Environmental effects on shear delamination of fabric-reinforced epoxy composites

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Composite materials are prone to delamination failure due to their laminated structure. Hence, this failure mode has been the subject of numerous investigations. In the present study the residual Mode II delamination fracture toughness was characterized following exposure to low cycle fatigue loading and moisture at 50°C. The composites investigated included balanced fabric-epoxy systems composed of glass, aramid and carbon fibres. Experimental results have shown that the matrix and fibre/matrix interface dominated behaviour in shear loading and was insensitive to short-term exposure to both fatigue and moisture. Moreover, the carbon/epoxy material exhibited enhanced resistance to Mode II delamination after short exposures due to relaxation of the processing induced thermal stresses. However, long exposure caused damage to both the matrix and the fibre matrix interface, manifested in reduction of the strain energy release rates at failure. Fractographs supported the experimentally determined results. While unconditioned failure surfaces were typified by shear hackles, fatigued specimens indicated gradual elimination of the hackles, fibre fracture and matrix abrasion. When the composites were moisturized, examination of the fracture surfaces revealed plasticization of the matrix following short-term conditioning with gradual disappearance of the typical shear striations and appearance of voids, following long-term exposure.

Key words: shear delamination; environmental effects; failure mode; fractography; fabric-reinforced epoxy composites

Interlaminar failure or delamination is a characteristic failure mode of composite materials due to their laminated structure. Consequently, this type of failure mechanism has been the subject of numerous studies.

Laminate failure may originate from a number of sources. These are the presence of interlaminar cracks and debonds in areas of stress concentrations due to discontinuities in the laminate structure, internal stresses as a result of differential thermal expansion coefficients of the composite constituents, or applied external stresses[1-3]. One of the ways to characterize the resistance of interlaminar crack propagation is by means of the classical linear elastic fracture mechanics (LEFM) which defines the critical strain energy release rate, G_c. Under controlled loading conditions delamination could be categorized in view of the loading modes. In Mode I, failure occurs by stress applied at right-angles to the laminate plane resulting in opening up of the laminate. Mode II takes place under in plane forward shear loading, and Mode III is in plane tearing mode. The first two loading modes are the most common ones. Mode I is usually characterized by the double cantilever beam type specimen, Mode II by the end notch flexure[4, 5] type specimen. Other types of specimens are employed for investigating the delamination fracture under combined Mode I and Mode II loading.

A large number of investigations have been dedicated to the study of the various factors affecting Mode I and Mode II delamination fracture mechanics. Among them, the specific fibre-matrix combination, the interfacial interactions between the fibre and the matrix, the fibre orientation and the type and rate of loading[6-8] are considered. Special emphasis has been given to the effects of humidity, temperature and fatigue conditioning prior to loading[6, 9-14].

Most the above mentioned studies have been performed using unidirectionally reinforced

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composites. The investigations related to woven fabric composites are sparse, even though fabric-reinforced composites are commonly used in high performance applications. In a previous study the authors[15] investigated the strain energy release rates under Mode I and Mode II loadings of glass, aramid and graphite fabric epoxy composites. Experimental results supported by microscopy have indicated that the Mode I delamination is governed by the fibre matrix interfacial strength while Mode II delamination is determined by the matrix shear strain, which is highly dependent on the delamination angle with respect to the fibres' direction. The latter study has shown that the highest resistance to delamination, in the case of fabric composites, is obtained when the axes of the fabric form an angle of 45° with respect to the delamination direction. Consequently, the present study concentrates on Mode II loading at 45° of similar fabric-reinforced composites, subjected to conditions of cyclic fatigue loading and moisture, prior to delamination under Mode II loading.

Experimental

Composite materials

Three fabric reinforced epoxy composites were included in the shear delamination study

- E-glass plain woven fabric (Style 7500 by Clark Schwebel) with Volan A finish. The glass fabric was impregnated with a blend of DGEBA epoxy resin and polyamide hardener (Epon 815 and Versamid 140 by Miller Stephenson) having a ratio of 70 to 30, respectively and 24 layers of the impregnated fabric were placed in a metal mould and cured at 100°C and 100 psi for 4 hours. The resulting laminate was 6 mm thick and was cut at 45° into 10 mm wide strips. The resin volume fraction of the glass-epoxy composite was determined to be 51%.
- Commercial graphite fabric-epoxy prepreg based on T-300 fibres, plain woven (W3T 282 weave style and F155 epoxy by Hexcel). Twenty-eight layers of the prepreg were cured in a metal mould at 121°C (250°F) and 100 psi for 2 hours. The cured laminate was 6 mm thick and was cut at 45° into 10 mm wide specimens. The resin volume fraction was found to be 53%.
- Commercial aramid fabric-epoxy prepreg made of crow foot fabric (Style K 285 and F 155 resin by Hexcel). Fourteen layers of the prepreg were compression cured in a metal mould at 121°C (250°F) and 100 psi for 2 hours. The resulting laminate was 3 mm thick, and was cut at 45° into 10 mm wide samples. The resin content was 43%.

