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Brittle matrices reinforced with polyalkene films of varying elastic moduli

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The inclusion of polyalkene films of different moduli in a cement-based matrix has shown the benefits to be gained, in terms of increased stress at a given strain, from the use of films of high elastic modulus. Further, the concept of load-bearing cracks is used to explain the transition region between the limit of proportionality and the bend-over point on the tensile stress–strain curve, which is found to exist with high film modulus composites. This transition region could be an important factor affecting the choice of film to be used in a commercial composite.

Nomenclature

E_c	uncracked composite modulus
E_m	matrix modulus
E_f	film modulus
V_m	matrix volume-fraction
V_f	film volume-fraction
$V_{f(crit)}$	$(E_c\epsilon_{mu})/\sigma_{fu}$
A_c	cross-sectional area of composite
α	$(E_m V_m/E_f V_f)$
ϵ_m	matrix strain
ϵ_{mu}	matrix cracking strain
$\bar{\epsilon}_{mu}$	average matrix cracking strain, $(\bar{\sigma}_{co})/E_c$
ϵ_{mc}	strain at end of multiple cracking
σ_{fu}	ultimate fibre stress
σ_{cu}	ultimate composite stress
$\bar{\sigma}_{co}$	average composite cracking stress (assumed at a strain of $\epsilon_{mc}/2$)
S4	8:1 draw ratio polypropylene film
S8	18:1 draw ratio polypropylene film
E3H	polyethylene film
LOP	limit of proportionality (stress at first crack, assumed to be a departure from linearity of the tensile stress–strain curve of a perfectly straight and uniform test specimen. However, this point cannot be reliably determined from the stress–strain curve because of the clamping strains induced in warped specimens)
BOP	bend-over point (stress at which the approximately horizontal portion of multiple cracking region commences. The BOP is generally higher than the LOP and is a much more reliable point to determine experimentally than the LOP)

1. Introduction

The concept of reinforcing brittle matrices with continuous, fibrillated polyalkene films has been described elsewhere [1, 2]. It has been shown that the composite exhibits fine multiple cracking and is capable of complying with the British Standard [3] for asbestos cement sheeting.

Polyalkenes are distinct from the more common fibres used to reinforce cement (asbestos, glass and steel) in that their tensile behaviour is non-linear and that their modulus is generally less than that of the cement matrix. However, the production of ultra-high modulus polyalkenes, particularly polyethylene with initial moduli higher than that of the matrix has been the subject of much current research.

The work has been concentrated within three fields, solid-state deformation [4–8], solution

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[9, 10] and melt [11] processes. Of particular interest is the highly drawn polyethlene [4] since it is currently undergoing commercial evaluation by the Metal Box Company [12] and is available in a form suitable for the reinforcement of cement pastes. Also of interest is the seed-grown 100 GPa polyethylene [9] since its tensile stress–strain curve is approximately linear to failure.

The solid-state and solution-process derived materials are highly prone to fibrillation, a desirable property for cement reinforcement. However, the melt process material has been designed to avoid fibrillation and hence may not be suitable in its present form, for use as a reinforcement for brittle matrices.

This paper reports a comparison of two polypropylene films (8:1 and 18:1 draw ratios) supplied by H. and A. Scott Ltd, and the experimental polyethylene film of the Metal Box Company Ltd.

2. Theory

2.1. Tension

Aveston *et al.* [13–15] have given a clear description of the principles involved in calculating the tensile stress–strain curve of a fibre-reinforced brittle matrix, where it is assumed that the fibre is continuous, aligned and is linear elastic, that the fibre–matrix bond is linear and that the matrix exhibits a single-valued failure stress.

The tensile curve can be divided into three zones (Fig. 1). In Zone 1 the matrix is dominant until matrix cracking occurs and the composite properties can be calculated by the Law of Mixtures. Providing that the fibre concentration exceeds the critical fibre volume-fraction the composite will break down into blocks of length between x' and $2x'$ (where x' is the transfer length required to transfer the extra load carried by the fibre at a crack back into the matrix.) This constitutes Zone 2. The strain at the end of multiple cracking, ϵ_{mc}, can be predicted by Equation 1 where it is assumed that the average crack spacing is $1.364\,x'$

$$\epsilon_{mc} = (1 + 0.659\,\alpha)\,\epsilon_{mu}. \quad (1)$$

In Zone 3 the increase in strain is assumed to be due to the extension of the fibres alone and the tangent modulus becomes, $E_f V_f$. In this condition the ultimate strength is given by

$$\sigma_{cu} = \sigma_{fu} V_f. \quad (2)$$

2.2. Flexure

As discussed previously [1], the usual method of calculating moduli of rupture from flexural tests is unjustifiable theoretically as it assumes a linear stress distribution with the neutral axis at half of the beam depth. However, this approach will be used here in order to provide a comparison with other fibre cements, the calculated stress being a "notional stress".

