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AGING STUDY OF EPDM O-RING MATERIAL FOR THE H1616 SHIPPING PACKAGE – Three Year Status

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DISCLAIMER

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Acronyms

ANSI	American National Standards Institute
CSR	compression stress relaxation
CV	containment vessel
DOE	Department of Energy
EPDM	ethylene propylene diene monomer
HAC	hypothetical accident conditions
NCT	normal conditions of transport
NNSA	National Nuclear Security Administration
SNL	Sandia National Laboratory
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
TTS	time temperature superposition

SUMMARY

This is the 3-year status report for tasks carried out per Task Technical Plan SRNL-STI-2011-00506 [1]. A series of tasks/experiments were performed at the Savannah River National Laboratory (SRNL) to monitor the aging performance of ethylene propylene diene monomer (EPDM) O-rings used in the H1616 shipping package. The test data provide a technical basis to extend the annual maintenance of the H1616 shipping package to three years and to predict the life of the EPDM O-rings at bounding service conditions.

O-rings were aged at elevated temperature for three years, and tests were conducted to determine (A) leak performance, (B) compression stress relaxation (CSR) behavior, and (C) mechanical properties.

Leak Performance

O-rings were aged in H1616-1 Containment Vessels (CV) at temperatures ranging from 160 to 300 °F. The vessels were helium leak tested initially and were re-tested periodically depending on the aging temperature to determine if they continued to meet the leak-tight criterion defined in ANSI standard N14.5-97 (< 1E-07 std cc air/sec) [2].

- The O-rings in the vessel aged at 160 °F remained leak-tight after three years at temperature.
- The outer O-ring installed in the vessel aged at 235 °F was leak-tight and would reseal at 788 days. This same O-ring was leak-tight after 858 days, but would not reseal at that time. The inner O-ring installed in this vessel remained leak-tight and would reseal after 1120 days, at which time it was removed from test conditions.
- The outer O-ring installed in the vessel aged at 270 °F was leak-tight and would reseal at 327 days. It was leak-tight but would not reseal after 364 days. The inner O-ring installed in this vessel remained leak-tight and would reseal after 364 days at test temperature, at which time it was removed from test conditions.
- The inner O-ring installed in the vessel aged at 300 °F was leak-tight and would reseal at 174 days. It was leak-tight but would not reseal after 196 days. The outer O-ring installed in this vessel remained leak-tight for 253 days.

The vessel tests provide a solid demonstration that the H1616 O-rings will remain leak-tight at temperatures up to 160 °F for at least three years. Significantly longer periods of leak-tight service are expected at the lower temperatures actually experienced in service.

Compression Stress Relaxation Behavior

The CSR behavior of H1616 shipping package O-rings was evaluated to develop an aging model. O-ring segments were aged at temperatures from 175 °F to 350 °F. Samples aging at the three highest temperatures reached a CSR failure criterion of 90% loss of sealing force, while samples at the two lower temperatures did not. These collective data were used to develop a predictive model for extrapolation of CSR behavior to relevant service temperatures (approximately 152 °F and below). This model indicates a service life of approximately 5 years can be expected at 152 °F (peak O-ring temperature at Normal Conditions of Transport [NCT]), with longer lifetimes expected at more realistic service conditions. This range is consistent with that suggested by available literature data, which largely derive from mechanical property and

CSR behavior. Although the relationship between CSR behavior and leak-tight performance has not been established for this design, CSR predictions are conservative relative to leak-tight performance at 235 and 300 °F.

Mechanical Properties
Baseline physical and mechanical properties of the O-rings (namely hardness, thickness and tensile strength) were found consistent with O-ring specifications [3]. Mechanical properties of aged O-rings show significant degradation can occur, but an inert atmosphere (argon backfill) greatly reduces the rate of degradation.

Recommendation
Based on the collective data presented in this report, it is recommended that the maintenance interval for the H1616 package be extended to three years. These data further suggest that a maintenance interval up to 5 years might be justifiable. However, additional testing is recommended if maintenance extension beyond three years is desired.

BACKGROUND

The H1616 is a certified "Type B" package for the transport of radioactive tritium reservoirs by the Department of Defense (DoD), United Kingdom (UK) Atomic Weapons Establishment (AWE), and Authorized Users (Savannah River Site (SRS), Pantex), Figure 1 [4a]. The H1616 package was initially designed and produced by Sandia National Laboratory and was first certified in 1992. The packages are re-certified every 5 years by DOE/NNSA/Packaging Certification Division. There are two variations of the package, designated H1616-1 and H1616-2, which are of essentially the same design, except the H1616-2 has a slightly thicker containment vessel wall and has an option feature to include a getter between the two EPDM O-rings. Both H1616-1 and H1616-2 utilize the same O-ring seal design.

Aluminum packing material used to position the tritium reservoir is added to the CV which is constructed of Type 304 stainless steel. The CV is placed inside a 16 gallon stainless steel drum (Figure 1). Two EPDM O-rings are housed between the CV lid and body and are used to seal each containment vessel. The seals are compressed nominally 25% (+/-3%) in concentric face seal grooves with a surface finish of 32 μinches or 32 RMS. No significant stretch is imposed, and no seal lubricant is used on the O-rings. All containers undergo annual maintenance, including containment vessel leak testing to verify leak-tightness to <1E-07 ref cc/sec air as per ANSI N14.5 [2].

H1616

Figure 1. H1616 container showing O-ring location (not to scale)

The H1616 O-ring compound formulation cited in Reference [4b] was assumed to be the current formulation at the beginning of this study and as listed in Reference [5] (EPDM aging data review). However, other documents [6] indicate that the O-ring formulation is currently based on Royalene® 580HT due to availability issues with Nordel® 1440/1470 polymers in 1999.

The following EPDM compounds are currently approved for the H1616 seals per Sandia specification SS395668: SR792B-80 (Stillman Seal), XP-0375 (Wynn's Precision, Inc.) and EXP-5225A (R&S Processing Co., Inc.). Both older and current formulations are given in Table 1. The effective date of the compound change is unknown to SRNL. SS395668 Change Issue K (original formulation) was effective in April, 2006, with Change Issue L (current formulation) effective in May, 2010. The cure date shown on the packaging of the O-rings provided to SRNL for testing is L08 (MFG code=BJY, lot#23881, December 2008). The O-ring composition was not analyzed or verified as part of this task.

Table 1. H1616 EPDM seal compound SS#395668 [4b, 6]

Ingredients	Original Formulation (*phr)	Current Formulation (*phr)	Approved Alternate
Nordel 1470	100		
Royalene 580 HT		100	
Zic Stick 85	5	5	Zinc oxide
N-990 carbon black	40	40	
N-539 carbon black	25	25	N-550 carbon black
Dicup 40C	12	12	
Flectol TMQ or Flectol H	2	2	Agerite Resin D (RT Vanderbilt)
SR-350 Sartomer	10	10	PLB-5405 (Synthetic Products), 13.3 phr

*phr = parts per hundred rubber (by weight)

Physical/Mechanical Properties

Thickness, inches	0.21 +/- 0.005
Hardness, Shore A	78 +/- 5
Tensile Strength, psi	1200 minimum
Elongation, percent	100 minimum
Compression Set	12% max (70 hours @ 125 °C)

The O-rings in the H1616-1 and the H1616-2 vessels are of slightly different major diameter but they have the same thickness, experience no stretch, and are compressed the same amount in service. The O-rings see essentially the same environmental conditions. Therefore, only the H1616-1 vessel was used in these tests, and the results are considered equivalent for H1616-2 vessels.

The safety analysis report for the H1616 package describes the following environmental conditions [4]. The service temperature range for the H1616 vessel is -40 °F to 169 °F. The peak vessel NCT temperature (169 °F) is based on an ambient temperature of 100 °F with solar heating. The maximum temperature at the flange closest to the O-rings is 152 °F with solar heating and 116 °F in the shade. Therefore, the maximum temperature judged to be most applicable to the O-rings is 152 °F. This is recognized to be a non-chronic condition, and typical seal temperatures are expected to be lower. Bounding radiation dose rates for the H1616 O-rings are not significant relative to seal performance.

The inner O-ring of the H1616-1 and H1616-2 without getter is credited as a containment boundary for transport purposes. For the H1616-2 with getter, the outer O-ring is credited as a containment boundary for transport purposes. The leak rate criterion for the EPDM seals is leak-tight per ANSI N14.5, or < 1E-07 ref cc/sec air, which is measured at ambient temperature. A nitrogen or argon backfill gas is approved for the H1616 containment vessels per the SARP. No lubricant is used for the O-rings.

