DIAMOND-FIBRE REINFORCED PLASTIC COMPOSITES

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Diamond fibre reinforced poly(methylmethacrylate) (PMMA, perspex) composite blocks (100×10×4 mm) have been fabricated by embedding CVD diamond coated tungsten wires within a perspex matrix. Various volume fractions of diamond have been used, as well as varying positions of the fibres within the composite. We find that even 1% fibre volume fraction can lead to an increase in the Young's modulus of the composite of a factor of 6.

1 Introduction

The ability to deposit thin films of polycrystalline diamond on different substrates has enabled scientists and engineers to exploit some of its superlative properties in a variety of electronic devices and mechanical applications [1]. However, one of the properties of diamond that has rarely been used to date is its extreme stiffness. One potential route to exploit the very high Young's modulus of diamond is in the form of thin wires or fibres, in which the chemically vapour deposited (CVD) diamond coatings are grown uniformly onto the surface of a metal or ceramic core wire [2]. These 'diamond fibres' have core diameters ranging from 10-200 μm, with the thickness of the diamond coating ranging from 10-100 μm. Some properties of these diamond fibres compared to fibres made of other materials are given in Table 1.

Table 1. Properties of diamond fibres measured using a resonance method [3], whilst the fracture strength was measured from a tensile testing experiment[4]. Literature values for selected alternative materials are given for a comparison [5].

Material	Young's Modulus / GPa	Fracture strength / GPa	Density / (g cm^{-3})
Natural diamond	1050	~ 3-5	3.51
CVD diamond	700-1000	0.5-1.4	3.5
Diamond fibres (50 μm diamond coating on 100 μm tungsten core)	800	1-2	7.4
Tungsten	362	2.3	19.3
SiC (Textron)	400 check	3.75	3.4
C fibre (PAN)	490	2.2-2.5	1.91
PMMA (Perspex)	4-5	~0.1	1.2

To exploit this exceptional stiffness, it is necessary to embed many fibres in a matrix material to produce a fibre-reinforced composite. Previously, such diamond fibres have been used to make stiff, lightweight metal matrix composites (MMCs), using Ti as the matrix material [5,6,7]. However, in order to obtain uniform fibre distribution within the Ti matrix, it was necessary to sputter coat each fibre with a thin uniform layer of Ti,

which added extra time and expense to the manufacturing process. Furthermore, it was necessary to add substantial numbers of diamond fibres to the metal matrix in order to obtain any appreciable benefit, and this high fibre volume fraction made Ti MMCs prohibitively expensive. But one way to reduce the costs of diamond fibre reinforced composites is to reduce the volume fraction of fibres required. This can be achieved in two ways, first, by using a matrix material such as plastic, which has a much lower Young's modulus than diamond, so that even a small number of fibres make a substantial difference to the overall composite mechanical properties. Such a stiff, strong, but extremely lightweight, and relatively inexpensive plastic matrix composite (PMC), might find numerous uses as a structural material in, say, aerospace applications, where high performance for minimum weight is paramount. If continuous fibres are used, aligned in the same direction, then the properties of the composite become anistropic. Different values for the Young's modulus will be obtained perpendicular to the fibre direction to those parallel to it. The second way to reduce the costs of fibre reinforcement is to simply use less fibres, but position them in ways that maximise their effect. For example, it has been shown that placing fibres close to the edges of a composite, rather than evenly throughout the bulk, leads to a much greater stiffness [8].

In this paper we investigate the feasibility of using such continuous, aligned diamond fibres for reinforcing plastics in general, using poly(methylmethacrylate), also known as PMMA or Perspex, as a test case. Although PMMA is not generally used as a structural plastic, it was chosen because (i) it is easy to make in the laboratory using cheap easy-to-obtain reagents, (ii) it is transparent, allowing the position of the fibres within the PMC, and any cracks or defects to be seen, and (iii) it is reasonably rigid, but has a Young's modulus significantly less than that of diamond. We also investigate whether significant modulus increases can be achieved by using small volume fractions of diamond fibres arranged in different positions within the PMC.

2 Methods

The diamond fibres were deposited in a hot filament CVD reactor adapted for fibre substrates. The choice of substrate core material is limited to those materials which can survive the aggressive deposition conditions, and which can be extruded in the form of wires. Past experience [5] has shown that despite its unwanted high density and cost, tungsten is one of the very few materials which make suitable core materials. Forty tungsten wires of length 10 cm and diameter 100 μm were loaded into the CVD reactor, and clamped in a cage at a set distance of 5 mm from two 2 mm-thick tungsten wires, which acted as the hot filaments. Deposition conditions were process gases of 1%CH_4 in H_2 with a total flow 200 sccm at a pressure of 20 Torr, and the filament temperature was 2200 K (as measured by a two-colour optical pyrometer). These conditions deposited diamond uniformly around the circumference of the W cores at a rate of about 0.7 μm h^{-1}. Thus, for a 50 μm coating, we require 70 h (3 days) continuous growth. An example of some of the diamond fibres can be seen in Fig.1.

