JOURNAL OF MATERIALS SCIENCE 9 (1974) 1420–1428

# The influence of the interface on the strength and elastic properties of low aspect ratio fibre composites

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The strength and elastic properties of randomly distributed low aspect ratio fibre composites were investigated. The fibre-matrix interfacial conditions were varied by applying various surface treatments to the fibres. The influence of the interfacial bond on the strengths of such composites was demonstrated.

## 1. Introduction

The purpose of the present study was to investigate the influence of interface on the strength and elastic properties of a brittle matrix composite containing fibres of small aspect ratios (length to diameter ratios). The short fibres were dispersed in the matrix with a three-dimensional random orientation. The composite material thus obtained was macroscopically homogeneous and isotropic.

There have been numerous experimental investigations on the strength and elastic properties of aligned short fibre composites. However, the literature on randomly oriented short fibre composites is limited. Sambell *et al.* [1] reported the strength of both partially aligned and randomly oriented short carbon and zirconia reinforced brittle ceramic and glass composites. When the fibres were randomly oriented, the composites had lower strengths than the unfilled matrix materials and the strength decreased continuously with increasing fibre volume fraction. With partially aligned fibres, the strength increased with fibre volume fraction after an initial decrease up to about 10 vol %. Trachte and Dibenedetto [2] reported the properties of randomly oriented short glass and graphite fibre reinforced composites. The matrix, in their study, was a ductile polymer, polyphenylene oxide. The ultimate strength of the untreated glass fibre composite increased linearly up to about 20% volume fraction of fibres and then decreased slightly. The ultimate tensile strength and elongation at break were found to be a sensitive function of temperature and interfacial adhesion.

In this study, a brittle epoxy resin was selected as the matrix material. The reinforcing elements were milled "E" glass fibres. The interfacial bond between the glass fibres and the brittle polymer matrix was varied by applying various surface treatments to the fibres. The mechanical properties of the composites were determined experimentally, and the fracture surfaces were studied extensively using scanning electron microscopy. An attempt was made to correlate the experimental observations with theoretical models.

## 2. Material description

Epon 828 (Shell Chemical Co) was selected as a representative brittle epoxy resin in this study. The curing agent used was Curing Agent Z (also Shell Chemical Co). The milled "E" glass fibres were made by Owens Corning Fiberglas. The number average $l/d$ (length to diameter) ratio, also called the fibre aspect ratio, of the fibres was determined to be 8. This was measured by first taking a series of photographs of the fibres at × 100 magnification and then counting from these photographs the number of fibres in each fibre length interval of 21 μm. Fig. 1 shows a frequency distribution curve of the $l/d$ ratios of the glass fibres under consideration. The average diameter of the fibres (also determined from the photographs) was 13 μm.

The glass fibres, as received from the manufacturer, were coated with a commercial binder

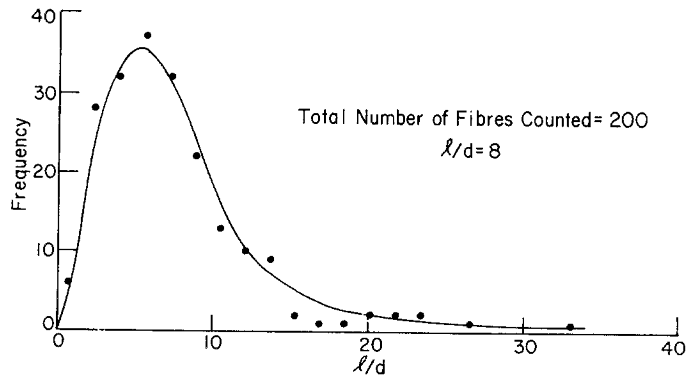


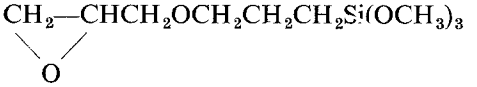
*Figure 1* Frequency distribution of *l/d* ratio.

