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Effect of woven structure on flexural and shear fracture behaviour of three-dimensional carbon–carbon composites

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The various structures and yarn-bundle spacing of 3D-5 (three directions, 5 mm), 5D-5 (five directions, 5 mm), and 3D-7.5 (three directions, 7.5 mm) preforms are fabricated into three-dimensional carbon–carbon (C/C) composites. The fracture behaviour is different due to the increased number of arrangement directions and fracture toughness. The flexural strength, short-beam shear strength, and microstructure of fractured specimens were investigated to study the mechanical behaviour of these composites. The results revealed that the strengths of the three-dimensional composites were larger than the five-dimensional ones in all reinforcement structures, but five dimensions were better than three dimensions in fracture toughness. The five-dimensional composites showed stepwise fracture, and the three-dimensional vertical fracture. In the aspect of yarn-bundle spacing, the flexural strength of the 7.5 mm type (spacing between two *Z* axial yarn bundles is 7.5 mm) was higher than the 5 mm type, while the short-beam shear strength was contrary, Furthermore, according to SEM observations, the fractural surface was ruptured along the *Z*-axis, indicating that a stress concentration would exist in the fringe of the *Z*-axis, and that the *Z*-directional fibres arrested delamination propagation.

1. Introduction

The idea of three-dimensional reinforcements was formed from the needled felts, pile fabrics, and stitched felts, because these structures lack a through-the-thickness strength. Accordingly, we expected to find that three-dimensional reinforcements gave interlaminar shear strength and impact toughness. Moreover, three-dimensional reinforcements can be used to change complex shapes and to afford the reductional capability of fabrication [1, 2]. Therefore, several investigations have been devoted to the development of three-dimensional and multi-directional reinforcements.

Carbon–carbon composite is an available engineering material for use above 2000 °C. It is composed of carbon (or graphite) fibre and carbon (or graphite) matrix. Generally, the matrices used are phenolic resin, petroleum pitch or coal tar pitch. The wide fabrication process for carbon–carbon composite is the first point at which to introduce carbon matrix into a multidirectional fibrous preform, through the multiple impregnation of the carbonaceous substrate, and subsequent carbonization or graphitization under an inert atmosphere, and then repeating the cycles to densification until the required density is achieved.

The conventional method used to increase the efficiency of impregnation and carbonization is through multi-cycles and long processing times at atmospheric pressure [3, 4]. Nowadays, hot isostatic processing (HIP) under high pressure and temperature is used, not only to reduce the processing time but also obtain better effects than the previous method [5].

To improve the thermal, mechanical, and physical properties of the carbon–carbon composites we may consider appropriate designs of weaving factors such as yarn-bundle spacing, weaving density, fibre orientation, types of fibre, and volume fraction of fibre in the required directions, etc. [6–10]. In the past, some literature mentioned combining these factors for investigation. Therefore, in this work we combined the parameters, to weave a triaxial arrangement of fibres in three orthogonal directions to obtain a three-directional reinforcement and added two more $\pm 45°$ direction fibres in the X–Y plane to give a five-directional reinforcement. The preforms were made into carbon–carbon composites. The various structure and yarn-bundle spacing were changed in order to investigate the effects of flexural and shear properties. Furthermore, optical microscopy and scanning electron microscopy (SEM) were employed to study the failure mechanisms in both three- and five-dimensional carbon–carbon composites.

2. Experimental procedure

2.1. Materials and specimens

The yarn bundles of carbon fibre, with an average width of band state of 4 mm, were purchased as the PAN precursor of Hysol/Grafil 12 K (density

= 1.82 g cm^{-3}). The properties and composition of petroleum pitch are shown in Table I.

Three-dimensional preforms with three directions (3D) and five directions (5D) were woven by the process described previously as shown in Figs 1 and 2 [11]. The structural geometries of 3D and 5D are shown in Fig. 3 and the preform size was 145 mm × 145 mm × 6.4 mm. The characteristics of various three-dimensional structures are listed in Table II.

2.2. Hot-isostatic pressing process

Three-dimensional preforms were impregnated under vacuum-pressure using a hot resin transfer system and cured at 250 °C. The previous impregnation was performed at low pressure to fix the shape and construction of substrates. After initial impregnation, the preforms were submerged in pitch, and sealed in a thin-walled stainless container, and were then ready for hot isostatic pressing (HIPing).

