USED FUEL DISPOSITION CAMPAIGN

Preliminary Report - Degradation Evaluation and Aging Management of Bolted Closure Joints

Fuel Cycle Research & Development

Prepared for
U.S. Department of Energy
Used Fuel Disposition Campaign

CG Verst, TE Skidmore, and
RL Sindelar
Savannah River National Laboratory

September 30, 2014

FCRD-UFD-2014-000318
SRNL-STI-2014-00449

EXECUTIVE SUMMARY

The bolted closure joint of a bare spent fuel cask is susceptible to age-related degradation and potential loss of confinement function under long-term storage conditions. The degradation phenomena for the components of the joint, including metallic and polymeric seals, and bolting are summarized, and the aging material activities to quantify and predict the degradation are described. An aging management program to demonstrate maintenance of confinement in consideration of this degradation in long-term storage at normal service conditions is suggested.

Metallic seals provide the primary physical barrier for leak tightness and confinement in storage casks with bolted closures. Testing and evaluation of metallic seals performed at the BAM Federal Institute for Materials Research and Testing has shown that metallic seals are subject to creep or relaxation that will manifest in a loss of the sealing force and seal resiliency. BAM has developed semi-empirical models to predict the loss of sealing force with time for several Helicoflex® metallic seal material systems used in the CASTOR® V/21 and also TN casks.

Polymeric secondary seals are often used to facilitate leak testing of primary metallic seals and protect metallic seals from the external ambient environment (air, water). Polymeric seals in spent fuel casks are susceptible to degradation over time by several mechanisms, principally via thermo-oxidation, stress-relaxation, and radiolytic degradation under time and temperature conditions in air.[1] Testing and evaluation of the ethylene-propylene diene monomer (EPDM) elastomeric seal material used in the CASTOR® V/21 cask for a matrix of temperature and radiation exposure conditions, and development of semi-empirical predictive models for loss of sealing force is in progress at SRNL.

Generic Aging Management Programs for the components of a Dry Cask Storage System have been developed,[2] and the AMP specific for the bolted closure joint is summarized in this report. The principal approach is to directly demonstrate leak tightness of the bolted closure joint through continuous on-line leak monitoring.

The experimentation and modeling being performed using the CASTOR® V/21 elastomeric seal material enable an estimation of the timeframe when loss of sealing function under relevant aging (temperature/radiation) conditions may occur. An update to this report will be issued in FY15 following the completion of elastomeric seal testing at SRNL and additional work on metallic and polymeric material aging testing by BAM.

---

[1] The degradation mechanisms and degradation kinetics of metallic gasket materials (creep and oxidation) and elastomeric materials are not related although their degradation under time/temperature conditions can be empirically described by Arrehnius models.

[2] FCRD-UFD-2013-000294, Managing Aging Effects on Dry Cask Storage Systems for Extended Long-Term Storage and Transportation of Used Fuel, Rev. 1, Argonne National Laboratory

With completion of this work, the gap to provide the technical bases to understand materials degradation and degradation kinetics, and to provide an approach for aging management to ensure the efficacy of the bolted closure joint under long-term storage conditions is effectively closed.

This report fulfills the M3 milestone M3FT-14SR0805063, Degradation Evaluation and Aging Management of Bolted Closure Joints.

1. Introduction

The bolted closure joint in a dry cask storage system provides a confinement function. The components of a bolted closure joint of a bare fuel cask (includes lids, bolting, metallic seals, and polymeric seals) are vulnerable to degradation under stressors of thermal and mechanical loads, and also radiation and corrosion conditions that could lead to a loss of confinement function during extended storage conditions.

The bolted closure joint has been recently re-ranked at a low-to-medium priority in the technical data gaps for extended storage and transportation in the DOE NE program [1]. This reflects that the understanding of the degradation and its management under extended storage and transportation conditions are relatively well-established versus the aging considerations for other components and degradation stressors for dry cask storage systems.

A compilation of the materials potentially used in bolted closure joints is given in Table 1 below.

Table 1 – Typical Materials of Bolted Closure Joints [2]

Metallic Seal Materials	Polymeric Seal Materials	Bolting Materials
Inconel® X750; Al-jacketed Nimonic® 90; Al-jacketed Inconel X750	Ethylene propylene copolymer (EPR), ethylene-propylene diene monomer (EPDM), silicone rubber, polypropylene, Viton® fluoroelastomer, PTFE (polytetrafluoroethylene)	304L SS, ASTM A193 Grade B7; ASTM A320 Grade L43 (AISI 4340); SB637 Grade N07718 (alloy 718); SA564 Type 630 (17-4PH, H1150 condition)

The potential degradation mechanisms of these materials and the impact on the confinement function under long-term storage have been discussed in other reports [3, 4, 5, 6]. This present report summarizes the degradation of these materials phenomenologically, and briefly describes the recent testing and analysis activities to quantify the degradation and degradation kinetics of the seal materials.

Recent testing and analysis activities have been performed for a specific bolted closure joint, that of the CASTOR® V/21 cask. This cask design is expected to be at the upper extreme of temperature and radiation exposure conditions for a loaded bare fuel storage cask.

A generic aging management program for the bolted closure joints has been recently suggested [6], and a summary of that program is provided.

This AMP for the bolted closure joint specifies the continued on-line leak monitoring of the cask that would assure the continued effectiveness of seals and bolting that could degrade under aging conditions. The experimentation and modeling being performed using the CASTOR® V/21 cask material enables, as an example bare fuel cask system, an estimation of the timeframe when loss of sealing function for the bolted closure joint under relevant aging (temperature/radiation) conditions may occur. An update to this report will be issued in FY15 following the completion of elastomeric seal testing at SRNL and additional work on metallic and polymeric material aging testing by BAM.

1.1 Materials and Configuration of the Bolted Closure Joint of the CASTOR® V/21 Cask

The CASTOR® V/21 (CASTOR = cask for storage and transport of radioactive material) is manufactured by GNS (Gesellschaft für Nuklear-Service mbH). The CASTOR® V/21 cask was selected as a representative cask with a bolted closure joint that is expected to be at the upper extreme of temperature and radiation exposure conditions for a bare fuel storage cask.

The general configuration of the bolted closure joint for the CASTOR® V/21 cask is shown in Figure 1 below.

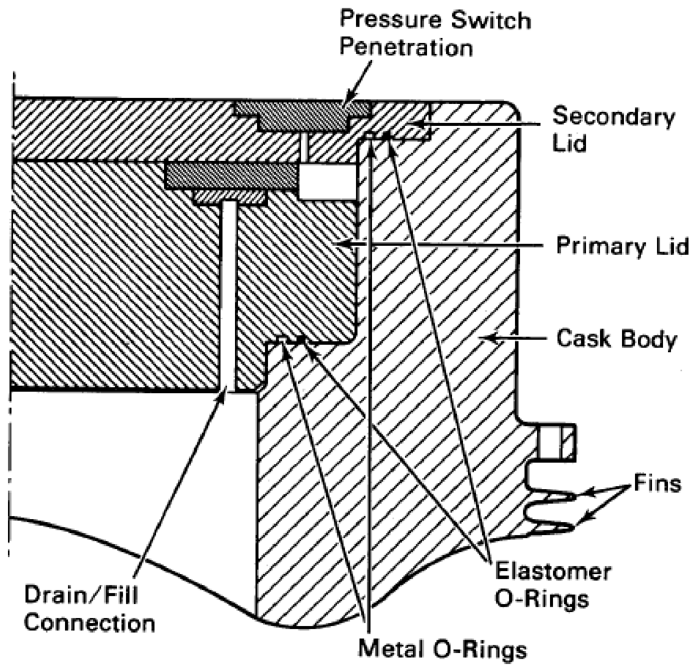


Figure 1 – CASTOR®–V/21 Cask Lid System

In Reference 7, the CASTOR® V/21 cask metallic seals are described as 1600 mm (5.25 ft) outside diameter x 1580.2 mm (5.19 ft) inside diameter, 9.9 mm (0.39 in.) cross section diameter, Helicoflex type HN 200, manufactured with an aluminum outside lining, a 304L or 316L stainless steel inside lining, and a Nimonic® or Inconel® spring. The elastomer seals are described as 1580 mm (5.19 ft) outside diameter x 1560.2 mm (5.12 ft) inside diameter x 9.9 mm (0.39 in.) cross section diameter, EPDM type 50049-2.2 supplied by the Helicoflex Company, Columbia, SC. The availability of this specific compound is discussed later in this report.

