

Mechanism of radiation-induced degradation in mechanical properties of polymer matrix composites

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Four kinds of polymer matrix composites (filler, E-glass or carbon fibre cloth; matrix, epoxy or polyimide resin) and pure epoxy and polyimide resins were irradiated with ^{60}Co γ -rays or 2 MeV electrons at room temperature. Mechanical tests were then carried out at 77 K and at room temperature. Following irradiation, the Young's (tensile) modulus of these composites and pure resins remains practically unchanged even at 170 MGy for both test temperatures. The ultimate strength, however, decreases appreciably with increasing dose. The dose dependence of the composite strength depends not only on the combination of fibre and matrix in the composite but also on the test temperature. A relationship is found between the composite ultimate strain and the matrix ultimate strain, thus indicating that the dose dependence of the composite strength is virtually determined by a change in the matrix ultimate strain due to irradiation. Based on this finding, we propose a mechanism of radiation-induced degradation of a polymer matrix composite in order to explain the dose dependence of the composite strength measured at 77 K and at room temperature.

1. Introduction

Polymer matrix composites are leading candidates for mechanical supports and electrical insulators in the construction of superconducting magnets for fusion reactors [1, 2]. This is because organic materials such as polymer matrix composites are superior to inorganic materials in terms of cost and processing [3]. Polymer matrix composites are also suitable materials for space vehicles, because they are light and strong. If polymer matrix composites are actually employed for these purposes, however, they will be subjected to substantial quantities of high-energy radiation over several years or decades, thus leading to significant degradation in the mechanical and electrical properties. For this reason, several studies have recently been performed on the irradiation effects in polymer matrix composites from the standpoint of their applications to fusion magnets [4-12] and to space vehicles [13, 14].

All of these studies are, indeed, useful sources of design data for fusion magnets and space vehicles. As pointed out by Brown [1], however, the goal of these studies should be to interpret the experimental data in terms of the constitutive properties of a composite, i.e. to try to understand the irradiation effects at a fundamental level. Not only will this permit better predictability and extrapolation of data, but it could also lead to procedures for increasing the radiation resistance of a composite. This goal has not yet been achieved, because mechanisms of radiation-induced degradation of polymer matrix composites are still not completely elucidated. Such a study has just been started by Klabunde and Coltman [6] using stereo-

microscopic techniques and by Egusa *et al.* [15-19] who have analysed the dose dependence of the mechanical properties of various composites.

In the present work, four kinds of polymer matrix composites and two kinds of pure resins were irradiated at room temperature with γ -rays from a ^{60}Co source or with 2 MeV electrons from an accelerator. Mechanical tests were then carried out at 77 K and at room temperature. This study revealed that the dose dependence of the mechanical properties of a polymer matrix composite depends not only on the combination of reinforcing filler and matrix resin in the composite but also on the temperature during the mechanical test. The present paper mainly describes the dose dependence of the mechanical properties of the composites and pure resins at 77 K and at room temperature. A mechanism of radiation-induced degradation of polymer matrix composites is proposed on the basis of a comparison of the dose dependence for the composites with that for the pure resins.

2. Experimental details

2.1. Materials

Four kinds of polymer matrix composites were prepared by Sumitomo Bakelite Co. Ltd, using E-glass or carbon-fibre cloth as reinforcing filler, and epoxy or polyimide as matrix resin. The epoxy resin used in these composites is Sumiepoxy ELM-434 (tetraglycidyl diaminodiphenyl methane, TGDDM) cured with diamino diphenyl sulphone (DDS), and the polyimide resin is polyamino**bis**-maleimide (Kerimid 601).

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TABLE I Specimens of polymer matrix composites and pure resins

Specimen	Volume fraction of fibres (%)	Average thickness (mm)
Glass/epoxy composite	63	1.81 ± 0.07
Glass/polyimide composite	62	2.05 ± 0.06
Carbon/epoxy composite	57	2.28 ± 0.03
Carbon/polyimide composite	62	1.69 ± 0.07
Epoxy resin	-	2.19 ± 0.20
Polyimide resin	-	2.18 ± 0.11

The E-glass fibre cloth (Kanebo KS-1210) is a plain-woven fabric having 53 and 48 yarns per 25 mm in the warp and fill directions, respectively, and each yarn is composed of about 200 fibres having a diameter of 7.36 μm. The carbon fibre cloth (Torayca no. 6142) is also a plain-woven fabric having 22.5 yarns per 25 mm in both directions, and each yarn is composed of about 1000 fibres having a diameter of 8 μm. The volume fraction of fibres and the average thickness of these composite sheets are shown in Table I.

Pure epoxy resin sheets (TGDDM cured with DDS) were obtained from Sumitomo Bakelite Co. Ltd. Pure polyimide resin sheets (Kerimid 1000) were obtained from Nippon Polyimide Co. Ltd. The average thickness of these sheets is also shown in Table I.

Sheets, ~2 mm thick, of these composites and pure resins were cut into specimens of 6.4 × 70 × ~2 mm³ dimensions, retaining the factory finish on both surfaces of the sheets. For composite specimens, the cutting was done so that the 70 mm axis was parallel to the warp direction of the reinforcing fabrics. These specimens were the same type as those used in previous work [20–22].

2.2. Irradiation

⁶⁰Co γ-ray irradiation was carried out in air at room temperature with an exposure rate of 2.25 × 10⁶ röntgen per hour (r h⁻¹). The exposure rate was determined using a cobalt glass dosimeter [23]. The dose absorbed in a matrix of a composite specimen or in a pure resin specimen was calculated by using conversion factors of 0.009 33 and 0.009 22 Gy r⁻¹ for the epoxy and polyimide resins, respectively [19].