The sealed container was placed in a furnace in a pressure vessel for HIPing. The impregnated preforms were heated at a programmed rate to 600 °C and the pressure was increased to 11 000 p.s.i. (75.85 MPa) after 7 h and maintained at 15 000 p.s.i. (103.43 MPa) for 2 h. The temperature and pressure profile of the HIPing are shown in Fig. 4. After HIPing, the composites were carbonized in nitrogen gas at 900 °C and further graphitized in vacuum and argon at 2200 °C, as shown in Fig. 5.

A schematic representation of the three-dimensional carbon–carbon HIPing process is shown in Fig. 6. The processes were repeated for multiple cycles until the desired density of composites was attained.

TABLE I Properties and composition of petroleum pitch

Softening point (°C)	90–130
Viscosity at 250 °C (cP)	25–30
Carbon content (%)	> 40
Density (g cm^{-3})	1.2–1.3
Xyline insoluble (%)	< 10
Quinoline insoluble (%)	< 1
Sulfur content (%)	< 3
Ash content (%)	< 0.15

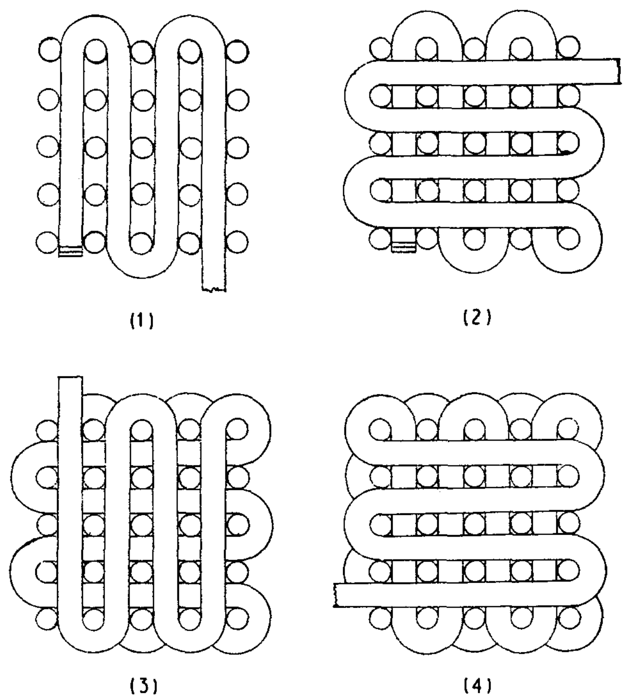

Figure 1 The woven process of orthogonal three-directional fabrics (3D-5 and 3D-7.5) [11].

