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Stiffness behaviour of injection moulded short glass fibre/impact modifier/polypropylene hybrid composites

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Abstract

The stiffness behaviour of injection moulded short glass fibre/impact modifier/polypropylene hybrid composites has been investigated in this work by theoretical predictions and experiments. Predictions from the self-consistent method were found to be in good agreement with test results for the impact modifier/polypropylene blends. By taking into account of the fibre orientation distributions in the skin and core layers, the values of Young's modulus for the skin and core layers were predicted by employing Eshelby's equivalent inclusion method and the average induced strain approach. The prediction of the values of Young's modulus for the whole sample was obtained by applying the simple mixture theory of laminated composites to the predicted results for the skin and core layers. Good correlation between predicted and experimental Young's modulus values were found. © 1999 Published by Elsevier Science S.A. All rights reserved.

Keywords: Stiffness behaviour; Glass fibre; Impact modifier; Hybrid composites

1. Introduction

Polypropylene (PP) has been used widely in a number of applications from economic and property considerations. One of the weaknesses of PP is its low toughness under low temperature or impact loading conditions. In order to improve the impact toughness of polypropylene, the most common method is to incorporate an elastomeric (impact modifier) phase into the PP matrix. Cavitation of the PP matrix around the impact modifier (IM) particles was observed to be the main toughening mechanism [1].

Although IM/PP blends have superior impact resistance to PP homopolymer, the stiffness and yield strength will generally be reduced [1,2]. The addition of short glass fibres (GF) into the IM/PP matrix will improve the stiffness and strength behaviour of the blends, whilst the IM in the matrix will contribute towards toughness. In this paper, a predictive model for the stiffness of IM/PP blends and GF/IM/PP hybrid composites will be derived and compared with experimental results.

A large number of theories, such as the Eshelby theory [3], the Mori–Tanaka method [4], and the self-consistent method [5] have been developed to predict the stiffness behaviour of particulate or fibre reinforced composite systems. In general, the modelling of composite with spherical inclusions is much simpler than that with short fibre reinforcements. This is because with short fibre reinforcements, both their aspect ratio and orientation have to be taken into account. Halpin and Pagano [6] considered a two-dimensional random fibre composite using the laminate theory. Christensen [7] and Chou and Nomura [8] applied the self-consistent approach to investigate the three-dimensional case by summing up the contribution to stiffness of all fibre orientations. Takao et al. [9] considered the interaction amongst fibres at different orientations by adopting the average induced strain method and the modified Eshelby theory.

In injection moulded short fibre composites, a three-layer (skin/core/skin) structure is usually developed [10,11]. Any predictive model must take into account the effect of fibre orientation and the layered structure of the injection moulded composites.

In this paper, the self-consistent method was first used to investigate the stiffness of IM/PP blends. Subsequently, the IM/PP blends were treated as an isotropic and homogeneous matrix reinforced by short glass fibres. The stiffness of the hybrid composites with different fibre orientation distribu-

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tions (corresponding to the skin and core layers of the injection moulded samples) were calculated following the method advanced by Takao et al. [9]. Finally, the stiffness of the injection moulded samples was calculated using laminate theory.

2. Experimental details

A summary of the materials used in this work to prepare the blends and hybrids is shown in Table 1, all of the materials being supplied by the Himont Technical Centre in Hong Kong. Hi-fax RA061, which is a polypropylene/polyethylene (PP/PE) copolymer, was used as the impact modifier (IM).

In the preparation of the various blends and hybrids, the pellets were mixed in the appropriate ratio so as to achieve the required IM, GF, and PP homopolymer concentration. The mixed pellets were subsequently compounded using a Brabender twin-screw compounder at a barrel temperature of 220°C.

A summary of the composition of the blends and hybrids as well as their designation is given in Table 2. The volume fraction for each constituent phase V_i (where i=PP, IM or GF) are related to the weight fraction W_i by

$$V_i=\frac{W_i/\rho_i}{(W_{\mathrm{PP}}/\rho_{\mathrm{PP}})+(W_{\mathrm{IM}}/\rho_{\mathrm{IM}})+(W_{\mathrm{GF}}/\rho_{\mathrm{GF}})},\qquad(1)$$

where the densities, ρ_i, for PP, IM and GF were taken as 0.90, 0.88 and 2.69 g/cm^3, respectively. In the hybrids, the GF volume fraction was kept at about 15%.