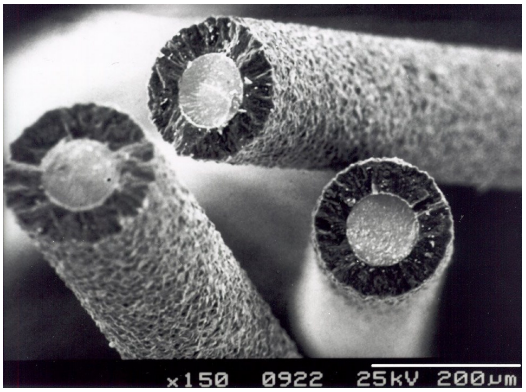

Figure 1. Electron micrograph showing some of the diamond fibres used in the PMCs. The W cores are 100 µm in diameter, with 50 µm of diamond deposited around them. The total fibre diameter is 200 µm, and the diamond volume fraction is 75%. The mechanical properties of these fibres are given in Table 1.

PMCs were made by embedding these fibres into a block of PMMA. Blocks of PMMA were cast in an aluminium mould (100×20×50 mm) using a standard bulk polymerisation process. The monomer reagent, methylmethacrylate (Aldrich) was stirred with 0.5% by weight of a benzoyl peroxide initiator (Aldrich) at 90-100°C for 10-20 mins, until the solution became thick and syrupy. In this state the solution could be allowed to cool, and would remain liquid until baked at a later date. The cool, syrupy solution was then poured into the mould to a depth of about a few mm (depending upon where the fibres were to be positioned) and then placed into an oven at 40°C for 10 h, which caused it to set solid. The setting process also caused the volume to shrink by about 15%. A number of diamond fibres were then placed lengthways into the mould and evenly spaced out. More liquid polymer was then poured over the fibres to embed them, and the mould was again baked at 40°C for 10 h. In order to test the effect of different geometries of fibres upon mechanical properties, various different fibre arrangements were produced, with one, two or more layers of fibres, as shown in Fig.2. After the PMC was complete, it was given a final oven bake at 80-90°C for 1 h to complete the polymerisation process. The resulting cast PMC was machined to create rectangular samples of dimensions 100×20×4 mm. PMCs were produced containing varying numbers of diamond fibres, and with different fibre core diameters. Two PMCs were also produced using commercially available (Textron) SiC fibres.

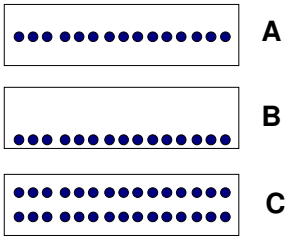

Figure 2. Schematic cross-sections through the PMCs showing the 3 different positions of fibres. (A) the fibres are embedded along the central line, (B) the fibres are embedded close to one edge, (C) multiple rows of fibres are embedded equidistant from the centre line.

The PMCs were tested to destruction using a screw-driven materials test machine. The samples were subjected to four-point loading as illustrated in Fig.3, and the central deflection, d, was measured as a function of the applied load, p. For such a system, the Young's modulus, E, is given by [9]:

$$E = \frac{p}{4dwh^2L}$$

where L is the distance between the upper and lower loading points, and w and h are the width and height of the PMC sample, respectively.

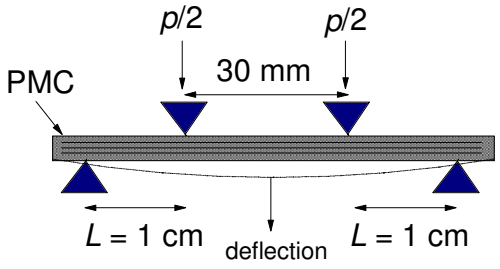

Figure 3. Schematic diagram of the 4-point bend test. Deflection is measured as a function of applied load, p.

3 Results

An example of one of the diamond fibre PMCs before testing is shown in Fig.4. Cross-sections through the PMCs (Fig.5) showed that the PMMA had uniformly coated each diamond fibre, with no obvious signs of voids or holes. The fibres were rigidly embedded, and showed no tendency to pull out.

Figure 4. A photograph of the top view of a diamond fibre PMC containing about 100 fibres.

Figure 5. A cross-section through one of the PMCs, showing two layers (indicated by the arrows) of 15 diamond fibres each equidistant from the centre of the block.

Curves of load against deflection for the PMCs were all linear up to the point at which the composite fractured. These curves provided an accurate estimate of the gradient, and hence Young's modulus. Repeating the measurement on identical perspex blanks allows us to estimate the uncertainty in each modulus value as better than 5%. Figure 5 shows the variation of Young's modulus of the PMCs against number of fibres, for the three fibre positions shown in Fig.2. The volume fraction of fibres, V_f, was estimated using:

$$V_f = \frac{\pi D^2 N}{4wh},$$

where w is the width and h the height of the PMC, D is the diameter of the fibres, and N is the number of fibres. Thus, 100 fibres corresponds to a fibre volume fraction of only 1%, and a diamond volume fraction of only 0.75%.

