Effect of 1.33 MeV γ Radiation and 0.5 MeV Electrons on the Mechanical Properties of Graphite Fiber Composites*

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Synopsis

Epoxy/graphite fiber, polyimide/graphite fiber, and polysulfone/graphite fiber composites were exposed to 1.33 MeV γ irradiation and 0.5 MeV electron bombardment for varying periods of time. The effects of the irradiation treatments on the breaking stress and Young's modulus were studied by a three point bending test. Effects were small; indeed, both electron radiation up to 5000 Mrad and γ radiation up to 350 Mrad resulted in slight increases in both stress and modulus.

INTRODUCTION

Graphite fiber reinforced composites are light-weight high-strength materials that are particularly suitable for space vehicles. Since some space experiments are scheduled over a period of several years, materials used in space may be subjected to substantial quantities of high-energy radiation. For this reason, it is important that materials considered for use in these experiments be evaluated with respect to their response to high-energy radiation.

Several sets of composite samples, fabricated at NASA Langley Research Center and supplied to us, have been irradiated using 0.5 MeV electrons and 1.33 MeV γ radiation. The effects of the irradiation treatments on breaking stress and Young's modulus were determined by a three point bending test.

Most of the cosmic radiation in regions near the earth is due to protons.[1] However, significant numbers of both protons and electrons are trapped in the radiation belts around the earth[2,3] and the predominant energy loss in matter of high-energy electrons found in geosynchronous orbit is by ionization.[3,4] Similarly, 1.33 MeV γ radiation would lose most of its energy by ionization through Compton scattering and the photoelectric effect.[5] Therefore, the experimental conditions of radiation exposure of materials to be investigated in this study should provide an excellent simulation of the actual effects of radiation on these materials when used in space applications. Moreover, it has been estimated[6] that the radiation dose for geosynchronous orbit in a thirty year lifetime should be on the order of 1000 Mrad for the 0.056–0.081 cm thickness of the samples we used in the experimental work reported here. This, coupled with the observation that radiation effects on solid polymers are dose rate independent to first order,[7] should indicate that the results described in this paper for doses up to 5000 Mrad should be applicable to the problem of estimating the radiation effects on space vehicles in geosynchronous orbit.

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EXPERIMENTAL PROCEDURES

Studies were made on samples of cured graphite fiber/epoxy composite (T300/520B Thornel graphite fiber/Narmco 5208 epoxy and AS/3501 graphite fiber/epoxy by Hercules), graphite fiber/polyimide composite (C6000/PMR-15 Celion graphite fiber/polyimide matrix), and graphite fiber/polysulfone composite (C6000/P1700 Celion graphite fiber/polysulfone matrix). The samples were cured and cut at NASA Langley Research Center. Samples of each type were subjected for different periods of time to electron irradiation, and samples of graphite fiber/epoxy were γ irradiated for different periods of time.

The mechanical tests were made on an Instron using a "three-point bending tester" attachment.[8] The specimens, 1.27 by 2.54 cm and 0.056–0.071 cm thick, were tested at a constant rate of elongation, perpendicular to the plane of the composites, at a speed of 0.254 cm/min (0.1 in./min) and with a span length of 1.40 cm (0.55 in.). The specimens were four-ply uniaxially oriented with the preferred axis aligned along the span direction during testing. The ultimate stress and average modulus at each exposure condition were determined by using the standard equations for small bending deformations of elastic bodies.[8]