Reference 7 describes examination of these seals after 14 years of storage (examined in 1999-2000 at INEEL). The specific seal conditions were not stated in the report, but the 1999 measured temperature of the exterior cask surface ranged from 103 °F (39 °C) to 141 °F (61 °C) at an ambient temperature 91.2 °F (32.9 °C). The hottest of selected fuel assemblies measured 309 °F (154 °C). This compares to 109 °F (43 °C) to 221 °F (105 °C) on the exterior cask surface, and a peak internal cask guide tube temperature of 676 °F (358 °C) measured in 1985 with a nitrogen backfill, with lid in place and cask in the vertical orientation.

Due to size, the O-rings are fabricated with a 45 degree splice joint. The O-rings examined were still resilient with no obvious evidence of significant oxidation or degradation. Bending, pulling, twisting, and coiling the elastomer into a 30 cm (12 in.) diameter coil did not cause fracture or stress failure. Remote camera and visual inspection revealed that the splice joint was slightly misaligned and partially open, suggestive that the splice adhesive did not completely fill the joint. However, the joint retained good strength and could not be pulled apart manually.

The compressed dimensional configuration of the elastomer O-ring after removal suggests an approximate installed percent compression of 13%, assuming no elastic recovery before measurement, metal-to-metal contact (zero gap between mating surfaces) and an initial O-ring thickness of 0.39" (9.9 mm). The actual percent compression is dictated by actual O-ring part thickness, groove dimensions, applicable tolerances, and any gap that may exist between mating surfaces after O-ring installation.

For most static seals, compression of 15-30% is typically recommended, with 25% used in most test standards for comparison unless otherwise specified [8-10]. Too little compression can lead to more extensive or rapid compression set, or may be insufficient to account for vibration or thermal variations, while higher compression values can overstress the elastomer. ID stretch and gland fill are also important factors, with minimal installed stretch values (<5%) preferred to minimize tensile stress on the elastomer during service.

1.2 Thermal and Radiation Conditions at the Bolted Closure Joint of the CASTOR® V/21 cask

Estimation of the heat load and radiation conditions at the joint in the CASTOR® V/21 cask were previously described in Reference 11 and the results are summarized below.

1.2.1 Temperature

The aging temperatures for an elastomer seal[3] for the CASTOR® V/21 cask were based on assumptions drawn from the CASTOR® V/21 opening & examination documented in NUREG/CR-6745 [7]. Thermocouple readings taken from various locations in the cask in 1985 and 1999 suggest that a freshly loaded cask would experience temperatures between 200 to 250 ˚F at the location of the primary lid elastomer seal. These values represent the initial inputs for the proposed test matrix thermal conditions. The secondary lid seals will be subjected to lower temperatures.

A more rigorous evaluation of the O-ring time-temperature profile can be performed. The thermal power of a fully loaded CASTOR® V/21 basket was estimated using SCALE6 to generate actinide and fission product inventory of 21 PWR assemblies burned to 23 GWD/MTU.

---

[3] Ethylene-propylene diene monomer (EPDM) elastomers are typically rated at a limit of 250°F for "continuous" service, although some formulations may be rated for higher temperatures. Also, the ratings are typically based on relatively short-term data, not years of aging performance.

This heat source term is being used to develop thermal models which can accurately predict temperature at specified locations as a function of time. These heat decay data are represented in Table 2.

Table 2 - Thermal power of 21 PWR assemblies with 23 GWD/MTU burn-up

Basket Heat								
Time Since Discharge	0 [years]			5 Yr	10 Yr		50 yr	100 yr
Cooling Time [Days]	1 [days]	100	1000	1825	3650	10000	18250	36500
Fission Products [Watts]	1.24E+06	1.69E+04	2.80E+03	1.47E+03	9.28E+02	5.61E+02	3.24E+02	9.81E+01
Actinides [Watts]	7.10E+04	1.43E+03	2.69E+02	2.49E+02	2.57E+02	2.64E+02	2.54E+02	2.20E+02
Total per MTU [Watts]	1.31E+06	1.83E+04	3.07E+03	1.72E+03	1.18E+03	8.26E+02	5.78E+02	3.18E+02
Total per Cask [Watts]	1.28E+07	1.79E+05	3.01E+04	1.69E+04	1.16E+04	8.10E+03	5.67E+03	3.12E+03
Total per m^3 [Watts]	3.03E+06	4.24E+04	7.10E+03	3.99E+03	2.74E+03	1.91E+03	1.34E+03	7.37E+02

1.2.2 Radiation

The dose rate with time at the seal location for a CASTOR® V/21 storage cask loaded with commercial PWR assemblies at the design conditions was evaluated. A fully loaded CASTOR® V/21 cask was modelled by the Monte Carlo transport code, MCNP. The model was simulated to store its maximum allowable fuel loading as per the CASTOR® V/21 license requirements. This consists of 21 W17x17 assemblies burned to 23 GWD/MTU during three 550 day cycles allowing 30 days of cooling in between cycles. Additionally, the spent fuel was allowed to cool 5 years before being loaded into the cask.

Dose detector tallies were used in the MCNP model to determine gamma dose rates at the elastomer O-ring location in the primary lid. The dose rate was calculated at 5, 10 15, 30, 50, and 100 years after fuel discharge, and integrated over time to yield the total accumulated dose between 5 and 100 years after discharge. The absorbed dose rate profile estimated for the seals in a CASTOR® V/21 cask loaded as described is shown in Figure 2.

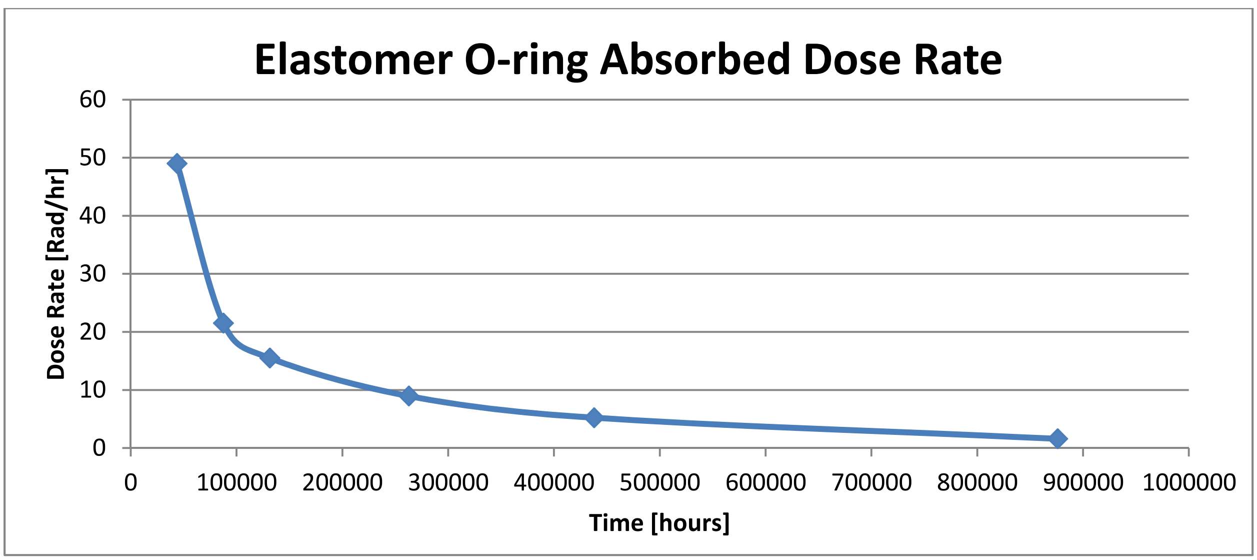


Figure 2 - Dose Rate at Elastomeric Seal Location for CASTOR® V/21 Cask

The absorbed radiation dose profile for the elastomeric seal in the CASTOR® V/21 cask primary lid during the first 95 years of storage is a cumulative dose of ~ 5.9 Mrad (5.9E+04 Gy or 59 kGy). The dose rate drops off severalfold in the first 10 years and more gradually thereafter. At 95 years, the dose rate is approximately 1.6 rad/hr