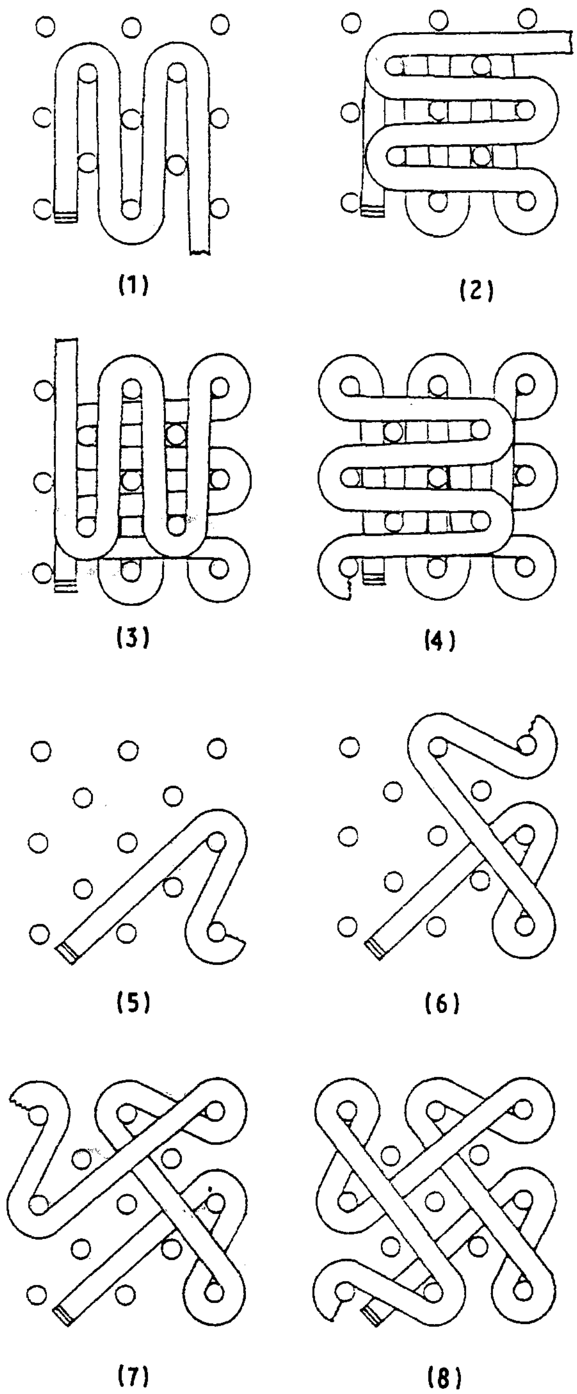

Figure 2 The woven process of five-directional fabrics (5D-5) [11].

2.3. Examination of properties

2.3.1. Flexural test

According to ASTM D790-84a standard, the flexural properties of composites were measured in a three-point bending test with 130 mm × 13 mm × 6.4 mm test pieces which had a span of 100 mm.

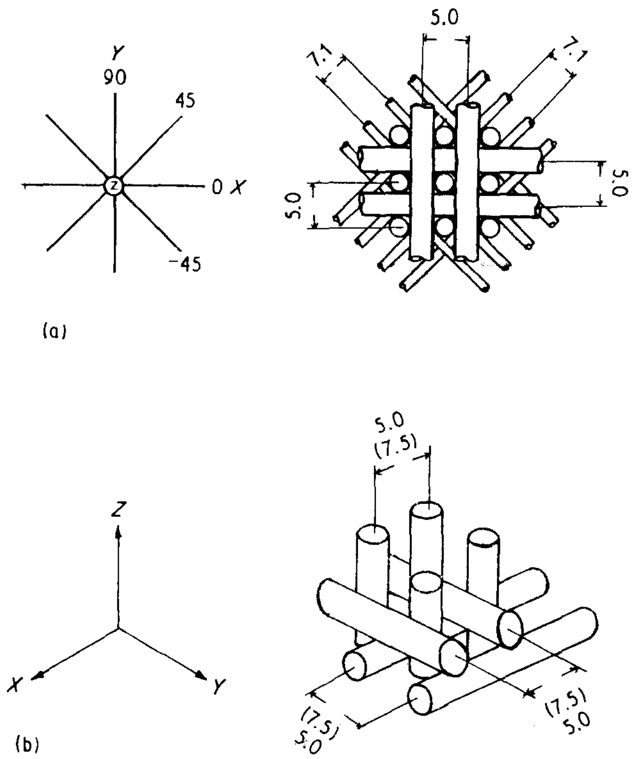

Figure 3 Structural geometry of the three-dimensional fibrous preforms. (a) 5D structure, (b) 3D orthogonal weave. Dimensions in millimetres.

TABLE II Characteristics of various three dimensional structures

		3D-5	5D-5	3D-7.5
Number of fibres[a] in *X*, *Y* axial (0° or 90°) (ends/mm^2)		4552	2897	4491
Number of fibres in *X*, *Y* axial (+ 45° or − 45°) (ends/mm^2)		0	1665	0
Number of fibres in *Z* axial (ends/mm^2)		1026	514	470
Number of yarn layers:	0°/90°	11	7	16
	± 45°	0	8	0
Fibre volume fraction (%)		50.6	50.1	49.8

[a]*X*, *Y* axial: *X* direction of the *Y–Z* plane or the *Y* direction of the *X–Z* plane.

