Performance Characteristics of O-Ring Seals for Radioactive Material Packages When Subjected to Extreme Temperatures*

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INTRODUCTION

Performance requirements for radioactive material (RAM) packages are specified in Title 10, Code of Federal Regulations, Part 71 (NRC 1993). Package components that form the containment boundary must function in both high- and low-temperature environments characteristic of the hypothetical fire accident and the -40°F (-40°C) normal transport condition, respectively. Seals that provide the containment system interface between the packaging body and closure(s) are routinely a source of special consideration when designing, testing, and licensing a RAM package. Seals are most often elastomeric O-rings and can be considered delicate by cask component standards.

A research and testing program has been conducted at Sandia National Laboratories to examine the performance of elastomeric seal materials commonly used in RAM packages during temperature extremes. Performance characteristics examined included leakage rate versus temperature, physical property inspections, and tracer gas permeation. This paper presents the results and findings of the test program (Bronowski 1994). Of particular interest are the use of modified O-ring groove widths and the high temperature testing in excess of manufacturers' standard ratings.

TEST DESCRIPTION

Tests were performed on face seal configuration fixtures under static conditions. In the face seal configuration (shown in Figure 1), compressive force was applied across the O-ring thickness. Fabricated from 304 stainless steel, fixtures consisted of a bottom plate, which contained two concentric O-ring grooves, and a flat top plate. Initially the square grooves were of nominal dimensions matching Parker Seal Company specifications for O-rings with a 1/4-inch nominal cross-sectional diameter (Parker 1991). The grooves provided a nominal compression of 25% as specified by most O-ring manufacturers for vacuum/gas service. These groove designs were modified for tests performed later in the series.

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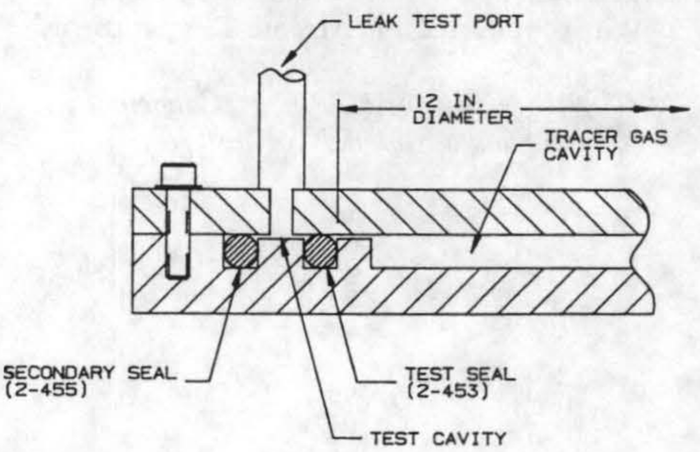


Figure 1. Seal Test Fixture

Figure 2 is a simplified schematic of the test configuration. A leak detector was plumbed to the cavity between the two seals. The tracer gas evacuation pump and supply were plumbed to the central cavity of the fixture. The tracer gas cavity was evacuated and then backfilled with tracer gas while monitoring the detector for leakage.