3. Experimental details

3.1. Specimen manufacture

The specimens were manufactured using a superplasticized high-strength cement-based matrix, applied to layers of aligned film by a hand lay-up technique until the desired film volume-fraction and specimen thickness were attained. Curing was for 28 days under water at 20° C prior to cutting to the required test dimensions.

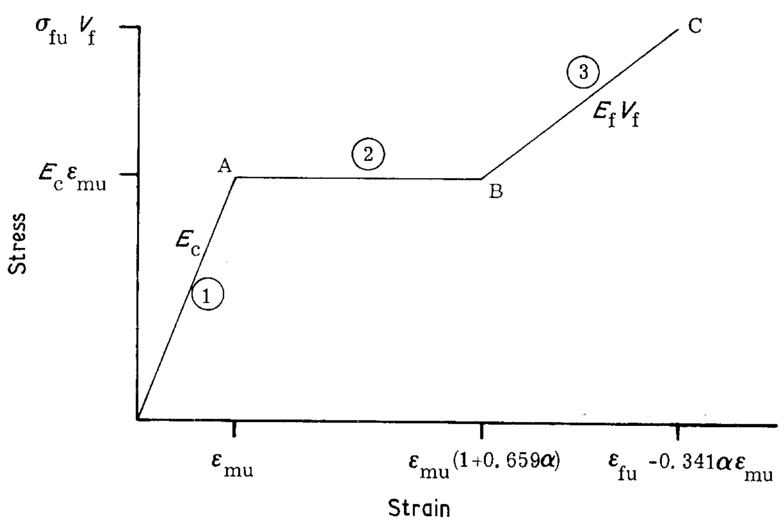

Figure 1 Theoretical tensile stress–strain curve.

3.2. Film tests

There is no British Standard for establishing the elastic modulus of polyalkene films. However, an ISO Recommendation [16] has been published and this has been used as the basis for the technique adopted in this work.

Ideally, 600 mm lengths of film (of width 3 mm) were prepared from an unfibrillated sample of the subsequently fibrillated film. Masking tape was applied to the ends of the strips in order to minimize film damage within the grips. An Instron 1122 machine was used to apply a strain-rate of approximately 5% min^{-1} and the complete load–cross-head displacement was recorded graphically. The cross-sectional area of the film, A, was established by weighing the test piece and assuming a specific gravity of 0.91 for polypropylene and 0.96 for polyethylene,

$$A = \frac{M}{\rho l}, \tag{3}$$

where M is the mass of film of length l and ρ is the density of film.

However, only the E3H polyethylene was supplied in the unfibrillated form. Hence, modulus determination of S4 and S8 polypropylene films was approximated using lengths of the fibrillated films carefully folded to a width of 25 mm. The limitations of the preparation are realized and the discrepancies involved are exhibited by the comparison of the E3H films shown in Fig. 2.

