8.5 Radiation Effects on Epoxy Composites at Cryogenic Temperatures

H. Yamaoka[1], K. Miyata[2], S. Nishijima[3] and T. Okada[3]

[1]Department of Polymer Chemistry, Kyoto University, Kyoto 606-1, Japan, [2]Research Reactor Institute, Kyoto University, Kumatori, Osaka 590-04, Japan and [3]Institute of Scientific and Industrial Research, Osaka University, Osaka 567, Japan

Abstract Radiation effects on glass-fiber reinforced epoxy composites at cryogenic temperatures has been studied by measuring the changes in interlaminar shear strength of the specimens. The scanning electron microscope observation has also been performed on fracture surface of the specimens. At 8.5 MGy of absorbed dose, only 10 % decrease of the strength was observed in the case of gamma irradiation, whereas over 80 % decrease of the strength was found on the reactor irradiated specimen. The difference of degradation behavior between gamma and reactor irradiations is attributed to the additional absorbed dose in the latter from the nuclear reaction due to boron-10 contained in the glass fibers by capture of thermal neutrons.

INTRODUCTION

Design studies for several fusion reactors and devices have become more detailed in recent years. One of the important aims of design engineering is to select suitable materials which perform reliably under the most severe conditions. Among several conceptual designs of fusion reactors, the machines based on magnetic confinement are planning to utilize a variety of superconducting magnets which are operated at cryogenic temperatures. The components of superconducting magnets are exposed to intense radiation of fast neutron and secondary γ-rays. Because of the brittleness and the difficulty of fabrication techniques in inorganic materials, the application of various organic materials has been considered for electrical insulators, thermal insulators and a part of structural supports in the magnets (Clinard and Hurley, 1981; Coltman, 1982; Hurley and Coltman, 1984).

Most of organic materials in fusion reactors must be designed to employ organic-matrix composites as it appears doubtful that conventional polymers can withstand the severe radiation environment for a long operation time. In the present paper, radiation effects on glass-fiber reinforced epoxy composites at cryogenic temperatures have been studied with mechanical measurements and scanning electron microscope observations.

EXPERIMENTAL

Materials

The specimens used in this work were basically glass-cloth-reinforced epoxy laminates. Both Lamiverre-A^R (Sample A) and Hoxan^R (Sample B) were commercially available diglycidyl ether of bisphenol A liquid-type epoxy resin laminates, and contained 60 to 62 % of E-glass woven fabrics by weight. Sample C and Sample D were specially prepared epichlorohydrin-bisphenol A solid-type epoxy resin laminates reinforced with E-glass cloth and T-glass (boron-free) cloth, respectively. E-glass contains the element of boron as a form of B_2O_3. Glass cloth content of both specimens were 50 % by weight.

Irradiation

γ-Irradiation of the specimens was done at room temperature and 77 K at a dose rate of 2 x 10^4 Gy/h with a ^{60}Co source of Osaka University. Reactor irradiation of the specimens was performed at a low-temperature irradiation facility of Kyoto University Reactor, the average temperature of which was about 20 K under reactor operation at 5 MW. The highest fast ($E > 0.1$ MeV) and thermal neutron fluences of the facility were 2.5 x 10^{15} n/m^2/s and 2.3 x 10^{16} n/m^2/s, respectively. The γ-ray dose rate at the same position was 1.2 x 10^5 Gy/h. After irradiation, the specimens were stored in liquid nitrogen (77 K) without warming up to room temperature and subjected to mechanical tests.

Measurements

Measurements of interlaminar shear strength (ILSS) of composites were performed with the use of specially prepared specimen shown in Fig. 1. The mean value of ILSS was obtained by compressing the specimen, although there are many problems of cryogenic ILSS testing arrangements (Becker, 1990). The displacement speed of Shimadzu Autograph Testing Machine, Model AG-500, was set at 1 mm/min. The fracture surface of specimens was observed with a JXA-50A scanning electron microscope (Japan Electron Optics Lab.) operated at 50 KeV.

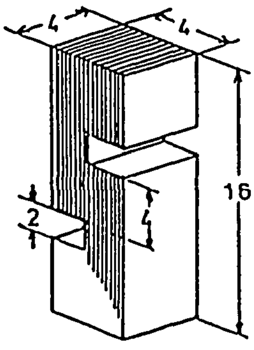

Fig. 1. Shape and size of specimens (in mm).

