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Fracture toughness and mechanical properties of glass fibre–epoxy composites

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The present study investigated fracture properties and various mechanical properties of a set of unidirectional glass fibre–epoxy resin composites. This set was comprised of samples with volume fraction of fibres in the range 0.29 to 0.75. An identical set of composites was water-boil treated for 7 days, and the effect of this treatment on the above properties was examined. The work of fracture (γ_F) and the fracture surface energy of initiation (γ_I) results were compared with existing theoretical models for the prediction of fracture toughness. It was discovered that the γ_F results agreed with the pull-out model [6], suggesting that this was the major contribution to the fracture energy of the complete process. The γ_I values corresponded generally with the surfaces formation model [9], proposing that the creation of new fibre, matrix and fibre–matrix surfaces controls the stage of fracture initiation.

1. Introduction

A number of researchers have looked into the problem of fracture in fibre-reinforced composite materials. It seems that the more meaningful studies are those which characterize fracture process in terms of fracture energies (see for example [1-4]), rather than by measuring ultimate properties such as strength. This results from the fact that whereas ultimate properties are usually a function of the constituent material properties (i.e. expressed by a "rule of mixtures"), the fracture energies relate to the composite structure and to the fracture mechanism. This explains why values of fracture energy are 2 to 4 orders of magnitude greater than those of the constituent materials [5].

Until recently, four theories of fracture of composites have been proposed to account for the main contributions to the fracture energy. These are fibre pull-out [6], debonding [5], fibre failure and relaxation [7, 8], and a new theory by Marston *et al.* [9]. The relationships proposed by these theories are all functions of the fibre volume fraction (V_f) and of the fibre critical length (l_c) (see Section 4.2).

In view of the assumed dependence on V_f and l_c of the fracture toughness, it was felt that an attempt should be made to examine fracture toughnesses of a set of composites with a wide range of V_f values, and also, to study the properties of similar composites which were exposed to boiling water, assuming that this treatment would affect the value of l_c. At the same time other properties such as the modulus, the ultimate strength and the shear strength were also determined in order to obtain a wider perspective.

2. Experimental procedure

2.1. Materials

Composites were manufactured from epoxy resin (Ciba-Geigy Ltd – Araldite F/HT972) reinforced by silane treated E-glass fibres (Vetrotex Ltd – EC14-300-K937). The glass fibre was wound and impregnated by an acetone solution of the resin on a winding machine drum. After a complete evaporation of the acetone, the "pre-pregs" were cut, laid in a mould and pressed under 100 to 400 psi* at 170° C to form 0.5 cm thick plates. The volume fraction of the fibre was controlled by the number of "pre-preg" sheets introduced into the mould, and by the intensity of the applied moulding pressure. The plates were post cured for 2 h at 180° C and then cut into test specimens. Specimens destined for the water-

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*10^3 psi = 6·89 N mm^{-2}.

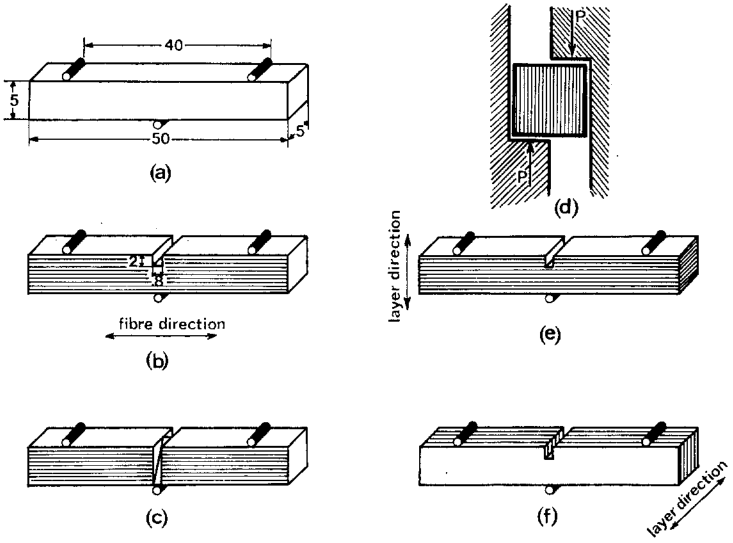

Figure 1 The geometry of the test specimens used for measuring mechanical properties and fracture toughness. All measurements are in millimeters.

boil treatment were continuously boiled in water for 7 days.

