Extrusion- and pultrusion-compounded carbon fibre reinforced polyamide 6,6 composites: Impact properties of injection moulded specimens

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ABSTRACT Processing of discontinuous fibre reinforced thermoplastic composites normally involves compounding and moulding, whereby fibre breakage problem could not be avoided. Pultrusion compounding technique has proven to improve tensile and fibre length characteristics of injection moulded specimens over extrusion technique. In this work, both pultrusion and extrusion techniques were employed to compound carbon fibre with polyamide 6,6 matrix, followed by injection moulding and testing for their impact properties. The values of G_c and K_c are increased with increase in fibre volume fraction. These values are also increased with increase in fibre length at lower fibre loadings (V_f of 0.20 and 0.21) but decreased with an increase in fibre length at higher fibre loadings (V_f of 0.31 and 0.32).

ABSTRAK Pemerosesan komposit termoplastik diperkuat gentian tak selanjar biasanya melibatkan penyebatian dan pengacuan; dan masalah pemutusan gentian tidak dapat dielakkan. Kaedah penyebatian pultrusi terbukti memberi kebaikan kepada sifat kekuatan regangan dan panjang gentian spesimen acuan suntikan dibandingkan dengan kaedah ekstrusi. Dalam kerja ini, kedua-dua kaedah pultrusi dan ekstrusi digunakan untuk penyebatian gentian karbon dengan matrik poliamida 6,6, diikuti dengan pengacuan suntikan dan seterusnya diuji untuk sifat impek. Nilai G_c dan K_c bertambah dengan bertambahnya pecahan isipadu gentian. Nilai-nilai ini juga bertambah dengan bertambahnya panjang gentian pada kandungan gentian rendah (V_f 0.20 dan 0.21) tetapi berkurang dengan bertambahnya panjang gentian pada kandungan gentian tinggi (V_f 0.31 dan 0.32).

(Processing, extrusion, pultrusion, impact properties, $G_{\text{\tiny c}}$ and $K_{\text{\tiny c}})$

INTRODUCTION

Polymer matrix composite (PMC) materials are becoming more acceptable in industrial applications, such as aircraft, automobile, transport industries, etc. Compared to more conventional engineering materials, such as metals and alloys, polymer matrix composite materials has added advantages in term of high specific modulus and high specific strength. Thus, less weight of materials is used to achieve the required properties [1].

In the processing of discontinuous fibre reinforced thermoplastic composites, moulding the dry blend of chopped fibres with polymer matrix has some disadvantages to the finished article such as wetting problem, variable

strength, shrinkage, poor surface finish, etc. Subsequently, the processes involve compounding and moulding. Short fibre reinforced thermoplastic composites are normally manufactured by extrusion process incorporated chopped strands/fibres into the plastic melt on a compounding extruder producing granules for injection moulding. Subsequent injection moulded product is called short fibre composite (SFC) [2]. The pultrusion process however, enables the reinforcement of continuous fibre bundles/tows with polymer matrix [3-5]. The pultruded product/pre-moulded composites were then pelletised to a required length for injection moulding. Subsequent injection moulded product is called long fibre composite (LFC) [6].

In this work, both techniques i.e. extrusion and pultrusion were employed in the compounding of polyamide 6,6 matrix with chopped- and continuous-carbon fibre, respectively. The extruded and pultruded composites were then pelletised and injection moulded, followed by property characterisation. In our previous publication [7], we reported that both tensile and fibre length properties of LFC are improved compared to the SFC counterpart at the same fibre loading. In the present paper, the work has been extended to study the fracture behaviour of SFCs and LFCs under impact loading compared to the tensile failure.

In designing materials against the possibilities of premature fracture, one of the most important properties of a material is its ability to withstand impact. With many metals and most plastics, the susceptibility of the material to shock loading is often the most critical parameter considered in material selection [8]. It has often been stated that impact strength is one of the least understood of the mechanical properties of polymers and composites in spite of its great technological importance. This is partly because impact strength is not as well defined a mechanical property as, for example, modulus in that its definition includes a description of how it is measured. This means that the use of a standard specimen shape, although necessary to compare different materials, causes a severe limitation on the amount of useful information obtained on these materials [9]. In this work, the relationship between the impact properties with compounding routes for composites manufacturing has been characterised and established.