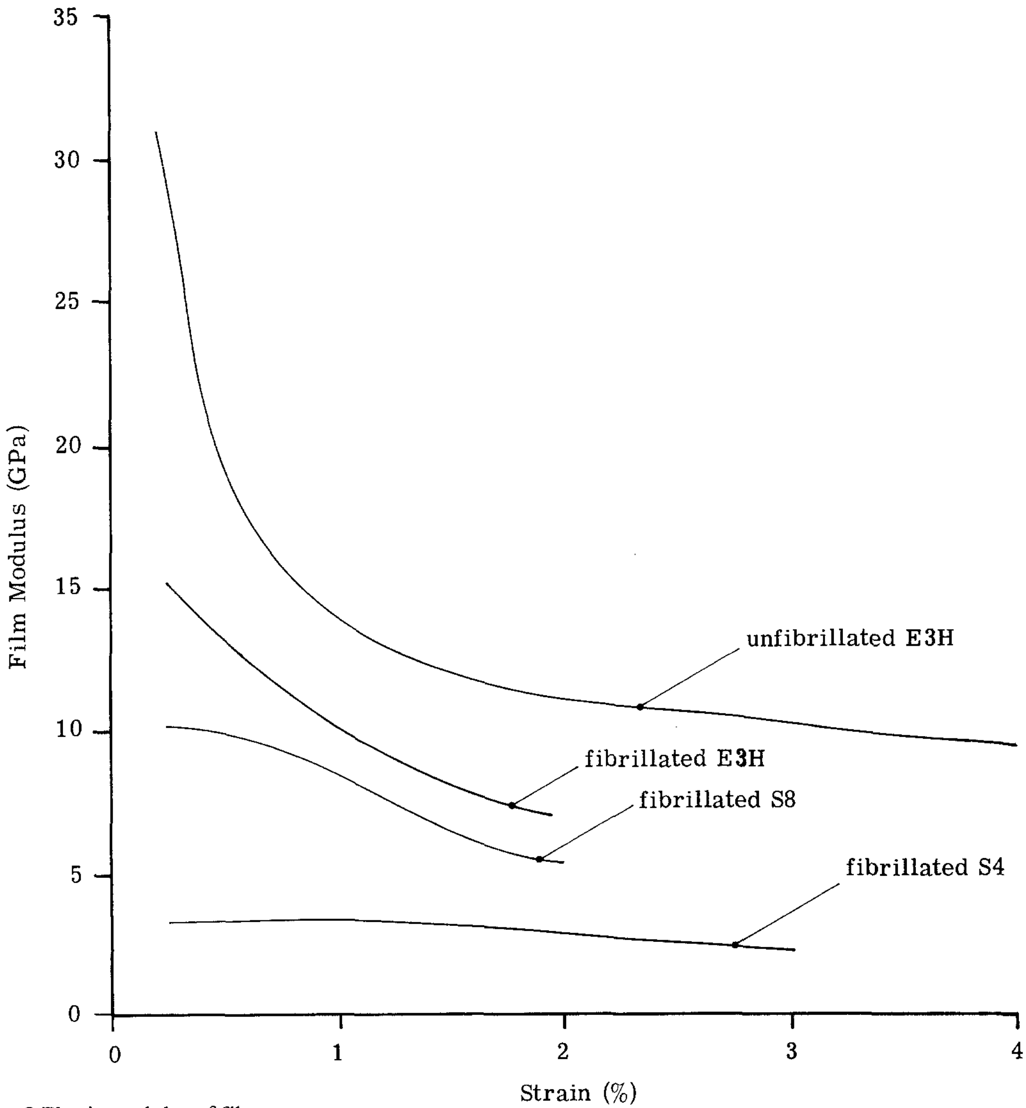

Figure 2 Elastic modulus of films

3.3. Composite tests

3.3.1. Tension

Strips, of dimensions 300 mm × 25 mm × 6 mm nominal thickness, were tested in an Instron 1122 machine at 10 mm min^{-1} cross-head speed. Lead sheet was placed between the specimen and each face of the grips in order to reduce localized stress concentrations. Strain monitoring was undertaken via a clip-on extensometer [17] equipped with two a.c. linear variable differential transformers and the signals were recorded simultaneously on two X–Y–Y recorders calibrated for strain ranges 0 to 0.5% and 0 to 5%, respectively. An increased strain range of 0 to 10% was used for the S4 composite. The strains on each face of the specimen were monitored individually face of the specimen were monitored individually and only averaged electronically for display on the coarse scale X–Y–Y recorder.

3.3.2. Flexure

Coupons, of dimensions 150 mm × 50 mm × 6 mm nominal thickness, were tested in third-point loading over a span of 135 mm [18] and at a cross-head speed of 10 mm min^{-1}. A load–cross-head displacement record was obtained.

4. Results

4.1. Film

The results of the measurements of film moduli are presented in Fig. 2, in the form of a tangent modulus determined at specified strains. It should be noted that the results for the fibrillated films are only reported up to 2 to 3% strain since progressive failure commenced at this stage.