Electron irradiation of 2 MeV was carried out with a Cockcroft-Walton type accelerator (Nisshin High Voltage Co. Ltd). Each specimen was wrapped in aluminium foil, and was attached with a conducting adhesive to the irradiation table. The table was cooled by cold water during irradiation, so that the specimen temperature was kept below 53°C under the present irradiation conditions of 2.0 MV and 5 mA. The absorbed-dose rate was measured using a cellulose triacetate (CTA) film dosimeter (FTR-125) manufactured by Fuji Picture Film Co. Ltd [24]. The dose rate was 2.60 and 3.76 kGy sec⁻¹ on the front and back sides of a composite specimen, respectively. For this reason, we tentatively determined the dose rate to be 3.18 kGy sec⁻¹ from the mean value. Similarly, the dose rate for a specimen of pure epoxy and polyimide resins was determined to be 2.88 and 2.98 kGy sec⁻¹, respectively.

2.3. Mechanical tests

The mechanical properties were examined by performing three-point bend tests. The bend rig was especially constructed so that the tests could be made at span lengths of 60, 40, and 20 mm at liquid nitrogen temperature as well as at room temperature. The radius of the loading and support noses was 3 mm. This bend rig was used with a mechanical test machine of Shimadzu Servopulser Model EHF-UC5-10L. All measurements were conducted at a cross-head speed of 0.6 mm min⁻¹. The load was applied in the direction normal to the widest surfaces of a specimen having 6.4 × 70 × ~2 mm³ dimensions.

3. Results

3.1. Comparison between γ-ray and electron irradiations

In the present work, 2 MeV electrons were used in place of ⁶⁰Co γ-rays for some specimens, because the irradiation time for a desired dose can be shortened considerably by electron irradiation. For this reason, the irradiation effects of electrons were compared with those of γ-rays for the glass/epoxy composite. This comparison was made for the ultimate strength of a composite, σ_{cu} , calculated from [25]

$$\sigma_{cu} = 3P_f(l/h)/2bh \quad (1)$$

where P_f is the applied load at failure in the three-point bend test, l is the span length, b is the specimen width, and h is the specimen depth (thickness). The failure test was made at 77 K with the span length of 20 mm. The ultimate strength thus determined is plotted in Fig. 1 as a function of absorbed dose. Each data point indicates the average value of three failure tests, and the error bar shows the standard deviation. A missing error bar means that the deviation is too small to be shown.

Comparison of the data points for the electron and γ-ray irradiation (Fig. 1) demonstrates that the dose dependence of the composite strength follows an identical pattern regardless of the type of radiation. This result strongly suggests that the irradiation effects of ⁶⁰Co γ-rays can be simulated by electron irradiation so far as the mechanical properties of a polymer matrix composite are concerned. This will also be the case for the irradiation of pure resins. Thus

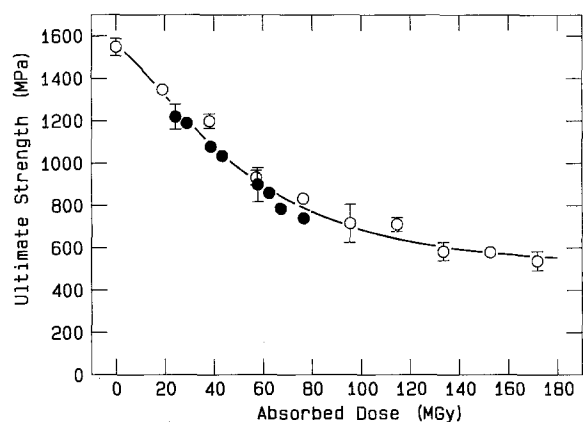


Figure 1 Plot of the ultimate strength at 77 K against the absorbed dose in matrix for the glass/epoxy composite irradiated with (○) 2 MeV electrons and with (●) ⁶⁰Co γ-rays, at room temperature.

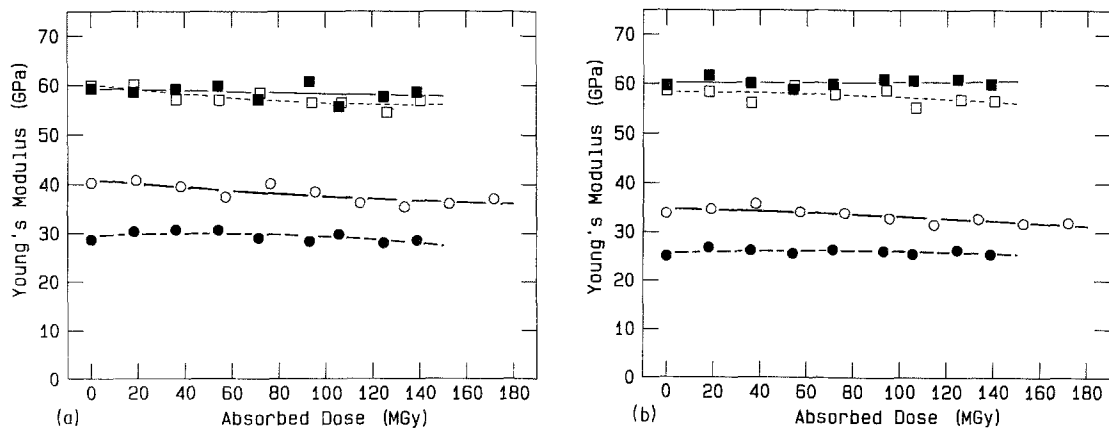


Figure 2 Plot of the Young's modulus at (a) 77 K and (b) room temperature against the absorbed dose in matrix for (○) the glass/epoxy, (●) glass/polyimide, (□) carbon/epoxy, and (■) carbon/polyimide composites.

it is reasonable to neglect the difference in the type of radiation in the present work. In the following sections, the data for the glass/epoxy composite and the pure epoxy and polyimide resins are for electron irradiation, while for the other specimens the data are for γ -ray irradiation.

