

Hybrid effects in the bending stiffness of graphite/glass-reinforced composites

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The flexural modulus of graphite/glass-reinforced hybrids exhibits deviations from the rule-of-mixtures base-line. The deviation increases as the segregation of the glass and graphite layers increases, and it reaches a maximum when the two fibre types are arranged in 3 layers. When the stiffer fibres (graphite) are in the outer layers the deviation is positive but it is negative with the less stiff fibres (glass) on the outside. The extent of deviation is shown to be predictable by the analysis.

1. Introduction

A recent presentation by Hashin [1] proposed a new viewpoint on composites, claiming that they are to be considered as structures and not as materials. The fundamental argument for this concept is that even in a "simple" composite the properties do not merely depend on those of the constituent materials, but they strongly relate to the internal geometry, i.e. the arrangement of fibres. It will be shown later that hybrid effects provide an excellent example of the validity of this viewpoint. To realize this, let us first recall the definitions of a number of relevant terms.

The general definition of hybrids presents them as composite structures containing at least two types of reinforcements. This commonly recognized definition obviously does not make any reference at all to the packing arrangement of the reinforcements in the matrix. Thus, it covers the complete range from an intimate fibre mixture to a segregated laminated structure typical of the lay-up and of the filament winding techniques.

A rule-of-mixtures expresses a composite property as a weighted average of the properties of its constituents. The weighting is proportional to the volume-fractions of the constituents also disregarding the internal geometry.

Combining these definitions it can be seen that if a rule-of-mixtures is valid in two-phase composites it will also be applicable to hybrids. Bearing this in mind, let us now recall the definition of a hybrid effect. This is expressed as a positive or a negative deviation from the rule-of-mixtures base-line [2].

A very important parameter which controls the existence of hybrid effects is the internal arrangement of the two types of reinforcements. This of course makes the hybrids comply to the concept of "structures" described above. As an example let us consider the dependence of fracture properties on the internal geometry. This topic was investigated in two recent studies, the first of which [2] showed that the deviations of the work-of-fracture values, for instance, from the rule-of-mixtures, is governed by the lay-up sequence of the graphite and of the glass fibres. When the fibres formed an intimate mixture, the glass fibres exhibited a smaller pull-out length compared with those of the graphite, resulting in a negative hybrid effect. However, when the two fibre types were contained in distinct and segregated layers, the rule-of-mixtures value was recorded. The second study [3] showed that the effect of the stacking sequence is in fact combined with that of the loading

TABLE I Sources and properties of the constituent materials

Source/Property	Graphite fibres	Glass fibres	Matrix
Description	Grafil HTS 1000 filaments (Courtaulds)	S-glass 904 20 ends (Owens-Corning)	Epoxy ERL2256 (Union Carbide) Hardener Toxon 60/40 (Uniroyal)
Density (g cm ⁻³)	1.76	2.49	1.17
Flexural modulus (GPa)	235.3	85.3	3.45

configuration, i.e., translaminar or interlaminar.

Along much the same lines the present study investigates the effect of the internal fibre arrangement on the flexural modulus of graphite/glass-reinforced hybrids. It is widely accepted [4-7] that the modulus of unidirectional hybrid composites follows the rule-of-mixtures behaviour although some reservations regarding the flexural modulus have been pointed out. Kalnin [8], for example, claimed that the flexural modulus depended strongly on the arrangement of the plies with respect to the plane of symmetry. Only when the plies were uniformly distributed was the rule-of-mixtures applicable to hybrids. He also stated that placing the stiffer plies in the outer portion of the laminate greatly increased the flexural modulus.

In view of this, the first part of our study [9] was devoted to an analysis of the elastic properties of laminated beams in bending. It showed that the occurrence of hybrid effects in the stiffness depended on the number of layers, on the volume-fractions of the materials, and on their modulus ratio. An intimate mixture of the reinforcements, however, yielded a rule-of-mixtures behaviour. The present study has been undertaken to provide an experimental verification of the analytical results. The work was carried out with rings cut of zero-angle filament wound glass/graphite-reinforced epoxy pressure vessels. While the various material and processing parameters remained unaltered during the preparation of the specimens, the stacking arrangement was changed through the winding sequence of the fibres.

