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Evaluation of interfacial properties in glass fibre–epoxy resin composites – reconsideration of an embedded single filament shear-strength test

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An embedded single-filament shear-strength test was employed to explore the effect of the reinforced interface on the fracture of single filaments in glass fibre–epoxy resin composites. The interfacial effect was evaluated not only by critical fibre length but also by the fracture process of an embedded single filament observed by applying tensile load to the composites. The mean fragment length, measured at different tensile stresses, decreased with an increase in stress and finally reached a value correlated with critical fragment length. Interfacial reinforcement by silane treatment affects the fracture process rather than the critical fibre length. The behaviour was examined in several factors, such as filament diameter, the tension of filament on the moulding of composites, and the scatter of the composites in mechanical properties. A useful method was proposed to exclude such scatter: specimens treated on a half-length of embedded filament were subjected to this test. The present method leads us to confirm that the reinforced interface has no effect on the critical fibre length, while it does promote the progress of the fracture.

1. Introduction

Silane-coupling agents are applied to the processing of glass fibre–epoxy resin (GFRP) composites to reinforce interfacial strength through the chemical function of the coupling agents. The reinforcement mechanism is still not completely elucidated in connection with the interfacial structure formed by the silane-coupling treatment. The interfacial reinforcement mechanism is virtually based on the chemical bonds of silane-coupling agents to resin and glass surface. The chemical bonding theory [1, 2], however, is not consistent with all of the facts: it does not explain some mechanical properties of GFRP, such as relaxation properties for stress and water susceptibility for strength. Many other theories have been proposed: the deformable layer theory; the surface wettability theory; the restrained layer theory; and the reversible hydrolytic bond mechanism, as summarized in reviews [2, 3]. These theories have been discussed in relation to the molecular structures of silane-coupling agents at the interface, to be correlated to the mechanical properties of GFRP.

In spite of these efforts, we have found no quantitative theory which relates the molecular structure to the mechanical properties at the interface, although such a theory is required to consider GFRP performance. We should evaluate the silane interphase not only on the basis of determination of molecular structures, but also in relation to some mechanical technique giving information on the mechanical properties of the interphase.

An embedded single-filament shear-strength test is a useful technique to evaluate the stress transferability of the interphase from matrix to glass fibre. This technique has been employed to determine critical fibre length and interfacial shear strength from the lengths of fibre fragments in the ultimate fracture state occurring under sufficient tensile strain [4–9].

However, little attention has been paid to the preparation of specimens, in particular to the pre-tension of filaments. Pre-tension means that a load is subjected to a filament prior to embedding. Another important point is the critical fibre length, that is, the critical or ultimate state. The single-filament test is ambiguous for the determination of the ultimate fracture state. It is difficult to judge whether the shear strain is sufficient to break the embedded filament down to the final state.

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In this study we discuss the effect of pre-tension on the critical fibre length. The critical fibre length is greatly affected by pre-tension of the filament. In the case of high pre-tension, we find no difference between treated and untreated filaments in critical fibre length. A new technique is proposed to make the interfacial effect clear.

2. Experimental procedure

2.1. Silane treatment

Glass fibres used in this study were E-glass filaments of 7 and 24 μm in diameter. The filaments were obtained from E-glass yarns supplied by Nippon Glass Fibre Co. Prior to use, the yarns were heat-cleaned at 600 °C for 2.5 h to remove binders and any other organic impurities. The silane-coupling agent, γ-anilinopropyltrimethoxysilane (AnPS), was purchased from Shin-etsu Chemical Industries. The silane agent was used without further purifications.

The silane treatment was performed as follows. The filament was dipped in liquid AnPS and then cured at 80 °C for 40 min. After curing, the filament was washed with distilled water and dried in air. The silane-treated filament was then used to make specimens of the single filament composite.

2.2. Preparation of specimens

The matrix material used in this study was a bisphenol-A-type epoxy resin (Epikote 828, Yuka-Shell). The resin was mixed with 11 parts of a hardening catalyst, triethylenetetramine. The mixture was agitated thoroughly and then defoamed in vacuum for ~ 12 min. This mixture was poured into a mould holding a glass filament with tension and subjected to curing at 50 °C for 100 min, and post curing at 100 °C for 60 min. The dimensions of the mould resulted in a specimen 1 mm thick, 10 mm wide and 100 mm long.