3.2. Young's modulus

The Young's (tensile) modulus, E , was calculated from [25]

$$E = (P/\Delta)l^3/4bh^3 \quad (2)$$

where P is the applied load and Δ is the midspan deflection. The initial slope (P/Δ) measurement for this purpose was made at the span length of 60 mm for a composite specimen and at 20 mm for a pure resin specimen. The Young's modulus is plotted as a function of absorbed dose in Figs 2a and b for composite specimens tested at 77 K and at room temperature, respectively. Comparison of unirradiated specimens in Figs 2a and b shows that the Young's modulus of the carbon/epoxy or carbon/polyimide composite is appreciably higher than that of the glass/epoxy or glass/polyimide composite for both test temperatures, thus reflecting a higher Young's modulus of carbon fibres compared to glass fibres. Comparison of Figs 2a and b, on the other hand, reveals that the Young's modulus at 77 K is about 17% higher than that at

room temperature for the glass fibre composites, whereas for the carbon fibre composites the modulus is almost independent of test temperature. Following irradiation the Young's modulus of these composites is seen to scarcely change even at 170 MGy.

The Young's modulus for pure resin specimens is plotted in Fig. 3 as a function of absorbed dose. Each data point indicates the average value of six tests at 77 K and three tests at room temperature, and the error bar shows the standard deviation. It is seen that the Young's modulus of unirradiated specimens at 77 K is about twice as high as that at room temperature for both resins of epoxy and polyimide. Following irradiation the Young's modulus of these resins remains practically unchanged even at 160 MGy, although the modulus of the polyimide resin at 77 K seems to increase slightly at the beginning of irradiation. With this exception, the Young's modulus is approximately the same for the epoxy and polyimide resins regardless of the absorbed dose and the test temperature.

3.3. Ultimate strength

The ultimate strength of a composite or a pure resin was calculated from Equation 1 with the P_f value obtained at the span length of 20 mm. The ultimate strength is plotted as a function of absorbed dose in Figs 4 to 7 for composites and in Fig. 8 for pure resins. Each data point in Figs 4 to 7 and that for a room-temperature test in Fig. 8 indicate the average value of

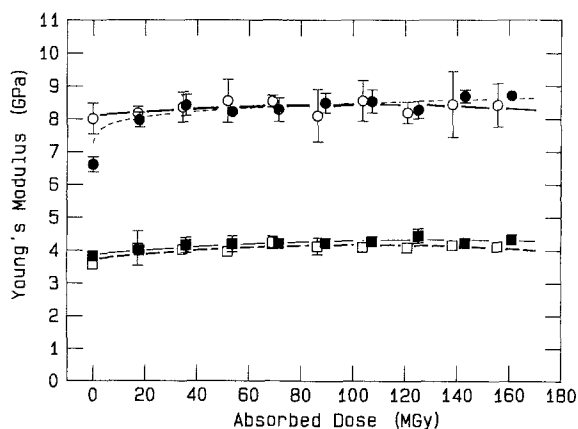


Figure 3 Plot of the Young's modulus against the absorbed dose for (○, □) the pure epoxy and (●, ■) the pure polyimide resins tested at (○, ●) 77 K and (□, ■) room temperature.

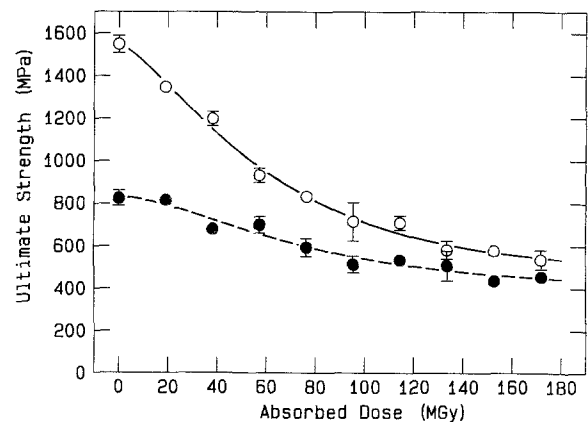


Figure 4 Plot of the ultimate strength at (○) 77 K and (●) room temperature against the absorbed dose in matrix for the glass/epoxy composite.

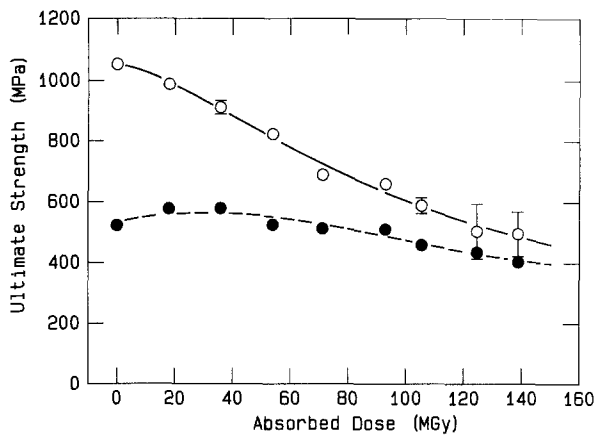


Figure 5 Plot of the ultimate strength at (○) 77 K and (●) room temperature against the absorbed dose in matrix for the glass/polyimide composite.

three failure tests, while that for a 77 K test in Fig. 8 indicates the average value of six failure tests.

The plots for the glass/epoxy or glass/polyimide composite in Figs 4 and 5 demonstrate that the ultimate strength of unirradiated specimens at 77 K is about twice as high as that at room temperature. Following irradiation the ultimate strength of these composites decreases monotonically with increasing dose for both test temperatures, except that a slight increase in the ultimate strength is observed at low doses for the glass/polyimide composite at room temperature (Fig. 5). Comparison of the 77 K and room-temperature data points shows that a decrease in the ultimate strength by irradiation is appreciably greater in the 77 K test than in the room-temperature test.