The samples were treated in a vacuum desiccator at 80°C for 7 days, then sealed in aluminum foil (Reynolds Wrap, heavy duty, thickness of 0.025 mm) by first securing the ends of the samples in place with a thin layer of Scotch tape and sealing the edges of the folded aluminum wrap with an epoxy glue (Devcon 5 min Epoxy®). An open glass tube was inserted prior to sealing the foil to permit a vacuum line to be connected for further vacuum treatment. These packages were placed in a vacuum desiccator at 80°C for at least an additional 4 days; then the glass tube was attached to a vacuum line and heat sealed. The packages then were taken immediately to the electron accelerator and exposed to 0.5 MeV electrons at a current of 8.3 mA. Each package was placed in a ziploc baggie by Dow Chemical (10 by 10 in.) that was prefilled with N_2 to reduce oxidative degradation in case of pin hole leaks, the packages clamped to the conveyor on the accelerator, and passed through the electron beam. Each revolution of the conveyor through the beam resulted in a 10 Mrad dosage. Following the electron irradiation treatment in a 500 kV Electron accelerator made by High Voltage Engineering Corporation, the specimens were removed from the packages and placed in open petri containers in a controlled laboratory (relative humidity 65%, temperature 20°C) where they remained from 3 to 10 weeks prior to mechanical testing. Following each 400–500 Mrad exposure, the Ziploc baggies were replaced and reflushed with N_2. After 2500 Mrad, the Al foil in regions of high stress concentrations (sharp bends) showed evidence of degradation and the Devcon epoxy seal on the aluminum foil showed evidence of appreciable discoloration, so the specimen were vacuumed and repackaged in new Al foil as described above.

Two sets of samples, T300/5208 and AS/3501, were exposed to 1.33 MeV γ radiation for periods up to 500 and 1070 h, respectively, in a Gamma Cell 220 made by Atomic Energy of Canada. These samples were vacuum desiccated at 80°C for a minimum of 3 days then placed in the vacuum chamber of the gamma cell (a $\frac{1}{3}$ Mrad/h ^{60}Co source), prevacuumed treated for 24 hr and exposed to γ radiation under a continuous vacuum (approximately 5×10^{-6} Torr). The samples, after γ exposure, were tested using the same procedures for equilibration and mechanical testing as described above.

RESULTS AND DISCUSSION

The theoretical prediction of stopping power of high energy electrons is given as by Segre.[5]

However, due to a secondary scattering, the effective irradiation dosage in a material will first increase and then is followed by a rapid monotonic decrease in radiation dosage.[9] Since the electron accelerator used in our experiments is designed to pass samples across the beam twice during each revolution of the conveyor (once on the front side of the sample and once on the back side), the approximate dosage experienced by the composite specimens as a function of penetration depth is as shown in Figure 1. The density of the composite is about 1.55 g/cm^3. The effective thickness of the baggie and aluminum foil is approximately 0.013 cm. Thus, the effective radiation dosage in the center of the specimen is approximately 40% higher than on the edge.

The load deformation curves of the three point bending tests were approximately linear in all cases and the deflections small so that equations for small bending deformations give excellent approximations of ultimate stress and Young's modulus.[8] Effects of electrons and γ radiation on these parameters for the samples investigated are summarized in Figures 2–5. A minimum of ten replication measurements were made for each exposure condition. All samples exposed to 5000 Mrad of 0.5 MeV electrons showed a slight increase in both stress and modulus compared with the control. In each case, the increases were essentially monotonic. The increases at 5000 mrad to the control were 13%, 10%, and 11.5% in stress and 11%, 12%, and 12% in modulus for samples T300/5208, C6000/PMR15, and C6000/P1700, respectively.

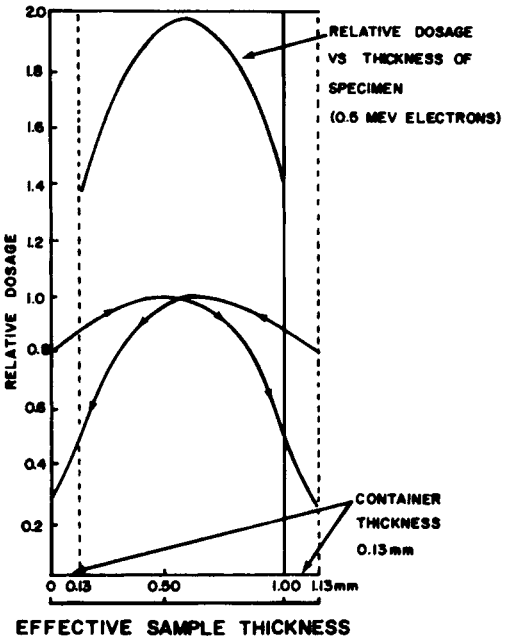

Fig. 1. Estimate of relative composite sample dosage vs. thickness when exposed to 0.5 MeV electrons in the sample holders when both sides of sample are exposed to the beam. The dose-distance relationship is adjusted to unit density material by multiplying thickness by specific gravity.

