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Short- and long-glass fibre reinforced polyamide 6,6 composites: Fibre length characteristics of injection moulded specimens

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Abstract. Injection moulding of the dry blend of fibre and polymer matrix usually results in composite materials with poor physical properties. Therefore, the processing of fibre reinforced composite materials usually involved compounding and moulding. These processes, however, are associated with the problem of fibre breakage. In this work, short and long glass fibre reinforced polyamide 6,6 composites, prepared by extrusion and pultrusion compounding respectively, were injection moulded. Test pieces were then subjected to fibre length distribution characterisation. It was found that pultrusion compounded composites showed superior fibre characteristics compared to the extrusion compounded composites counterpart. These fibre length characteristics were also in agreement with the improved tensile strength and tensile modulus of long fibre composites over the short fibre composites.

Abstrak. Pengacuan suntikan campuran kering diantara gentian dan matrik, biasanya menghasilkan bahan komposit yang kurang baik sifat-sifatnya. Oleh itu, kebiasaannya pemerosesan bahan komposit diperkuat dengan gentian melibatkan mengkompaun and pengacuan. Proses ini, walaubagaimanapun menyebabkan pemutusan gentian. Dalam kerja ini, bahan komposit poliamida 6,6 diperkuat dengan gentian kaca pendek dan panjang yang dikompoun dengan peralatan ekstruder dan pultrusi masing-masingnya, dilakukan pengacuan suntikan. Pengujian dilakukan untuk menentukan taburan panjang gentian. Didapati komposit yang dikompoun dengan kaedah pultrusi menunjukkan kelebihan cirri gentian dibandingkan dengan komposit yang dikompaun dengan dengan kaedah ekstrusi yang setara. Ciri panjang gentian ini juga setara dengan peningkatan kekuatan regangan dan modulus Young yang ditunjukkan oleh komposit gentian panjang yang setara.

(pultrusion, extrusion, injection moulding, fiber length characteristics)

INTRODUCTION

Discontinuous short fibre reinforced thermoplastic composites have become attractive engineering materials since they offer improved performance over unreinforced materials at limited incremental cost [1]. In addition, these materials can also be processed by conventional means such as extrusion or injection moulding, as those for unreinforced materials [2].

In the processing of this composite, attempts to mould the dry blend of fibres and matrix granules usually result in some disadvantages to the moulded product or finished article. Consequently, the processing normally involved compounding in which the fibre was introduced into the polymer matrix, followed by moulding to the finished article. These processes lead to fibre degradation. In the case of short fibre composites (SFC), with the initial fibre length of 3-6 mm before the compounding, average fibre length in the injection moulded part is well below 1 mm. This relatively short fibre length retained in the final product limits the improvement achievable in the properties of the moulded part. However, with the development of processing technology called pultrusion [3,4], pre-moulded composite feed stock in the form of pellet with the length of up to 12 mm can be prepared. This long fibre composite (LFC) material is then subjected to only one stage of fibre degradation, during injection moulding.

The final composite properties are very much influenced by the microstructure of their constituents, such as fibre, matrix, fibre-matrix interface/phase, fibre volume fraction (V_f), etc. In this work, microstructure of composites was studied by means of characterising fibre length characteristics of the injection moulded specimens.

EXPERIMENTAL

Commercial materials used for the characterisation are Maranyl® A100 (unreinforced polyamide 6,6), Maranyl® A190 (short glass fibre reinforced polyamide 6,6 composite, 18% V_f) and Verton® RF7007 (long glass fibre reinforced polyamide 6,6 composite, 19% V_f).

Methods of injection moulding of specimens and fibre extraction from the composite specimen are as described previously [5]. The list of materials under investigation are given in Table 1.

Table 1. Specimens abbreviation and formulation.

Sample	V_f	Fibre	Description
S10	0.10	short	Maranyl® A190, diluted with Maranyl® A100
L10	0.10	long	Verton® RF7007, diluted with Maranyl® A100
S18	0.18	short	Maranyl® A190, used as received
L18	0.18	long	Verton® RF7007, diluted with Maranyl® A100

Table 2. L_n and L_w of injection moulded short- and long-fibre composites.