This phenomenon is also observed for the carbon/epoxy and carbon/polyimide composites shown in Figs 6 and 7, respectively. For these composites, in fact, such a decrease in the ultimate strength is quite small or practically nil even at 140 MGy when tested at room temperature, whereas at 77 K the decrease is significant. On the other hand, no appreciable difference in the ultimate strength before irradiation is found between the 77 K and room-temperature tests for these carbon fibre composites, in contrast to the glass fibre composites of epoxy and polyimide matrix resins. This fact can be seen more clearly from

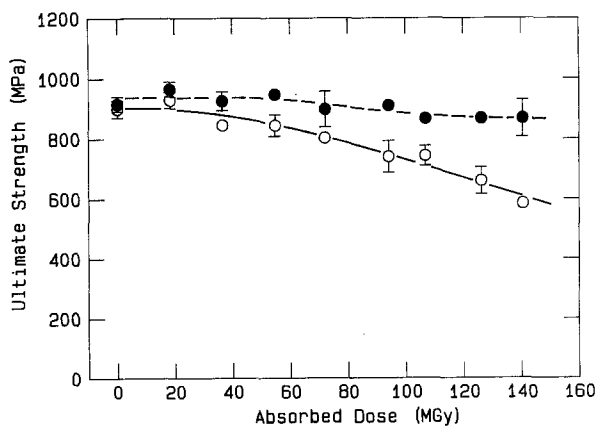


Figure 6 Plot of the ultimate strength at (○) 77 K and (●) room temperature against the absorbed dose in matrix for the carbon/epoxy composite.

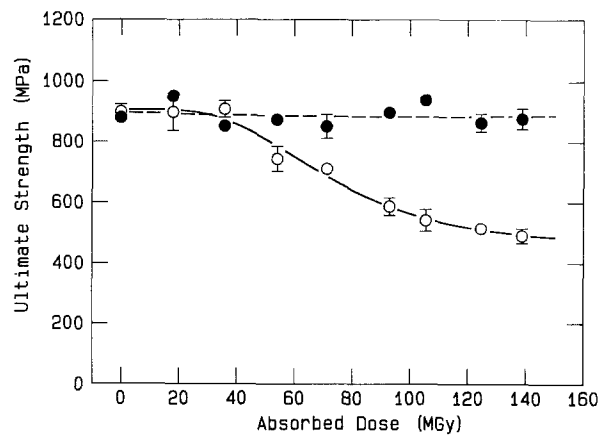


Figure 7 Plot of the ultimate strength at (○) 77 K and (●) room temperature against the absorbed dose in matrix for the carbon/polyimide composite.

the load-deflection curves shown in Fig. 9 for the glass/epoxy and carbon/epoxy composites before irradiation.

The plots for the pure epoxy or polyimide resin in Figs 8a and b demonstrate that the ultimate strength does not differ strikingly between the 77 K and room-temperature tests, considering that the Young's modulus of these resins increases by a factor of about two on cooling from room temperature to 77 K (see Fig. 3). Following irradiation, the ultimate strength of these resins decreases monotonically with increasing dose. Comparison of Figs 8a and b reveals that such a decrease by irradiation is much smaller for the polyimide resin compared to the epoxy resin, thus confirming a high radiation resistance of the polyimide resin [4, 5].

4. Discussion

4.1. Young's modulus

The Young's modulus of a composite was found to change little even after irradiation up to 170 MGy (Fig. 2). This result is explained by the rule of mixtures [26]

$$E_c = \alpha E_f V_f + E_m V_m \quad (3)$$

where E is the Young's modulus, V is the volume fraction, α is the coefficient dependent on the form of fibres (about 0.5 for the present composites), and the subscripts c, f, and m stand for the composite, fibre, and matrix, respectively. This equation predicts that the Young's modulus of a composite does not change as long as the Young's modulus of the fibre and matrix materials remains unchanged. The Young's modulus of the matrix materials was found to remain practically unchanged up to at least 160 MGy (Fig. 3). For the fibre materials, on the other hand, the radiation resistance of inorganic materials such as glass and carbon fibres is known to be much higher than that of organic materials such as epoxy and polyimide resins [3]. Accordingly, the dose-independent Young's modulus of the composites studied here (Fig. 2) is most likely ascribed to the radiation insensitiveness of the modulus for the fibre and matrix materials.

4.2. Ultimate strength

The ultimate strength of a composite was found to

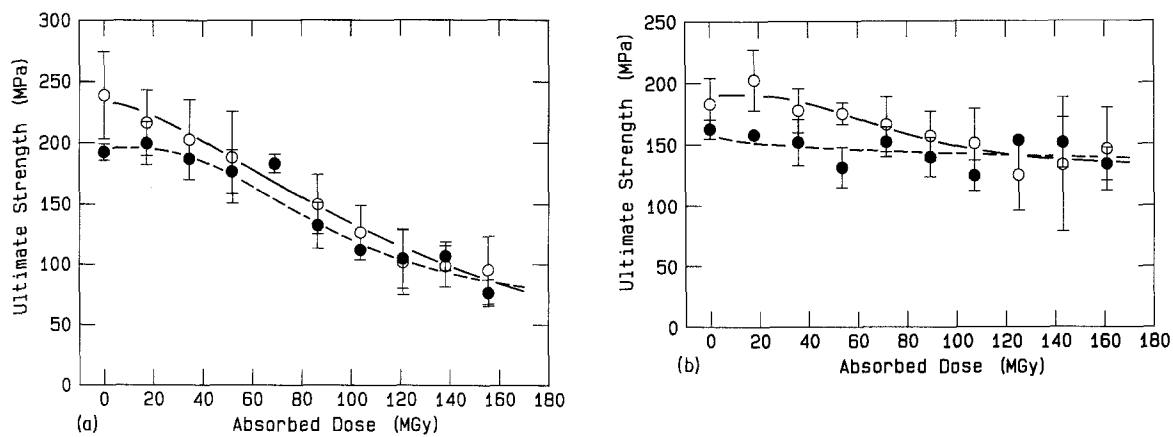


Figure 8 Plot of the ultimate strength at (○) 77 K and (●) room temperature against the absorbed dose for (a) the pure epoxy and (b) the pure polyimide resins.

