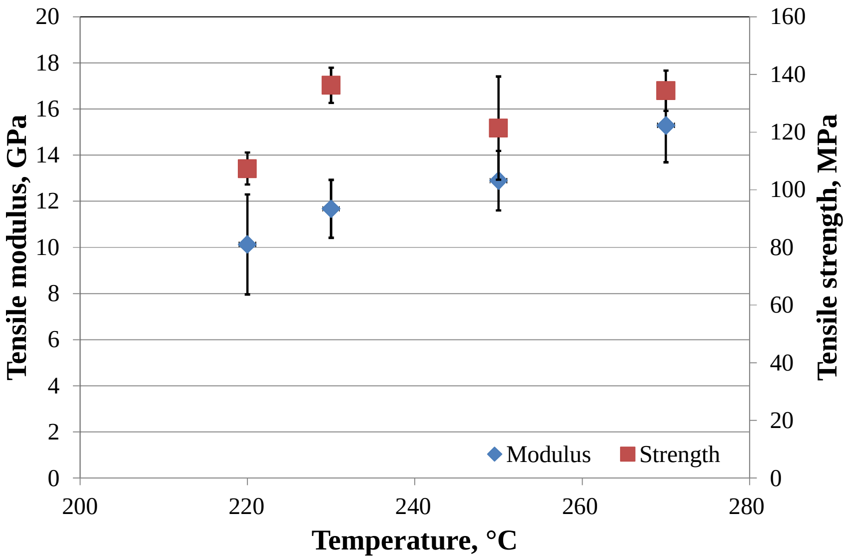


Figure 5: Relationship between moulding temperature and composite mechanical tensile properties

3.2 Mechanical performance of commingled composites

3.2.1 Composite tensile properties

The values of tensile properties of the four commingled composites are compared in a cluster column chart shown in Figure 6. The tensile properties were measured along the machine direction of the composites. The chart reveals that the VR100 composite has a higher tensile performance compared to the data obtained from the pre-optimised moulding condition as discussed in the Section 3.1, this positive result suggested a reduction in void content. However, in a comparison between VR100 and VA20 composites, no significant benefit was observed from the aligned structure of the VA20 sample. Fibre architecture of the VA20 mat was later analysed using a microscope and a sample image is given in Figure 7. Unlike the straight carbon fibre, the nylon fibre is curly and elastic and thus prevents a tightly packed structure. As a result, the aligning efficiency of the papermaking process is largely compromised. A more detailed analysis of the anisotropic property of the composite is covered in Section 3.2.2.

The tensile strength of the composites containing recycled carbon fibre is lower. A study on interfacial bonding between the fibre and polymer matrix was performed by observing SEM images taken from tensile-fractured cross section of the composites. According to Figure 8, it can be seen that despite some evidences of fibre pull-out, both virgin and recycled fibres show similar adhesion to the PA matrix. As a result, it is suggested that the reduction of composite strength can be attributed to the use of the recycled carbon fibre, which has a lower strength and shorter average fibre length compared to the virgin counterparts. Other possible factor including the presence of undispersed fibre bundles in the commingled mats, as shown in Figure 9. The bundles might act as stress concentrators that promote earlier composite failure. However, more tests are required to understand the reason for a higher reduction in composite modulus, in particular for the RA20 mat, which has the lowest tensile performance.

