O-ring Lifetime of the SAVY-4000 Nuclear Material Storage Container

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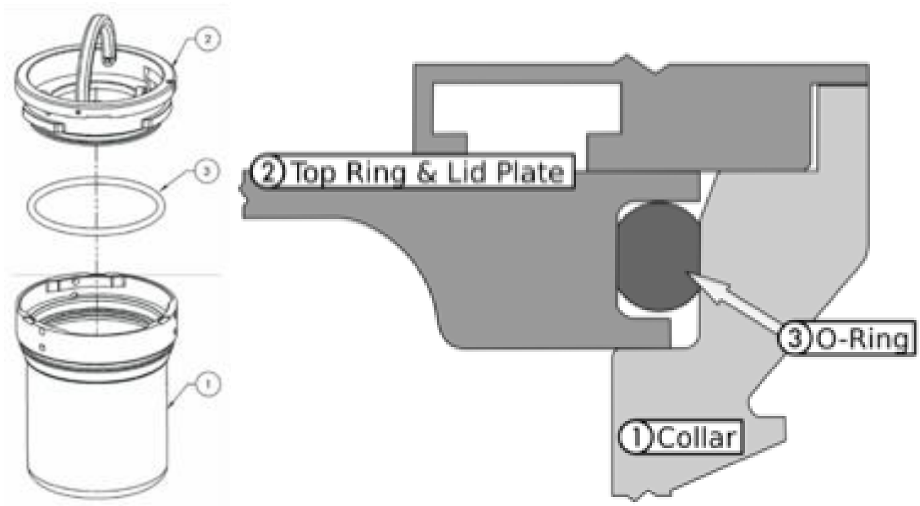
1. Abstract

The typical use conditions of rubber O-ring plays at least as important a role in the effectiveness of the seal they form as the physical properties of the O-rings themselves. Under normal use conditions, O-rings are subject to wear, environmental contaminants such as hair, dirt and metal filings, and mishandling by the users. We systematically examine how each of these factors impacts the leak-tightness of a nuclear material storage container, and the likelihood that any one of these factors will allow the inadvertent release of radioactive material.

2. Introduction

The motivation for this project begins with DOE M 441.1-1, the Nuclear Material Packaging Manual, issued in March of 2008. The manual details design requirements for nuclear material storage containers, and covers numerous aspects of the expected performance of the containers. In their implementation of DOE M441.1-1, Los Alamos National Laboratory (LANL) and Nuclear Filter Technology, Inc. (Nuc-Fil) developed the SAVY-4000 container.

The SAVY-4000 design includes several sizes of container, ranging from 1-quart up to 10-gallon, and all feature a filtered vent in the lid to allow for gas exchange and pressure equalization. Although this is a vented container, a seal is needed between the lid and body of the container to prevent any particulate matter that may be inside the container from escaping and possibly exposing workers. In the SAVY-4000, that seal is provided by a fluoropolymer O-ring that is compressed between a piston groove in the lid and the inside of a collar on the body of the container, as shown in figure 1. The SAVY-4000 Safety Analysis Report includes a calculation of the expected dose to a worker given a release of particulate material from a container in the worst-case, and concludes that to ensure worker safety, leaks from the container must be lower than 1×10^{-5} atm $cm^3\ s^{-1}$ of helium into vacuum.



(a) *(b)*

Figure 1. (a) Exploded view of a SAVY-4000 container, with the body (a1), lid, (a2) and O-ring (a3) shown. (b) Cross-section of an assembled container.

We are, of course, concerned with the service lifetime of a container, and as the contianers are opened and closed over many years of service, we expect the O-ring to be one of the lifetime-limiting components, especially because they will experience elevated temperatures and ionizing radiation while in use. To study the effects of radiation, temperature and compression on the O-rings, and to determine an expected lifetime for the O-rings, we developed and implemented an accelerated aging plan. The aging studies detailed in that plan involved several experiments. One, was to hold segments of O-rings under compression at a range of higher temperatures, and to check the effect that had on compression set and hardness as measured by durometer. Another was to hold whole assembled containers at higher temperatures and periodically check them for leaks. A third study examined the combined effects of elevated temperature and ionizing radiation on O-ring segments under compression and whole O-rings in assembled containers.

