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Hybrid effects in composites: conditions for positive or negative effects versus rule-of-mixtures behaviour

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A positive or negative hybrid effect in hybrid composites is defined as a positive or negative deviation of a certain mechanical property from the rule-of-mixtures behaviour. The question of hybrid effects is first examined with special hybrids which have been chosen so that the effect of the fibre–matrix interface is minimized. The hybrids examined consisted of two types of carbon fibres with different mechanical properties but similar surface treatments. The results of all the mechanical properties examined (modulus, strength, stress intensity factor, fracture energies) under quasi-static and fast testing conditions do not show any synergism. In view of these results a second hybrid system of E-glass fibre/AS carbon fibre-reinforced epoxy has been chosen. In this system both the mechanical properties of the fibres and the interface which they form with the resin are entirely different. None of the mechanical properties, excluding the fracture energies, show any signs of a hybrid effect. The fracture energy results, however, show the existence of a negative hybrid effect. A theory which sets upper and lower bounds for the hybrid effect is proposed, and the conditions for the occurrence of either a positive or a negative effect are discussed.

1. Introduction

Hybridization with more than one fibre type in the same matrix provides another dimension to the potential versatility of fibre-reinforced composite materials. It would seem possible with hybrid composites to have greater control of specific properties, achieving a more favourable balance between the advantages and disadvantages inherent in any composite material. Thus, for example, by combining two types of fibres a reduction in modulus might be acceptably traded for increased fracture resistance and reduced cost for the composite.

Of considerable interest is the quantitative description of the behaviour of hybrid composites as affected by the amount and type of hybridization. Phillips [1] reviews the lively controversy surrounding the suggestions of possible synergistic hybrid effects, in which the properties of the hybrid composite might not follow from a direct consideration of the independent properties of the individual components. A positive or negative hybrid effect can be defined as a positive or negative deviation of a certain mechanical property from the rule-of-mixtures behaviour, respectively. It is commonly agreed that the elastic moduli follow rule-of-mixtures behaviour [2–4]. In glass/carbon-reinforced epoxy systems, hybrid effects are reported for the strain [2, 3, 5] and the work of fracture [6, 7]. The hybrid effect for the strain has been observed as an increase in the maximum linear strain, as well as an increase in the maximum strain at failure, when comparing the respective strains for the two-phase composite containing the stiffer fibre. The ultimate strength is reported to exhibit a negative hybrid effect [2, 8], sometimes as a result of the synergistic improvement in the maximum strains [2]. Harris and Bunsell [4] did not observe a hybrid effect for the work of fracture in a glass/carbon hybrid; however, the arrangement of the fibres within their hybrid (intimate contact) is significantly different from that examined in references [6, 7] (distinct layers).

In this paper, we investigate some possible parameters, other than the most commonly studied relative volume fraction of fibre types, which might control the hybrid effect in unidirectional three-phase hybrid composites over a broad range of loading rates. These include the relative moduli and strengths of the fibres, the nature of the fibre–matrix interface, and the arrangement of the fibres within the composite. Comparisons are made between the experimentally observed values and predictions based on the rule-of-mixtures for the hybrid modulus, fracture strength, fracture toughness, and fracture energies. Positive and negative deviations from the rule-of-mixtures for the fracture energies can be described by a model defining upper and lower bounds for the hybrid effect.

2. Experimental

2.1. Materials

Carbon/carbon hybrids (c/c) were constructed from alternating unidirectional "prepegs" of either HMS/AS (Courtaulds Ltd) or Type I treated/Type II treated (Fothergill and Harvey Ltd). Unidirectional glass/carbon hybrids (g/c) were constructed from "prepregs" of E-glass fibre and AS-carbon fibre. Three types of such hybrids were prepared and designated by 1/1, 2/2 and 5/5, describing the number of sequential layers of "prepregs" of each reinforcement in the hybrid. A rather special technique for the preparation of the 1/1 hybrid was adopted. According to this technique the carbon fibre "prepregs" were laid on a winding machine drum, and the glass fibre was wound and impregnated by an acetone solution of the epoxy resin on the carbon fibre "pregreg". The epoxy solution which plasticized the resin of the "prepreg" and the tension on the wound glass fibre helped the latter penetrate between the carbon fibres and form an intimate fibre mixture. The "prepregs" were cut to size, laid in a mould and pressed for 20 min at 175° C under 0.69 GPa to form 0.6 cm thick plates, which were additionally postcured at 180° C for 2 h. The total volume fraction of the fibres (V_f) in the Type I/Type II hybrids was 60% and that of the HMS/AS hybrids and of the g/c hybrids was 50%. In each hybrid the relative volume fractions of the constituent fibres were equal (50%).

