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FY2018 Status Report: Model 9975 O-Ring Fixture Long-Term Leak Performance

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Summary

A series of experiments to monitor the aging performance of Viton® GLT and GLT-S O-rings used in the Model 9975 shipping package has been ongoing since 2004 at the Savannah River National Laboratory. One approach has been to periodically evaluate the leak performance of O-rings being aged in mock-up 9975 Primary Containment Vessels (PCVs) at elevated temperature. Other methods such as compression stress-relaxation (CSR) tests and field surveillance are also ongoing to evaluate O-ring behavior. Seventy tests using PCV mock-ups with GLT O-rings were assembled and heated to temperatures ranging from 200 to 450 ºF. They were leak-tested initially and have been tested periodically to determine if they continue to meet the leak-tightness criterion defined in ANSI standard N14.5-97. Due to material substitution, a smaller test matrix with fourteen additional tests was initiated in 2008 with GLT-S O-rings heated to temperatures ranging from 200 to 400 ºF.

Leak test failures have been experienced in all of the GLT O-ring fixtures aging at 350 ºF and higher temperatures, and in 8 fixtures aging at 300 ºF. The 300 °F GLT O-ring fixtures failed after 2.8 to 5.7 years at temperature. The remaining GLT O-ring fixtures aging at 300 ºF were retired from testing following more than 5 years at temperature without failure. No failures have yet been observed in GLT O-ring fixtures aging at 200 ºF for 10 to 11.6 years, or in GLT O-ring fixtures aging at 270 ºF for 6.5 to 6.7 years. These aging temperatures bound O-ring temperatures anticipated during normal storage in K-Area Complex (KAC).

Leak test failures have been experienced in all of the GLT-S O-ring fixtures aging at 300 ºF and above. No failures have yet been observed in GLT-S O-ring fixtures aging at 200 and 250 ºF for 8 to 8.5 years.

Data from the O-ring fixtures are generally consistent with results from compression stress relaxation testing, and provide confidence in the predictive models based on those results. However, uncertainty exists in extrapolating these elevated temperature results to the lower temperatures of interest for normal storage in KAC. Oxygen consumption testing, which includes results from temperatures near KAC normal storage temperatures, is ongoing to provide further confidence and corroborate these extrapolations. The collective data from these test efforts suggest the minimum O-ring service life at KAC normal storage conditions should be at least 34 years for GLT O-rings and 77 years for GLT-S O-rings.

Measurement of compression set in O-rings removed from failed fixtures, compared to that from KAC surveillance O-rings, indicates margin remains for O-rings still in service. Aging and periodic leak testing will continue for the remaining PCV fixtures.

Background

This is the FY2018 status report for PCV O-ring fixture experiments carried out per Task Technical Plan [1], which is part of the comprehensive 9975 package surveillance program [2].

PCV test fixtures were assembled with either Parker Seals V0835-75 (hereafter referred to as Viton® GLT) O-rings or Parker Seals VM835-75 (hereafter referred to as Viton® GLT-S) O-rings, and are being aged in environments that provide varying degrees of margin over KAC normal storage conditions (O-ring behavior during and following short-term accident conditions is beyond the scope of this work). The purpose of these experiments is to characterize the performance of the O-ring seals, and then correlate the data to lifetime predictions of primary (PCV) and secondary (SCV) containment vessel O-ring seals in 9975 packages being stored in KAC. O-ring performance in these tests is defined by leak-tightness, per ANSI standard N14.5-97 [3] at room temperature. The 9975 shipping package, including the O-rings, is currently approved for storage in KAC for up to 20 years [4]. The oldest packages have been in storage for approximately 16 years.

The data from these fixtures are scoping in nature, although most of the controls under which they were collected are typical of baseline data. Accordingly, care should be used to assess the overall quality of the data prior to use in baseline applications. Within the 9975 surveillance program, these data will be used for information only, to compare to baseline data from other testing and build confidence in the overall predictions of O-ring service life.

Experimental Method

This task involves the aging of containment vessel O-rings as-installed in a mock-up PCV, and does not incorporate any influence from the remainder of the package. While other package components (such as the fiberboard overpack and lead shield) may degrade over time under storage conditions, the O-rings are isolated within the containment vessels and continue to experience the same service environment. The O-ring temperature is calculated to change no more than 2 ºF during 20 years of bounding degradation of the fiberboard and lead shield [5]. Therefore, these aging effects experienced by the remainder of the package are not replicated in the O-ring aging tests.

Test Matrix

Testing has evolved to include 3 test matrices. These address Viton® GLT O-rings aged at 200 or 300 ºF, Viton® GLT O-rings aged at 270 – 450 ºF, and Viton® GLT-S O-rings aged at 200 – 400 ºF.

The first test matrix was developed to determine the importance and effect of several variables on the condition of the PCV O-rings over time inside the KAC storage facility. The variables believed to be the most relevant to O-ring performance in storage were O-ring temperature, radiation/dose rate, O-ring lubrication, and internal PCV atmosphere (internal PCV atmosphere was subsequently dropped as a test variable). Two different dose rates were selected to evaluate potential dose rate effects. A total of 62 tests, with 22 separate sets of conditions were developed. Replicates of tests were developed based on a modified full-factorial statistical design. The test variables and the basis for variable selection are given in Table 1. A 63rd test (designated 62-2007) was added in 2007, in which fixture 62 conditions were repeated, but was heated primarily in an oven. This test was terminated after less than 3 months for reasons that were not documented.

The interior of the test fixture is accessible through a tube connected to the bottom. This tube includes a T connection to facilitate leak testing of both O-rings simultaneously or separately. With this arrangement, data are obtained on both O-rings installed in each fixture. Although only the outer O-ring is credited for containment, testing both provides twice the information under nearly identical conditions.

Twenty seven fixtures have been removed from the first test matrix for reasons other than failing the room temperature leak test. Further details of these fixtures are provided in Reference 6. Several fixtures conditioning at 300 ºF have failed to remain leak-tight, beginning in April 2010. The remaining 300 ºF fixtures were retired in July 2012, still leak-tight after aging for more than 5 years. The status of each fixture, along with its test parameters, is summarized in Table 2.

Fixtures in the first test matrix were initially leak tested on a nominal 6-month schedule. Once the first of these began failing, the test frequency for fixtures heated to 300 °F was increased to every 3 months.

In the second test matrix, five fixtures were placed into test in October 2008 with new Viton® GLT O-rings. These fixtures were aged at temperatures ranging from 350 to 450 °F. They were intended to provide some O-ring failures in a shorter time frame to enhance the predictive value of the original test matrix and to determine the time to failure at the vendor's service temperature rating (400 °F). The predictive model assumes that the time to leakage at all temperatures is a function of a common mechanism. With the expectation that these would fail in a much shorter time than the original fixtures, they were leak tested on a nominal 3 week frequency.