2.2. Testing

Fig. 1 presents the geometry of the specimens used for measuring (a) modulus and ultimate strength, (b) fracture surface energy of initiation, (c) work of fracture [10], and (d) shear strength. Fig. 1e and f specify interlaminar and translaminar modes of loading. These were used since properties of lay-up composites were expected to depend on the direction of loading relative to the layer direction. Testing was carried out by three-point bending (except for shear tests) on a floor model Instron at a cross-head speed of 0.05 cm min^{-1}. Each property was determined using at least seven specimens.

The work of fracture was calculated by the expression $\gamma_F = U/2A$, where U is the integrated load-deflection curve and $2A$ is the area of the newly formed surfaces. The fracture surface energy for initiation was calculated by the expression $\gamma_I = \sigma^2 Y^2 c/2E$, where σ is the fracture stress, c is the notch depth, E is Young's modulus, and Y is a geometrical factor [11].

3. Mechanical properties

3.1. Young's modulus

The results of the modulus are presented in Fig. 2, where they are compared with the "rule of mixtures" (see for example [12]). It is seen that the theoretical line sets higher values compared with the experimental results, and also, the interlaminar values ($E_c{}^i$) are in general slightly greater than those of the translaminar ($E_c{}^t$) values. The first observation is in agreement with the suggestion that the "rule of mixtures" only sets an upper bound to the axial modulus values [12]. The latter observation may be explained by Tsai's [13] modified expression $E = k(E_f V_f + E_m V_m)$, where E_f and E_m are the moduli of the fibre and the matrix, respectively, and $V_m = 1 - V_f$. The factor k accounts for different states of fibre misalignment which may exist in the interlaminar and the translaminar directions.

It should be pointed out that the modulus values are not affected by the water-boil treatment, in agreement with an observation made on SiO_2 fibre–epoxy resin composite [4].

3.2. Ultimate flexural strength

Fig. 3 presents the experimental translaminar u.f.s. values (σ_c). Interlaminar values could not be measured because the specimens failed by delamination. The results in Fig. 3 are compared with the simple "rule of mixtures" expression, drawn for $\sigma_f = 1.2$ GN m^{-2}, which is the value of the fibre strength after the compounding process [14]. Unboiled samples exhibit u.f.s. values which agree with the "rule of mixtures", except for high V_f values. It may, however, appear that the real value of σ_f for the glass fibre in the resin is even smaller than the above. Water-boiled samples exhibit lower u.f.s. values along the whole examined V_f range. This results