(Binder 701). In order to remove this binder without degrading the fibres, they were heat-cleaned in an oven at 260°C for 2 h. After heat-cleaning, the following surface treatments* were applied to the fibres:

1. Z-6020: *n*(trimethoxysilylpropyl) ethylene-diamine,

$H_2N(CH_2)_2NH(CH_2)_3Si(OCH_3)_3$

2. Z-6040: γ-glycidoxypropyltrimethoxysilane,



$CH_2\text{—}CHCH_2OCH_2CH_2CH_2Si(OCH_3)_3$ (with O bridging $CH_2$ and $CH$)

3. Z-6076: γ-chloropropyltrimethoxysilane,

$ClCH_2CH_2CH_2Si(OCH_3)_3$

4. XF-13-563: polydimethylsiloxane, a silicone fluid.

The first three surface treatments are commercially available coupling agents which, when used with compatible resin systems, improve the adhesive bond strength of the glass-resin interface. Z-6020 and Z-6040 treatments are known to be used with epoxy and other thermosetting resins, whereas Z-6076 treatment is primarily used with thermoplastics. The XF-13-563 compound, on the other hand, is a mould release treatment and is intended to prevent adhesion between the glass and the resin. In addition to the fibres with the four surface treatments mentioned above, heat-cleaned (no treatment) and as-received (Binder 701) fibres were also used to prepare the composite plates. Thus, in total, six different interface conditions with varying degrees of adhesive bond strength were used in the present study.

The pre-heated resin and glass fibres were mixed with a hi-torque stirrer for about 10 min before adding the curing agent to the mix. The mixing was then continued in a vacuum chamber for at least 30 min. After thorough vacuum blending, the mix was poured into a 10 in. × 10 in. × 1/8 in. aluminium mould. In order to obtain a uniform distribution and avoid any settling of fibres, the mould was rotated at 2 to 3 rpm in a rotation fixture for approximately 20 h. During this time, the resin mix was transformed into a solid gel (B-stage). For complete curing, the mould was placed in a hydraulic press at 60°C for 2 h and then at 148°C for another 2 h.

## 3. Experiments and results

The flexural properties of the unfilled and filled epoxy resins were measured by three-point flexural tests (span to depth ratio = 16) on an Instron Universal Testing machine. All tests were performed at room temperature and at a deflection rate of 0.05 in $\text{min}^{-1}$. Figs. 2 and 3 show the typical stress-strain curve obtained from the flexural tests of the present composite systems.

Flexural strengths of the composite systems are shown in Fig. 4 as a function of fibre volume fraction. Each point on Fig. 4 represents an average of at least five observations. Table I shows the range of strength values obtained in the flexural tests. The ultimate flexural strengths for all composites except those with Z-6040 treated fibres decrease first with volume fraction, exhibit a minimum value and then increase again. The volume fraction at which the minimum strength is exhibited depends on the surface treatment. The mould release treatment and

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*All the surface treatment compounds were supplied by Dow Corning Corporation, Midland, Michigan, USA.

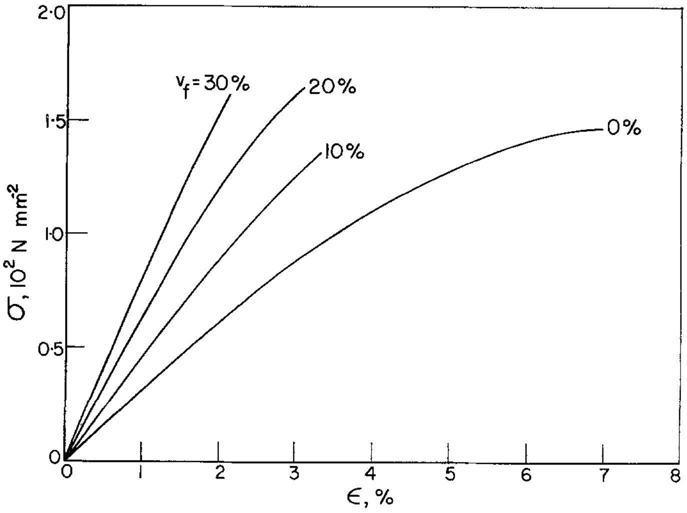

*Figure 2* Typical flexural stress-strain curves of Z-6020 treated glass fibre composites.