An additional two fixtures with Viton® GLT O-rings were added to the second test matrix in April 2011, and began aging at 270 °F. With leak test failures experienced at aging temperatures of 300 ºF and above, and no failures projected to occur at 200 °F for many years yet, it was anticipated that these two intermediate temperature fixtures might experience leak failures sooner that the 200 ºF fixtures. This would provide additional confirmation of the extrapolation model for leak test data at an earlier date than the 200 °F fixtures.

All of the second matrix fixtures were assembled with the normal O-ring lubricant and contained no backfill gas (i.e. filled with air). Three of them (one each at 350, 400 and 450 °F) were irradiated to 2 E5 rad at a high dose rate (approximating a 10-year service dose at a bounding rate of 2 rad/hr). Gamma irradiation of fixtures in all test matrices is performed in a cobalt-60 cell [1].

The third test matrix repeats much of the variety of the first two matrices with Viton® GLT-S O- rings, but on a smaller scale. Fewer fixtures were used for this alternate O-ring material since it was expected they would demonstrate the same parametric variations as the GLT O-rings. Seven separate sets of conditions were developed, and tested in duplicate for a total of 14 fixtures. The status of these fixtures, along with their test parameters, is summarized in Table 2.

Several additional irradiations were performed in 2014, as proposed in Reference 7, as follows:

- Three of the 200 °F GLT fixtures received an additional 2 E5 rad (at high dose rate), and will continue to receive an additional 2 E5 rad every 4 years at the aging temperature.
- Three of the 200 °F GLT fixtures received an additional 1 E5 rad (at high dose rate), and will continue to receive an additional 1 E5 rad every 4 years at the aging temperature. These two steps will maintain an approximate balance relevant to storage conditions between the thermal and radiation degradation mechanisms.

- One retired, but unopened 300 °F GLT fixture was irradiated to total doses of 1 E6, 2 E6 and 3 E6 rad (bounding 50, 100 and 150 year doses). Each irradiation cycle was preceded and followed by a leak test, and also followed by a 24 hour cycle to 300 °F and another leak test. This provides a bounding radiation dose commensurate with the thermal degradation (5 years at 300 °F is comparable to about 155 years at the more realistic temperature of 166 °F, based on compression stress-relaxation predictive models).
- Two un-used GLT O-rings were installed on a cone-seal plug and irradiated incrementally until significant damage was observed at 240 Mrad. The same was done for two GLT-S O-rings. The maximum dose rate received by the inner O-ring on the side oriented up in the irradiator was 3.5 E5 rad/hr. These irradiations provide a comparison between the two compounds in their radiation response.
- It was anticipated that the previous test would be followed by installing new O-rings in a fixture and repeating the irradiations, to identify whether the compression experienced within the O-ring groove would extend the useful service life under high radiation dose. This option was dropped after the O-rings failed to degrade in the manner expected.

Initial Assembly and Setup

The two-piece lid of the mock-up PCV, consisting of the cone seal nut and cone seal plug, was machined to be identical to the actual PCV lid. The body of the mock-up PCV was shortened to 3.5 inches from the original design of 18.6 inches and a threaded hole was machined in the bottom to provide a port for evacuating and filling the vessel with gas and for in-situ leak testing of the O-rings. A PCV test fixture with the O-rings installed in the lid is shown in Figure 1.

The mock-up PCV fixtures were assembled per the requirements described in the 9975 Safety Analysis Report for Packaging (SARP) [8]. After installation of the O-rings and assembly of the mock-up PCV test fixture, an initial leak test was performed while the fixture was at room temperature to verify leak-tightness to 1 E-7 ref·cc/sec air or better. If the fixture required irradiation, it was placed in a Co-60 gamma cell and irradiated at one of two dose rates to reach a total dose of 2 E5 rad. This is equivalent to a ten year dose at the bounding dose rate expected for the PCV O-rings (2 rad/hr). The fixture was irradiated at either a "slow" dose rate of approximately 667 to 830 rad/hr (lasting approximately 240 hrs) or a faster rate of ~1.7 E5 rad/hr (lasting 72 minutes), as Viton® and other elastomers/polymers are known to be sensitive to the dose rate [9, 10]. After irradiation, the fixture was leak tested again to ensure that irradiation alone did not affect leak-tightness, and then heated to test temperature.

The vessels are heated with a flexible, wound-wire heater wrapped around the vessel circumference. Ceramic fiberboard and fiber batting are used to insulate the exposed ends of the fixtures. Stainless steel tubing is attached to the port on the top of the fixture lid via a high-pressure fitting and to the hole machined into the bottom of the PCV body. A thermal fuse was added to each heater to prevent excessive temperature excursions. The heaters are controlled by a desktop computer running LabView™ software, with feedback via a type-K thermocouple attached to the PCV body. The final assembled fixture is shown in Figure 2.

Fixture Leak Testing

The O-ring fixtures are leak-tested after initial setup, after irradiation, and periodically thereafter to the same leak-tight criterion as the 9975 PCV and SCV. A room temperature leakage rate of no more than 1 E-7 ref·cc/sec air (2 E-7 ref cc/sec He) demonstrates leak-tightness when measured according to the requirements outlined in ANSI Standard N14.5-97 [3]. The outer O-rings of the 9975 PCV and SCV are credited with being leak-tight while in transport and are credited with maintaining containment while in storage in the KAC [1, 8].

Leak testing is conducted using a Varian 959 helium mass spectrometer leak detector. A gas filled envelope test, as defined in ANSI N14.5-97 Section A.5.3 is used for the mock-up PCV fixtures [3]. Both O-rings are tested simultaneously, with failure of either O-ring causing a failure of the test. Although this approach differs from annual certification testing, it gives results that are valid and comparable, and accommodates the difference in set up of the actual PCV and SCV and the mock-up PCV fixture. If a leak is found, it is possible to determine which O-ring is leaking by selectively directing the helium to either the fixture interior or exterior, thus testing one O-ring at a time.

The O-ring fixture leak test program was reviewed in December 2008, prompting reconsideration of the methodology used for leak testing the mock-up PCV fixtures. One important change that was made in the conduct of the leak test involved extending the test duration until permeation of helium through the O-ring was detected [11].

Observing a permeation signal for each test provides positive evidence that the fixture and test setup are capable of transmitting a helium signal (i.e. no part of the flow path is blocked), and that helium was actually introduced into the fixture. Once a permeation signal was observed for each fixture, subsequent testing has been conducted without the extended duration to demonstrate permeation, since no actions are performed that might disrupt the flow path during aging and leak testing. All fixtures in test since December 2008 have demonstrated permeation. The time to permeation ranged from 4 to 75 minutes.

<u>Results</u>

PCV fixtures have been assembled and aged to identify the time to failure of GLT O-rings (70 tests), and GLT-S O-rings (14 tests). This report summarizes results for these fixtures through June 1, 2018.