2. Summary of Aging Degradation Phenomena of Components of Bolted Closure Joint of a Storage Cask

Loss of confinement could occur if either the sealing capability of the metallic seal was compromised due to viscous flow or creep, or the bolt closure stresses were reduced by processes such as stress corrosion cracking of the bolting.

Polymeric/elastomeric seals are used in some spent fuel cask designs to facilitate leak testing of the primary seal and to provide a secondary confinement barrier and weather seal. These materials are subject to several degradation mechanisms including thermo-oxidation, stress-relaxation and radiolytic degradation. These mechanisms and their relative potential to occur are summarized.

2.1 Metallic Seal Degradation

Metallic seals are an essential component in the confinement of a bolted closure, and are potentially subject to degradation in service. One scenario of failure would be if a seal relaxed to the point that insufficient sealing forces were retained on the closed joint, causing the seal to fail required leak tests or to provide the confinement function. Internationally, long-term full-scale tests with monitoring of leak resistance of metallic seals have been performed in France (CEA Atomic Energy Commission), Japan (Central Research Institute of Electric Power Industry), and Germany (BAM Federal Institute for Materials Research and Testing).

Testing and analysis to quantify the loss of sealing force of four types of metallic Helicoflex® seals under aggressive test conditions and correlate the sealing force loss to leakage have been conducted at BAM [12]. Helicoflex® seals are constructed of a helical spring, surrounded by an inner jacket (of stainless steel and an outer jacket of a malleable material, such as aluminum or silver. These seals are capable of sealing to leakage rates of less than 10^{-7} atm cc s^{-1}.

Loss of sealing force of metallic seals is expected to occur over time because of plastic deformation of the outer jacket of the seal. Creep/relaxation behavior of aluminum and silver metallic seal linings were studied [12]. Tests longer than 10,000 hr were implemented only for silver. For each lining, two different section diameters were investigated. A primary goal of the work was to determine the residual linear load that can be guaranteed for a seal after a particular time of relaxation at specific temperatures.

To test this process, seals were tightened between two flanges and exposed to constant temperatures, including 100 and 200 °C. Residual load and 'useful' recovery was measured after the exposure. Results were interpreted according to two methods: a time extrapolation, and a time–temperature equivalence parameter. Both methods are based on linear relationships and were assessed through statistical analysis (calculation of scatter) which is also used to determine a minimum guaranteed residual closure load.

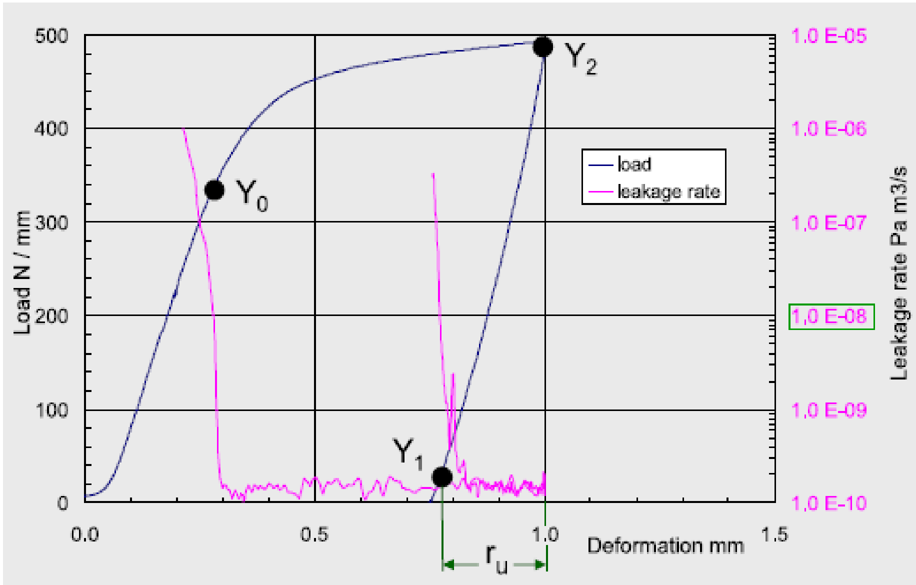
For silver-lined seals, it was concluded that use of a time– temperature equivalence parameter equal to T (11 + log10 (t)) is justified [13]. This relationship allows one to assess the maximum temperature at which seals can be used. This relationship assumes that a residual linear load of at least 100 N/mm will be retained to maintain a seal after closing the cask. Though a reasonable assumption, this may not be true for all metallic seal materials and cask designs.

In addition to the potential problems with stress relaxation, the crevices formed in the seal joint are subject to possible crevice corrosion conditions including potential galvanic effects, if moisture can migrate to and condense in the crevice of the joint. The kinetics of crevice corrosion are difficult to predict reliably but crevice corrosion is supported by the same environmental conditions (water film and aggressive species) that support SCC. The likelihood of crevice corrosion occurring within a given cask depends on the materials, the crevice geometry, and environment. Similar to SCC, only at temperatures conditions allowing a water film to exist at the joint would crevice corrosion occur.

The BAM program includes testing with water (borated pool water at 2400 ppm B; 10^{-3} M NaCl solution) in the gap between the inner and outer seal jacket. No corrosion-induced failure has been observed since testing began in 2001.

Figure 3 shows the comparison of sealing force with leakage for the BAM testing [12]. Figure 4 shows the test results and an extrapolation of the 150 °C data, indicating that sealing force will likely continue to decline, but a significant force will be retained after 40 years, approximately 22% (aluminum) or 33% (silver). Further extrapolation of these data indicate that at least 10,000 weeks (192 years) is required to reach the 100 N/mm value needed for leak tightness assumed for the silver jacket seal.

Linear extrapolation goes well beyond the 10,000 week point, and is closer to 20,000-30,000 weeks (384-576 years). Linear extrapolation over this range is tenuous but significant lifetimes appear possible, assuming no significant changes in the relaxation mechanism or rate are observed and crevice corrosion or other service-limiting mechanisms do not occur.



