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THE EFFECT OF A 100 MGy (10^{10} RADS) GAMMA-RAY DOSE AT 5 K ON THE STRENGTH OF POLYIMIDE INSULATORS

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3.2 THE EFFECT OF A 100-MGy (10^{10} RADS) GAMMA-RAY DOSE AT 5 K ON THE STRENGTH OF POLYIMIDE INSULATORS — R. R. Coltman, Jr., C. E. Klabunde, and C. J. Long (Oak Ridge National Laboratory)

3.2.1 SPM Task

Radiation Behavior of Magnet Insulation

3.2.2 Objective

This study seeks to determmine the strength of polyimide materials as a function gamma-ray irradiation dose at 5 K. It compares new results with those from previous studies of epoxies made under the same conditions.

3.2.3 Summary

The most recent efforts in this program have examined the strength and other properties of pure and glass-fiber-filled polyimide materials irradiated to a dose of 100 MGy (10^{10} rads). At this dose the losses in strength measured at 78 K were less than 40%, and at 300 K slight increases were observed. Overall, the glass-fiber-filled polyimide materials are 5 to 10 times more radiation resistant than glass-fiber-filled epoxy materials.

3.2.4 Progress and Status

The aim of this program is to understand the irradiation behavior of materials that may be used in the construction of large superconducting coils which provide magnetic containment for the plasma in a fusion reactor. The use of such materials provides an economical method for producing not only electrical insulation but also mechanical support needed in coil construction. Although many studies on the effects of radiation on organic insulators do exist, there are very few studies on the most recently developed materials or on the simultaneous effects of radiation and low temperatures.

Progress in the conceptual design of fusion reactors has led to the consideration of higher plasma power densities, and hence higher irradiation doses to superconducting coils and their components than had first been considered.[1] In addition, recent calculations by B. A. Engholm[2] show that without additional protective shielding even the first test reactors such as the Engineering Test Facility (now known as the Fusion Engineering Device) will produce lifetime doses at insulator locations that exceed the radiation tolerance[3] of glass-filled epoxies in present use. For these reasons, new irradiations were carried out on pure and glass-filled polyimide materials. The superior radiation resistance of pure polyimides at room temperatures has been known for some years. Only recently, however, have bulk glass-filled polyimides become available. These materials have the strengths which are needed in magnet construction.

This is an interim report giving results for a gamma-ray dose of 100 MGy (10^{10} rads) on various polyimide materials. These results will be combined in a later report with those obtained from intermediate doses now being carried out. The experimental procedures and conditions that apply to this study are the same as those used for recent epoxy studies, which have been given in an earlier report.[3] For the sake of completeness, however, some experimental details are repeated in this report.

3.2.4.1 Materials

Glass-fiber-filled materials were especially prepared by cooperating manufacturers. The unfilled polyimide, Vespel, is available commercially. The following list gives the material designation as used in this report and a brief description of each. For a description of the glass-fiber-filled epoxies to which the present results are compared, see Ref. 3.

1. Vespel — Unfilled polyimide; E. I. DuPont de Nemours and Company.
2. Spauldite — SPAULDITE® SPAULDRAD™; Spaulding Fibre Company; a high-pressure laminate composed of aromatic polyimide resin reinforced with continous filament E glass-woven fabric 70-71% by weight.
3. Kerimid — Kerimid® 601 laminate; Norplex Division, VOP Inc.; a balanced resin of bismaleimide and aromatic diamines reinforced with E glass woven fabric 40-60% by weight.

3.2.4.2 Types of Tests and Sample Specifications

Specimens for testing flexure and compressive strengths were divided into two identical groups, one for irradiation and the other for unirradiated controls. Flexure tests were made at both 300 and 78 K, while compressive tests were made only at 78 K. The compression tests of Spauldite were made only in the direction parallel to the glass fiber laminations, while tests of Kerimid were made both parallel and perpendicular to the laminations. Three tests were made for each of the indicated test conditions of material, temperature and orientation, making a total of 30 control and 30 irradiated specimens. Flexure tests at 78 K on Spauldite and Kerimid were made in 3-point bending, while all other flexure tests were made in a 4-point bending with 25 mm between supports for both cases. Values for fracture stress, linear flexure modulus, and flexure strain were calculated using formulations given in the ANSI/ASTM D 790-71(78) code. The nominal dimensions of the specimens in mm were 1.6 × 3.2 × 50 for the flexure sticks and 6.4 d × 12.7 ℓ for the compression rods.

