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LONG-TERM LEAK TIGHTNESS OF O-RING SEALS IN THE 9975 SHIPPING PACKAGE

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ABSTRACT

O-ring seals in the 9975 shipping package containment vessels are fabricated from a Viton GLT or GLT-S compound. Long-term testing of these O-rings has been performed to support service life predictions for packages used for long-term storage. Since the only criterion for O-ring performance is to maintain a leak-tight seal, leak testing is the primary indicator of service life. Fixtures have been aging at elevated temperatures to provide data for service life predictions. Limited leak test failures have been observed at the higher temperatures. This provides the opportunity for comparison to trends based on other O-ring properties, such as compression stress relaxation. Initial data suggest that the CSR data have some predictive value for a leak-tight service life, but other factors can complicate efforts to draw definitive conclusions.

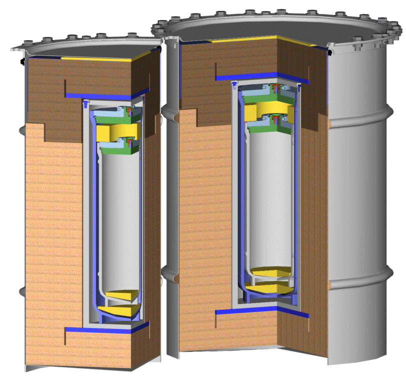

Figure 1 Illustration of 9975 package including fiberboard overpack, lead shielding, and double containment vessels.

INTRODUCTION

The Savannah River Site (SRS) is storing plutonium (Pu) materials in the K-Area Materials Storage (KAMS) facility. The Pu materials are packaged according to the standard DOE-STD-3013 which requires nested, welded, stainless steel containers. Within KAMS, the welded 3013 containers are stored in Department of Transportation (DOT) Type B 9975 packages [1]. The 9975 package consists of two nested stainless steel containment vessels (CV) closed with threaded cone-seal plugs, surrounded by a lead shielding body and fiberboard overpack, all contained within a 35 gallon stainless steel drum, Figure 1. The 9975 package is part of the approved storage configuration for Pu materials in KAMS. As such, it will be continuously exposed to the service environment for a period of time greater than the approved transportation service period. The leak rate studies were undertaken to verify the integrity of the containment vessels' O-ring seals for an initial storage assessment period of 10 years. It is anticipated that the packages may be used for up to 50 years. Accordingly, the replacement of aged components may be necessary based on the results of service life predictions..

Each containment vessel is sealed with O-rings (compounds V0835-75 or VM835-75, Parker Hannifin Corporation, Lexington, KY) based on Viton® GLT/GLT-S fluoroelastomer (Dupont Performance Elastomers, Wilmington, DE). Viton® fluoroelastomer O-rings are placed on the cone-seal plugs in each package: two O-rings, inner and outer, sealing the primary containment vessel (PCV) and two O-rings, inner and outer, sealing the secondary containment vessel (SCV). In transportation service, the O-rings are replaced on an annual basis and leak tested to a 1 x 10^{-7} ref cc air/s (leaktight) criterion per ANSI N14.5 [2]. However, while in storage the O-rings are not replaced and cannot be leak tested. The outer O-ring in each vessel is credited for containment. The inner O-ring provides a volume for leak testing and a secondary barrier to product release. To function as a seal, the O-rings must maintain mechanical elasticity and exert a compressive force against the sealing surfaces.

EXPERIMENTAL METHOD

Experiments to monitor the aging performance of Viton GLT and GLT-S based O-rings used in the Model 9975 package have been ongoing since 2004 at the Savannah River National Laboratory. Sixty-two mock-ups of 9975 PCVs, Figure 2, were assembled with GLT based O-rings and heated to 93 or 149 ºC. The vessels are heated with a flexible, wound-wire heater wrapped around the vessel circumference. Ceramic fiber batting is used to insulate the exposed ends of the fixture. Stainless steel tubing is attached to the leak test port on the top of the fixture lid via a high pressure fitting and to a threaded hole machined into the bottom of the fixture body. This arrangement allows for healium leak testing of both O-rings, either individually or in combination. Both sets of tubing are capped until needed for leak testing. A thermal fuse was added to each heater to prevent excess temperature excursions. The heaters are controlled by a desktop computer running