TABLE II The lamination arrangement

Designation	Layer sequence*	Nominal thickness (mm)
3G	g/c/g	0.75/1.50/0.75
3C	c/g/c	0.75/1.50/0.75
5G	g/c/g/c/g	0.50/0.75/0.50/0.75/0.50
5C	c/g/c/g/c	0.50/0.75/0.50/0.75/0.50
7G	g/c/g/c/g/c/g	0.25/0.50/0.50/0.50/0.50/0.50/0.25

*g denotes glass-epoxy layer, c denotes graphite-epoxy layer.

2. Experimental procedure

2.1. Specimen preparation

Glass/graphite-reinforced epoxy hybrid tubes were manufactured by a filament winding technique, employing the materials whose properties are listed in Table I. A constant winding angle of 0° (perpendicular to the tube axis) was maintained, using, however, five different lamination sequences described in Table II and Fig. 1. Equal total volumes of glass-epoxy and of graphite-epoxy layers were retained throughout, although these layers were divided into different thicknesses arranged symmetrically with respect to the centre of the wall thickness. The volume-fractions of the two fibres and of the resin were determined by a resin etching and graphite burning-off procedure. The resulting volume-fractions were 35, 28 and 37% for the glass, the graphite and the epoxy, respectively, in each tube.

The resulting tubes had an internal diameter of

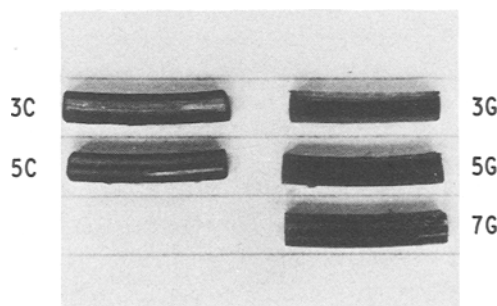


Figure 1 Sections of the ring specimens showing the lamination sequences.