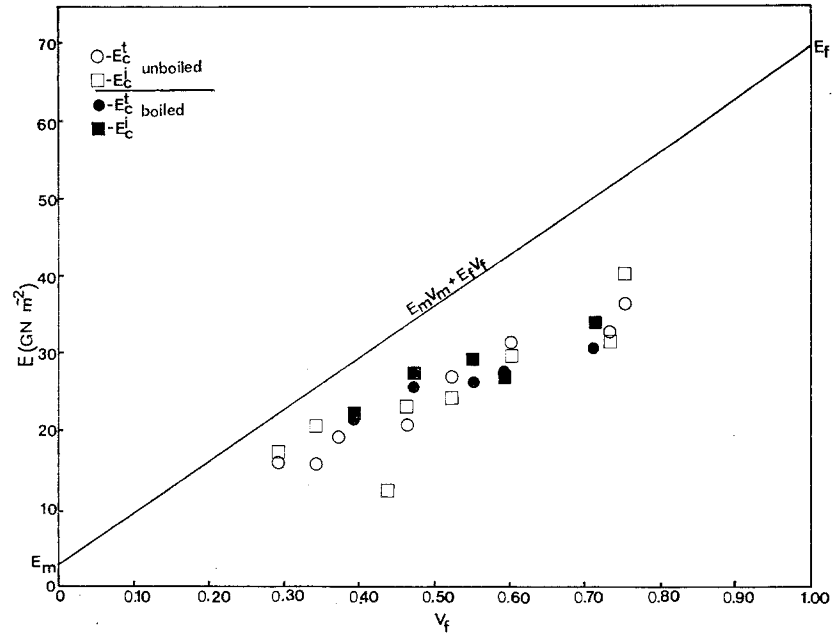

Figure 2 The results of Young's modulus compared with the rule of mixtures.

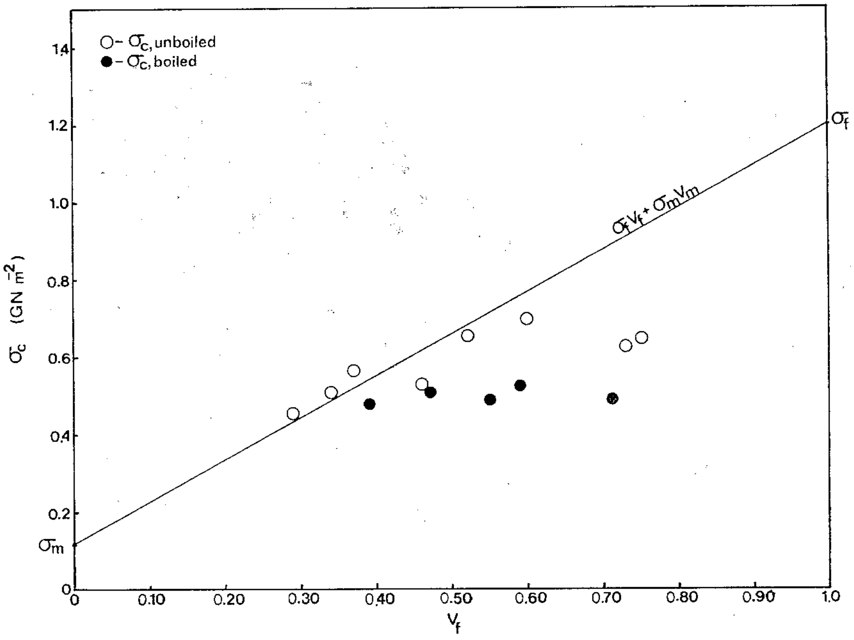

Figure 3 The results of the ultimate fracture stress compared with the rule of mixtures.

from a deterioration in the strength of the glass fibre due to the boiling process.

3.3. Scissors shear test

Translaminar shear strengths ($\tau_c{}^t$) were mostly greater than interlaminar ($\tau_c{}^i$) values. This results from the composites manufacturing technique, which involved a hand lay-up procedure of "pre-pregs", whereupon the possibilities of translaminar fibre misalignment were greater than interlaminar. The average value of $\tau_c{}^t$ and $\tau_c{}^i$ for each sample was taken to represent its shear strength. The results are shown in Fig. 4. It can be seen that the exposure of the samples to boiling water results in a 20% decrease in the value of τ_c. This decrease is attributed to the