EXPERIMENTAL

For the preparation of short fibre composite, the required amount (weight) of chopped carbon fibres (Toray® FT300B) with length of 6 mm and polyamide 6,6 matrix (Technyl® A216) were pre-blended using a Z-blade mixer for about twenty minutes. The blends were then passed through a twin-screw extruder with zone temperatures set at 250, 260, 265, 270 and 280 °C for zones 1, 2, 3, 4 and 5, respectively. The resulting strand extrudate was then pelletised for injection moulding.

long fibre composites preparation, compounding between the continuous carbon fibres with polyamide 6,6 matrix was carried out using novel experimental rig. Three continuous carbon fibre tows (total of 18,000 filaments) were spread and passed through the cross-head die of the extruder where impregnation with polymer melts occurred. For this purpose, a polyamide single-screw plastic extruder was used to melt the polymer. Temperature settings along the barrel of the extruder were 260, 270 and 300°C for the rear, centre and front zones, respectively. The impregnated continuous fibre composite was then cooled down by passing through a water bath, and then pelletised to a length of ca. 10 mm for injection moulding. The same grade of fibre and matrix as those for short fibre composite preparation were used for this compounding. Materials prepared are listed in (Table 1).

In the injection moulding process, a single gated double cavity, impact and tensile standard test bar mould was used in the moulding of the test specimens.

For the fibre extraction, samples from the central portion of injection moulded test pieces were cut, accurately weighed and placed in a long neck reaction flask. Formic acid was added to dissolve the polyamide matrix. The residual fibres were then collected by filtration using dried preweighed filter paper through a sintered glass crucible. The extracted fibres were then washed with distilled water, dried at 100° C in a vacuum oven for two hours, cooled and dried in desiccator and then weighed. Fibre volume fraction (V_f) and fibre length distribution (FLD) were then determined.

Impact test bars were notched at the centre on one edge to produce a single edge notch (SEN) impact test specimen. For each batch, specimens were notched with four different notch to depth ratios (a/D) i.e. 0.1, 0.2, 0.3 and 0.4. Throughout the test, a support span to depth ratio (S/D) was adjusted and maintained at 4. An instrumented falling weight impact tester was used for the impact testing. Measurements were made of the force required to break a notched test bar, supported at either end by striking it with a striker at a prescribed velocity at a point midway

between and on the opposite side of the two supports. The test was conducted at room

temperature.

Table 1. List of specimens and impact properties of injection moulded short- and long-carbon fibre composites.

Specimen	Fibre	V_{f}	G _c (kJm ⁻²)	K _c (MPa m ^{1/2})	Property index	
					G_{c}	K _c
SC21	short	0.21	6.98	8.37	std	std
LC20	long	0.20	7.67	9.58	1.15	1.2
SC31	short	0.31	8.85	10.86	std	std
LC32	long	0.32	8.06	10.26	0.88	0.92

Table 2. Fibre length characteristic of injection moulded short- and long-carbon fibre composites.

Specimen	$\mathbf{L}_{\mathbf{n}}$	L _w (mm)	Property index	
	(mm)		L _n	$L_{\rm w}$
SC21	0.25	0.34	std	std
LC20	0.57	0.88	2.39	2.72
SC31	0.22	0.29	std	Std
LC32	0.43	0.62	2.05	2.24

RESULTS AND DISCUSSION

Fibre volume fraction, V_f was calculated using the following equation:-

$$V_{f} = \frac{\frac{M_{f}}{\delta_{f}}}{\frac{M_{f}}{\delta_{f}} + \frac{M_{m}}{\delta_{m}}}$$
(1)

where M and δ are weight and density, respectively; and subscripts f and m refer to fibre and matrix, respectively. Long fibre composites were calculated to have V_f of 0.20 and 0.32, designated as LC20 and LC32, respectively. Short fibre composites with V_f of 0.21 and 0.31, were designated as SC21 and SC31, respectively. Results of this calculation are given in Table 1. With 1% different in V_f between SFC and LFC, they were in good agreement for the purpose of comparison.

Impact test results provide the values of fracture energy (W) and peak load (P) to break the specimens. The plots of these values against notch to depth ratios (a/D) are given in (Figures 1

and 2), respectively. Fracture energy and peak load are increased with increase in fibre volume fraction. At lower fibre loading, these properties are also increased with increase in fibre length. At higher fibre loading, however, W and P of long fibre composites are slightly lower compared to the short fibre composites. With regard to the effect of increasing notch to depth ratio (a/D), the fracture energy and peak load are decreased for all composites. This is expected due to the reduction in fracture area. With regard to the decrease in W and P of LFC compared to SFC (i.e. with an increase in fibre length), it is expected that, LFC to fail in a more brittle manner by fibre breakage mechanism.

The relationship between fracture energy (W), the critical strain energy release rate (G_c) and specimen geometry parameter (BD Φ) is given by:-

$$W = G_c BD\phi$$
 (2)

where B and D are the width and depth of the specimen, respectively. A correction factor, Φ is given by:-

$$\phi = \frac{1}{2} \left(\frac{a}{D} \right) + \frac{1}{18\pi} \left(\frac{S}{a} \right) \tag{3}$$

where a and S are notch depth (or crack length) and span of the specimens, respectively.

A plot of W against BD Φ (Figure 3) produced a straight line, where its slope is equal to the G_c of the materials.