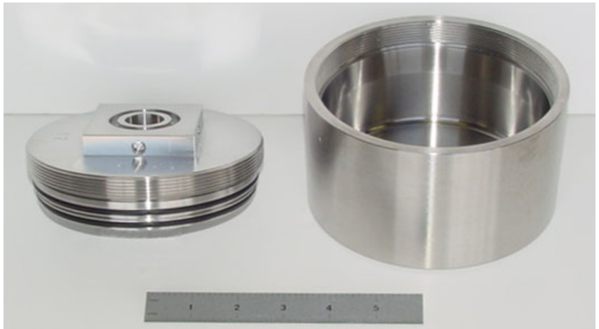

Figure 2 Mock-up PCV test fixture lid and body.

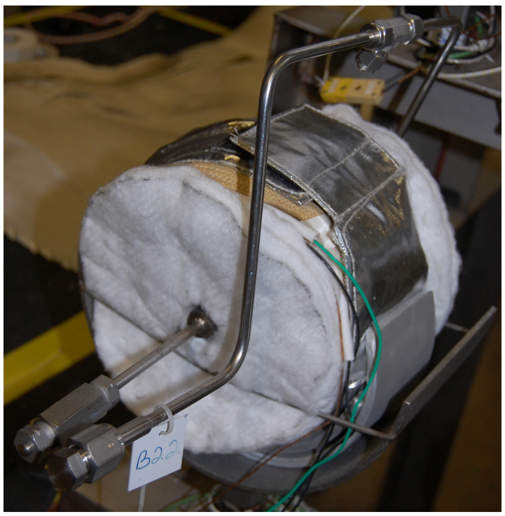

Figure 3 Assembled mock-up PCV.

LabView™ software, with feedback via a type-K thermocouple attached to the PCV body, see Figure 3.

Each fixture was leak-tested initially with a helium leak detector (Varian, Inc. Vacuum Technologies, Lexington, MA) and has been tested at nominal six month intervals to determine if it meets the criterion of leaktightness defined in ANSI standard N14.5 [2]. The test matrix was developed to determine the effect of temperature, gas composition, and irradiation dose and dose rate on the condition of the PCV O-rings over time. A total of 62 tests, with 22 separate sets of conditions were developed. Replicates of tests were performed based on a modified full-factorial statistical design. The test variables are listed in Table 1.

An additional five mock-up fixtures were more recently assembled in 2008 and exposed to higher temperatures (177, 204, and 232 ºC) in an attempt to accelerate aging of the GLT based O-rings, resulting in failures to improve the predictive capability of the first set of leak tests. Leak testing was performed in shorter intervals of 3 weeks or less on the elevated temperature fixtures with the expectation that these would fail in a much shorter time than the original fixtures. All of these fixtures were assembled with the normal O-ring lubricant and contained no backfill CO_2. Three of them, one at each aging temperature, were irradiated to 2 x 10^5 rad in 72 minutes.

Table 1 Test Matrix Variables for O-ring Experiment

Test Variable	Values Tested	Basis for Values Tested
Temperature	93 ºC	With loss of ventilation in the KAMS facility, the maximum ambient temperature is 58 ºC, and the corresponding PCV O-ring temperature is 92 ºC.
	149 ºC	The maximum allowable temperature for the PCV P-rings for continuous operation is 149 ºC.
	177 ºC 204 ºC 232 ºC	Elevated temperatures added to increase the likelihood of seeing O-rings failures in a timely manner.
Radiation Dose	$2x10^5$ Rad in 72 min	The bounding dosse rate for the PCV is 2 rad/hr. A total dose of $2x10^5$ rad represents ten years of storage.
	$2x10^5$ Rad in > 200 hr	Longer-term exposure may reveal the added effect of diffusion-limited oxidation (DLO) that only occurs with long-term exposure.
	None	Many packages will have little radiation exposure. This also serves as an experimental control.
Internal Atmosphere	≥75% CO_2 with a balance of Air	The free volume of the PCV is filled/diluted with CO_2 as a cover gas. A small portion of the air originally in the vessels may remain.
	Air	It supplies comparative data and acts as a control.
O-ring Lubriaction	Silicone high-vacuum grease	It is specified in assembly of the 9975 package.
	Krytox ® 240AC	It has been used on the 9975 O-rings at DOE facilities. It is used on lid components of the 9975 PCV and SCV.
	None	It supplies comparative control data. Also, it is possible that the O-rings may be mistakenly installed without grease.