Test specimens

Mode II fracture toughness was studied using the end notch flexure specimen (Fig. 1). The initial crack length a_0 was varied between 3 and 6 mm, the loading span was between 15 and 30 mm and the thickness was between 3 and 6 mm. The lower values were used for the aramid composites and the higher ones for the glass and carbon composites. In all cases the specimen width was fixed at 10 mm. The ratio ℓ/t was chosen to ensure shear failure and to avoid tensile or compressive failure. The ratio $a_0(\ell/2)$ was between 0.4 and 0.6.

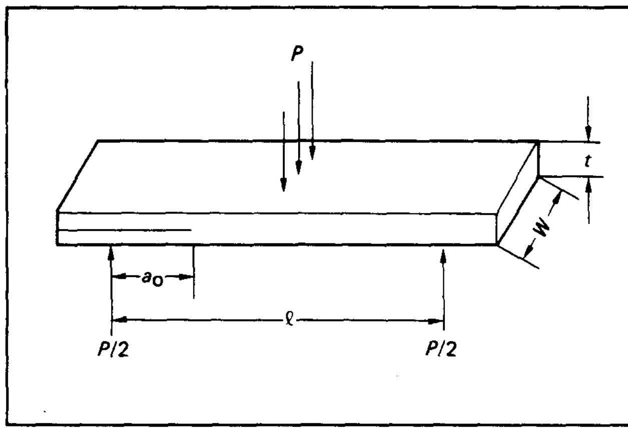

Fig. 1 End notch flexure specimen for Mode II delamination loading

Following exposure to different environmental conditions the specimens were loaded in flexure using an Instron mechanical tester at a crosshead speed of 1 cm min^{-1}. Two types of fatigue loading were used. In propagation was followed by a Vernier microscope and an acoustic emission probe at 375 Hz and amplification of 95 dB.

Environmental conditions

Prior to Mode II loading the pre-cracked specimens were exposed to fatigue loading or immersed in water at 50°C. Fatigue loading was carried out using the three point bending fixture (Fig. 1) at a rate of 1 cm min^{-1}. Two types of fatigue loading were used. In the first mode each specimen was subjected to 100 cycles at a constant stress amplitude, each one being a different fraction of the ultimate delamination strength. In the second loading mode each specimen was fatigued under a stress amplitude of approximately 50% of the ultimate shear delamination strength and the number of loading cycles was varied. In the cases where the effect of moisture pick-up was investigated, notched specimens, immersed in water at 50°C, were taken out of the immersion bath after a predetermined duration. The moisture level in the specimens was calculated prior to the fracture toughness testing.

Fractography

Shear delamination loading to failure was complemented with fractographic examination to analyse the surface morphology. A scanning electron microscope (SEM) was used for this purpose.

Calculations of fracture toughness

Strain energy release rates in shear Mode II loading, G_{IIc}, were calculated using the compliance method:

$$G_{IIc} = \frac{P_c^2}{2W} \frac{dc}{da} \tag{1}$$

where P_c is the critical load at the start of crack propagation and the compliance, C, is defined for the case of three point bending by

$$C = \frac{\ell^3 + 12a^3}{4E\,Wt^3} \tag{2}$$

where E is the composite Young's modules.

Differentiation of Equation (2) with respect to a and substitution into Equation (1) yields

$$G_{IIc} = \frac{18\, a^2 P_c^2 C_b}{W(\ell^3 + 12a^3)} \qquad (3)$$

Where C_b is the compliance in flexure defined by

$$C_b = \delta / P \qquad (4)$$

where δ is the measured deflection and P the respective load.

Results and discussion

The resistance of glass, graphite and aramid fabric-epoxy composites to Mode II delamination at 45° was characterized by the strain energy release rate, G_{IIc}, following fatigue or wet conditioning.

Fig. 2 presents a typical load–deflection curve of a glass-epoxy specimen and its respective acoustic emission response. As can be seen, the acoustic measurements indicate an increase in voltage at 130 kg while the visual inspection with the microscope points out that crack propagation takes place at 135 kg.

Effect of fatigue on shear delamination

Generally it has been shown[16] that cyclic loading is effective in causing microdelamination in front of the delamination notch, and an increase in the size of the damage zone, as well as abrasion of the matrix and fibre breakage in the delamination plane. Figs 3 and 4 summarize the results for testing with a constant number of cycles and a constant amplitude, respectively. As could be distinguished, preconditioning of the pre-notched specimens to fatigue loading resulted in deterioration of fracture energy of delamination. The three material systems under investigation behaved differently. The glass-epoxy composite had the lowest resistance to delamination compared with the aramid and carbon composites. It should be emphasized that the resin system used in combination with the glass fabric was a common DGEBA and a polyamide curing agent, while the commercial prepregs used were based on a toughened epoxy resin (F155). Furthermore, the resin content of the carbon-epoxy material was higher than the aramid-epoxy composite, 53% compared to 43%, respectively. The difference in matrix toughness and its volume fraction influences the initial resistance to shear delamination as this characteristic property is highly dependent on the interlaminar matrix[15]. However, the deterioration of the performance with pre-fatigue

