

## PREPARATION AND MECHANICAL PROPERTIES OF HYBRID PLASTIC COMPOSITES OF ARAMID AND CARBON FIBRES

S. A. DOGBE

*Chemistry Department, National Nuclear Research Institute, GAEC, P.O. Box 80, Legon, Ghana*

### Abstract

Hybrid plastic composites are considered materials of great potential for engineering applications. For the study of the potential of these materials, a simple method was developed to prepare unidirectional, intimate mixtures of different volume fractions of aramid and carbon fibres for reinforcement of epoxy resin. Composites of different volume fractions of only aramid fibre and only carbon fibre were also prepared. Mechanical properties flexural strength and modulus, tensile modulus and short beam interlaminar shear strength and fracture energy of the plastic composites were measured to determine any "hybrid effect". For better appreciation of the hybrid effect, the mechanical property values of the composites containing different fibre volume fractions, were normalized to 50 per cent volume. The normalized values show that mixing carbon and aramid fibres for reinforcement of plastics provided much more improved mechanical properties (hybrid effects) of the composites than using either carbon fibre or aramid fibre alone.

### Introduction

Fibrous fillers are widely employed in thermosetting resins for dilution purposes and for reinforcement. In the latter category of materials, strong, stiff fibres are dispersed in thermosetting matrix with a view to increasing stiffness and strength. Stiffness and strength of fibre-reinforced plastics (FRP) are functions of fibre properties and quantity of fibre incorporated. For a composite made of continuous uniaxially-aligned fibres, properties in the direction of fibre may be estimated from properties of the components by law of mixtures (Bader & Bowyer, 1977).

$$E_c = E_f V_f + E_m (1 - V_f)$$

where  $E_c$ ,  $E_f$  and  $E_m$  are the moduli of the composite, fibre and matrix respectively, and  $V_f$  is the total volume fraction of fibre. In practice, the law gives a reasonable prediction at very low levels of composite strain but becomes increasingly optimistic as strain increases. Incorporation of two or more different fibres into a single matrix is known as hybridization and the resulting material is generally referred to as a 'hybrid' or 'hybrid composite'.

There is increasing interest in hybrid compos-

ites because these materials offer a range of properties such as fracture toughness and impact resistance that cannot be obtained with a single type of reinforcement. At the same time, material cost is reduced substantially by careful selection of reinforcing fibres (Zweben & Norman, 1976; Zweben, 1977). "Hybrid effect" is the attribute of composites of hybrid construction with properties better than the sum of the components (Philips, 1976a).

Several forms of hybrid construction can be identified (Philips, 1976b), and these are principally:

- (i) Those in which the fibres are intimately mixed throughout the resin and having no intentional concentration of either type of fibre. The present study deals solely with this form of hybrid construction.
- (ii) Those in which the material is formed as discrete layers of a mixture of individual fibres, for example, by the use of hybrid tapes.
- (iii) Those in which each layer is a sequence of plies of a single tape of fibre symmetrical

about the neutral axis of the composite.

- (iv) A specialization of type (iii) in which the core of the component is made entirely of one of the constituent fibres and the shell (outer ply) is of a second fibre. This is referred to as "sand-wiched composite".

In all cases, the resulting material is anisotropic and the properties are depended on the constituent materials and on the structure of the component, i.e. to lay-up sequence.

Carbon fibre offers high stiffness and strength but the impact resistance of composites made from it is relatively low. It has been shown (Moore & Sturgeon, 1973; Beaumont, Reiwald & Zweben, 1975; Reiwald & Zweben, 1975) that resistance to impact damage and improved fracture toughness can be obtained by addition of fibres of high strain-to-failure, such as aramid and glass fibres. Carbon fibre (CF) and aramid fibre (AF) are particularly well suited for hybridization because their coefficient of thermal expansion are similar (Zweben & Norman, 1976). This minimises internal thermal stresses.

The objective of this present work, therefore, was to use a simple technique to examine the existence of hybrid effects in plastic composites prepared from epoxy resin and unidirectional intimate mixture of aramid and carbon fibres.

