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CRYOGENIC RADIATION EFFECTS ON ELECTRIC INSULATORS*

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The radiation allowable at the superconducting magnets of a fusion reactor may be limited by the radiation tolerance of organic and composite electrical insulators. The allowable dose at the magnets will in turn influence the thickness of shielding necessary and therefore the size of the magnets. The response of organic substances to radiation at 4 K cannot be confidently predicted from irradiations at higher temperatures. Therefore, epoxy-based, polyamide, and polyimide insulators chosen from superconducting magnet practice were irradiated in liquid helium to a total gamma dose of 20 MGy (2 × 10^9 rd). Electrical resistivity was measured during and after irradiation and electrical breakdown was measured after irradiation. Mechanical properties measured at 77 K after irradiation included flexure strength, shear strength (of adhesives between copper), and compressive strength. Electrical properties were generally degraded, but the materials should still be useful as insulators. Mechanical strengths, however, were sufficiently decreased to require re-evaluation of some designs. Aluminized Mylar (superinsulation) was severely embrittled.

1. INTRODUCTION

Despite radiation shielding outside the blanket, the superconducting coils of a magnetic fusion power reactor will receive sufficient nuclear radiation to degrade some of the materials used in them [1,2]. The materials whose radiation tolerance may limit the allowable dose, and therefore set a minimum thickness for the shield, are the superconductor itself (Nb-Ti or Nb_3Sn), the stabilizer (Cu or Al), and organic electrical insulators (any of several polymers and composites). This paper describes current results in a program to measure the resistance of a number of technically significant insulation systems to irradiation in liquid helium, the environment in which the magnets will operate. Earlier results at a lower dose (2 MGy) have been reported [3]. Further details of the current experiment are found in ref. [4].

Considerable knowledge about the effects of radiation on polymers at room temperature has been accumulated [5,6]. Less is known about behavior at cryogenic temperatures, although some work has been done [7–11], especially at 20 K [7–9]. Our results show that the most radiation-resistant polymers and polymer-matrix composites appear to be degraded at or below the doses calculated [12,13] for the toroidal magnets of a power reactor over its lifetime. In general, radiation sensitivity in polymers does not seem to be appreciably affected by temperature, but it has been suggested [1] that different damage modes may operate at very low temperatures or during postirradiation annealing than during room-temperature irradiation.

2. EXPERIMENTAL PROCEDURE

The radiation used in this experiment was provided by the Bulk Shielding Reactor (BSR), a swimming-pool-type fission reactor at ORNL. Its core can be moved next to a cryostat at one side of the shielding pool; a heavy-water tank can be interposed to highly moderate the neutron spectrum. The cryostat is supplied with process liquid helium by a continuously operating refrigerator. The usable cold space is 25 mm diam by 200 mm high. In this experiment, a cadmium shield was used around the samples to absorb thermal neutrons and increase the gamma ray flux, which is by far the principal source of deposited energy. The materials irradiated and the tests used to evaluate them were chosen to make the most efficient use of limited cold space, to use prior knowledge about radiation effects, and to represent typical applications for insulators in superconducting magnets. The materials, which are shown in Table 1, included adhesives, rigid and flexible sheet materials, and films. The aluminized Mylar, while not an electrical insulator, was included because of its importance as thermal insulation for magnets. The tests are shown in Table 2. Controls were made at the same time as the irradiated samples and exposed to the same temperatures. Wherever possible, multiple tests were conducted. Results from the in-situ compression tests (in which materials were irradiated under load) and the

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Table 1
Materials irradiated