2.3. Measurement of the fracture process

The specimens were subjected to tensile stress at a test speed of 0.05 mm^{-1} min^{-1} with the aid of an Instron-type machine (Yonekura, CATY-500 BH). The specimens were pulled to different tensions and then the fragment lengths and fracture positions along the fibre length were measured with an accuracy of 0.0025 mm by a measuring system equipped with a polarization microscope (Nippon Kougaku, S-Po).

3. Results and discussion

3.1. Mean fragment length

Single filament composites with a reinforced interface by silane treatment were subjected to tensile testing to measure the mean fragment length at various extents of the stress. Composites without silane treatment were also subjected to tensile testing as a reference. Fig. 1 shows the change in mean fragment length in the stress of specimens embedded in 7 μm diameter of filament. The mean fragment length, $\langle l \rangle$, decreases with an increase in stress and finally reaches a constant value about 0.2 mm, regardless of silane treatment.

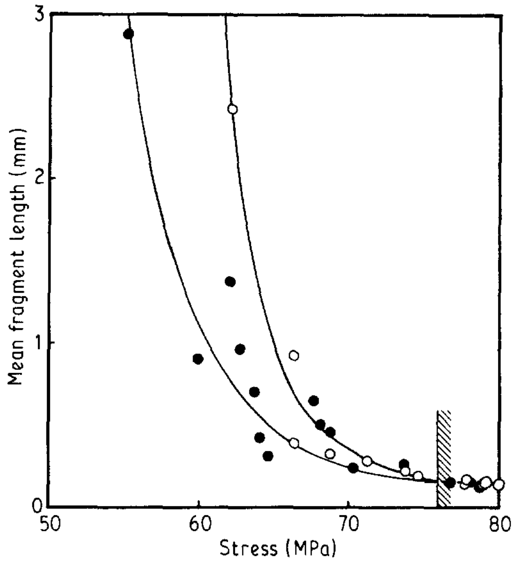

Figure 1 Effect of silane treatment on mean fragment length in fracture process by tensile stress for an embedded filament of 7 μm diameter with 3.3 g of tension. ●, treated; ○, untreated.

As shown in earlier reports for the single-filament test [4, 5], the critical fibre length, l_c, has been evaluated from the mean length of the final broken pieces, $\langle l_f \rangle$, by the following equation

$$l_c = 4/3 \langle l_f \rangle \qquad (1)$$

In this study, $\langle l_f \rangle$ is comparable to the ultimate mean value of about 0.2 mm, so that the critical fibre length has the same value regardless of silane treatment. This result suggests that the interfacial strength is ineffective on the final fracture state, though it has been pointed out that the interfacial effect reflects the critical fibre length. An explanation for these inconsistent results may relate to two different factors: one is the diameter of filament, and the other the tension of the filament on the moulding of the single-filament composite.

A filament > 10 μm in diameter was employed in the previous studies, while a filament of 7 μm was used in this study. According to the theory for critical fibre length [5–11] the shorter the diameter, the shorter the critical fibre length. Hence the ultimate mean value obtained in this study may include such an experimental error that the interfacial effect cannot be regarded as significant.

3.2. Effect of pre-tension of filament

The tension of the filament on the moulding affects the fracture process in stress. As shown in Fig. 2, the fracture begins at lower stress as the tension is higher. This means that insufficient tension makes it impossible to observe the ultimate fracture state before the composites are broken down in the tensile test. The stress range giving the rupture of specimens, as shown in Fig. 1, overlaps with the final fracture state.

In order to confirm these factors in the tensile test, we have examined the fracture process for the composite embedding a single filament 24 μm in diameter.

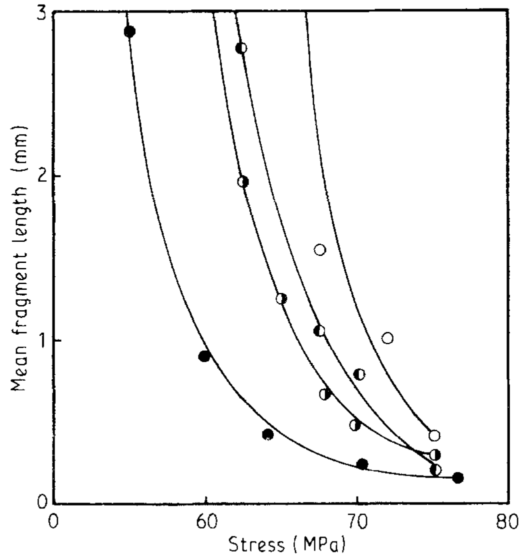

Figure 2 Effect of filament tension on fracture process for an untreated filament of 7 μm diameter. Tension: ●, 3.3; ◑, 2.2; ◐, 1.2; ○, 0 gf.