Several other studies geared toward establishing the mechanism of the O-rings' degradation in performance, such as a study of the O-rings' consumption of oxygen as a function of temperature, and a study of changes in the mean molecular weight of the cross-linked polymer of the O-rings by means of solvent swelling, and in the spectroscopic changes observed in aging O-ring pieces, were also conducted, but not detailed here.

3. Whole-Container Thermal Aging Study

In order to correlate a leak rate failure with other diagnostic tests, O-rings were installed in 1- Quart SAVY containers that were then placed in aging ovens. Leak tests were conducted with a frequency of between 2 weeks and 6 months during a 9-month period. The containers passed each leak test conducted immediately after oven storage, as shown in Table 1.

Table 1. Estimated compression set and leak test result of the whole-container aged O-rings.

Aging Temperature	Compression Set	Leak Test Result Before O- ring Removal	Leak Test Result Immediately After O-ring Removal	Leak Test after O-ring Relaxation
210°C	93% ± 19%	Pass	Fail	Fail
160°C	13% ± 20%	Pass	Pass	NA
120°C	36% ± 14%	Pass	Pass	NA
90°C	18% ± 13%	Pass	Pass	NA
70°C	21% ± 8%	Pass	Pass	NA

The compression set was estimated for each of the O-ring in this experiment. The initial thickness was assumed to be the nominal O-ring thickness with the uncertainty taken as the manufacturer's tolerance, 0.210 ± 0.005 inches. The compression thickness was taken as the nominal gland depth and tolerance from the SAVY-4000 1-Qt engineering drawings, 0.170 ± 0.003 inches. The thickness of each O-ring was measured with a laser micrometer, as the average of eight measurements from around the O-ring. The 210°C O-ring was re- measured and leak tested again after two weeks, and found to fail the leak test and to have maintained a compression set. Since the 210°C O-ring maintained a compression set of up to 88 ± 20% for at least two weeks, and still failed a leak test, the compression set limit was conservatively set to 65% for other aging determinations.

4. Compression Set Study

Compression set measurements for O-ring pieces aged under compression at temperatures between 70 – 210°C were taken. For each aging condition, three O-ring pieces were measured. Only a small number of samples in the accelerated aging study show compression set values above the 0.65 limit, and those were aged at the highest temperatures (190 and 210°C).

A statistical model was developed to use the compression set data to predict the lifetime of the O-ring based on knowledge of the temperatures of aging, the aging times and the 65% compression set limit. A graphical depiction of the predicted compression set over long times is shown in figure 2.