A total of 23 GLT O-ring fixtures and 4 GLT-S O-ring fixtures remain in test. All of the GLT O-ring fixtures currently conditioning at 200 ºF have remained leak-tight, with total times at temperature ranging from 10 to 11.6 years (at the time of their last leak test). Two fixtures began conditioning at 270 ºF in 2011. They have remained leak-tight, with total time at temperature of 6.5 and 6.7 years (at the time of their last leak test). GLT O-rings in the remaining fixtures aging at 300 ºF and higher were retired or failed previously, as noted in prior status reports. The times to failure for each GLT O-ring fixture are summarized in Table 3. Recent leak rate histories can be found in Table 4 for fixtures in test since the last status report [12].

All of the GLT-S O-ring fixtures conditioning at 200 and 250 ºF have remained leak-tight, with total times at temperature of 8 or 8.5 years. GLT-S O-rings aging at 300 ºF and higher have failed previously, as noted in prior status reports. The times to failure for each GLT-S O-ring fixture are summarized in Table 3. Leak rate histories can be found in Table 4 for fixtures in test since the last status report [12].

Discussion

As noted in previous interim reports, sufficient data are available to compare the time to failure for GLT and GLT-S O-rings at 3 temperatures. At 300 and 400 ºF, most of the GLT-S O-ring fixtures remained leak-tight longer than the GLT O-ring fixtures. At 350 ºF, the trend is reversed, with 3 of the 4 GLT O-rings in two fixtures remaining leak-tight longer than the GLT-S O-ring fixtures. With the scatter in this varying trend, it is not practical to conclude whether one O-ring material would perform better than the other, especially at the lower temperatures typical of storage service. This conclusion remains unchanged from the previous status report.

No influence on time to failure has been observed from the irradiations performed on test fixtures. The additional irradiations added to the test matrix as the fixtures continue aging are similarly not expected to be severe enough to influence failure times. It is recognized that these irradiations present a simplistic approach that ignores potential synergistic effects from the simultaneous exposure to elevated temperature and radiation.

Recent thermal analysis of the 9975 package [5] assumed 95 ºF as the long-term average ambient temperature and a maximum heat load of 19 W, and calculated an O-ring temperature of 156 ºF at beginning of life, and 158 ºF after 20 years of service with bounding package degradation. In packages with lower internal heat loads, the O-ring temperatures would be correspondingly less.

The times at temperature for the fixtures to fail the leak test are shown graphically in Figure 3. In addition, the times at temperature for the fixtures still in test are also shown. A trendline provides a lower bound to the failure data to illustrate a potential extrapolation of the observed behavior to temperatures below 300 ºF. In addition, the O-rings aging at 270 ºF have now accumulated sufficient time at temperature to exceed the lower bound trendline, demonstrating that the trendline maintains a lower bound to the data at least to that temperature. Note, however, that the degree of scatter in the data would suggest significant uncertainty for extrapolations over a large temperature range. Nevertheless, this trendline suggests the possibility that O-rings aging at 200 ºF might maintain a leak-tight seal for up to 77 years, and O-rings at KAC storage temperatures have the potential for even greater service life.

The lower bound trendline is repeated in Figure 4, and compared to model predictions based on compression stress-relaxation (CSR) data [13]. Each line is extrapolated to suggest possible behavior at the temperatures the O-rings will experience in storage. The coarse dashed lines extrapolate the O-ring behavior through temperatures for which partial data exist (samples have not yet reached a failure point), while the fine dashed lines extrapolate the O-ring behavior further to temperatures for which no data exist. Given the range of scatter observed in both fixture and CSR testing, it is recommended that any extrapolated service life estimates not exceed the minimum of these projections at this time.

Tests based on measuring the rate of oxygen consumption of aging O-ring material samples at elevated and KAC storage temperatures are underway to provide further confidence in these extrapolations or indicate an alternate extrapolation approach. Oxygen consumption results were generally in agreement with CSR and leak testing results, although there were slight deviations from extrapolated CSR data at temperatures below 175 ºF [13]. The oxygen consumption testing includes results from temperatures near KAC service temperatures and corroborates and adds confidence in the predictive models. Considering both oxygen consumption and CSR results, the extrapolations predict potential service life at 158 ºF of up to 34 years and 570 years for GLT and GLT-S O-rings, respectively. Given these collective results, it would be conservative to assume GLT O-rings will perform their required function in KAC storage conditions for at least 34 years (based on the more limiting CSR and oxygen consumption rate data), and GLT-S O-rings will perform their required function in KAC storage conditions for at least 77 years (based on the more limiting leak test data). As further data is collected from each of these tasks and the aging models further refined, these conservative service life estimates may be revised. However, at present, both GLT and GLT-S O-rings are expected to provide leak-tight service well in excess of the current approved 20 year KAC storage life.

The compression set data from the failed fixtures are plotted in Figure 5, showing compression set values as a function of the aging time. Compression set values based on O-ring dimensions right after removal are shown in Figure 5a, while Figure 5b shows the final compression set values based on dimensions measured 26 days or more after removal. Also shown in Figure 5 are compression set values from surveillance O-rings. There is a clear separation in compression set values from surveillance and from failed fixtures. This separation is larger for the final compression set values than for the initial values.

Compression set for the surveillance O-rings is calculated using ASTM D395, Method B. This parameter relates to the degree of O-ring degradation by showing how much the O-ring can spring back from its compressed state. A new O-ring should have a compression set of approximately 0%, while a heavily degraded O-ring will have a compression set approaching 100%. There is currently no demonstrated limit on compression set at which the O-ring will not remain leak-tight. Such a limit would be highly variable depending on the specific O-ring environment.

A nominal initial O-ring thickness is assumed in calculating compression set of surveillance O-rings since the actual initial dimensions are unknown. This introduces an uncertainty band for the surveillance O-rings of +/- 12 percentage points. Note that some of the compression set values in Figure 5b are negative (as low as -10%). Since negative compression set values are not physically meaningful for this O-ring configuration, these data points likely reflect an initial O-ring thickness less than nominal. If a similar scatter exists for initial O-ring thickness greater than nominal, then the maximum final compression set values for surveillance O-rings could be well under 20%. Whatever degree of scatter has resulted from variation in the initial O-ring thickness is equally applicable to both the initial and final compression set values.

The surveillance O-ring compression set values are compared to those from the test fixtures to show that there is margin between these two sets. While there are no data to specifically show how long it will take at storage conditions for the compression set to increase to that seen from the test fixtures, so long as surveillance O-rings continue to show a similar behavior, they likely retain significant margin to reaching a failure condition.

Conclusions

The GLT O-ring fixtures aging at 300 ºF and above have experienced leak test failures, or they have been removed from test for other reasons. No failures have yet been observed in GLT O-ring fixtures aging at 200 ºF and 270 ºF for 10 – 11.6 years and 6.5 to 6.7 years, respectively, which is more representative of (but still bounding to) O-ring temperatures during normal storage in KAC. These 23 fixtures with GLT O-rings continue aging at 200 or 270 ºF.