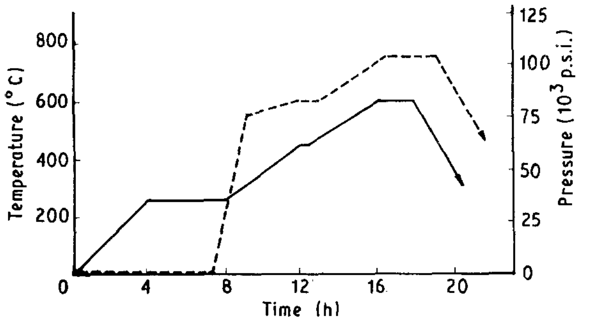

Figure 4 The (——) temperature and (– – –) pressure profiles of the HIPing process.

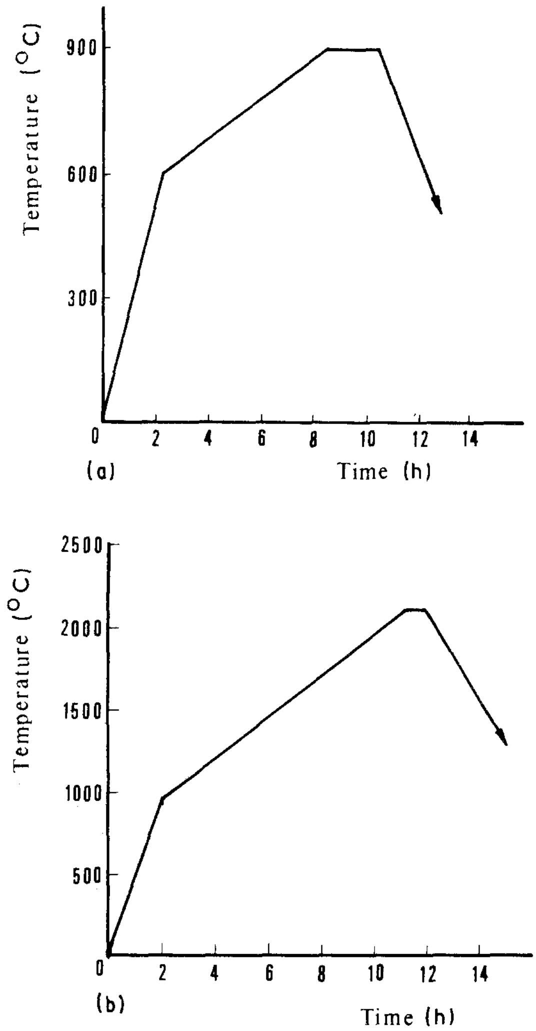

Figure 5 Relation between time and temperature. (a) Carbonization cycle, (b) graphitization cycle.

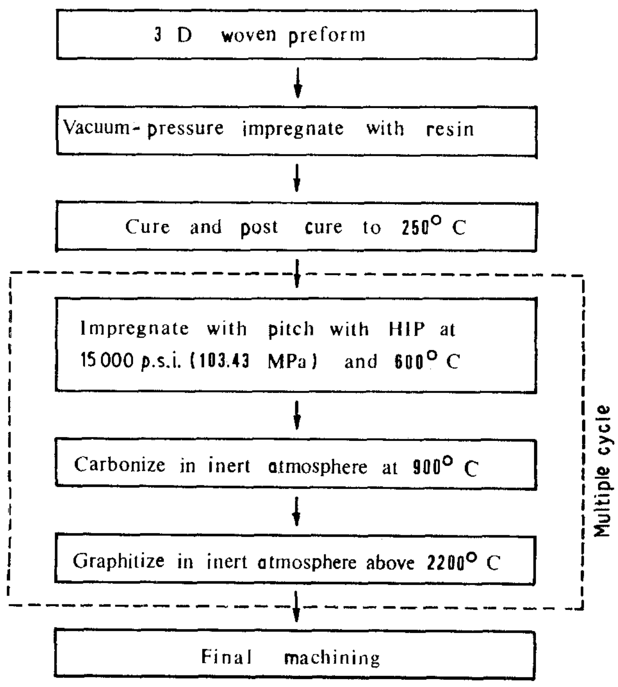

Figure 6 Schematic representation of the three-dimensional carbon–carbon HIPing process.

2.3.2. *Short-beam shear strengths test*

The short-beam shear strengths were examined using ASTM D2344-84 standard. Specimen sizes were 38.4 mm × 6.4 mm × 6.4 mm and the span length was 25.6 mm.

2.3.3. *Analysis of fractural surface and microstructure*

The fractural surface and microstructure of composites were examined using optical microscopy and a Cambridge S360 scanning electron microscope.