$Q_{He/St}$ = $\leq 10^{-8}$ Pa·m³·s⁻¹

- Y_0 = Achievement of $Q_{He/St}$ during pressing process
- Y_1 = Exceeding $Q_{He/St}$ during load relieving
- Y_2 = Optimal operation point according to manufacturers specification
- r_u = usable resilience

Figure 3 – Characteristic load-deflection curve of a Helicoflex® silver metal seal and correlation with helium leakage rate for the BAM test setup [reproduced from 12]

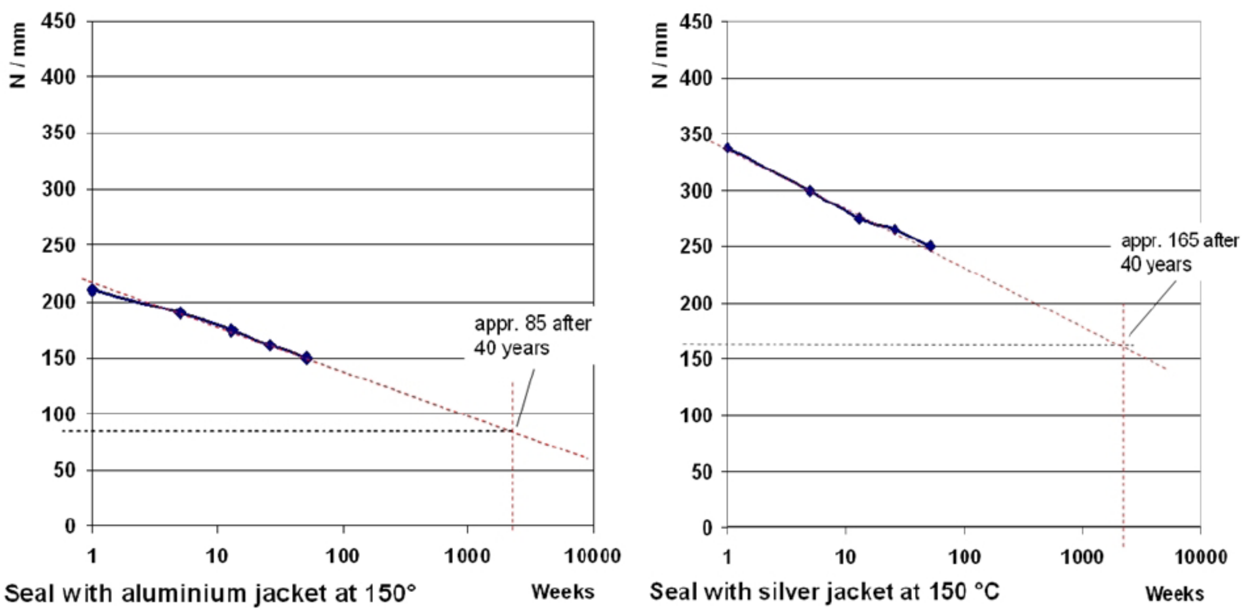


Figure 4 – Extrapolation of seal force decrease over 40 years under constant conditions [reproduced from 12]

2.2 Elastomeric Seal Degradation

Elastomer seals often provide secondary seals in bolted closure joints in dry fuel storage cask designs. They are primarily used to facilitate leak testing of primary metallic seals and to provide a secondary barrier from the exterior environment.

Polymeric seals in dry fuel storage casks are susceptible to degradation by several mechanisms, including thermo-oxidation, stress-relaxation and radiolytic breakdown. Several degradation mechanisms may be on-going simultaneously within a given cask environment, with the dominant mechanism likely to vary with the specific service conditions relative to a particular cask design and seal location. At low dose rates and higher temperatures relative to material resistance, thermo-oxidation and antioxidant depletion are likely to dominate seal degradation. At higher dose rates, or after longer cumulative storage periods, radiation damage may be more dominant. The dominant manifestations of these degradation mechanisms are stress-relaxation and/or possible embrittlement. Stress-relaxation is driven by both mechanical and chemical processes.

Several elastomer types may be used as secondary seals in spent fuel cask designs, depending on service conditions. Types that are typically used include ethylene-propylene diene monomer (EPDM), silicone, FKM fluoroelastomer (Viton® or similar) and butyl rubber. Each generic elastomer type has inherent advantages and limitations, with specific compounds often tailored for specific applications. The primary advantage of fluoroelastomers and silicone compounds is resistance to heat, thermo-oxidation (antioxidants not typically needed) and resistance to ozone/ultraviolet degradation. Silicone elastomers can sometimes have less desirable mechanical properties and higher permeability. Butyl rubber generally offers excellent (low) gas permeability, but is also less resistant than other types to heat and/or ionizing radiation.

Thermo-oxidation of polymers generally follows the general Arrhenius-type relationship for reaction rate as a function of temperature. However, the presence of antioxidants and other formulation additives can significantly complicate the aging/degradation process. Arrhenius-type models and predictions based thereon can be overly optimistic if antioxidant depletion and "inhibition" periods are not accounted for.

An example of this behavior is shown in Figures 5 and 6.

Figure 5 shows an example of sealing force decay of an EPDM compound over time at different temperatures, with the data for both temperatures superposed using time-temperature superposition principles. The advantage of this method is that all experimental data at various aging temperatures can be superposed into a "master" curve using shift factors that can be translated to other service temperatures of interest. A limitation of this approach is that the degradation mechanism (and thereby activation energy) is assumed not to change within the temperature range of interest.

Figure 6 shows that the activation energy of oxygen uptake or oxygen consumption is lower at lower aging temperatures than the activation energy observed for compression stress-relaxation (CSR) behavior at higher aging temperatures. This results in lower lifetime values than would be predicted by CSR data alone. In this particular case, the CSR aging model alone would predict a lifetime of many thousands of years at room temperature, while the data from oxygen consumption would predict a lower but still very acceptable lifetime of at least 150 years. These same data can be translated to other temperatures using the appropriate shift factors. In the case discussed, seal lifetime was defined as the time to lose 90% of initial sealing force, or to reach a 10% retained sealing force value. The relationship between this parameter and leakage has not been universally established, but it is likely reasonable and conservative for many static seal designs.

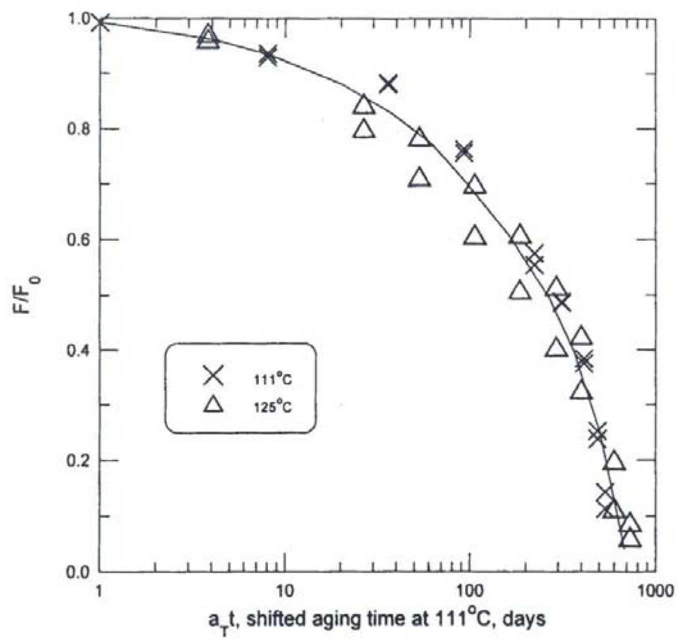


Figure 5 - Time-Temperature Superposition of CSR data for EPDM seals [reproduced from reference 14]