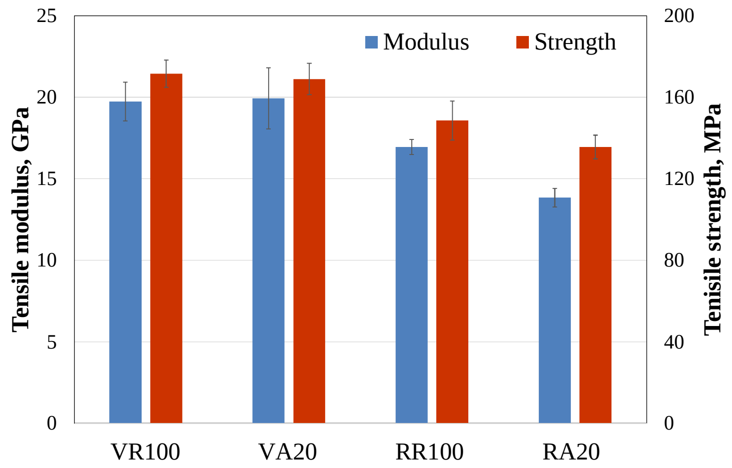


Figure 6: Values of tensile modulus and strength for four different commingled composites

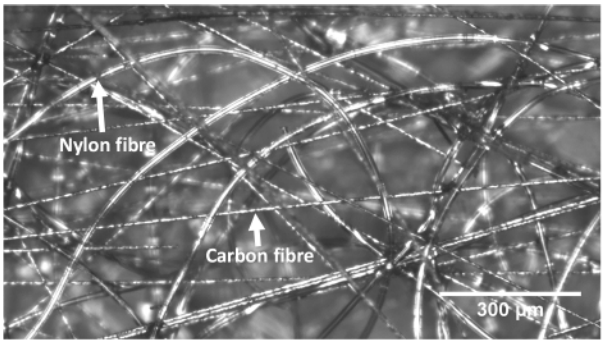


Figure 7: Micrographic image of the VA20 mat

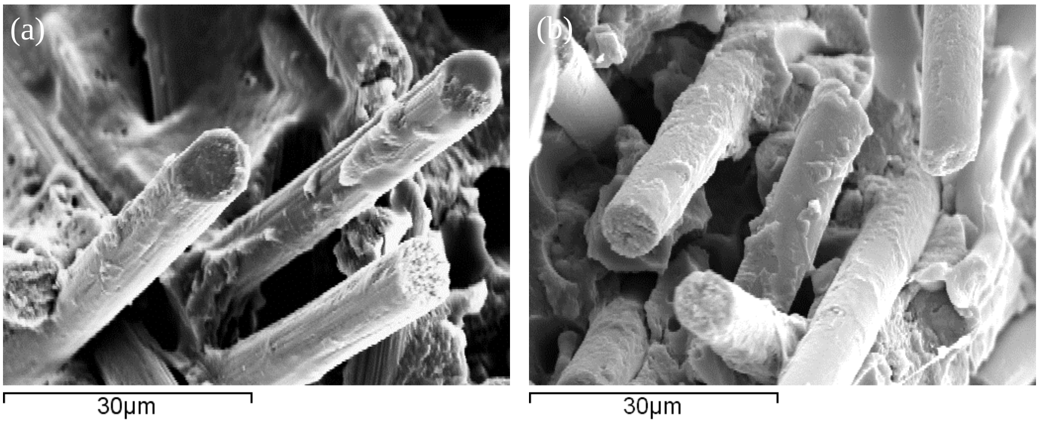


Figure 8: SEM images of tensile-fractured composite containing (a) VR100 and (b) RR100 mat

Figure 9: Undispersed fibre bundles in the RA20 mat

3.2.2 Composite flexural properties

Flexural properties of the commingled composites, which were measured along and normal to the mat's machine direction, are plotted in Figure 10. Anisotropic flexural properties are evidently shown, even for the random structures, such as the VR100 and RR100 mats. This concludes that the process used in this study does not fabricate mat with a fully random structure for the employed processing conditions. From Figure 10(c), a very similar strength ratio was obtained from the four mats, suggesting they shared a very similar fibre architecture i.e. fibres were distributed preferentially along the machine flow direction. More process optimization is required to produce a truly random mat. In general, Figure 10 shows that the recycled carbon fibre is more efficient in strengthening composite flexural properties as compared to tensile properties, depicted in Figure 6.