### Experimental

#### Materials

Aramid fibre, carbon fibre, epoxy resin and a curing agent were the experimental materials. Aramid fibre (Kevlar 49) of 7888 denier was supplied by DuPont Company (UK), and described by the manufacturer as an aromatic polyamide. Carbon fibre ('Grafil' E/A-S) was supplied by Courtaulds Company (UK). The fibre has a tow containing 3,000 individual parallel filaments of approximately 8  $\mu$  in diameter, and sized with about 0.5 per cent epoxy resin as an aid to consumer handling. Epoxy resin (Epikote DX-210-B-80), supplied by Shell Chemicals (UK), is a solution containing 80 per cent by weight of a specially-developed semi-solid

epoxy resin (Epikote DX-210) in methyl ethyl ketone (MEK). The curing agent (Epikure BF<sub>3</sub>-400) supplied by Shell Chemicals (UK) was described as a boron trifluoride monoethylamine complex.

#### Methods

The important aspect of this study was to develop a simple but suitable method for mixing aramid and carbon fibres for reinforcement of epoxy resin. The set up of equipment for mixing of the two fibres, carbon and aramid, and preparation of pre-

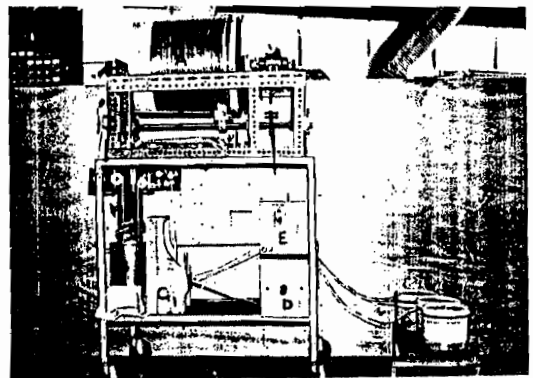


Fig. 1. A complete set up of equipment for preparation of pre-preg showing the drum A, resin bath B, fibre mixing unit C, and the reels for aramid D, and carbon E, fibres

preg is shown in Fig. 1. A similar set up was used for the preparation of all carbon and all aramid pre-pregs.

*Mixing of resin.* Six hundred and fifty grams (650 g) of the resin and 15 g of curing agent were transferred into a plastic container and the mixture stirred until a homogenous system was obtained. Additional MEK, about 200 g, was added to adjust the resin concentration to produce pre-preg with the desired resin content and also to prevent excessive breaking of the brittle carbon fibre. Stirring was continued with magnetic stirrer until a clear resin solution was obtained.

*Mixing of fibres and preparation of pre-preg.* Presence of excessive moisture on the surface of aramid fibre can adversely affect quality of com-

posites, especially interlaminar shear properties (DuPont Company, 1973). For the control of this problem, the fibre was dried at 120 °C in an oven for a minimum of 16 h before use. The fibre was used within 1 h after removal from the oven.

Releasing the twist in the former to produce five separate tows facilitated mixing of the aramid and carbon fibres. As shown in Fig. 2, the five tows were separately passed through eyelets in a plastic plate. Three separate tows of the carbon fibre were simultaneously passed through three of the inner eyelets carrying the aramid fibre. After pass-

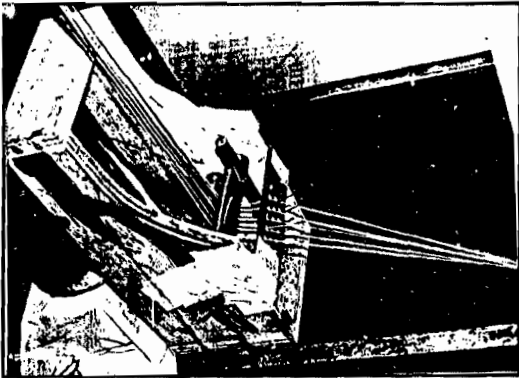


Fig. 2. The fibre mixing unit showing the eyelets through which the carbon fibres and the aramid fibres are passed