Specimen	L_n (mm)	L_w (mm)	Property index	
			L_n	L_w
S10	0.31	0.39	std	std
L10	0.63	1.06	2.03	2.72
S18	0.28	0.35	std	std
L18	0.58	1.02	2.07	2.91

std = standard

Table 3. Percentage of fibre with L less than 0.2, 0.4 and 0.6 mm of injection moulded short- and long-fibre composites.

Specimen	Fibre with length, L (%)			Property index		
	L < 0.2 mm	L < 0.4 mm	L < 0.6 mm	L < 0.2 mm	L < 0.4 mm	L < 0.6 mm
S10	23	76	95	std	std	std
L10	9	36	54	0.39	0.47	0.57
S18	27	81	97	std	std	std
L18	17	48	67	0.63	0.59	0.69

std = standard

Table 4. Fibre length (L) range and fibre aspect ratio (L/D) range of injection moulded short- and long-fibre composites.

Specimen	Fibre length, L range (mm)		Fibre aspect ratio, L/D range	
	$L < L_{1/2}$	$L > L_{1/2}$	$L < L_{1/2}$	$L > L_{1/2}$
S10	0.00 - 0.28	0.28 – 0.90	0 - 28	28 – 90
L10	0.00 - 0.48	0.48 – 1.80	0 - 28	28 - 106
S18	0.00 - 0.26	0.26 – 0.80	0 - 26	26 - 80
L18	0.00 - 0.42	0.42 – 1.70	0 - 25	25 - 100

Table 5. Fibre characteristics of injection moulded short- and long-fibre composites, computed for one arbitrary unit of fibre volume.

Property	Unit	LFC	SFC
Diameter		17	10
Length	arbitrary	1	2.89
Surface area	arbitrary	1	1.7
No. of fibre	arbitrary	1	6 [a]
			8 [b]

[a] : calculated using L_n

[b] : calculated using L_w

For determination of fibre length distribution (FLD), the extracted fibres [5] were immersed in a beaker containing a microscope glass slide, water and a small amount of detergent to reduce surface tension. In order to ensure uniform mixing of fibres, the solution was placed in an ultrasonic bath for a period of about 60 seconds. The solution was removed from the beaker using a pipette, leaving a small amount of solution with fibres on the glass slide. The slides with fibres on one side were dried in an oven. Lengths of at least five hundred fibres were measured and the histogram of percentage of fibre counts (frequency) against fibre length range was plotted.

RESULTS AND DISCUSSION

Short fibre composites (SFC) having V_f of 0.10 and 0.18 (designated as S10 and S18 respectively) which were calculated as described in [5], were analysed for fibre length distribution (FLD). Similar analysis was carried out for long fibre composites having V_f of 0.10 and 0.18 (designated as L10 and L18 respectively). Histograms of of these composites are given in Figures 1 and 2.

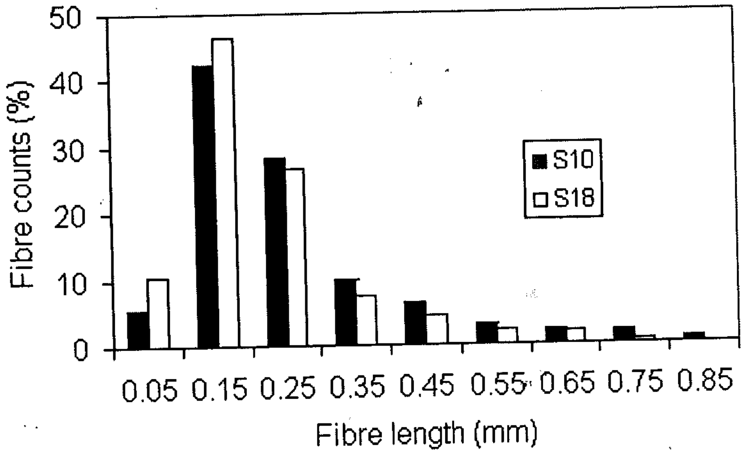

Figure 1. FLD of injection moulded short fibre composites.