RESULTS AND DISCUSSION

The study on the mechanism of radiation-induced deterioration in organic composites has elucidated that the essential property which contributes the deterioration is the change in ILSS of composites (Okada *et al.* , 1986). An example of radiation effect on the mechanical properties of epoxy composites at room temperature is shown in Fig. 2. After γ-irradiation of Sample A to 20 MGy, the ILSS of the specimen was reduced to approximately 75 % of the initial value, although the tensile strength of the same specimen kept constant (Nishijima *et al.*, 1988). This means that the tests on ILSS are more sensitive as a measure of radiation-induced deterioration of composites than those on tensile strength.

The changes in ILSS of Sample B at different irradiation conditions are represented in Fig. 3, where the absorbed doses due to fast neutrons were estimated under the conversion factor of 1 Gy = 10^{15} n/m^2 (E > 0.1 MeV). When this conversion factor was used, the absorbed dose due to neutrons was given to be one order of magnitude smaller than the absorbed dose of -radiation from the reactor, indicating that the absorbed dose due to fast neutron irradiation can be neglected. Nevertheless, the remarkable difference between - and reactor irradiations was observed. After cryogenic irradiation up to 8.5 MGy, over 80 % decrease of strength was found on the reactor irradiated specimen, although only 10 % decrease of the strength was observed in the case of -irradiation.

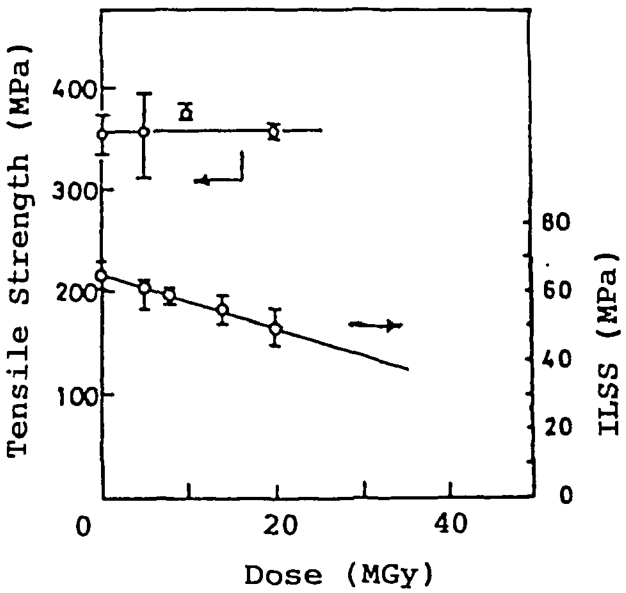

Fig. 2. Effect of γ-irradiation on tensile strength and ILSS of Sample A.

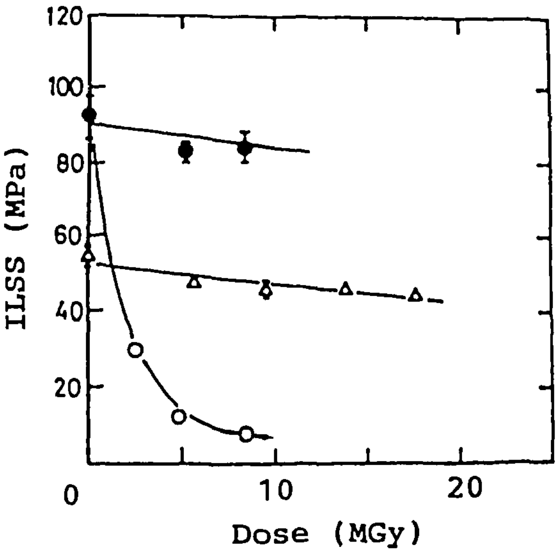

Fig. 3. Changes in ILSS of Sample B at different irradiation conditions. (o : Reactor irradiation at 20 K, tested at 77 K; ● : γ-irradiation at 77 K, tested at 77 K; Δ : γ-irradiation at room temperature, tested at room temperature.)