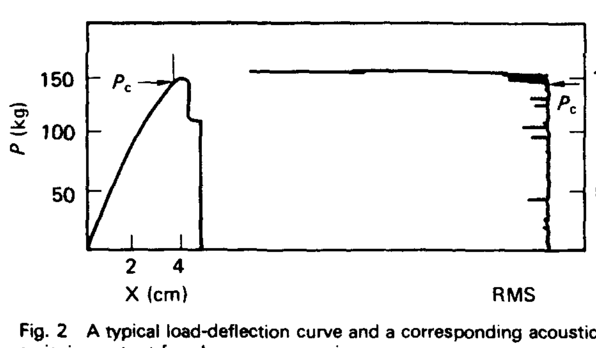

Fig. 2 A typical load-deflection curve and a corresponding acoustic emission output for glass-epoxy specimen

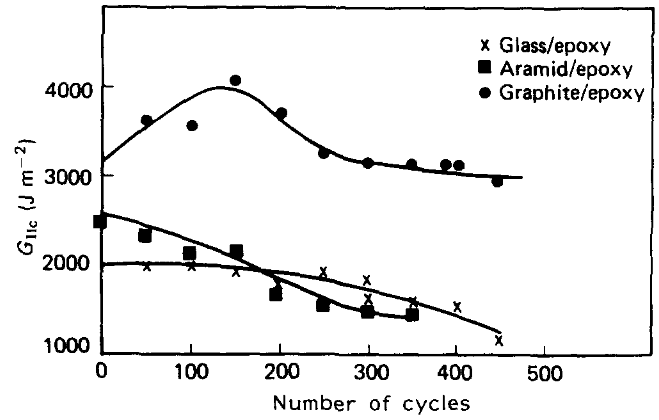

Fig. 3 Strain energy release rates following constant stress amplitude and varying number of loading cycles for glass, aramid and graphite epoxy composites

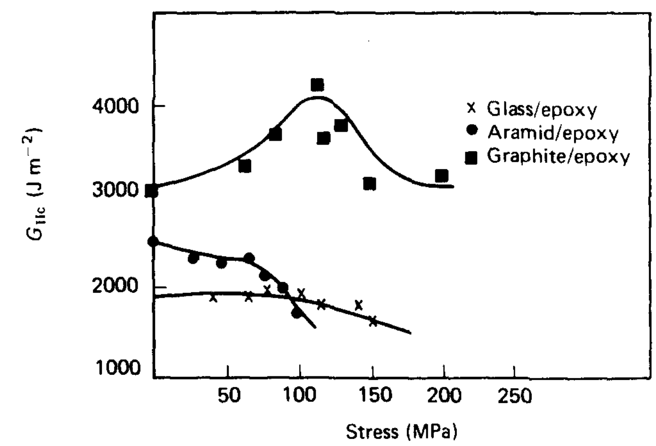

Fig. 4 Strain energy release rates following constant number of loading cycles and varying stress amplitudes for glass, aramid and graphite epoxy composites

loading could be investigated, taking into account the experimental details.

The glass-epoxy composite showed a similar behaviour when loaded either at constant stress amplitude or at constant number of loading cycles. In both cases the strain energy release rates declined monotonously with increasing loading cycles or stress amplitudes, indicating that a similar damage mechanism takes place upon cyclic stressing. A closer examination of Figs 3 and 4 indicates that in the case of glass-epoxy composites up to a threshold level of damage no noticeable reduction in fracture toughness is discernible. The same phenomenon was reported by Laksimi and Bathias[17]. They have found that a minimum loading level exists, below which delamination is inhibited.

The aramid-epoxy material, while exhibiting a similar trend, shows a higher rate of toughness deterioration with pre-fatigue loading. As evident from Figs 3 and 4, a large drop in fracture toughness takes place upon cyclic stressing to the point of more than 50% drop compared to the initial toughness values.

The carbon-epoxy composite exhibited a completely different behaviour compared to the glass and aramid composites. As shown in Figs 3 and 4, the Mode II strain energy release rate increases at the initial stages of fatigue stressing followed by a sharp decline and levelling off at the highest fatigue stresses or number of loading cycles investigated in the present study. A similar behaviour has been reported elsewhere[14, 18, 19].

The initial increase in delamination resistance has been attributed to release of internal stresses which are caused by the differences of the thermal expansion coefficients between the matrix (positive) and the graphite fibres (slightly negative). However, the fracture toughness enhancement is only temporary and, due to interfacial damage, the resistance to Mode II delamination deteriorates upon continuation of fatigue loading. This phenomenon is observed with the carbon-epoxy composite and not with the aramid-epoxy composite despite the much larger negative thermal expansivity of the aramid fibre, because of the weaker interfacial bond and lower restraint in the latter system.

Effect of moisture on shear delamination

To investigate the effect of moisture content on Mode II delamination resistance the pre-notched 45° specimens were immersed in water at 50°C for periods up to 8 weeks. Table 1 summarizes the moisture levels as functions of immersion time. The moisture contents reported are calculated assuming that the fibres are inaccessible to moisture penetration, and that moisture is absorbed in the matrix alone. For the case of the aramid composites, it has been assumed that the moisture level in the matrix is identical to the case of the carbon composites as their matrices are identical (F155).