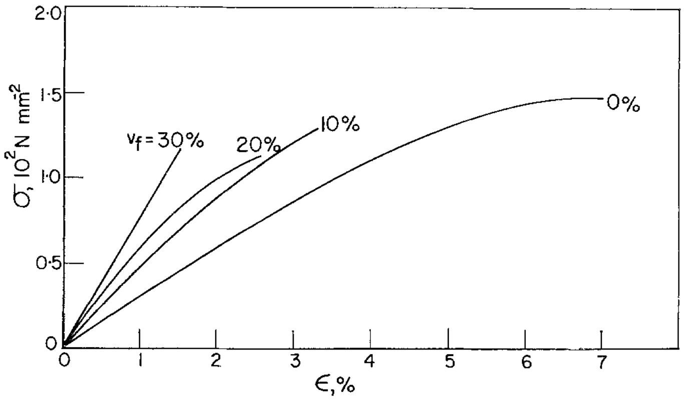

*Figure 3* Typical flexural stress-strain curves of mould release agent treated glass fibre composites.

Z-6076 treatment appear to have the most severe effect on the flexural strength. The untreated, heat-cleaned fibres also give low values of flexural strength. The strength values for these composites are lower than the strength of the unfilled epoxy at all volume fractions studied. The fibres in the as-received form (Binder 701) also lower the composite strength at low fibre volume fraction, but the strength recovers at higher volume fractions. In the case of Z-6020 treatment, the strength decreases up to about 10 vol %, then increases and even exceeds the unfilled epoxy strength. At about 20%, the composite strength exceeds the unfilled epoxy strength by about 10%; however, the strength does not increase any further. The Z-6040 treated fibres increase the composite strength only slightly over the unfilled epoxy strength, but unlike other treatments there is no decrease in strength at any vol %.

The variation in flexural strength with fibre volume fraction indicates that there is a minimum volume fraction below which the strength decreases and above which it increases with increasing fibre volume fraction. For matrix reinforcement to occur, a critical volume fraction is needed so that the composite strength is higher than the unfilled matrix strength. Both of the minimum and critical volume fractions are strongly dependent on the type of surface treatment used.

Although, even the best of the surface treatments did not show any major improvement of strength, it can be concluded from the above results that surface conditions of the fibres do have influence on the flexural strength of the

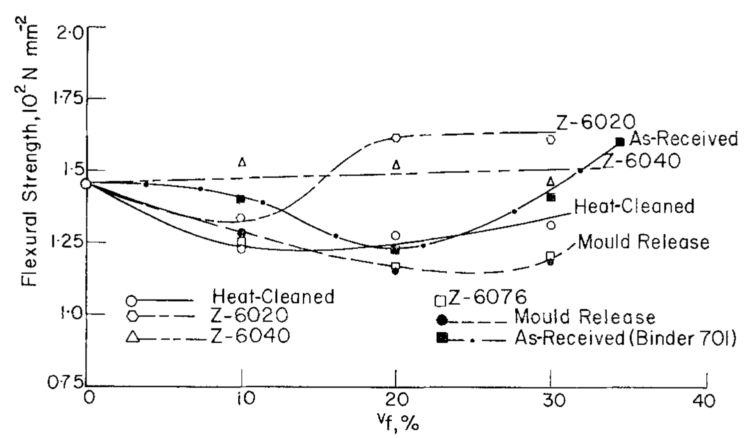

*Figure 4* Flexural strengths of glass fibre composites.