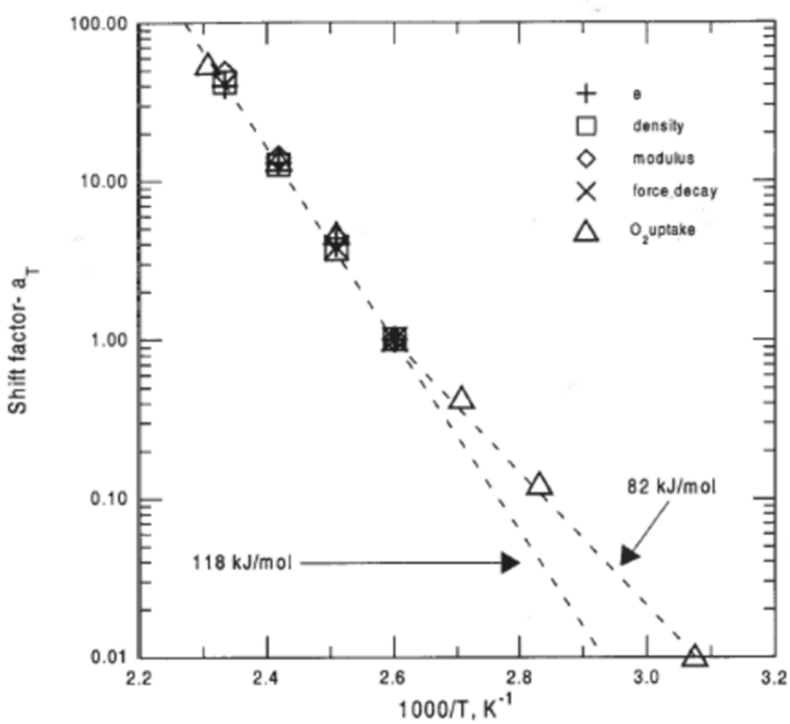


Figure 6 - Non-Arrhenius aging behavior of EPDM O-rings indicated by oxygen uptake data [reproduced from reference 14]

2.3 Bolt Degradation

In most nuclear service, the bolting is a replaceable component that can be replaced. A compendium of bolted closure maintenance for is provided in reference 15.

The gap reports and aging review of the components of the DCSS [3-5] have concluded that the cask bolting is subject to corrosion and potential loss of joint loading function. The US operating experience on bolted closure joints, summarized in reference 6 captures the reported observation of bolt corrosion (and metallic seal corrosion) at the Surry Independent Spent Fuel Installation.

Thus, with time, bolt failure may be anticipated to occur, and if sequential (Figure 6), a loss of sealing force for gasket compression would be expected. However, a failure of one or several bolts may not cause a sealing (confinement) failure unless the failed bolts are adjacent to each other in the bolt pattern, and it is highly unlikely that this would occur in cask storage [6]. It is beyond the scope of this report to evaluate the leakage expected from a pattern of failed bolts.

Periodic monitoring for bolt torque and corrosion are typically part of a maintenance program for bolting in NPP [15]. The on-line leakage monitoring program and the visual observation for corrosion at the closure region as suggested in reference 6, and summarized below in section 3, would provide a sufficient indicator of incipient bolt failure and loss of sealing for aging management under extended storage conditions.

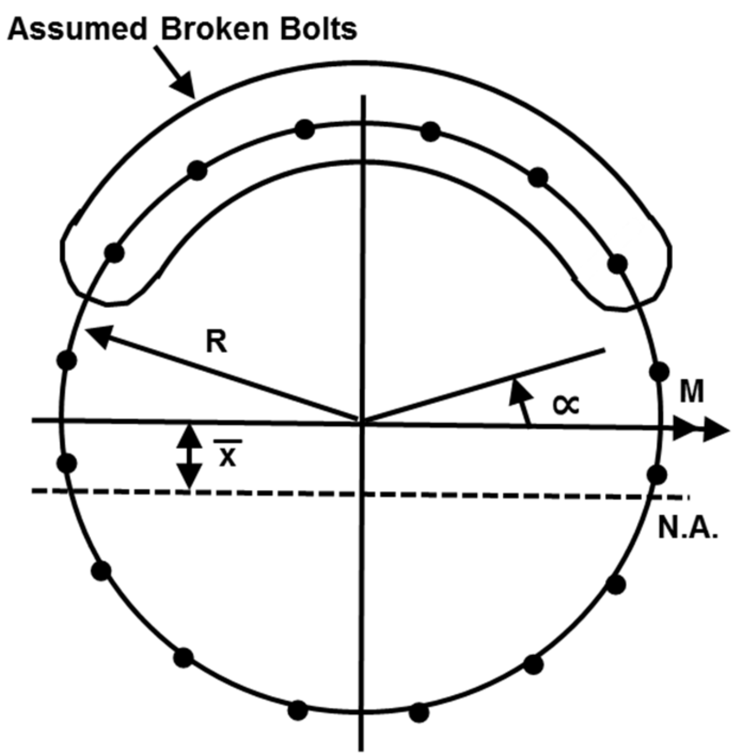


Figure 6 – Generic condition for loss of sealing force from multiple adjacent bolt failure

3. Aging Management Program for the Bolted Closure Joint

A bare fuel cask with a non-weld-seal is required to provide for continuous monitoring for leakage as part of its license. Continuation of this monitoring is the approach recommended for an AMP under extended storage (re-licensing).

3.1 Requirements for Continuous Leakage Monitoring

Federal requirements in 10CFR72 and the further stipulations in the Standard Review Plan govern the standing monitoring for leakage of the bare fuel cask bolted closure joint.

10 CFR72.122 states: "Storage confinement systems must have the capability for continuous monitoring in a manner such that the licensee will be able to determine when corrective action needs to be taken to maintain safe storage conditions. For dry spent fuel storage, periodic monitoring is sufficient provided that periodic monitoring is consistent with the dry spent fuel storage cask design requirements. The monitoring period must be based upon the spent fuel storage cask design requirements."

10 CFR 72.236(e) states: "The spent fuel storage cask must be designed to provide redundant sealing of confinement systems." For bolted cask designs, this usually consists of two concentric metallic seals between the cask lid and body. For welded canisters, two weld joints are required with at least one of them subjected to a helium leak check after fabrication. In bolted casks, the space between the seals may be provided with a seal monitoring system to detect failure of either seal.

According to standard review plan NUREG-1536, rev. 1: "Typically, this means that field closures of the confinement boundary should either have two seal welds or two metallic O-ring seals. The NRC staff has found that casks closed entirely by welding do not require seal monitoring. However, for casks with bolted closures, the staff has found that a seal monitoring system is required to adequately demonstrate that seals can function to limit releases and maintain a helium atmosphere in the cask for the term of the 10 CFR Part 72 general license. A seal monitoring system, combined with periodic surveillance, enables the licensee to determine when to take corrective action to maintain safe storage conditions."

According to NUREG-1567: "Although the details of the monitoring system may vary, the general design approach has been to pressurize the region between the redundant seals, with a non-reactive gas, to a pressure greater than that of the cask cavity and the atmosphere. The monitoring system is leakage tested to the same leak rate as the confinement boundary. A decrease in pressure between these seals indicates that the non-reactive gas is leaking either into the cask cavity or out to the atmosphere."

3.2 AMP Overview

This proposed program manages aging of closure bolting for the confinement function of the cask and the program is adopted from reference 6. Reference 6 followed an aging management review and developed aging management programs for the components of the

Dry Cask Storage System, similar to the Generic Aging Lessons Learned program for generic AMPs for NPP.

The monitoring of the non-reactive gas leakage at the bolted closure joint, including the acceptance criteria, required in the existing program for cask storage, is recommended to be continued in the cask bolted joint AMP for extended storage throughout the licensing period.

In addition visual monitoring of the closure seal components for evidence of corrosion is also recommended. Specifically, the AMP from reference 6 consists of the following activities:

The Bolted Cask Seal and Leakage Monitoring Program is an aging management program (AMP) that consists of the following activities [6]:

> (a) An overpressure leakage monitoring system for continuous monitoring of the pressure between the two seal assemblies in some cask designs or inside the cask cavity for other cask designs. In the TN, NAC-I28, and CASTOR metal casks, an overpressure leakage monitoring system provides continuous monitoring of pressure in the region between the redundant metallic seal assemblies, which is pressurized with a non-reactive gas to a pressure greater than the helium pressure in the cask cavity. Therefore, for these casks, the licensee does not have to specify the maximum allowed leakage rate because leakage of radioactive contents through the seals is not a credible event. An overpressure leakage monitoring system in the MC-10 casks provides continuous monitoring of pressure inside the cask cavity, and a decrease in pressure indicates a leakage from the cask cavity through both of the O-ring lid seals or through the vent lid welds. For this cask design, the applicant should specify the maximum allowed leakage rate, as recommended by NRC ISG-5, Rev. 1. Furthermore, as discussed above, the breach of the cask confinement boundary from causes other than degradation of the inner metallic seal or the vent lid seals is not considered credible.
>
> (b) Periodic visual inspection of the closure seal components and maintenance of the overpressure leakage monitoring system and associated instrumentation.