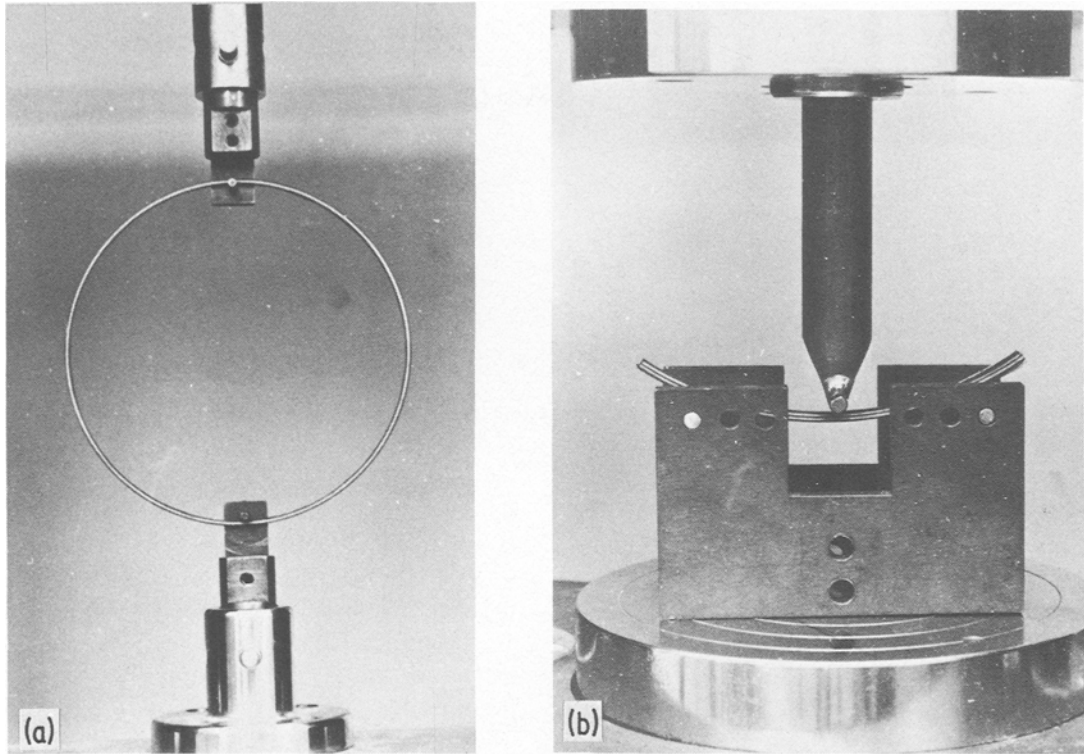


Figure 2 The testing set-ups: (a) ring testing, (b) curved beam testing.

195 mm and a nominal wall thickness of 3 mm, and they were over 1 m long. Two types of specimens were cut from the tubes. These were 3 mm wide rings and 120 mm (measured along the outer fibre) long curved beams.

2.2. Testing

The configuration of the ring testing and the device used are shown in Fig. 2a. The testing speed was 5 mm min^{-1} . The flexural Young's modulus for that loading configuration is given by [10],

$$E = k \frac{R^3}{I} \left(\frac{\pi}{4} - \frac{2}{\pi} \right), \quad (1)$$

where E is Young's modulus, k is the slope of the load–displacement curve, and R is the internal radius of the ring. I is the moment of inertia of the cross-section of the ring and is given for the rectangular cross-section by $I = bt^3/12$, where b and t are the ring width and thickness, respectively. Between 6 to 10 rings were tested for each hybrid material.

Curved beam testing was carried out in three-point bending as shown in Fig. 2b. The testing

speed was 0.5 mm min^{-1} , and the loading span was 80 mm. The flexural modulus was calculated using

$$E = \frac{k l^3}{4bt^3}, \quad (2)$$

where l is the loading span. This equation disregarded the shear effects which were considered negligible for the span-to-depth ratio employed [11]. Also, the effect of the beam curvature was ignored. Five specimens were tested for each hybrid material.

3. Results and discussion

Fig. 3a and b present the results of the ring and of the curved beam testing, respectively. They exhibit similar characteristics as follows:

(a) The moduli of the hybrids with the graphite fibres in the outside layers (3C and 5C) are always higher than those of the hybrids with glass fibres in the outside layers (3G, 5G and 7G).

(b) The deviation (negative or positive) from the mean value decreases as the total number of layers increases, indicating that the modulus approaches this mean value as the layer arrange-

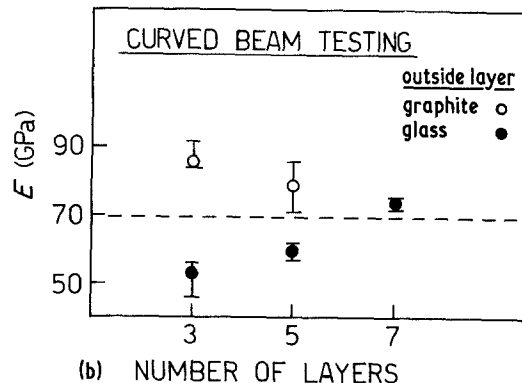
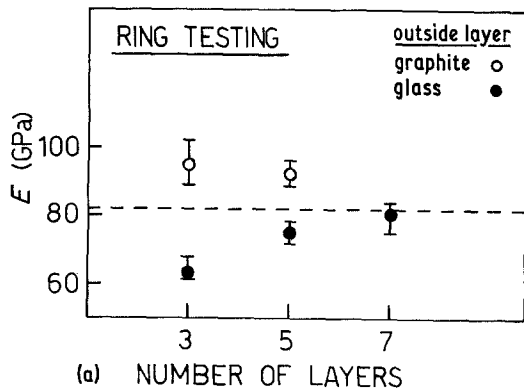


Figure 3 The experimental flexural moduli: (a) from ring testing. (b) from curved beam testing.

ment becomes less segregated and the fibres form a more intimate mixture.