TABLE I Flexural strength data

| Treatment | $v_f$ (%) | Flexural strength | |
|---|---|---|---|
| | | Range (10³ psi) | Average (10³ psi) |
| Unfilled matrix | 0 | 20.7–21.3 | 21.0 |
| Heat-cleaned (no treatment) | 10 | 16.7–20.1 | 17.8 |
| | 20 | 17.3–19.6 | 18.5 |
| | 30 | 16.8–21.2 | 19.0 |
| Z-6020 | 10 | 18.5–20.3 | 19.3 |
| | 20 | 22.9–24.4 | 23.4 |
| | 30 | 22.5–24.5 | 23.4 |
| Z-6040 | 10 | 18.8–23.8 | 22.1 |
| | 20 | 21.5–23.0 | 21.9 |
| | 30 | 20.0–22.2 | 21.2 |
| Z-6076 | 10 | 17.6–18.8 | 18.2 |
| | 20 | 15.4–19.5 | 16.9 |
| | 30 | 15.0–22.2 | 18.4 |
| Mould release | 10 | 18.1–20 1 | 18.7 |
| | 20 | 16.0–17.4 | 16.6 |
| | 30 | 16.2–18.0 | 17.1 |
| As-received (Binder 701) | 10 | 19.9–20.6 | 20.4 |
| | 20 | 16.5–19.9 | 17.7 |
| | 30 | 20.0–20.5 | 20.4 |
| | 40 | 23.0–24.6 | 23.9 |

composites, particularly at high fibre volume fractions. At 20% volume fraction, the strength of the mould release treated fibre composites is about 25% lower than that of the Z-6020 treated composite. Since the fibres are three-dimensionally oriented, any dramatic improvement of strength cannot be expected.

The variation of the flexural modulus is shown in Fig. 5. The modulus for all composite systems increases monotonically with increase in fibre volume fraction. There is not much appreciable difference in modulus values for different surface treatments. Since the modulus is measured at low strains, the influence of the interface is not manifested at these strains. The modulus values were compared with two theoretical solutions (Fig. 5). The experimental values follow the theoretical solution for the effective modulus of three-dimensionally oriented fibre composites presented by Christensen and Waals [3] and Halpin-Tsai. [4] up to the volume fractions investigated.

The ultimate flexural strains are shown in Fig. 6. For all composite systems, the ultimate strain is less than the ultimate strain of the unfilled epoxy. The strain value decreases very rapidly as

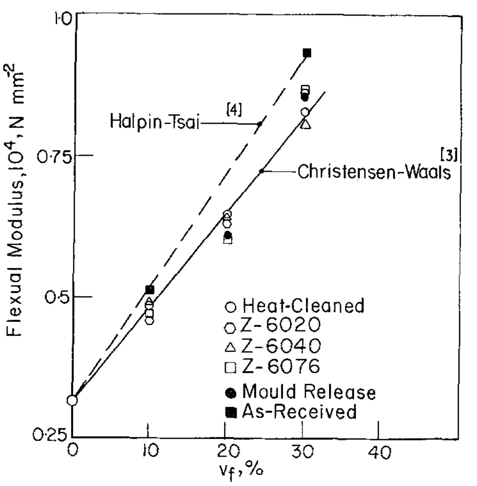

*Figure 5* Flexural moduli of glass fibre composites.

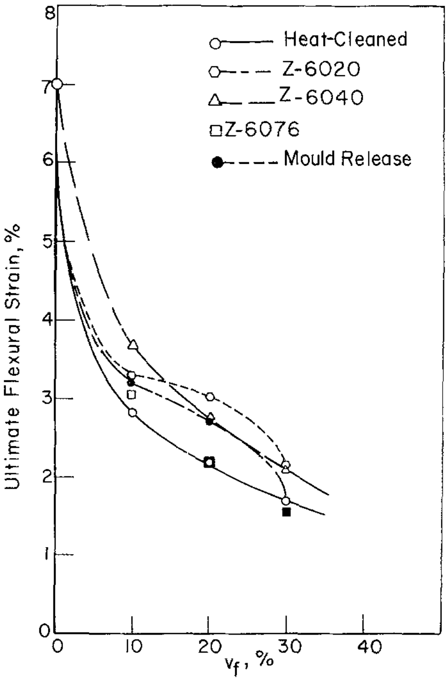

*Figure 6* Ultimate flexural strains of glass fibre composites.

the fibre volume fraction is increased from 0 to 10%, the rate of decrease being smaller at higher fibre volume fraction. The untreated heat-cleaned fibre composite has the lowest ultimate strain at all volume fractions.