2.2. Testing

The specimens, oriented for deformation in the translaminar mode, were tested in three-point bending. Slow and fast loading rates of 8.3×10^{-6} m sec^{-1} and 1.3 m sec^{-1} were obtained using an Instron Testing Machine and DYNATUP Instrumented Impact System, respectively. Notched and unnotched specimens were tested and five specimens were used to obtain an average for each type of test.

The unnotched specimens had a cross-section of 0.6 cm width and 0.5 cm depth and were tested over a loading span of 8.0 cm for the slow and 4.0 cm for the fast loading rates, respectively. The notched specimens for both slow and fast loading rates had cross-sectional dimensions as above and contained a 0.1 cm wide × 0.2 cm deep notch. The loading span was 4.0 cm for all notched specimens. For one group of samples, i.e., Type I/Type II materials, the dimensions of the test specimens were different as following: notched and unnotched specimens were 0.8×0.8 cm^2 in cross-section, and tested over a loading span of 4.0 cm; notched specimens contained a 0.1 cm wide × 0.3 cm deep notch. These different dimensions prevent a comparison between this group of materials to the other groups examined. However, a comparison between samples within the group is significant as shown later.

The flexural modulus and strength were calculated using linear elastic beam theory from the unnotched specimen data. The results from the notched specimens were used to calculate the fracture toughness by the equation $K_{IC} = Y\sigma c^{1/2}$, where Y is a function of the specimen geometry, σ is the ultimate stress at fracture, and c is the notch length. The work of fracture (γ_F) was also determined with notched specimens from the ratio of the integrated fracture energy to the total new fracture surface area. The fracture surface energy (γ_I) was calculated through the standard relationship $\gamma_I = K_{IC}^2/2E$. Although E in the last relationship is a complex function of the elastic constants of the composite, we used in our calculations either the experimental value of the longitudinal modulus of the composite, or the value calculated by the simple rule-of-mixtures. This is based on findings by Sih *et al.* [9], who compared the use of the orthotropic model, i.e., the use of the complex E, with the use of the isotropic homogeneous relationship between K_{IC} and γ_I. They showed that the isotropic model provided the best model for glass fibre composites up to volume fractions of 60%, while both models resulted in similar discrepancies for graphite fibre specimens. In addition to the above properties, γ_σ, the integral of the load–deflection curve to maximum load per new fracture surface area, and the ductility index $(\gamma_F/\gamma_\sigma - 1)$ were also calculated.

3. Results and discussion

The results of all the mechanical properties examined (modulus, strength, fracture toughness, fracture energies) under quasi-static and fast testing conditions are presented in Tables I and II. For each type of reinforcement the properties of the two-phase composites are presented followed by the properties of the hybrid. The designation 1/1, 2/2, or 5/5 describe the number of sequential layers of "prepregs" of each reinforcement in the hybrid. The coefficients of variation for the results in Tables I and II were generally about 0.1 for $(\sigma, K_{IC}$, E and γ_F and 0.1–0.2 for γ_σ. In the next sections the results for the two types of hybrids, i.e., carbon/carbon (c/c) and glass/carbon (g/c) are analysed. Also, the strain-rate dependence of the properties is examined by comparing between slow and fast testing results.