Leak test failures have been experienced in all of the GLT-S O-ring fixtures aging at 300 ºF and above. No failures have yet been observed in GLT-S O-ring fixtures aging at 200 and 250 ºF for up to 8.5 years. These 4 fixtures with GLT-S O-rings continue aging at 200 or 250 ºF.

The PCV O-ring temperature expected in the KAC is 156 - 158 °F, based on an average ambient temperature of 95 °F and the maximum payload (19 W). Cooler ambient temperatures and reduced payloads are less challenging to the seals in storage. The data available to date suggest the GLT and GLT-S O-rings aging at 200 ºF might maintain a leak-tight seal for up to 77 years, and O-rings in KAC storage at temperatures of 158 °F or less have the potential for even greater service life. Further consideration should be given to results from CSR testing which give different predictions of O-ring service life.

Data from the O-ring fixtures are generally consistent with results from compression stress relaxation testing, and provide confidence in the predictive models based on those results. However, uncertainty exists in extrapolating these elevated temperature results to the lower temperatures of interest for storage in KAC. Oxygen consumption testing, which includes results from temperatures near KAC storage temperatures, are ongoing to corroborate and provide further confidence in these extrapolations. Measurement of compression set in O-rings removed from failed fixtures, compared to that from KAC surveillance O-rings, indicates margin remains for O-rings still in service.

Aging and periodic leak testing will continue for the remaining fixtures.

References

[1] SRNL-TR-2014-00057, "Task Technical and Quality Assurance Plan for Characterization and Surveillance of Model 9975 Shipping Package O-Rings and Fiberboard Materials", April 2014.

[2] WSRC-TR-2001-00286, Rev. 9, "The Savannah River Site Surveillance Program for the Storage of 9975/3013 Plutonium Packages in KAC", Oct 2017.

[3] ANSI Standard N14.5-97, "American National Standard for Radioactive Materials - Leakage Tests on Packages for Shipment", American National Standards Institute, New York, NY, February 1998.

[4] G-ESR-K-00204, Revision 0, 9975 Shipping Package/3013 Container 20 Year Life Extension Summary, June 2017.

[5] M-CLC-K-00788, “The Initial and 20-year Service Thermal Performances of the 9975 Shipping Packages due to Fire-Drop-Smoldering Accidents in KAC”, B. B. Kiflu, January 17, 2017.

[6] SRNL-TR-2009-00186, “Fourth Interim Status Report: Model 9975 PCV O-Ring Fixture Long-Term Leak Performance”, W. L. Daugherty and T. M. Stefek, June 2009.

[7] SRNL-TR-2014-00170, “Ninth Interim Status Report: Model 9975 PCV O-Ring Fixture Long-Term Leak Performance”, W. L. Daugherty, August 2014.

[8] WSRC-SA-2002-00008, Rev. 0, “Safety Analysis Report for Packaging – Model 9975”, December 2003. (Note –this revision is cited for historical reasons. The current revision is S-SARP-G-00003, Rev 4, “Safety Analysis Report for Packaging – Model 9975”, Feb 2016.)

[9] Nuclear Instruments and Methods in Physics Research B, vol 208, pp 448-453, “Dose Rate Effects in Radiation Degradation of Polymer-Based Cable Materials, V. Placek, B. Bartonicek, V. Hnat and B. Otahal, 2003.

[10] Journal of Nuclear Materials, Vol 131, pp 197-207, “Prediction of Service Lifetimes of Elastomeric Seals During Radiation Ageing”, S. C. Burnay and J. W. Hitchon, 1985.

[11] SRNL-L1400-2008-00038, “Characterization of Leak Test System for O-Ring Studies”, D. J. Trapp, December 18, 2008.

[12] SRNL-TR-2017-00257, “FY2017 Status Report: Model 9975 O-Ring Fixture Long-Term Leak Performance”, W. L. Daugherty, July 2017.

[13] SRNL-STI-2018-00201, “Compression Stress-Relaxation and Oxygen Consumption Behavior of 9975 Shipping Package O-rings”, T. T. Truong and W. L. Daugherty, April 2018.

[14] SRNS-TR-2008-00290, Rev. 0, “Summary and Matrix 9975 Shipping Package Qualification Program for Extended Storage of Plutonium in the K Area Complex”, J. A. Radder, Savannah River Nuclear Solutions, Aiken, SC, November 2008. (Note – later revisions to this document exist, but this revision is cited for historical reasons.)

[15] S-CLC-K-00194, Rev. 2, “Integrated Thermal Analysis of the 9975 Shipping Packages for KAMS Phase 6”, J. K. Norkus, March 16, 2011.

[16] L9.4-10500, Rev. 2, “Annual Maintenance and Leak Testing for the 9975 Shipping Package,” Instrumentation and Equipment Systems Section, SRNL, August 29, 2007. (Note – later revisions to this document exist, but this revision is cited for historical reasons.)

Table 1. Test Matrix Variables for O-Ring Fixtures

Test Variable	**Values Tested**	**Basis for Values Tested**
Temperature	200 °F (93 °C)	With loss of ventilation in the KAC facility, the maximum ambient temperature is 137 °F [14], and the corresponding PCV O-ring temperature is 199 °F [15].
	300 °F (149 °C)	The maximum allowable temperature for the PCV O-rings for continuous operation is 300 °F [8].
	270, 350, 400, 450 °F (132, 177, 204, 232 °C)	Elevated temperatures added to increase the likelihood of seeing O-ring failures in shorter test periods.
Radiation Dose	2 E5 rad in 72 min	The bounding (high) dose rate for the PCV is 2 rad/hr. A total dose of 2 E5 rad represents ten years of storage (the initial period to be validated).
	Additional periodic 2 E5 rad at high dose rate	Additional dose after 8 years, and every 4 years subsequently, to maintain relative balance between degradation mechanisms (radiation, thermal)
	Additional periodic 1 E5 rad at high dose rate	Additional dose after 8 years, and every 4 years subsequently, to maintain relative balance between degradation mechanisms (radiation, thermal)
	2 E5 rad in >200hr	Longer-term exposure may reveal the added effect of diffusion-limited oxidation (DLO) that only occurs with long-term exposure. (lower dose rate)
	None	Many packages will have little radiation exposure. This also serves as an experimental control.
O-Ring Lubrication	Silicone high-vacuum grease	It is specified in assembly of the 9975 package [16].
	Krytox® 240AC	It has been used on 9975 O-rings at DOE facilities. It is used on lid components of the 9975 PCV and SCV [16].
	None	It supplies comparative control data. Also, it is possible that the O-rings may be mistakenly installed without grease.