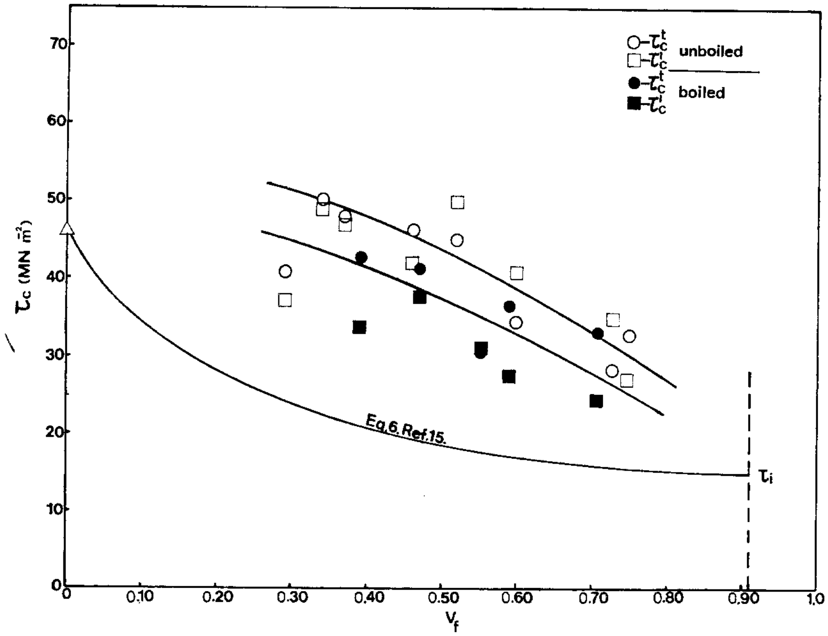

Figure 4 Scissors shear stress values presented as a function of the fibre volume fraction.

destruction of the fibre-matrix bond. The trend of the shear strength values as a function of V_f deviates from the expression [15] $\tau_c = \tau_m (1 - \chi) + \tau_i \chi$, which expresses τ_c by proportional contributions of the matrix and the interface (τ_m and τ_i are the shear strength of the matrix and interface respectively, and χ is the fraction of the fracture area occupied by the fibres). Also, some composite shear strength values exceed that of the pure matrix. These observations agree with those made by other investigators for various types of composites (see for example [16]), and may be a result of contribution of fibres misaligned with respect to the shear plane [16].

4. Fracture toughness

4.1. Measurements of critical length

The value of l_c was measured by a scanning electron microscope (SEM) using two methods. By the first, the pull-out lengths of fibres sticking out from the fracture faces were measured (Fig. 5). This was carried out for specimens of all the composite samples, but no significant difference in the value of l_c was noticed, either as a result of a different V_f value or as a result of the water-boil treatment (see Section 4.2). The observed pull-out length varies between 0 and $l_c/2$, hence the mean pull-out length ($\bar{x}$) is equal to $l_c/4$. This method produced an average value of $l_c = 2.4$ mm.

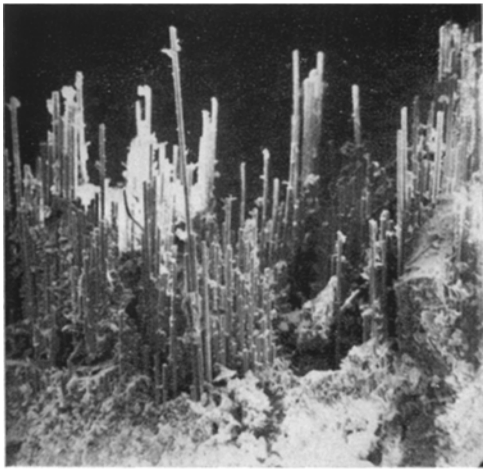

Figure 5 SE micrograph of a fracture surface showing the pull-out fibres (× 43).

By the second method, the debonded length (y) was determined. This was done by measuring the ultimate width of the "milky zone" formed in the specimens (Fig. 6). Dynamic observations of three-point bending tests performed on the SEM enabled to detect the stage of fracture initiation, which relates to the ultimate debonded

Figure 6 SE micrograph of the debonding zone at the instance of fracture initiation (× 40).

length. If we assume that y will be about twice the mean pull-out length, i.e. $y = 2\bar{x}$ (see for example [18]), and that $l_c = 4\bar{x}$, we obtain $l_c = 2y$. The average critical length calculated from this relationship was 2.2 mm. The final value used in the present work was 2.3 mm.