3.1. c/c hybrids

These hybrids which comprise two types of carbon fibres have been chosen so that the effect of the fibre–matrix interface is minimized. In these composites the surfaces of the constituent fibres are similar, while their mechanical properties differ greatly. Hence, the source of any possible hybrid effect reduces to the different mechanical properties of the fibres. However, Table I presents a general picture with a few exceptions whereby values of the mechanical properties, excluding γ_I, of the hybrids generally obey the rule-of-mixtures; and since the hybrids contain 50% of each type of fibre, a mechanical property in question is an aver-

TABLE I Results of the carbon fibre hybrids.

Reinforcement	σ (GPa)		E (GPa)		K_{IC} (MN m$^{-3/2}$)		γ_I(kJ m^{-2})		γ_σ(kJ m^{-2})		γ_F(kJ m^{-2})	
	slow	fast	slow	fast	slow	fast	slow	fast	slow	fast	slow	fast
Carbon fibre Type I	0.50	0.43	206*	–	12.0	10.9	0.35	0.30	1.4	1.9	3.6	3.0
Carbon fibre Type II	0.90	0.95	124*	–	47.2	51.6	9.0	10.8	22.2	26.5	27.6	43.6
Hybrid 1/1	0.70	0.66	165*	–	27.7	33.8	2.3	3.4	10.2	14.2	16.0	21.1
Rule-of-mixtures	0.70	0.69	165	–	29.6	31.3	4.7	5.5	11.8	14.2	15.6	23.3
Carbon fibre HM-S	0.85	0.53	137	–	31.4	30.4	3.6	3.4	11.8	11.5	27.6	32.3
Carbon fibre A-S	1.2	1.1	97	–	50.8	55.8	13.3	16.0	33.5	38.1	38.0	46.4
Hybrid 1/1	0.80	0.60	117	–	35.4	39.0	5.4	6.2	13.9	21.1	34.4	38.0
Rule-of-mixtures	1.03	0.82	117	–	41.1	43.1	8.5	9.7	22.7	24.8	32.8	39.4

* Calculated by the rule-of-mixtures

TABLE II Results of the glass/carbon hybrids.

Reinforcement	σ (GPa)		E (GPa)		K_{IC} (MN m$^{-3/2}$)		γ_I(kJ m^{-2})		γ_σ(kJ m^{-2})		γ_F(kJ m^{-2})		$\gamma_F/\gamma_\sigma^{-1}$	
	slow	fast	slow	fast	slow	fast	slow	fast	slow	fast	slow	fast	slow	fast
Glass fibre E	0.70*	–	25*	–	27.0*	39.7	14.6*	31.5	27.6	64.2	54.0	134.6	1.0	1.1
Carbon fibre A-S	1.2	1.1	97	–	50.8	55.8	13.3	16.0	33.5	38.1	38.0	46.4	0.1	0.2
Hybrid 1/1	0.85	–	61	–	34.7	35.7	9.9	10.5	21.7	29.9	30.6	60.6	0.4	1.0
Hybrid 2/2	0.83	–	61	–	32.4	29.5	8.6	7.1	17.8	25.4	27.9	51.7	0.6	1.0
Hybrid 5/5	0.80	–	62	–	34.7	35.6	9.7	10.2	20.6	19.0	46.0	82.0	1.2	3.3
Rule-of-mixtures	0.85	–	61	–	38.9	43.8	13.9	23.8	30.6	51.2	46.0	90.8	0.55	0.57

* Taken from reference [10]

age of the corresponding properties of the two-phase composites. The exceptions are the lower than the rule-of-mixtures strength of the 1/1 HMS/AS hybrid, which is expected [2, 8], and the slow testing γ_σ value of the same material.

With regard to the value of γ_I, since γ_I is related to the modulus and the stress intensity factor by $\gamma_I = K_{IC}^2/2E$, and since K_{IC} and E of the hybrid are determined by the rule-of-mixtures, it can easily be shown that γ_I of the hybrid must be smaller than the rule-of-mixtures value. For the present hybrids, γ_I is given by the expression

$$\gamma_I = \frac{(K_{I1} + K_{I2})^2}{4(E_1 + E_2)} \tag{1}$$

where the subscripts 1 and 2 denote the two types of fibres, respectively. This expression results in a γ_I value of 2.4 kJ m^{-2} for the first hybrid and of 7.2 kJ m^{-2} for the second. Both values are smaller than the rule-of-mixtures values, and they generally agree with the respective experimental results of 2.3 and 5.4 kJ m^{-2}.