Table 2. Summary of test parameters for fixtures

Temp. °F	Gamma Dose (rad) / Dose Rate	Lubricant	Fixtures Still in Test	Fixtures Removed from Test		
				Failed Leak Test	Retired July 2012	For Other Reasons**
GLT O-ring Fixtures – First Test Matrix						
200	~2 E5 High	Normal	5, 6, 9, 27, 36, 37, 40			15, 16, 23, 24
200	~2 E5 High +2 E5 after 8 years	Normal	41, 42			
200	~2 E5 High +1 E5 after 8 years	Normal	53, 54, 55			
200	~2 E5 Low	Normal	10, 11			
200	None	Normal	1, 3, 43, 44, 56, 57			13, 28, 29
300	~2 E5 High	Normal		8, 12, 26, 31	7, 52	17, 22, 25, 39, 45, 46, 47, 58, 59, 60
300	~2 E5 High +2.8 E6 after retirement	Normal			51	
300	~2 E5 Low	Normal		32	18, 30*	21, 38
300	None	Normal		49, 33	4, 61	2, 14, 48, 50, 62, 62-2007
300	~2 E5 High	None				19
300	None	None				34
200	None	Krytox	35			
300	~2 E5 Low	Krytox			20	
GLT O-ring Fixtures – Second Test Matrix						
270	None	Normal	14W, 21W			
350	~2 E5 High	Normal		18D		
350	None	Normal		19D		
400	~2 E5 High	Normal		14D		
400	None	Normal		21D		
450	~2 E5 High	Normal		23D		
GLT-S O-ring Fixtures – Third Test Matrix						
200	None	Normal	13H, 15H			
250	None	Normal	22H, 16H			
300	None	Normal		29H, 34H		
350	None	Normal		38H, 39H		
400	None	Normal		45H, 58H, 60H, 62H		
400	2 E5 High	Normal		28H, 50H		

* Fixture 30 has 1 failed O-ring (inner).

** Other reasons fixtures were removed from test include high temperature leak test difficulties (11 fixtures), and unplanned temperature excursion (14 fixtures). Two additional fixtures were removed from test for reasons that were not documented.

Table 3. Summary of GLT and GLT-S O-ring leak failures

		Days at temperature to failure *	
Fixture	**Temp (ºF)**	**Inner**	**Outer**
GLT O-ring Fixtures			
8	300	2009 - 2082	2009 - 2082
12	300	957 - 1020	957 - 1020
26	300	1273 - 1366	1261 - 1273
30	300	1279 - 1392	1902 – no fail
31	300	1280 - 1291	1280 - 1291
32	300	1271 - 1352	1271 - 1352
33	300	1360 - 1466	1924 - 1979
49	300	1101 - 1276	1323 - 1360
18D	350	481 - 497	304 - 324
19D	350	573 - 594	560 - 571
14D	400	29 - 45	29 - 45
21D	400	8 - 28	8 - 28
23D	450	10 - 12	0 - 8
GLT-S O-ring Fixtures			
29H	300	2370 - 2455	2370 - 2455
34H	300	2013 - 2096	2013 - 2096
38H	350	338 - 358	338 - 358
39H	350	95 - 114	95 - 114
45H	400	65 - 99	14 - 33
58H	400	62 - 75	62 - 75
60H	400	33 - 50	33 - 50
62H	400	34 - 50	34 - 50
28H	400	33 - 50	33 - 50
50H	400	260 - 281	260 - 281

* The first time at temperature is the last successful leak test. The second time at temperature is the failed leak test. Failure occurred at some point between these two times.

Table 4. Leak rate data since last status report

Test 1 (GLT), 200 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
1/18/17	10.5	2.7 E-8
8/1/17	11.0	5.4 E-9
2/1/18	11.5	5.0 E-9

Test 3 (GLT), 200 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
6/28/17	11.0	1.3 E-8
1/5/18	11.6	5.5 E-9

Test 5 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
2/16/17	10.5	1.6 E-8
9/6/17	11.0	5.6 E-9
2/27/18	11.5	5.7 E-9

Test 6 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
2/16/17	10.5	9.7 E-9
9/6/17	11.1	1.1 E-8
2/27/18	11.5	8.5 E-9

Test 9 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
5/9/17	10.0	6.9 E-8
11/9/17	10.5	2.4 E-8
5/23/18	11.0	2.5 E-9

Test 10 (GLT), 200 ºF		
2E5rad /240 hr, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
6/28/17	10.5	7.3 E-9
1/5/18	11.1	<1.9 E-9

Test 11 (GLT), 200 ºF		
1.4E5 rad /479 hr, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
12/20/16	10.0	7.6 E-9
7/11/17	10.5	9.4 E-9
1/17/18	11.0	6.2 E-9

Test 27 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
2/16/17	9.0	1.6 E-8
9/6/17	9.5	8.5 E-9
2/27/18	10.0	8.5 E-9

Test 35 (GLT), 200 ºF		
No rad., Krytox grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
1/18/17	10.0	2.5 E-9
8/1/17	10.5	1.4 E-8
2/1/18	11.0	5.0 E-9

Test 36 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
1/18/17	10.0	2.7 E-8
8/1/17	10.5	1.1 E-8
2/1/18	11.0	7.5 E-9

Test 37 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
1/18/17	10.0	3.7 E-8
8/1/17	10.5	2.7 E-9
2/1/18	11.0	5.0 E-9

Test 40 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
5/9/17	10.0	4.6 E-9
11/9/17	10.5	1.8 E-8
5/23/18	11.0	2.5 E-9

Table 4. (cont) Leak rate data since last status report

Test 41 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
12/20/16	10.0	5.0 E-9
7/11/17	10.5	1.9 E-8
1/17/18	11.0	6.2 E-9

Test 42 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
6/28/17	10.5	2.1 E-8
1/5/18	11.0	1.0 E-8

Test 43 (GLT), 200 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
1/18/17	10.0	9.9 E-9
8/1/17	10.5	1.1 E-8
2/1/18	11.0	1.0 E-9

Test 44 (GLT), 200 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
6/28/17	10.5	1.5 E-8
1/5/18	11.1	8.2 E-9

Test 53 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
6/28/17	10.5	1.7 E-8
1/5/18	11.1	<1.9 E-9

Test 54 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
6/28/17	10.5	2.8 E-8
1/5/18	11.0	<1.9 E-9

Test 55 (GLT), 200 ºF		
2E5rad /72 min, silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
12/20/16	10.0	5.0 E-9
1/17/18	11.1	6.2 E-9

Test 56 (GLT), 200 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
2/16/17	10.0	9.7 E-9
9/6/17	10.5	1.1 E-8
2/27/18	11.0	2.8 E-9

Test 57 (GLT), 200 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
1/18/17	10.0	1.5 E-8
8/1/17	10.5	8.1 E-9
2/1/18	11.0	2.5 E-9

Test 14W (GLT), 270 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
5/24/17	5.7	3.5 E-8
8/22/17	5.9	9.6 E-9
11/28/17	6.2	1.6 E-8
3/27/18	6.5	8.2 E-9