SUMMARY OF BASELINE AGING DATA REVIEW

A literature/data review was performed on EPDM seals as part of this task. Focus was given to data believed to be the most relevant to sealing applications similar to the H1616 application. An abundance of aging data exists for EPDM elastomer in other applications, including outdoor weathering, roofing membranes and seals in fluid service, but these were not considered appropriate for comparison. The literature/data review was documented in Reference [5]. These data generally involve testing of mechanical and CSR properties, with some data on leak-tightness and oxygen consumption rates.

A summary of the EPDM seal lifetimes observed or predicted at various temperatures from different references is given in Figure 2. Direct comparison is difficult as the basis for performance or failure in each reference varies. Data points represented by solid symbols (references 13, 15 and 21 cited in the literature/data review) are not actual failures as defined by CSR or leakage, but represent samples removed from test prior to failure.

The trend line shown in Figure 2 is a lower bound of all of the reviewed failure data. This is intended to show conservative lifetimes that might be predicted from the available data, though actual lifetimes could vary. At 67 °C (152 °F), the peak temperature closest to the O-rings, the trend line suggests a lifetime of approximately 5.8 years. At 47 °C (approximately 116 °F), the peak O-ring temperature in shade (100 °F ambient), the trend line suggests a lifetime of approximately 16.4 years. Additional discussion of the literature data is provided in References [5] and [7].

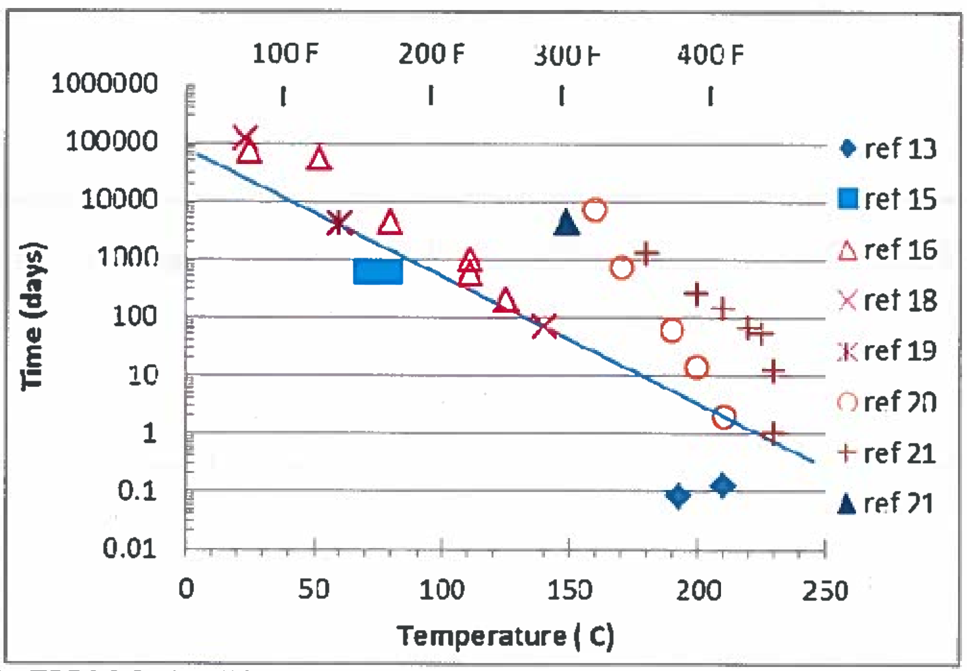

Figure 2. EPDM O-ring lifetime data from literature references [5]. The open symbols are failure points, while the solid symbols denote samples that did not fail.

EXPERIMENTAL APPROACH

Favorable O-ring performance was identified in the literature review, however, there is no substitute for real-time aging at realistic or bounding conditions. Therefore, the periodic examination and testing of actual seals from a select group of H1616 shipping packages aged at bounding conditions were used to validate assumptions and make predictions based on accelerated-aging methodology. Such examinations cannot predict time to failure until failure is actually observed, but advance notice of degradation may be provided. A description of the tests performed is included.

H1616 Vessel Aging and Leak Performance [1]

Vessel aging and leak tests were performed using three full-size containment vessels. Since aging at room temperature is not expected to produce leak failure in the time allotted for testing, these test CVs were aged at conditions that bound operating temperatures with the intent to drive the O-rings to failure within the test period. This approach used actual vessels and the leak-tight acceptance criterion for service. Failure of the CV seal is a leak rate greater than the leak-tight criterion of 1E-07 cc/sec dry air at 25 °C and 1 atm pressure differential. The vessels were heated externally such that O-ring temperatures were maintained at 160 °F, 235 °F, 270 °F or 300 °F. The use of four temperatures provides confidence in the extrapolation of results to support life prediction model development. These temperatures are based on bounding service temperatures and bounding continuous temperature ratings as indicated from typical EPDM compounds. Again, the 160 °F temperature is bounding for normal service as it accounts for solar heating superimposed on a 100 °F ambient temperature, neither of which are chronic conditions.

Each CV was placed in a round stainless steel support pan within an aluminum test stand (Figure 3). Two Watlow® heaters, 6 inches wide x 20 inches long were wired in series and wrapped around the upper half of the vessel. These heaters provided 300 watts (2.5 amps) of heating to the vessel. 24 layers of aluminum foil were placed between the heater and the vessel OD to accommodate the heater length without overlap. The foil, heaters and one layer of silica insulation were secured to the vessel with band clamps. Additional silica insulation was added to the side, top and bottom of the vessel to help regulate vessel temperature. Additional band clamps held the side insulation in place. Ring supports, bolts, and braces were fabricated to hold the vessel secure in the stand (Figures 3, 4).

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[1] Although the Task Technical Plan originally identified the leak testing as Scoping data, it was performed in accordance with Technical Baseline data requirements at the request of the customer (Paul Lari, Sandia National Lab, December 2011).

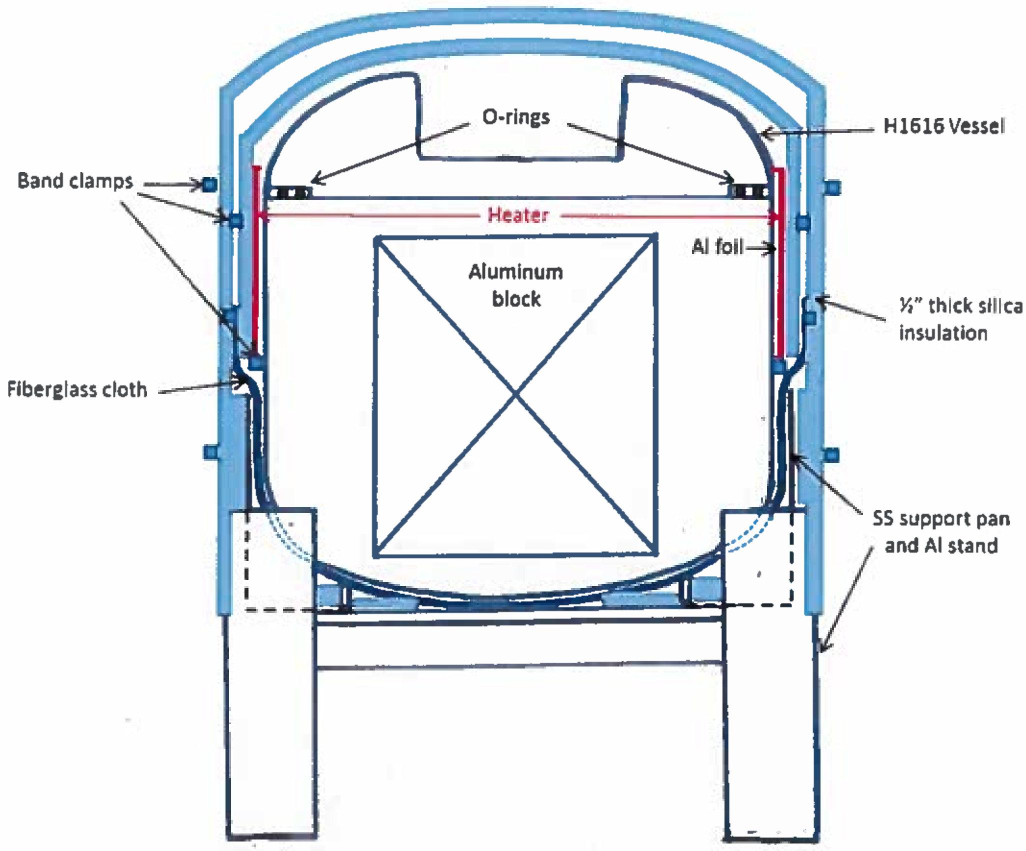

Figure 3. Cross section sketch of vessel and insulation arrangement

Figure 4. Containment vessel leak test setup

Four thermocouples were placed at 90 degree intervals between the aluminum foil and the outside of vessel, approximately 1.5 inches from the top surface of the CV lid. Thermocouples were verified with a calibrated Dry Well Calibrator and thermocouple reader. The heaters were controlled by a LabView® software program. One thermocouple from each vessel was used to control the heater, while two additional thermocouples provided assurance that the vessel was heating uniformly. The fourth thermocouple provided a signal to an over-temperature controller which prevented an unplanned thermal excursion.