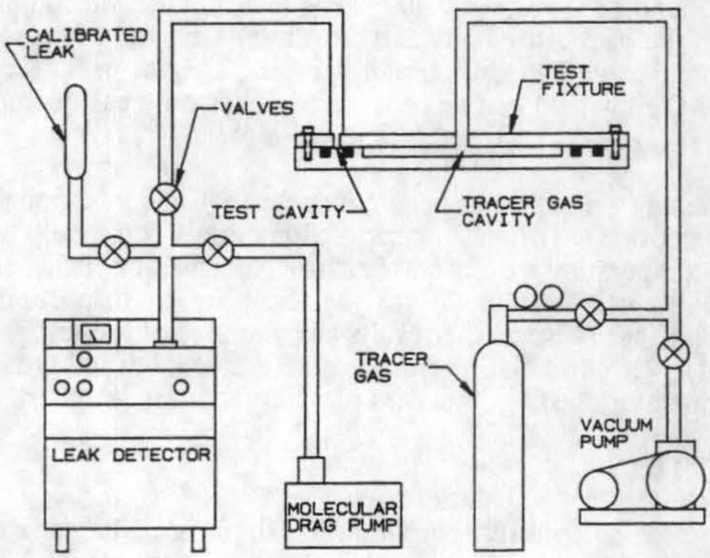


Figure 2. Leak Test Schematic

All measurements taken were made in cubic centimeters per seccond (cm^3/s). For the purposes of this program, a leakage rate of less than $1x10^{-7}$ cm^3/s was considered leaktight, a common reference in the RAM packaging industry. A seal with a rate between $1x10^{-7}$ and $1x10^{-4}$ cm^3/s was as leaking, while a rate in excess of $1x10^{-4}$ cm^3/s was deemed a failure. Since the primary purpose of this test program was to provide guidance to cask designers in material selection, it must be noted that scaling laws for leakage rates do not exist. Measurements from this program were intended to be used as a qualitative rather than a quantitative measure of seal performance. A seal determined to be leaking or failed in this program does not mean that that material should not be considered, as each application has its own criteria.

The leak test system allowed up to three fixtures to be placed in the environmental chamber and piped to an manifold system. Data acquisition was provided by a Hewlett Packard computer and data acquisition unit. Software was written specifically for this system and test program. The program recorded leakage, pressure, and thermal data, and maintained detailed data bases and logs. The program also provided remote control of the manifold system valves and maintained a real time history of every valve manipulation for quality assurance purposes. Fixtures were thermally conditioned using an environmental chamber with operating range of -100°F to 950°F (-73°C to 510°C).

LOW TEMPERATURE TESTS

Low temperature testing was initially performed using fixtures with standard groove dimensions as recommended by Parker Seal Company. Material selection was based on a review of manufacturers' literature, cask designers survey responses, and a review of materials used in currently licensed packages. A total of 26 compounds (Table 1) selected included those from 7 manufacturers and 9 parent chemical groups.

Table 1. Candidate Materials for Low Temperature Tests

Butyl		Neoprene	
B0612-70	Parker	C0873-70	Parker
R0403-50	Rainier	C1124-70	Parker
R0404-70	Rainier		
Fluorocarbon		Ethylene Propylene	
V0747-75	Parker	E0540-80	Parker
V0835-75	Parker	E0740-75	Parker
R1429-70	Rainier		
19657-GLT	Wynns	Fluorosilicone	
Kalrez 4079	Dupont	L0677-70	Parker
Silicone		Polyphosphazene	
S0383-70	Parker	F0953-70	Parker
S0604-70	Parker	R1801-70	Rainier
S0613-60	Parker		
S0899-50	Parker	Teflon^{tm}	
		NPTFE	Parker
Miscellaneous Composite Materials			
Teflon^{tm}/Silicone	Chicago Gasket	Teflon^{tm}/Silicone	Row Co.
Teflon^{tm}/Viton^{tm}	Chicago Gasket	Teflon^{tm}/Viton^{tm}	Row
Teflon^{tm}/Silicone	Creavey	Astro^{tm}/Teflon^{tm}	Creavey

Fixtures were cooled to an initial temperature of +20°F (-7°C) and individual fixture leak tests sequentially performed. Fixtures were then cooled in 10°F (5.5°C) steps with a leak test being performed on each fixture at each step. Fixtures were tested to failure or to -90°F (-68°C), the lower limit of the chamber. The rationale for the extreme low temperatures (in excess of the -40°F regulatory requirements) was to obtain data for comparison to manufacturers' usage ratings.

A summary of low-temperature leak test data is presented in Table 2. With few exceptions, the seals did not regularly remain leaktight to the manufacturers' low temperature ratings. It must be noted that most elastomeric seal applications are in the automotive or hydraulics industries and usually have a different criterion for performance. The gas leakage tests discussed herein were much more stringent and sensitive than a typical test for liquid leakage; for example, no visible water will leak from a known leak that passes dry air at a rate of 1×10^{-4} cm^3/s.