Unfilbrillated E3H exhibited the highest initial modulus of 31.5 GPa. The form of the modulus of the fibrillated E3H is similar but possessed a lower initial value of 15.4 GPa. Obviously, the poor test technique employed for the latter film will account to some extent for the lower modulus. Also, in determining the cross-sectional area of a fibrillated film by gravimetric methods, an over estimate of the effective area is obtained since any discontinuous fibrils attached to the primary network are included but remain non-load carrying. Further, some of the load carrying elements are angled with respect to the molecular orientation and would be expected to yield lower modulus values. Hence, modulus values derived from fibrillated film are regarded as a lower bound.

Polypropylene films S4 and S8 exhibited initial moduli of 2.5 GPa and 10.2 GPa, respectively.

An important feature of Fig. 2 is the reduction of the modulus as the strain increases. Hence, when incorporated within a cement matrix the film will not exhibit a unique value of modulus. Indeed, at any specified composite strain the film will have a range of moduli depending upon the film–matrix stress transfer function.

4.2. Composite

The properties of the composites are dependent on the included volume of film and therefore a precise analysis of the test results requires a knowledge of the film volume in each test specimen. This was achieved for each specimen by acid dissolution and the variation of film volume across a sheet was thus obtained. In order to provide a direct comparison of the tests for different film types in the same matrix it was necessary to compare them at the same film volume-fraction, V_f, and a V_f-value of 10% has been chosen for this purpose. Fig. 3 shows tensile curves for both polypropylene composites containing 10% film by volume, each obtained from 20 individual specimens with film volumes between approximately 8.5% and 11.5%. Fig. 4 shows two types of tensile ene composite. Flexural curves are shown in Fig. 5 for composites containing 10% film by volume, each obtained from about 9 individual specimens with film volumes between approximately 8% and 12%.

5. Discussion

5.1. Composite (tension)

It can be seen from Fig. 3 that the inclusion of films of varying moduli to reinforce identical matrices has affected the performance of the polypropylene film composites in all three zones described in Section 2.1. Also, Fig. 4 shows that the polyethylene composite has a higher BOP and lower strain at the end of multiple cracking than the polypropylene composites in Fig. 3.

5.1.1. Initial Cracking

Since the LOP determination is seriously affected by the measuring and plotting technique and the planarity of the specimen it is not justifiable to allot specific stresses to the LOP. However, recent use of acoustic emission techniques has enabled the "first crack" to be detected and has shown that the LOP is lower than the BOP and is approxi-