3.2.4.3 Irradiation and Test Procedures

As in previous experiments[3] the specimens were contained in a basket made of Al screen which in turn was surrounded by a 0.13-mm-thick cylindrical Cd shield. This capsule was then inserted into the sample chamber of the ORNL Low Temperature Radiation Facility (LTIF) located at the Bulk Shielding Reactor. The Cd shield converted 92% on the thermal neutron flux present in the LTIF into gamma rays via an (n,γ) reaction.

Prior to the initial cool-down of the LTIF the sample chamber was evacuated to less than 7 Pa (0.05 torr) each of several times after backfilling with clean dry helium. Immediately after the irradiation, the liquid He in the sample chamber was removed by pumping to 47 kPa (350 torr) without sample warm-up above 5 K. The liquid-He refrigerator was then turned off and the samples warmed to room temperature. Twenty-four hours after termination of the irradiation, the contents of the chamber were pumped slowly through an external trap immersed in a laboratory vessel of liquid He where all gases except He were frozen out. The trap was then sealed off, warmed to room temperature, and the contents analyzed. After backfilling the sample chamber again, a second gas sample was taken 144 hours (6 days) after irradiation.

The conditions present during irradiation in liquid He at 4.96 K were nearly identical to those present during previous studies.[3] It was shown that the small fast neutron fluence (8.7×10^{20} n/m^2 > 0.1 MeV) accompanying the γ-ray dose had no detectable effect upon the mechanical strength of glass-filled epoxies. We expect the same result would be true in this experiment for the glass-filled polyimides. This is not to say that fast neutrons are unimportant. In many fusion reactor magnet locations, fast neutron fluences will greatly exceed those we have studied. The response of the strength of polyimide materials to fast neutron fluence remains as an important matter for new study.

As in earlier irradiations, the major sources of damage were gamma rays and energetic fission fragments. The latter are produced only in the glass-filled materials as a result of the fission of ^{10}B atoms (present as B_2O_3 in the glass) by capture of thermal neutrons that leak through the Cd shield. In our earlier report[3] we incorrectly stated the glass content in the composite epoxies as 33.7 wt %. We now know by reconfirmation from the manufacturers that the glass content was 66.3 wt %, and hence the energy deposition rate from ^{10}B fission was 1.25 times that due to gamma rays, and not 0.64 as was stated. In the present experiment the glass content of the Spauldite was 70 wt %, which is very close to that in the previously studied epoxies. In the case of Kerimid a value between 40-60% is given by the manufacturer, and, of course, Vespel has no glass. All dose values in the report are given only in terms of energy deposition by gamma rays, which is common to all the materials. It is important to note that while we can indicate the additional dose received by each material from ^{10}B fission, there is insufficient data to determine if energy deposition is a valid criterion for predicting property changes which are produced by radiations having different damage production mechanisms such as gamma rays and ^{10}B fission fragments. The following list gives the pertinent irradiation conditions:

gamma ray dose		100 MGy (10^{10} rads)
$\frac{^{10}\text{B fission dose}}{\text{gamma ray dose}}$	Vespel	0
	Spauldite	1.33
	Kerimid	0.95
Thermal Neutron Fluence		3.1×10^{21} n/m^2
Fast Neutron Fluence		8.7×10^{20} n/m^2 > 0.1 MeV
Irradiation Time		189 hrs

3.2.4.4 Flexure Test Results

The flexure-fracture-stress results are shown in Fig. 3.2.1 and compared with previous results for two glass-filled epoxies. Several features are noted:

1. In the unirradiated conditions the glass-filled Kerimid and Spauldite are respectively 2.2 and 3.1 times stronger at 78 K than the unfilled Vespel, and the best, Spauldite, has 90% of the strength of the glass-filled epoxies.

2. At 100 MGy (10^{10} rad) Vespel loses only 8%, Kerimid 30%, and Spauldite 38% of initial strength, but the latter remains slightly stronger than Kerimid. For this dose the glass-filled polyimides are at least 2.5 times stronger than the glass-filled epoxies, which after a dose 1/4 as large have insufficient strength for practical use.

3. The well-known temperature dependence of the strength of polyimides (increasing strength with decreasing temperature) is observed in both unirradiated and irradiated specimens. It is interesting to note, however, that the temperature dependence is notably reduced by irradiation as a result of the fact that the strength at 300 K is actually increased by irradiation.