Table 1
Summary of the materials used to prepare the blends and hybrids

Material code	Description
Pro-fax 6331	PP homopolymer
Hi-fax RA 061	PP/PE copolymer
Hi-glass PF062-2	20 wt% glass fibre reinforced PP
Hi-glass PF062-3	30 wt% glass fibre reinforced PP
Hi-glass PF062-4	40 wt% glass fibre reinforced PP
Hi-glass PF062-5	50 wt% glass fibre reinforced PP

Table 2
Compositions of the IM/PP blends and IM/GF/PP hybrids used in this work

Designation of the blends and hybrids used in this work	Volume % of PP homopolymer	Volume % of impact modifier	Volume % of glass fibres
PP	100	0	0
9IM	91.0	9.0	0
17IM	83.4	16.6	0
30IM	70.8	29.2	0
0GF	84.8	0	15.2
9GF	75.8	9.0	15.2
17GF	68.2	16.6	15.2
30GF	55.6	29.2	15.2

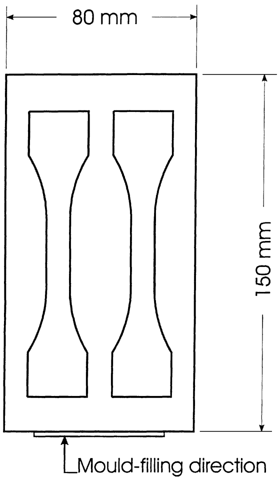

Fig. 1. Schematic diagram showing the injection moulded plaque and the location of the tensile bars.

The blended extrudates were pelletised and injection moulded into plaques of dimensions 6×80×150 mm^3 using a Chen Hsong Mark II-C injection moulding machine. A schematic diagram showing the injection moulded plaques with the location of the tensile specimens is shown in Fig. 1. Due to the skin–core structure and the fibre orientation effect [10,11], a complicated microstructure is expected for the hybrid samples. It is therefore important to control the position and orientation of the specimens. In this work, the longitudinal direction of the specimens was parallel to the melt flow direction (MFD).

For the hybrid samples, the IM and PP phases were burnt off in a laboratory furnace. The remaining short glass fibre bundles were dispersed in water with the aid of an ultrasonic bath. The dispersed fibres were collected and dried on a filter paper. The lengths of 1000 fibres for each type of hybrids were measured using an image analyser. It was observed that the average fibre lengths in all of the hybrids were approximately 327 μm, whilst the fibre diameters were about 12 μm.

Tensile tests were carried out using an Instron 4206 tensile tester at a temperature of about 20°C. The cross-head speed was 5 mm/min. A clip-on extensometer was used to measure the longitudinal strain, which allowed an accurate determination of the value of Young's modulus.

3. Theoretical considerations

The formulation to predict the value of Young's modulus of GF/IM/PP hybrid composites can be separated into a number of steps. Firstly, a predictive model for the Young's modulus of IM/PP blends will be developed. Subsequently, the hybrids will be treated as composites consisting of an isotropic and homogeneous IM/PP matrix reinforced by

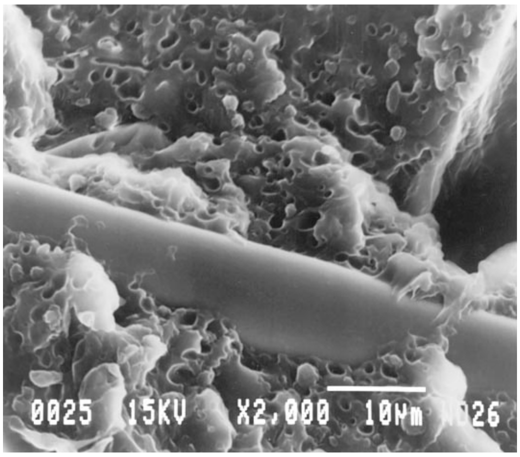

Fig. 2. The fracture surface of a hybrid sample.

short glass fibres. The stiffness of the hybrid composites with different fibre orientation distributions (corresponding to the skin and core layers of the injection moulded samples) were calculated following the method advanced by Takao et al. [9]. Finally, the stiffness of the injection moulded samples was calculated using laminate theory.

3.1. Prediction of Young's modulus of IM/PP blends

Fig. 2 shows the fracture surface of a hybrid composite. It can be seen that the IM domains were essentially spherical in the PP matrix. A key problem in the prediction of Young's modulus for in a two-phase composite is to determine the stress distributions in the two phases. The Eshelby theory [3] is a useful tool for this purpose. As an extension to the Eshelby theory for high filler volume fraction and the subsequent interaction between fillers, the self-consistent method can be used.

When a blend is subjected to an external loading, the stress distribution in the filler and matrix phase is:

$$\sigma^{\mathrm{IM}} = \left(\frac{\underline{C}_{\mathrm{IM}}}{\underline{S}\,\underline{C}_{\mathrm{IM}} + (\underline{I} - \underline{S})\underline{C}_{\mathrm{blend}}}\right)\sigma^{\mathrm{A}}, \tag{2}$$

$$\sigma^{\mathrm{PP}} = \left(\frac{\underline{C}_{\mathrm{PP}}}{\underline{S}\,\underline{C}_{\mathrm{PP}} + (\underline{I} - \underline{S})\underline{C}_{\mathrm{blend}}}\right)\sigma^{\mathrm{A}}, \tag{3}$$

where $\underline{C}_{\mathrm{IM}}$, $\underline{C}_{\mathrm{PP}}$ and $\underline{C}_{\mathrm{blend}}$ are the elastic constants of the IM, PP and the IM/PP blend, respectively. $\underline{S}$ is the Eshelby tensor. σ^{IM} and σ^{pp} denote the stresses in the IM and PP matrix, and σ^{A} is the loading applied to the blend.