Table 4. (cont) Leak rate data since last status report

Test 21W (GLT), 270 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
5/24/17	5.7	1.9 E-8
8/22/17	6.0	1.4 E-8
11/28/17	6.3	<1.8 E-8
2/28/18	6.5	2.8 E-9
5/23/18	6.7	5.0 E-9

Test 13H (GLT-S), 200 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
3/20/17	7.5	5.5 E-9
10/3/17	8.0	1.7 E-8
3/27/18	8.5	5.5 E-9

Test 15H (GLT-S), 200 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
2/16/17	7.5	3.2 E-9
9/6/17	8.0	1.4 E-8
2/27/18	8.5	2.8 E-9

Test 16H (GLT-S), 250 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
12/20/16	6.9	5.0 E-9
8/1/17	7.5	1.4 E-8
2/1/18	8.0	5.0 E-9

Test 22H (GLT-S), 250 ºF		
No rad., silicone grease		
Date	Time at Temp (years)	Leak Rate (std cc He/sec)
5/9/17	7.5	<1.6 E-9
11/9/17	8.0	2.9 E-9
5/23/18	8.5	5.0 E-9

Table 5. Summary of compression set data* from O-ring fixtures

Fixture ID & History	Time since Opening	Comp. Set – Inner / Outer O-ring	Time since Opening	Comp. Set – Inner / Outer O-ring	Time since Opening	Comp. Set – Inner / Outer O-ring
GLT O-Ring Fixtures reported previously with high temperature leak test difficulties						
2 (392 days at 300 ºF)					30 days	62% / 59%
29 (283 days at 200 ºF)					30 days	30% / 18%
38 (473 days at 300 ºF)	<30 min.	85% / 87%				
39 (456 days at 300 ºF)	<30 min.	77% / 81%				
45 (291 days at 300 ºF)					30 days	60% / 71%
46 (493 days at 300 ºF)	<30 min.	76% / 75%				
47 (394 days at 300 ºF)	1 hour	80% / 81%	5 days	77% / 73%	34 days	68% / 72%
48 (490 days at 300 ºF)	<30 min.	84% / 84%				
50 (265 days at 300 ºF)					30 days	42% / 38%
60 (454 days at 300 ºF)	<30 min.	88% / 89%				
62 (282 days at 300 ºF)					30 days	50% / 54%
GLT O-Ring Fixtures removed after failing leak test (at room temperature)						
8 (2082 days at 300 ºF)	15 minutes	90% / 92%	13 days	94% / 91%	30 days	94% / 89%
12 (1020 days at 300 ºF)	7 minutes	82% / 70%	14 days	75% / 55%	30 days	74% / 49%
14D (45 days at 400 ºF)	21 minutes	51% / 77%	9 days	54% / 74%	85 days	45% / 66%
18D (497 days at 350 ºF)	23 minutes	91% / 96%	14 days	93% / 97%	30 days	92% / 97%
19D (594 days at 350 ºF)	13 minutes	95% / 94%	14 days	98% / 98%	30 days	97% / 97%
21D (27 days at 400 ºF)	27 minutes	66% / 77%	9 days	58% / 69%	80 days	53% / 66%
23D (12 days at 450 ºF)	21 minutes	65% / 70%	14 days	53% / 63%	90 days	54% / 59%
26 (1410 days at 300 ºF)	10 minutes	90% / 91%	14 days	88% / 89%	30 days	88% / 88%
31 (1292 days at 300 ºF)	15 minutes	84% / 78%	14 days	80% / 67%	31 days	78% / 65%
32 (1352 days at 300 ºF)	14 minutes	93% / 83%	14 days	90% / 73%	31 days	89% / 71%
33 (1979 days at 300 ºF)	10 minutes	88% / 82%	12 days	84% / 74%	29 days	84% / 73%
49 (1360 days at 300 ºF)	14 minutes	84% / 81%	14 days	82% / 80%	30 days	81% / 79%
GLT O-Ring Fixtures removed for other reasons						
7 (2167 days at 300 ºF)	19 minutes	93% / 84%	12 days	90% / 78%	29 days	90% / 75%
28 (630 days at 200 ºF)	4 hours	68% / 62%	10 days	31% / 28%	230 days	28% / 24%
30 (1902 days at 300 ºF)	16 minutes	94% / 82%	12 days	90% / 73%	29 days	89% / 71%
52 (1848 days at 300 ºF)	12 minutes	92% / 87%	12 days	90% / 80%	29 days	88% / 79%
62-2007 (~3 months at 300 ºF)	4 hours	66% / 77%	11 days	35% / 35%	230 days	32% / 31%
GLT-S O-Ring Fixtures removed after failing leak test (at room temperature)						
28H (50 days at 400 ºF)	10 minutes	84% / 91%	11 days	80% / 88%	26 days	80% / 88%
29H (2455 days at 300 ºF)	7 minutes	83% / 80%	14 days	79% / 79%	30 days	79% / 79%
34H (2096 days at 300 ºF)	21 minutes	90% / 88%	14 days	90% / 86%	28 days	88% / 89%
38H (358 days at 350 ºF)	20 minutes	92% / 92%	14 days	90% / 88%	30 days	88% / 87%
39H (114 days at 350 ºF)	15 minutes	78% / 90%	11 days	74% / 89%	26 days	72% / 88%
45H (99 days at 400 ºF)	12 minutes	93% / 93%	11 days	91% / 92%	26 days	91% / 91%
50H (281 days at 400 ºF)	14 minutes	95% / 82%	14 days	93% / 76%	30 days	93% / 76%
58H (75 days at 400 ºF)	10 minutes	83% / 87%	11 days	81% / 84%	26 days	78% / 84%
60H (50 days at 400 ºF)	7 minutes	84% / 93%	11 days	80% / 90%	26 days	79% / 89%
62H (50 days at 400 ºF)	7 minutes	89% / 91%	11 days	86% / 89%	26 days	85% / 89%

* Compression set is calculated per ASTM D395, Method B, as follows:

$$\text{comp. set (\%)} = (t_i - t_f) / (t_i - \text{groove depth}) * 100$$

If the initial radial thickness was not recorded, 0.139 inch is assumed.

Figure 1. Mock-up PCV test fixture lid and body.