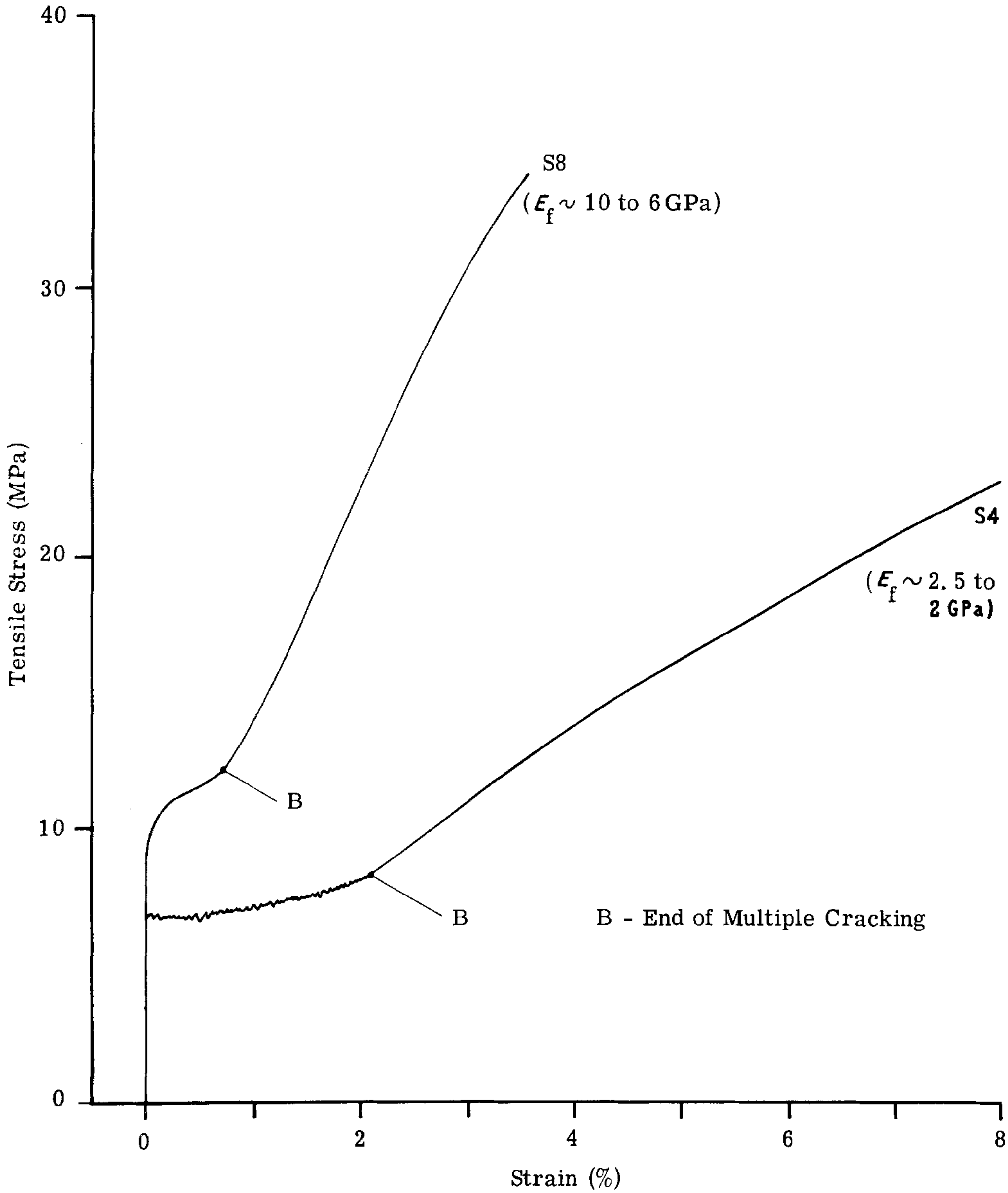

Figure 3 Tensile stress–strain curves of two polypropylene film composites ($V_f = 10\%$)

mately constant for film volumes up to 10%. The LOP can therefore be defined as the first burst of acoustic noise from the specimen, but the matrix stress at this point is still somewhat speculative because of the stresses induced in the specimen by clamping in the jaws of the test machine.

It can clearly be seen in Figs 3 and 4 that an increase in E_f is accompanied by an increase in BOP, leading to a transition region between the approximately constant LOP and the increased BOP. It has also been noted that the BOP is increased for a given composite type as V_f is increased (Fig. 6) leading to an increase in the average matrix-cracking strain, $\bar{\epsilon}_{mu}$. This is a similar type of behaviour to that reported for glass-reinforced cement (GRC) and may well also occur in asbestos–cement.

Possible explanations of this behaviour may be sought in the failure mechanism of the matrix. Since cement matrices are known not to possess a single-valued failure stress, the initial crack may propagate across the complete cross-section of the composite or may possibly be arrested by the weak fibre–matrix interface [20]. It has been suggested by Higgins and Bailey [21, 22] that, due to particles interlocking across the newly-formed crack, a