Table 2. Low Temperature Leak Test Data

Material	Number of Tests	Failure Temp-range °F (°C)	Failure Temp - avg °F (°C)	Mfg's Low Temp Rating °F (°C)
Butyl				
B0612-70	12	-10 to -83 (-23 to -64)	-68 (-56)	-75 (-60)
R0403-50	6	-50 to -68 (-46 to -56)	-63 (-53)	-65 (-54)
R0404-70	5	-40 to -68 (-40 to -56)	-53 (-47)	-65 (-54)
Polyphosphazene				
F0953-70	15	-1 to -85 (-18 to -65)	-60 (-51)	-85 (-65)
R1801-70	3	-60 to -80 (-51 to -62)	-73 (-58)	-85 (-65)
Ethylene Propylene				
E0540-80	9	-11 to -61 (-24 to -52)	-40 (-40)	-70 (-57)
E0740-75	6	-49 to -81 (-45 to -68)	-58 (-50)	-70 (-57)
Fluorocarbon				
V0747-75	6	+10 to -20 (-12 to -29)	-1 (-18)	-15 (-25)
V0835-75	17	+20 to -42 (-7 to -41)	-24 (-31)	-40 (-40)
R1429-70	6	+10 to -30 (-12 to -34)	-16 (-26)	-40 (-40)
19657-GLT	9	-19 to -31 (-28 to -35)	-27 (-33)	-40 (-40)
Kalrez 4079	5	+20 to +10 (-7 to -12)	+16 (-9)	-60 (-52)
Fluorosilicone				
L0677-70	9	-10 to -90 (-23 to -68)	-60 (-51)	-100 (-73)
Neoprene				
C0873-70	6	-30 to -41 (-34 to -41)	-34 (-37)	-45 (-43)
C1124-70	12	-36 to -71 (-38 to -57)	-51 (-46)	-65 (-54)
Silicone				
S0383-70	9	-1 to -90 (-18 to -68)	-46 (-43)	-100 (-73)
S0604-70	9	-1 to -65 (-18 to -54)	-35 (-37)	-65 (-54)
S0613-60	15	-70 to -71 (-56 to -57)	-70 (-56)	-60 (-51)
S0899-50	18	-41 to -92 (-41 to -69)	-85 (-65)	-100 (-73)
Teflon				
NPTFE	6	-17 to -90 (-27 to -68)	-52 (-47)	-40 (-40)
Composites				
Tef/Sil -C-G	6	-49 to -65 (-45 to -54)	-54 (-48)	Not Available
Tef/Sil -Crvy	23	+20 to -90 (-7 to -68)	-45 (-43)	-80 (-62)
Tef/Sil -Row	6	-40 to -60 (-40 to -51)	-49 (-45)	-40 (-40)
Tef/Viton -C-G	9	+10 to -31 (-12 to -35)	-11 (-24)	Not Available
Tef/Viton -Row	6	0 to -50 (-18 to -46)	-38 (-39)	-40 (-40)
Astro/Tef -Crvy	6	-31 to -80 (-35 to -62)	-54 (-48)	-80 (-62)

Many compounds did remain leaktight to the regulatory -40°F (-40°C) temperature. Several others had average failure temperatures at or below the -40°F (-40°C) target, with a single failure keeping them from routinely passing the target criterion. Fluorocarbon materials did not perform well, with very few individual tests passing the -40°F (-40°C) test step and average failure temperatures significantly higher. This was as expected, as most of the compounds only had a -40°F (-40°C) rating.

A statistical analysis was performed on the test data. This study found that one of the test fixtures used produced appreciably higher failure temperatures than the other fixtures. After analyzing the data set with this fixture removed, an estimated survival probability was established for each material. This information was then used to select candidate materials for the high-temperature test series.