The relationship between the critical stress intensity factor (K_c) with nominal fracture stress (σ) , geometry correction factor (Y) and crack length (a) is given by [8]:-

$$\sigma Y = K_c a^{-1/2} \tag{4}$$

In three-point bend test, σ is given by simple bending theory as:-

$$\sigma = \frac{6 \,\mathrm{PS}}{4 \,\mathrm{B} \,\mathrm{D}^2} \tag{5}$$

For the three-point bend test specimen, where S is equal to 4D, Y is given by:-

$$Y = 1.93 - 3.07 \left(\frac{a}{D}\right) + 14.53 \left(\frac{a}{D}\right)^{2}$$
$$- 25.11 \left(\frac{a}{D}\right)^{3} + 25.80 \left(\frac{a}{D}\right)^{4}$$
 (6)

A plot of σY against a^{-1/2} (Figure 4) produced a straight line, where its slope is equal to the K_c of the materials. This method of G_c and K_c determination has also been employed by Carling and Williams [10] in their work.

G_c and K_c values of these composites, determined by using the method described above, are tabulated in Table 1. Their property index (PI) is calculated using the following equation:-

$$PI = \frac{\frac{P_{c}}{V_{f}}}{\frac{P_{c,r}}{V_{f,r}}}$$
(7)

where $P_{c,r}$ and $V_{f,r}$ are respectively the property and fiber volume fraction of a reference composite, and; P_c and V_f are the corresponding property and fiber volume fraction of the composite from which a comparison is to be made.

Histograms of G_c and K_c values against specimen types are given in Figures 5 and 6, respectively. Both G_c and K_c values are increased with increase in fibre volume fraction. This is as expected since more fibre will be controlling the fracture behaviour with increase in fibre loading.

In previous work [7], it was reported that long fibre composites has an added advantage in retaining longer fibre length in injection moulded specimens. Their number average fibre length, L_n and weight average fibre length, Lw are tabulated in Table 2. L_n values of LFCs are increased by 139% and 172% at lower and higher fibre volume fraction, respectively; whereas L_w values of LFCs are increased by 105% and 124% at lower and higher fibre volume fraction, respectively. The values of W and P are increased with an increase in fibre length at lower V_f (LC20 compared to SC21) but decreased at higher V_f (LC32 compared to SC31). This is thought to be due to the effect of high V_f and longer fibres. At higher V_f, the existence of longer fibres further restricts matrix movement and these composites become more brittle. Upon fracture, composite tends to fail in a brittle manner by fibre breakage and contributing less energy.

In impact testing, too long fibres generally tend to break in more brittle manner by fibre breakage mechanism and contribute less fracture resistance to the composite compared to the shorter fibres that tend to break in more ductile manner by fibre pull-out mechanism and contribute more fracture resistance to the composite.

Crosby, et al. [11], Jones, et al. [12] and Gupta, et al. [13] also reported that an improvement in impact properties of glass fibre reinforced composites is associated with an increase in fibre length.

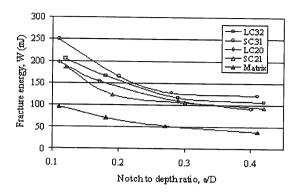


Figure 1. Variation of fracture energy with notch to depth ratio.

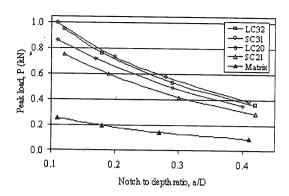


Figure 2. Variation of peak load with notch to depth ratio

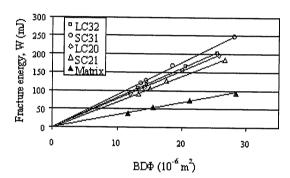


Figure 3. Variation of fracture energy with specimen geometry function.

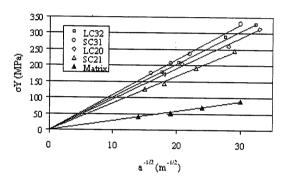


Figure 4. Variation of σ Y with a^{-12}

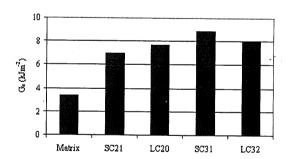


Figure 5. Gc of polymer matrix and short- and long- fiber composites.

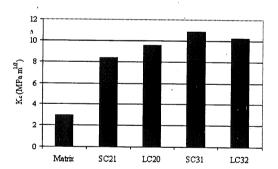


Figure 6. Kc of polymer matrix and short- and long- fiber composites.

CONCLUSION

Fracture energy and peak load of SFCs and LFCs are increased with an increase in fibre volume fraction.

At lower fibre volume fraction, fracture energy and peak load also increased with increase in fibre length; however, at higher fibre volume fraction, the opposite effect is seen.

Both fibre volume fraction and fibre length also affect $G_{\rm c}$ and $K_{\rm c}$ values in the same trend as they do on the fracture energy and peak load.

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