ing through the eyelets, the two fibres were combined by introducing a slight twist in them and then passed round a number of pulleys (Fig. 1). The mixed fibre was slowly drawn through a resin bath 160 mm deep and 140 mm diameter. The wet mixed fibre was passed round another set of pulleys and finally round a drum 429 mm wide and 380 mm diameter (Fig. 1), previously covered with PTEE release sheet. The number of turns of fibre around the drum per minute depended on the speed of rotation of the drum. Hence, by altering the rotation speed of the drum, the total number of turns per minute of fibre around the drum was varied to produce pre-pregs of different fibre volume fractions. Rotation of the drum was done manually.

The resin-impregnated-mixed fibre was left on the drum for at least 18 h when most of the solvent should have evaporated. The PTFE sheet, together with the pre-preg, was then removed from the drum and allowed to dry in a well-ventilated area for a further 48 h or more. This drying schedule removed residual solvent and gave a pre-preg with good handling characteristics. It was also important to protect the pre-preg from dirt particles and direct sunshine during drying as these adversely affected aramid fibre properties in particular. Storing them at temperatures 0 °C or lower until required for curing extended the shelf life of the pre-pregs.

For all aramid and all carbon fibre pre-pregs, a set up similar to that used for the mixed fibres was employed except that the unit for fibre mixing was not utilized. The aramid fibre was impregnated with resin in its original twisted form. For the carbon fibre pre-preg, three separate tows were slightly twisted together before passing through the resin bath.

*Curing procedure.* The precise conditions necessary to produce satisfactory composites of the desired fibre content from a given pre-preg depend on the type and dimensions of the mould. For this work, the following schedules were used for moulding 305 mm × 114 mm × 2 mm plaques:

|                    |                             |
|--------------------|-----------------------------|
| Mould temperature: | 24 °C                       |
| Press temperature: | 170 °C                      |
| Dwell time:        | 20 - 25 min                 |
| Curing pressure:   | 0.5 - 0.7 MN/m <sup>2</sup> |
| Curing time:       | 4 h                         |

A hydraulic cutter was used to cut the dried pre-pregs to fit into the mould. Five or six of the pre-preg laminates were placed together in the mould, as the case may be, to produce a cured plaque 2 mm thick. It is essential that fibre direction of laminates placed in the mould be the same in order to produce a unidirectional fibre composite. After the cold mould had been racked with the desired number of pre-preg laminates, it was closed and placed in a heated press (170 °C). The press was then closed gently until the mould was just in contact with the top platen of the press.

A dwell time (20-25 min) was allowed for the mould to reach the temperature of the press and also for any excess resin to be forced out of the pre-preg laminate. At the end of the dwell time, when the resin should begin to set, the full curing pressure,  $0.6\text{MN/m}^2$ , was applied for 4h. It was

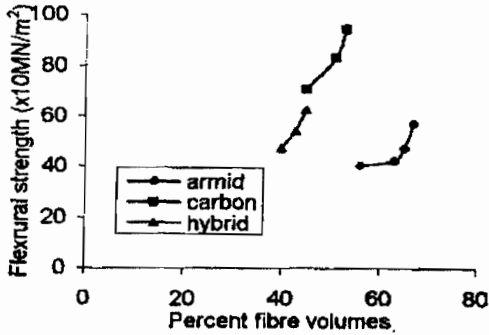


Fig. 3. Flexural strength of plastic composites vrs percent fibre volumes

important to use a minimum pressure during the dwell time so as not to force out more resin from the pre-preg than necessary. The time for application of full pressure should also be controlled to prevent voids and a cure composite plaque thicker than desired. At the end of the curing period (4h), the pressure was released and press allowed cooling with mould. The mould was later removed and the cured composite trimmed and kept for preparation of test pieces.

Determination of mechanical properties. The mechanical properties investigated for composites were:

- i. Flexural strength and modulus,
- ii. Short beam interlaminar shear strength,
- iii. Tensile modulus.