A value of $l_c = 2.3$ mm as calculated by those methods is in agreement with that of 4 mm reported for an S-glass-epoxy system [19] and with that of 2 mm reported for an E-glass-polyester system [2]. However, such a value of l_c is much smaller than that of $\sim$ 13 mm obtained from measurements of strengths [15] and thermal expansion [21] of discontinuous fibre composites of a similar composition. Also, a value of $l_c =$ 2.3 mm when substituted into the relationship $\tau_i = \sigma_f r / l_c$ yields a value of τ_i of 3.6 MN m^{-2}. This value is low compared with that of $\sim$ 15 MN m^{-2} obtained by extrapolation of τ_c (Fig. 4) to the point of maximum V_f (0.91). The reason for these discrepancies may appear to lie in different fibre–matrix shear transfer processes, which affect fracture properties, strength or thermal expansion differently, resulting in different l_c values. Thus, a particular l_c value reflects an interfacial shear strength which exists during a specific mechanical configuration. For example, a value of $l_c \simeq 13$ mm, measured from strength of discontinuous fibre composites, reflects a concomitant value of $\tau_i \simeq 12$ MN m^{-2} [15], while a value of $l_c \simeq 2$ mm, measured from fracture tests, reflects a concomitant value of $\tau_i \simeq 4$ MN m^{-2}. This consideration also indicates that none of these τ_i values ought to coincide with that of $\simeq$ 15 MN m^{-2} measured by scissors type testing. (See also Section 5.)

4.2. The work of fracture

The results of the work of fracture are shown in Fig. 7. It is obvious that the values of the translaminar work of fracture ($\gamma_F{}^t$) are in general slightly lower than those of the interlaminar values ($\gamma_F{}^i$). The water-boil treatment results in an average reduction of 65% in the value of γ_F. This phenomenon is discussed later. The results of the two sets exhibit a linear trend with V_f. This behaviour agrees with all the theories for the prediction of γ_F.

According to Kelly [17] the major contribution to the value of the work of fracture is from the fibre pull-out process [6], which relates to a complete separation of the two formed specimen parts. This energy contribution is expressed by Equation 1.

$$\gamma_{po} = \frac{V_f \sigma_f l_c}{24} . \tag{1}$$

Similarly, the debonding process [5], expressed by Equation 2, is claimed to contribute to the fracture surface energy [17].

$$\gamma_d = \frac{V_f \sigma_f{}^2 y}{4 E_f} . \tag{2}$$

Another proposed explanation for the toughness of composites is the fibre failure and relaxation process [7, 8] (also called stress redistribution). The energy dissipated in this process is given by

$$\gamma_r = \frac{V_f \sigma_f{}^2 l_c}{6 E_f} . \tag{3}$$

Marston *et al.* [9] have recently proposed a new theory, by which the fracture toughness is given as a sum of separate contributions, namely:

$$\gamma_F = \gamma_{po} + \gamma_r + \gamma_s . \tag{4}$$

γ_s is the fracture toughness of the three types of surfaces formed during fracture: fibre, matrix and fibre–matrix interface. γ_s is given as follows:

$$\gamma_s \simeq \frac{V_f \gamma_m l_c}{d} . \tag{5}$$

where d is the fibre diameter and γ_m is the fracture toughness of the matrix.

According to [8], the process of fibre failure and relaxation includes the process of debonding. In [9], however, it is claimed that γ_d is implicit in γ_s. Thus, Equation 4 appears to include the