The area under the stress-strain ($\sigma$-$\epsilon$) curve is a conventional measure of specific toughness of the material. For a brittle material, this represents the elastic strain energy stored in the material at fracture. For the composite systems studied, the area decreases at first very rapidly and then at a much slower rate (Fig. 7). The area under the $\sigma$-$\epsilon$ curve shows a trend similar to the ultimate flexural strain variation for different surface treatments (Fig. 6).

## 4. Microscopic studies

Figs. 8 to 11 show four photomicrographs of the fracture surfaces observed under a scanning electron microscope. Figs. 8 and 9 show the extremely good bond that can be obtained by using Z-6020 and Z-6040 surface treatments. The fibre-matrix bonds are still intact even after severe and complex loading conditions experienced during crack propagation. Figs. 10 and 11, on the other hand, show the poor bonding

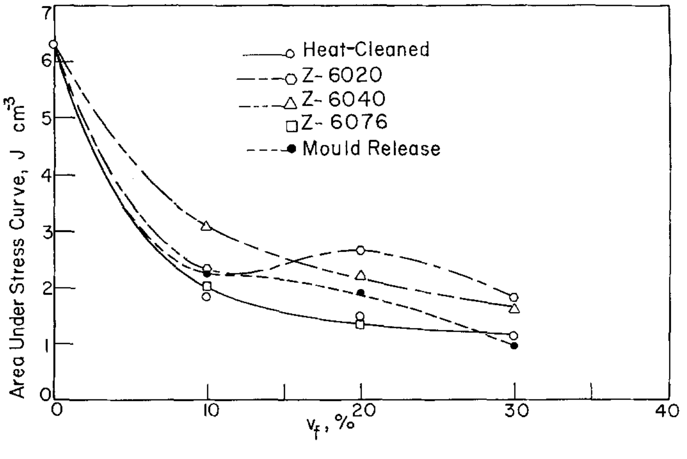

*Figure 7* Area under stress-strain curves for glass fibre composites.

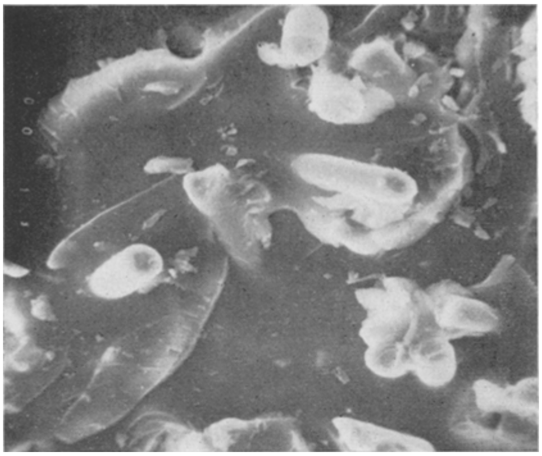

*Figure 8* A close-up view of the fracture surface of a 20% Z-6020 treated glass fibre composite showing good bond between the fibre and the matrix (× 400).