The above observations are important in the sense that the discussion on the existence of a hybrid effect is now focused on the fibre–matrix interface as a possible cause for such an effect. This point is strengthened even further by some previous observations made on a three-phase composite system comprising glass beads, glass fibres and epoxy resin [10], and on another system of asbestos whiskers, glass fibres and epoxy resin [11]. In principle such systems are hybrids where the two types of reinforcements also vary by their geometry. In both cases it was observed that, by and large, the mechanical properties of the three-phase system were determined by proportional contributions from the constituent materials, including processes which take place in the individual two-phase systems. This again is a rule-of-mixtures behaviour.

3.2. g/c hybrids

In view of the failure to observe any hybrid effect as a result of the different mechanical properties of the two types of reinforcements, when they form an approximately identical fibre–matrix interface, a second system has been chosen. In this system of epoxy reinforced by E-glass fibre/AS-carbon fibre both the mechanical properties of the two kinds of fibres and the two types of fibre–matrix interfaces are different.

Examination of Table II reveals that the moduli of the hybrids coincide with the rule-of-mixtures value, while σ exhibits values which are slightly below the rule of mixtures prediction. These observations are consistent with those made in references [2, 8]. Table II also shows that K_{IC} of the hybrids as well are smaller than the rule-of-mixtures. It is evident that E, σ, and K_{IC} do not depend on the construction of the layers in the hybrid. These observations are very significant since they show that even a complex system which contained entirely different fibres does not result in a synergistic effect as far as the above properties are concerned.

An entirely different behaviour is exhibited by the values of the fracture energies. The observed values of γ_I, γ_σ, and γ_F are smaller than the respective values of the corresponding two-phase glass fibre or carbon fibre composites. This, in fact, suggested that a negative hybrid effect governs the fracture behaviour of the g/c hybrids. This phenomenon of a negative hybrid effect can best be explained through examining the γ_F values. Assuming that the main contribution to the work of fracture for either glass fibre [12] or carbon fibre composites [13] is from pull-out (γ_{po}), then the rule-of-mixtures expression for γ_F of the hybrid is

$$\gamma_F^h = \gamma_{po}^h = \frac{V_{f1}\sigma_{f1}l_{c1}}{24} + \frac{V_{f2}\sigma_{f2}l_{c2}}{24} \tag{2}$$

where V_{f1} and V_{f2} are the volume fractions of the two types of fibres so that $V_f = V_{f1} + V_{f2}$, σ_{f1} and σ_{f2}, and l_{c1} and l_{c2} are the strength and the critical length of the two fibres, respectively. When $l_{c1} \gg l_{c2}$ various possibilities for the pull-out lengths of the constituent fibres in the hybrid exist. In one extreme case the pull-out length of the glass will reduce to that of the carbon fibre, and in the other extreme case the pull-out length of the carbon fibre will increase to that of the glass fibres. These two extreme cases result in lower and upper bounds for the hybrid effect as expressed by Equations 3 and 4

$$\text{lower bound, } \gamma_{po}^h = \frac{V_{f1}\sigma_{f1}l_{c2}}{24} + \frac{V_{f2}\sigma_{f2}l_{c2}}{24} \tag{3}$$

$$\text{upper bound, } \gamma_{po}^h = \frac{V_{f1}\sigma_{f1}l_{c1}}{24} + \frac{V_{f2}\sigma_{f2}l_{c1}}{24} \tag{4}$$

A negative hybrid effect will yield a γ_F value between the rule-of-mixtures value and the lower bound, while a positive hybrid effect will result in