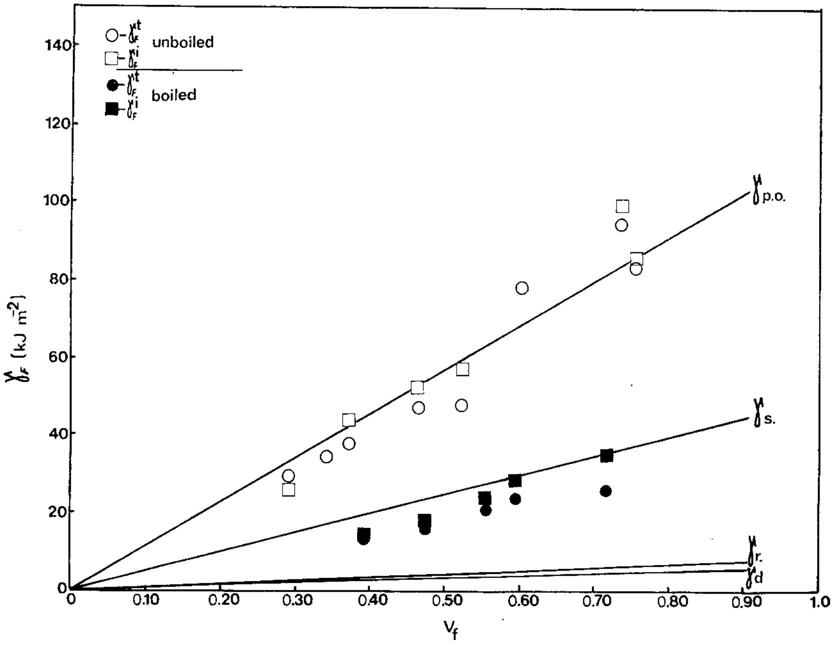

Figure 7 Work of fracture values as a function of V_f. A comparison with theoretical models for the prediction of fracture toughness.

contribution of γ_d twice, once through its contribution to γ_s and again through its contribution to γ_r. Fig. 7 compares the experimental data with the proposed models. The value of γ_m taken in the calculation of γ_s was 0.3 kJ m^{-2} [20]. It is seen that energies dissipated in debonding, failure and relaxation, and surfaces formation are too low to account for the experimental values. The sum of $\gamma_s + \gamma_{po}$ exhibits much too high values. However, the fibre pull-out model exhibits a very good agreement with the experimental values of the unboiled set, suggesting that this is the main contribution to the average fracture energy of the complete process. This result, although in agreement with Kelly's [17] anticipation, contradicts a suggestion by Beaumont and Phillips [19] that a debonding model agrees more closely with their observed glass fibre composite data. The data given in [19] for S-glass fibre–epoxy resin composite is: $V_f = 0.76$, $\sigma_f = 3.44$ GN m^{-2}, $E_f = 85.5$ GN m^{-2}, and $\bar{x} = 1$ mm. Substituting these data into Equation 2, taking $y = 2\bar{x}$, gives $\gamma_d = 52.5$ kJ m^{-2} compared with a value of 105 kJ m^{-2} quoted in [19]. Also, careful examination of the data shows that rather unrealistic σ_f values were employed in the calculation. The experimental mean tensile strength for the composite as given in [19] is 1.45 GN m^{-2}. With V_f being as high as 76% it is reasonable to assume that the ultimate strength of the fibre in the composite would not be much greater than 1.5 GN m^{-2}. With this value for σ_f we obtain $\gamma_d = 10$ kJ m^{-2} and $\gamma_{po} = 190$ kJ m^{-2}. Now, their experimental γ_F value of 121 kJ m^{-2} agrees more closely with γ_{po}, similar to our observation.

Assuming that the main contribution to the value of γ_F is from γ_{po}, and using the relationship $l_c = d\sigma_f/2\tau_i$ to rewrite Equation 1, it is seen that γ_F is inversely proportional to τ_i

$$\gamma_{po} = \frac{V_f \sigma_f^2 d}{48\tau_i} \,. \qquad (6)$$

This seems to contradict the observation that the water-boiled set exhibits lower γ_F values. However, the results in Figs. 3 and 4 indicate that the water-boil treatment should be expected to result in a simultaneous effect on both σ_f and τ_i to reduce their values. This is further supported by the fact that the measured values of l_c ($l_c \propto \sigma_f/\tau_i$) did not change significantly in water-boiled specimens (Section 4.1). Even if τ_i decreases more than σ as a result of boiling, γ_{po} will still be reduced since it is directly proportional to σ_f squared.

