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Interfacial component of glass fiber in ternary composites of GF/PC/PP: effect of the preferential encapsulation of glass fiber

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Abstract

We investigated the encapsulation phenomena of glass fibers for a ternary composite system of glass fiber, polycarbonate and polypropylene by impregnating fibers with a selected resin using a newly developed process. On the basis of our observations of the morphology and mechanical properties, we report that viscosity ratio may very likely play an important role in the final interfacial structure of this system and that the improved wetting of fibers by preferential encapsulation would be a good approach in cases where the combinations of the resins or processing conditions lead to the similar relative melt viscosity of the component resins. © 2000 Published by Elsevier Science Ltd.

Keywords: Interfacial structure; Glass fiber/polycarbonate/polypropylene; Preferential encapsulation

The phase structure of polymer composites has a great influence on their behavior [1–3]. The composition dependence of tensile and impact properties always reflect the prevailing structure, which is determined by the interaction of the components [1]. Mutual miscibility and interfacial tension and the components' viscosity ratio are the crucial factors determining structure and properties of multi-phase systems [4]. For ternary reinforcing immiscible blend systems, widely differing structures were observed ranging from the separate dispersion of the filler within both polymeric components to the encapsulated filler within either the matrix or the dispersed phase and the prediction of properties was more complicated [2–4]. Earlier studies on such ternary systems have produced contradictory results. In some studies, separate dispersion of the components was observed even when encapsulation was thermodynamically favored owing to the use of compatibilized component, while in others the encapsulation of fillers was reported even though the kinetic barrier to displacement of the component appeared to be too high [4–6].

Recently, we investigated the encapsulation phenomena of glass fibers by adding the components sequentially, rather than simultaneously [7]. The glass fiber was preimpregnated with one of the component using a newly developed process to preparing a continuous fiber reinforced thermoplastic composites or prepregs with the fiber bundles cut to any desired length [8]. It was found that the ternary system of the GF/polyamide (PA) + polypropylene (PP), in which the polyamide phase persisted to be preferentially located surrounding the GF, were clearly superior to those of the GF/PP + PA as far as the tensile strength and impact properties were concerned. In samples designated GF/X + Y, X refers to the impregnation resin of GFs and Y represents the matrix resin of the ternary systems mixed with the impregnated GFs(e.g. GF/PA + PP indicates the ternary composite containing a PP matrix and the GF impregnated by polyamide). Our observation of the mechanical properties enhanced by the preferential encapsulation is contrary to the results for a similar blend system reported by Benderly et al. [2,3,5]. They concluded that variations in glass surface treatment, blending time, and order of addition did not affect the rapid encapsulation of glass fiber by PA. Accordingly, further studies are required to isolate practical implications of the preferential encapsulation of GF with a selected resin. In this paper, we investigated the effect of viscosity ratio of the blend components along with the interfacial properties on the persistence of pre-encapsulated resin. The ternary system of GF/polycarbonate (PC)/PP was chosen to achieve better control of the viscosity ratio of the components.

We used unmodified PP(Topilene J700, Hyosung T and C) and PC(Trirex 3022 IR, Samyang co.) as blend resins. First, we measured the melt viscosity of PP and PC as a function of shear rate at various temperatures. Fig. 1 shows melt flow curves of the resins obtained with a capillary rheometer at 260°C. The capillary used had a length/

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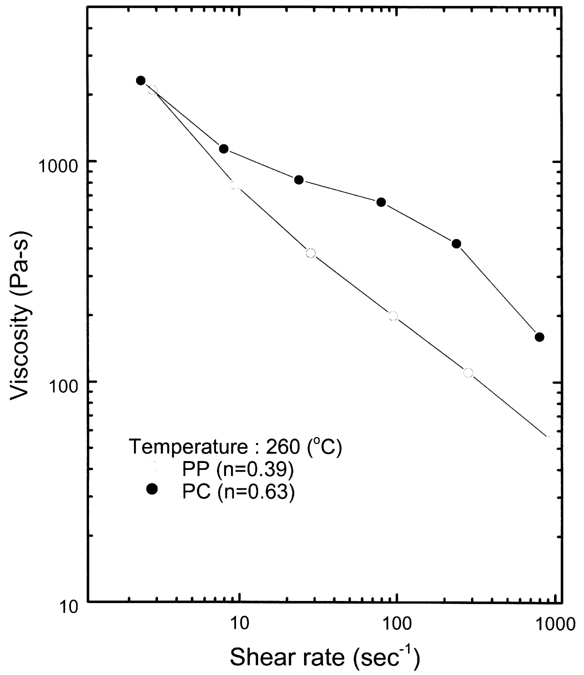