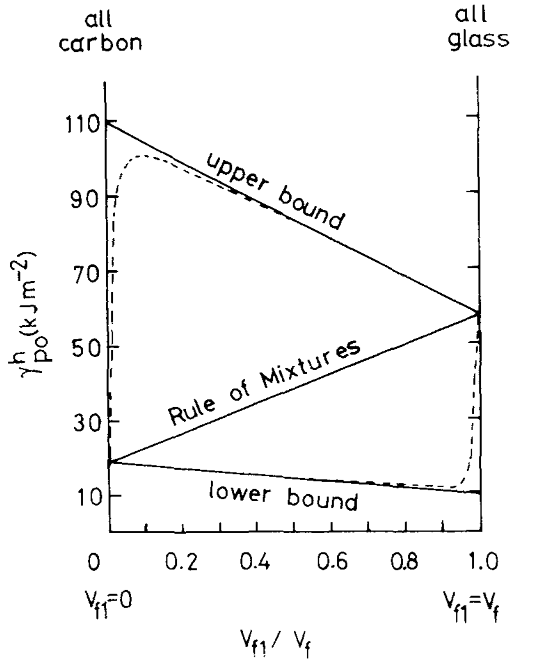

Figure 1 Upper and lower bounds for the work of fracture of the hybrids as a function of the relative fibre content.

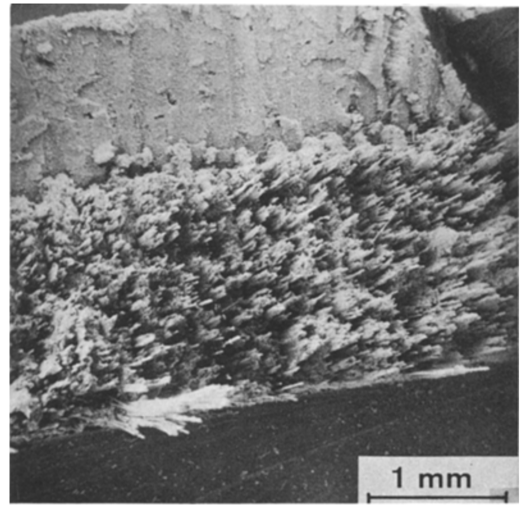

Figure 2 A low magnification scanning electron micrograph of the fracture surface of the g/c 1/1 hybrid, showing the notch (at the bottom), the pull-out zone, and the compressive fracture zone.

a γ_F value between the rule-of-mixtures value and the upper bound.

As an example, let us consider a hybrid of $V_f = 0.50$ composed of E-glass fibre and AS-carbon fibre, where for the first fibre $l_{c1} = 23.0 \times 10^{-4}$ m and $\sigma_{f1} = 1.2$ GN m^{-2} [12], and for the second $l_{c2} = \sim 4.0 \times 10^{-4}$ m and $\sigma_{f2} = 2.3$ GN m^{-2}. The results are shown in Fig. 1, where the broken lines indicate the expected real behaviour near the points of discontinuity of $V_{f1} = 0$ and $V_{f1} = V_f$.

Similar consideration may apply to γ_I, whereby, assuming that the contribution to its value comes from the debonding mechanism, bounds can be worked out according to the different debonded lengths of each type of fibre.

The experimental results provide sufficient evidence to support the above analysis. Fig. 2 is a low magnification scanning electron micrograph of the fracture surface of a 1/1 g/c hybrid. It shows the notch (at the bottom of the picture), the pull-out zone, and the compressive fracture zone with its typical features [14]. We concentrate our attention on the pull-out zone which is relevant to the above analysis. Fig. 3 is a scanning electron micrograph of the pull-out zone. It is possible to distinguish between the two types of fibre by their different diameters (the glass fibre is wider), however it is difficult to trace any difference in the pull-out lengths of the two fibres. Fig. 3 also shows that due to the special preparation technique, by which the glass fibres were wound and impregnated on the carbon fibre "prepregs", the fibres in the 1/1 hybrid are intimately mixed. Fig. 4 is a picture of the pull-out fibres taken with an optical microscope at right angle to the fibres near the bottom of the notch. Here it is easier to distinguish between the glass and the carbon fibres by their different colours. Fig. 4 clearly shows that the pull-out length of the glass fibres is smaller than expected, and in fact it is even smaller than that of the carbon fibres. Thus the reason for the observed negative hybrid effect is the lower pull-out length of the glass fibres, which is caused by the fact that in the 1/1 g/c hybrid the glass fibres are in close contact with the carbon fibres.