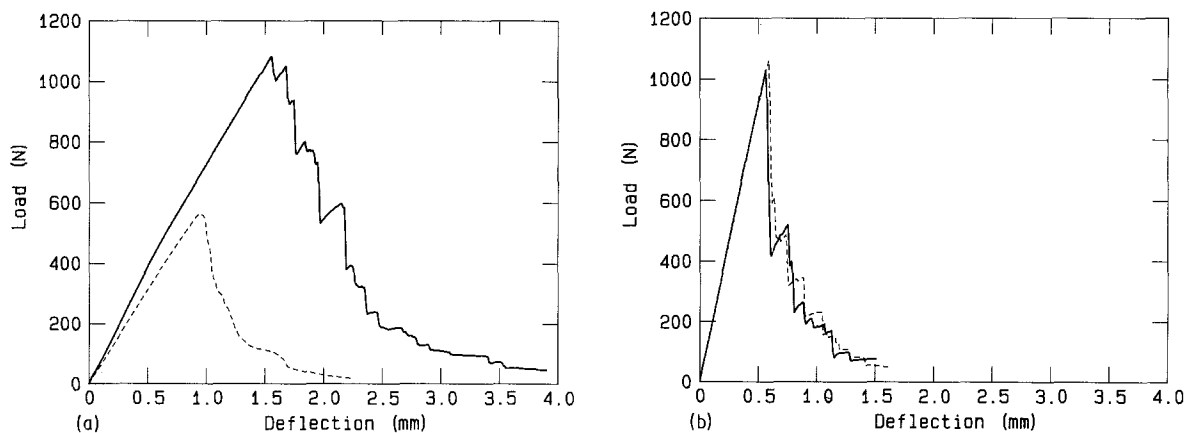


Figure 9 Load-deflection curves obtained in three-point bend tests at (—) 77 K and (---) room temperature for (a) the glass/epoxy and (b) the carbon/epoxy composites, before irradiation.

decrease appreciably with increasing dose for all the composites studied here, except for the carbon/epoxy and carbon/polyimide composites tested at room temperature (Figs 4 to 7). This result should be interpreted in terms of radiation damage in the matrix, the fibre/matrix interface, or both, because such damage in the fibre can be neglected within the dose range covered in the present work [3]. At the present stage, however, it is unknown whether the matrix or the fibre/matrix interface is the more important factor for the radiation sensitivity of a polymer matrix composite. In this connection, our previous work demonstrated that as long as the matrix resin is identical, the dose dependence of the composite strength follows a similar pattern regardless of the kind of reinforcing filler (E-glass, S-2 glass, and alumina fibres) and its surface treatment (see Figs 5 and 6 in [19]). It is reasonable to assume, therefore, that the radiation-induced degradation of a polymer matrix composite is virtually determined by the radiation damage in the matrix rather than that at the fibre/matrix interface.

Comparison of Figs 4 and 8a shows that the ultimate strength of the glass/epoxy composite and that of the pure epoxy resin may be correlated with each other for both the 77 K and room-temperature tests. This correlation suggests that a change in the matrix strength by irradiation virtually determines the dose dependence of the composite strength. Comparison of Figs 6 and 8a, however, reveals that this suggestion is

not correct, because the ultimate strength of the carbon/epoxy composite and that of the pure epoxy resin are not correlated with each other at all for the room-temperature test. It is concluded, therefore, that the dose dependence of the composite strength is determined by factors other than the matrix strength in a composite. As one possibility, let us consider the ultimate strain of a matrix in the next section.

4.3. Ultimate strain

The ultimate strain, ϵ_u , was calculated from [25]

$$\epsilon_u = 6\Delta_f h/l^2 \quad (4)$$

where Δ_f is the midspan deflection at failure in the three-point bend test. This calculation was made by using the same test data as those used in the evaluation of the ultimate strength. The ultimate strain thus determined is plotted as a function of absorbed dose in Fig. 10 for the pure epoxy and polyimide resins and in Fig. 11 for the glass/epoxy and carbon/epoxy composites. For comparison, the plots for the pure epoxy resin at (---) 77 K and (---) room temperature are also reproduced in Fig. 11.

The plot for the pure epoxy and polyimide resins in Fig. 10 demonstrates that the ultimate strain of unirradiated specimens decreases appreciably on cooling from room temperature to 77 K, in contrast to the Young's modulus of these resins (see Fig. 3). Following irradiation the ultimate strain decreases monotonically with increasing dose, although the

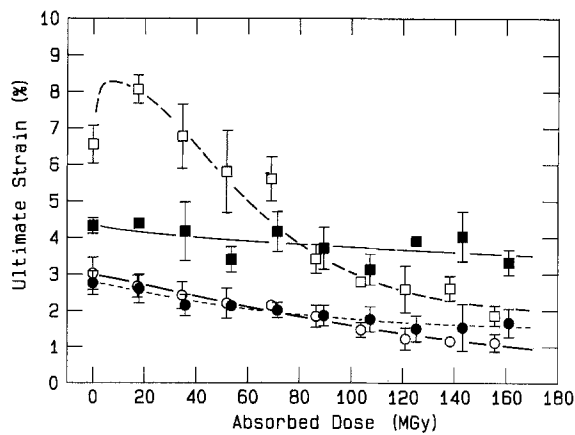


Figure 10 Plot of the ultimate strain against the absorbed dose for (○, □) the pure epoxy and (●, ■) the pure polyimide resins tested at (○, ●) 77 K and (□, ■) room temperature.

ultimate strain of the epoxy resin at room temperature seems to increase slightly at the beginning of irradiation. Comparison of the epoxy and polyimide resins at each dose reveals that the ultimate strain of the epoxy resin is higher than that of the polyimide resin only up to about 80 MGy for both test temperatures, thus reflecting a higher radiation resistance of the polyimide resin than that of the epoxy resin.