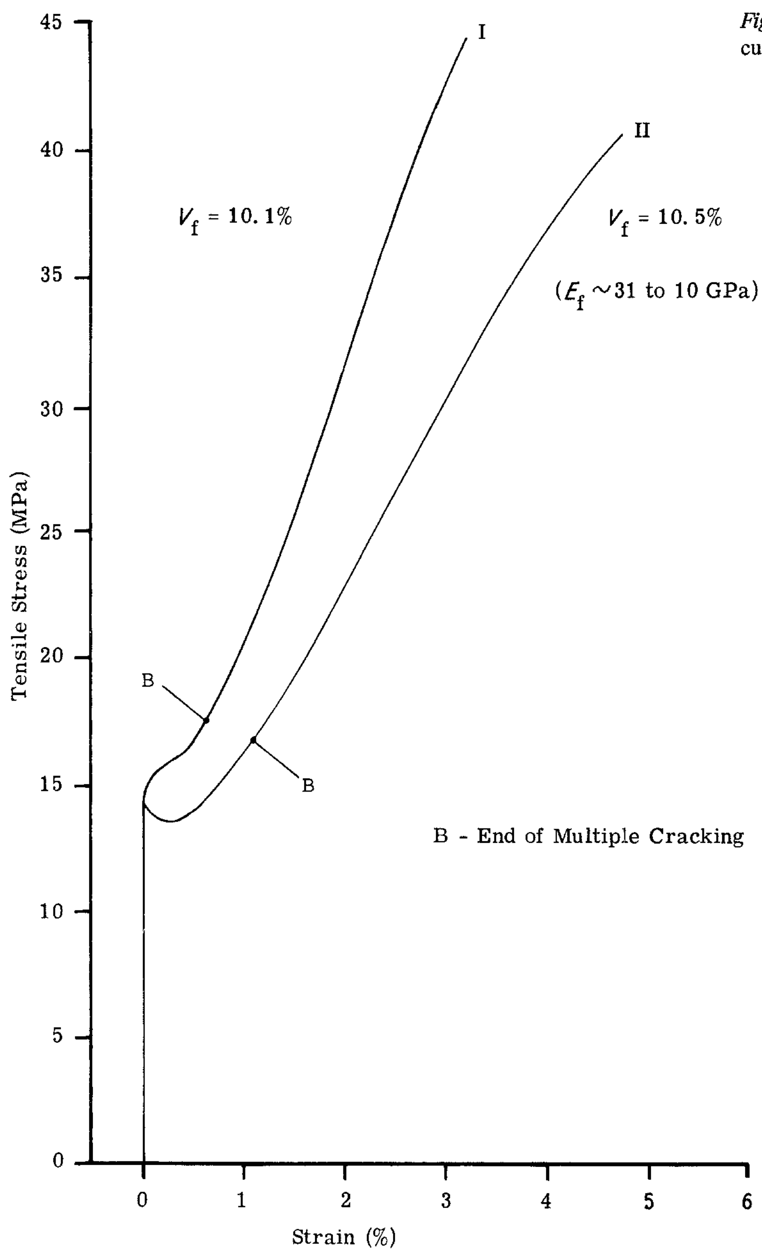

Figure 4 Typical tensile stress–strain curves of a polyethylene film composite.

tied crack exists up to a crack-width of approximately 2 μm. Hence, the load-carrying capacity of a cracked composite (taken as the load applied to the total cross-section) may be derived from four separate components, i.e., the matrix and the fibres in the uncracked section of composite, the tied crack and the fibres bridging the crack. The relationship between crack-width and the load carried by the crack is at present unknown and, hence, a valid quantitative description cannot be given for stable crack growth.

5.1.2. Multiple cracking region

Due to the fact that, for cement pastes, the matrix does not have a single-valued cracking stress and that the specimen cross-section is not precisely uniform, the stress at the end of multiple cracking (Point B in Fig. 1) is, in practice, generally higher than the BOP. The end of multiple cracking is sometimes difficult to establish precisely but, for these results, is taken to be the beginning of the mainly-linear portion of Zone 3 of the stress–strain curve (see Fig. 1). It was shown in Equation