It is further recommended that bolts removed from a cask be subjected to VT/UT inspection, and that seals be examined for corrosion (metallic seals) and cracking/crazing (elastomeric seals) and that the results be recorded.

4. CASTOR®/V21 Elastomeric Seal Irradiation and Test Program

Testing and analysis is in progress to evaluate time/temperature/dose exposure conditions at which a polymeric seal may lose its ability to provide a seal function. The testing is being performed for a specific elastomer seal compound, Parker Seals EPDM E740-75, as used in other cask designs.

This compound is currently being used as a surrogate for the specific EPDM compound in the CASTOR® V/21 cask as the V/21 cask compound has proved elusive. Variations in the EPDM compounds are possible, though significant variations in aging behavior are not expected.

Reference 11 provides a full description of the irradiation and test program including:

- EPDM compound
- Special Compression Stress Relaxation test assembly for irradiation testing
- Irradiation and test matrix
- Initial results

Additional testing using this approach is in progress to develop a data set with a mechanistically-based empirical model. The objective is to enable prediction for time/temperature/dose conditions for this particular EPDM compound.

An update to the irradiation and CSR results is provided in Section 5.

5. Testing Matrix and Results

The experimental test matrix was designed to evaluate seal responses to a variety of dose and temperature combinations which range from conservative representations of realistic conditions within a cask to well-beyond to identify certain failure conditions.

The conditions for each sample set may include irradiation at a prescribed dose rate, exposure to high temperatures or a combination of both. For each temperature considered, several accompanying dose rates will be applied simultaneously. The variations in these combinations should provide insight into the quantitative damaging effect of both radiation and elevated temperatures on the EPDM O-ring compound studied.

Additionally, the degradation due to radiation alone and its dependence on dose rate will be tested at ambient temperature. Similarly, several samples will be exposed to high temperature in air without radiation exposure. The proposed Test Matrix is in Table 3**.** These conditions may be revised as new data are obtained.

Table 3 - Test Matrix for EPDM O-ring Aging Exposure Conditions (Radiation/Temperature)

Set Name : Description	Radiation Dose	Temperature
A : Absolute Failure , Extremely High Temperature Only	None	350°F
B : Absolute Failure , Extremely High Dose Rate, High Temperature	100 Mrad - 240 kRad/hr	240°F
C : Absolute Failure , High Dose Rate, High Temperature	100 Mrad - 100 kRad/hr	240°F
D : Target Dose, High Temp, Low Dose Rate	6 MRad – 10 kRad/hr	240 °F
E : Target Temp , High Temperature Only	None	240°F
F : Absolute Failure , Extremely High Dose Rate, Med Temp	100 Mrad - 240 kRad/hr	200°F
G : Absolute Failure , High Dose Rate, Med Temp	100 Mrad - 100 kRad/hr	200°F
H : Target Dose, Med Temp, Low Dose Rate	6 MRad – 10 kRad/hr	200 °F
I : Target Temp , Med Temperature Only	None	200°F
J : Absolute Failure , Extremely High Dose Rate, Low Temperature	100 Mrad - 240 kRad/hr	100°F
K : Absolute Failure , High Dose Rate, Low Temperature	100 Mrad - 100 kRad/hr	100°F
L : Target Dose, Low Dose Rate, Low Temperature	6 MRad – 10 kRad/hr	100 °F
M : Target Dose, Extremely Low Dose Rate, Low Temperature	6 MRad – 1 kRad/hr	100 °F

All samples undergo testing at their specified conditions for a period of 25 consecutive hours. It is desirable to minimize the frequency heating/cooling cycles experienced by the samples as this more accurately reflects the intransient thermal conditions within a cask seal. However, multiple data points must be collected to draw a reasonable trend of the data. The 25-hour duration provides a practical balance between the drawbacks of rapid heating and cooling and the need for resolution on the sealing force decay plots. A 25 hour cycle also deposits exactly the 95 year cumulative absorbed dose value of 6 MRad to all samples undergoing the “Extremely High Dose Rate” condition.

At the conclusion of a 25 hour testing cycle, samples are removed from their specified environment and allowed to cool to room temperature before being loaded into the CSR jig and evaluated for counterforce measurements. The time allowed for cooling is typically 1 to 2 hours, although samples generally reach ambient conditions within 30 minutes. It would be beneficial to determine if extended cooling time or aggressive cooling rates affect the samples’ counterforce measurements.

Several consecutive CSR measurements are recorded for each sample, averaged, and adjusted for the inherent break force of the jig used for the measurements. Counterforces are measured in units of Newtons and denote the force required to further compress an O-ring sample. Although it is difficult to directly relate sealing force to a leak rate analytically, a plot of the sealing force decay does reveal the relationship between damaging factors and loss of seal effectiveness.

As of September 2014, eight of the sample sets from the proposed test matrix (Table 3) have begun testing and will continue to provide sealing force data until their target conditions are met or they reach ultimate failure (90% loss of sealing force). These eights sets were initiated first due to their expected rapid decay in the presences of harsh testing conditions. The remaining sample sets will begin testing concurrently as equipment and irradiator space becomes available. Select results of Sets B, C, E, F, G, and I are tabulated in Table 4 below.

Table 4 - Compression Stress Relaxation Measurements for Test Sets to Date

Set : I **Sample: 10** Position / Dose Rate: None
Temperature: 200 ˚F / 93 ˚C Anticipated Total Dose: None

Measurement Number	Date	Unadjusted Counterforce Measurements			Average	Breakforce	Adjusted Counterforce	Time at Temperature
		1st	2nd	3rd				
Baseline	8/7/2014	594	589	593	592	4	588	N/A
1	8/11/2014	527	529	533	530	4	526	25 Hrs
2	8/14/2014	403	412	413	409	5	404	50 Hrs
3	9/3/2014	481	500	491	491	4	487	75 Hrs
4	9/4/2014	472	467	463	467	5	462	100 Hrs
5	9/18/2014	450	451	462	454	5	449	125 Hrs

Set : I **Sample: 11** Position / Dose Rate: None
Temperature: 200 ˚F / 93 ˚C Anticipated Total Dose: None

Measurement Number	Date	Unadjusted Counterforce Measurements			Average	Breakforce	Adjusted Counterforce	Time at Temperature
		1st	2nd	3rd				
Baseline	8/7/2014	609	603	606	606	4	602	N/A
1	8/11/2014	550	542	546	546	4	542	25 Hrs
2	8/14/2014	518	513	511	514	5	509	50 Hrs
3	9/3/2014	507	505	497	503	4	499	75 Hrs
4	9/4/2014	490	490	486	489	5	484	100 Hrs
5	9/18/2014	472	460	464	465	5	460	125 Hrs

Set : E **Sample: 12** Position / Dose Rate: None
Temperature: 240 ˚F / 115 ˚C Anticipated Total Dose: None

Measurement Number	Date	Unadjusted Counterforce Measurements			Average	Breakforce	Adjusted Counteforce	Time at Temperature
		1st	2nd	3rd				
Baseline	8/7/2014	632	630	636	633	4	629	N/A
1	8/11/2014	561	561	565	562	4	558	25 Hrs
2	8/14/2014	524	536	534	531	5	526	50 Hrs
3	9/3/2014	513	505	523	514	4	510	75 Hrs
4	9/4/2014	494	490	488	491	5	486	100 Hrs
5	9/18/2014	457	463	468	463	5	458	125 Hrs