The plot for the glass/epoxy and carbon/epoxy composites in Fig. 11 indicates that the dose dependence of the ultimate strain is quite similar to that of the ultimate strength of each composite for both the 77 K and room-temperature tests (see Figs 4 and 6). This similarity of the dose dependence is due mostly to the dose-independent Young's modulus of these composites (Fig. 2). Comparison of the ultimate strain of the glass/epoxy composite with that of the pure epoxy resin at each dose (Fig. 11a) reveals that the composite ultimate strain is always lower than the matrix ultimate strain at room temperature, whereas at 77 K the situation is reversed at every dose studied here. For the carbon/epoxy composite at 77 K (Fig. 11b), however, the composite ultimate strain is lower than the matrix ultimate strain only at low doses below about 80 MGy. At room temperature, the situation for this composite is exactly the same as that for the glass/epoxy composite.

From a simplistic standpoint, the composite ultimate strain lower than the matrix ultimate strain will be regarded to imply a composite failure mode that the failure of a composite occurs when the composite strain is reached at the fibre ultimate strain [27]. Thus this failure mode will be termed the fibre failure mode. The reverse case of the composite ultimate strain higher than the matrix ultimate strain, on the other hand, will be regarded to imply another failure mode that the failure of a composite occurs when the composite strain is reached at the matrix ultimate strain [27]. The failure mode for this case will be termed the matrix failure mode. These considerations lead to a conclusion that the polymer matrix composites studied here have at least two different failure modes, and that the most important factor for the dose dependence of the composite strength is a change in the matrix ultimate strain due to irradiation.

In order to discuss this point quantitatively, we calculate a relationship between the composite ultimate strain and the matrix ultimate strain by using the test data like those shown in Fig. 11. The relationship thus obtained is plotted in Fig. 12. The plot for the glass/epoxy and glass/polyimide composites (Fig. 12a) demonstrates that the composite ultimate strain increases with an increase in the matrix ultimate strain in a similar pattern for both of these composites at 77 K. At room temperature, however, the composite ultimate strain appears to increase at first and then levels off at the matrix ultimate strain of about 4% or above. This tendency is observed also for the carbon/epoxy and carbon/polyimide composites at 77 K and at room temperature (Fig. 12b), although a discrepancy is found between the two composites. This discrepancy may be due, at least in part, to a difference in the volume fraction of fibres for these composites (see Table I).

In Figs 12a and b, the initial region at low values of the matrix ultimate strain is most likely assigned to the matrix failure mode of a composite, because the composite ultimate strain is comparable with the matrix ultimate strain in this region. The other region at high values of the matrix ultimate strain, on the other hand, is assigned to the fibre failure mode of a composite, because the composite ultimate strain is lower than the

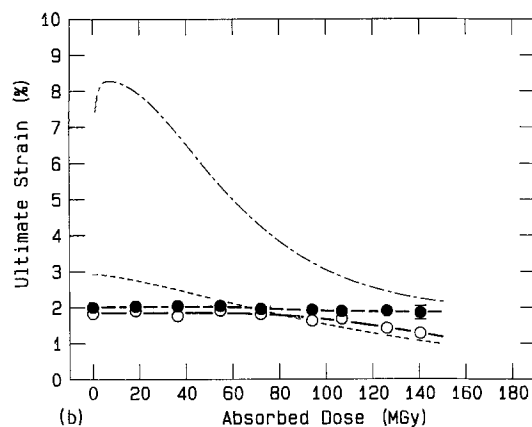
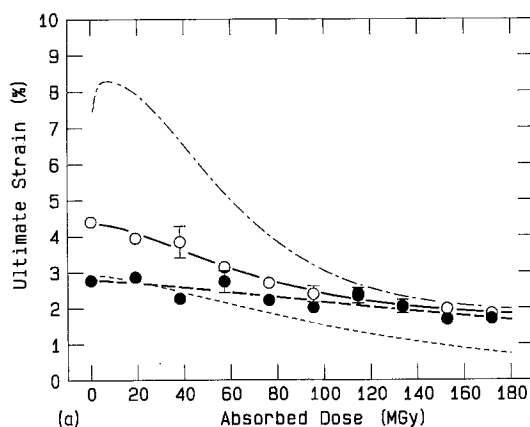


Figure 11 Plot of the ultimate strain at (○) 77 K and (●) room temperature against the absorbed dose in matrix for (a) the glass/epoxy and (b) carbon/epoxy composites. For comparison, the plots for the pure epoxy resin at (---) 77 K and (---) room temperature (Fig. 10) are also reproduced here.