A third set of 14 mock-up fixtures were assembled with GLT-S based O-rings and heated (93 ºC to 204 ºC). Testing variables included temperature, radiation dose (2 x 10^5 rad in 72 minutes or no exposure), and internal atmosphere of either air or a backfill of 75% CO_2. Leak testing was performed nominally every 3 weeks.

Once fixture leak rate surpassed the leak criterion, diagnostic leak testing was performed to determine whether one or both O-rings had failed. If only one had failed, the fixture was typically returned to test until the second O-ring failed. Once both O-rings had failed, the fixtures were disassembled and O-rings visually examined. O-rings were immediately measured for dimensions. O-ring thicknesses were measured again 1-2 weeks after disassembly, and 30 days after disassembly. Dimensions were taken at four locations along the O-ring in both the axial and radial directions. Hardness values were also measured.

RESULTS

Within the original set of fixtures containing GLT based O-rings exposed to 93 ºC, no failure in leak criterion at room temperature has been observed up to four years at temperature for both 93 and 149 ºC fixtures. The outer O-ring on one of the fixtures exposed to 149 ºC failed at 3.5 years. This fixture was CO_2 backfilled, irradiated at the low dose rate, and assembled with silicone grease on the O-rings.

Four fixtures with GLT based O-rings exposed to 177, 204 and 232 ºC have failed. The experimental parameters of the GLT based fixtures failures are shown in Table 2. In the 232 ºC fixture, the outer O-ring failed the leak test after 8 days at temperature, and the inner O-ring failed after 12 days at temperature. In the two 204 ºC fixtures, failure of both O-rings, inner and outer, in each fixture was noted after 28 and 45 days, respectively, at temperature. The outer O-ring in one of the two high temperature fixtures aging at 177 ºC failed after 10.6 months (324 days).

Table 2 Experimental parameters and time to failure detection for fixtures containing GLT based O-rings.

Temp (ºC)	Fill Gas	Rad Dose (rad)	Time to Outer O-ring Failure Detection (days)	Time to Inner O-ring Failure Detection (days)
149	75/25 CO_2/Air	$2x10^5$ in 300 hrs	1270	N/A
177	Air	$2x10^5$ in 72 mins	324	N/A
204	Air	$2x10^5$ in 72 mins	45	45
204	Air	None	28	28
232	Air	$2x10^5$ in 72 mins	8	12

Three fixtures aging at 204 and 232 ºC were disassembled after failing the room temperature leak test. For each fixture, both O-rings were verified to have failed to maintain leak-tightness. General observations made during disassembly include O-rings adhering to the body and/or plug, the body and plug retained black deposits from the O-rings, white and brown deposits indicative of oxidized grease were observed on the metal and O-ring. A scuff mark was discovered on one of the 204ºC O-rings in one location. The cross-sections of both O-rings from the 232 ºC fixture, were more squared than round after removal, Figure 4.

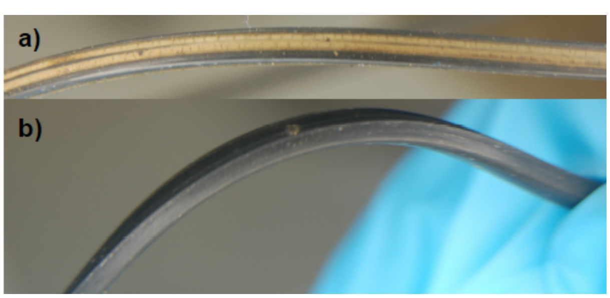

Figure 4 GLT O-rings from 232 ºC fixture after removal (a) inner O-ring (b) outer O-ring.