A floor model Instron TT-C testing machine was used for carrying out all tests. For the preparation of the test pieces, a "clipper" masonry-cutting machine was used.

Flexural strength and modulus of the composites were determined in accordance with ASTM

Method D 7907-70 (ASTM, 1976a). In this procedure, the Instron machine was used to subject rectangular test pieces (of dimension 150 mm long  $\times$  20 mm wide  $\times$  2 mm thick, prepared from the composites) to a three-point load over a 100-mm span. Cross head speed was 20 mm/min, and applied load at full deflection 5,000 N. Three test pieces were tested from each plaque of the same total fibre volume fraction. Since aramid composites exhibit a non-linear stress-strain behaviour or yield after about half the ultimate stress (DuPont Company, 1973), the modulus of the hybrid composites were determined on the bases of the linear, or initial portion of the stress-strain curve. Values for flexural strength and modulus recorded for the hybrid composites were the mean of nine test pieces prepared from three plaques of the same total volume fraction. For the aramid and carbon fibre composites, the recorded values for the flexural strength and modulus were the mean of six test pieces prepared from two plaques of the same fibre volume fraction.

The interlaminar shear strength of the composites were determined in accordance with ASTM Method D 2344 (ASTM, 1976b). For this test, the Instron machine was used to subject sample pieces, 15 mm long along fibre direction, 12 mm wide across fibre and 2 mm thick, to three-point loading over 10 mm span. This short span (hence the name 'Short Beam Shear') caused the bending moment to be directed perpendicular to the direction of load application, and the shear stress to be developed parallel to the longitudinal axis of the specimen. Equipment parameters used were:

|                                |            |
|--------------------------------|------------|
| Cross head speed:              | 2 mm/min   |
| Chart speed:                   | 100 mm/min |
| Load at full-scale deflection: | 5,000 N    |

The load at which failure first occurred was recorded and the mode of failure of test piece also noted. All values recorded were the mean of six measurements.

Tensile modulus of all the composites was determined by using straight-sided test pieces. The samples were 25 mm long  $\times$  10 mm wide  $\times$  2 mm

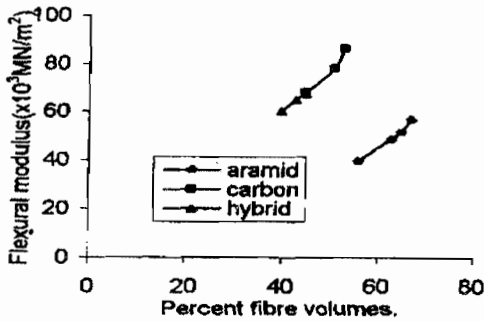


Fig. 4. Flexural modulus of plastic composites vrs percent fibre volumes

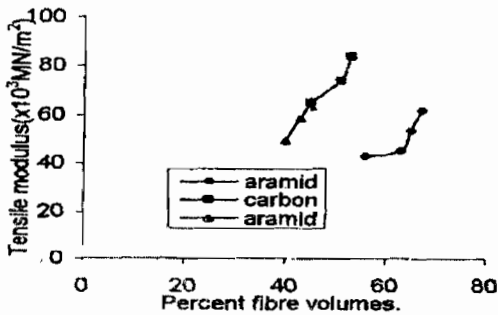


Fig. 5. Tensile modulus of plastic composites vrs percent fibre volumes

thick. Tensile modulus was determined by using the Instron machine. Jaw separation rate was 0.1 mm/min. Maximum load was determined directly from the calibrated Instron chart, and elongation was measured with strain-gauge extensometer connected to the machine X-Y chart drive amplifier. Gauge length of sample was 20 mm and chart speeds 50 mm/min. All values recorded were the mean of six measurements.

### Results and discussion

In order to use a material confidently, we need to understand how it behaves under applied load. The primary objective in this work, therefore, was to provide some insight into existence of hybrid effects in the mechanical properties of compos-

ites reinforced with two types of fibres that have different stiffness and strain characteristics. To do this, idealized models of composites were constructed which incorporated the major phenomena involved.