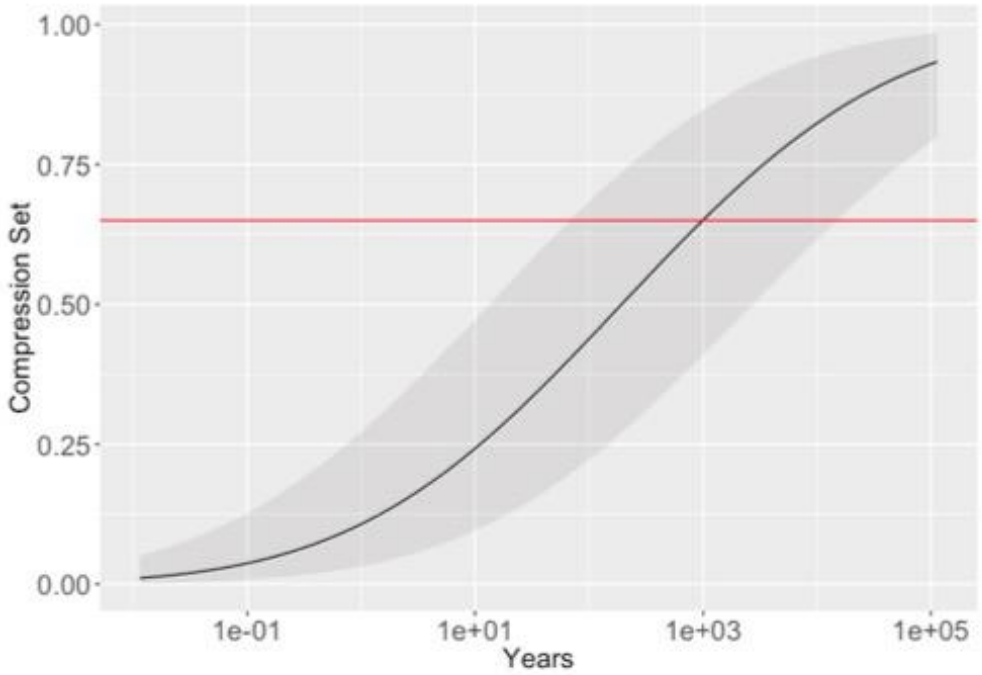


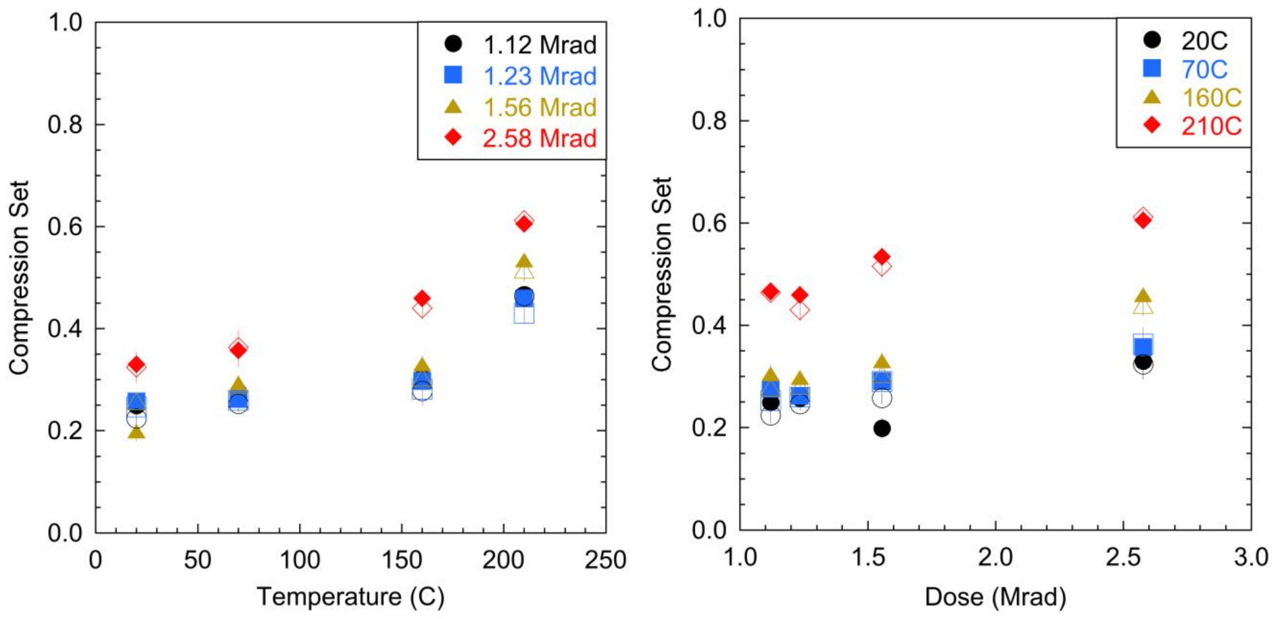
Figure 2 Predicted compression set at 80°C over long times, shown in black, with the failure threshold of 0.65 compression set shown in red.