Moisture effects on composites have been shown to be dependent on the exposure duration. In the initial stages of exposure plasticization of the matrix is dominant resulting in a drop of the glass transition temperature. Upon removal of the water after short exposures the properties are regained and the whole process is reversible. However, long durations of exposure to humidity caused irreversible effects due to deterioration of the constituent materials, especially of the fibre-matrix interface, to the point of fibre debonding[20, 21]. Figure 5 depicts the relationship between the strain energy release rates and the moisture level in the epoxy matrix for the three composites studied.

As in the case of pre-fatigue loading, the glass-epoxy composite exhibits moderate insensitivity to moisture

Table 1. Matrix moisture content following immersion in water

Immersion time (weeks)	Moisture content of the matrix (wt%)	
	Glass-epoxy	Carbon or aramid-epoxy
0	0	0
1	1.07	1.39
2	1.39	1.95
3	1.77	2.28
4	2.02	3.01
5	2.22	3.95
6	2.45	4.01
7	2.45	4.07
8	-	4.57

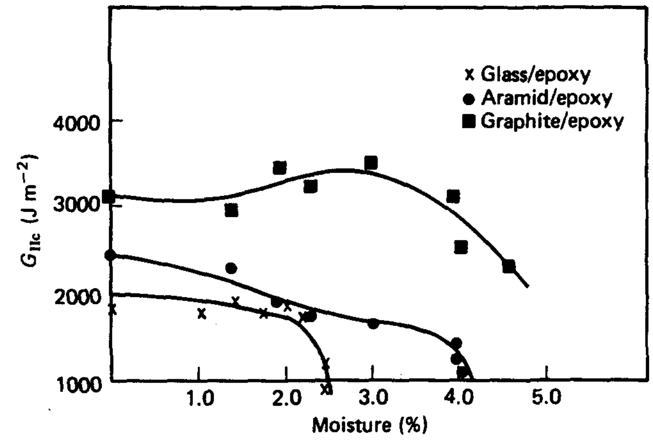

Fig. 5 Strain energy release rates following moisture absorption for glass, aramid and graphite epoxy composites

up to 2% (four weeks) followed by a very large drop. This behaviour conforms to the common observation of initially reversible hygroelastic effects of moisture, followed by permanent damage caused to the fibre-matrix interface.

The same general behaviour could be observed for the aramid-epoxy composite. In this case the shear delamination resistance gradually decreases. Following a critical immersion time of 6 weeks (equivalent to 4% moisture pick-up) gross damage is evident which may be due to a combined moisture absorption effect of the matrix and the fibre, which may lead to fibre debonding.

The carbon-epoxy composites responded differently to water absorption, as was the case for the low cycle fatigue loading. As shown in Fig. 5, the resistance to delamination increases up to a moisture level of 4% followed by a drop after exposure for 7 and 8 weeks. Once again, the differences in thermal expansion coefficients between the graphite fibres and the epoxy matrix coupled with strong interfacial bonding result in an initial stress relaxation (of the residual thermal stresses) followed by long-term deterioration of the fibre-matrix interface.

Again the absence of an initial increase in toughness of the aramid composites, as found with the carbon-

Fig. 6 Fracture surface of unexposed glass/epoxy specimen (× 300) Delamination is in arrow direction ↑

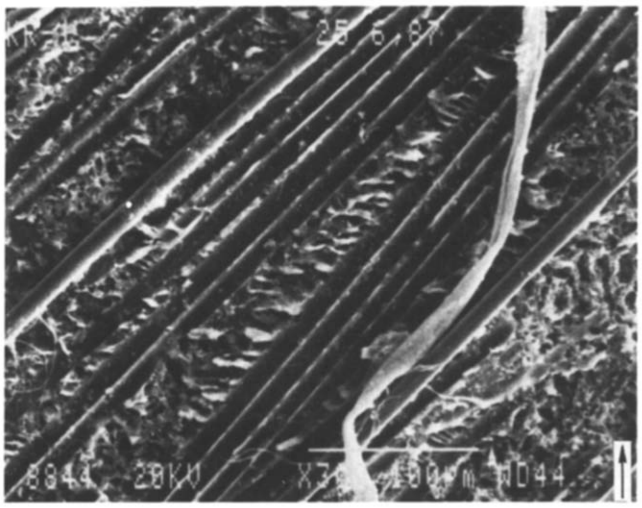

Fig. 7 Fracture surface of unexposed aramid/epoxy specimen (x 300) Delamination is in arrow direction ↑

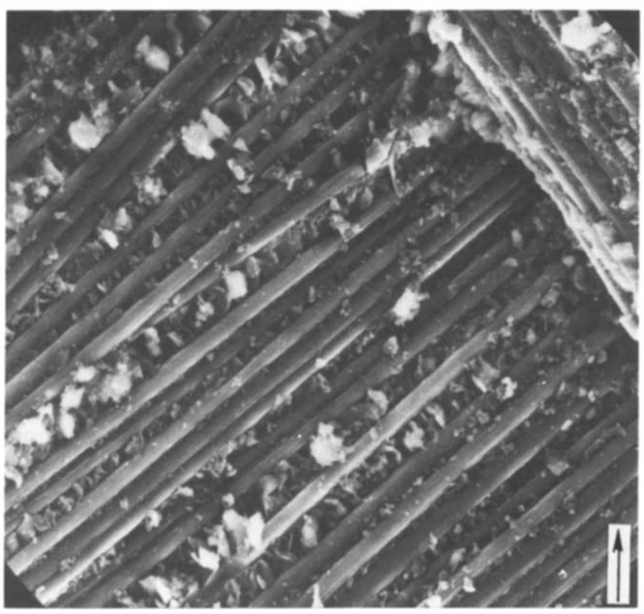

Fig. 8 Fracture surface of unexposed carbon/epoxy specimen (x 300) Delamination is in arrow direction ↑

epoxy material, even though the aramid fibre is characterized by a relatively large negative coefficient of thermal expansion, is attributed to poor fibre-matrix bonding which is dominant.