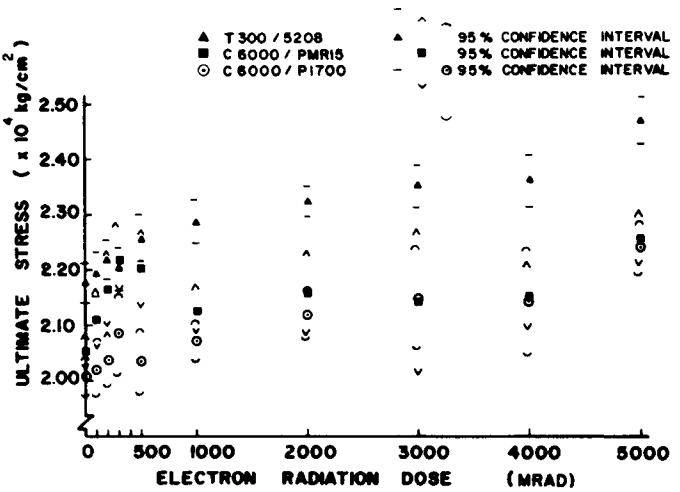

Fig. 2. Ultimate stress vs. 0.5 MeV electron irradiation dosage for graphite fiber composite samples.

Results are shown for the γ irradiated samples in Figures 4 and 5. At the dose levels applied in these experiments, no large changes were observed in either stress or modulus. (The maximum difference from the control for any of the treatments were <10%.)

To test for significant difference in the stress or modulus as a function of irradiation dosage, analysis of variance was done using a statistical analysis system (SAS).[10] In a sample set the data for two irradiation treatments were compared at the 5% confidence levels using the Duncan Multiple range test. In all cases, the 5000 Mrad treatment was significantly different from the control for both the stress and the modulus. Earlier work has been reported by Parkinson and Sisman[11] on the effects of radiation on the mechanical properties of a number of plastics. Their work suggests that polymers containing aromatic rings are highly resistant to radiation and they attribute this to the absorption and dis-

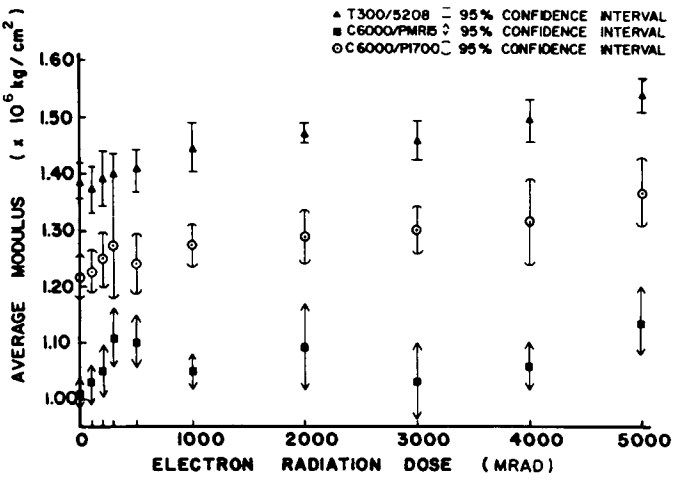

Fig. 3. Average modulus vs. 0.5 MeV electron irradiation dosage for graphite fiber composite samples.