Tritium Packaging Operations personnel initially replaced the sample valve O-ring, the sample fitting O-ring, and the hex head plug O-ring on all three vessels after the trial runs and before new CV O-rings were installed to begin testing. The new CV inner and outer O-rings were installed by SRNL personnel.

A thin strip of 0.005 inch thick stainless steel shim was placed on the vessel flange between the O-rings to prevent a possible metal-to-metal seal from masking the leak behavior of the O-rings. Initial tests were performed to show permeation (gradual increase in detector response over time) to prove helium access to/through the O-rings. This step eliminates the possibility of false positive results (no leakage path). In addition, vessel O-ring temperature/vessel gradient heat-up trials were performed to determine the correct temperature control settings, as shown in Table 2.

Table 2. Vessel O-ring/control temperature comparison

Vessel Body ID	Vessel Lid ID	Desired O-Ring Aging Temperature (°F)	Vessel Exterior Control Temperature (°F)
13784	13774	160	165
11274	11296	235	241
13772	13896	270	280
13772	13896	300	313

An aluminum block (8 inches diameter x 10 inches long) was placed inside each vessel to reduce the amount of gas space and facilitate leak testing.

Argon (at least 99.99%) was used as a backfill gas. Leak tests were performed periodically, after the vessel cooled down to room temperature. Leak-tightness was verified per ANSI N14.5-97 with a hood test performed by a certified Level II Leak Test Specialist. The vessel was bagged, and the vessel interior and exterior were evacuated and backfilled with helium (Figure 5). A helium mass spectrometer leak detector was connected to the test volume between the two O-rings to detect leakage from either O-ring. When necessary, vessel connections were re-configured to test each O-ring individually.

To understand the effect of oxidation due to the isolation provided by the O-ring groove and lid design, the 300 °F vessel was retested with new O-rings and no inert gas backfill.

Figure 5. Hood test on containment vessel

The test CV's were opened periodically in conjunction with leak testing, keeping the same O-rings in service. These openings were intended to prevent artificial "sticking" of the O-rings, which could provide a false positive result, and provide a more realistic representation of actual in-service practice. This artificial sticking has been observed in other studies, particularly at higher aging temperatures [8].

Compression Stress Relaxation Testing

Compression stress relaxation (CSR) tests were performed on the H1616 O-rings at several aging temperatures to develop an aging model based on sealing force decay. In the absence of fluid or significant radiation exposure, thermo-oxidative aging and sealing force decay is expected to be the most relevant degradation mechanism for the H1616 O-rings. CSR tests are a common industry method for evaluating the mechanical performance of seals over time at various aging temperatures. CSR tests were performed per ASTM D6147, using the periodic measurement approach.

A single H1616 O-ring was carefully cut into several segments approximately 0.75 inch long, with three segments placed into each CSR jig (Shawbury-Wallace type). The CSR jigs were initially developed to compress large disc samples, but several studies have shown that diffusion-limited oxidation effects are possible with larger samples, and disc samples do not ideally represent actual O-ring behavior, even if made of the same elastomer compound [9]. Three segments were selected to balance the contact area of the compression platens and the surface area of the exposed material. The segments were then compressed approximately 25% from 0.21 inch cross-section thickness to approximately 0.1575 inch as in the H1616 design. A fabricated shim was used to verify compression.

The jigs with compressed samples were placed into aging ovens, with three jigs per temperature for repeatability. Aging temperatures (175 °F, 235 °F, 270 °F, 300 °F and 350 °F) were selected to bound expected normal service temperatures and to challenge the seals within a reasonable aging period. Excessive aging temperatures should be avoided, but failures must be observed in order to predict failure at more reasonable service temperatures. Prior to compressing samples in the jigs, the break force for each jig was determined using the relaxometer. The break force is subtracted from later total force measurements to give sealing force measurements only.

After initial compression and heat up, but prior to aging, the jigs were placed into the relaxometer to obtain an initial sealing force measurement. During aging, the CSR jigs were periodically removed from the aging ovens and placed in the relaxometer (Figure 6) for measurement of sealing force, which is the force required to deflect the compressed seals approximately 0.0001-0.0002 inch. As the elastomer relaxes and ages over time, this counterforce value is reduced. During periodic sealing force measurements, five sequential readings were typically taken, with time and temperature of the jig monitored. Measurements are taken as quickly as possible to capture the sealing force at the aging temperature, rather than waiting for the fixture to cool to room temperature. CSR values can be taken at any temperature, but should be taken as close to the aging temperature as possible for direct comparisons and interpretation of sealing behavior at the actual temperature of interest. Using the average sealing force value for all five measurements per test period minimizes concern over sealing force fluctuations with temperature due to cooling effects.

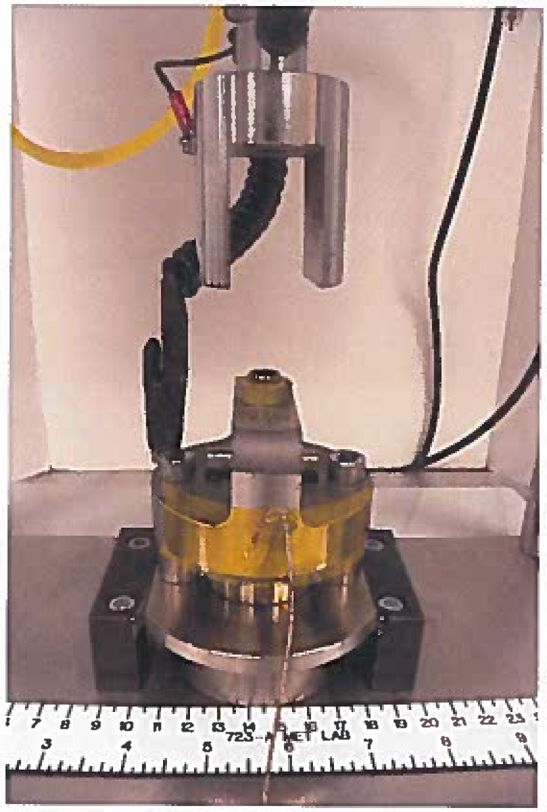

Figure 6. CSR jig in relaxometer for sealing force measurement

The main goal of the CSR tests is to age the seals to a significant degradation point (loss of sealing force) at multiple temperatures, which allows development of an aging model based on Arrhenius theory and Time Temperature Superposition (TTS) of the collective data. Ideally, it is desirable for failure to be reached at all aging temperatures within the planned aging period for model development. For CSR testing, a 90% loss in sealing force (F/F_o=0.10) was selected as the failure or end-of-life parameter. This parameter has been used for other critical seal applications such as nuclear weapon components [10]. However, it is emphasized that CSR failure does not inherently mean that the seal is no longer functional or leak-tight. The use of 90% sealing force loss leaves 10% of the initial sealing force remaining for margin. This is expected to be more than sufficient for most static seal designs. The relationship between CSR and leak rate has not been established for this design and was not part of the scope for this task.

One advantage of the TTS principle is that all of the data can be used in the aging model, rather than using only a few time to failure data points. Another advantage of the TTS principle is that if the same degradation mechanism exists over the full aging temperature range and at the expected service temperatures, the aging model can be translated to any temperature of interest.

Baseline O-Ring Characterization

Confirmatory measurements were previously performed on a limited number of O-ring samples to verify the O-ring physical and mechanical properties were consistent with the O-ring specification. Thickness measurements were verified at four equally-spaced points along the circumference in both top-to-bottom and side-to-side orientations. Hardness measurements were verified with a calibrated Durometer Shore Hardness tester measured on the top of the O-ring at 5 points along the circumference using two scales, Durometer Shore A (per specification) and Durometer Shore M for comparison. Durometer Shore M scale (microhardness) is required per ASTM D2240 for O-rings less than 0.25 inch thick. M-scale and A-scale values tend to be very close but there is no direct conversion.