Material	Form	Polymer System	Manufacturer	Comments
Stycast 2850 ft blue	Adhesive	Filled epoxy	Emerson-Cumming	Stycast 2850 ft blue[a]
Epon 828	Adhesive	Filled epoxy	Shell	Epon 828[b]
G-10 CR	Rigid sheet	Glass/epoxy composite	Spaulding	High-pressure laminate
EF-527	Adhesive sheet	Glass/epoxy composite	Synthane-Taylor	B-staged; 1 h 175°C under pressure
Nomex 410	Flexible sheet	Aramid	DuPont	Paper
Kapton F	Film	Polyimide	DuPont	PTFE-coated
Superinsulation	Al-coated Mylar film	Polyamide	DuPont	Thermal insulation
Formvar	Varnish	Vinyl acetal	Westinghouse	25–30 μm on 18-AWG Cu
Omega	Varnish	Polyimide	Westinghouse	30 μm on 18-AWG Cu

[a] Stycast 2850 ft blue + 7% 24 LV hardener; cure 24 h, RT.

[b] Epon 828 + 20% Z curing agent + 0.5% Z 6020 Silane + 40 wt % 400-mesh SiO_2; 2 h 80°C + 2 h 150°C.

Table 2
Postirradiation tests performed

Test	Environment	Temperature (K)
Mechanical:		
Lap shear[a]	Nitrogen	77
3-point flexure[b]	Nitrogen	77
Compressive strength[c]	Nitrogen	77
In-situ compression[d] [~14 MPa (2000 psi)]	Nitrogen	77
Through-thickness compression	Nitrogen	77
Varnish scrape test	Air	300
Electrical:		
In-situ resistivity	Helium	4.9–300
Postirradiation resistivity	Dry nitrogen	300
Breakdown	Air	300
Dielectric loss factor	Air	300
Miscellaneous:		
Gas analysis[e]		300
Weight loss		
Visual observation		

[a] Between Cu substrates.

[b] 25.4 mm between supports.

[c] Cylinders 12 mm high by 6.1 mm diam.

[d] Load applied by Belleville washers.

[e] Gas collected in 4-K cold trap.

through-thickness compression tests were inconclusive and are not reported. Radiation-produced gaseous species were collected in a 4-K cold trap after warmup following each irradiation and 35 d after the second irradiation.

The irradiation schedule was similar to that used previously [3], but the total dose was 10 times as great. After specimen loading and cooldown, the reactor core was moved near the cryostat and brought to power. When the test materials had absorbed 6 MGy (6×10^8 rd) at 4.9 K, the irradiation was stopped and the samples were warmed to room temperature. Resistivities of appropriate specimens were measured, the cryostat was again cooled down, and the irradiation continued to a total dose of 20 MGy (2×10^9 rd) and a total irradiation time of 143 h. Because of the low irradiation temperature, dose rate effects are believed to be negligible. Based on the calculations of Santoro et al. [13], this dose corresponds to a first wall loading of about 8 MW y m^{-2} if penetrations are neglected and about 0.04 MW y m^{-2} if an unshielded penetration is immediately adjacent to the magnet. While these correlations are necessarily imprecise and subject to many variations in reactor design, they do demonstrate that the doses used are, at best, at the low end of those expected during a reactor lifetime. No thermal instabilities in the form of surges in sample chamber pressure or temperature were observed during either warmup.

3. RESULTS

Results of the mechanical property tests are shown in Figs. 1 and 2 and Table 3. Some materials were significantly degraded. All

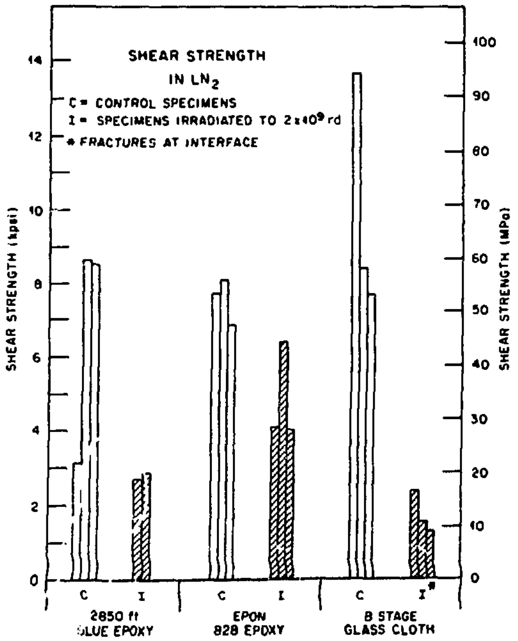

Fig. 1. Changes in shear strength due to irradiation at 4.9 K for 20 MGy (2 × 10^9) rd.