Set : E **Sample: 13** Position / Dose Rate: None
Temperature: 240 ˚F / 115 ˚C Anticipated Total Dose: None

Measurement Number	Date	Unadjusted Counterforce Measurements			Average	Breakforce	Adjusted Counterforce	Time at Temperature
		1st	2nd	3rd				
Baseline	8/7/2014	580	583	570	578	4	574	N/A
1	8/11/2014	503	506	507	505	4	501	25 Hrs
2	8/14/2014	469	458	488	472	5	467	50 Hrs
3	9/3/2014	441	465	442	449	4	445	75 Hrs
4	9/4/2014	424	423	417	421	5	416	100 Hrs
5	9/18/2014	400	399	394	398	5	393	125 Hrs

Set : G		Sample: 15			Position / Dose Rate:		100 krad/hr	
Temperature:		200 ˚F / 93 ˚C			Anticipated Total Dose:		100 Mrad	
Measurement Number	Date	Unadjusted Counterforce Measurements			Average	Breakforce	Adjusted Counterforce	Cumulative Dose
		1st	2nd	3rd				
Baseline	9/4/2014	535	545	555	545	5	540	0
1	9/18/2014	474	475	476	475	4	471	6 Mrad
2	9/23/2014	466	465	467	466	4	462	12 Mrad
3	9/24/2014	451	452	451	451	4	447	18 Mrad
4	9/28/2014	431	431	430	431	5	426	24 Mrad
5	9/30/2014	427	416	418	420	5	415	30 MRad

Set : F		Sample: 16			Position / Dose Rate:		240 krad/hr	
Temperature:		200 ˚F / 93 ˚C			Anticipated Total Dose:		100 Mrad	
Measurement Number	Date	Unadjusted Counterforce Measurements			Average	Breakforce	Adjusted Counterforce	Cumulative Dose
		1st	2nd	3rd				
Baseline	9/4/2014	496	497	497	497	5	492	0
1	9/18/2014	418	420	418	419	4	415	2.5 Mrad
2	9/23/2014	392	393	392	392	4	388	5 Mrad
3	9/24/2014	345	350	352	349	4	345	7.5 Mrad
4	9/28/2014	320	331	326	326	5	321	10 Mrad
5	9/30/2014	322	325	321	323	5	318	12.5 Mrad

Set : B		Sample: 17			Position / Dose Rate:		240 krad/hr	
Temperature:		240 ˚F / 115 ˚C			Anticipated Total Dose:		100 Mrad	
Measurement Number	Date	Unadjusted Counterforce Measurements			Average	Breakforce	Adjusted Counterforce	Cumulative Dose
		1st	2nd	3rd				
Baseline	9/4/2014	502	508	497	502	5	497	0
1	9/18/2014	401	404	397	401	4	397	6 Mrad
2	9/23/2014	365	370	363	366	4	362	12 Mrad
3	9/24/2014	333	328	334	332	4	328	18 Mrad
4	9/28/2014	303	301	306	303	5	298	24 Mrad
5	9/30/2014	272	278	280	277	5	272	30 Mrad

Set : C		Sample: 18			Position / Dose Rate:		100 krad/hr	
Temperature:		240 ˚F / 115 ˚C			Anticipated Total Dose:		100 Mrad	
Measurement Number	Date	Unadjusted Counterforce Measurements			Average	Breakforce	Adjusted Counterforce	Cumulative Dose
		1st	2nd	3rd				
Baseline	9/4/2014	502	508	520	510	5	505	0
1	9/18/2014	455	459	459	458	4	454	2.5 Mrad
2	9/23/2014	403	404	406	404	4	400	5 Mrad
3	9/24/2014	380	372	375	376	4	372	7.5 Mrad
4	9/28/2014	363	364	362	363	5	358	10 Mrad
5	9/30/2014	342	350	345	346	5	341	12.5 Mrad

These results verify the expected trend of sealing force decay and reveal the falloff rate which can be correlated with exposure time to produce predictive models that extrapolate data out to the service lifetime of seal components. These data must of course be corrected for non-Arrhenius behavior factors before resulting in absolute sealing force values (i.e. 1N/cm) for a given exposure condition and service time, but when the current results are normalized to initial CSR measurements and plotted versus exposure time, the relative effects of elevated temperature and gamma radiation exposure become clear. The sealing force decay plots are shown in Figures 7 and 8.

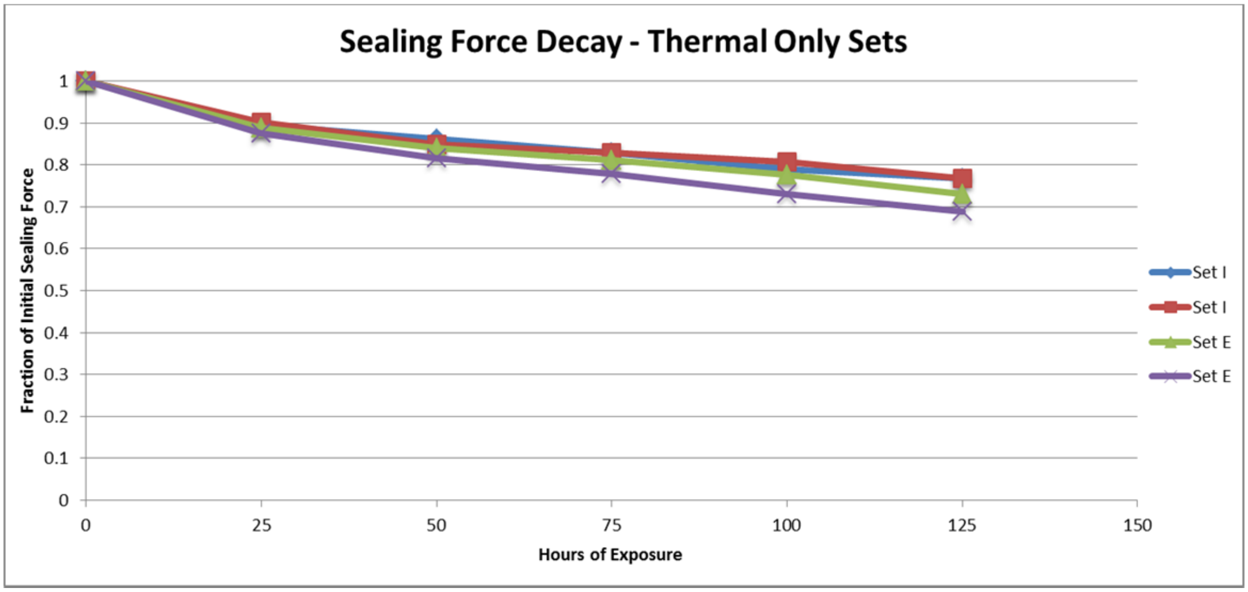


Figure 7 – Sealing force decay of sample sets E and I (thermal load only)

Figure 7 shows a slow nearly linear, decay in the sealing force of samples exposed to high temperatures of 200 °F (Set I) and 240 °F (Set E). The divergence in the fraction of sealing force remaining between Sets E and I illustrates the influence that 40 °F may have in the functionality of the seal. It is important to note, however, that divergence also exists between two samples of Set E which were both exposed to identical conditions simultaneously, perhaps owing to intrinsic variations in O-ring segments and sample loading (i.e. asymmetric loading, edge effects). It is expected that both samples within Set E will continue a sharper decline in sealing force than Set I as exposure increases.