The results of mechanical properties investigated for the hybrid composites, carbon fibre reinforced plastics (CFRP) and aramid fibre reinforced plastics (AFRP) are presented in Fig. 3-7. For a meaningful interpretation of the experimental results, they were normalized to 50 per cent total fibre volume, which are presented in Table 1.

At 50 per cent fibre volume, flexural strengths of the CFRP, hybrid and AFRP were 783, 594 and 363 MN/m<sup>2</sup> respectively. From the law of mixtures, flexural strength of the hybrid composite was 573 MN/m<sup>2</sup>. The normalized value (594 MN/m<sup>2</sup>), therefore, suggests some "hybrid effect" in the flexural strength of the hybrid composite. Consideration of the actual experimental values indicates that the energy level at which flexural damage occurred in the hybrid composite was lower than that of the CFRP and the AFRP (Fig. 3).

With respect to the normalized values, the flexural modulus of the hybrid was 75 GN/m<sup>2</sup>. It was 76 GN/m<sup>2</sup> and 35 GN/m<sup>2</sup> for the CFRP and AFRP respectively. Comparison of the flexural modulus value of 56 GN/m<sup>2</sup> obtained by the law of mixtures to the normalized value of the hybrid composite (75 GN/m<sup>2</sup>) suggests "hybrid effect" in the flexural modulus of the hybrid composite. The lower flexural modulus value of the hybrid composite compared to that of the CFRP, as observed in this work (Fig. 4), points to ductile behaviour of the hybrid. That is, whenever damage occurs in the hybrid, its extent will be more localized than in the CFRP. Since residual strength of materials decreases as the size of damaged zone increases, the localization of damage provided by the prepared hybrid is a result of greater structural reliability. In fact, the improved ductility of the hybrid compared to the CFRP was shown by their modes of failure under flexural loading. Flexural damage was catastrophic in all the CFRP samples but was observed to be non-catastrophic in the

hybrid composites.

The normalized tensile modulus of the CFRP was 72 GN/m<sup>2</sup>. Similar values obtained for the hybrid and the AFRP were 63 GN/m<sup>2</sup> and 38 GN/m<sup>2</sup> respectively. The law of mixtures predicted a tensile modulus of 55 GN/m<sup>2</sup> for the hybrid, which is lower than the normalized value (63 GN/m<sup>2</sup>). The indication is that mixing the two fibres produced "hy-

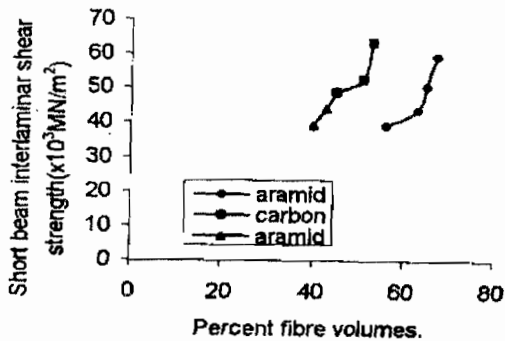


Fig. 6. Short beam interlaminar shear strength of plastic composites vrs percent fibre volumes  
 "brid effect" in tensile modulus of the hybrid composite. Besides, comparing the experimental values recorded for the hybrid, the CFRP and the AFRP (Fig. 5), it can be said that the hybrid is less stiff than the CFRP but more stiff than the AFRP.

The interlaminar shear strength of the hybrid composite with 50 per cent total fibre volume was 49 GN/m<sup>2</sup>. The normalized value for the CFRP was 54 GN/m<sup>2</sup> and 35 GN/m<sup>2</sup> for AFRP. Interlaminar shear strength is largely determined by fibre to resin bond strength since this failure usually occurs at the interface. The low values recorded for the hybrid composite would, therefore, suggest that hybridization is not necessarily the best method of improving interlaminar shear strength of CFRP although it seems possible for AFRP. This is also observed in the actual experimental results (Fig. 6).