This leads to the idea that a positive hybrid effect is possible in hybrids in which the layers of the different reinforcements are more distinct and segregated. The results of the 5/5 g/c hybrids are in accord with this idea. Figs. 5–7 present various details of the fracture surface of the 5/5 g/c hybrid. Fig. 7 shows that near the tip of the notch the pull-out length of the glass fibres is 3–4 times bigger than that of the carbon fibres. However, the pull-out length of the glass decreases while that of the carbon increases gradually, as the fracture propagates across the specimen. In fact, near the

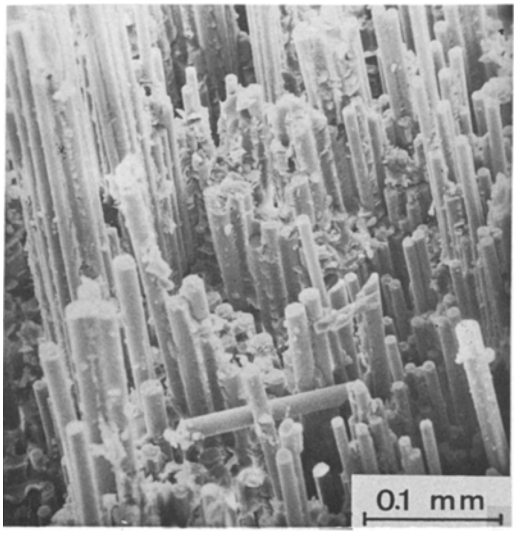

Figure 3 Scanning electron micrograph of the pull-out zone of the g/c 1/1 hybrid, showing an intimate mixture of glass and carbon fibres.

compressive fracture zone the pull-out of the carbon is slightly bigger than that of the glass fibre. On average, the pull-out of the glass is longer than that of the carbon fibre, indicating that γ_F of the hybrid will be closer to the rule-of-mixtures value, as indeed is the case (Table II). This is also seen by examining the load–deflection curves of the hybrids in Fig. 8, or by comparing the ductility indices ($\gamma_F/\gamma_\sigma - 1$) in Table II. More energy is required for fracture propagation in the 5/5 g/c hybrid, and it is related to its higher glass fibre pull-out lengths. Recent results by Mallick and Broutman [7] and by Steg and Tuler [6] show that when the hybrid is composed of a laminated structure with only a few distinct and thick layers of each type of reinforcement, γ_F exhibits a significant positive hybrid effect, and the shape of the experimental curve of γ_F as a function of glass fibre content resembles that of the theoretical upper bound proposed here [6].

It is important to note that the original hypothesis of the importance of the fibre arrangement in the hybrid was raised by Harris and Bunsell [4]. They even suspected that hybrids in which the different types of reinforcements were more intimately mixed would not perform as well in terms of fracture resistance. The results reported above based on the comparison between the behaviours of the 1/1 and the 5/5 hybrids prove their hypothesis correct. Also, whereas the variation of the pull-out length of the glass fibres is reported here to result from the variation of the layer sequence, a similar variation is reported by Harris and Bunsell to occur as the relative fibre volume content is varied. Thus, the occurrence of a negative or a positive hybrid effect will depend on these two factors, i.e., on the arrangement of the fibres within the hybrid and on their relative volume fraction.

3.3. Strain-rate effect

The strain-rate dependence of the mechanical properties of glass fibre-reinforced epoxies was recently studied and discussed [15]; the corresponding results in Table II entirely agree with that study. With regard to the carbon fibre-reinforced composites, Table I shows that σ is probably independent of strain-rate because the differences between the fast and slow testing values may be insignificant in view of the five orders of magnitude difference between the slow and fast conditions. Hence, by analogy with the