(c) The moduli obtained by the ring testing are higher than those obtained by the curved beam testing.

The deviations exhibited by the above results from the rule-of-mixtures base line obviously correspond with the general anticipation that if the stiffer component is on the outside of a laminated beam then the stiffness will exceed rule-of-mixtures prediction, and vice versa. The important point, however, is to foresee how the *magnitude* of these deviations varies with the *arrangement* and the *number* of the layers. This point is answered by the analysis derived in our recent study [9]. It presented the flexural elastic properties of symmetrically laminated beams composed of m materials arranged in n layers with a regular stacking sequence.

The particular structure of two materials arranged in n layers was solved as an example, and the results were found to be in agreement with those appearing in the literature [10, 12], namely:

$$E = E_2(\alpha \bar{E} \phi_1^3 + \beta \bar{E} \phi_1^2 + \gamma \bar{E} \phi_1 + 1), \quad (3)$$

where $\bar{E} = (E_1 - E_2)/E_2$ and E_1 and E_2 are the moduli of the two types of layers so that the

TABLE III The values of the coefficients α , β , γ as a function of the total number of layers

n	α	β	γ
3, 7, 11, 15, ...	$\frac{8}{n^2 - 1}$	$\frac{4(n-3)}{n^2 - 1}$	$\frac{(n-3)(n-1)}{n^2 - 1}$
5, 9, 13, 17, ...	$\frac{8}{n^2 - 1}$	$\frac{-4(n+3)}{n^2 - 1}$	$\frac{(n+3)(n+1)}{n^2 - 1}$

subscript 1 always refers to the central layer, ϕ_1 is the volume-fraction in the hybrid of the layers containing Material 1 [13], and α , β and γ are numerical coefficients which depend on the total number of layers, n . Two special cases are distinguished which relate to the layer arrangements of the hybrid laminates tested in the present study. In the first case $n_1 = n_2 = (n-1)/4$, where $n = \{5, 9, 13, \dots\}$; and in the second case $n_2 = n_1 + 1 = (n+1)/4$, where $n = \{3, 7, 11, \dots\}$. For these special cases the values of α , β and γ are presented in Table III.

Taking the relevant values of the modulus of a glass-epoxy layer to be 60.75 GPa and that of a graphite-epoxy layer to be 133.30 GPa, and $\phi_1 = 0.50$, Equation 3 results in the calculated values presented in Fig. 4. It is seen that the trend of the experimental results is in complete agreement with the analytical prediction. It can also be shown [9] that when $n \rightarrow \infty$ then $\alpha \rightarrow 0$, $\beta \rightarrow 0$ and $\gamma \rightarrow 1$, and Equation 3 reduces to:

$$E = E_1 \phi_1 + E_2 (1 - \phi_1) \quad (4)$$

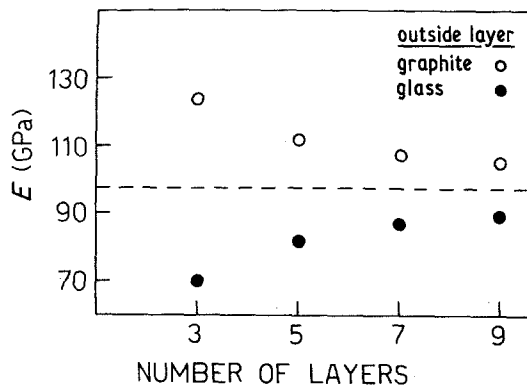


Figure 4 The calculated flexural moduli as a function of the number of layers.