RESULTS / DISCUSSION

H1616 Vessel Aging and Leak Testing

Three containment vessels which had seen actual service were transferred to SRNL. The vessels were known to have been fabricated per specification and were not removed from service due to any known defects or non-conformances. Each vessel received a baseline leak test by a certified Level II Leak Test Specialist, and began heating at the target temperature in January 2012. Leak tests were performed on each vessel after one month and at varying intervals thereafter, depending on the aging temperature (Table 3). After each leak test the vessel lid was removed, a visual inspection was performed on the vessel body and lid O-ring grooves, and visual and hardness tests were performed on the inner and outer O-rings (Figure 7). A post-examination leak test was then performed to confirm the O-ring's capability to reseal had not been compromised.

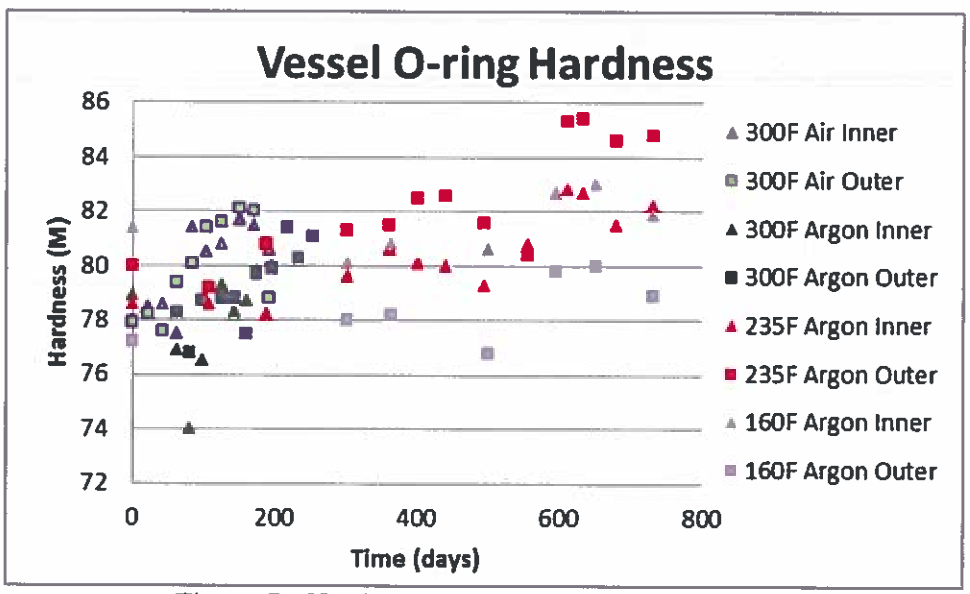

Figure 7. Hardness of O-rings in containment vessels

Table 3. Vessel leak test intervals (days at test temperature)

300 °F Air	300 °F Argon	270 °F Argon	235 °F Argon	160 °F Argon
0	0	0	0	0
22	25	40	25	25
43	47	76	108	109
64	64	118	189	303
85	81	158	302	363
105	99	194	362	403
127	127	240	401	499
152	145	288	440	595
171	161	327	495	650
193	174	364* outer fail	556	664
both o-rings	196* inner fail	inner leak tight	612	734
leak tight	218		633	821
	234		679	910
	253* outer fail		732	1001
			788* outer fail	1097
			856	both o-rings
			904	leak tight
			959	
			1033	
			1074	
			1120	
			inner leak tight	
*At failure, O-ring was leak tight but would not reseal				

After approximately 64 days, the O-rings in the vessel aging at 300 °F became sticky and an oily residue was observed on the O-rings and in the O-ring grooves. The O-rings and the O-ring grooves were cleaned using isopropyl alcohol wipes to remove the residue, and the same O-rings were placed back in the vessel. Another leak test was performed to verify that leak-tightness was not compromised by the inspection. After 85 days at 300 °F, the inner and outer O-rings were beginning to "flatten" and lose their round shape. The vessel remained leak-tight and the O-rings resealed after 174 days. The vessel was leak-tight, but failed to reseal after 196 days at temperature.

For the 300 °F vessel retested without inert gas backfill, a sticky, oily residue was observed on the O-rings and in the grooves, and the O-rings became square during this test as well. This additional test successfully demonstrated no significant decrease in O-ring service life resulting from the absence of inert backfill.

After approximately 40 days, the outer O-ring in the vessel aging at 270 °F was sticky. After approximately 76 days, both O-rings had become square and deposits were observed in the vessel grooves. The stains/deposits were cleaned after the visual, hardness, and thickness measurements and another leak test was performed to verify that leak-tightness was not compromised. After 364 days at test temperature when the outer O-ring reached an end of life condition, various surface indications were observed which could impact resealing of the O-ring (Figure 8).

After approximately 99 days, the O-rings in the vessel aging at 235 °F started showing some evidence of flattening, however no oily deposits were observed on the O-rings or in the O-ring grooves until after approximately 302 days at temperature. As was done with the 300 °F vessel, the O-rings and the O-ring grooves were cleaned using isopropyl alcohol wipes to remove the residue, and the same O-rings were placed back in the vessel. Another leak test was performed to verify that leak-tightness was not compromised by the inspection.

After 303 days, the O-rings in the vessel aging at 160 °F became a little sticky, however based on visual observation there was no residue and no flattening of the O-rings even after 1097 days.

Figure 8. Outer O-ring in 270 °F vessel showing surface indications

Compression Stress Relaxation Testing

CSR jigs with compressed H1616 O-ring segments were placed in aging ovens at the following temperatures: 175 °F, 235 °F, 270 °F, 300 °F and 350 °F. CSR measurements were recorded approximately every 30 days, depending on the aging temperature. A shim fabricated to 75% of nominal O-ring thickness (0.1575 inch) was used to check the degree of compression in each jig. All O-ring segments came from a single O-ring so thickness for each segment was assumed to be constant. These original samples provided the following results.

- After 9 weeks aging time, some of the 350 °F CSR O-ring segments were observed to be split vertically across the compressed cross-section. The 350 °F samples reached an end-of-life condition (90% loss of initial sealing force) after 11 weeks aging time.
- The 300 °F samples reached an end-of-life condition after 21 - 23 weeks aging time. These samples did not exhibit any cracking.
- As a result of early failures at 350 °F, additional O-ring segments were placed into the same three CSR jigs from the 350 °F tests and aged at 270 °F to maximize data for aging model development. These jigs reached an end-of-life condition after 37 - 43 weeks aging time. The 270 °F temperature was selected as an intermediate temperature between the 300 °F and 235 °F aging temperatures for convenience.
- The 235 °F samples experienced a loss of initial sealing force of approximately 75% after 68 weeks aging time. At that time, the 235 °F aging oven experienced an unplanned temperature excursion, compromising any further test data from these samples, and they were removed from test.
- The 175 °F samples remained in test through September 2015, with a loss of initial sealing force of 11 – 39% after 3.4 years aging time.

A second set of samples was initiated to supplement the original set of data and resolve inconsistencies in the results. All of these additional samples were taken from the same O-ring as the original samples, with one exception noted below.

- Following the unplanned temperature excursion of the 235 °F oven, additional samples began aging at 270 and 300 °F, with three jigs at each of these temperatures. These samples were intended to rapidly age to approximately 75% loss of initial sealing force, and then continue aging at 235 °F to fill in the remaining portion of the original 235 °F decay curve from the original samples. These samples reached an end-of-life condition and were removed from test. These samples provide additional valid data for CSR behavior at 270 and 300 °F. However, due to the unique thermal stress history caused by changing the aging temperature, their behavior at 235 °F did not match that of the original samples aged at that temperature (Figure 9), and was considered inappropriate for further use.
- After it became apparent that the replacement samples aging at 270 and 300 °F prior to shifting to 235 °F did not produce the same behavior at 235 °F, three jigs with new samples began aging at 235 °F. These samples remained in test through September 2015, with a loss of initial sealing force of 50 – 56% after 56 weeks aging time.
- Based on comparison of the original CSR data at 270 and 300 °F to that from the second set of samples at the same temperatures, it became apparent that these two sets of data were not consistent with each other. To further explore this condition, additional samples began aging in three jigs at 350 °F. These samples were removed from test after 4 - 9 weeks aging time after reaching or nearing an end-of-life condition. Unlike the original 350 °F samples, they did not exhibit any cracking. Two of these jigs used segments from the same O-rings as all the prior CSR testing. However, this depleted all remaining material from this O-ring, so the third jig used segments from a different O-ring.

Sealing force decay curves (CSR curves) for all samples (except the 235 °F samples following higher temperature pre-aging) at aging temperatures are given in Figure 10. In these curves, the sealing force for each jig is normalized to the initial maximum sealing force.

