Transcrystallization at glass fibre/polypropylene interface and its effect on the improvement of mechanical properties of the composites

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Transcrystallization has been reported to occur in semicrystalline polymers such as isotactic polypropylene (i-PP) in contact with carbon fibres (CF) and aramid fibres [1, 2]. On the other hand, transcrystallization has not been considered to be observed in PP in the presence of glass fibres (GF) [3]. However, it was recently demonstrated that transcrystallinity could form at the interface between GF and an i-PP matrix [4–8]. Thomason and Van Rooyen reported that application of stress at the interface between GF and supercooled PP melt by using pulling apparatus resulted in the growth of a transcrystallized interface, independent of crystallization temperature ranging from 140 to 180 °C [4]. Wagner *et al.* demonstrated that transcrystallinity could form at the interface between GF and i-PP if appropriate nucleating agents were used to coat the fibres [5]. However, transcrystallization of i-PP in contact with GF is not sufficiently understood compared to transcrystallization in the presence of CF and aramid fibres. Therefore, we investigated transcrystalline behaviour at the interface between GF and i-PP, particularly without application of stress at the interface and without nucleating agents to coat GF.

Commercial i-PP (melt flow index = 10) was used as a matrix. E-glass fibres coated with an aminosilane coupling agent were used. The fibres were used with or without coating with acid-modified polypropylene (AMPP, $M_w > 10^4$, acid value = 70 mg KOH g^{-1}) in tetralin solution at 120 °C, in order to study the improvement of the compatibility with matrix PP by coating GF with AMPP. A hot-stage microscope fitted with charge-coupled device (CCD) camera and video recorder was used to observe the crystalline behaviour of PP. The samples, prepared by hot pressing GF with two sheets of matrix PP films, were heated to 205 °C and held for 5 min to ensure complete melting. The samples were then cooled under nitrogen to 120 °C at a constant cooling rate ranging from 5 to 40 °C min^{-1}, in order to observe the crystallization behaviour. Furthermore, a fragmentation test [9] was done to evaluate the interfacial shear strength between GF and PP. The test pieces were prepared by attaching the samples for crystallization observation to dumbbell-shaped epoxy resin by adhesive.

Fig. 1 shows polarized micrographs of crystalline morphology observed using GF with and without AMPP coating. Transcrystalline region was found to be induced around GF coated with AMPP, while no transcrystalline region was found around GF without AMPP coating. It was demonstrated that transcrystallization could be induced by coating GF with AMPP without application of stress at the interface and without nucleating agents to coat GF. It was thought possible that some impurities such as metallic compounds in AMPP acted as nucleating agents.

Table I shows the effect of cooling rate on transcrystallization. Transcrystallization could be observed for GF coated with AMPP at different cooling rates ranging from 5 to 40 °C min^{-1}. These cooling