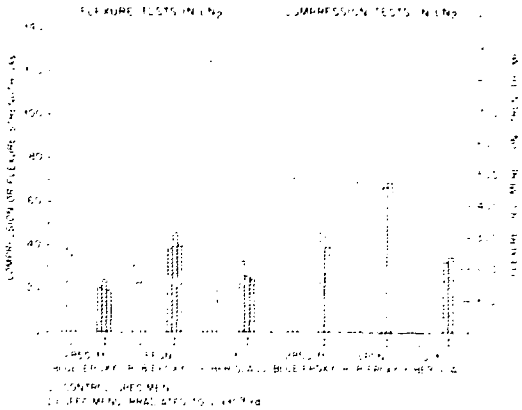

Fig. 2. Changes in flexure and compression strength due to irradiation at 4.9 K for 20 MGy (2 × 10^9 rd).

the materials tested are probably still strong enough to be usable but may require derating. The results of individual tests show the experimental variability. In some cases, behavior changed qualitatively as a result of irradiation. In the flexure test on G-10 CR, the control failed at the tension surface but the irradiated specimen failed at the compression surface. This is consistent with matrix (epoxy) degradation. The cylindrical compression specimens failed by the shear mode characteristic of brittle materials tested in this way.

Table 3
Mechanical property measurements

Material	Lap Shear Strength (MPa)		Flexure Strength (MPa)		Compressive Strength (MPa)	
	Individual Values	Mean	Individual Values	Mean	Individual Values	Mean
Stycast 2850:						
Control	22.8, 59.7, 58.9	46.8	259, 266, 238	254	500, 489	495
Irradiated	18.7, 19.8	19.2	137, 162, 132	143	322, 273	298
Epon 828:						
Control	53.4, 55.9, 47.4	52.2	207, 152, 315	225	511, 479	495
Irradiated	28.4, 44.3, 28.1	33.6	263, 315, 270	283	465, 483	474
G-10 CR:						
Control			921, 860, 805	862	602, 837^a	769^a
Irradiated			229, 176, 168	191	225, 237^a	231^a
EF 527:						
Control	94.4, 58.4, 53.2	68.7				
Irradiated	16.7, 10.6, 9.1	12.1				

^{a}G-10, not G-10 CR; rod tested in axial direction.

Electrical resistivities and breakdown potentials, shown in Tables 4 and 5, decreased slightly in several instances and severely in one. However, in no case are the postirradiation properties poor enough to reduce these materials' usefulness in magnet design. The lower values for resistivity in-situ than after irradiation reflect principally the difficulty of making electrical measurements in an environment of ionizing radiation. The data obtained to date suggest that a given materials' mechanical properties become unacceptable before its electrical properties.

Weight changes are shown in Table 6 with all sample types of each material combined. The noticeable weight loss in the Kapton F probably reflects damage in its PTFE (Teflon) coating.

Results of gas analyses taken as indicated earlier are shown in Table 7. Nitrogen and carbon monoxide are not distinguished by the analytical method. The decrease in the relative abundance of H_2 from after irradiation to 35 d later points to its more rapid diffusion rate out of specimen materials.

No quantitative tests were performed on the aluminized Mylar superinsulation. When removed from the sample chamber, it was obviously embrittled and broke into several pieces.