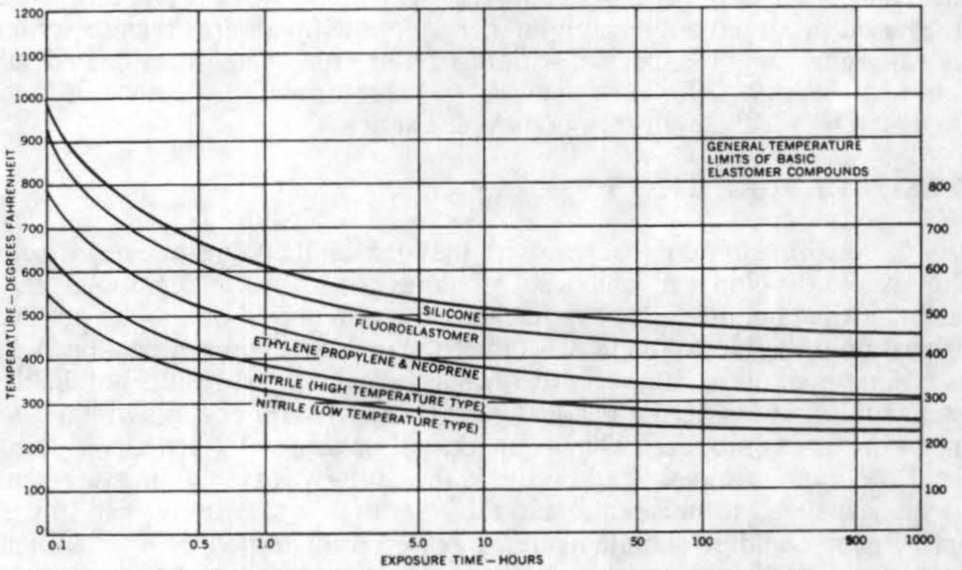
HIGH-TEMPERATURE TESTS

The first task performed in conjunction with the high-temperature test series consisted of scoping tests to develop a reliable test technique using a residual gas analyzer and a tracer gas other than helium. The new technique was required due to the problem of rapid permeation of helium through elastomeric materials. This permeation, which increases with temperature, can mask real leakage, making test results unreliable. The final system configuration used a residual gas analyzer in conjunction with a helium/neon tracer gas mixture. The equal partial pressure mixture served two functions: 1) the helium signal would provide the primary leak rate measurement since it had the highest signal-to-noise ratio and thus the highest sensitivity; and 2) the mass 22 isotope of neon could be simultaneously monitored for response. A simultaneous rise in both signals indicated a real leak, while a delayed response between the two signals denoted permeation.

Candidate materials for the high temperature tests were selected primarily based on the statistical analysis of the low-temperature data. Materials that had a high probability of passing -40°F (-40°C) tests were initially included. Added to the list were V0835 and E0893 materials. In spite of not having performed well in the low temperature testing, V0835 is a widely used material, and additional information was desired. E0893, an ethylene propylene compound, was added, as this material had good temperature ratings but had not been available for the initial test series.

Next, target test temperatures were selected from seal life estimates from Parker Seal Company. Manufacturers' upper temperature limit values typically relate to a 1000-hour life at that temperature. Parker literature presents data that relates estimated life to elevated temperatures. Target test temperatures were selected based on seal life estimates for 10-hour use (Figure 3). These values exceeded the published standard ratings by 50 to 90°F (28 to 50°C), depending on compound family. The intent was to select temperatures for which there were high probabilities for success rather than testing to failure. The rationale for exceeding the standard 1000-hour life rating was that the usual concern for a RAM package is survival during and after the fire accident scenario, a limited duration event.

One observation from the high temperature scoping tests showed that seals were expanding and often completely filling fixture grooves. This was due to the differences in the coefficient of thermal expansion of the seals versus that of the fixtures. Thermal expansion coefficients for candidate materials were identified and maximum expansions based on target temperatures were calculated. Fixture groove widths were redesigned, increasing groove widths to allow a maximum fill of 95%. This resulted in three different fixtures for high temperature tests: a standard width of .305/.310-inch (in.), and two widened designs that were 0.010-in. and 0.020-in. oversized. Groove depths were not changed, leaving nominal compression unchanged.