Fourteen fixtures were taken out of test after a computer controller failure resulted in a temperature excursion. Computer communication with the heaters was lost for a period of two hours, with the result that fourteen fixtures overheated and their thermal fuses melted. During that period, the maximum temperature recorded for each of these fixtures ranged from 172 to 408 ºC. With the temperature recorded at 30 minute intervals, actual peak temperatures could have exceeded the recorded values. Most of the overheated fixtures were opened and the O-rings discarded without examination. Four of them (Fixtures 16, 23, 24, and 34) were disassembled. Observations made on the fixtures are in Table 3.

Table 3 Observations made on fixtures disassembled after temperature excursion.

Fixture No.	Maximum Recorded Temperature (ºC)	Observations
16	221	No findings
23	222	Excess grease on/around O-rings
24	172	No findings
34	382	Significant amount of O-ring material extruded out of groove. Brown residue on bottom of plug. O-ring residue adhering to body and plug surfaces.

Initially, leak testing was performed at the aging temperatures. The testing led to inconclusive results and the removal of several fixtures before actual failure in room temperature tests.

Six fixtures of the highest aging temperature in the GLT-S based series have failed. All of the fixtures aged at 204 ºC failed. One fixture aged at 177 ºC failed. Experimental parameters of the GLT-S based fixture failures are shown in Table 4.

Table 4 Experimental parameters and time to failure detection for GLT-S based O-rings

Temp (ºC)	Fill Gas	Rad Dose (rad)	Time to Outer O-ring Failure Detection (days)	Time to Inner O-ring Failure Detection (days)
177	Air	None	113	113
204	Air	None	52	98
204	Air	None	74	74
204	>75/25% CO_2/Air	None	50	50
204	>75/25% CO_2/Air	None	52	52
204	>75/25% CO_2/Air	$2x10^5$ in 72 min	58	58
204	>75/25% CO_2/Air	$2x10^5$ in 72 min	280	280

Compression set measurements taken within 30 minutes after opening the fixture for failed O-rings removed from each fixture ranged from 60-75% and were comparable between GLT and GLT-S based O-rings.

DISCUSSION

Aging temperature is the most critical parameter for O-ring performance in both the GLT and GLT-S based O-rings when compared with gamma radiation dose, lubricant composition, and atmosphere. The lifetimes based on leak rate of the inner and outer O-rings both experience exponential decay with aging temperature, see Figure 5.

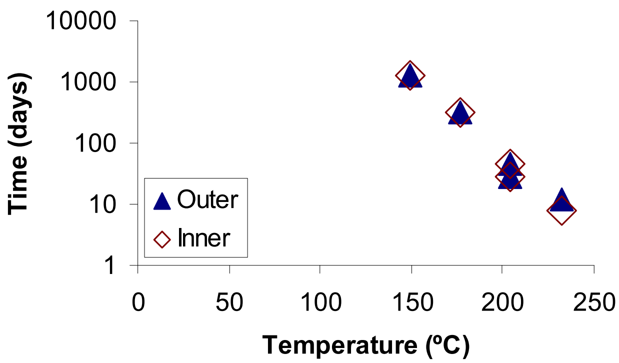

Figure 5 Time of O-ring failure detections versus temperature for both inner and outer GLT based O-rings.

A more extensive study was performed on the GLT based O-ring compared to the GLT-S based O-rings. However, with the current data, it appears that the lifetime for GLT and GLT-S based O-rings is similar. The lifetimes based on leak rate of the inner and outer GLT-S O-rings both experience decay with aging temperature, see Figure 6.