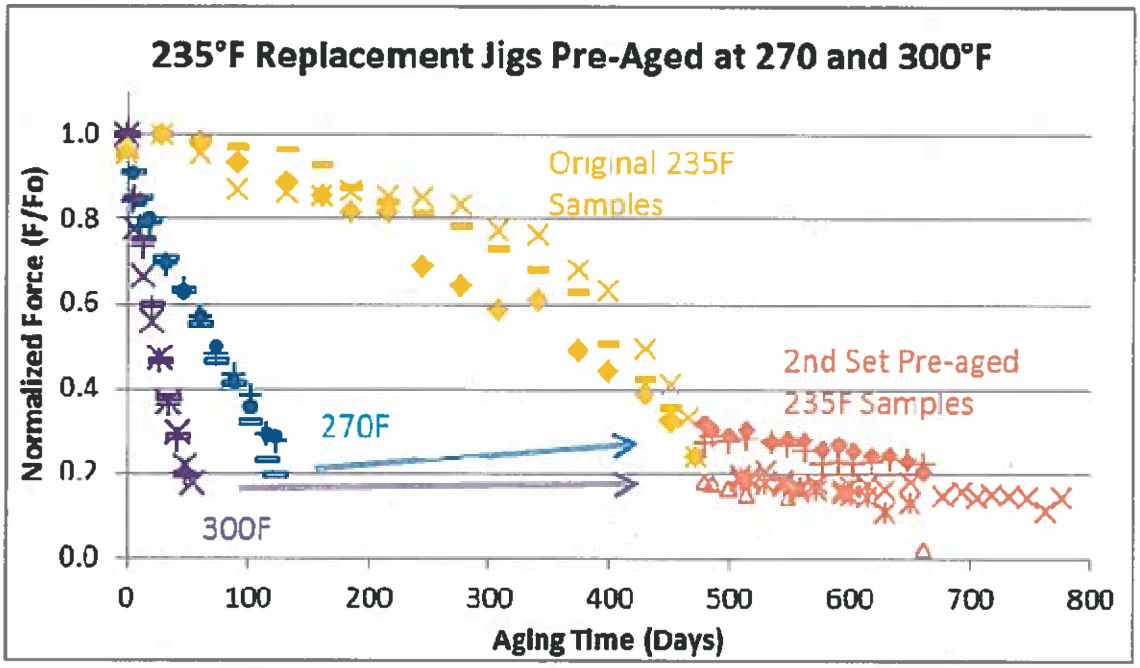

Figure 9. CSR data for the 235 °F original and replacement samples, including accelerated pre-aging at 270 and 300 °F. The time scale for the 235 °F aging is shifted to align with the 235 °F original jig data.

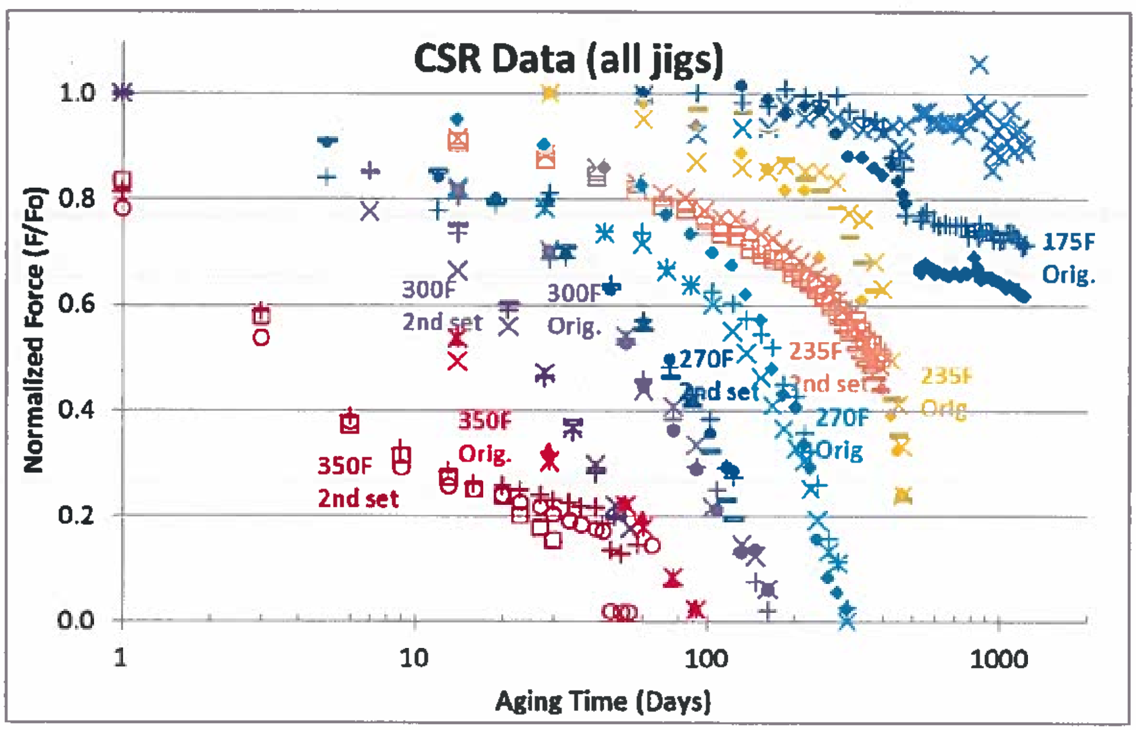

Figure 10. CSR data at all aging temperatures

Data from the original samples provided the basis for the analysis reported in Reference [7]. The subsequent data, labelled "2nd Set" in Figure 10, were generated after O-ring aging and testing were consolidated into a new laboratory building. For reasons that are not yet understood, the collective data from the original laboratory and from the second laboratory each show a degree of internal consistency, but do not compare favorably to each other. The 175 °F samples are the only samples to be aged and tested in both locations. They show a distinct change in behavior after aging for approximately 500 days, which is when they were transferred to the second laboratory.

Numerous actions were undertaken to understand the difference in CSR behavior between the two data sets, including:

- The same equipment was used to measure the sample counterforce in each laboratory, and the same test protocols were followed. While different personnel made the measurement in each laboratory, the same person provided training to the test personnel at both laboratories, and subsequently observed measurements to verify they were conducted the same in each laboratory.
- Oven temperatures were tracked with calibrated thermocouples, and were consistently observed to match the desired setpoint.
- The magnitude of potential temperature gradients within each oven was small, based on the recorded jig temperatures. Three jigs are aged at each temperature, and are located near the center with the oven verification thermocouple. Any significant gradient would appear as a difference between these four readings.
- It is observed that the relative difference in the rate of counterforce decay from the two data sets changes with aging temperature. This indicates that the overall difference in behavior is not the result of a simple bias in aging temperature, since such a bias would similarly increase or decrease at all temperatures.
- There were no obvious environmental factors (chemical vapors, ozone, radiation, etc) in either laboratory which might affect the degradation rate of the EPDM material.
- O-ring segments aged at 270 °F in each laboratory, and non-aged segments that had been stored in each laboratory were examined by Fourier-transform infrared (FT-IR) spectroscopy [11]. This did not identify any specific difference in composition or internal structure that would correlate to varying CSR behavior.
- Measurement of the relaxation modulus using dynamic mechanical analysis (DMA) was also performed on these segments but the data available from this limited effort were inconclusive in showing whether these samples are comparable to each other.
- In parallel with testing the H1616 O-ring segments, parallel tests were also being performed on Viton® O-rings for a separate program. These tests also included aging in the two laboratories. However, the Viton® samples did not display a similar change in behavior. As was done with the EPDM samples, additional Viton® samples were also re-aged in the newer laboratory at higher temperatures and produced data comparable to the original Viton® data.
- The data were reviewed by an internal panel of subject matter experts, and no specific cause for this behavior was identified.

Additional O-ring aging mechanical tests

The primary degradation mechanism for the H1616 O-rings is expected to be thermo-oxidation [4]. It was suspected that the argon backfill limits oxidation of the O-rings, thereby significantly extending the useful service life. As a check on this effect, two sets of O-ring segments were placed in each of the aging furnaces in order to track changes in mechanical properties. One set of these samples was exposed to air, while a second set was placed inside an enclosed container backfilled with argon. The observed changes in hardness are shown in Figure 11 for each atmosphere.

The tensile strength and elongation for O-rings aging at 300 and 350 °F decreased significantly faster than the O-rings aging at 235 and 175 °F, and this effect was magnified for samples aged in air relative to those aged in argon.