Fractography

The experimentally determined G_{IIc} values following low cycle fatigue loading or moisturization of the glass, aramid and graphite composites provides a quantitative measure of resistance to shear delamination. A fractographic study of the fracture surfaces complements the experimental investigation with respect to failure mechanisms and their correlation with the G_{IIc} values. Figs. 6, 7 and 8 depict typical shear delamination failure morphologies of unexposed glass, aramid and carbon specimens, respectively. In all cases the shear hackles of the matrix typical of Mode II loading in 45° aligned fabric-reinforced specimens are evident[15].

SEM micrographs of pre-fatigued samples of glass, aramid and carbon composites are presented in Figs 9, 10 and 11 respectively. For each material, fractographs are presented for treatments below and above the threshold values of G_{IIc}, indicating a significant deterioration in delamination resistance. As shown in Fig. 9(a) for the glass-epoxy case loading to 75 MPa for 100 cycles resulted in slight damage, some broken fibres being observed while the basic shear striations remain unchanged. Fig. 9(b) indicates that following 400 cycles at 80 MPa the number of broken fibres increased. The resin shear bands have largely disappeared due to the abrasive action of cyclic loading at the crack zone. The fractographs given in Figs. 9(a) and 9(b) correspond to the relatively high and low G_{IIc} values, respectively.

In the case of aramid-epoxy composites, damage is observed even at a lower level of fatigue loading. Fig. 10(a) shows that at 100 44 MPa cycles the matrix is already abraded and some of the aramid fibres fibrillate in a typical splitting failure mode.

Fig. 10(b) demonstrates that massive damage has taken place following 350 cycles at 75 MPa, with a large number of broken fibres and almost complete

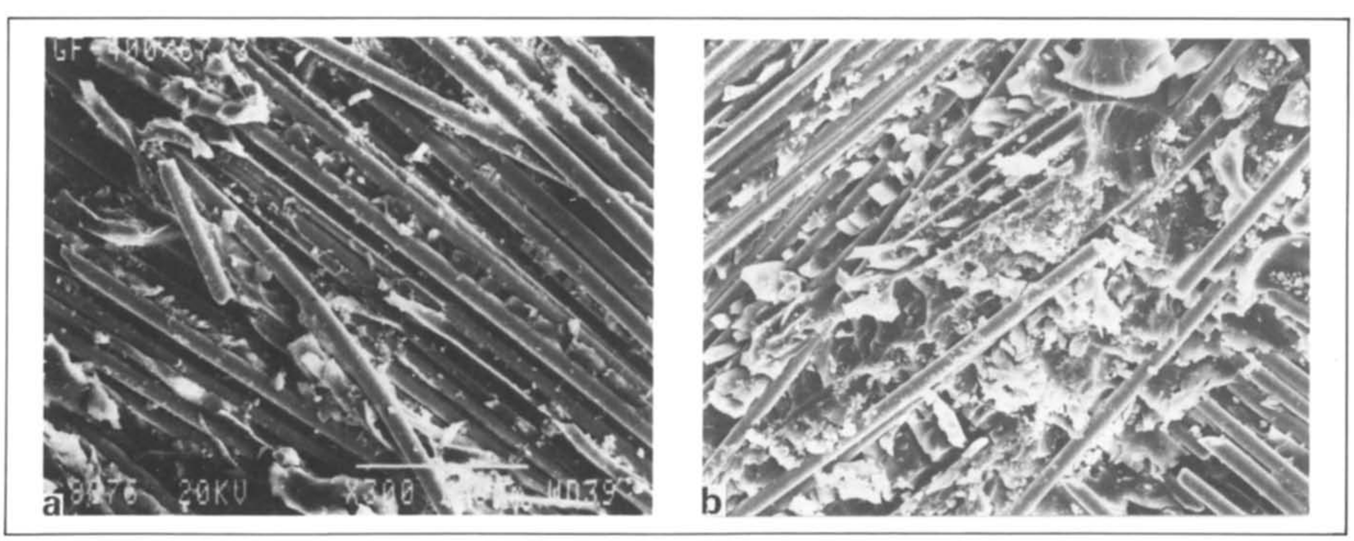

Fig. 9 Fractographs of delaminated glass/epoxy specimens (x 300): (a) after 100 cycles at 75 MPa; (b) after 400 cycles at 80 MPa