In an attempt to obtain some information about fracture toughness of the composite materials, an arbitrary measure, the 'fracture energy', was introduced. The values recorded were computed as the product of the area under the stress-strain curve for flexural loading and the maximum load applied to the test pieces. The reason for this procedure was to determine the maximum energy required for the ultimate failure of the composites, especially the hybrid composite. By this methods, the hybrid material with 50 per cent fibre volume fraction recorded fracture energy of 236 units for complete failure; the CFRP failed at 139 units and the AFRP at 300 units. These values indicate improvement in the fracture toughness of the CFRP

TABLE 1  
 Mechanical properties of CFRP, hybrid and AFRP  
 (Values were normalized to 50 per cent fibre volume)

| Mechanical property  | CFRP | Hybrid      | AFRP |
|--|------|-------------|------|
| Flexural strength (MN/m <sup>2</sup> )                                     | 783  | 594 (573)** | 363  |
| Flexural modulus ( $\times 10^3$ MN/m <sup>2</sup> )                       | 76   | 75 (56)     | 35   |
| Tensile modulus ( $\times 10^3$ MN/m <sup>2</sup> )                        | 72   | 63 (55)     | 38   |
| Short beam interlaminar shear strength ( $\times 10^3$ MN/m <sup>2</sup> ) | 54   | 49          | 35   |
| Apparent work to failure (arbitrary unit)                                  | 139  | 236         | 300  |

\*\* Values in brackets were obtained by the law of mixtures.

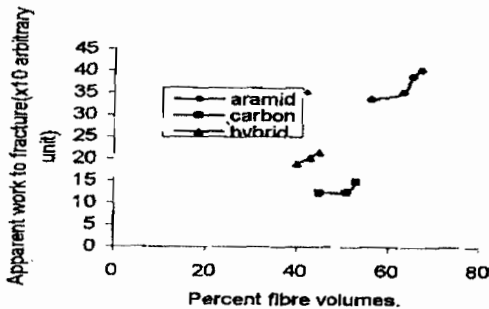


Fig. 7. Apparent work failure of plastic composites vrs percent fibre volumes

by hybridization. The improved fracture toughness of the hybrid was contributed by the aramid fibre, which alone produced a composite with 300 fracture energy units. The actual experimental results (Fig. 7) also show this behaviour.

The work of fracture of composite materials is the sum of the separate energies expended in the fracture process (Harris, 1972). These components are summarized as follows:

- i. Work of fracture of the matrix.
- ii. Work of fracture of the fibres.
- iii. Energy required to de-bond fibres from the matrix.
- iv. Elastic energy released after a fibre/matrix de-bond.
- v. Frictional work required to pull broken fibres from the matrix after the latter has cracked.
- vi. The plastic work of shearing fibres that does not lie normal to the crack face.

These factors contribute to the fracture toughness of various types of composites and, indeed, suggest what steps may be taken in an attempt to improve the toughness of any composite. In the case of AFRP, the first two factors will contribute very significantly to its toughness. For the CFRP, the important factor will be the fifth. The important factors contributing to fracture toughness of

the hybrid composite will, therefore, be the first, second and fifth.

### Conclusion

The values recorded for all the mechanical properties investigated in this work show that carbon-aramid hybrid composites have lower strength and stiffness, lower interlaminar shear strength and higher fracture energy than CFRP. The low stiffness of the hybrid composites implies that they are ductile materials with improved fracture toughness, a hybrid effect, contributed by the aramid fibre. The measured 'fracture toughness' values support the claim that high fracture energy is required for complete failure of hybrid composites. Mixing carbon and aramid fibres for reinforcement of plastics, therefore, provided improved mechanical properties (hybrid effect) of the composite materials. The lower interlaminar shear strength of our hybrid composites compared to that of the CFRP indicates poor fibre/resin bond formation in the hybrid. Hybridization, however, improved the strain properties of the hybrid composite by changing the catastrophic failure in the all-CFRP to a ductile failure in the hybrid. Aramid and carbon fibre hybrid plastic composites should, therefore, have the potential as suitable material for engineering applications:

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