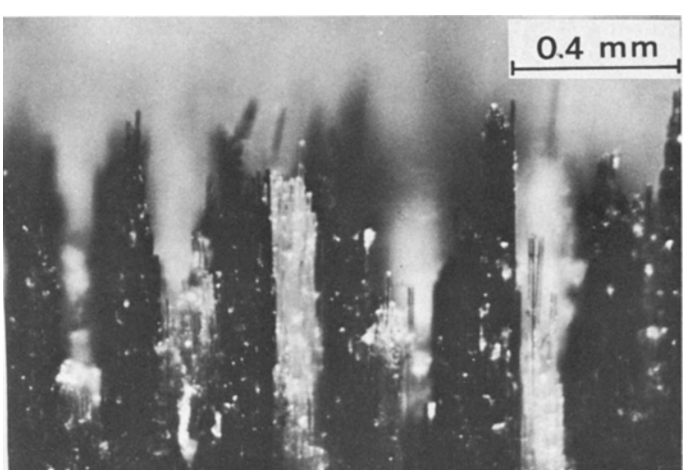

Figure 4 The pull-out zone of the g/c 1/1 hybrid near the tip of the notch viewed at a right angle to the fibres. The pull-out length of the carbon fibres is bigger than that of the glass fibres.

considerations raised previously [15], and since γ_I and K_{IC} are related to σ, they are also considered not to be significantly dependent on the strain-rate. However, γ_F is strain-rate dependent as already shown in [16]. The values of γ_F of the carbon-fibre composites increase by an average of about 20% as the testing speed is increased by five orders of magnitude. Although this increase is significant it is small compared with that of about threefold exhibited by the glass fibre-reinforced composites. In view of γ_F being determined by γ_{po} which is time-dependent [15] it is maintained that the reason for the difference between the glass and the carbon composites is the much smaller pull-out length of the latter.

Regarding the above discussion, the experimental fracture energy results show clearly that the carbon fibres dominate the strain-rate dependence of the hybrid at the stage of fracture initiation (i.e., γ_I and γ_σ), while the glass fibres dominate its strain-rate dependence at the stage of fracture propagation (indicated by γ_F). Thus γ_F and the ductility index of the hybrid are strongly dependent on the strain-rate and exhibit a twofold increase between slow and fast testing conditions, while γ_I and γ_σ are strain-rate insensitive.

4. Conclusions

The main conclusions can be summarized as follows. Among the mechanical properties examined, only the fracture energies exhibit a hybrid effect. A prerequisite for the occurrence of such an effect is that the two types of fibres will differ by both their mechanical properties and by the interface which they form with the matrix. The hybrid effect may be positive or negative according to the

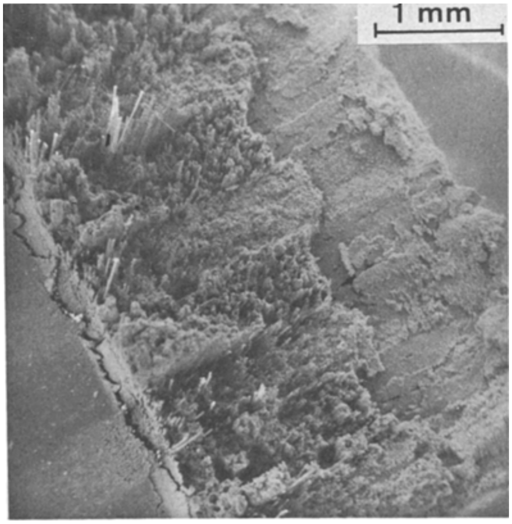

Figure 5 A low magnification scanning electron micrograph of the fracture surface of the g/c 5/5 hybrid.

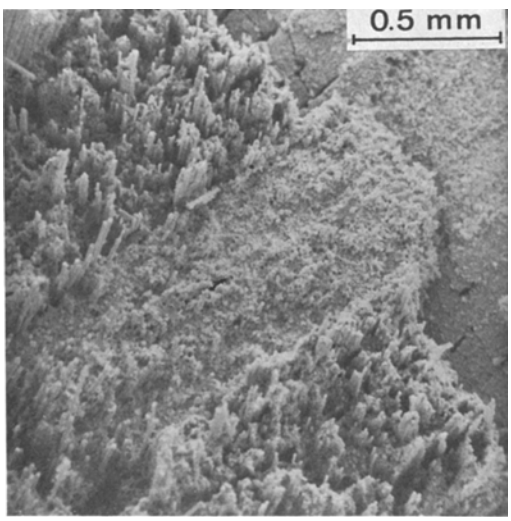

Figure 6 Scanning electron micrograph of the pull-out zone of the g/c 5/5 hybrid, showing the distinct glass and carbon layers.