Based on stress equilibrium, the following equation can be obtained:

$$V_{\mathrm{PP}}\sigma^{\mathrm{PP}} + V_{\mathrm{IM}}\sigma^{\mathrm{IM}} = \sigma^{\mathrm{A}}. \tag{4}$$

By substituting Eqs. (2) and (3) into Eq. (4), and assuming that the fillers are spherical in shape, the following equation is obtained:

$$V_{\mathrm{PP}}\left(\frac{15E_{\mathrm{PP}}}{7E_{\mathrm{PP}} + 8E_{\mathrm{blend}}}\right) + V_{\mathrm{IM}}\left(\frac{15E_{\mathrm{IM}}}{7E_{\mathrm{IM}} + 8E_{\mathrm{blend}}}\right) = 1. \tag{5}$$

The value of Young's modulus for the IM/PP blends (E_{blend}) can be obtained by solving Eq. (5).

3.2. Prediction of Young's modulus of GF/IM/PP hybrid composites

Once the Young's modulus for the IM/PP blends has been determined from Eq. (5), the blends are treated as a homogeneous matrix reinforced by short glass fibres. The problem is now reduced to the determination of Young's modulus for a short fibre reinforced composite. Takao et al. [8] has carried out an analysis on the effective longitudinal Young's modulus of misoriented short fibre reinforced composites, the analysis being based on the Eshelby equivalent inclusion method and the average induced strain approach. The approach is adopted in the present analysis to account for the fibre reinforcement effect.

In order to obtain an accurate prediction of Young's modulus of the hybrids, the microstructural details of the composite have to be taken into account. It is known that injection moulded short fibre reinforced composites will have a multi-layered structure. In general, a three-layer structure will be formed, with the fibres in the surface layers (top and bottom) being highly oriented parallel to the melt flow direction. In the core layer, the fibres are oriented randomly. An orientation density function is used to describe the fibre orientation in the skin and core layers of the hybrid composites, which is

$$g(\theta) = \frac{\sin\theta}{1 - \cos\alpha}, \quad 0 \le \theta \le \alpha, \tag{6}$$

where α defines the maximum fibre orientation in the respective layers. It is expected that the values of α will depend on the viscosity of the polymer matrix as well as on the injection moulding conditions.

By employing the Eshelby equivalent inclusion method and the average induced strain approach, Takao et al. [9] derived the following expression for the effective longitudinal Young's modulus E_{hybrid} of a short fibre composite

$$\frac{E_{\mathrm{hybrid}}}{E_{\mathrm{blend}}} = \frac{1}{1 + (E_{\mathrm{blend}}/\sigma_0)(1/V_{\mathrm{D}})\int_{\Omega} e_{33}^{*}\mathrm{d}V}, \tag{7}$$

where E_{blend} is the Young's modulus of the matrix, σ_0 is the applied tensile stress, and e_{33}^{*} is the normal Eigenstrain along the loading direction. The volume integral is evaluated taking into account the fibre orientation density function $g(\theta)$ given in Eq. (6). In Eq. (7), E_{blend} is calculated using Eq. (5) with V_{IM} and V_{PP} calculated from the following equations:

$$V_{\mathrm{IM}}=\frac{\text{Volume \% of impact modifier}}{\text{Volume \% of PP}+\text{Volume \% of impact modifier}}, \tag{8}$$

$$V_{\mathrm{PP}}=\frac{\text{Volume \% of PP}}{\text{Volume \% of PP}+\text{Volume \% of impact modifier}}, \tag{9}$$

where the Volume % of PP and the Volume % of impact modifier are given in Table 2 for the various hybrid samples.

Using Eq. (7), the Young's modulus for the skin (E_{skin}) and core (E_{core}) layers of the hybrid samples can be calculated. The effective longitudinal Young's modulus (E_{C}) for the whole of the hybrids can be calculated from the simple mixture theory of laminated composites

$$E_{\mathrm{C}}=\left(\frac{W_{\mathrm{skin}}}{W_{\mathrm{total}}}\right)E_{\mathrm{skin}}+\left(\frac{W_{\mathrm{core}}}{W_{\mathrm{total}}}\right)E_{\mathrm{core}}, \tag{10}$$

where W_{skin} is the total thickness of the two skin layers, W_{core} the thickness of the core layer, and W_{total} is the total thickness of the tensile samples.