Details of these tests have been reported in Reference [7]. They indicate that an argon backfill can significantly reduce the degradation rate of the O-rings, which is intuitive. While this behavior suggests that the O-ring performance in service life is likely extended compared to O-rings aged in air, the extent to which this behavior can be credited has not been determined.

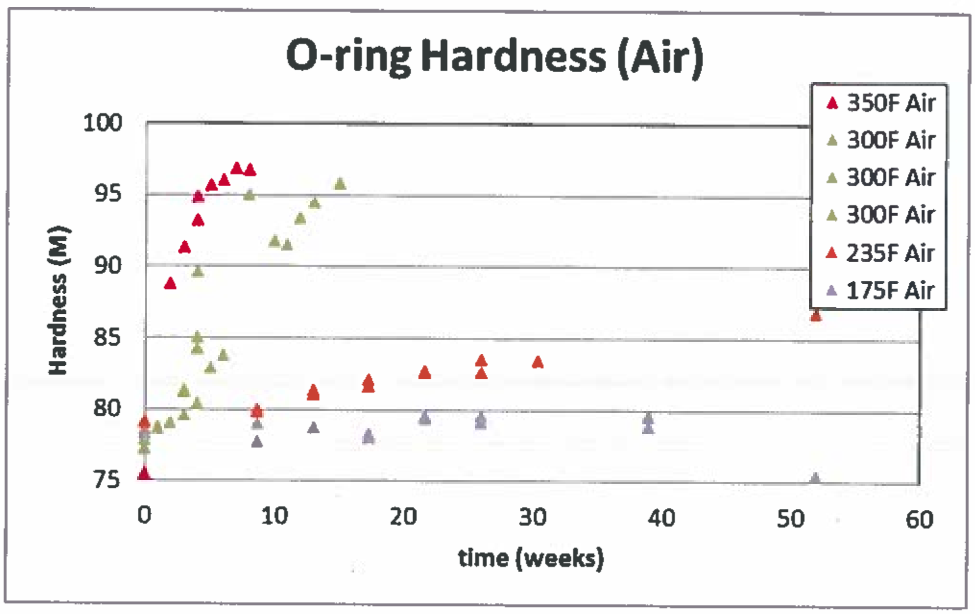

Figure 11a. Hardness of O-ring segments aged in air

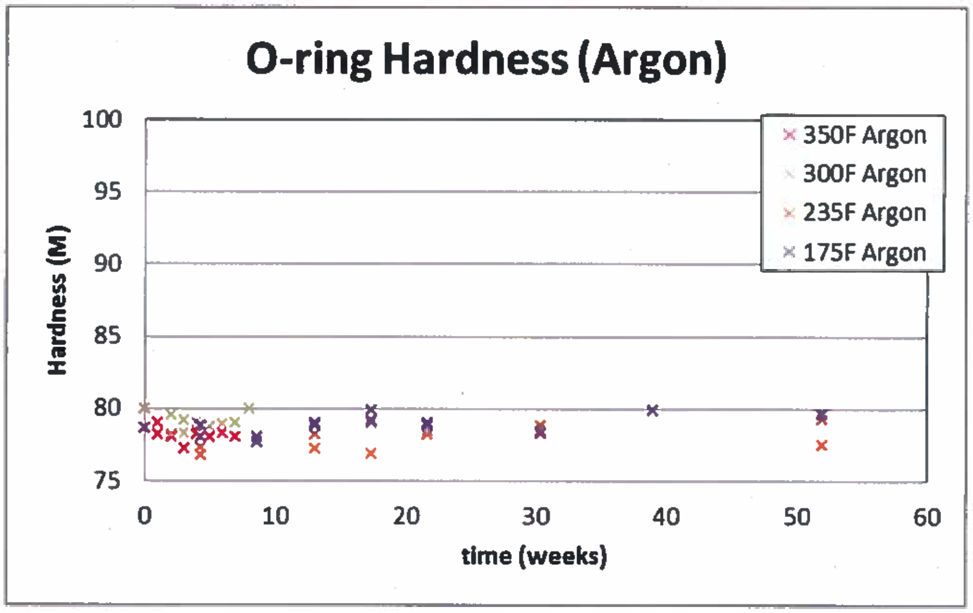

Figure 11b. Hardness of O-ring segments aged in argon

Baseline O-Ring Characterization

Confirmatory measurements on twelve new O-rings were performed to verify the material properties were consistent with existing published vendor data (Table 4). All O-ring thickness measurements were within the specified range of 0.21 +/- 0.005 inches. All O-ring hardness measurements were within the specified range of 78 +/- 5 "A" scale. Both the thickness and hardness measurements are non-destructive.

Since the tensile strength and elongation tests are destructive, a limited number of O-rings were tested using the Instron Tensile Tester. The O-rings were held in padded grips and pulled at a rate of 20 inches per minute. The O-rings tested met the specified requirements of greater than 1200 psi tensile strength, and greater than 100 percent elongation. The basis for the requirements in the O-ring specification is unknown, but the values are typical for similar compounds.

Table 4. Baseline confirmatory measurements of O-rings

Sample ID:	Average Thickness (inches) (top – bottom)	Average Thickness (inches) (side – side)	Average Hardness (A)	Average Hardness (M)	Tensile Strength (psi)	Elongation (%)
1 -1 Outer	0.2090	0.2120	80	80	2267	335
1 -1 Inner	0.2076	0.2120	80	80	2017	294
2 -1 Outer	0.2090	0.2123	80	80		
2 -1 Inner	0.2085	0.2113	80	79		
3 -1 Outer	0.2096	0.2123	80	80		
3 -1 Inner	0.2088	0.2121	79	79		
4 -1 Outer	0.2091	0.2118	80	77		
4 -1 Inner	0.2089	0.2120	80	81		
5 -1 Outer	0.2093	0.2113	80	78	2116	392
5 -1 Inner	0.2086	0.2126	80	79	2137	340
6 -1 Outer	0.2099	0.2123	80	79		
6 -1 Inner	0.2089	0.2130	80	79		

SERVICE LIFE DISCUSSION

No direct correlation has been established between CSR behavior and O-ring service life based on leak-tightness. While it is intuitive that a significantly reduced sealing force should correlate to an increased likelihood of leakage (especially for a dynamic application), very little data are available to indicate the actual sealing force needed to maintain leak-tightness. Reference [12] indicates the threshold sealing force to maintain a leak-tight seal is about 1 N/cm, but this value will change depending on the seal design. The criterion of 90% loss in initial sealing force has been adopted in some studies as a failure parameter, and is also used in the current effort. The CSR samples exhibit an initial counterforce of approximately 90 – 120 N/cm at room temperature, and higher values after heating to their aging temperature. They still exhibit >9 N/cm counterforce when the 90% loss in sealing force criterion is reached. Compared to the Reference [12] data, this indicates that this criterion maintains some margin for the H1616 O-rings, especially given the static nature of this seal.

A further consideration in use of the CSR data in a predictive model for leak-tight service life is the test environment. The CSR samples are aged at elevated temperature in an air environment, while the vessels are maintained with an inert gas backfill, limiting the O-ring exposure to oxygen. The hardness and tensile data show a significant difference in degradation rate for the two environments (air vs argon). The O-rings are further isolated within the vessel grooves, such that there would be very little replenishment of oxygen even without an inert backfill. The CSR data have been analyzed to develop a failure model for the H1616 O-rings. The limited vessel leak test failure points are then used to validate the results of the CSR model to develop additional confidence in its applicability.

In Reference [7], the CSR data were analyzed using time-temperature superposition, a technique that works best when the decay curves at each temperature share a common shape. It was recognized that there is some variation in the decay curve shapes, which often indicates a change in degradation mechanisms may be occurring within the range of aging temperatures. This conclusion is further supported by data from Sandia National Laboratory (SNL) on a similar EPDM compound, which shows an approximately 30% decrease in the activation energy for aging temperatures below 230 °F (110 °C) [9].