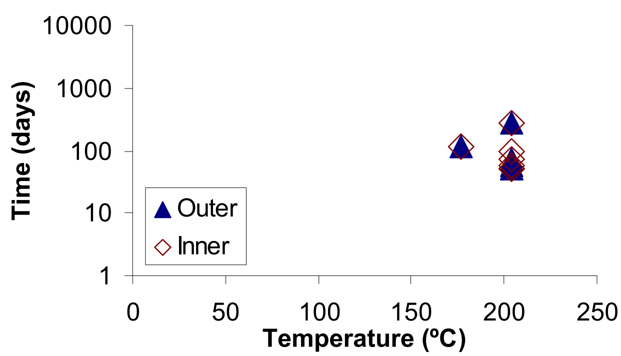

Figure 6 Time of O-ring failure detection versus temperature for both inner and outer GLT-S based O-rings.

When the leak testing results are compared to compression stress relaxation results [3], both suggest a decrease in seal performance with increasing temperature, see Figure 7.

Field 9975 packages containing plutonium materials were opened for surveillance activities and leak tested to a sensitivity of 10^{-3} ref cc air/s. This field leak testing was performed at room temperature by measuring pressure drop over time rather than using the He method. 164 packages have been evaluated with an average time in service of 1212 days or 3.3 years. Only two packages failed the post-load leak test. Both of the failures were attributed to debris on the O-

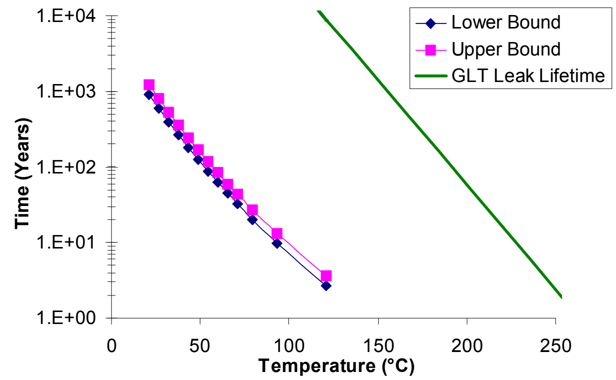

Figure 7. Comparison between CSR lower and upper bound lifetime limits versus the GLT O-ring lifetime based on leak testing.

ring opposed to degradation of the elastomer. The package with the longest time in service, 2469 days or 6.8 years, passed the post-load leak test.

Leaktightness is dependent not only on degree of O-ring degradation, but on the situation. Variables such as pressure gradient and debris contribute to the leaktightness of an O-ring. In this study, external variables were held constant within reason. Compression set is a measure of the elastomer's reduction in elastic response and can be used to evaluate the elastomer's degradation in mechanical response.

Compression set was calculated using O-ring thickness measurements immediately, 1-2 weeks, and 1 month after disassembly. The calculation for compression set was based on the average radial dimension (t_d) and taking into account that only the section of the O-ring thickness outside the metallic O-ring gland was capable of being compressed. The compression set equation used is:

$$CS\% = \left(\frac{t_d - t_i}{t_d - 0.0995}\right)100 \tag{1}$$

where t_i is the initial O-ring thickness and 0.0995 is the average depth of the O-ring gland on the cone seal plug. The original O-ring thickness was assumed to be a nominal value of 0.139" if baseline thickness data was not available.

Compression set results over time for inner and outer GLT based O-rings are shown in Figure 8 and 9.

The colors on Figures 8 and 9 indicate the fixture source. Brown indicates fixtures removed based on high temperature leak test results. Purple indicates fixtures removed after overheating. Red indicates high temperature fixtures removed following room temperature leak test failure. Blue indicates fixtures removed for other reasons.

No significant trend was found between the compression set response of the inner and outer O-rings. All fixtures experienced some degree of drop in compression set over time, including the O-rings from fixtures that were removed due to overheating. A drop in compression set over time

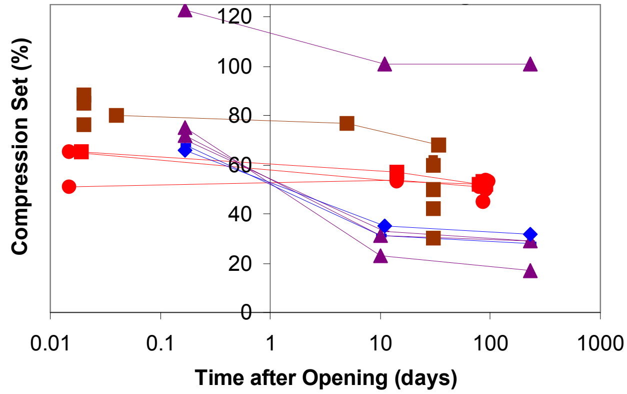

Figure 8 Compression set over time for GLT based inner O-rings.