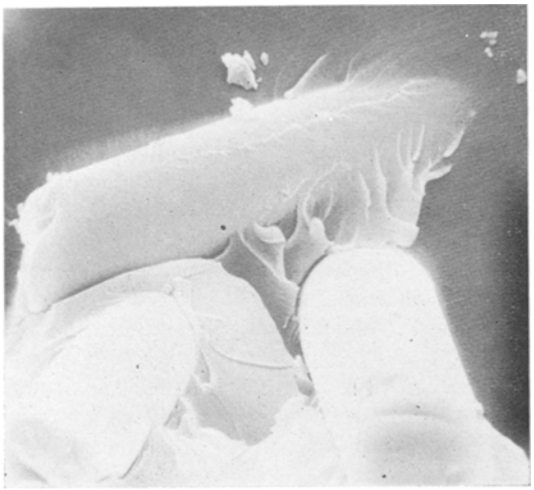

*Figure 9* A close-up view of the fracture surface of a 10% Z-6040 treated glass fibre composite showing good bond between the fibre and the matrix (× 1000).

obtained by using the mould release treatment and Z-6076, respectively. The bond, in these cases, has either failed during loading or there was no bond present before loading. Additional evidence of the good bond showing in Figs. 8 and 9 is the presence of a thin layer of matrix material covering the pulled-out fibres which cannot be seen on the fibres in Figs. 10 and 11. A more detailed description of the microscopic observations are given in [5].

## 5. Discussion of the strength results

The strength values of the glass fibre composites can be summarized as follows:

1. Of all the surface treatments, the mould release treatment gives the lowest strength at all the volume fractions studied.
2. There is a minimum fibre volume fraction below which the composite strength decreases with increasing volume fraction and above which the strength increases with increasing volume fraction.
3. There is a critical volume fraction below which the composite strength is lower than the unfilled matrix strength. Reinforcement is

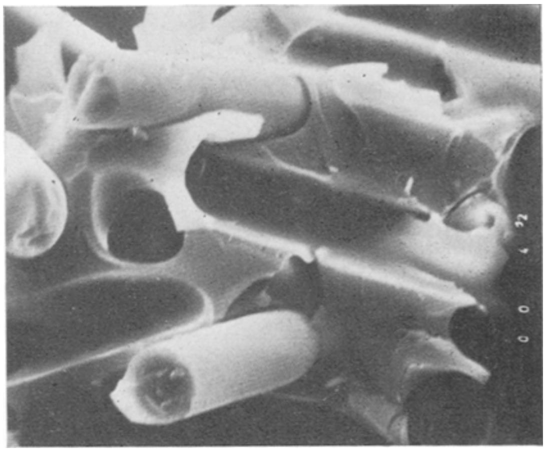

*Figure 10* A close-up view of the fracture surface of a 30% mould release agent treated glass fibre composite showing debonded fibres and unconstrained matrix (× 800).

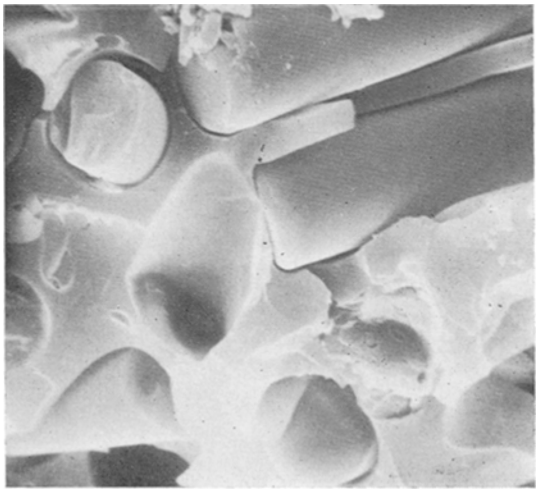

*Figure 11* A close-up view of the fracture surface of a 30%, Z-6076 treated glass fibre composite showing debonded fibres and unconstrained matrix (× 1600).

obtained only if the volume fraction of the fibres exceeds the critical volume fraction.

4. Not only does the strength depend strongly on the surface treatment of the fibres, the minimum and critical volume fractions also are dependent on the strength of the bond achieved by various surface treatments.

For aligned discontinuous fibre composites, many theoretical solutions exist to predict the composite strength [6, 9]. These solutions will predict higher strength values than could be expected from three dimensionally oriented random fibre composites like the ones used here. However, these theories can be used to analyse the present observations in a qualitative manner.