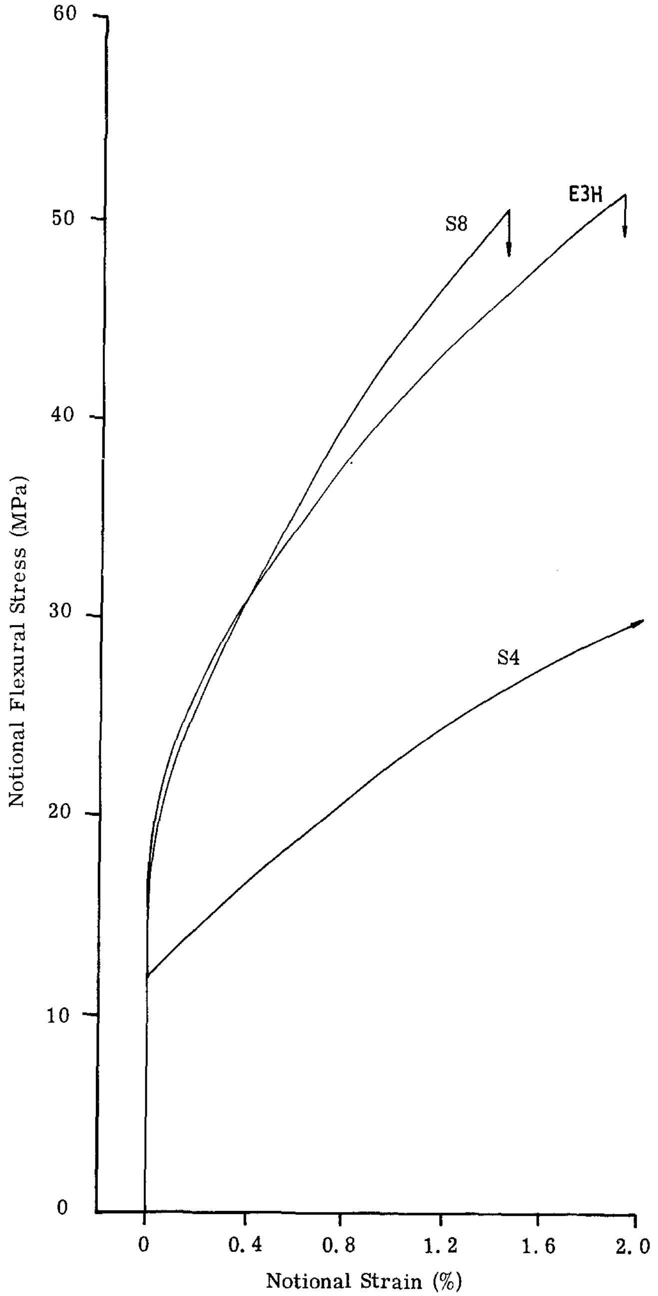

Figure 5 Notional flexural stress–notional strain curves for composites containing 10 vol% film. (Strain calculated assuming neutral axis at mid-depth of beam.)

1 that for a given film volume-fraction and matrix the strain at the end of multiple cracking, ϵ_{mc}, is theoretically inversely proportional to the film modulus, E_f. However, this is an over-simplification because α is itself continually varying as a function of the decreasing film modulus and, also, the changing average matrix cracking strain, $\bar{\epsilon}_{mu}$, apparently increases with the inclusion by the higher modulus films, as indicated by the higher stress level at which most of the multiple cracking occurred (assumed to be a strain of $\epsilon_{mc}/2$). Using the representative values of $\bar{\epsilon}_{mc}$ and α, values of ϵ_{mc} were calculated for S4, S8 and E3H composites as shown in Table I.

The polypropylene films S4 and S8 (see Fig. 3) and Curve I of the E3H polyethylene composites (see Fig. 4) give good agreement with the theoretical values for ϵ_{mc}. Fig. 4 will be discussed in more detail in Section 5.1.4.

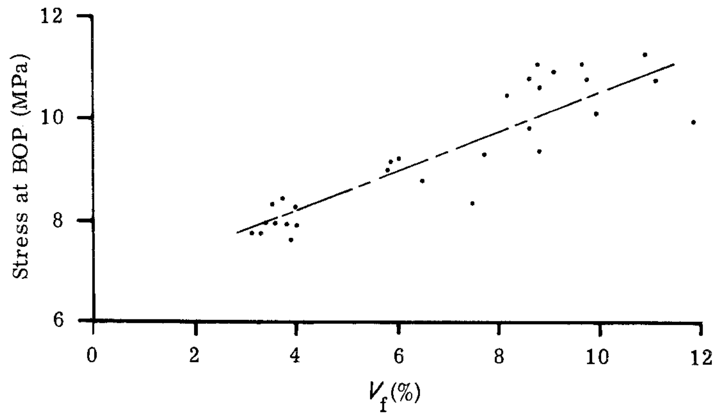

Figure 6 Experimental relationship between BOP in tension and V_f-film bar 112 (Polypropylene film, $E_f = 8.7$ GPa).

5.1.3. Post-crack region (polypropylene composites)

In order to determine the film modulus effective within the post-crack region (Zone 3) the slope of the curve is divided by the film volume-fraction. This yields values of 2.29 GPa and 8.36 GPa for the polypropylene films S4 and S8 respectively, approximately values which would be expected from Fig. 2 at these strains.