Given the shortcomings of applying time-temperature superposition to the H1616 CSR data, and the variation between the original and second CSR data sets, the following approach will be used in analyzing the data:

- Both the original CSR data [7] and the SNL data [9] indicate a change in activation energy occurs near 230°F. Therefore, the 175 °F data will not be grouped with the higher temperature (235 °F and greater) data for analysis.
- Based primarily on the SNL data, and consistent with the qualitative trend for the original 175 °F data [7], it will be assumed that the activation energy decreases by approximately 30% below 235 °F.
- The 350 °F data are extreme in regards to the temperature rating of the EPDM material, as demonstrated by some of the original samples splitting at this temperature, and the significantly different decay curve shape for the second set of samples aged at this temperature. Therefore, the 350 °F data will be excluded from the analysis. The impact of this omission will be minimal since these highest temperature data are the furthest removed from the temperature range of interest, and therefore would be least relevant in extrapolating the behavior to lower temperatures.
- The CSR data will be treated as two distinct data sets (original, second), and each set will be analyzed separately. In the absence of a rationale for favoring one set over the other, the more conservative results will be used to develop service life estimates for the H1616 O-rings.
- Given the range of shapes for the CSR decay curves, a variation will be introduced to the time-temperature superposition approach. Rather than comparing and shifting entire decay curves to obtain the temperature dependence of the CSR behavior, only the time to failure will be considered. For the case where the decay curves have the same shape, this approach is equivalent to time-temperature superposition. However, when the curve shapes differ, using the entire decay curve to compare behavior becomes counterproductive. Using only the time to failure will focus on the portion of data most relevant to service life.

The time for each jig to reach the failure criterion is summarized in Table 5. Since the decay curves display a range of shapes and some of them do not extend all the way to / past the failure criterion, several different approaches were used to identify a failure time for each jig. The specific basis used to estimate the failure time for each jig is also identified in Table 5.

Table 5. Summary of times to reach the failure criterion for each CSR jig

CSR jig	Temperature (F)	Test dates	Failure time (days)	Basis for failure time *	Average failure time (days)
Original CSR data					
11H	350	Nov 2011 - Feb 2012	76	A	76.3
65H	350	Nov 2011 - Feb 2012	76	A	
88H	350	Nov 2011 - Feb 2012	77	B	
81H	300	Nov 2011 - Apr 2012	142	A	147
89H	300	Nov 2011 - Apr 2012	151	A	
84H	300	Nov 2011 - Apr 2012	148	A	
11H	270	May 2012 - Mar 2013	280	A	272
88H	270	May 2012 - Mar 2013	277	A	
65H	270	May 2012 - Mar 2013	259	C	
83H	235	Nov 2011 - Mar 2013	491	D	502
85H	235	Nov 2011 - Mar 2013	499	D	
86H	235	Nov 2011 - Mar 2013	516	D	
80H	175	Nov 2011 - Mar 2013	1255	E	1255
82H	175	Nov 2011 - Mar 2013	1255	E	
87H	175	Nov 2011 - Mar 2013	1255	E	
Second set of CSR data					
9	350	Apr 2015 - May 2015	37	D	53.3
83H	350	Apr 2015 - Jun 2015	54	D	
86H	350	Apr 2015 - Jun 2015	69	D	
64	300	May 2013 - Jun 2013	57	D	58.7
84H	300	May 2013 - Jul 2013	62	D	
89H	300	May 2013 - Jun 2013	57	D	
20	270	May 2013 - Sep 2013	163	D	155.3
18	270	May 2013 - Sep 2013	162	D	
7B	270	May 2013 - Sep 2013	141	D	
7B	235	Jul 2014 - present	808	F	808
64	235	Jul 2014 - present	808	F	
89H	235	Jul 2014 - present	808	F	
82H	175	Mar 2013 - present	13500	G	12933
80H	175	Mar 2013 - present	16000	G	
87H	175	Mar 2013 - present	9300	G	

* Definition of failure time basis codes:

A – Linear fit to last (linear) portion of CSR decay curve

B – Linear fit to the last 2 points of the CSR decay curve using the slope from the other jigs at this temperature

C – Linear fit to the 2 points that straddle the failure criterion

D – Linear fit to last portion of CSR decay curve plus extrapolation to failure criterion

E – 2.4 times average failure time at 235 °F based on average shift factor needed to align the 175 °F behavior to jig 85H (235 °F)

F – 5.2 times average failure time at 270 °F based on average shift factor needed to align the 235 °F behavior to jig 85H (270 °F)

G – Extrapolate the average slope from the limited decay curve from 100% to 10% (gives an upper bound estimate of failure time)

The times for the CSR decay curves to reach the failure criterion are plotted in Figure 12 as a function of reciprocal temperature. The activation energy is a parameter that describes the temperature dependence of a process (such as the time to failure), and remains constant so long as the process is governed by a consistent mechanism(s). Exponential trend lines fit to these data (for the 235 to 300 °F range) can be used to get the activation energy for this temperature range using the Arrhenius relationship, which is described by the equation:

$t = A \exp(-E_a / RT)$

where t = time to reach failure criterion
- A = constant
- E_a = activation energy
- R = ideal gas constant, 8.3145 J/K-mol
- T = absolute temperature (K)

In the Figure 12 trend line equations, the coefficient in the exponential term is equal to the activation energy divided by the ideal gas constant. Therefore, the original data set gives an activation energy estimate of (5509 * 8.3145 =) 45800 J/mol. Similarly, the second data set provides an estimate of activation energy of (11890 * 8.3145 =) 98900 J/mol.

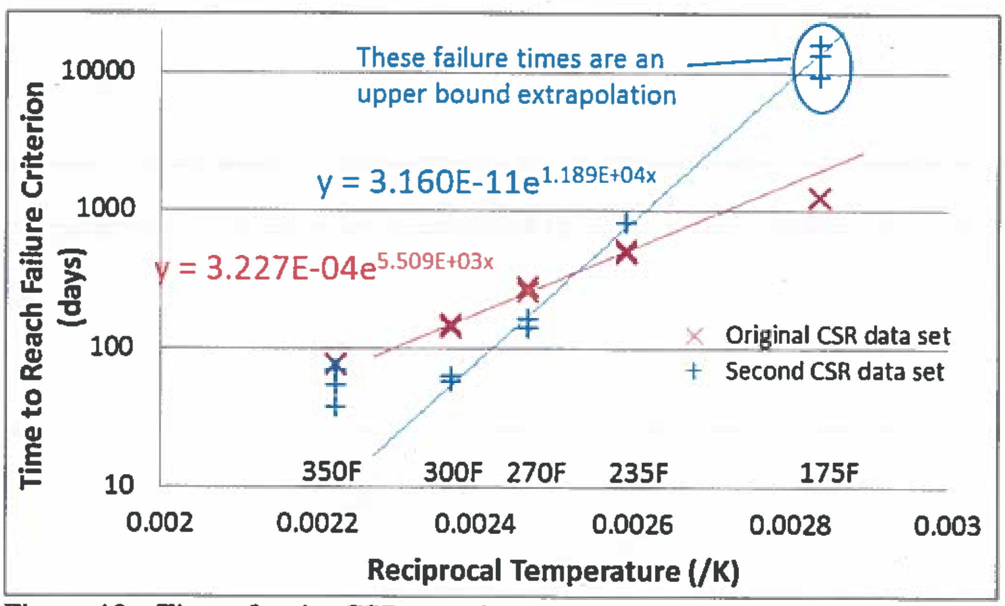

Figure 12. Times for the CSR samples to reach the failure criterion (from Table 5) plotted as a function of reciprocal temperature. The trend lines are developed separately for data from the two lab locations using the 235 – 300 °F data only.

Given the likelihood that the activation energy decreases near 230 °F, an additional trend line is added to the original set of CSR data, as shown in Figure 13. This shows the reduced slope suggested by the 175 and 235 °F data. From the coefficient in Figure 13, this gives an activation energy estimate of 31100 J/mol. This value is 32% lower than the activation energy for the higher temperature data, in good agreement with the trend indicated by the SNL data. In the second CSR data set, the failure times for the 175 °F data are identified as upper bound estimates based on the approach used to derive these estimates from a very limited portion of the decay curve. Based on the overall shape of the decay curves at other temperatures, it is judged that the actual failure time at 175 °F might be significantly less than these upper bound estimates. An additional trend line in Figure 13 shows the effect of reducing the activation energy for the second data set by 32% below 235 °F. This trend line suggests a failure time at 175 °F of approximately 5800 days (15.9 years).

