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EFFECTS OF TRITIUM IN ELASTOMERS

by

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EFFECTS OF TRITIUM IN ELASTOMERS*

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ABSTRACT

Elastomers are used as flange gaskets in the piping system of the Savannah River Plant tritium facilities. A number of elastomers is being examined to identify those compounds more radiation-resistant than the currently specified Buna-N rubber and to study the mechanism of tritium radiation damage. Radiation resistance is evaluated by compression set tests on specimens exposed to about 1 atm tritium for several months. Initial results show that ethylene-propylene rubber and three fluoroelastomers are superior to Buna-N. Off-gassing measurements and autoradiography show that retained surface absorption of tritium varies by more than an order of magnitude among the different elastomer compounds. Therefore, tritium solubility and/or exchange may have a role in addition to that of chemical structure in the damage process. Ongoing studies of the mechanism of radiation damage include 1) tritium absorption kinetics, 2) mass spectroscopy of radiolytic products, and 3) infrared spectroscopy.

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THE EFFECTS OF TRITIUM IN ELASTOMERS

Polymeric materials serve as seals and packings in the Savannah River Plant tritium separation facility. In particular, nitrile (also called buna-n) rubber is specified for flange gaskets. The beta radiation resulting from decay of tritium to helium-3 causes this, and many other elastomers, to become hard and brittle. To study the effects of tritium on elastomers and to identify compounds with high beta radiation resistance, nine elastomer compounds were exposed to 83 kPa (620 torr) of tritium gas for 125 days. All compounds underwent significant radiation damage. Results of compression set measurements show that ethylene propylene rubber and Kalrez® (Du Pont) fluoroelastomer are the most radiation-resistant of the compounds tested. Both are superior to the tested nitrile compound.

BACKGROUND

The degradation by radiation of the useful mechanical properties of elastomers is more severe than the degradation of the properties of any other group of engineering materials.¹ Much research on the response of elastomers to cobalt-60 gamma radiation has shown that a dose of 10^8 rad renders most of the materials unusable. However, the sealing of fluid-handling components usually requires

elastomers, even in radiation environments. Some elastomers, through their polymer chemistry or formulation, show significantly higher resistance to gamma radiation than do others. Therefore, a program is in progress at the Savannah River Laboratory to identify elastomers with high tritium beta radiation resistance and to investigate the mechanism and effects of beta radiation damage.

Damage to polymers from any high-energy radiation proceeds from the ionization of atoms and consequent severing of chemical bonds. What typically distinguishes damage from different sources of radiation is the distance that the irradiating particle or ray can travel in the polymer and the amount of energy deposited per unit path length. Tritium damage is particularly distinguished by the ability of tritium to dissolve into the polymer component and exchange with the hydrogen atoms of the polymer molecules. Since the mean free path of a tritium beta particle with average kinetic energy is less than 1 mm in gas, radiation damage occurs where the tritium atom is located. Therefore, the solubility of tritium in a material and the tendency for exchange with hydrogen atoms can strongly influence the amount and distribution of the radiation damage.

The evaluation of elastomers for flange gasket service was made with ASTM Standard Test method D2309-68 "Compression Set Induced by Nuclear Radiation." When compressive displacement is applied to a specimen, compression set is defined as the percentage

not recovered 30 minutes after the removal of the compressive load. Although stress relaxation of a compressed elastomer specimen directly measures its utility as a sealing material, it is a more difficult measurement than compression set. It has been shown in gamma-radiation damage studies of O-ring materials that compression set and stress relaxation measurements correlate well.² In Reference 2, a compression set value of 75% or more in the test environment was interpreted as indicative of seal failure. Because elastomeric seals may actually hold with a higher compression set, compression set values are more properly used as indicators of relative response to an environment rather than as predictions of absolute performance.

Experimental Procedure

The elastomers tested are listed in Table 1. All but Kalrez[®] were obtained from Parker Seal Group of Parker-Hannifin Corp., and the appropriate Parker compound numbers are also listed. The cylindrical specimens are specifically prepared for compression set tests; they measured 1.12 in. in diameter and 0.50 in. thick. They were compressed in aluminum fixtures to 0.375 in. thick. Irradiated specimens were exposed to a gas mixture of 93% T and balance D and ³He at a pressure of 83 kPa (620 torr) for 125 days. Control specimens were stored in air at room temperature. The compression set measurements were made two months after the exposure to allow

sufficient tritium off-gassing for safe handling of the specimens. Tritium desorption was measured with a flowing air Kanne monitor, and autoradiographic exposures of the tritiated specimens were made.

Results and Discussion

The results of the compression set measurements are shown in Table 1. The table lists the total compression set and the radiation-induced compression set. The latter values were obtained by subtracting the compression set values measured on the appropriate control specimens. The elastomers are listed in order of total compression set, since it reflects the of the response elastomers to both time and environment. Ethylene propylene (E740-75) has the lowest total compression set and third lowest radiation-induced compression set. Although Kalrez® shows a lower radiation-induced compression set than does ethylene propylene, it is a much more expensive compound. Viton B® (Du Pont), a fluoroelastomer like Kalrez®, also shows good radiation performance. In contrast, nitrile rubber shows poor resistance to tritium beta radiation as measured by compression set. Because the elastomers with the highest compression set values are essentially damaged to the maximum extent, the difference between the best and worst performer may be underrepresented by the present data. Future testing will be of shorter duration (<100 days) and may reveal a wider difference.

