Effect of gamma and neutron irradiation on the mechanical properties of Spectralon™ porous PTFE

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Abstract

We establish a correspondence between the mechanical properties (maximum load and failure elongation) of Spectralon™ porous PTFE irradiated with 14 MeV neutrons and 1.17 and 1.33 MeV gammas from a cobalt-60 source. From this correspondence we infer that the effects of neutrons and gammas on this material are approximately equivalent for a given absorbed dose.

Introduction

The National Ignition Facility (NIF) at the Lawrence Livermore National Laboratory (LLNL) was conceived as a means to achieve contained fusion of isotopes of hydrogen in an experimental setting up to an energy yield of approximately 20MJ, corresponding to a neutron yield of 7.1×10^{18} . Although yields of this magnitude have not yet been realized, materials used in the target chamber of the NIF are nevertheless exposed to appreciable fluences of gamma radiation and 14 MeV neutrons from fusion events that occur in lower-energy experiments. Of these materials, polymers, which are used as elastomeric seals, structural elements and electrical insulators among other applications, are at greatest risk of deterioration from exposure to this ionizing radiation. The effects of gamma irradiation [1,2,3,4,5,6], combined gamma and thermal (0.025eV) [7,8] or fast (>1 MeV)⁹ neutron irradiation, combined gamma and electron irradiation [10], electron [11], and ion irradiation [12,13,14] on a wide variety of polymers are well characterized, principally because of the ready availability of cobalt-60, nuclear reactor, and accelerator sources that can conveniently provide radiation doses up to 10^6 Gray (Gy) or more. The effects of 14 MeV neutrons alone on polymers, however, are not well characterized, because of the lack of controlled sources sufficiently intense to produce appreciable damage in an exposure period of acceptable duration.

In this paper we report our study of optical grade Spectralon™ [15] porous polytetrafluoroethylene (PTFE). Although PTFE is known to have very poor resistance to damage

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from ionizing radiation [16,17] large panels of Spectralon™ are used in a diffuse reflector assembly (“scatter plate”) in the NIF target chamber where their high reflectivity over a broad spectral range is essential to the performance of the near-backscatter imager (NBI) diagnostic instrument. The lifetime of the scatter plate in the presence of ionizing radiation is of great interest because the scatter plate is located only 4.1m from the center of the NIF Target Chamber and the Spectralon™ panels are difficult to replace. Apart from the practical need to estimate the lifetime of the panels, however, PTFE is a convenient material for study because its poor radiation resistance allows the effects of damage to be observed at neutron fluences and exposure times that can practically be achieved in available facilities under well controlled conditions. Our approach is to employ the maximum (failure) load and failure elongation as proxies for the internal state of and degree of radiation damage in the samples. Hence we assume that doses of neutrons and gammas that produce comparable sample failure loads and elongations are approximately equivalent with regard to the damage they produce.

Experimental

Tensile test samples of Spectralon™ were prepared as specified in ASTM D638-10 [18], Figure 1, Type V. This sample type was chosen because of its small size, which accommodated both the limited amount of material available and the equipment used for irradiation. The samples had nominal gauge lengths of 7.62 mm (0.3”) and gauge cross-sections of 3.38mm wide by 3.12mm thick (0.133” x 0.123”). The nominal material properties of Spectralon™ and Teflon™ PTFE [19] are given in Table 1.

Table 1: Nominal properties of Spectralon™ and fully dense Teflon™ PTFE

Material	Density (g/cm ³)	Hardness (Shore D)	Ultimate stress (psi)	Elongation at failure (%)
Spectralon™	1.25 – 1.5	20-30	891	91
Teflon™ PTFE	2.16	55	4,500	400

Gamma irradiation was performed in the cobalt-60 pool at LLNL. Samples were placed inside of a water-tight stainless steel container filled with either air or high-purity argon gas at 1 atm pressure that was lowered into a water-filled pool to a position adjacent to an intense cobalt-60 source. The tensile samples were arranged in the canister in such a manner as to assure that the centers of the gauge lengths received uniform, intense doses of 10, 100, 10³, 10⁴, and 10⁵ Gy. Exposure times for each dose were calculated using a dose rate of approximately 15.4 Gy/minute (1.54 krad/minute). Our experience is that PTFE has negligible strength and elongation for gamma doses appreciably larger than 10⁴ Gy when the irradiation is performed in air at 1atm pressure. In high purity argon, however, samples were irradiated to doses of 10⁴ and 10⁵ Gy because the ultimate loads and elongations were expected to be enhanced relative to samples irradiated in air because of the greatly reduced amount of oxygen.