Fig. 1. Melt viscosities of PP and PC as a function of shear rate at 260°C.

diameter ratio of 33/1. End effects were neglected but a wall shear rate correction was made following the Rabinowitch procedure. The viscosity of the two polymers are nearly identical at lower shear rates around $5\,s^{-1}$ but at increasing shear rates, the viscosity of the PC resin is significantly higher than the PP resin. This viscosity behavior implicates that the viscosity ratio can be varied through only speed modification of the rotating screw of mixing. For an example, to obtain the viscosity ratios of about unity and the highest that can be processed, we have run the extruder at two rotation rates: 10 and 110 rpm. Drive rotation rate of 10 rpm corresponded to a maximum shear rate of $5\,s^{-1}$ in the extruder and 110 rpm corresponded to a shear rate of $55\,s^{-1}$. These nominal shear rates were found by using the minimum gaps in the extruder and assuming that the shear rate in the channel is that due to drag flow.

The choice of glass fiber was prompted by the following interfacial considerations between the GF and preimpregnation resins. Since the two resins, PC and PP are very different in their affinity to GFs, we can deliberately change the interfacial structure by using two types of the GFs with different surface treatments. The glass fibers used in this study, as received, were coated with a coupling agent designed to improve adhesion to polyester and polypropylene: (a) polyester-sized roving with diameter of 17 μm (GF1); and (b) polypropylene-sized roving with diameter of 13 μm (GF2).

A continuous roving of these GFs were preimpregnated with each of the two resins, PC and PP by using an impregnation die specifically designed in this study and then cut into chopped pellets of 6 mm length. These pellets were mixed with an appropriate amount of the other virgin resin to fabricate four different interfacial types of GF/PC/PP composites, GF1/PC + PP, GF1/PP + PC, GF2/PC + PP, GF2/PP + PC which contain about 30 wt% of GF and then extruded through sheet die or molded into the mechanical specimens by injection machine. Consider a ternary GF composite, GF1/PP + PC for example. If the GF1 is preimpregnated with PP, interphase cannot be formed by PP which do not wet the GF1. As a result, dewetting is anticipated in a later stage of mixing by the resin of PC if the migration of the resin from the earlier formed resin interface is supported kinetically. However, it is not easy to confirm experimentally such an interfacial interaction. At present, no direct methods are available which could be used to determine the extent of encapsulation surrounding the glass fiber. To be able at least to visualize the interfacial component, we have first observed the etched fracture surface of the numerous specimens using a SEM (Model XL-30, Philips Co.).

Fig. 2 shows the representative micrographs of the izod impact fractured surface for the samples, GF1/PC + PP prepared at the shear rate of $55\,s^{-1}$. They demonstrate that the pre-existing PC layer surrounding GFs was replaced by PP since the polymer sticking to the fiber remained after etching. Similar observations (not shown here) were also made for the other samples of GF1/PP + PC, GF2/PC + PP, GF2/PP + PC, prepared at the same condition. These results indicate that the apparent affinity of the GFs as well as the initial morphology of encapsulated GFs does not influence the interfacial structure and the lower viscosity PP is primarily located within the interphase around the GFs at the higher shear rate.

However, at the lower shear rate of mixing ($5\,s^{-1}$), a noticeable change in the interfacial component was not observed for the same composite, GF1/PC + PP. Comparison of the Fig. 3a and b reveals that PC still remains around the GFs as the polymer sticking to the fibers disappears after etching. Similarly, in the other samples, interfacial changes are not observed as the viscosity ratio approaches unity even though it is thermodynamically unfavored. Thus we conclude that the pre-existing PC layer persists during the mixing process at the lower shear rate. It is interesting to note that the preferential encapsulation of fibers by sequential impregnation protected the fiber to allow significant increase in average fiber length to be achieved in the injection molded specimens [7]. So, we expect large aspect ratios of the fibers occurring in the present composites, thereby allowing the shear force to exceed adhesion. This final structure of the composites, which is dependent on the relative magnitude of the viscosity, suggests that real thermodynamic equilibrium cannot be attained by the end of mixing. Thus, the interface in these types of ternary composites is controlled by kinetic parameters.