In order to clarify this different effect, the reactor irradiations of Sample C and Sample D were carried out at 20 K. The results obtained are shown in Fig. 4. The ILSS of Sample C reinforced with E-glass cloth markedly decreased with an increase of dosage, whereas that of Sample D reinforced with T-glass showed rather small effect on irradiation. The difference of radiation damage between two samples seems to be attributed to the additional absorbed dose in Sample C from the fission of boron-10 in E-glass through a nuclear reaction of $^{10}B(n,\alpha)^{7}Li$ by capture of thermal neutrons (Okada *et al.*, 1992). Similar effects were also observed in the flexural tests of epoxy composites (Coltman and Klabunde, 1983) and the fatigue tests of various composites (Schmunk *et al.*, 1984).

The results of scanning electron microscope (SEM) observation on the fracture surface of irradiated specimens of Sample D revealed that the good adhesive nature with the matrix resin and the T-glass cloth were still maintained even after cryogenic reactor irradiation of 7.4 MGy. On the contrary, the SEM photograph of the specimens of Sample C showed that the surface of E-glass cloth at the same irradiation conditions was very smooth, indicating the debonding with the matrix resin. These fact are consistent with the results obtained in ILSS tests.

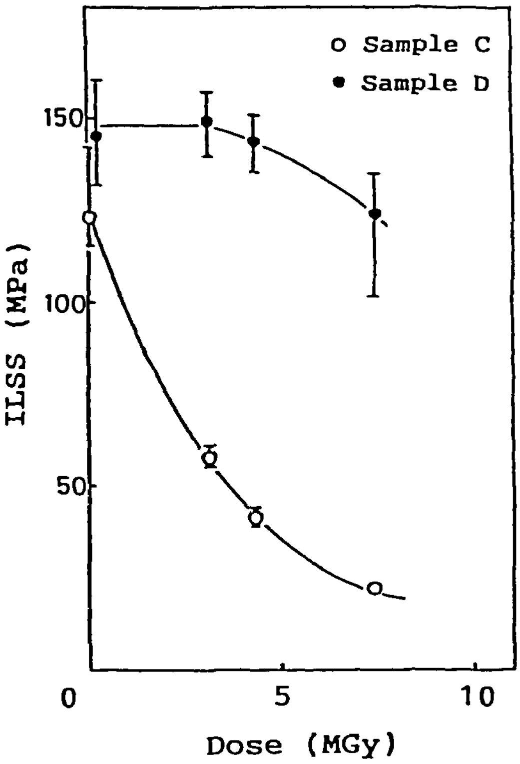

Fig. 4. Reactor irradiation of Sample C and Sample D at 20 K. (Tested at 77 K.)

CONCLUSION

In the epoxy composites reinforced with E-glass cloth, the remarkable difference in deterioration behavior was observed between gamma and reactor irradiations at cryogenic temperatures.

The drastic reduction in the ILSS of S-glass reinforced epoxy composites arose from a marked debonding in the matrix-glass cloth interface due to the additional absorbed dose resulted in the thermal neutron fission of boron-10 atoms. The SEM observation of fracture surfaces of irradiated specimens gave another evidence for the difference in radiation damage between E-glass and T-glass reinforcements.

The results obtained in this work suggest that the boron-free glass as reinforced fibers should be applied for polymer composites used in intense neutron fields.

REFERENCES

Becker H. (1990) Adv. *Cryog. Eng. (Mater.)* 36, 827

Clinard F. W. and Hurley G. F. (1981) *J. Nucl. Mater.* 103/104, 705

Coltman, Jr. R. R. (1982) *J. Nucl. Mater.* 108/109, 559

Coltman, Jr. R. R. and Klabunde C. E. (1983) *J. Nucl. Mater.* 113, 268

Hurley G. F. and Coltman, Jr. R. R. (1984) *J. Nucl. Mater.* 122/123, 1327

Nishijima S., Okada T., Miyata K. and Yamaoka H. (1988) *Adv. Cryog. Eng. (Mater.)* 34, 35

Okada T., Nishijima S. and Yamaoka H. (1986) *Adv. Cryog. Eng. (Mater.)* 32, 145

Okada T., Nishijima S., Nishiura T., Miyata K and Yamaoka H (1992) *Adv. Cryog. Eng. (Mater.)* 38, 241

Schmunk R. E., Miller L. G. and Becker H. (1984) *J. Nucl. Mater.* 122/123, 1381