Irradiations with 14 MeV neutrons at fluences of 10^9 , 10^{10} , 10^{11} , 10^{12} , and 10^{13} neutrons/cm² (n/cm²) were conducted in the SAMES facility [20] shown in Figure 1. SAMES operates by accelerating deuterium ions (D) and impacting them on a tritium (T) -containing target. The classical D-T reaction produces 14 MeV neutrons. The fluences of 14 MeV neutrons were uniform over a circle approximately 3cm in diameter centered between two tensile samples placed side-by-side, although only one sample was irradiated at the highest fluence. This positioning assured that the gauge lengths of the samples received uniform exposures. Unlike the experiments of Chabane et al. [21], our samples were located approximately 4 cm from the source. This close placement assured that the contribution of gammas and low-energy scattered neutrons to the overall dose (“room scatter”) was negligible. No shielding was used during the exposure. The different neutron fluences were obtained by using different neutron fluxes and exposure durations (Table 2). For this set of experiments, the neutron fluxes were adjusted by varying the accelerator current to minimize the uncertainties of the fluences deposited on the samples. A flux of 4.5×10^8 n/cm²/s ($10^{13}/22200$) is close to the maximum available at SAMES (10^9 n/cm²/s) and was therefore used to obtain the highest fluence. The flux was reduced incrementally to a value of 1.67×10^7 n/cm²/s ($10^9/60$) for the lowest fluence to ensure better control of both the run duration and the deposited fluence at each fluence level. The flux was approximately constant during each exposure. The target used to produce the 14 MeV neutrons was completely consumed during the six hours and ten minutes of exposure necessary to achieve the fluence of 10^{13} n/cm². A calibrated alpha particle counter, used to count particles emitted through the reaction $D + T \rightarrow n + \alpha$, allowed accurate on-line monitoring of the neutron beam.

Table 2: Neutron exposure durations in the SAMES facility

Fluence (n/cm²)	10^9	10^{10}	10^{11}	10^{12}	10^{13}
Duration (s)	60	80	240	2280	22200

The samples were pulled to failure at LLNL on an Instron™ Model 5500R test machine equipped with a 30,000 pound frame and a 0 – 2000 Newton (0 – 450 lbf) load cell. Elongations were measured by crosshead displacement, and the data was reduced using Instron™ Bluehill™ 2 software. The measurement accuracy specified for the test machine by the manufacturer is $\pm 0.5\%$ in both load and displacement. The maximum load occurred either at or just prior to failure in all cases. The failure elongation is the elongation just prior to failure.

Results

The maximum load and elongation at failure are shown as functions of gamma dose and neutron fluence in Figures 2 and 3 respectively. The data for the gamma-irradiated samples are averages of values from five tensile samples at each dose. The uncertainties in load and elongation are reported as 2σ . The data for the neutron-irradiated samples are averages of values from two tensile samples at each fluence except for 10^{13} n/cm² for which only one sample was available.

The reported uncertainties in load and elongation for the neutron-irradiated samples are 2σ estimates.

Data for tensile samples taken from six locations in the NBI scatter plate are also plotted in Figures 2 and 3. Each datum is the average of five measurements and the uncertainties are reported as 2σ . The total cumulative yield inside of the NIF target chamber while the NBI scatter plate was in place is estimated to be 6.7×10^{16} neutrons. Using this value, the total fluence of neutrons at the scatter plate can be estimated from the calculated [22] neutron fluence map in Figure 4 to be 7.5×10^{10} n/cm². This fluence is assumed to have been uniform over the entire scatter plate. The largest fraction (~60%) of this fluence is from neutrons scattered from the first wall steel panels that line the Target Chamber and the aluminum wall of the Target Chamber itself. The balance (~40%) is from 14 MeV neutrons generated at the center of the Target Chamber. The fluence of 14 MeV neutrons is $\sim 3.2 \times 10^{10}$ n/cm² and the corresponding dose is 0.65 Gy. The dose from the scattered neutrons is ~ 0.25 Gy and the dose from neutron generated gammas is ~ 0.2 Gy. Hence the total dose at the NBI scatter plate from all neutrons and gammas is ~ 1.1 Gy.