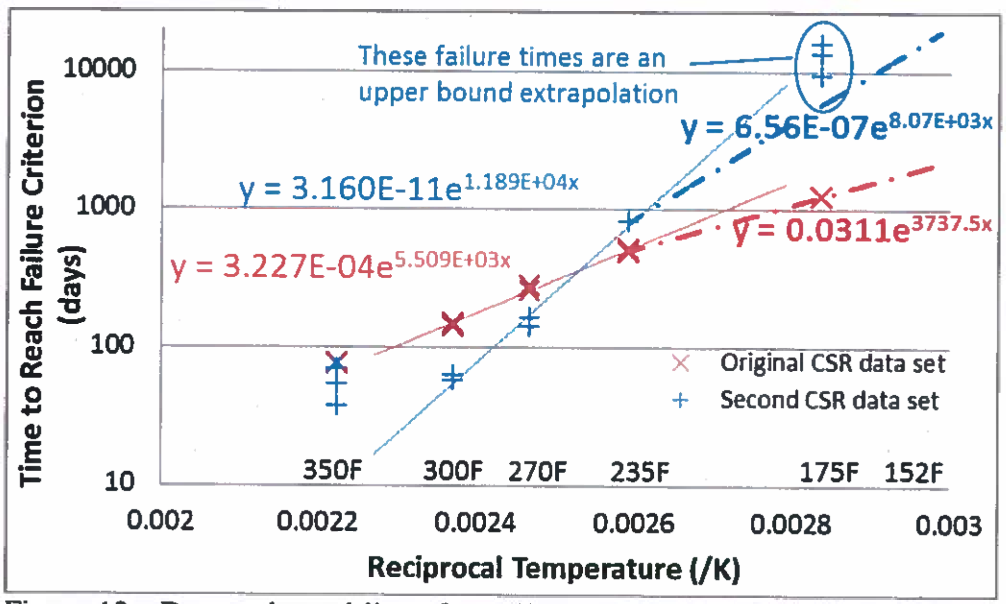

Figure 13. Data and trend lines from Figure 12, with the addition of trend lines below 235 °F (dashed lines) as suggested by the SNL data.

The temperature dependence of the CSR failure time is described by these activation energy estimates. Separate models are available from the two data sets. In each model, the behavior for temperatures between 235 and 300 °F is described by the trend lines in Figure 12, while the behavior at lower temperatures is described by the additional dashed trend lines in Figure 13. At the H1616 maximum O-ring temperature of 152 °F, the original data set model indicates a failure time of 1868 days, or 5.1 years. The second CSR data set predicts a failure time of 13600 days, or 37 years. Of these two model estimates, the more conservative value of 5.1 years is recommended for consideration.

It is emphasized that these failure times require chronic exposure at the bounding temperature, which is conservative to a typical shipping/transport application. In the above analysis, it was assumed that the times for the CSR samples to reach the failure criterion are representative of the leak-tight service life of the H1616 vessel. This assumption can be examined with the vessel leak test data. In Figure 14, the vessel leak data are compared to the CSR data. The trend line provides an activation energy estimate of 55400 J/mol within the temperature range of 235 – 300 °F. If it is assumed that the activation energy for leak tightness decreases below 235 °F similarly to that for the CSR data, then the service life estimate at 152 °F based on the vessel leak data is 11.4 years. Within the temperature range of the vessel leak data (235 – 300 °F), the CSR-based service life model based on the original CSR data is conservative to actual leak-tight behavior. However, without additional vessel failure data, the degree of conservatism at lower temperatures cannot be assessed.

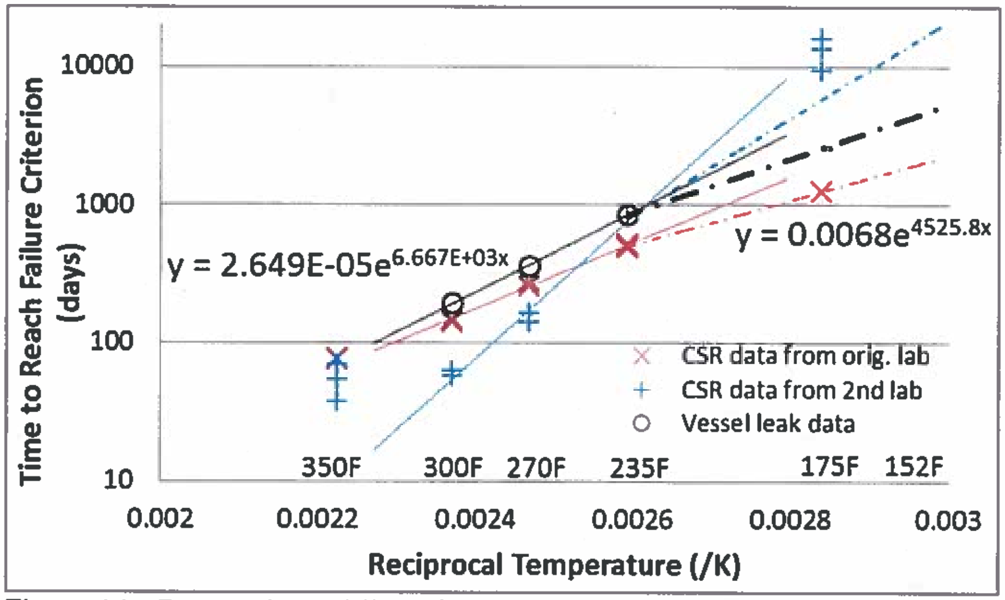

Figure 14. Data and trend lines from vessel leak tests compared to CSR data. The trend line is extrapolated to temperatures below 235 °F (dashed line) assuming a 32% decrease in the activation energy.

CONCLUSIONS

Aging data for EPDM elastomers developed within this task demonstrate that the H1616 O-rings remain functional (leak-tight) for at least three years at 160 °F, and will provide a longer service life at the bounding O-ring service temperature of 152 °F (the maximum temperature at the flange closest to the O-rings with solar heating and 100 °F ambient). In addition, a conservative interpretation of the literature data suggests that the H1616 O-rings can have lifetimes of 6 years at 152 °F. This bounding service temperature is conservative relative to typical O-ring temperatures in service.

Leak testing has demonstrated acceptable leak-tight performance up to the three year mark for the 160 °F vessel, and the continued ability to reseal at that time. The vessel aging at 300 °F maintained leak-tightness and the capability to reseal after 174 days, however it would not reseal after 196 days. Following failure of the O-rings aging in the 300 °F vessel, new O-rings were installed and began aging without an argon backfill. This was intended to demonstrate the extent to which air exposure might influence O-ring degradation. These O-rings remained leak-tight and capable of re-sealing after 193 days. With the limited free space in the O-ring groove and the close fit between the vessel and lid, it is reasonable to assume that the O-rings are sufficiently isolated from a renewed oxygen source to behave similarly to O-rings in an inert atmosphere.

Accordingly, the behavior of the O-rings in service is expected to be much more similar to that observed in an inert environment than in fresh air (as in CSR testing).

CSR test samples aging at 270, 300 and 350 °F in an air environment reached a failure condition after about 40, 22 and 11 weeks, respectively. Significant degradation was also observed at 235 °F, but the failure condition (90% loss in initial sealing force) was not yet reached. Two aging models based on these collective data were developed to predict the O-ring behavior at service conditions. Based on the more conservative of these models, the H1616 O-ring service life at a bounding temperature of 152 °F is conservatively estimated to be approximately 5 years. A definitive correlation between CSR behavior and leak-tight performance has yet to be demonstrated.

Baseline characterization was limited to physical and mechanical properties, namely hardness, thickness, tensile strength and elongation. These properties are consistent with O-ring specifications. The hardness and tensile properties degrade as the material ages, as expected. In addition, this degradation occurs significantly faster in an air environment than when the material is maintained in an inert atmosphere.

The data and analysis developed within this task have produced several estimates of the service life of EPDM O-rings in the H1616 vessel:

- Literature data suggest a conservative service life of approximately 6 years at 152 °F.
- The CSR data (in air) indicate a service life of approximately 5 years at 152 °F.
- Vessel aging tests have demonstrated leak-tight performance for at least 3 years at 160 °F, without reaching a failure condition.
- Extrapolation of vessel aging tests at 235 °F and above provides a service life estimate of 11.4 years at 152 °F.

It is expected that leak-tight performance of the O-rings will be significantly longer at service temperatures (152 °F and below) than at 160 °F. The vessel tests have demonstrated that the model predictions based on CSR data are conservative relative to actual leak performance at 235 and 300 °F, although no correlation has been established to compare these two approaches at lower temperatures. Nevertheless, these collective data demonstrate that the EDPM O-rings in the H1616 vessel are fully capable of providing leak-tight performance with significant margin for a period of three years at service temperatures of 160 °F and below. After three years at temperature, the CSR data show a minimum 61% retained sealing force at 175 °F, indicating a significant degree of margin to leakage failure. An even greater margin would be expected at the peak O-ring service temperatures (152 °F). Therefore, an extension in the maintenance interval for the H1616 package to three years is recommended. The data also provide high confidence that a longer service life, possibly 5 years or longer, would likely be supported by further testing.

If an increase in the maintenance interval beyond three years is desired, the following additional testing is recommended:

- Continue the vessel aging and leak tests at 160 °F (to failure and/or up to 5 years)
- Continue the CSR tests at 175 and 235 °F (to failure and/or up to 5 years)

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