The compression set experiments predict that the O-rings will begin to leak after about 993 years, in the range of 68 to 15,000 years at the 95% confidence interval.

5. Synergistic Effects: Accelerated Aging with Radiation and Temperature

All results described above pertain to thermal aging alone, but we also needed to understand how the O-ring behaves when subjected to radiation and elevated temperature. The synergistic effects experiments were conducted at the Gamma Irradiation Facility (GIF) at Sandia National Laboratory in Albuquerque. Samples of compressed O-rings accumulated 1.12, 1.23, 1.56, or 2.58 Mrad, while being held at an elevated temperature of 20, 70, 160, or 210°C. Whole O-rings in SAVY-4000 containers were exposed to the same schedule of irradiation and high-temperature. There was some interest in studying aging affects on samples that had been pre-heated for a period of time as well as virgin samples, and the pre-heating is referred to as annealing.

Figure 3 shows the compression set measured as a function of aging temperature and dose, respectively, with open markers for annealed samples and filled markers for unannealed samples. Both factors cause an increase in compression set, but no difference can be observed between annealed and unannealed samples.



(a) *(b)*

Figure 3. Compression set for O-ring pieces as a function of (a) temperature and (b) total dose for samples exposed to both elevated temperature and radiation at the GIF. Filled markers correspond to unannealed samples, while open markers are for annealed samples.

To determine if there is a synergistic effect between temperature and radiation with respect to compression set, a two-way analysis of variance (ANOVA) was performed on the data from the unannealed experiments and from the annealed experiments separately. For these analyses, temperature and dose are the factors and compression set is the response; the results are reproduced in Tables 4.5 and 4.6. In these tables, the values for “Sum_sq,” “F,” and “$\boldsymbol{\omega}^2$” can be used to determine if a factor (temperature or dose) has an effect on the response (compression set); for each of these, higher values indicate greater effects. “Sum_sq” is the sum, over all observations, of the squared difference of each observation from the overall mean. “F” compares the sum of squared values to the "Residual," which represents the error (or noise) in the model. "**ω^2**" estimates the effect size by calculating the variance in the response; a value of zero indicates that the factor does not affect the response. "Df" represents the degree of freedom (or the number of values that are independent from each other), while "p" compares the data for a given factor to the "Residual." A p-value of 0.01 corresponds to a 99% confidence range.

For the unannealed samples, the ANOVA shows a significant effect for temperature, dose and the combined effects of temperature and dose on the compression set attained during each experiment, because the p-value for each is much smaller than the typical 0.05 value. The effect size, conservatively estimated as ω^2, however, shows that the temperature has a greater effect on the compression set than the dose. Moreover, the independent effects of temperature and dose are greater than the synergistic effect of both.

Table 4.6. ANOVA for compression set on unannealed O-ring pieces.

	Sum_sq	**Df**	**F**	**p**	**ω^2**
Temperature	1.4013	3.0	414.7551	1.0×10^{-65}	0.7059
Dose	0.3666	3.0	108.5173	5.3×10^{-35}	0.1834
Temp:Dose	0.0670	9.0	6.6106	1.0×10^{-7}	0.0287
Residual	0.1442	128.0			

For the annealed samples, the significance of the effect is about the same as for the unannealed samples, as is the effect size. The p-value for the synergistic effect is much larger than the rest, though it is still below the typical 0.05 cutoff.

Given that there is a weak synergistic effect, we can find an equation describing the relationship between compression set C_B (as a fraction), temperature (T in °C) and dose (D in Mrad) by performing a non-linear least-squares fit of the function below to the experimental data:

$$C_B = F(T,D) = a + b(T) + c(D) + d(T \times D)$$

For unannealed samples, the coefficients are $a = 0.12 \pm 0.03$, $b = 0.0008 \pm 0.0002$, $c = 0.06 \pm 0.02$, and $d = 0.0003 \pm 0.0001$. For the annealed samples, the coefficients are $a = 0.11 \pm 0.03$, $b = 0.0007 \pm 0.0002$, $c = 0.06 \pm 0.02$, and $d = 0.0003 \pm 0.0001$. There are no substantial differences between the annealed and unannealed results.

