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Journal of Nuclear Materials 304 (2002) 83–85

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Letter to the Editors

Irradiation performance of polytetrafluoroethylene (Teflon®) in a mixed fast neutron and gamma radiation field

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Received 17 December 2001; accepted 1 April 2002

Abstract

Samples of polytetrafluoroethylene have been irradiated with a mixed field of fast neutrons and gamma rays using the MIT Research Reactor. Dose levels from ~0.3 to ~50×10^6 Gy for gamma and from ~0.13 to 80×10^4 Gy for fast neutrons were used. Weight loss, fluorine loss, and swelling were measured quantitatively. Subjective mechanical property tests were also performed to assess embrittlement. Aside from high levels of embrittlement, no large changes, $\gtrsim$ 1.5%, were observed in the properties which were assayed even at the highest doses.

1. Introduction

Our primary motivation for irradiation testing of polytetrafluoroethylene (PTFE) (Teflon®) was related to its applications in neutron filters used to produce high purity epithermal neutron beams, 1 eV $\lesssim E_n \lesssim$ 10 keV, for neutron capture therapy applications [1–3]. The desirable neutronic properties for this application are primarily related to the fluorine content of the PTFE. High radiation doses from gamma rays and neutrons are expected in the filter application and it was desirable to demonstrate that the physical and chemical properties of the PTFE were sufficiently stable for this application. In particular, the swelling or shrinkage and the fluorine loss should be limited to a few percent. Tensile properties are not of great importance in this application as long as the material does not crumble.

Most previous irradiation performance studies of PTFE have focussed on physical, electrical and mechanical properties such as shear, tensile, elastic modulus, weight loss, gas evolution and electrical resistivity [4–9]. However, no results for irradiation-induced swelling or shrinkage appear to exist in the published literature. Fluorine loss as a function of radiation dose has been reported by Ryan [10]. In this paper we report post irradiation measurements of swelling, weight loss, fluorine loss and embrittlement for a range of doses from ~30×10^6 to ~5×10^9 Rads. The lower range of these doses overlaps the doses reached in previous measurements and the higher range significantly exceeds the doses used in Refs. [4–10]. Some of our results have previously been published in a symposium proceeding, which emphasized the neutronic performance of PTFE for epithermal neutron beam production [11]. The results presented here also include substantially improved fast neutron dosimetry.

2. Irradiation and post-irradiation testing

A series of irradiations of PTFE samples were carried out in the 5 MW, MIT Research Reactor. Samples were irradiated in mixed fields of fast neutrons and gamma rays to several dose levels. Bars of virgin grade Teflon®, about 0.6 cm square by 10 cm long were irradiated in a pneumatic tube irradiation facility near the MITR's core tank. The gamma dose rate was approximately 3.3×10^4 Gy/min while the fast neutron flux ($E > 1$ MeV) ranged from 3.9×10^{15} to 1.8×10^{16} n/m^2 s, corresponding to a

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0022-3115/02/$ - see front matter
PII: S0022-3115(02)00849-8

dose rate range of approximately 1.2×10^2–5.4×10^2 Gy/min. Sample temperature during irradiation was determined to be in the range of 93–107 °C by using temperature indicating waxes [12].

Post-irradiation testing was performed to determine the degree of swelling, weight changes, embrittlement and loss in fluorine content. To assess swelling the sample thickness was measured with a micrometer. Weight changes were determined with a sensitive balance. Embrittlement was subjectively assessed by physical tests, bending and cleaving. Assays of fluorine content were made by slow neutron activation analysis. Pre-irradiation and post-irradiation properties were assayed with each type of test.