3. Results and discussion

3.1. Flexural properties

The flexural properties of various woven structures are shown in Fig. 7. The flexural strength of the 3D structure is higher than the 5D structure, due to the effect of axial fibre contents, as shown in Table II. The axial fibre contents of the 3D-5 and 3D-7.5 are both higher than the 5D-5. Although the 5D structure has about equal fibre numbers with 3D, the force is divided into different directions ($\pm 45°$) for the 5D structure, giving declining reinforcement. Therefore, the flexural strengths of the 3D structure are better. Moreover, scanning electron micrographs of fracture surfaces also confirmed the results. Fig. 8 shows that the fracture surface of the 3D structure is more orderly than the 5D structure. The latter appears to show yarn bundles debonding along $\pm 45°$ directions, so the flexural strength of the 3D structure is naturally preferable.

The effect of yarn-bundle spacing for the flexural strength of the 7.5 mm type (spacing between two *Z* axial yarn bundles is 7.5 mm) is stronger than for the 5 mm type, because the 7.5 mm type radius of curvature of the axial yarn bundles is larger than that of the 5 mm type, i.e. the angle of bending of the former is smaller.

The load of both 3D-5 and 3D-7.5 is larger than 5D-5. Fig. 9 shows the load–deflection curves of the flexural tests, where the area under the curve for the 5D structure has more deflection than for the 3D structure. Therefore, the amount of absorbed energy occurring before failure is larger than in the latter, because 3D deformation is mainly controlled by the axial yarn bundles and, therefore, it obviously shows brittle fracture, while the 5D structure has two more $\pm 45°$ direction fibre reinforcements, and exhibits a tough zone behind the rupture point. Therefore, multidirectional reinforcements will distribute the loading stress more, and lead to the better resistance to deformation.

The fracture behaviour for 3D and 5D composites is shown in Fig. 9. We found that the source of strength for the 3D structure was primarily on the axial yarn bundles, and then crack propagation initiates from the axial yarn bundles of the outer layer and extends in an orderly manner to the off-axis yarn bundles. As a result, the force of resistance of the off-axial yarn bundles is so weak that they all depend on the axial yarn-bundle condition which will create only one fracture behaviour. On the other hand, the 5D structure comprises the axial and the off-axial ($\pm 45°$ directions) yarn bundles. Therefore, after the axial yarn bundles of the outer layer are destroyed, the yarn bundles of the sublayer in the $\pm 45°$ directions redistribute the dissemination of loading force and create the defence effect. Accordingly, two fracture peaks clearly appear. We could infer that the 5D

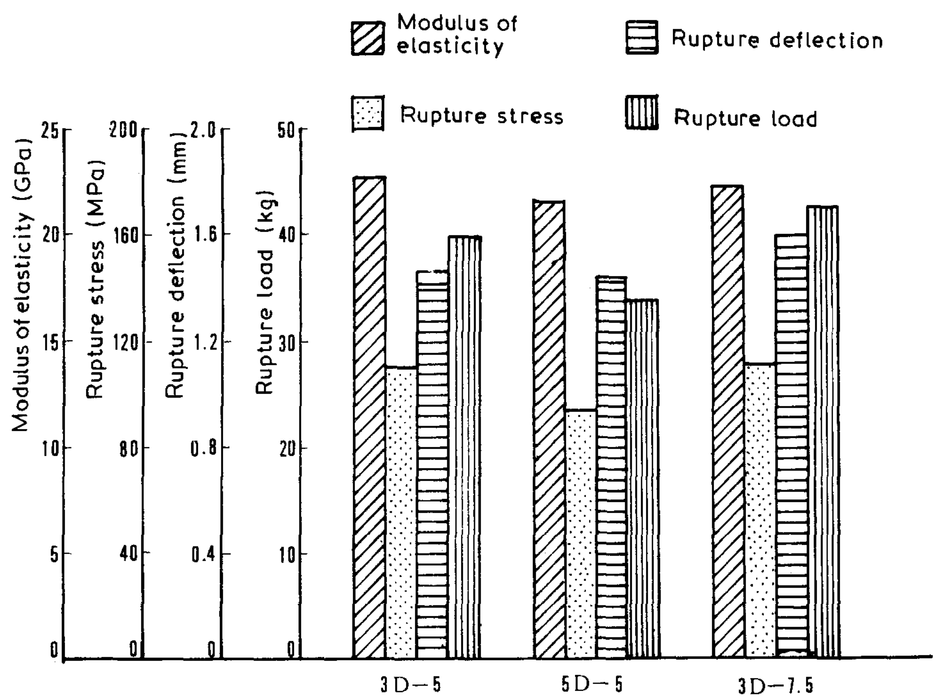

Figure 7 Flexural properties of three-dimensional carbon–carbon composites with various woven structures.