Notice that a similar observation in carbon fibre-polyester composites, where σ_f is not affected considerably by the water-boil treatment, was explained differently [18].

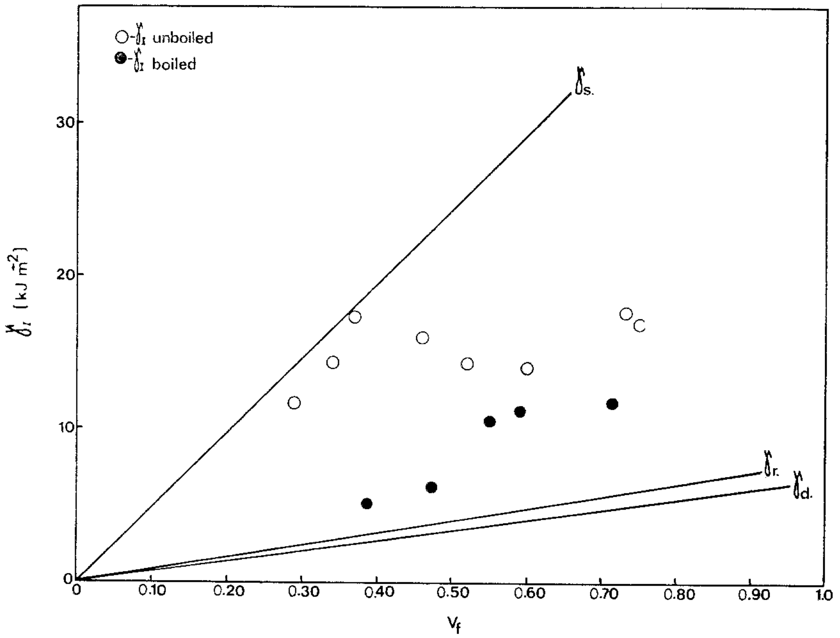

Figure 8 Fracture surface energy of initiation values as a function of V_f. A comparison with theoretical models predicting fracture toughness.

4.3. The fracture surface energy of initiation

Fig. 8 presents the results of fracture surface energy of initiation (γ_I) of the two composite sets with the unboiled set exhibiting higher values. A comparison between γ_F (Fig. 7) and γ_I shows that $\gamma_I \ll \gamma_F$. This finding agrees with the controlled fashion of the observed fracture process.

The results of γ_I are five-fold lower than those of γ_F (Fig. 7), thus, the possibility of contribution from γ_{po} is excluded. Yet, the values set by γ_r or γ_d are too low to account for the experimental data of the unboiled set. It is seen that the surfaces formation model provides the best explanation for the experimental data. It is suspected that when the term γ_m (in Equation 5) is replaced by the true term, γ_{if}, the fracture toughness of the interface (see [9]), an even better agreement will prevail, since it is thought that in the present composite system $\gamma_{if} < \gamma_m$.

The drop in the values of γ_I due to the water-boil treatment may be explained as follows. Assuming that the main contribution to the value of γ_I is from γ_s and rewriting Equation 5 for $l_c/d = \sigma_f/\tau_i$ it results that γ_I is proportional to σ_f/τ_i:

$$\gamma_s = \frac{V_f \sigma_f \gamma_m}{\tau_i} . \tag{7}$$

Since the water-boil treatment affects both σ_f and τ_i (Section 4.2) this treatment is not expected to affect γ_s markedly. However, it is expected that γ_m and even more so γ_{if} deteriorate as a result of such a treatment. This is followed by a drop in γ_s.