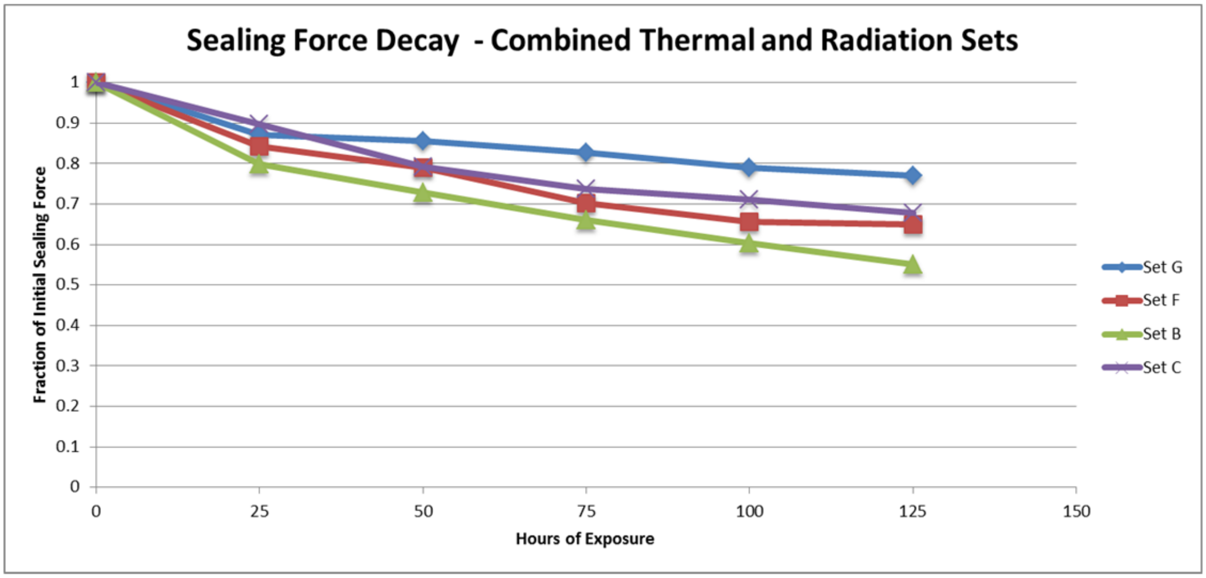


Figure 8 – Sealing force decay of sample sets G, F, B, and C exposed to elevated temperature and high gamma flux

Figure 8 shows sealing force decay plots for four different sample sets, each consisting of a combination of thermal and radiation conditions. For convenience, the test matrix for these four sample sets is reproduced here in Table 5.

Table 5 – Test conditions for sample sets B, C, F, and G

Set Name : Description	Radiation Conditions	Thermal Conditions
B : Absolute Failure , Extremely High Dose Rate, High Temperature	100 Mrad - 240 kRad/hr	240°F
C : Absolute Failure , High Dose Rate, High Temperature	100 Mrad - 100 kRad/hr	240°F
F : Absolute Failure , Extremely High Dose Rate, Med Temp	100 Mrad - 240 kRad/hr	200°F
G : Absolute Failure , High Dose Rate, Med Temp	100 Mrad - 100 kRad/hr	200°F

The results suggest that the dose accumulated by Sample Sets C and G following 125 hours of exposure (12.5 MRad) have a slightly greater sealing force loss as compared to Sample Sets E & I which experienced the same temperature without any gamma flux. However, the dose rate of 240 kRad/hr imparted to Sample Sets B and F did appear to have a significant effect on the sealing force over the range of data collected to this date. It is unknown at this point if the apparent radiation damage to seal is influenced heavily by dose rate, but this will be revealed as the low dose rate samples (10 kRad/hr & 100 kRad/hr) approach the same target absorbed dose (100 Mrad) as the high dose rate samples (240 kRad/hr).

6. Path Forward

Testing will be continued in FY15 to develop a nomograph to predict loss of sealing force of the EPDM seal material as a function of time/temperature/dose and dose rate for ambient air conditions. This will establish an approach with the example of a specific elastomeric material system to enable the timeframe at which an elastomeric seal may lose its sealing capability.

A final report will be issued in FY15 with latest test information from BAM. An assessment will be made for other elastomeric seals with regards to expected performance relative to EPDM.

7. References

1. FCRD-UFD-2014-000050, "Used Nuclear Fuel Extended Storage and Transportation Research and Development Review and Plan," August 9, 2014.
2. ASTM C-1562-10, Standard Guide for Evaluation of Materials used in Extended Service of Interim Spent Nuclear Fuel Dry
3. USED FUEL DISPOSITION CAMPAIGN, Gap Analysis to Support Extended Storage of Used Nuclear Fuel, Rev. 0, Brady Hanson (PNNL), Halim Alsaed (INL), Christine Stockman (SNL), David Enos (SNL), Ryan Meyer (PNNL), Ken Sorenson (SNL), January 31, 2012, FCRD-USED-2011-000136, Rev. 0, PNNL-20509.
4. Draft Report on Indentification and Prioritization of the Technical Information Needs Affecting Potential Regulation of Extended Storage and Transportation of Spent Nuclear Fuel, U.S. NRC, May 2, 2012 (ML12122A922).
5. NUREG/CR-7116, SRNL-STI-2011-00005, Materials Aging Issues and Aging Management for Extended Storage and Transportation of Spent Nuclear Fuel, U.S. Nuclear Regulatory Commission – Office of Nuclear Materials Safety and Safeguards, November 2011.
6. USED FUEL DISPOSITION CAMPAIGN, Managing Aging Effects on Dry Cask Storage Systems for Extended Long-Term Storage and Transportation of Used Fuel, Chopra OK, D Diercks, D Ma, VN Shah, S-W Tam, RR Fabian, Z Han, and YY Liu, 2013. FCRD-UFD-2013-000294, Rev.1, ANL-13/15. Prepared for the U.S. Department of Energy Used Fuel Disposition Campaign, Washington, D.C.
7. NUREG/CR-6745, INEEL/EXT-01-00183, Dry Cask Storage Characterization Project-Phase 1: CASTOR® V/21 Cask Opening and Examination, September 2001.
8. Parker O-Ring Handbook, ORD 5700.
9. ASTM D395 - 03(2008), Standard Test Methods for Rubber Property—Compression Set.
10. ASTM D6147 - 97(2008), Standard Test Method for Vulcanized Rubber and Thermoplastic Elastomer—Determination of Force Decay (Stress Relaxation) in Compression.
11. USED FUEL DISPOSITION CAMPAIGN, Preliminary Report: Effects of Irradiation and Thermal Exposure on Elastomeric Seals for Cask Transportation and Storage, Verst GC, TE Skidmore, and WL Daugherty, FCRD-UFD-2014-000317, May 30, 2014, SRNL-2014-00231,
12. Völzke, H. and Wolff, D., "Aspects on Long Term Storage of Used Nuclear Fuel and High Active Waste in Germany," BAM Federal Institute for Materials Research and Testing, presentation at PATRAM 2010, October 07, 2010.

http://www.tes.bam.de/de/umschliessungen/behaelter_radioaktive_stoffe/dokumente_veranstaltungen/patram_2010/PATRAM2010-PanelP7-Voe-LongTermStorage.pdf

13. Völzke, H. and Wolff, D., “Spent Fuel Storage in Dual Purpose Casks Beyond the Original Design Basis,” in Proceedings of the International High-Level Radioactive Waste Management Conference (INLRWMC), April 28-May2, 2013, American Nuclear Society, 2013.
14. SAND98-1942, “New Methods for Predicting Lifetimes in Weapons. Part 1-Ultrasensitive Oxygen Consumption Measurements to Predict the Lifetime of EPDM O-Rings”, K.T. Gillen, M. Celina, R.L. Clough, G.M. Malone, M. Keenan, J. Wise
15. Bolted Joint Maintenance and Applications Guide, TR-104213, EPRI, December 1995