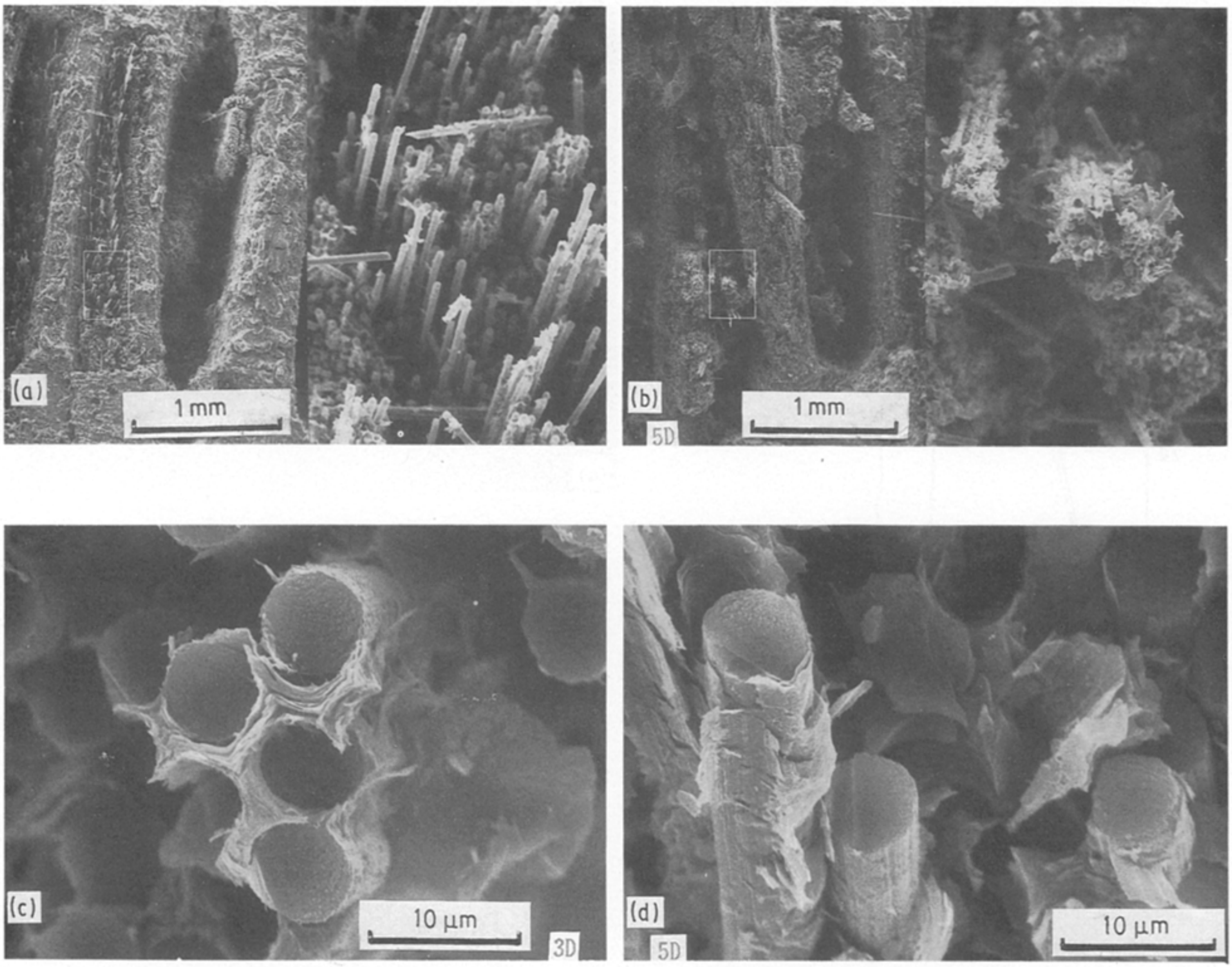

Figure 8 Scanning electron micrographs of three-dimensional carbon–carbon composites for 3D and 5D structures. (a, b) Dual images of flexural rupture; (c, d) microstructure of the flexural rupture section.

structure possessed a larger toughness effect than the 3D structure.