Fig. 3 shows the change of mean fragment length in stress for the filament 24 μm in diameter. The final fracture state is rarely observed at lower tension of the filament. Moreover, in the stress range rupturing the specimens, the apparent final value for the filament with 3.3 g of tension is apparently different for silane treatment and no treatment. The results suggest that the embedded single-filament test may make the critical fibre length misleading if the diameter is large and the filament tension is insufficient. It is necessary to fix an adequate filament tension for comparison of the interfacial effect in the single filament test.

From Fig. 3, the final rupture stress in the case of pre-tension of 3 gf is larger than that of 13 gf. Fig. 4 shows the fracture surface of a specimen. It was clear that the fracture initiated from the filament. In the case of high pre-tension, the number of fracture points was increased so that the probability of rupture was high: therefore the final rupture stress was decreased.

3.3. Half-length-treated specimen

As shown in Figs 1 and 3, the effect of interfacial reinforcement appears not in the ultimate state but in the initial and middle states of the fracture process. We should deal with the fracture process in detail to investigate the interfacial effect. However, the data points are so scattered as to include a considerable error in measurement. The scatter may result from that of the specimens in mechanical properties. In order to make the interfacial effect clear, a half-length of single filament embedded in the resin was treated with a coupling agent. Here we call this specimen a "half-length-treated specimen". This technique is expected to exclude the error resulting from the scatter of mechanical properties. The fracture state in an untreated part of the filament can be used as a reference, for the actual tensile stress of the filament results in the corresponding fracture state.

Fig. 5 shows the result obtained by a technique of silane treatment on a half-length-treated specimen.

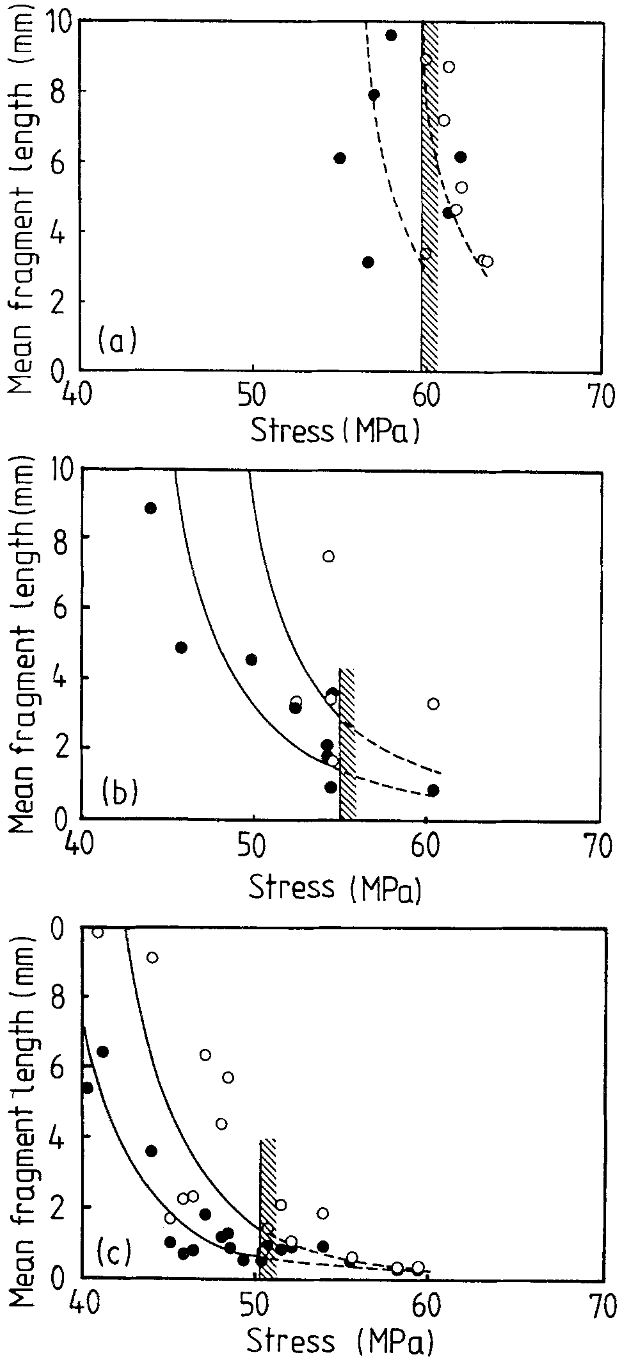

Figure 3 Fracture processes for a filament of 24 μm diameter: ●, silane-treated; ○, untreated. Vertical line, approximate failure stress for specimen in each tensile test. Tension (a) 3, (b) 8 and (c) 13 gf.