Table 4
Results of electrical measurements[a]

Material	In-Situ Resistivity (TΩ·m)		Postirradiation Resistivity (TΩ·m)		Breakdown (MV/m)		Dissipation Factor (tan δ) at 1 kHz	
	C	I	C	I	C	I	C	I
Stycast 2850	10	3.5	140	50	32[b]	32[b]	0.024	0.023
Epon 828	15	7.5	100	60	24[b]	23[b]	0.026	0.028
G-10 CR	7	5	70	70	22[b]	22[b]	0.022	0.024
EF 527			120	0.05	31	11	0.025	
Nomex 410	35	15	130	140	38	39	0.008	0.008
Kapton F			150	190	70[b]	57[b]	0.008	0.008

[a] C = control, I = irradiated.

[b] Surface flashover, not breakdown.

Table 5
Effects of radiation on wire varnishes

	Electric Breakdown (kV)	Scrape Test, Cycles to Fail	
		Average	Standard Deviation (%)
Formvar:			
Standard	5.9	32.0	9
Control	3.0	35.0	33
Irradiated	1.8	12.2	31
Omega:			
Standard	>11.8	50.0	35
Control	10.0	58.8	15
Irradiated	4.0	21.2	14

Table 6
Weight change as a result of irradiation

Material	Weight Change (%)	
	Control	Irradiated
Stycast 2850	+0.19	−0.19
Epon 828	+0.36	+0.15
G-10 CR	+0.16	+0.18
EF 527	+0.32	+0.70
Nomex 410	+1.9	+1.9
Kapton F	<0.1	−5.6

Table 7
Off-gas analyses

Gas	Content (%) after each dose in MGy (rd)		
	$6(6 \times 10^8)$	$20(2 \times 10^9)$	$20(2 \times 10^9)$[a]
H_2	58.9	93.7	35.8
CH_4	1.7	0.3	29.9
H_2O	21.7	3.2	3.1
N_2 + CO	9.0	1.4	18.2
O_2	0.2	<0.1	0.1
Ar	0.1	<0.1	0.05
CO_2	6.9	0.7	8.9
Organics[b]	1.4	~0.2	1.8
Fluorocarbons	0.2	~0.5	2.0

[a] Taken 35 d later.

[b] Heavier than methane up to molecular weight about 100.

4. DISCUSSION

Comparison of Tables 3 and 4 shows little correlation between degradation of mechanical and electrical behavior. Further, while several materials' electrical properties were degraded, even the as-irradiated properties appear to be good enough at this dose not to inhibit design. This is in contrast to mechanical strength, which in most of the materials dropped enough to require design revisions. For these materials, at least, it appears that the allowable radiation dose will be set by mechanical rather than electrical requirements.

The reduced surface electrical breakdown strength and the weight loss of the Kapton F, visual observations of the surface, and the presence of fluorocarbons in the off-gas lead one to conclude that the Teflon surface layer was significantly damaged. Kapton H (uncoated) is probably preferable for applications in which the radiation dose is appreciable.

Standard wire-coating tests [14] were performed on Westinghouse Formvar and Omega coatings by the Westinghouse Research and Development Center. Both materials were degraded in approximately the same proportion; the Omega's initial superiority appears to persist.

Because pumpout of the vacuum space and cooldown flex superinsulation, the usefulness of aluminized Mylar superinsulation at this dose is questionable.

The off-gas species detected are typical of those produced when polymers are irradiated. The total gas produced in a particular magnet could probably be calculated by using the weight losses of Table 6 and considering the amount of each insulation material present. Refrigeration designers must be prepared to cope with the release (during warmup) of a variety of gases in their helium and insulating vacuum systems.

5. CONCLUSIONS

1. Electrical behavior of the polymeric materials tested, which were taken from systems with good radiation resistance at room temperature, was degraded by a dose of 20 MGy in several instances but is still probably adequate for design.

2. In several instances, mechanical behavior of the materials tested was degraded by 20 MGy sufficiently to require derating for design.

3. The two wire coatings tested were degraded but are still usuable after 20 MGy.

4. At 20 MGy, the flexibility of aluminized Mylar superinsulation was sufficiently degraded that its usability is questionable.

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