During this 125-day exposure about 3.2×10^{-3} moles of atomic tritium decayed, with a release of about 1.7×10^{11} rad of energy. Mass spectrographic analysis at the end of the exposure showed exposure gas to be 61% T₂ and 33% H₂, indicating substantial solution of tritium into the specimens and release of hydrogen from them. The mass spectrographic data suggest therefore that about one-third of the energy of decay was deposited within the specimens. A relative indication of the solubility of tritium in particular specimens and thus the radiation dose to the specimen is given by tritium off-gassing measurements (Table 2). A comparison of Tables 1 and 2 suggests that ethylene propylene may have a lower compression set because it received a lower radiation dose. However, only a very weak correlation could be drawn between compression set values and off-gassing rates for the present data.

The tritium off-gassing measurements do correlate with autoradiographs of the specimens (Figure 1). The ethylene propylene specimens produced much less darkening of a plate than the silicone or urethane specimens. It can be seen in Figure 1 that the darkening from tritium decay is localized at the edge of the specimen. During exposure the compression set fixture covers the flat surfaces of the cylindrical specimen, so that those surfaces are not exposed to tritium gas. However, the diffusion coefficient for hydrogen in elastomers is high enough to produce a homogeneous distribution of tritium in a small specimen. The distribution seen in

the autoradiographic images suggests some surface or near-surface radiolytic reaction prohibits the expected homogeneous distribution.

REFERENCES

1. R. W. King et al. "Polymers" **Effects of Radiation on Materials and Components**, ed. by. J. F. Kircher and R. E. Bowman, Reinhold, New York (1964).
2. R. G. Wensel. "Resistance of O-Ring Seal Compounds to Heat and Ionizing Radiation in Air and in Water." Report AECL-5697, Atomic Energy of Canada Limited, Chalk River Nuclear Laboratories, Chalk River, Ontario (1977).

TABLE 1

Compression Set Values*

<u>Elastomer</u>	<u>Total Compression Set, %</u>	<u>Radiation- Induced Compression Set, %</u>
Ethylene Propylene (E740-75)	52	44
Styrene Butadiene (G244-70)	75	68
Kalrez® 1018	83	34
Viton® B (V494-70)	88	42
Viton® E60C (V747-75)	93	69
Ethylene Acrylic (A936-70)	93	77
Nitrile (N741-75)	97	90
Urethane (P642-70)	98	77
Silicone (S604-70)	99	90

* Experimental error of +3 percentage points.

TABLE 2

Tritium Offgassing Rates

<u>Elastomer</u>	<u>Rate in cm³(STP)/sec x 10⁷</u>
Ethylene Propylene	3.6
Styrene Butadiene	9.1
Viton® B	11
Kalrez®	11
Nitrile	14
Viton® E60C	20
Ethylene Acrylic	39
Silicone	120
Urethane	140

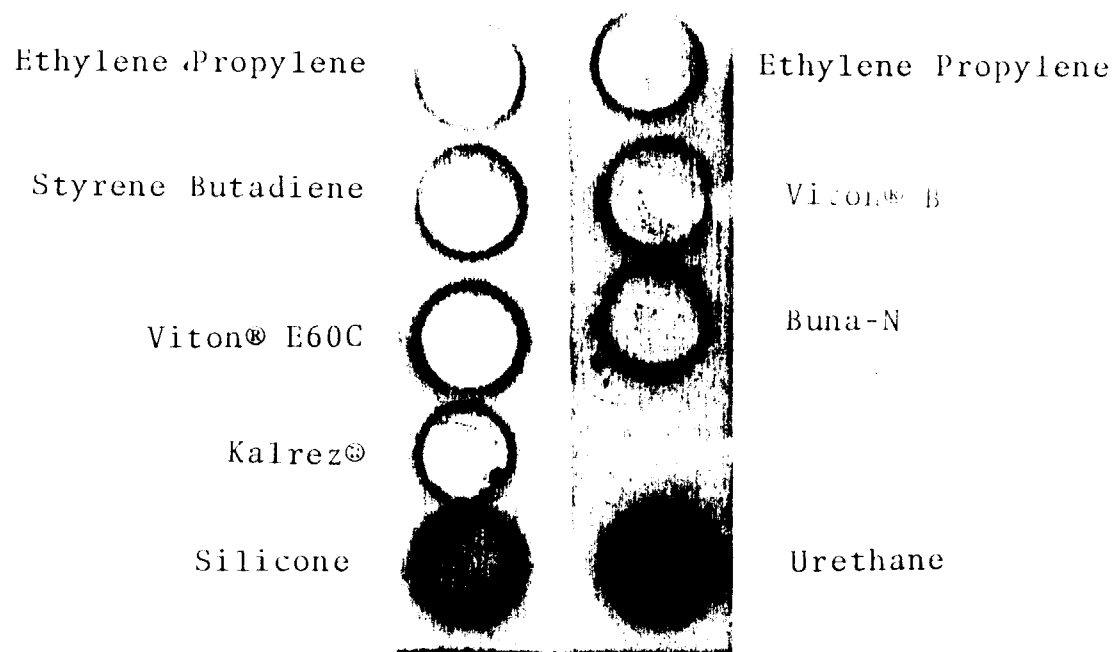


Figure 1. Autoradiograph of Tritium-Exposed Specimens

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Attached is a copy of the following:

DP-MS-82-54, "The Effects of Tritium in Elastomers" by P. E. Zapp, G. L. Tuer, and R. G. Derrick.

This paper is proposed for presentation at the Tenth Annual DOE Compatibility Meeting to be held in Albuquerque, New Mexico on October 26-29, 1982 and for publication in the Proceedings.

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