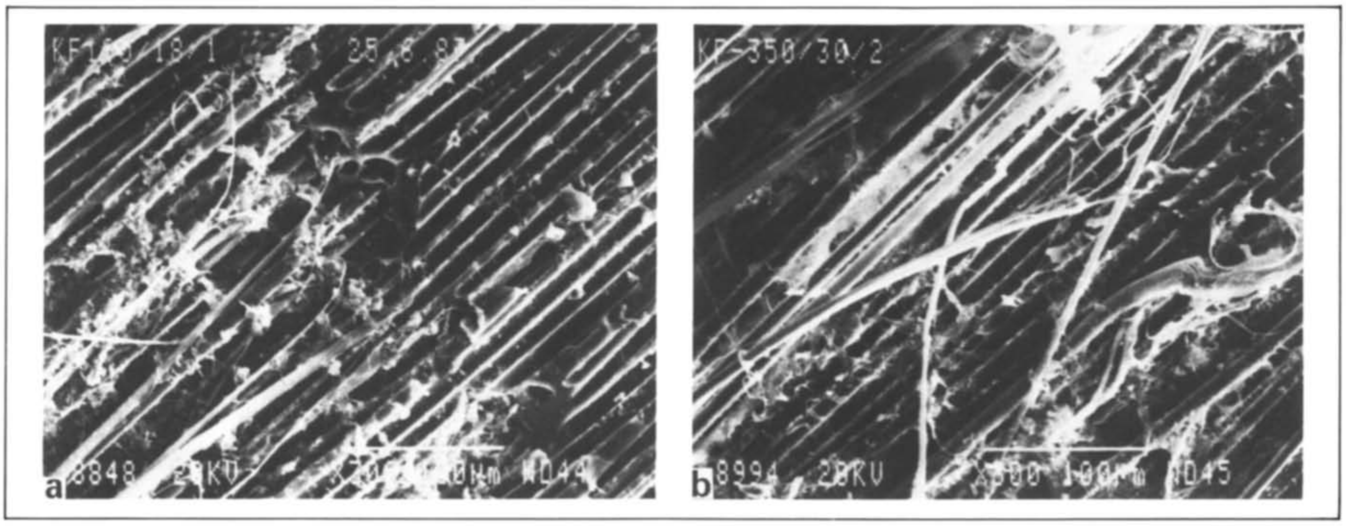

Fig. 10 Fractographs of delaminated aramid-epoxy specimens (x 300): (a) after 100 cycles at 44 MPa; (b) after 350 cycles at 74 MPa

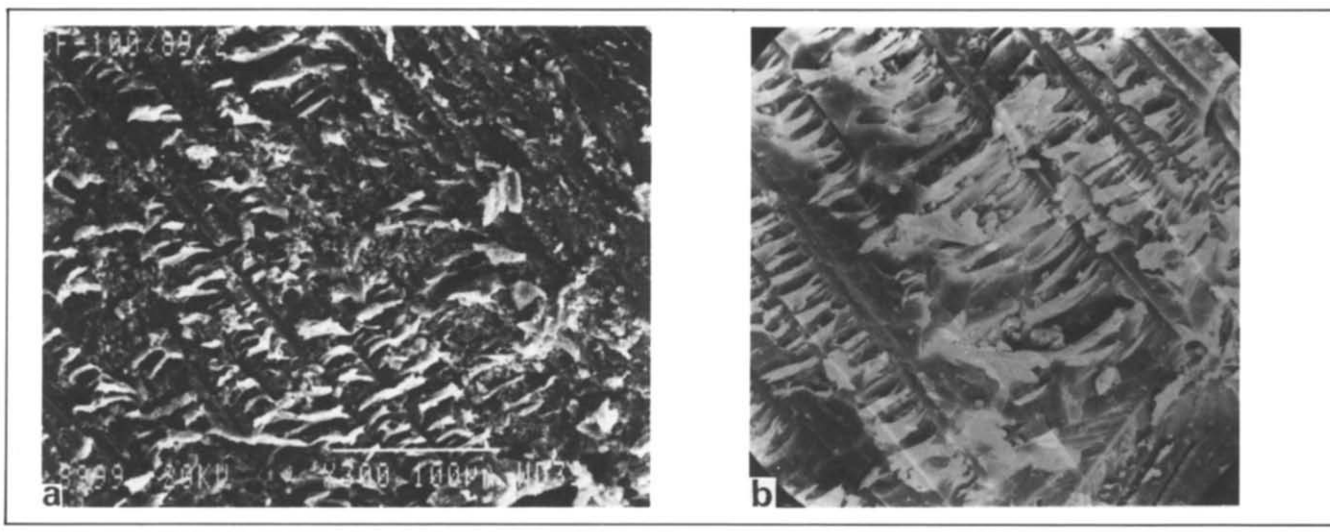

Fig. 11 Fractographs of delaminated carbon-epoxy specimens (x 300): (a) after 100 cycles at 110 MPa; (b) after 450 cycles at 114 MPa