From the scanning electron micrographs, it is clear that the Z-6020 and Z-6040 treatments provide strong bonds between the fibres and the matrix, whereas the other treatments provide only weak bonds. The surface treatments of the fibres influence the interfacial adhesion which, in turn, influences the critical fibre length ($l_c$). The critical fibre length ($l_c$) is given by

$$l_c = \frac{\sigma_f d}{2\tau_i} \qquad (1)$$

where $\sigma_f$ = fibre strength, $d$ = fibre diameter, $\tau_i$ = interfacial shear strength. According to Equation 1, a strong bond gives a smaller $l_c$ value than a weaker bond. This suggests that, in the cases of Z-6020 and Z-6040 treatments, more and more fibres have lengths greater than or equal to $l_c$ and in the cases of mould release and other surface conditions, a large proportion of fibres have lengths less than $l_c$. A qualitative discussion of the fracture energy results [7] also confirms this postulation.

For aligned discontinuous fibres with $l > l_c$, Kelly and Tyson [8] predicted strengthening of a matrix at fibre volume fractions $v_f > v_{\text{critical}}$. Kelly [9] has shown that for $v_f < v_{\min}$, the composite strength decreases with increasing values of $v_f$. The variation of composite strength, $\sigma_c$, with fibre volume fraction, $v_f$, in a composite containing discontinuous fibres of length $l \geqslant l_c$ is shown schematically in Fig. 12. The expressions for $v_{\text{critical}}$ and $v_{\min}$ are given in [9]. These volume fractions are indirectly controlled by the interfacial shear strength, $\tau_i$. When $l = l_c$, the composite strength is given by

$$\sigma_c = \tfrac{1}{2}\sigma_f v_f + \sigma_m(1 - v_f)\,. \qquad (2)$$

In this case, the reinforcement of the matrix occurs at all volume fractions provided $\sigma_f > 2\sigma_m$ – which is the case for the glass fibre-epoxy system.

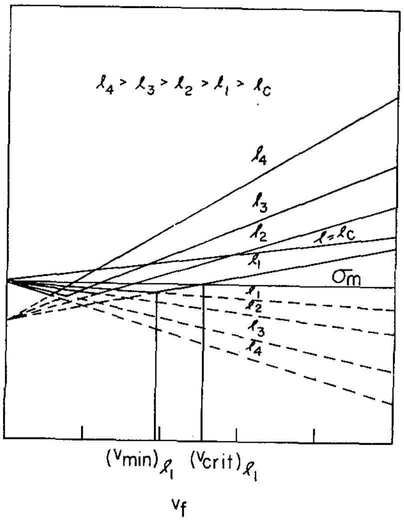

*Figure 12* Theoretical variation of composite strength $\sigma_c$, with volume fraction, $v_f$, for reinforcement with discontinuous brittle fibres, $l \geqslant l_c$.

In the present study, the strength values for Z-6020 and Z-6040 treated fibres appear to follow a trend shown similar to that shown in Fig. 12. In the case of Z-6040 treatment, no minimum value was observed and reinforcement, however small, occurred at all volume fractions considered. This indicates that the fibre lengths may be closer to the $l_c$ value for this particular treatment. In the case of the Z-6020 treatment, there is is a minimum volume at about 10% after which the strength of the composite increases and at about 15% volume fraction, the composite strength exceeds the matrix strength. Lee [10] reported the tensile and flexural strength values for random chopped strand reinforced thermoplastics. The strength results, in this case, also appeared to follow the trend predicted by the Kelly and Tyson equation.