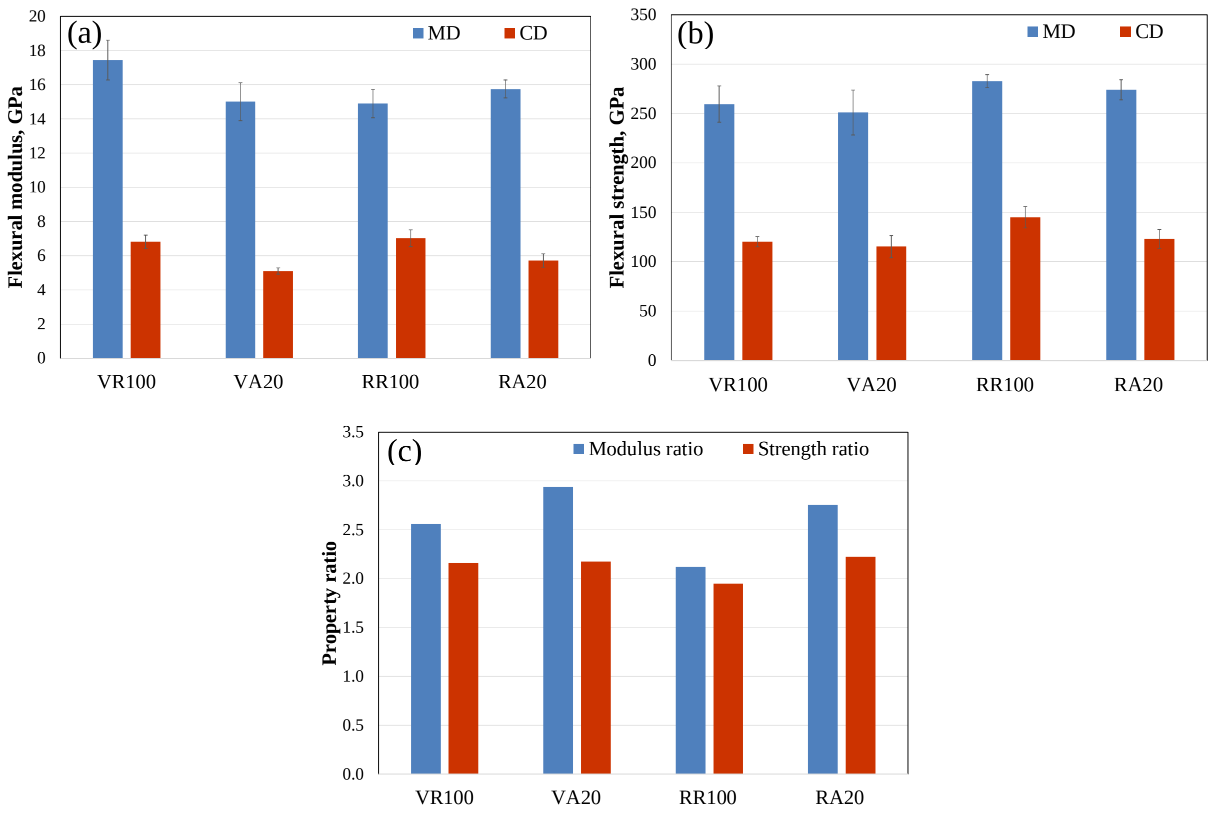


Figure 10: Values of tensile modulus and strength for four different commingled materials

4 Conclusions

The work performed has shown that the selected bi-component polyamide system was not suitable for the production of commingled mat due to its two distinct melting temperatures which complicated the control of resin impregnation at the melting stage. Single phase thermoplastic fibre should be used and curly physical form should be avoided if fibre alignment is required for achieving close-packed structure. Shorter input thermoplastic fibre length is preferable to provide higher alignment result. Nevertheless, the work demonstrated that the tensile, flexural and adhesion properties of composites reinforced with the recycled carbon fibre were comparable to the virgin fibre.

Acknowledgements

The authors wish to acknowledge financial support from the EPSRC Centre for Innovative Manufacturing in Composites.

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