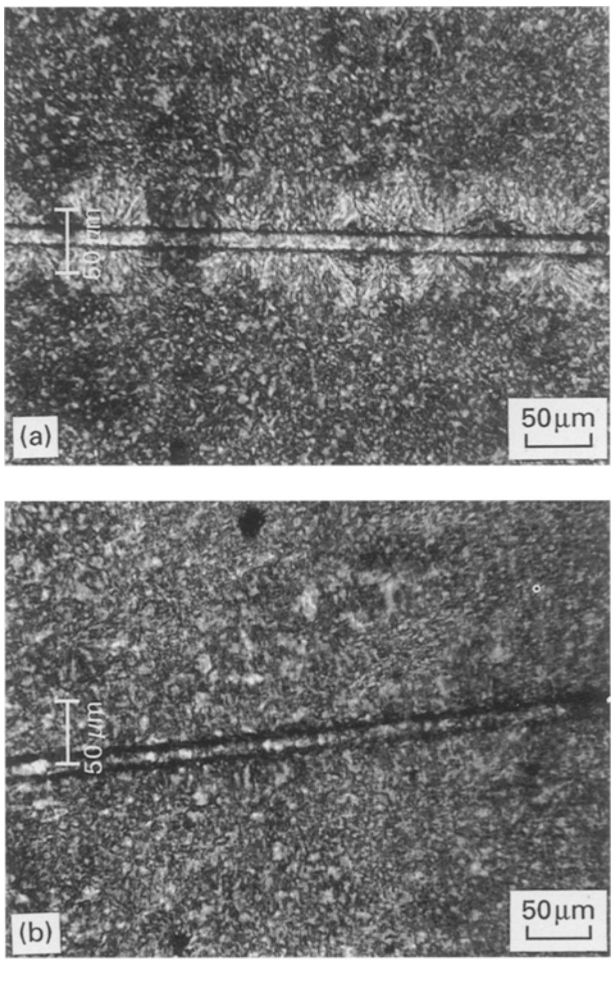

Figure 1 Polarized optical micrographs of crystalline morphology: (a) GF coated with AMPP and (b) GF without AMPP coating.

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TABLE I Effect of cooling rate on transcrystallization

	Cooling rate (°C min^{-1})		
	5	20	40
Coated with AMPP	Yes	Yes	Yes
Without AMPP coating	No	No	No

TABLE II Effect of coating GF with AMPP on interfacial shear strength

Glass fibres	Interfacial shear strength (MPa)
GF without AMPP coating	6
GF coated with AMPP	24

rates were found to be much lower than those reported by Thomason and Van Rooyen (100–280 °C min^{-1}) [4] or by Wagner *et al.* (10 °C s^{-1}) [5]. Our experimental results suggest that transcrystallization could occur even at a low cooling rate, similar to that of the actual moulding process of the composites.

Fig. 2 shows polarized optical micrographs of the fracture surfaces of GF with and without AMPP coating at fragmentation test. It was shown that the interfacial adhesion between GF coated with AMPP and matrix PP was so good that fracture at the surface was transferred to matrix PP. On the other hand, pull-out of GF was found to take place in the case of GF without AMPP coating, because of poor interfacial adhesion. The improvement of interfacial shear strength by coating with AMPP studied by the fragmentation test is shown in Table II. The interfacial shear strength was drastically improved from 6 to 24 MPa.