Figure 3.2.2 shows the results obtained for the linear flexure modulus. It can be noted that for tests performed at 78 K Vespel shows a very slight increase in the modulus after irradiation, while the glass-filled polyimides show small decreases in modulus; even less than the epoxies. At 300 K Vespel shows no change, while both Spauldite and Kerimid show substantial increases with irradiation, giving modulus values close to those found at 78 K.

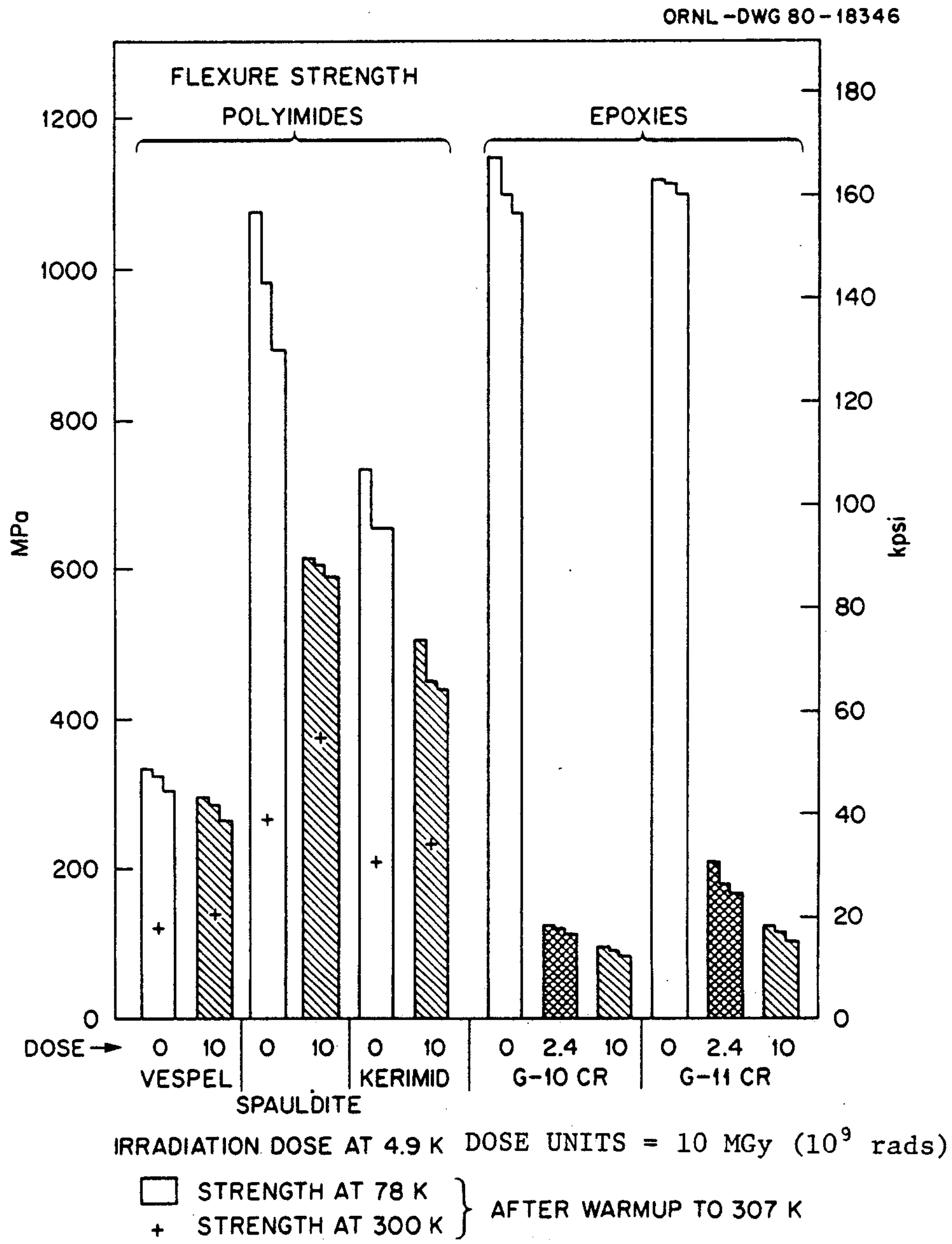


Fig. 3.2.1 Results of Flexure Fracture Tests

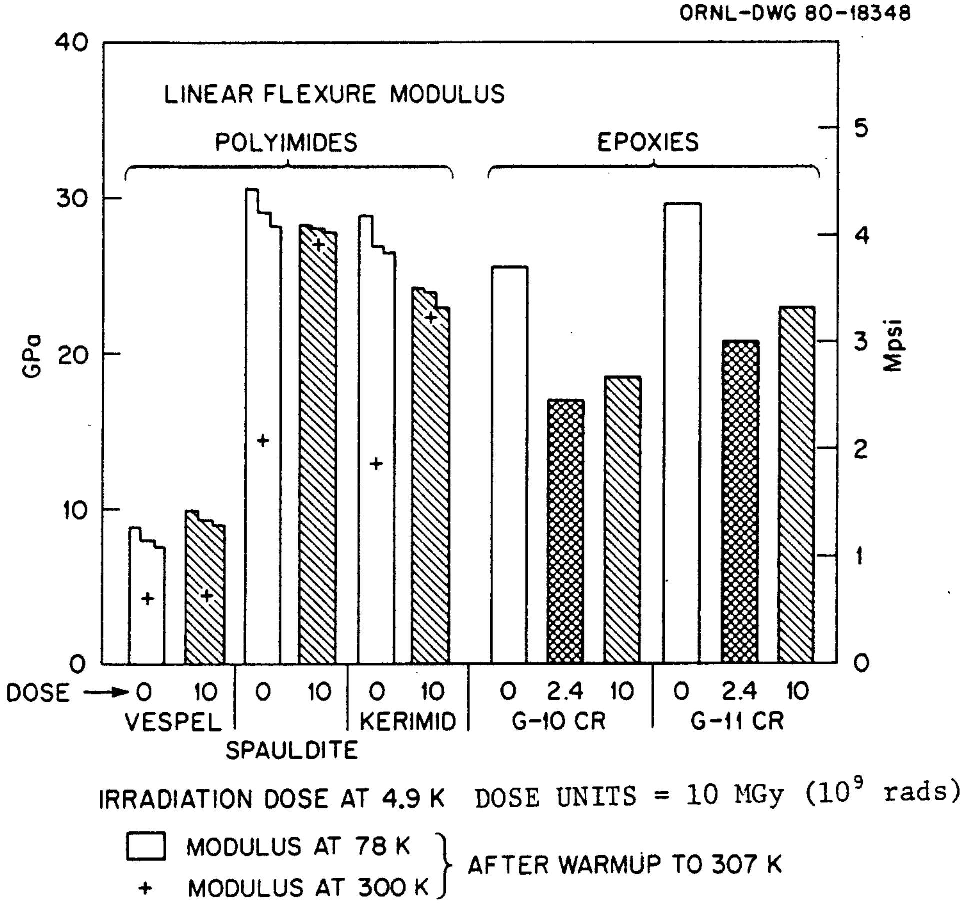
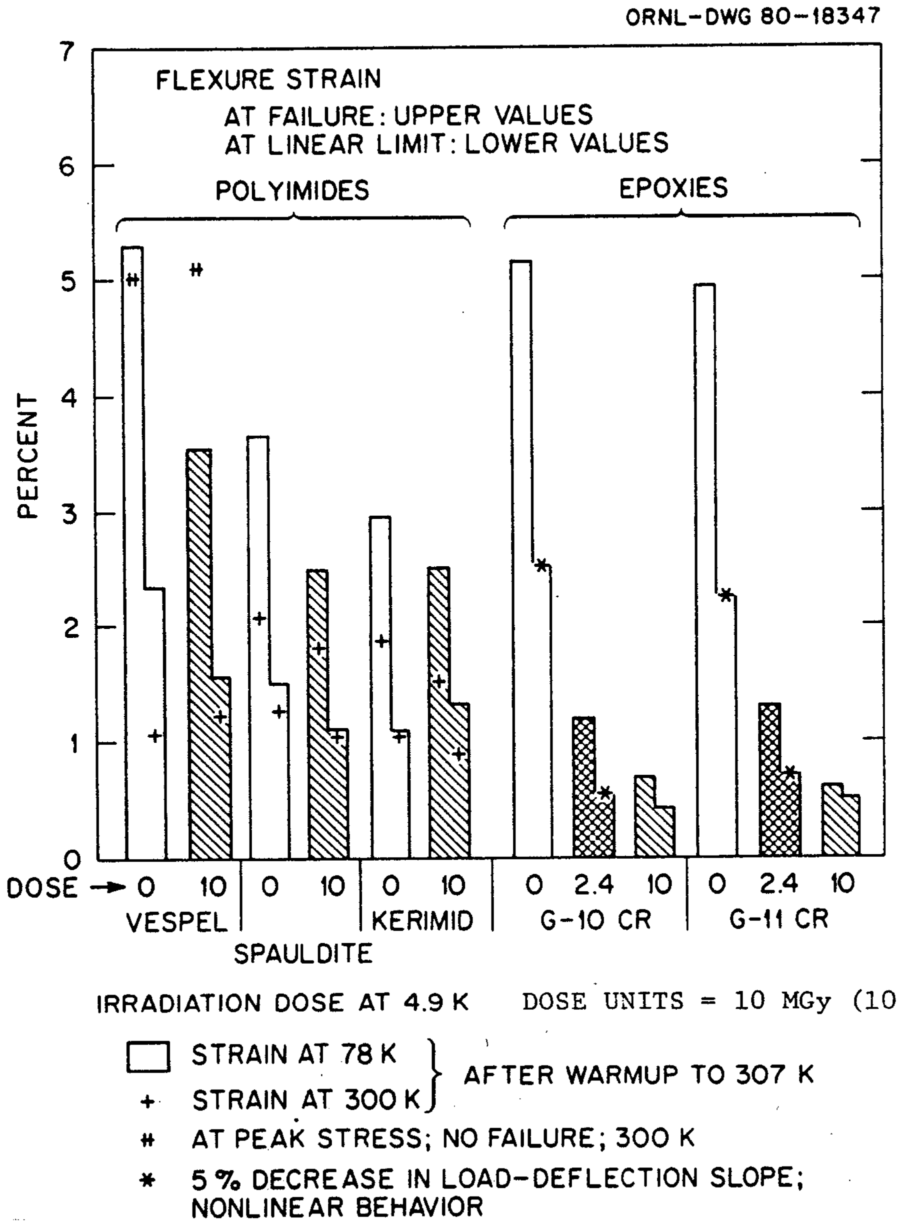