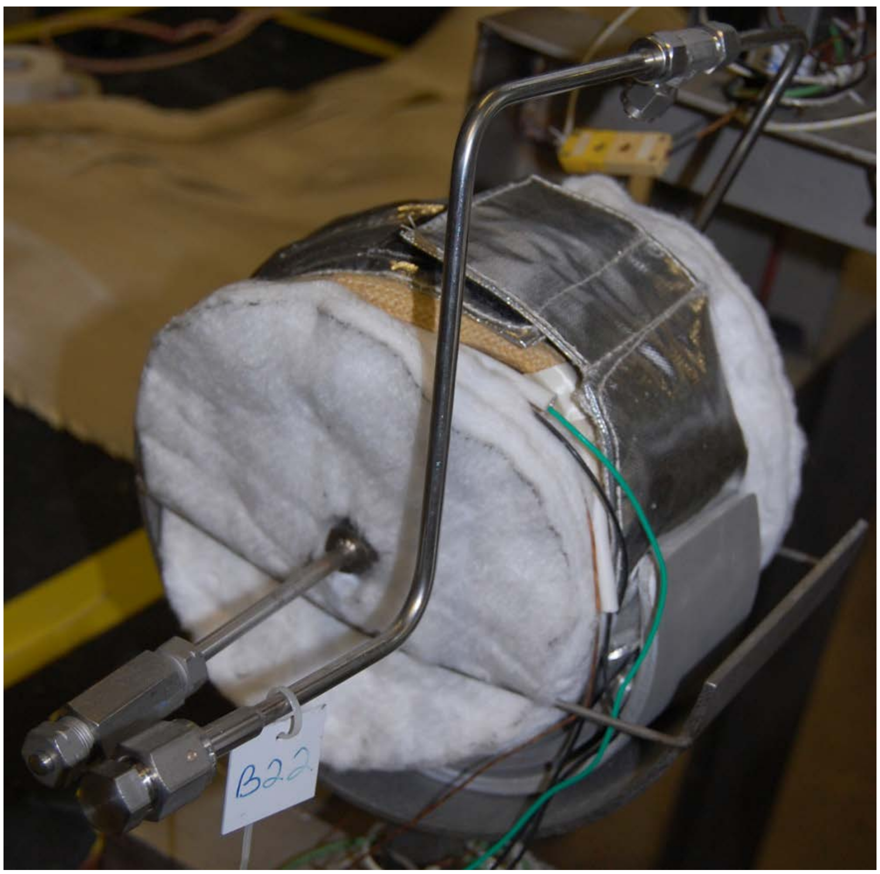


Figure 2. Assembled mock-up PCV test fixture with heater and insulation.

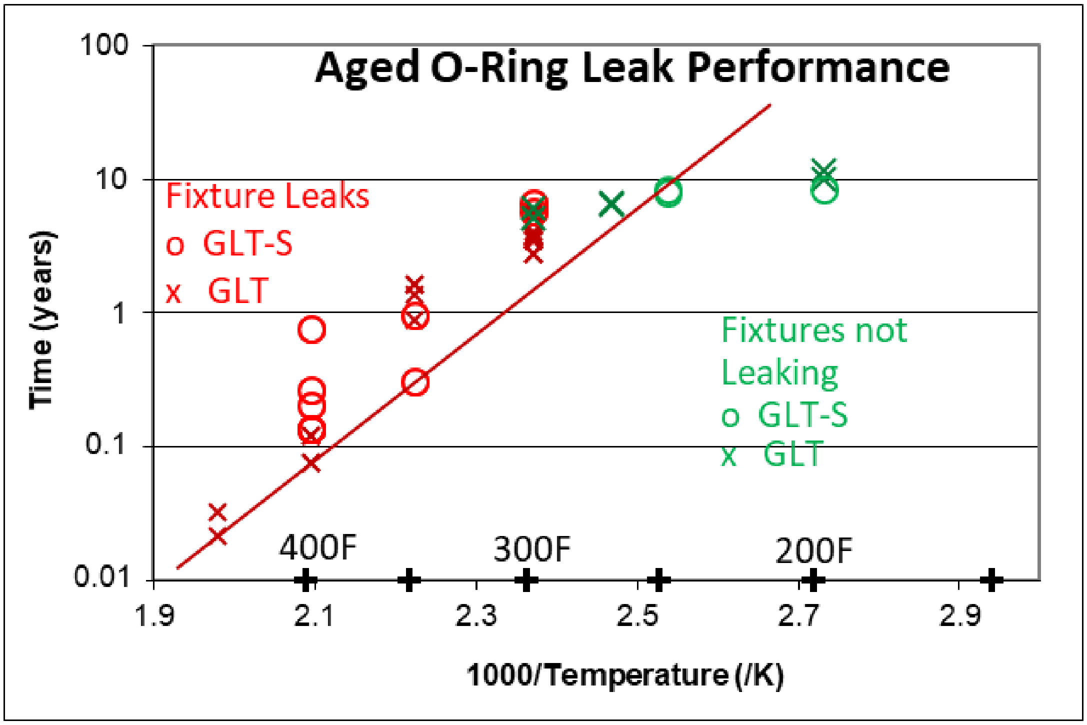


Figure 3. Summary of behavior for fixtures that failed the leak test and for fixtures still in test. The trendline (defined by y=1.005 E-11 * exp(10.86x)) illustrates a lower bound projection.