4. Results and discussions

Fig. 2 shows a SEM micrograph of a tensile-fractured hybrid sample. It can be seen that the glass fibre diameter is much larger than the IM particle size. From microscopic examination, the values of α as mentioned in Eq. (6) was found to be 30° and 90° for the skin and core layers, respectively.

The variation of Young's modulus for the IM/PP blends (E_{blend}) against IM vol% predicted using Eq. (5) is shown in Fig. 3. The experimental values are also plotted and can be seen to be in good agreement with theory. The values for E_{PP} and E_{IM} used in Eq. (5) are given in Table 3. Both experiment and Eq. (5) predict a decrease in Young's modulus with increasing IM content. This is expected, as the IM used has a lower Young's modulus than that of PP (see Table 3).

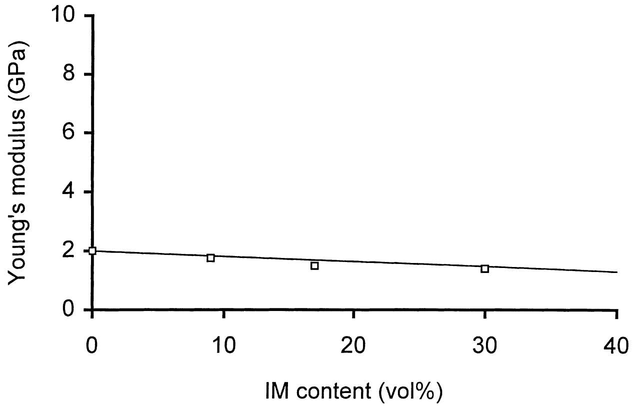

Fig. 3. Variation of Young's modulus with IM content for the IM/PP blends. The solid line shows prediction obtained using Eq. (5), whilst the experimental values are represented by □.

Table 3
Young's modulus for PP, IM and glass fibre used in the theoretical prediction

Material	Polypropylene	Impact modifier	Glass fibre
Young's modulus (GPa)	2.0	0.8	70

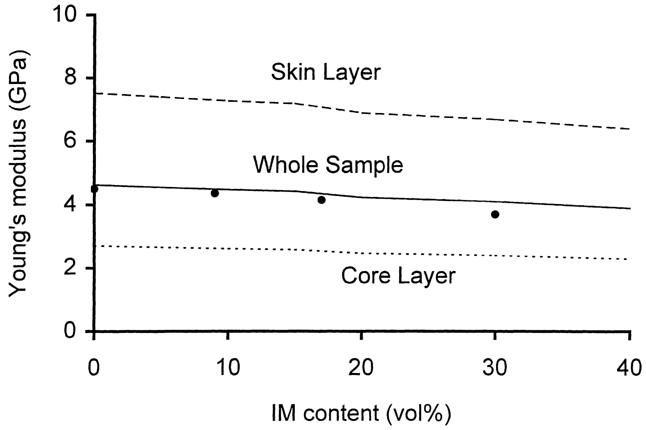

Fig. 4. Variation of Young's modulus with IM content for the IM/GF/PP hybrids, the experimental values being represented by •.

For the IM/GF/PP hybrids, prediction (using Eqs. (6) and (7)) of the variation of Young's modulus with IM content for the core layer is shown in Fig. 4. In the prediction, the α value is taken to be 90°. The prediction of the variation of the Young's modulus with IM content for the skin layer is also shown in this figure, with α equal to 30°. It can be seen that the predicted Young's modulus for the skin layers is significantly greater than that for the core layers. For both the skin and core layer samples, Young's modulus decreases with increasing IM content.

Once the Young's moduli for the skin and core layers are determined, Young's modulus for the whole samples (E_{C}) can be determined from Eq. (10). As expected, due to the rule-of-mixture nature of Eq. (10), E_{C} lies between the prediction for the skin and core layer samples. Similar to the skin and core layer samples, Young's modulus for the whole samples decreased mildly with increasing IM content.

5. Conclusions

The stiffness behaviour of injection moulded short glass fibre/impact modifier/polypropylene hybrid composites has been investigated in this work. Microscopic examination showed that a skin/core/skin structure exists in the hybrid samples. In the skin layers the fibres tend to align parallel to the melt flow direction. In the core layer the fibres take up a more random orientation distribution. Predictions from the self-consistent method were found to be in good agreement with test results for the IM/PP blends. By taking into account of the fibre orientation distributions in the skin and core layers the values of Young's modulus for the skin and core layers were predicted by employing the Eshelby equivalent inclusion method and the average induced strain approach. Prediction of Young's modulus for the whole samples were obtained by using the simple mixture theory of laminated composites. Good correlation between prediction and experiment was seen.

Acknowledgements

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