Reprinted with permission of Parker Seal Group. This chart is intended only as a rough guide; it cannot be used for precise predictions of seal life.

Figure 3. Estimated Seal Life Versus Temperature

Before proceeding with the high temperature test series, an abbreviated low temperature series was conducted on the candidate materials. This series utilized the widened groove fixtures to observe whether the new designs had a negative effect on seal performance. All composite materials were deleted from the test series after failing all tests. All conventional compounds performed as expected based on data from the initial low-temperature series. The only standard elastomer to not regularly pass the test series to -60°F (-51°C) was V0835, which failed two of three tests in a range correlating with previous data. Final candidate materials selected for high-temperature testing are listed in Table 3.

Table 3. Candidate Materials For High Temperature Tests

Material	Target Test Temperature °F (°C)	Mfg. High Temp Rating °F (°C)	Coefficient of Expansion in./in./°F	Fixture Design/Groove Width (in.)
B0612-70	300 (149)	250 (121)	$6.2\text{x}10^{-5}$	Std. (.305/.310)
R0403-50	300 (149)	250 (121)*	$6.2\text{x}10^{-5*}$	Std. (.305/.310)
R0404-70	300 (149)	250 (121)*	$6.2\text{x}10^{-5*}$	Std. (.305/.310)
E0540-80	380 (193)	300 (149)	$8.9\text{x}10^{-5}$	+.010 (.315/.320)
E0740-75	380 (193)	300 (149)	$8.9\text{x}10^{-5}$	+.010 (.315/.320)
E0893-80	380 (193)	300 (149)	$8.9\text{x}10^{-5}$	+.010 (.315/.320)
C0873-70	380 (193)	300 (149)	$7.6\text{x}10^{-5}$	+.010 (.315/.320)
C1124-70	380 (193)	300 (149)	$7.6\text{x}10^{-5}$	+.010 (.315/.320)
F0953-70	380 (193)	350 (177)	$9.0\text{x}10^{-5*}$	+.010 (.315/.320)
R1801-70	380 (193)	350 (177)*	$9.0\text{x}10^{-5*}$	+.010 (.315/.320)
V0835-75	470 (243)	400 (204)	$9.0\text{x}10^{-5}$	+.020 (.325/.330)
S0383-70	520 (271)	430 (221)	$1.0\text{x}10^{-4}$	+.020 (.325/.330)
S0604-70	520 (271)	450 (232)	$1.0\text{x}10^{-4}$	+.020 (.325/.330)
S0613-60	520 (271)	450 (232)	$1.0\text{x}10^{-4}$	+.020 (.325/.330)
S0899-50	520 (271)	430 (221)	$1.0\text{x}10^{-4}$	+.020 (.325/.330)
L0677-70	520 (271)	400 (204)*	$1.0\text{x}10^{-4}$	+.020 (.325/.330)

* Estimated by compound family; data unavailable from manufacturer.
Coefficient of thermal expansion for SS = $9.6\text{x}10^{-6}$ in./in./°F.

Test sequence for the high-temperature test series consisted of an assembly test at room temperature, a test upon reaching target temperature, a test after holding at target temperature for 2 hours, another test after cooling to ambient, and a final leak test at -40°F (-40°C). The first test at the target temperature was included to verify that the seal did not fail during the temperature transient. The 2-hour dwell period simulated an extended fire scenario. Ambient, post-high-temperature tests were performed to show integrity after cooling. The -40°F tests were added for informational purposes, as this step is not normally part of a hypothetical accident sequence.

With one exception, all seals remained leaktight, i.e., a leakage rate of less than 1.0×10^{-7} cm^3/s, throughout the series. All test data from this series are presented in Table 4. The single seal (of nine tested) with measured leakage was of the S0899 silicone compound. This seal failed with a 6×10^{-6} cm^3/s rate for the -40°F (-40°C) test. Upon disassembly of the seal from the test fixture, a minor abrasion was noted. It is not known whether this damage was present during the test or caused at disassembly.