5. Conclusions

The major conclusion of the present study regards the correspondence between the proposed fracture toughness models and the experimental values of the work of fracture and of the fracture surface energy for initiation of glass-epoxy composites. γ_F, the average fracture energy of the complete process, is expected to include some contribution from γ_I. However, the finding that γ_F is five-fold greater than γ_I indicates that totally different fracture mechanisms control the stages of fracture initiation and fracture propagation. The very good agreement between the pull-out model and the γ_F results suggests that the stage of fracture propagation is concerned with extracting the broken fibres from the matrix, and dissipating energy against the fibre–matrix friction stress. The general agreement between the γ_I results and the surfaces formation model suggests that the creation of new fibre, matrix and fibre–matrix surfaces is the process which occurs during the stage of fracture initiation. The energy required to create the new surfaces includes that of the debonding process [9].

The above conclusions are in a general

agreement with results by Beaumont and Phillips [2] for glass fibre–polyester composites of $V_f = 0.15$. These authors postulate that γ_I is determined by a debonding mechanism, while γ_F is the sum of a debonding mechanism plus pull-out contribution.

The very good agreement of γ_F values with γ_{po} based on our observed $l_c = 2.3$ mm strengthens the hypothesis brought forward in Section 4.1, that this l_c value applies to fracture, and cannot be replaced by l_c values obtained from different mechanical or physical testing.

Long-term exposure to boiling water of glass fibre–epoxy resin composites results in a prominent decrease in the fracture toughness and the mechanical properties excluding the modulus. The decrease in the values of γ_F and γ_I is attributed to a simultaneous destruction of the fibre–matrix bond and deterioration of the fibre strength, and also, in the case of γ_I to a drop in the fracture toughness of the matrix, γ_m and the interface, γ_{if}.

References

1. G. A. COOPER, *J. Mater. Sci.* **5** (1970) 645.
2. P. W. R. BEAUMONT and D. C. PHILLIPS, *ibid* **7** (1972) 682.
3. M. FILA, C. BREDIN and M. R. PIGGOTT, *ibid* **7** (1972) 983.
4. C. D. ELLIS and B. HARRIS, *J. Composite Mater.* **7** (1973) 76.
5. J. O. OUTWATER and M. C. MURPHY, 24th Annual Technical Conference, Composites Division, Society of Plastics Industry, Inc. Paper IIc (1969).
6. A. H. COTTRELL, *Proc. Roy. Soc.* **A282** (1964) 2.
7. M. PIGGOTT, *J. Mater. Sci.* **5** (1970) 669.
8. J. FITZ-RANDOLPH, D. C. PHILLIPS, P. W. R. BEAUMONT and A. S. TETELMAN, Paper presented at the 5th St. Louis Symposium on Advanced Fibre Composites (1971).
9. T. U. MARSTON, A. G. ATKINS and D. K. FELBECK, *J. Mater. Sci.* **9** (1974) 447.
10. H. G. TATTERSALL and G. TAPPIN, *ibid* **1** (1966) 296.
11. W. F. BROWN and J. E. SRAWLEY, ASTM, STP410 (1967).
12. L. R. CALCOTE, "The Analysis of Laminated Composite Materials" (Van Nostrand Reinhold, USA, 1969).
13. S. W. TSAI, NASA, CR-71 (July 1964).
14. M. G. BADER and W. H. BOWYER, *J. Phys. D: Appl. Phys.* **5** (1972) 2215.
15. P. HANCOCK and R. C. CUTHBERTSON, *J. Mater. Sci.* **5** (1970) 762.
16. W. REYNOLDS and N. L. HANCOX, *J. Phys. D: Appl. Phys.* **4** (1971) 1747.
17. A. KELLY, *Proc. Roy. Soc. Lond.* **A319** (1970) 95.
18. B. HARRIS, P. W. R. BEAUMONT and E. MOCUNILL DE FERRAN, *J. Mater. Sci.* **6** (1971) 238.
19. P. W. R. BEAUMONT and D. C. PHILLIPS, *J. Composite Mater.* **6** (1972) 32.
20. G. MAROM and E. F. T. WHITE, *J. Mater. Sci.* **7** (1972) 1299.
21. G. MAROM and A. WEINBERG, *ibid* **10** (1975) 1005.

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