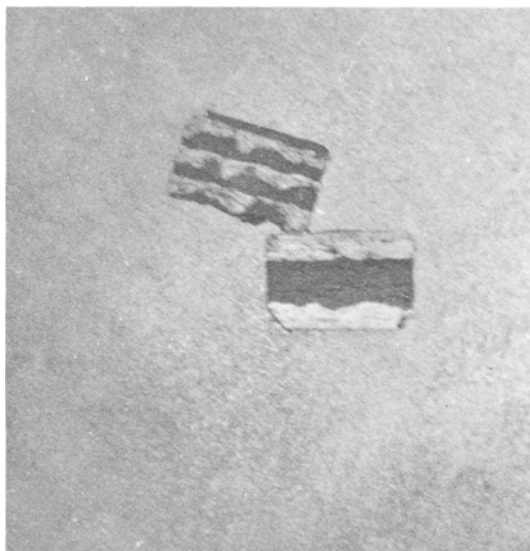


Figure 5 Polished cross-sections of 3C and 7G specimens, showing non-uniform layer segregation.

which is the rule-of-mixtures expression. The rule-of-mixtures value calculated with the above data is 97.02 GPa, also indicated in Fig. 4.

It is maintained that the correspondence of the trends is by itself a sufficient proof for the relevance and for the applicability of the analytical treatment. The differences between the various sets of results are attributed to experimental errors in the determination of the volume-fractions, and to other errors resulting from neglecting the shear contribution and the beam curvature in the three-point bending experiments. Another source of error is the fact that the segregation of the layers as obtained by the filament winding is not perfect (see Fig. 5), as assumed by the analytical treatment.

Further evidence for approaching a rule-of-mixtures value with increasing the number of the layers can be found in our previous work [2], where the three lay-up sequences investigated (designated 1/1, 2/2 and 5/5) exhibited the exact rule-of-mixtures value.

4. Conclusions

The flexural modulus of graphite/glass-reinforced composites, where the reinforcement is arranged in segregated layers, deviates from the rule-of-

mixtures base line. When the stiffer fibres are placed in the outer portion of the laminate the deviation is positive, and it is negative when the less stiff fibres are placed there. The deviation (positive or negative) increases as the number of layers decreases, and it reaches a maximum for the minimum number of layers of 3. As the number of layers increases, and the mixture becomes more intimate, the flexural modulus approaches the rule-of-mixture value.

Acknowledgement

The authors thank Mr J. Goobich for the volume-fraction determinations.

References

1. Z. HASHIN, USA-Italy Joint Symposium on Composite Materials: The Role of the Polymeric Matrix on their Processing and Structural Properties, Capri, June 1981, edited by J. C. Seferis and L. Nicolais (Plenum Press, New York), to be published.
2. G. MAROM, S. FISCHER, F. R. TULER and H. D. WAGNER, *J. Mater. Sci.* **13** (1978) 1419.
3. S. FISCHER, G. MAROM and F. R. TULER, *ibid.* **14** (1979) 500.
4. B. HARRIS and A. R. BUNSELL, *Composites* **6** (1975) 197.
5. A. R. BUNSELL and B. HARRIS, *ibid.* **5** (1974) 157.
6. L. N. PHILLIPS, Proceedings of the 10th International Reinforced Plastics Conference, Brighton, UK, 1976 (British Plastics Federation, 1976) Paper 21.
7. I. STEG and F. R. TULER, *Polim Vehomarin Plast.* **6** (2) (1976) 12 (in Hebrew).
8. I. L. KALNIN, ASTM STP 497 (1972) 551.
9. H. D. WAGNER, I. ROMAN and G. MAROM, *Fibre Sci Tech.*, to be published.
10. J. H. FAUPEL, "Engineering Design" (John Wiley and Sons, Inc., New York, 1964) p. 470.
11. S. FISCHER, I. ROMAN, H. HAREL, G. MAROM and H. D. WAGNER, *J. Testing Evaluation*, **9** (1981) 303.
12. N. J. HOFF, in "Engineering Laminates", edited by A. G. H. Dietz (John Wiley and Sons, Inc., New York, 1949) pp. 36.
13. H. D. WAGNER and G. MAROM, *Composites*, to be published.

Received 11 August

and accepted 30 September 1981