The measured mechanical properties confirmed these

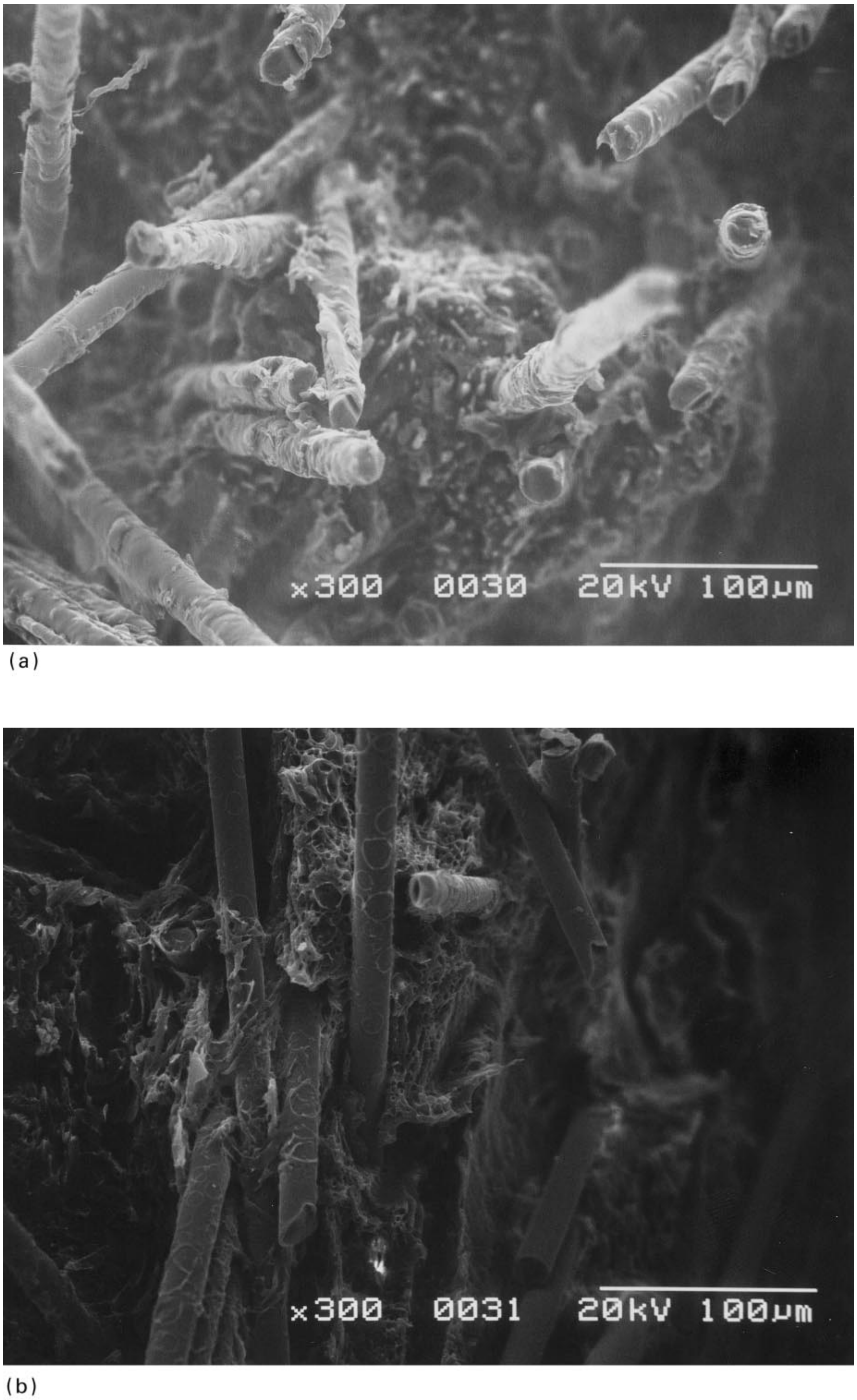

Fig. 2. SEM for the izod impact fractured surface: (a) before, and (b) after etching PC for the composites, GF1/PC + PP prepared at a shear rate of 55 s^{-1}.

morphological behaviors. According to the observed stress–strain behaviors for the four composite samples, GF2/PP + PC sample exhibited the best tensile properties. This result is consistent with the morphological observations. The initial morphology of the encapsulated GFs with PP persists during processing (Fig. 4a and b) regardless of the mixing rate and good wetting results in enhanced properties. Unlike the best properties, the GF1/PC + PP and GF2/PC + PP exhibited the poorest tensile behaviors at the high and low shear rate, respectively. There are several interesting aspects to these results. First, the expected interfacial interaction between PC and polyester