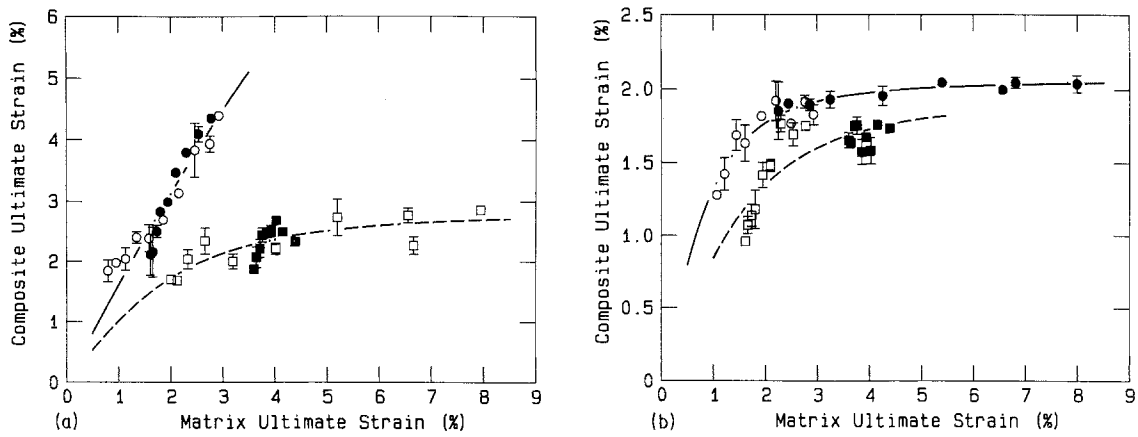


Figure 12 Relationship between the composite ultimate strain and the matrix ultimate strain for (a) the glass and (b) the carbon fibre composites. The data points are as follows for pairs of the 77 K and room-temperature tests: (O, □) glass/epoxy; (●, ■) glass/polyimide; (○, ●) carbon/epoxy; (□, ■) carbon/polyimide.

matrix ultimate strain in this region. Then it is seen from Fig. 12 that the composite ultimate strain is almost independent of the matrix ultimate strain for the fibre failure mode. This is the case, for instance, for the carbon/epoxy composite at room temperature whose ultimate strain or strength is scarcely changed by irradiation (Fig. 11b or 6). For the matrix failure mode, on the other hand, the composite ultimate strain is seen to decrease with a decrease in the matrix ultimate strain. This is the case, for instance, for the glass/epoxy composite at 77 K whose ultimate strain or strength is decreased significantly by irradiation (Figs. 11a or 4). For the other composites at 77 K or at room temperature also, the dose dependence of the ultimate strain or strength can likewise be interpreted based on the failure modes of a composite described above.

4.4. Modelling of composite failure modes

In order to discuss the composite failure mode more quantitatively, we have tried to derive an expression for the relationship between the composite ultimate strain and the matrix ultimate strain. First let us consider the simplest mode by which failure of a composite occurs when the composite strain is reached at the lowest value of the matrix and fibre ultimate strains. This simplest mode can be written as follows for the matrix failure mode

$$\varepsilon_{cu}/\varepsilon_{mu} = 1 \quad (\varepsilon_{cu} \geq \varepsilon_{mu}) \quad (5)$$

and for the fibre failure mode

$$\varepsilon_{cu}/\varepsilon_{fu} = 1 \quad (\varepsilon_{cu} \leq \varepsilon_{fu}) \quad (6)$$

where ε_{cu} , ε_{mu} , and ε_{fu} are the ultimate strain of the composite, matrix, and fibre, respectively. These equations are shown by the broken lines in Fig. 13. In this figure, the $\varepsilon_{cu}/\varepsilon_{fu}$ ratio is plotted as a function of the $\varepsilon_{mu}/\varepsilon_{fu}$ ratio. This is a meaningful way of plotting, because it is expected that the ε_{fu} change on irradiation will be negligibly small in the present work. The broken lines indicate that the composite ultimate strain increases in proportion to the matrix ultimate strain up to the fibre ultimate strain, and then it levels off.

Let us now consider a complex failure mode that the failure of a composite is caused by interactions

between the matrix and fibre failure modes. This failure mode may be written as

$$(\varepsilon_{cu}/\varepsilon_{mu})^2 + (\varepsilon_{cu}/\varepsilon_{fu})^2 = 1 \quad (7)$$

This equation is shown by the solid curve in Fig. 13. The solid curve indicates that the composite ultimate strain increases with an increase in the matrix ultimate strain at first, and then gradually approaches the fibre ultimate strain. This characteristic of the complex failure mode is, in fact, observed for the composites studied in the present work (See Fig. 12). It seems reasonable to conclude, therefore, that the failure mode of actual composites can be approximated more adequately by the complex failure mode (Equation 7) rather than the matrix or fibre failure mode (Equation 5 or 6).

It should be pointed out here that the ε_{fu} value in Equation 7 is for fibres in a composite. The ultimate strain or strength of such fibres will be affected by many factors such as the form of reinforcing fibres, the volume fraction of fibres, and the kind of matrix resin. In fact, according to the statistical analysis for the strength of a bundle (yarn) of fibres in a composite, the bundle strength increases with increasing hardness of the matrix resin [27]. Thus the bundle strength in the composites studied here is expected to increase on cooling from room temperature to 77 K, because the hardness of the epoxy and polyimide resins increases

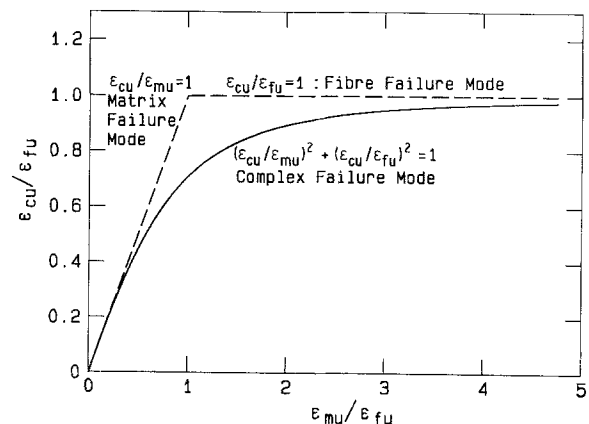


Figure 13 Modelling of composite failure modes based on a relationship between the composite ultimate strain and the matrix ultimate strain.

on cooling, as seen from Fig. 3. In addition, the strength of an E-glass fibre itself is known to increase by about 50% on cooling from room temperature to 77 K [28]. As a result, it is possible that the bundle strength of E-glass fibres in a composite increases by a factor of two or above on cooling to 77 K. If this is also the case for the ultimate strain of the bundle, then the resulting difference in the ϵ_{fu} value in Equation 7 could be responsible for a discrepancy observed between the 77 K and room-temperature tests of the glass/epoxy and glass/polyimide composites (Fig. 12a).