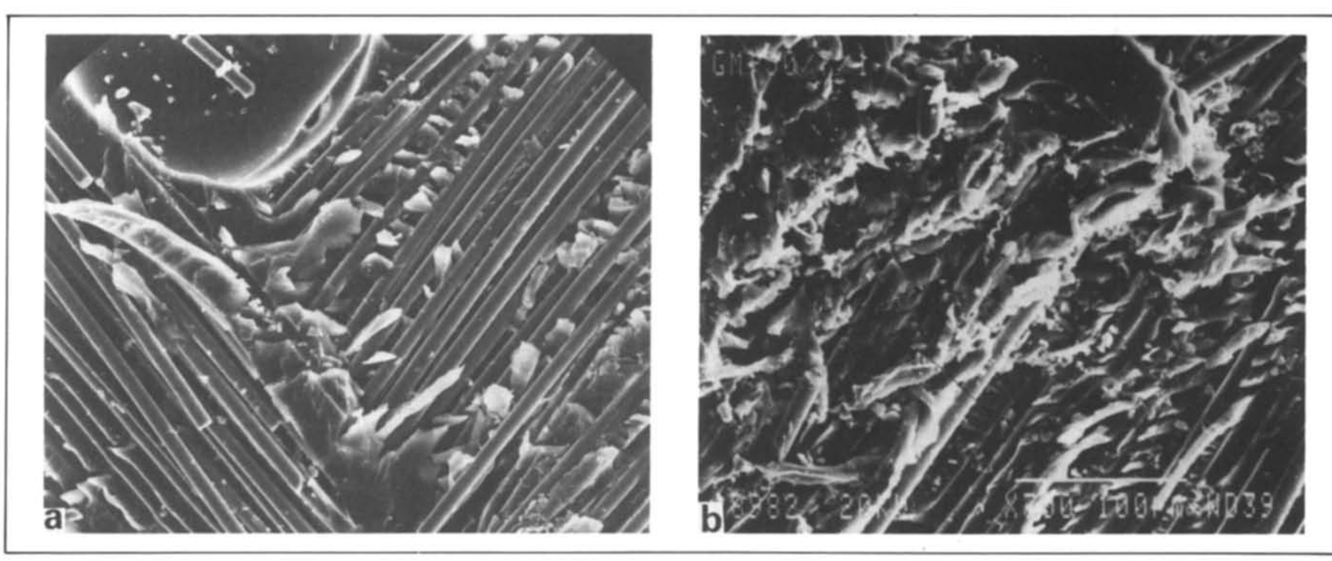

Fig. 12 Fracture surfaces of delaminated glass-epoxy specimens (x 300): (a) after exposure to water at 50°C for 4 weeks; (b) after exposure to water at 50°C for 7 weeks

disappearance of the typical shear hackles. This failure topography corresponds to the low G_{IIc} value measured after the higher pre-fatigue loading.

A completely different morphological effect has been observed in the case of carbon-epoxy composites. Figs. 11(a) and 11(b) conform to the G_{IIc} values determined. The basic shear hackles are unchanged even after the highest level of fatigue loading. The shear bands are

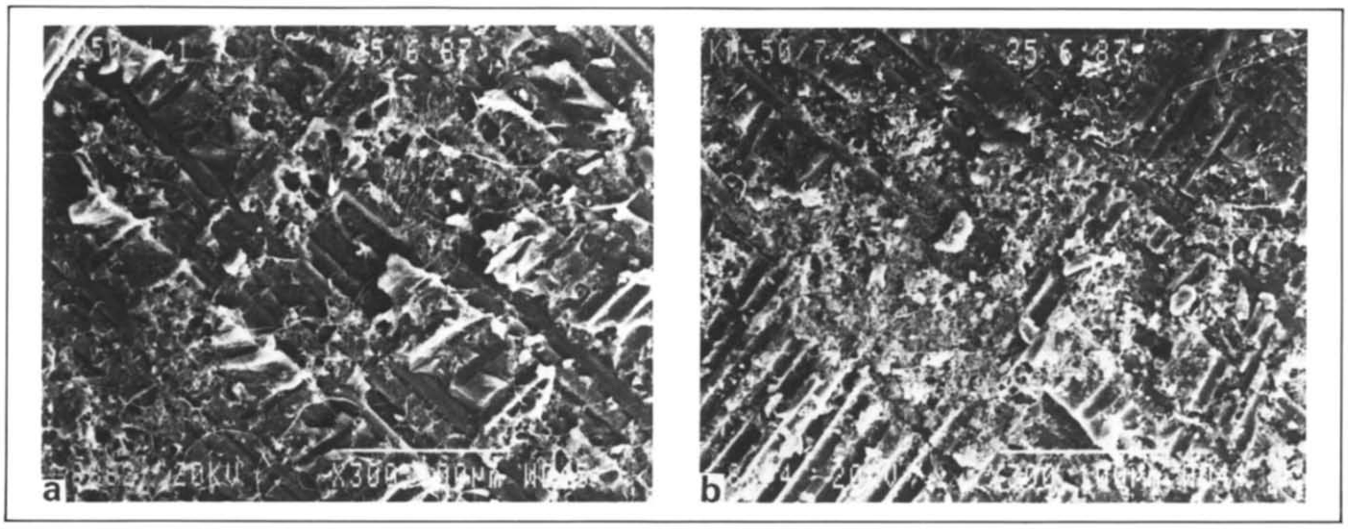

Fig. 13 Fracture surfaces of delaminated aramid-epoxy specimens (× 300): (a) after exposure to water at 50°C for four weeks; (b) after exposure to water to 50°C for 7 weeks