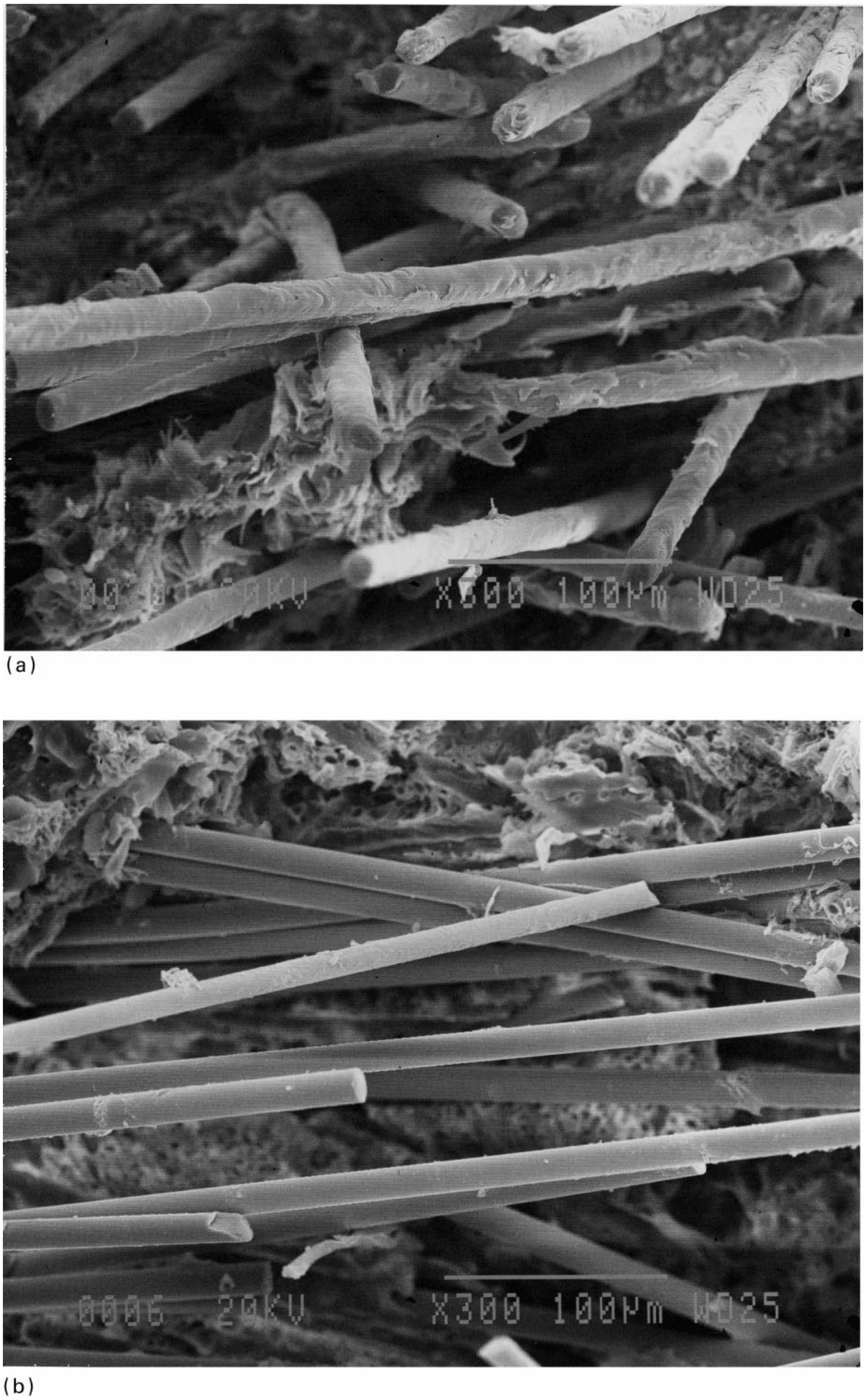

Fig. 3. SEM for the izod impact fractured surface: (a) before, and (b) after etching PC for the composites, GF1/PC + PP prepared at a shear rate of 5 s^{-1}.

sized GFs did not occur during mixing in case of the composite, GF1/PC + PP. Second, poor wetting in GF2/PC + PP between PC and PP sized GFs persisted during mixing at low shear rate. Third, these interfacial structures indicate that the migration of the interfacial PC component occurs at high shear mixing while it does not at low shear mixing.