3.2. Short-beam shear strength

In various woven structures, the short-beam shear strengths of the 3D structure were greater than the 5D structure, as shown in Fig. 10, for the same reasons as in the flexural test, because the 5D structure has components in $\pm 45°$ directions which partially share the load. The yarn-bundle spacing experiments showed that the shear strength of the 5 mm type is higher than that of the 7.5 mm type due to the reinforcement numbers of the Z-axis increasing with increasing weaving density, which can help to arrest delamination propagation (Table II).

Another reason is that the span length of the shear test specimens is only one-quarter (25.6 mm) the length of the flexural testing specimens. Therefore, it is unsuitable for the derivation of flexural moment theory. Moreover, the connective distance of each axial yarn of the 5 mm type is shorter than that of the 7.5 mm type, so a connective effect of whole structure can develop. In addition, increasing the weaving density could increase fibre contents of the X–Y plane.

3.3. Analysis of fracture surface and microstructure

Analysis of the fracture surface by SEM is shown in Fig. 8. We observe that the fracture surface of the 3D structure is more orderly than that of the 5D structure. Subsequently, according to the high magnification microstructure, the adhesive strength between fibre and matrix of the 5D structure in $\pm 45°$ directions is smaller than the loading force. Therefore, the fracture behaviour of the 5D structure in $\pm 45°$ directions shows the debonding phenomenon. We can confirm that the previous flexural and shear strengths of the 3D structure are both larger than for the 5D structure. Thus the shear fracture tests show the stepwise fracture of the 5D structure and vertical fracture of the 3D structure, as shown in Fig. 11.

4. Conclusions

Both 3D and 5D preforms were woven by the arrangement of fibres in three orthogonal directions, and with the addition of two more $\pm 45°$ directions fibres, respectively. The fracture behaviour of three-dimen-

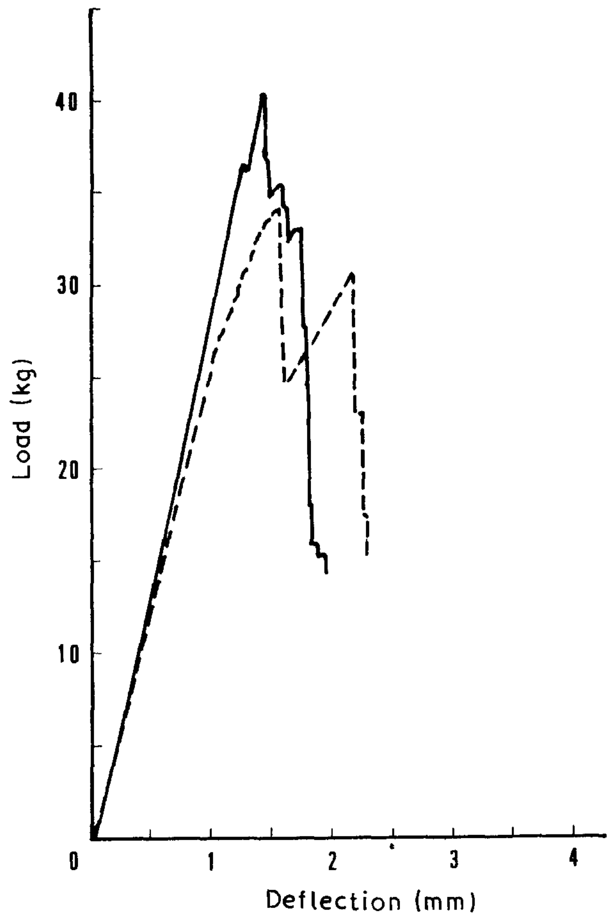

Figure 9 Flexural load–deflection curve of three-dimensional carbon–carbon composites for (——) 3D and (– – –) 5D structures.