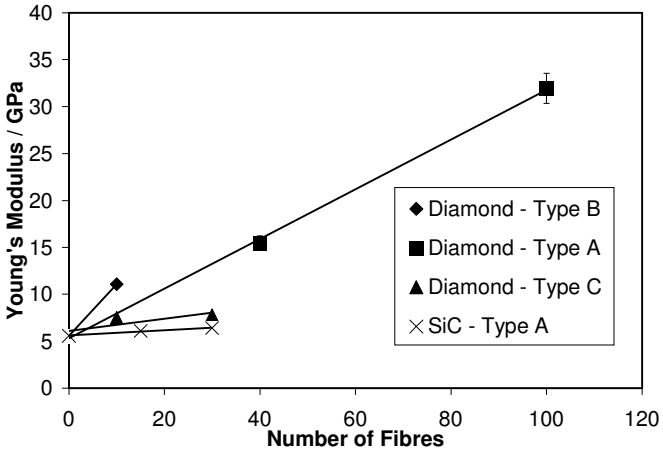

Figure 6. A plot of Young's modulus of the PMCs against number of fibres, for the three different types of fibre position depicted in Fig.2. Also shown for comparison are data for a PMC made with Textron SiC fibres.

It is clear from Fig.6 that even a very small volume fraction of diamond fibres has caused a substantial increase in the stiffness of the PMCs. For the fibres embedded along the centre line (A), which is not the optimum configuration for reinforcement, we still observe an increase in Young's modulus of over 6 times for an addition of only 1% fibre volume fraction. This is a significant improvement, and shows the benefits of using diamond for plastic reinforcements, especially when compared to the SiC fibres which (by extrapolation) only show a factor of 1.5 improvement. Further improvements can be achieved if the fibres are placed at one edge (B). Although only one sample was tested, we can see that the gradient is much steeper than for the other two fibre positions, and this shows great promise as a way to obtain maximum benefit from the fibres with minimum cost. An obvious extension to this is to use two layers of fibres, one at each edge, and this is ongoing work. For diamond fibres spread evenly throughout the PMC, or in layers equidistance from the centre line, the improvement in stiffness is not so dramatic, and is comparable to the SiC fibres.

4 Concluding Remarks

We have shown that even very small volume fractions of diamond fibres can substantially increase the Young's modulus of a plastic composite. Even greater increases can be achieved if the fibres are arranged in layers at the edges of the PMC. This means that the number of diamond fibres required for a given modulus increase can be reduced to a minimum, so reducing the overall cost of the PMC. Thus, such composites may finally have an economical route to allow them to be used in some advanced structural applications. Further work still needs to be done, however, particularly on the effect upon

the modulus of the number and position of the fibres within the PMC. Also, other plastics need to be investigated, such as epoxies which are more commonly used as structural materials.

Another potential use for such PMCs is as electrically insulating 'heat-pipes', since the high thermal conductivity of diamond could be used to transfer heat rapidly along the length of the composite without heat leaking out sideways.

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References

1. May P. W., Diamond Thin Films: a 21st Century Material, *Phil. Trans. R. Soc. Lond. A* **358** (2000) pp. 473-495.
2. May, P. W, Rego, C. A., Thomas, R. M., Ashfold, M. N. R., Rosser, K. N. and Everitt, N. M. E., CVD diamond wires and tubes, *Diam. Relat. Maters*. **3** (1994) pp. 810-813.
3. Nicholson, E. D., Baker, T. W., Redman, S. A., Kalaugher, E., Rosser, K. N., Everitt, N. M., Ashfold, M. N. R. and Partridge, P. G., Young's Modulus of Diamond-Coated Fibres and Wires, *Diam. Relat. Maters*. **5** (1996) pp. 658-663.
4. Everitt, N. M., Shatwell, R. A., Kalaugher, E. and Nicholson, E. D., Mechanical Properties of Diamond Fibres, *Mat. Res. Soc. Symp. Proc*. **383** (1996) pp. 379-384.
5. Partridge, P. G., May, P. W. and Ashfold, M. N. R., Chemical Vapour Deposited diamond fibres: Manufacture and potential properties, *Maters. Sci. Technol*. **10** (1994) pp. 177-189.
6. Partridge, P. G., May, P. W., Rego, C. A. and Ashfold, M. N. R., Potential for diamond fibres and diamond fibre composites, *Maters. Sci. Technol*. **10** (1994) pp. 505-512
7. Nicholson, E. D., Meaden, G., Kaulaugher, E., Partridge, P. G., Ashfold, M. N. R., May, P. W. and Wisbey, A., Continuous Diamond fibres and diamond fibre reinforced composites, *Diam. Films Technol*. **6** (1996) pp. 217-240.
8. Smith, D. J. and Partridge, P. G., Flexural stiffness envelopes for planar and multilayered systems containing dissimilar materials (Ti-alloy and Ti-MMC), *Proc. Inst. Mech. Eng*. **213**, part L, (1999) pp. 1-20.
9. Gere, T. M. and Timoshenko, S. P., *Mechanics of Materials* (Brooks Cole, Monterey, CA, 1984).