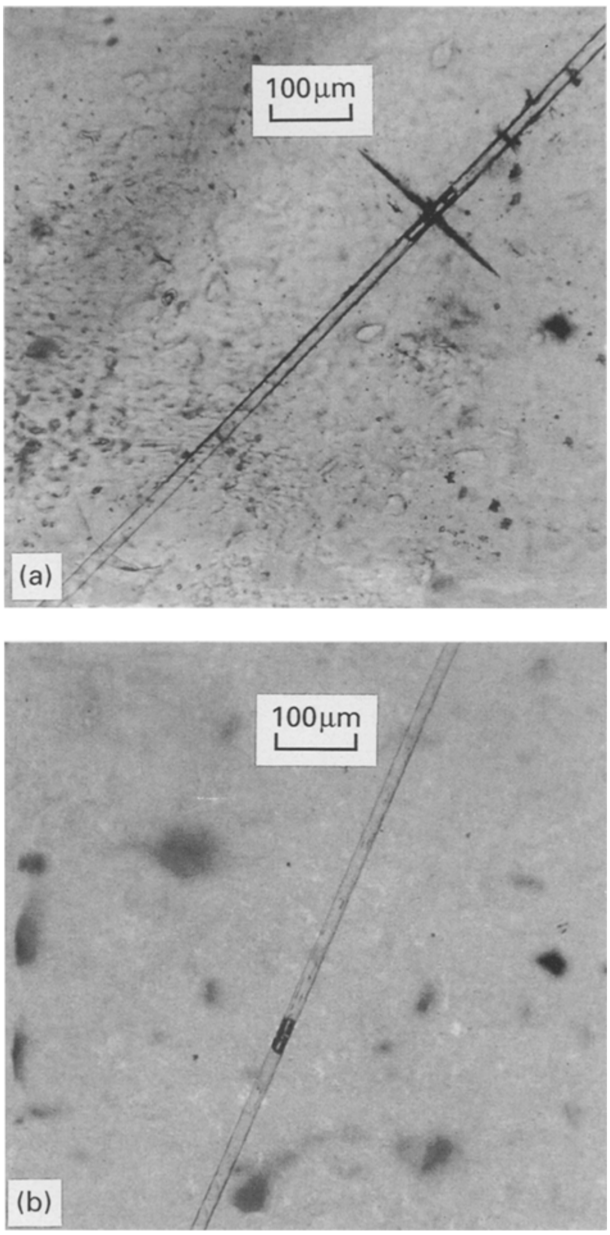

Figure 2 Polarized optical micrographs of fracture surface of GF at fragmentation test: (a) GF coated with AMPP and (b) GF without AMPP coating.

The mechanical properties such as flexural strength and notched Izod impact strength were investigated for the composites prepared using GF with and without AMPP coating, because the improvement of interfacial shear strength was expected to lead to improvement in the mechanical properties of the composites. For example, Folkes and Hardwick [7] and Rijsdijk *et al.* [8] reported that the improvement of interfacial shear strength contributed to the improvement of flexural strength of GF/PP laminates. Fig. 3 shows our experimental results of the mechanical properties of the composites. Both flexural strength and notched Izod impact strength were significantly improved by coating GF with AMPP.

In order to study the morphology of the composites, a cross-section of the PP film sandwiching GF was observed by scanning electron microscopy (SEM) following the etching procedure reported by Aboulfaraj *et al.* [10]. As shown in Fig. 4, transcrystalline morphology could be observed in the case of the composites prepared using GF coated with AMPP. It is considered that transcrystallization of PP around glass fibres is closely related to the

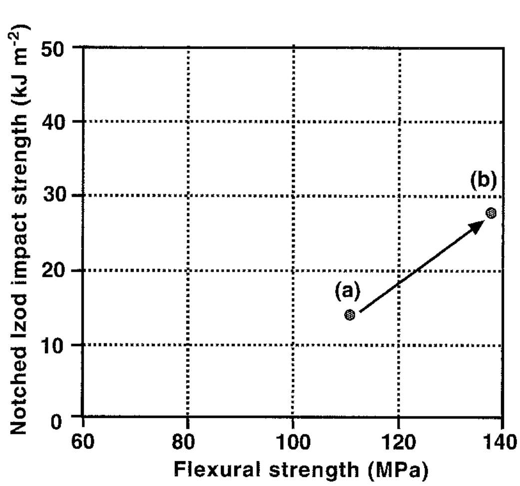

Figure 3 Effect of AMPP coating on mechanical properties of composites: (a) GF without AMPP coating and (b) GF coated with AMPP.

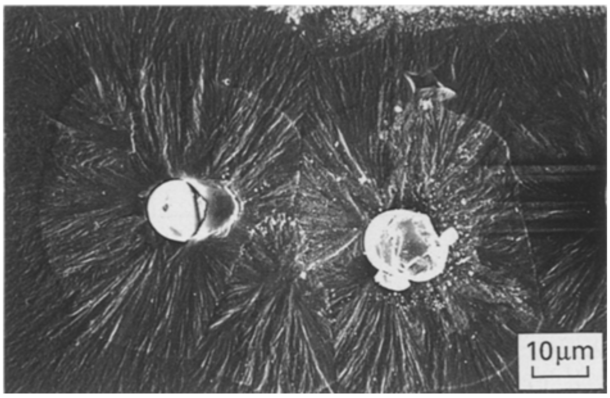

Figure 4 SEM micrograph of cross-section of GF/PP composite.

improvement of the mechanical properties of the composites. However, further study is necessary to clarify the mechanism of transcrystallization and its effects on the improvement of mechanical properties of the composites.

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