Maximum loads and failure elongations for Spectralon™ gamma-irradiated in air and high-purity argon are compared to data for fully dense PTFE in Figures 5 and 6. Each datum is the average of five measurements and the uncertainties are reported as 2σ .

The 2σ uncertainties for the gamma doses are 2-3% and for the neutron fluences approximately 5%. These values are too small to show legibly on the log scales used in Figures 2, 3, 5, and 6 and are not plotted. To accommodate the log scales used for gamma dose, the results for the sample that was not irradiated are plotted at a dose of 0.01 Gy (1 rad).

Discussion

Although the uncertainties in some of the measurements are substantial, the data for Spectralon™ gamma-irradiated in air shown in Figures 2 and 3 clearly show declines in both maximum load and failure elongation commencing at a dose of approximately 100 Gy. The data for the neutron-irradiated samples show declines in both maximum load and failure elongation commencing at fluences between 10^{12} and 10^{13} n/cm². If we assume that equivalent mechanical responses (maximum loads and failure elongations) indicate approximately equivalent degrees of material modification caused by irradiation, then registration of the x-axes for neutron fluence and gamma dose in Figures 2 and 3 suggests that a fluence of 14 MeV neutrons of 10^{13} n/cm² corresponds to a gamma dose of the order of 200 Gy and that the dose-to-fluence conversion factor for 14 MeV neutrons is approximately 5×10^{10} n/(cm²-Gy). We recognize that this registration depends upon measurements of the single sample irradiated at a neutron fluence of 10^{13} n/cm². In the absence of any other measurements at this fluence, we have applied the maximum uncertainties in load and elongation found for the other neutron-irradiated samples.

Even with this appreciable uncertainty the data suggest a real decline at a fluence of 10^{13} n/cm² and we consequently believe that the registration of the data sets is valid.

Calculations [22] of the energy deposited in Spectralon™ by 14 MeV neutrons indicate that a fluence of 10^{13} n/cm² is equivalent to a dose of approximately 200 Gy. The registration of the data sets therefore suggests that the effects of neutrons and gammas on Spectralon™ PTFE are approximately equivalent for a given absorbed dose. Although the damage mechanisms for neutrons and gammas are different (neutrons have mass and produce bond scissions by displacing atoms through collisions, whereas the massless high-energy photons of gammas are absorbed directly and produce bond scissions by extreme excitation of bonding electrons), our observations indicate that this difference does not have appreciable consequences when averaged over the millimeter lengths relevant to macroscopic strength and elongation measurements. At this length scale the net effect of either type of damage appears to be characterized principally by the energy deposited in the material per unit mass, not the details of the deposition mechanism.

The load and elongation of fully dense PTFE show the same behavior with gamma dose as Spectralon™, Figures 5 and 6. The observation that the deterioration of mechanical properties begins at doses of the order of 100 Gy in air is consistent with previous studies [23] and indicates to us that the correspondence that we have established between 14 MeV neutron fluence and gamma dose for Spectralon™ applies to fully-dense PTFE as well, as does the conversion factor of 5×10^{10} n/(cm²-Gy). Although the presence of 30-42% porosity greatly reduces the load and elongation of Spectralon™ relative to fully-dense PTFE, the bonds are the same in both materials and will, presumably, be affected by gammas and neutrons in the same way.