Fig. 3.2.2 Linear Flexure Modulus

Figure 3.2.3 shows the flexure strain results. Values were obtained for both the failure condition and for the transition point between linear and nonlinear load-elongation behavior. In a few cases at 300 K no failure was observed and the strain at peak stress is indicated. All of the materials except the unfilled Vespel gave results which correlated quite well with the fracture-stress results; this correlation is also found in the epoxy results.[3] Although Vespel showed some loss in strain at 78 K due to irradiation, it remained unchanged at 300 K. This result, along with the generally resilient characteristics observed during handling, gave the impression that Vespel may serve as a very radiation-resistant gasket material.



DOSE UNITS = 10 MGy (10^9 rads)

Fig. 3.2.3 Flexure Strain Results

3.2.4.5 Compression Test Results

In the case of compression tests Kerimid material was available to prepare specimens that could be loaded parallel (Kerimid ⊥) or perpendicular (Kerimid ⊥) to the glass-fiber laminations. The results of the tests seen in Fig. 3.2.4 show that, except for Kerimid ⊥, there is very close agreement with the flexure-fracture strength results.

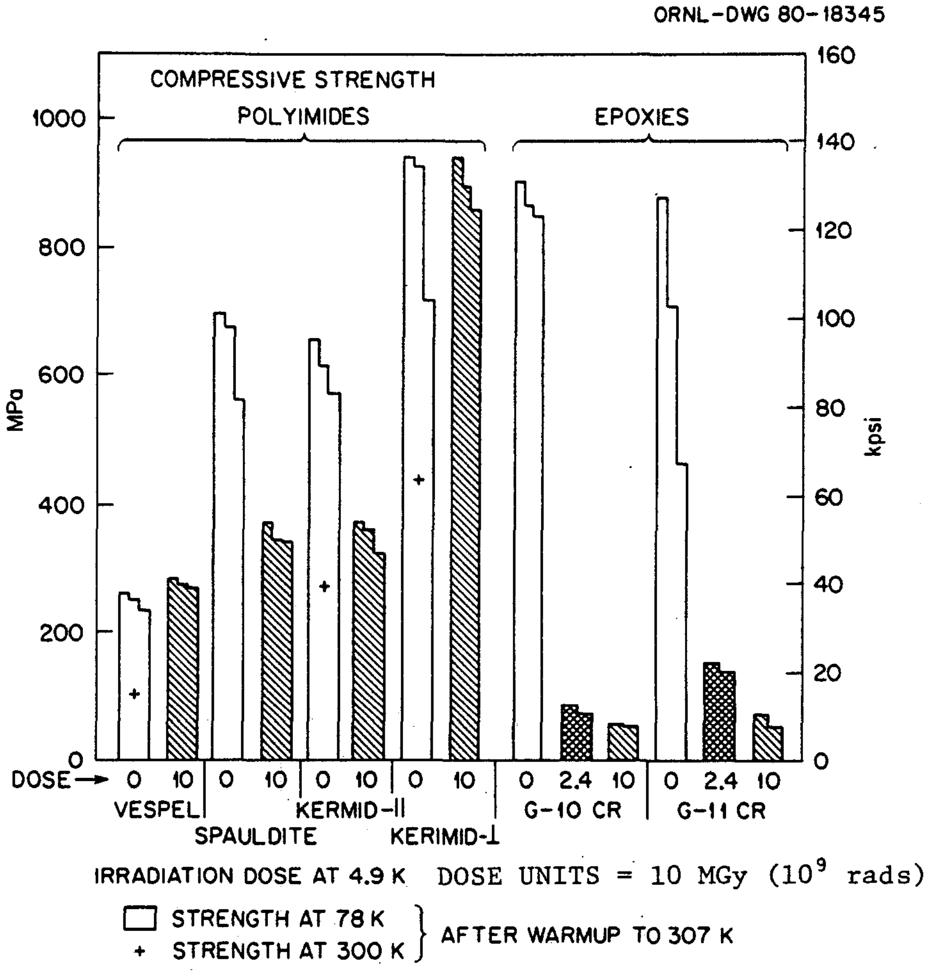


Fig. 3.2.4 Results of Compressive Strength Tests

In the case of Kerimid⊥ no loss in strength was observed for 100 MGy (10^{10} rads). While failure in all other glass-filled materials loaded in the parallel orientation was by delamination, that in Kerimid⊥ was by fracture, generally along an oblique plane that produced a fragment shorter than the sample length. Since in the application to fusion reactor magnets there are many locations where the principal stress on laminated insulation is perpendicular to the laminations, further investigation to higher irradiation doses is necessary to determine the radiation tolerance for this orientation.

3.2.4.6 Radioactivity

The radioactivity of the materials was monitored for some weeks following the irradiation. The results were obtained in the practical units of mrad/hr measured at the surface of a 75-mm-diameter paper-shell Cutie Pie (radiation-survey meter). Use was made of the flexure specimens for these measurements, since their stick-like shape was more suitable for accurate positioning parallel to the barrel of the meter. The maximum activity was 48 and 19 mrad/hr after one and four weeks respectively.