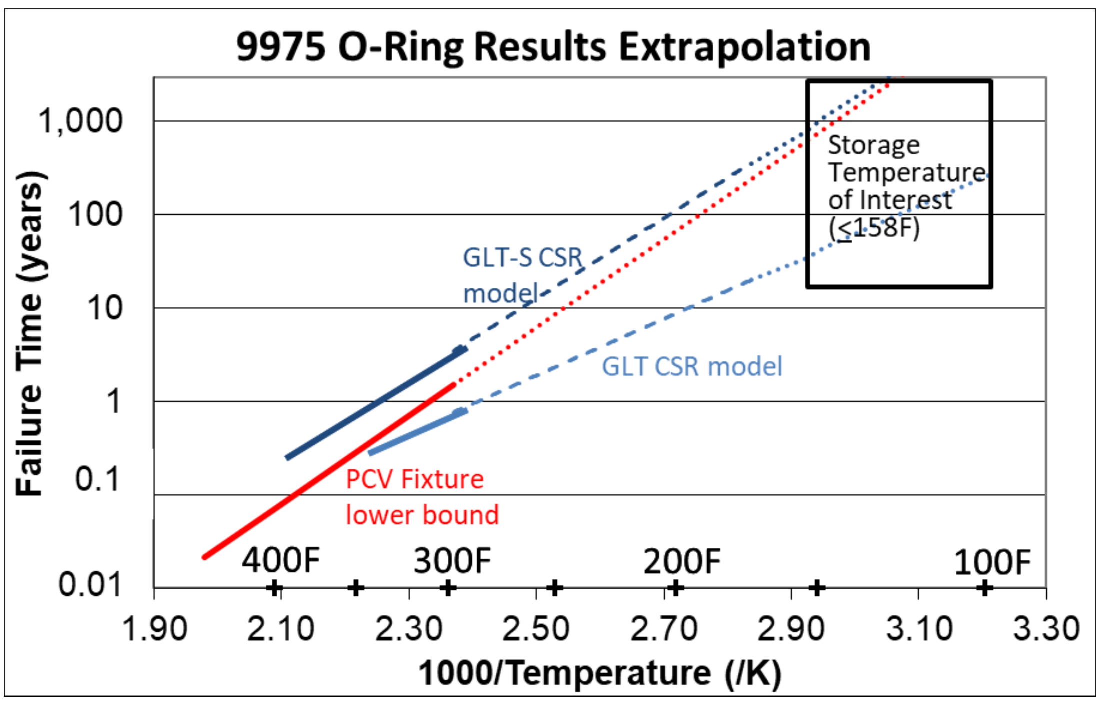
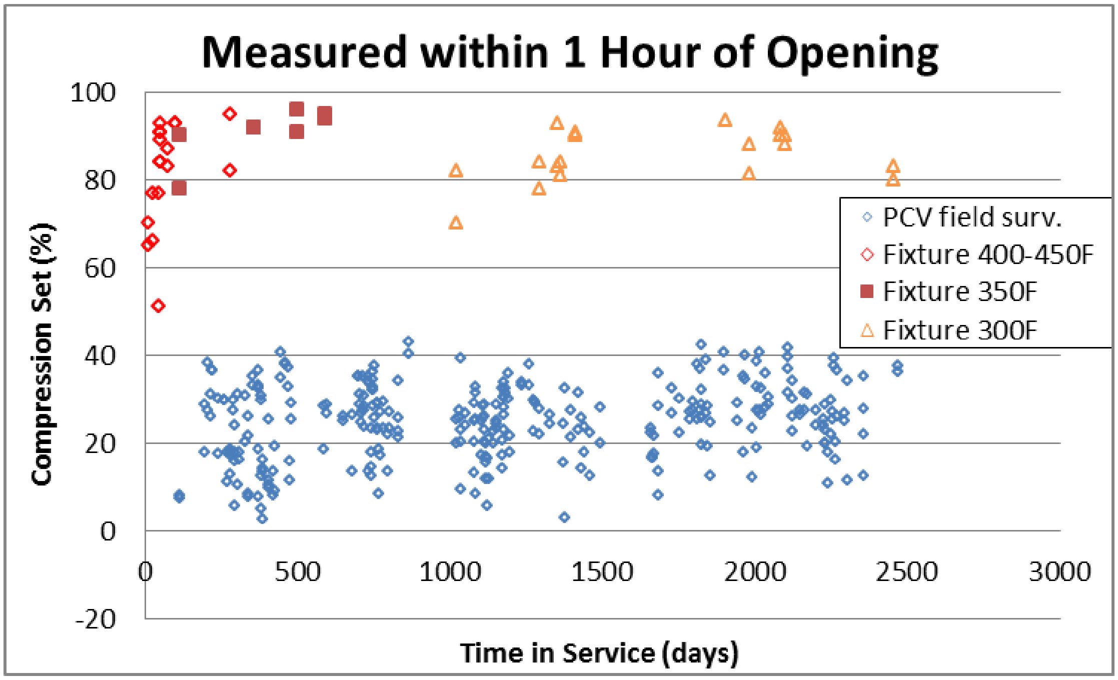
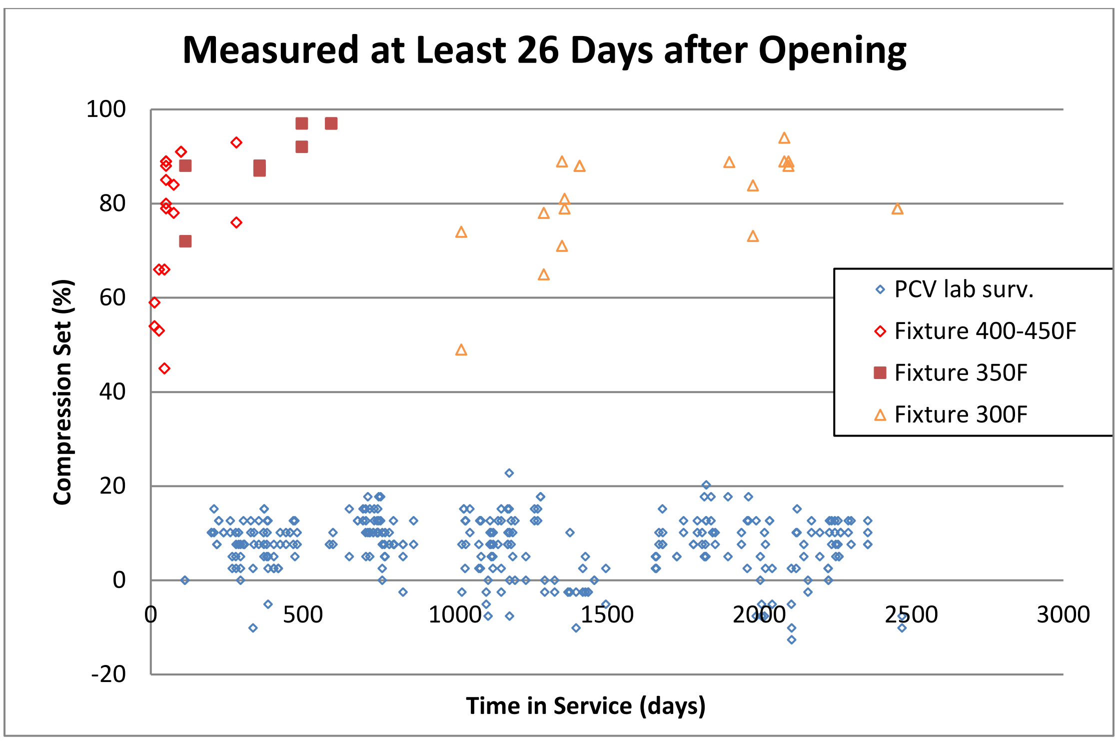


Figure 4. Lower bound trendline with CSR model predictions, showing extrapolation to the temperature of interest for O-rings in KAC storage. The extrapolated segments shown by coarse dashed lines are only partially supported by data which did not reach a failure condition. The extrapolated segments shown by fine dashed lines are currently not supported by any data.



(a) All data in this graph were taken within 1 hour of opening the PCV or fixture.



(b) All data in this graph were taken 26 days or longer after opening the PCV or fixture.

Figure 5. Compression set values for field surveillance PCV O-rings (blue) compared with O-rings from the PCV fixtures (red – orange).

Distribution

G. A. Abramczyk, 730-A
R. J. Bayer, 705-K
J. S. Bellamy, 730-A
W. L. Daugherty, 773-A
B. L. Garcia-Diaz, 773-A
S. L. Garrison, 704-2H
T. W. Griffin, 705-K
R. J. Grimm, 705-K
E. R. Hackney, 705-K
S. J. Hensel, 705-K
J. M. Jordan, 705-K
B. B. Kiflu, 705-K
D. R. Leduc, 730-A
P. L. Livengood, 705-K
J. W. McEvoy, 707-C
A. J. McWilliams, 773-A
P. M. Palmer, 705-K
W. E. Petty, 705-K
M. M. Reigel. 773-A
T. E. Skidmore, 730-A
T. T. Truong, 773-41A
K. E. Zeigler, 773-41A
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