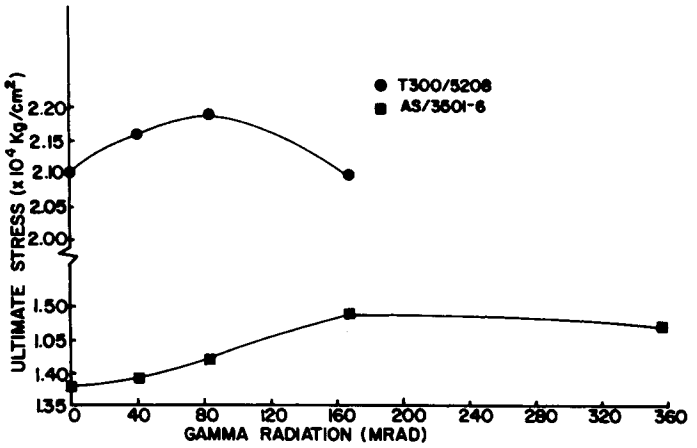

Fig. 4. Ultimate stress vs. 1.33 γ irradiation dosage for graphite fiber/epoxy composites.

sipation of energy, without bond disruption, of aromatic rings. Using neutron and γ radiation, they showed very little change in the mechanical properties of cured diaminodiphenyl methane epoxy and polyimide polymer at radiation dosages $\geq 10^9$ rad. Gamma irradiation experiments by Brown and O'Donnell[12] on aromatic polysulfone show that no deleterious effects occur in vacuum to dose levels of 600 Mrads. However, degradation of flexural properties did occur when irradiation of samples was done in air.

Bullock reported that fast-neutron irradiation of graphite fibers in air showed an increase in strength followed by a decrease (by as much as 25%) of the control.[13] However, irradiation in a inert environment showed only an increase in the strength.[14] Jones and Peggs[15] show a small increase in both the strength and modulus of graphite when irradiated with fast neutrons at elevated temperature. In addition, they reported an increase in the crystallite dimension suggesting that the elevated temperature induced recrystallization.

Graphite fiber/epoxy composites irradiated in air at 75°C with a combination of γ, fast neutrons, and thermal neutrons showed a decrease in flexural strength and modulus.[16] When samples were irradiated in liquid N_2, increases in the strength and modulus were observed when tested at liquid N_2 temperature while

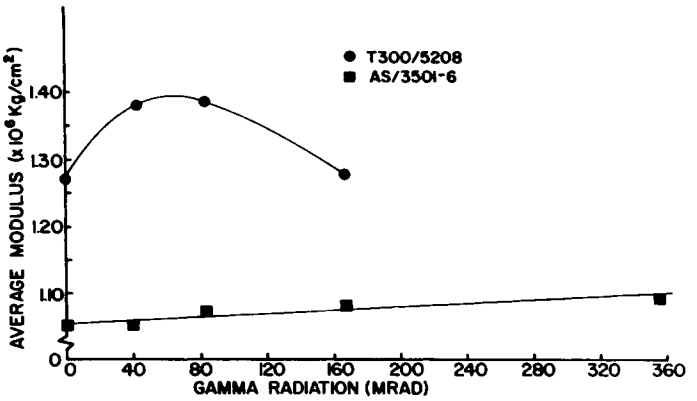

Fig. 5. Average modulus vs. 1.33 γ irradiation dosage for graphite fiber/epoxy composites.

a decrease in those parameters occurred when irradiated at liquid N_2 temperature but tested at room temperature.

The results reported here are consistent with earlier work on plastics, fibers and composites. All the composites that we have studied contain matrices which have an abundance of ring structures and none of the systems we studied showed any degradation in stress or modulus when subjected to ionizing radiation, under vacuum, to dose levels of 5×10^9 rad. The ring structures in the matrices and the fibers appear to protect the composite from radiation damage.

CONCLUSION

Graphite fiber/epoxy, graphite fiber/polysulfone, and graphite fiber/polyimide composites show no deleterious stress or modulus effects by the exposure of 0.5 MeV electron radiation in vacuum up to 5000 Mrad. At 5000 Mrad the stress and modulus increased by approximately 12% compared with the controls. Graphite fiber/epoxy composites show little change in stress and modulus when exposed to several hundred Mrad of γ radiation. Therefore, the results reported here indicate that graphite fiber composites would have a considerable lifetime in space (probably >30 years) before strength and stiffness properties would be affected significantly by high-energy radiation.

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