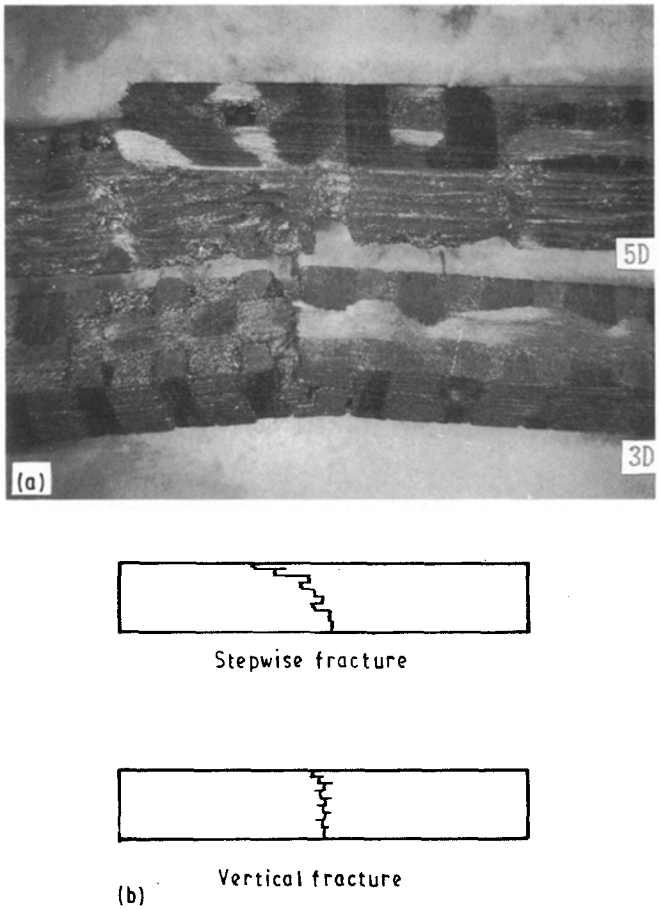

Figure 11 Photograph of shear fracture surfaces of 5D and 3D structures (×3.25).

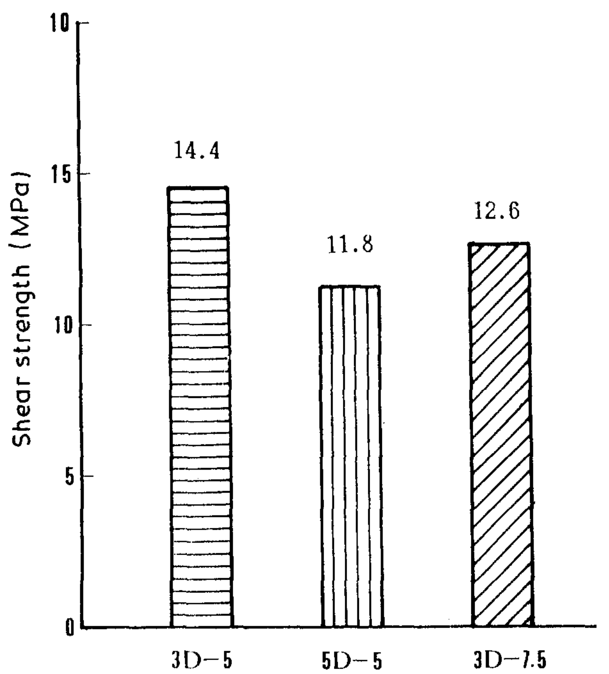

Figure 10 The short-beam shear strength of three-dimensional carbon–carbon composites with various woven structures.

sional carbon–carbon composites, based on the various woven structures and yarn-bundle spacing, was investigated. According to the flexural test, short-beam shear test, and analysing the fracture surface by SEM, we found that the strengths of the 3D structure were larger than those of the 5D structure. The latter absorbed energy before failure occurred to a greater extent than the former. The 5D structure had a higher fracture toughness.

The effect of yarn-bundle spacing on the flexural strength of the 7.5 mm type was higher than that of the 5 mm type while the short-beam shear strength was contrary. SEM analysis of the fracture surfaces which were ruptured along the *Z*-axis showed that a stress concentration existing in the fringe of the *Z*-axis and *Z*-directional fibres arrested the delamination propagation.

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