In view of the above observations, we conclude that viscosity ratio may very likely play an important role in the final interfacial behavior of the GF filled composite and a technologically stationary state of the system can be observed after a sufficiently long and effective mixing instead of equilibrium state. The interfacial structure around GFs can be, however, deliberately

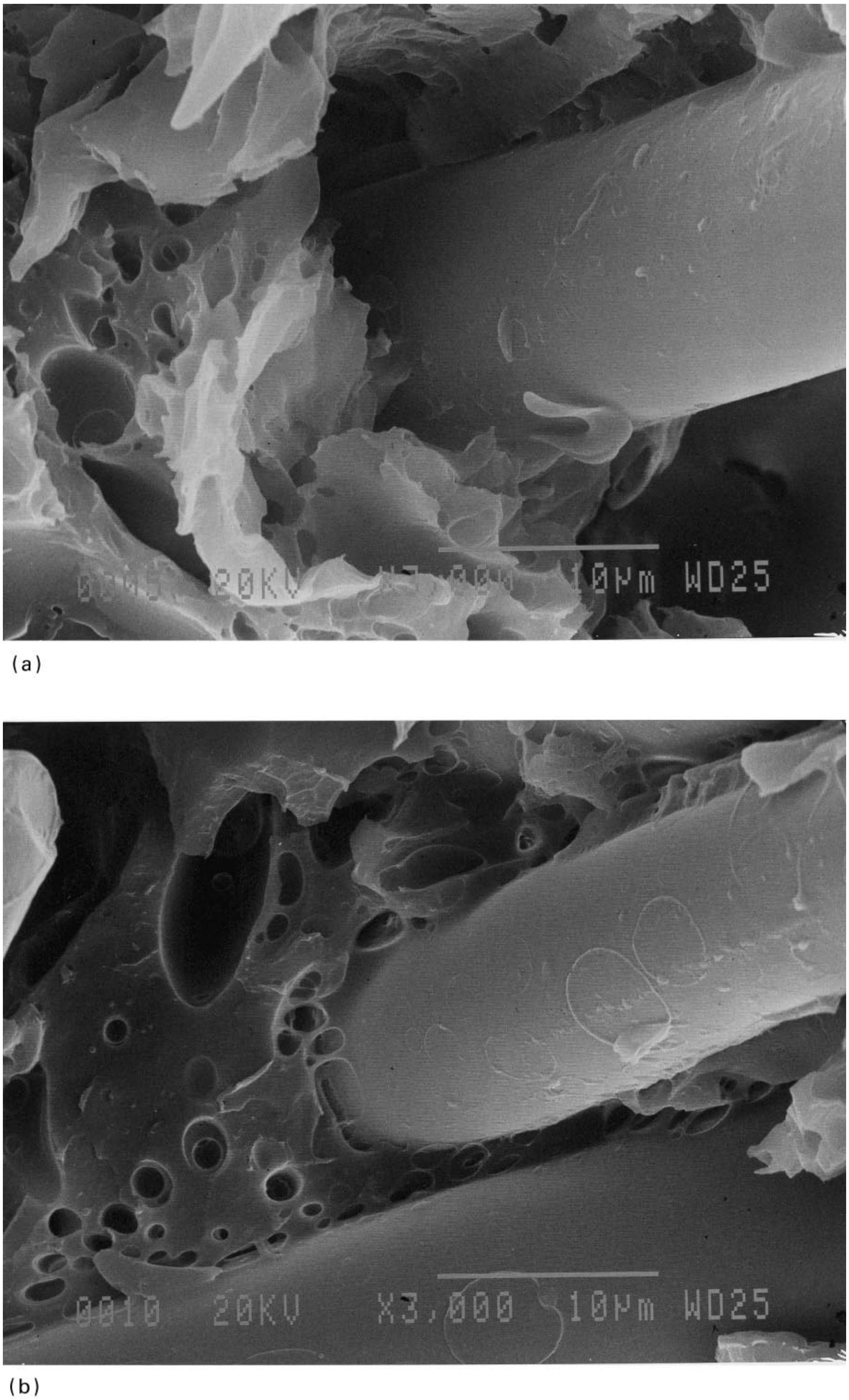

Fig. 4. SEM for the izod impact fractured surface after etching PC for the composites: (a) GF1/PC + PP, and (b) GF2/PP + PC prepared at a shear rate of 5 s^{-1}.

controlled by pre-encapsulation technique in the composite systems containing the resins with similar melt viscosities due to the thermodynamic work of adhesion between fiber and resin. Thus, the improved wetting of fibers by sequential impregnation in ternary composites would be a good approach in cases where the combinations of the resins or processing conditions lead to the similar relative melt viscosity of the component resins.

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