The load and elongation of samples of Spectralon™ and PTFE that were gamma-irradiated in high-purity argon are larger than for samples irradiated in air, as shown in Figures 5 and 6 for doses of 10^4 and 10^5 Gy (the load and elongation for samples irradiated to 10^5 Gy in air are so low that reliable measurements could not be made). We attribute this enhancement to the absence of oxygen in the argon. Oxygen reacts with bonds broken by irradiation thereby impeding bond repair and promoting material deterioration [24]. The effect is most pronounced for fully-dense PTFE, but is notable in Spectralon™ as well. At low gamma dose or neutron fluence the effect of the environment disappears, however, because damage levels are so low. Under these conditions of irradiation, therefore, we expect the loads and elongations of material irradiated in air, argon or vacuum to be comparable. The data shown in Figures 2 and 3 for the NBI scatter plate are consistent with this expectation. Although the variation among the six sample sets of maximum load taken from the scatter plate is considerable, the average of all of these measurements is 17.6 ± 6.6 lbf, which places it in the range of un-irradiated or mildly irradiated Spectralon™. We attribute the wide variation in mechanical properties of the NBI scatter plate to local variations in the porosity or bonding of the Spectralon™. We cannot, however, eliminate the possibility that such variations are the result of non-uniformities of irradiation of which we are unaware. Nevertheless, we conclude that the mechanical properties of the NBI scatter plate Spectralon™ have not yet been compromised by exposure to neutrons in

the NIF target chamber. Based on our data we believe that deterioration will likely begin when the cumulative neutron yield in the NIF target chamber reaches 10^{19} , corresponding to a neutron fluence at the NBI scatter plate of approximately 10^{13} n/cm².

Conclusions

Our observations suggest the following:

1. The effects of neutrons and gammas on Spectralon™ and fully-dense PTFE are approximately equivalent for a given absorbed dose.
2. For the purposes of estimating the effects of 14 MeV neutron- and gamma irradiation on the mechanical properties of Spectralon™ and fully-dense PTFE, a neutron fluence of 10^{13} n/cm² corresponds approximately to a gamma dose of 200 Gy and the dose-to-fluence conversion factor is approximately 5×10^{10} n/(cm²-Gy).
3. The maximum loads and failure elongations of Spectralon™ and fully-dense PTFE samples gamma-irradiated in a low-oxygen environment (high-purity argon) are enhanced relative to samples gamma-irradiated in air, although the effect is much less for Spectralon™ than for fully-dense PTFE.
4. The mechanical properties of the NBI scatter plate Spectralon™ have not yet been compromised by exposure to neutrons in the NIF target chamber but will likely begin to deteriorate when the cumulative neutron yield in the NIF target chamber reaches 10^{19} , corresponding to a neutron fluence at the NBI scatter plate of approximately 10^{13} n/cm².

Acknowledgments

The authors wish to thank John Moody for his interest in the condition of the NBI scatter plate, Jack Topper (LLNL) for providing the detectors used to measure fluences and O. Duhamel and M. Raine (CEA) for performing the neutron irradiations. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

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Figures

Figure 1: The SAMES facility used for neutron irradiations of Spectralon™ samples.

Figure 2: Maximum load of Spectralon™ as a function gamma- and neutron irradiation. To accommodate the logarithmic abscissa the data for material that was not irradiated is plotted at a dose of 0.01 Gy.

Figure 3: Failure elongation of Spectralon™ as a function gamma- and neutron irradiation. To accommodate the logarithmic abscissa the data for material that was not irradiated is plotted at a dose of 0.01 Gy.

Figure 4: Neutron fluence map of the NIF target chamber equator level for a yield at target chamber center (TCC) of 7.1×10^{18} neutrons. The resolution of the map is approximately 1ft (0.3m) and shows estimated neutron fluences in and around the NIF Target Bay for this yield. The position of the wall of the target chamber is indicated by the dashed line and the protrusions indicate the locations of diagnostic ports. Fluence varies linearly with yield so that fluences at any shot yield may be obtained by a simple scaling of the values shown in the map. The NBI scatter plate is located 4.1m from TCC, indicated by the black circle in the figure. For a yield of 7.1×10^{18} neutrons at TCC, the calculated fluence at this position is 8×10^{12} n/cm².

PTFE and Spectralon™ maximum loads in air and argon

Figure 5: Maximum loads of Spectralon™ and fully-dense PTFE as a function gamma irradiation. To accommodate the logarithmic abscissa the data for material that was not irradiated is plotted at a dose of 0.01 Gy.

PTFE and Spectralon™ in air and argon

Figure 6: Failure elongation of Spectralon™ and fully-dense PTFE as a function gamma irradiation. To accommodate the logarithmic abscissa the data for material that was not irradiated is plotted at a dose of 0.01 Gy.

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