It should also be pointed out that in the present work the modelling of composite failure modes is based on the assumption that the matrix ultimate strain for a pure resin specimen is the same as that for a composite specimen. Strictly speaking, however, this assumption would fail for actual composites, especially for cloth-filled composites. This is because the presence of reinforcing fibres in a composite makes the matrix strain distribution inhomogeneous on a microscopic scale, and hence the local strain can be much higher than the overall strain [27, 29]. In addition, it is generally accepted that a difference in the thermal expansion between the fibre and matrix materials generates a shear stress at the fibre/matrix interface in a composite even before the mechanical test [30]. This effect will also increase the microscopic inhomogeneity of the matrix strain distribution. As additional work, modelling of composite failure modes taking these points into consideration would be very useful for furthering the fundamental understanding of the degradation mechanism of polymer matrix composites.

5. Conclusions

The present work has shown that the dose dependence of the ultimate strength of a composite depends not only on the combination of fibre and matrix in the composite but also on the temperature during the mechanical test. A relationship found between the composite ultimate strain and the matrix ultimate strain indicates that the dose dependence of the composite strength is virtually determined by a change in the matrix ultimate strain due to irradiation. Modelling of composite failure modes based on this finding reveals that the failure mode of the composites studied here can be adequately approximated by the complex failure mode.

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References

1. B. S. BROWN, *J. Nucl. Mater.* **97** (1981) 1.
2. F. W. CLINARD Jr and G. F. HURLEY, *ibid.* **103**, **104** (1981) 705.
3. R. D. HAY and E. J. RAPPERPORT, Oak Ridge National Laboratory Report ORNL/TM-2643 (1976).
4. R. R. COLTMAN Jr and C. E. KLABUNDE, *J. Nucl. Mater.* **103**, **104** (1981) 717.
5. *Idem*, *ibid.* **113** (1983) 268.
6. C. E. KLABUNDE and R. R. COLTMAN Jr, *ibid.* **117** (1983) 345.
7. S. TAKAMURA and T. KATO, *ibid.* **103**, **104** (1981) 729.
8. *Idem*, *Adv. Cryogenic Engng Mater.* **30** (1984) 41.
9. G. F. HURLEY, J. D. FOWLER and D. L. ROHR, *Cryogenics* **23** (1983) 415.
10. D. S. TUCKER, J. D. FOWLER Jr and F. W. CLINARD Jr, *Fusion Technol.* **8** (1985) 2696.
11. H. W. WEBER, E. KUBASTA, W. STEINER, H. BENZ and K. NYLUND, *J. Nucl. Mater.* **115** (1983) 11.
12. H. YAMAOKA, K. MIYATA, Y. NAKAYAMA and H. YOSHIDA, in "Nonmetallic Materials and Composites at Low Temperatures", Vol. III, edited by G. Hartwig and D. Evans (Plenum, New York, 1986) p. 71.
13. R. E. FORNES, J. D. MEMORY and N. NARANONG, *J. Appl. Polym. Sci.* **26** (1981) 2061.
14. S. M. MILKOVICH, C. T. HERAKOVICH and G. F. SYKES, *J. Compos. Mater.* **20** (1986) 579.
15. S. EGUSA, M. A. KIRK, R. C. BIRTCHER, M. HAGIWARA and S. KAWANISHI, *J. Nucl. Mater.* **119** (1983) 146.
16. *Idem*, *Nucl. Instrum. Meth. Phys. Res.* **B1** (1984) 610.
17. S. EGUSA, M. A. KIRK, R. C. BIRTCHER and M. HAGIWARA, *J. Nucl. Mater.* **127** (1985) 146.
18. S. EGUSA, H. NAKAJIMA, M. OSHIKIRI, M. HAGIWARA and S. SHIMAMOTO, *ibid.* **137** (1986) 173.
19. S. EGUSA and M. HAGIWARA, *Cryogenics* **26** (1986) 417.
20. S. EGUSA, M. A. KIRK and R. C. BIRTCHER, *J. Nucl. Mater.* **126** (1984) 152.
21. *Idem*, *ibid.* **148** (1987) 43.
22. *Idem*, *ibid.* **148** (1987) 53.
23. R. TANAKA and Y. NAKAMURA, *Radioisotopes* **24** (1975) 11.
24. R. TANAKA, S. MITOMO, H. SUNAGA, K. MATSUDA and N. TAMURA, Japan Atomic Energy Research Institute Report JAERI-M 82-033 (1982).
25. L. E. NIELSEN, in "Mechanical Properties of Polymers and Composites" (Marcel Dekker, New York, 1974) p. 45.
26. R. H. KROCK and L. J. BROUTMAN, in "Modern Composite Materials", edited by L. J. Broutman and R. H. Krock (Addison-Wesley, Massachusetts, 1967) Ch. 1.
27. H. T. CORTEN, *ibid.*, Ch. 2.
28. N. M. CAMERON, *Glass Technol.* **9** (1968) 14.
29. C. C. CHAMIS, in "Composite Materials", Vol. 6, edited by E. P. Plueddemann (Academic, New York, 1974) Ch. 2.
30. D. A. SCOLA, *ibid.*, Ch. 7.

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