As mentioned earlier, for mould release, Z-6076 and untreated fibres, the interfacial bond is relatively weak, and, therefore, a large proportion of fibres have lengths $l < l_c$ due to the high values of $l_c$ for these treatments. For discontinuous fibres with $l < l_c$, no critical or minimum volume fractions have been predicted, since no interface failure was considered. In the present study, however, there exists a minimum volume for fibres with $l < l_c$. It is well known that for $l < l_c$, the ultimate strength of the fibre is not achieved. But provided that $v_f > v_{\min}$, the fibres will carry the major portion of the load through shear stress transfer across the interface. If the interfacial bond strength is not strong, the failure of the composite will occur as soon as the failure occurs at or near the interface. When this happens, the matrix will not be able to carry any load. At $v_f < v_{min}$, on the other hand, there is an insufficient number of fibres to effectively restrain the elongation of the matrix. Thus, the fibre-matrix bond is easily broken, particularly if the bond is weak. The interface may not be broken completely; the interface shear strength only reduces to a value $\tau_i'$, where $0 \leqslant \tau_i' < \tau_i$. After the interface debonding, the major portion of the load is carried by the matrix and the ultimate composite failure occurs only after matrix failure. The composite strengths, in these cases, are given by

$$\sigma_c = \frac{\tau_i l}{d} v_f + \sigma_m'(1 - v_f), \quad \text{for } v_f > v_{min} \qquad (3)$$

$$\sigma_c = \frac{\tau_i' l'}{d} v_f + \sigma_m(1 - v_f), \quad \text{for } v_f < v_{min} \qquad (4)$$

where, $\sigma_m$ = matrix ultimate strength; $\tau_i$ = interface shear strength; $\sigma_m'$ = stress in the matrix at composite ultimate strain; $\tau_i'$ = interfacial shear strength after partial debonding, $0 \leqslant \tau_i' < \tau_i$; $l'$ = fibre length still bonded to the matrix after partial debonding.

Combining Equations 3 and 4, $v_{min}$ can easily be calculated. The value of $v_{critical}$ can also be determined by equating Equation 3 to $\sigma_m$. When this is done, it can be seen that both $v_{critical}$ and $v_{min}$, in this case, are directly related to interfacial shear strength, $\tau_i$. For $l < l_c$ and a very strong interfacial bond, the composite failure occurs only after the matrix fails. The composite strength is then given by

$$\sigma_c = \frac{\tau_i l}{d} v_f + \sigma_m(1 - v_f) \qquad (5)$$

at all volume fractions. In this case, no minimum volume can be observed and matrix strengthening will occur provided $(\tau_i l/d) > \sigma_m$.

It should be noted that the qualitative discussion presented here is based on the theoretical solutions for aligned fibre composites. So these solutions will predict higher strength values than those observed for a three-dimensional randomly oriented fibre composite. In fact, Cox [11] predicted that the strength of an aligned fibrous composite is reduced to one third of its value by randomizing in two dimensions and to one sixth of its value in three dimensions. A three-dimensional orientation of the fibres produces an isotropic composite, but no real enhancement of strength can be achieved.

## 6. Conclusion

The strength of a discontinuous fibre reinforced composite depends on the relative values of the fibre aspect ratio and the critical fibre aspect ratio. For a given fibre, the critical aspect ratio can be manipulated by simply varying the fibre matrix bond strength and consequently a wide variety of strength values can be obtained depending on whether the given fibre aspect ratio is higher or smaller than the derived critical aspect ratio. For effective reinforcement to occur, a strong interfacial bond as well as a critical fibre volume fraction are necessary.

For the three-dimensional low aspect ratio fibres considered here, no real enhancement in strength was observed. However, the strength appears to follow the trend predicted for aligned composites. Both minimum and critical volume fractions were observed. These volume fractions are either directly or indirectly controlled by the interfacial bond strength. The elastic modulus of the composite systems with small aspect ratio fibres increases with fibre volume fraction and is not influenced by the interfacial conditions.

Finally, it should be pointed out that in view of the recent worldwide plastic shortage due to the energy crisis, the type of short fibres considered here could find greater use as filler/extenders in resins so as to reduce the resin requirement and to make use of their reinforcing characteristics.

## Acknowledgement

The authors gratefully acknowledge the financial support provided by the US Atomic Energy Commission for the pursuit of this investigation under Contract No. AT (11-1) 1794.

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Received 31 December 1973 and accepted 26 March 1974.