6. Conclusions

Overall, the lifetime extension program for the SAVY-4000 O-rings predicts a lifetime greater than 40 years, with no special interaction between temperature and radiation that limits the life of the O-ring.

7. References

1) DOE, “Nuclear Material Packaging Manual.” **DOE M 441.1-1,** https://www.directives.doe.gov/directives/current-directives/441.1-DManual-1/view

2) Smith, P.H.; Stone, T.A.; Veirs, D.K.; Anderson, L.L.; Blair, M.W.; Hamilton, J.; Kelly, E.; Moore, M.E.; Teague, J.G.; Weis, E.; Yarbro, T.F., DOE/NNSA review, “Safety analysis report for the SAVY 4000 container series” (2012).

3) Gladson, J.; Hagan, R., “Nuclear Material Container Testing,” Los Alamos National Laboratory (1999).

4) Weis, E.M., Blair, M.W., Hill, B.D., Stone, T.A., Smith, P.H., Winter, J.C., Reeves, K.P., and Veirs, D.K., “Durability and field condition study of seal of SAVY-4000

storage container.” (2014), Packaging, Transport, Storage & Security of Radioactive Material **25(1):** 24-9. 10.1179/1746510914y.0000000059

5) Blair, M.W., Weis, E., Veirs, D.K., Smith, P.H., Stone, T.A., and Ball, J.M., Service Life Prediction of Polymeric Materials: Vision for the Future ; 2013-03-03 - 2013-03-08 ; Monterrey, California, United States, “Accelerated Aging Studies for the Lifetime Extension of O-rings Used in the SAVY-4000 Unit,” (2013).

6) Weis, E.M., Welch, C.F., Smith, P.H., Blair, M.W.; Stone, T.A.; Veirs, D.K.; Reeves, K.P.; Karns, T.; Oka, J.M.; Keller, J.; Meincke, L.; Torres, J.A.; Herman, M.J.; Weaver, B.; Adams, J.C.; Trautschold, O.C., “Lifetime Extension Report: Progress on the SAVY-4000 Lifetime Extension Program,” Los Alamos National Laboratory report, LA-UR-16-27168 (2016).

7) ASTM International. *Standard Test Methods for Rubber Property — Compression Set*; ASTM D 395 – 03; 2008; Vol. 9.

8) Wise, J.; Gillen, K.T.; Clough, R.L., *Radiat. Phys. Chem.* **1997**, *49*, 565. 16) Gillen, K.T.; Bernstein, R.; Celina, M., *Rubber Chem. Tech.* **2015**, *88*, 1.

9) Weis, E.M.; Blair, M.W.; Herman, M.J.; Keller, J.; Torres, J.A.; Edwards, S.L.; Cordes, N.L.; Oka, J.M.; Karns, T.; Brown, A.D.; Veirs, D.K.; Smith, P.H.; Stone, T.A., “FY 2015 Annual Report: Progress on the SAVY-4000 O-Ring Certification and Lifetime Extension Program,” Los Alamos National Laboratory Report (2015).

10) Cardon, R. A., “Polymer Seals in Radiological Service,” Los Alamos National Laboratory Criterion 428 R1 (2010).

11) Welch, Cynthia F.; Weis, Eric; Smith, Paul Herrick; Stone, Timothy Amos; Reeves, Kirk Patrick; Karns, Tristan; Oka, Jude M.; Keller, Jennie; Herman, Matthew Joseph; Weaver, Brian Phillip; Adams, Jillian Cathleen; Trautschold, Olivia Carol; Van Buskirk, Caleb Griffith; Garcia, Michael David. "SAVY-4000 O-Ring and Filter Lifetime Extension Report." Los Alamos National Laboratory Report (2017).