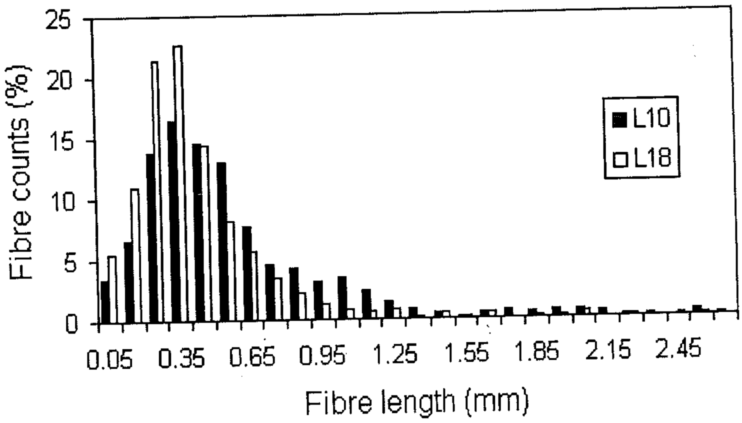

Figure 2. FLD of injection moulded long fibre composites.

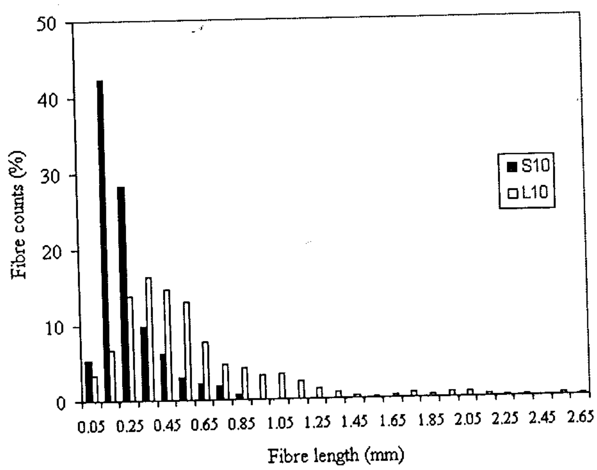

Figure 3. FLD of injection moulded short and long fibre composites.

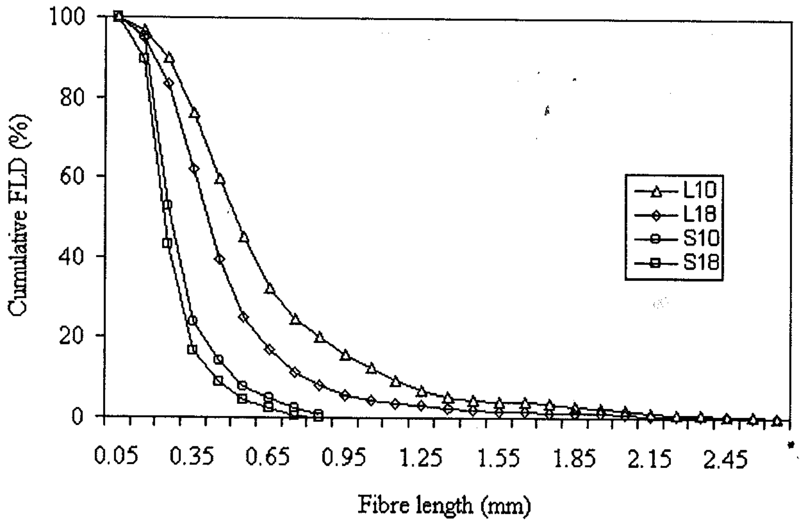

Figure 4. Cumulative FLD of injection moulded short and long fibre composites.

FLD histograms of both short and long fibre composites show quite normal distribution with long tail towards the longer fibre length. Both histograms also show that more fibre population with shorter length occurred in composites with higher V_f. In SFC, up to the fibre length of 0.2 mm, composite with higher V_f have more population of fibres compared to the composite with lower V_f, whereas in LFC, this trend occurs up to the fibre length of 0.3 mm. This means that composites with lower V_f will have more fibre population with longer length. This indicates that more fibre degradation takes place during compounding and moulding which could be due to fibre-fibre, fibre-metal and fibre-matrix friction. The same trend of behaviour was also reported by Denault *et al.* [6]. In explaining this, the authors quoted that high melt viscosity and increased tendency for high fibre contact are probably responsible for the higher fibre breakage in composites with a higher fibre volume fraction. Jones *et al.* [7] also reported that fibre degradation is due to an increase in fibre volume fraction of composites.

Figure 3 represents the effect of compounding route (extrusion vs pultrusion) on FLD of composite with the same V_f of 0.10. For short fibre composites, majority of the fibres have lengths between 0.1–0.5 mm compared to the long fibre composites with majority of the fibres are between 0.1–1.2 mm. This figure clearly shows the superiority of long fibre composites compared to the short fibre composite counterpart.