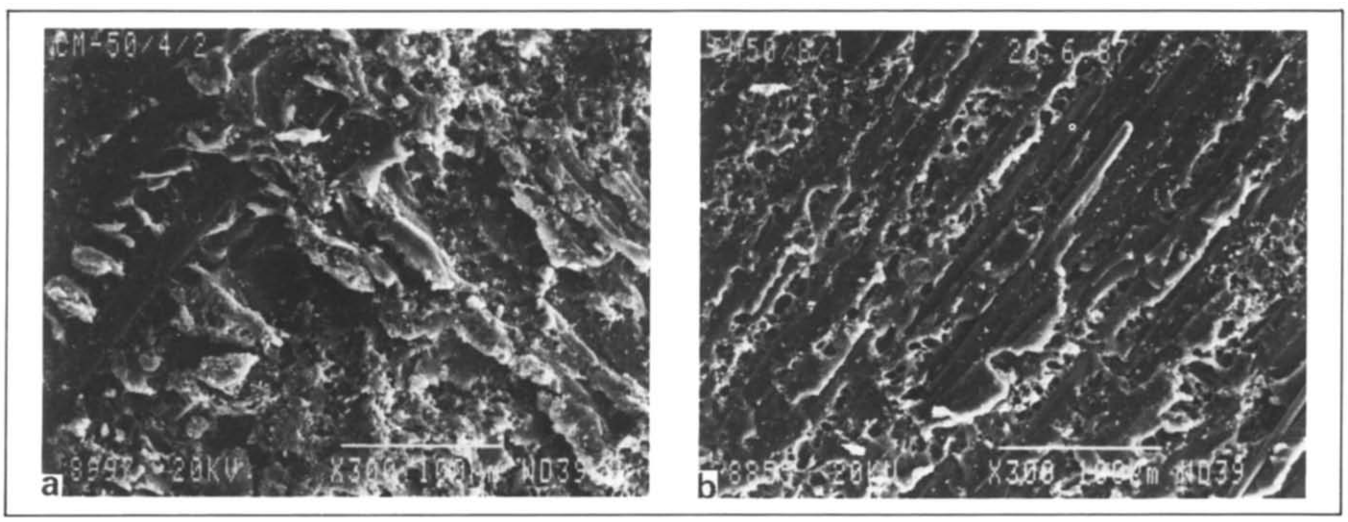

Fig. 14 Fracture surface of delaminated carbon-epoxy specimens (× 300): (a) after exposure to water at 50°C for 4 weeks (b) after exposure to water at 50°C for 8 weeks

extended upon increased loading, whilst the only damage is some broken fibres.

As mentioned earlier, the moisture effect on shear delamination has two different consequences. Short-term exposure to water results in plasticization of the epoxy matrix, while long-term conditioning results in a sharp decrease in the Mode II toughness.

Fig. 12(a) represents the delamination failure surfaces of glass-epoxy composites following exposure to water for 4 weeks. As can be seen, the shear hackles are enlarged and the failure is ductile compared to the unexposed case (Fig. 6). The fracture topography has a completely different appearance following exposure for 7 weeks. As seen in Fig. 12(b), the shear striations are even larger, due to the softening of the matrix[20].

The effects of moisture on the failure mechanism of aramid-epoxy composite are described by Figs. 13(a) and 13(b) for 4 and 7 weeks immersion in water, respectively. The fracture is ductile after 4 weeks due to matrix plasticization, which leads to complete softening of the matrix after 7 weeks with additional appearance of voids indicating massive damage due to leaching of the matrix.

The same general picture of fracture surfaces is obtained in the case of the carbon-epoxy material (Fig. 14). Following 4 weeks immersion the matrix appears to soften by the water-matrix interaction. Voidiness appearance, general matrix plasticization and disappearance of shear bands are the result of exposure to water for 8 weeks, corresponding to permanent damage. Garg[21] studied the effects of temperature and humidity on the interlaminar crack propagation of unidirectional graphite-epoxy material. He reported that the humidity plasticized the matrix, resulting in an increase in the damage zone at the crack tip.

Conclusions

In the present study, shear delamination resistance of glass, aramid and carbon fabric-epoxy composites was characterized with respect to conditioning in low cycle fatigue loading or moisture prior to Mode II loading to failure. Experimental results, substantiated with fractographs of the failure surfaces indicated that fatigue pre-loading of the notched specimens causes microdelamination and matrix abrasion in front of the delamination crack. When fatigue conditioning is limited in amplitude and number of loading cycles, the reduction in shear toughness is minimal in glass-epoxy and aramid-epoxy, while the carbon-epoxy exhibits an enhancement in delamination resistance. However, following relatively prolonged fatigue loading, the damage caused to the shear hackles is massive, accompanied by fibre breakage and manifested in low shear toughness.

Strain energy release rates, G_{IIc}, varied with moisture level absorbed by the matrix. In the initial stages of exposure to water, the matrix is plasticized and the measured delamination energies are insensitive to absorbed moisture. The carbon-epoxy material strain energy release rate increased initially due to release of thermal stresses. Upon extension of immersion times the shear toughness decreased sharply due to damage caused to the fibre-matrix interface, appearance of voids in the matrix and complete softening of the epoxy.

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