Table 4. High Temperature Leak Test Data

	10 Hour Ratings			3 Hour Ratings		
Compound/ Material	Test Temp °F (°C)	No of Tests	No of Failures	Test Temp °F (°C)	No of Tests	No of Failures
B0612-70	300 (149)	3	0	330 (166)	1	0
R0403-50	300 (149)	3	0	330 (166)	1	0
R0404-70	300 (149)	3	0	330 (166)	1	0
E0540-80	380 (193)	6	0	410 (210)	2	0
E0740-75	380 (193)	6	0	410 (210)	2	0
E0893-80	380 (193)	3	0	410 (210)	1	0
C0873-70	380 (193)	6	0	410 (210)	2	0
C1124-70	380 (193)	6	0	410 (210)	2	0
F0953-70	380 (193)	6	0	410 (210)	2	0
R1801-70	380 (193)	3	0	410 (210)	1	0
V0835-75	470 (243)	9	0	500 (260)	3	0
S0383-70	520 (271)	6	0	550 (288)	2	0
S0604-70	520 (271)	6	0	550 (288)	2	0
S0613-60	520 (271)	6	0	550 (288)	2	0
S0899-50	520 (271)	9	1	550 (288)	3	0
L0677-70	520 (271)	9	0	550 (288)	3	1

At this point a final high-temperature test series was performed. This abbreviated series tested a single seal from each compound and batch at a temperature 30°F (16.7°C) higher than the last test for each material. These temperatures related to an approximate 3-hour life expectancy. These tests were not intended to establish a precise upper limit of survivability for the seals, but rather to give a level of confidence to the previous testing. If seals could pass these tests with any regularity, it would indicate that they were not on the verge of failure in the earlier series.

All seals, with the exception of one fluorosilicone (L0677), performed well, passing all series tests (Table 4). Note that the test temperature for this material had already been increased significantly above the manufacturer's rating in the previous tests. Since the estimated 3-hour life temperature of approximately 460°F (238°C) was also exceeded, failures were not unexpected.

SUMMARY AND CONCLUSIONS

The low-temperature test series showed the need for careful material selection to meet RAM packaging criteria. Manufacturers' ratings typically relate to passage criteria significantly different from that required of RAM packaging and should therefore not be relied upon without careful examination. An added concern for low-temperature seal design that was not evaluated here is relative flange movement due to vibration or shock. A very hard or brittle seal would be unlikely to maintain leaktightness if interface surfaces moved. The majority of materials tested gave the indication of meeting the basic -40°F (-40°C) criteria for RAM packaging. The exception to this was fluorocarbon materials, which failed sufficient tests to be of major concern.

Manufacturers' nominal high-temperature ratings apply to an estimated seal life at 1000 hours. If a package were routinely subjected to a high temperature, as in the case of a high internal heat load, the manufacturers' 1000-hour rating might be a reasonable value for design; however, a hypothetical accident fire is most certainly a singular event of limited exposure time. These tests demonstrated that seals can remain leaktight when subjected to temperatures in excess of standard ratings. Additionally, estimates of life versus temperature can be used, at least as a rough guide, when increasing a seal's design temperature. While reasonableness must be maintained when applying this philosophy to cask design, a 50°F to 90°F (28 to 50°C) increase in usable temperature can greatly aid a designer. It also gives the designer additional options for seal material selection.

Seals performed well in the groove designs utilized in this test series. Specific controlled tests were not performed to compare performance of "standard" groove width fixtures to the modified groove design fixtures. It should be noted that groove designs from several other seal manufacturers define wider nominal groove widths, presumably with a wider temperature range in mind.

It is concluded that seal expansion at high temperature is an important factor in seal groove design in order to obtain satisfactory performance. Maximum temperature and thermal coefficient of expansion should be calculated to ensure that grooves are not overfilled. This is important not only for performance while at high temperature but also at ambient (and possibly lower) temperatures that may be encountered after an accident scenario.

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