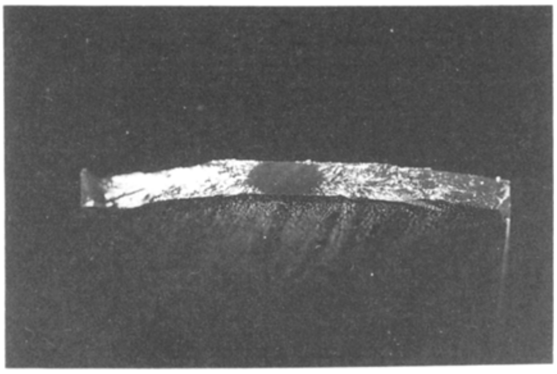

Figure 4 Fracture surface of the embedded single filament specimen.

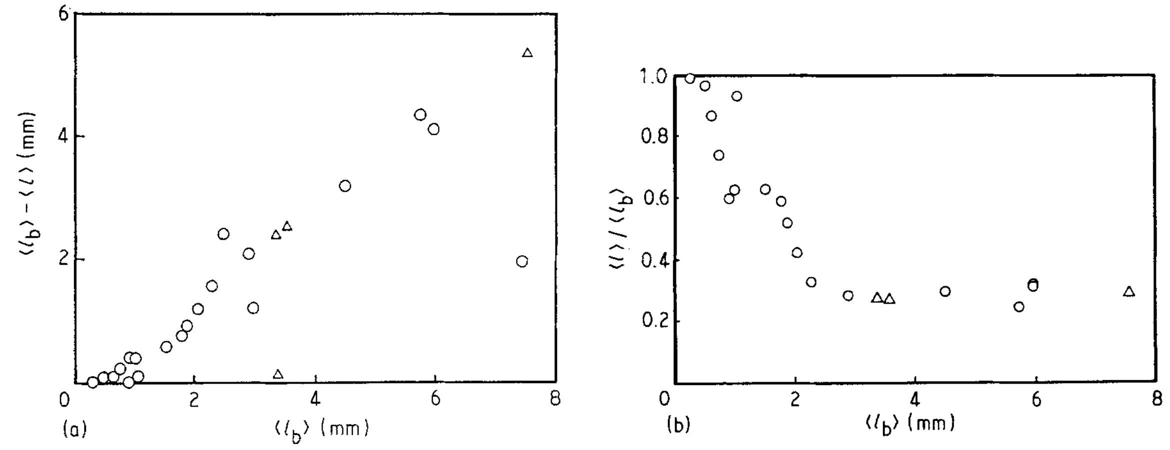

Figure 5 Interfacial effect on mean fragment length resulting from a technique of silane treatment on a half length of embedded filament of 24 μm diameter. Tension: △, 8; ○, 13 gf.

The mean fragment length in the untreated part, $\langle l_b \rangle$, was taken as an index expressing progress in the fracture process. The difference, $\langle l_b \rangle - \langle l \rangle$, and the ratio of mean fragment lengths, $\langle l \rangle / \langle l_b \rangle$, were plotted against $\langle l_b \rangle$. This procedure was found to depress the scatter of the data points. Also, the filament tension has no effect on the fracture process, as can be seen from the plots for the different tensions, 8 and 13 gf. It apparently shifts the fracture process to lower specimen stress. This means that the tension is ineffective in interfacial reinforcement. From the relation between $\langle l \rangle$ and $\langle l_b \rangle$, it is obvious that the mean fragment length becomes the same value in the final fracture state, regardless of silane treatment. On the other hand, the interfacial reinforcement appears in the initial and middle states. The present procedure is thus a useful way to evaluate the interfacial effect.

4. Conclusions

The fracture process of a single filament embedded in epoxy resin was investigated. A technique of silane treatment on a half-length of embedded filament was found to be a useful way to investigate the interfacial effect, due to the depression of scatter of composites caused by mechanical properties. All of the results show that interfacial reinforcement reflects the fracture progress rather than the final fracture state.

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