5.1.4. Polyethylene composites

Whilst the polypropylene composites exhibited a consistent behaviour, the polyethylene composites showed two distinct modes of behaviour for the same film and nominally similar film volume-fractions (Fig. 4). For stresses up to the BOP the performance was identical. However, once multiple cracking commenced, the stress–strain curve either increased (Curve I) or immediately decreased before commencing to rise at substantial strain (Curve II).

From Fig. 4 and Table I it can be seen that polyethylene composites showing the behaviour of Curve I completed multiple cracking at approximately the theoretical strain (0.75%). However, those composites showing the behaviour of Curve II consistently showed a higher value of ϵ_{mc}. This phenomenon was also exhibited by those composites showing an initially constant multiple cracking stress.

TABLE I Composite parameters used to calculate ϵ_{mc} ($V_f = 10\%$)

Film	E_m (GPa)	E_f (GPa)	α	$\bar{\epsilon}_{mu}$ (%)	ϵ_{mc} (%)
S4	32	2.5	115	0.0260	2.00
S8	32	9	32	0.0368	0.81
E3H	32	11	26	0.0415	0.75

A surprising feature of the post-crack slopes (Zone 3) of the polyethylene composites is their linearity approximately to failure, rather than showing a gradually decreasing tangent modulus as expected from Fig. 2. Indeed, polyethylene composites showing the Curve II-type behaviour show an approximate 10% increase in tangent modulus, $E_f V_f$, commencing at approximately 1.9% strain. This behaviour was not noted during tests on the film alone.

It can be seen from Fig. 4 that, although of similar film volume-fraction, the effective film modulus derived from Zone 3 (see Section 5.1.3.) is significantly different for the two curves. The effective film moduli are 9.67 GPa and 6.08 GPa for the Curve I and Curve II behaviour composites, respectively.

The lower effective modulus is compatible with a composite exhibiting a high ϵ_{mc} since it can be seen in Fig. 2 that film modulus decreases with increasing strain. However, the reason for the variability of ϵ_{mc} is not fully understood. Future research is aimed at expanding the spread of data (V_f ~4% to 12%) so that a fuller description of both the post-crack slope and the strain at the end of multiple cracking may be achieved.

5.2. Composite (flexure)

In considering a composite for commercial exploitation, the load-carrying capacity at low strains combined with fine crack spacing are important criteria for acceptance. It is shown in Fig.5 that the main advantage of the polyethylene lies in the high BOP of approximately 20 MPa. This value exceeds the minimum strength of 15.7 MPa specified in the asbestos–cement codes [3] and the composite would therefore appear to be uncracked at this stress level. After the BOP the extra load and strain capacity is available to absorb transient overloads without major distress to the products.

In Fig. 5 the maximum load on S8 and E3H composites occurred at an estimated tensile strain of approximately 1.5% and "failure" was due to a compression failure of the matrix above the neutral axis. Due to lower modulus of film S4, compression failure was not observable before the deflection capacity of the flexural rig was attained.

5.3. Bond

The subject of stress transfer between cement pastes and fibrillated polyalkene films will be discussed in detail in a future publication. However, bond strengths were adequate such that average crack spacings of 1.0, 1.2 and 0.6 mm were achieved for S4, S8 and E3H composites, respectively.

6. Conclusions

It has been shown that, in general, polyalkene-reinforced cement behaves in accordance with theory. However, as film of increasing modulus is incorporated, a transition region is introduced between the limit of proportionality and the bend-over point. It is suggested that this may be a result of microcracks either being constrained such that their width is less than 2 μm, and hence maintaining their ability to sustain loads, or that cracks are arrested by the weak fibre–matrix interface normal to the crack.

Nominally identical polyethylene composites exhibited surprisingly different modes of behaviour in the multiple-cracking region. A continually rising multiple cracking at constant stress or at a reducing end of multiple cracking and a higher post-crack slope, $E_f V_f$, than those samples which showed mulitple cracking at constant stress or at a reducing stress. The reason for this difference in behaviour is currently unexplained but the higher post-crack slope is thought to be related to the higher film modulus encountered at lower strains. Hence, for a composite containing film with a variable modulus, the post-crack slope is dependent upon the strain at the end of multiple cracking rather than simply dependent upon the film volume-fraction.

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