Figure 4 represents the cumulative FLD of short and long fibre composites at both V_f of 0.10 and 0.18. Despite more fibre degradation occurring during the processing of composite with higher V_f, it is interesting to note that cumulative FLD of LFC at V_f of 0.18 is still far better than that of SFC with V_f of 0.10. This behaviour further proves the advantage of long fibre composites in minimising fibre length degradation during moulding.

FLD data can also be explained from the statistical point of view by the number average fibre length, L_n that gives a measure of fibre ends density; and weight average fibre length, L_w that gives a greater emphasis to the proportion of long fibres in the distribution. These terms are given by equations (1) and (2) respectively:-

$$L_n = \Sigma(f_i L_i)/\Sigma f_i \quad (1)$$

$$L_w = \Sigma(f_i L_i^2)/\Sigma(f_i L_i) \quad (2)$$

where; f_i is the number of fibre count (frequency) of fibres of species i with length L_i. A mid-point of fibre length ranges, each at 0.1 mm interval was taken as an average value of fibre length, L_i in the calculation. From this calculation, L_n and L_w with their property index (PI) are given in Table 2. The percentage of fibres with L less than

0.2, 0.4 and 0.6 mm together with their property index (PI) are given in Table 3. The calculation of PI was carried out using the following equation:-

$$PI = (P_c/V_f)/(P_{c,r}/V_{f,r}) \quad (3)$$

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

In Table 2, L_n and L_w of LFC increased by 103% and 172% respectively at V_f of 0.10; and 107% and 191% respectively at V_f of 0.18. In Table 3, at any L below 0.6 mm, LFC have less percentage of fibre, indicating that there are more fibres were with L longer than 0.6 mm. For example at V_f of 0.10, SFC posseses only 5% fibre with L longer than 0.6 mm, whereas LFC still have 46%.

It was found that fibre diameter of SFC and LFC were 10 µm and 17 µm respectively [5]. From this, it can be computed that for one arbitrary unit of fibre volume, SFC has more fibre length, fibre surface area and number of fibres compared to their LFC counterpart with the same level of fibre loading. Results of this calculation are given in Table 5. For composites with the same fibre volume fractions, SFC posseses total fibre length, total fibre surface area and total number of fibre counts of 2.9, 1.7 and 6-8 units respectively compared to only one unit each for LFC. All these further prove the advantage of pultrusion compared to extrusion as compounding technique for the fibre reinforced thermoplastic composites.

When comparing two composites having the same fibre diameters, fibre length is a good indicator as point of reference for calculation. However, if comparisons were to be made between two composites with different fibre diameters, another factor, fibre aspect ratio (length/diameter, L/D) should be considered. This is due to, in tensile testing for example, the fact that the determining point of whether one particular fibre is going to break or pull-out upon fracture depends upon the interfacial shear strength (IFSS) between the fibre and matrix. This IFSS is very much dependent on the critical fibre length, L_c of composite. L_c is the minimum length that permits the fibre to contribute its full strength to the composite. Below this length, the fibre is too short to accumulate enough stress to be broken, and pulls out of the matrix instead. For this reason, fibre L/D range was also calculated. On cumulative FLD curve (Figure 4); at cumulative FLD of 50%, the equivalent fibre length was determined, termed as $L_{1/2}$. Aspect ratio, L/D range was determined for the two range of fibre counts, i.e. for the fibres with length, L of $0<L<L_{1/2}$ and $L_{1/2}\leq L\leq L_{max}$, where L_{max} is the maximum fibre length available. Results of these calculations are given in Table 4. It can be seen that, although the longest fibre available, L_{max} of LFC is about two times longer than that of SFC, its aspect ratio is about the same. This is due to the effect of larger diameter of fibre, 17 µm of LFC compared to 10 µm of short fibre composites. Therefore, the reinforcement efficiency is greatly reduced

CONCLUSION

- More fibre degradation has occurred during processing of fibre reinforced polymer composite materials as fibre volume fraction increased.
- Injection moulded specimens of pultrusion-compounded LFC, give superior fibre length characteristics compared to the extrusion compounded SFC of equivalent composition.
- Besides fibre length, fibre aspect ratio should also be considered in analysing composite properties when working with two or more composites reinforced with fibres having different diameter.

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