3. Results

The results of the irradiation tests are summarized in Fig. 1 for four groups of samples which have been exposed to increasing levels of gamma and fast neutron dose/fluence. PTFE samples were, as expected, strongly embrittled even at the lowest doses used in these tests, approximately $\sim 3.4 \times 10^5$ Gy gamma and 0.25–1.2×10^{19} n/m^2, (0.1–0.6×10^4 Gy) fast neutrons. No dimensional changes, weight loss or fluorine loss is observed for this group of samples. The intermediate dose group samples received approximately 3.7×10^6 Gy gamma and 0.28–1.2×10^{20} n/m^2 (1.4–6.1×10^4 Gy) fast neutrons. These sample were brittle, had weight loss of $\sim 0.1\%$, exhibited shrinkage in the 0–0.3% range and had no measurable fluorine loss. The samples in the third group were exposed to gamma doses of approximately 1.6×10^7 Gy and 1.2–5.5×10^{20} n/m^2 (5.9–27×10^4 Gy) fast neutron dose. These samples were very brittle, swelled 0–0.1% and had a fluorine loss of $<1.0\%$. Weight loss measurements for this dose group were inconclusive due to the high probability of chipping the embrittled material during handling. At the fourth and highest dose a sample was exposed to approximately 4.8×10^7 Gy gamma and 1.6×10^{21} n/m^2 (79.5×10^4 Gy) of fast neutron dose. This sample was highly embrittled, exhibited swelling of $\sim 1\%$ and fluorine loss of $\lesssim 1.5\%$. Weight loss measurements were inconclusive due to sample chipping.

The results of these irradiation tests of Teflon® show the embrittlement which has been observed by other investigators. However, the current results also show that only small levels of swelling, $\sim 1\%$, and $\lesssim 1.5\%$ of fluorine loss are experienced at gamma and fast neutron doses as high as 4.8×10^7 Gy and 7.95×10^5 Gy respectively. The fluorine loss is consistent with the results reported in Ref. [10] and extends those results to 5× higher dose. Although very severe embrittlement was observed at the higher doses, the PTFE samples maintained their shape, were self-supporting and did not degrade to a powder. Therefore, PTFE (Teflon®) appears to have adequate physical and chemical stability for use in neutron filter applications. The rectangle in the middle left hand side of Fig. 1 shows the expected lifetime radiation exposure for a high intensity epithermal neutron beam currently in operation at the MIT Research Reactor. This shows that PTFE should perform satisfactorily in the neutron filter application

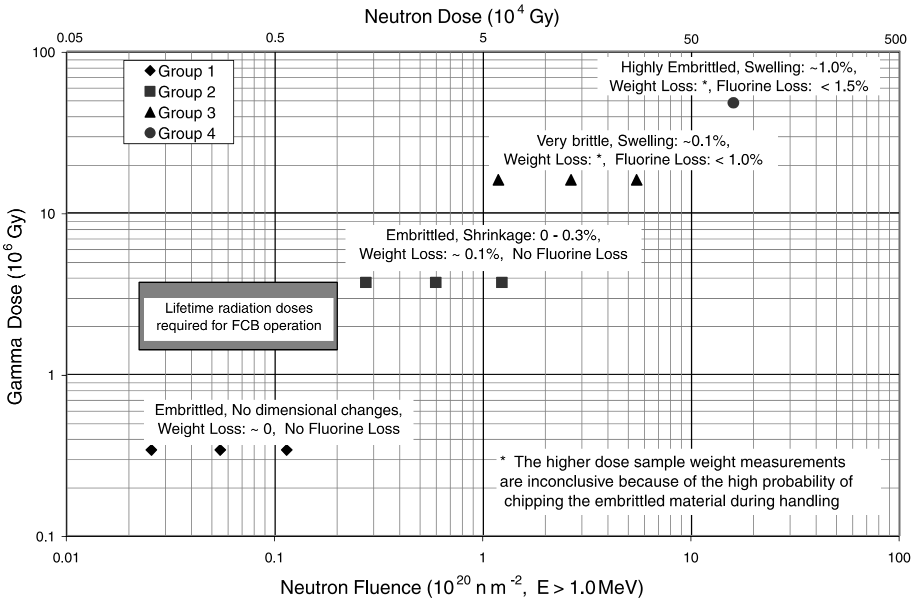

Fig. 1. Summary of the PTFE irradiation tests.

provided of course that the accelerated tests reported here produce representative results for a PTFE application where the dose rates are orders of magnitude lower than the dose rates used in the tests reported here. Funding for this project was provided by the US Department of Energy, Office of Science under contract numbers DE-FG02-97ER62489 for FCB construction and DE-FG02-96ER62193 for facility design activities.

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