By comparing the results for the unfilled Vespel with Spauldite and Kerimid, it was clear that almost all of the radioactivity originates in the glass fibers. Further, the glass-filled polyimides give almost the same results as G-10CR, which is described as having nearly the same glass content (see Ref. 3).

3.2.4.7 Gas Evolution

The percentage weight composition of the gas mixture evolved from all samples during warm-up to room temperature after irradiation is given in Table 3.2.1.

It can be noted that the off-gas composition changes markedly with time after irradiation. The first gas sample shows a much larger proportion of H indicating its more rapid diffusion out of the material compared to the heavier species.

Table 3.2.1 Analysis of Evolved Gases (Wt %)

	Days After Irradiation	
	1	6
H_2	44.2	0.5
CH_4	0.8	1.4
H_2O	1.95	21.0
N_2 and CO	49.4	66.6
O_2	0.05	0.3
CO_2	2.9	9.1
C_2H_4	0.7	1.1

3.2.5 Conclusions

1. The glass-filled polyimide materials show an average loss in strength of ~35% after irradiation at 4.9 K to 100 MGy (10^{10} rads). While more data is needed to make a precise comparison, the present results indicate that the glass-filled polyimides are about 5-10 times more radiation resistant than the glass-filled epoxies.

2. In order to provide designers with information that gives the maximum flexibility in the choice of type and quantity of insulation needed, polyimide data should be provided on the response of strength to dose for values less than 100 MGy (10^{10} rads). Experiments in this range are now in progress. In contrast, there is also a need for further study at higher doses of the compressive strength measured perpendicular to laminations (see 3.2.4.5), in order to determine the radiation tolearance for this orientation.

3. In the present experiments ^{10}B fission fragment damage accompanies that produced by gamma rays. In terms of energy deposition these two types of damaging radiation are determined to be about equal. In his neutronics calculations for the Fusion Engineering Device (FED), previously called the Engineering Test Facility (ETF), B. A. Engholm[2] finds a corresponding situation at magnet locations where energy deposition produced by fast neutrons is about equal to that produced by gamma rays. At the present time, however, there is not enough data to determine if energy deposition is a valid criterion for predicting property