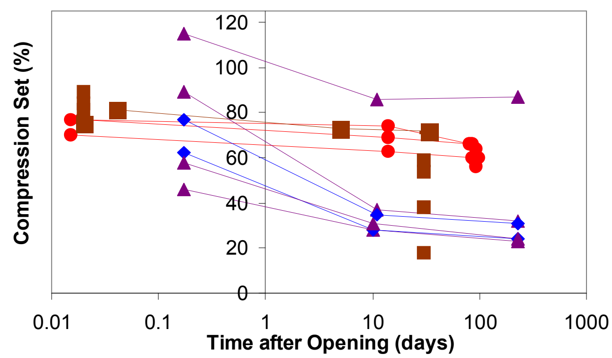

Figure 9 Compression set over time for GLT based outer O-rings.

suggests that the O-ring maintains some degree of elasticity. O-rings with the smallest change in compression set over time were the O-rings from fixtures aged at the higher temperatures.

Compression set results over time for inner and outer GLT-S based O-rings are shown in Figures 10 and 11.

Greater variation in compression set between the inner and outer GLT-S-based O-rings occurred compared to GLT. The change in compression set for both sets of O-rings was consistent between fixtures.

A typical compression set value for both PCV and SCV O-rings measured from surveillance activities is 10-15%. This compression set is less than the values measured in the

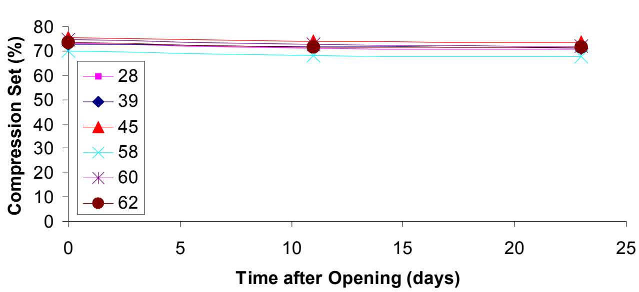

Figure 10 Compression set for outer GLT-S based O-rings.

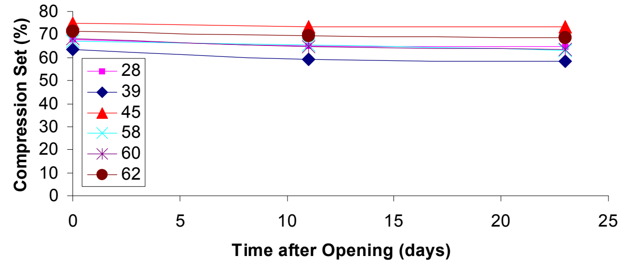

Figure 11 Compression set for inner GLT-S based O-rings.

laboratory testing leak fixtures. The higher aging temperatures used in the laboratory testing result in increased compression set in the laboratory O-rings. .

CONCLUSION

The lifetimes of Viton® GLT and GLT-S O-rings are strongly dependent on aging temperature. Typical service conditions that O-rings are exposed to in the KAMS facility are well below the accelerated aging testing temperatures used in this study. The CSR model predicts O-ring service life at the maximum predicted service temperature of 93 ºC of 10 years. Surveillance data on packages examined after 6 years in service show little degradation of the O-rings, with compression set of 15 % or less. This is consistent with predictions of a much longer service life. Comparison of service life predictions based on CSR data with time to failure due to leakage at elevated temperatures indicates the CSR-based predictions are conservative to the actual failure condition.

ACKNOWLEDGMENTS

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