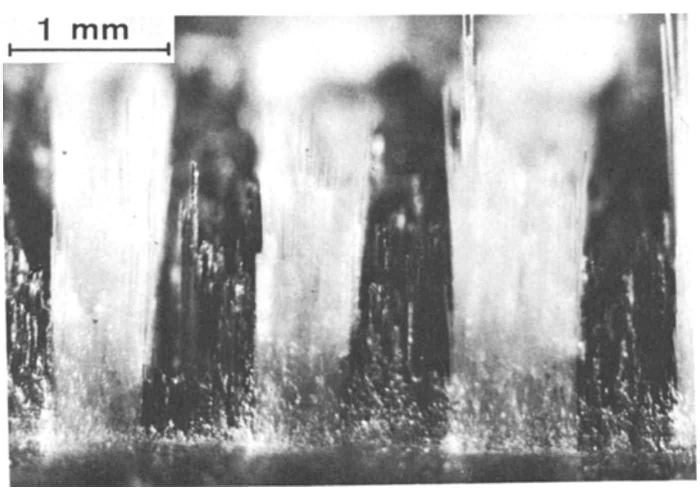

Figure 7 The pull-out zone of the g/c 5/5 hybrid near the tip of the notch viewed at a right angle to the fibres. The pull-out length of the glass fibres is bigger than that of the glass fibres.

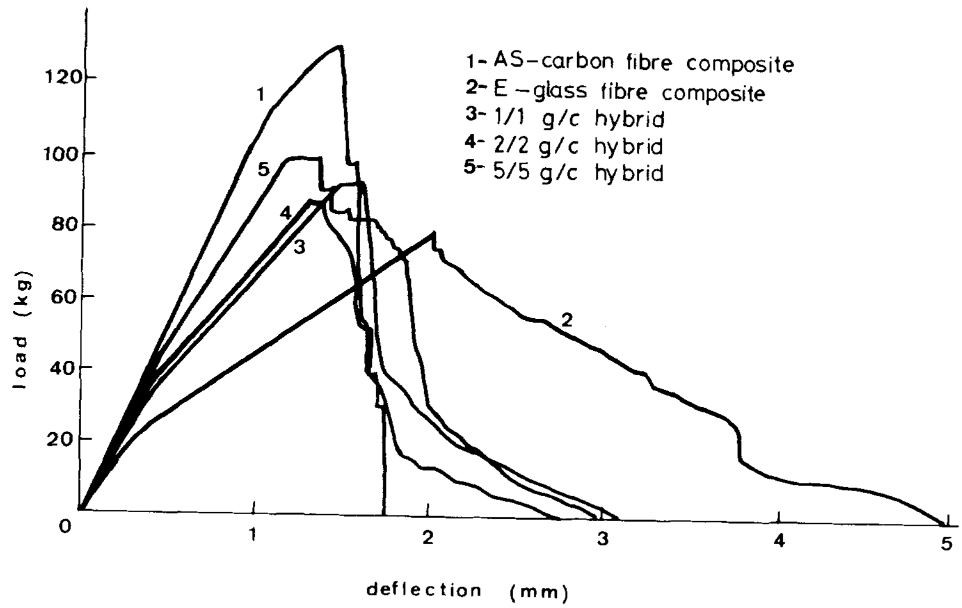

Figure 8. Load–deflection curves of glass fibre and carbon fibre composites and hybrids.

relative volume fraction of the two types of fibres, the construction of the layers in the hybrid, and, presumably, according to the loading configuration (e.g., translaminar or interlaminar).

The strain-rate dependence of the mechanical properties of the carbon fibre composites is very small compared with that of the glass fibre composites. The strain-rate dependence of the fracture energies of the hybrids is governed at the stage of fracture initiation by the carbon fibres, while at the stage of fracture propagation the glass fibres control the strain-rate dependence of the hybrid.

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