

RADIOACTIVE MATERIAL PACKAGE SEAL TESTS*

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ABSTRACT

General design or test performance requirements for radioactive materials (RAM) packages are specified in Title 10 of the United States (U.S.) Code of Federal Regulations Part 71 (U.S. Nuclear Regulatory Commission, 1983). The requirements for Type B packages provide a broad range of environments under which the system must contain the RAM without posing a threat to health or property. Seals that provide the containment system interface between the packaging body and the closure must function in both high- and low-temperature environments under dynamic and static conditions.

A seal technology program, jointly funded by the U.S. Department of Energy Office of Environmental Restoration and Waste Management (EM) and the Office of Civilian Radioactive Waste Management (OCRWM), was initiated at Sandia National Laboratories. Experiments were performed in this program to characterize the behavior of several static seal materials at low temperatures. Helium leak tests on face seals were used to compare the materials. Materials tested include butyl, neoprene, ethylene propylene, fluorosilicone, silicone, Eypel, Kalrez, Teflon, fluorocarbon, and Teflon/silicone composites.

Because most elastomer O-ring applications are for hydraulic systems, manufacturer low-temperature ratings are based on methods that simulate this use. The seal materials tested in this program with a fixture similar to a RAM cask closure, with the exception of silicone S613-60, are not leak tight (1.0×10^{-7} std cm^3/s) at manufacturer low-temperature ratings.

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INTRODUCTION

Type B packages for transporting RAM are required to contain their contents in both high- and low-temperature environments. General design or test performance requirements for RAM packages used in the U.S. are specified in Title 10 of the U.S. Code of Federal Regulations Part 71. Seals, which provide the containment system interface between the packaging body and the closure, must function through the temperature range of -40°C (-40°F) to the hypothetical accident condition maximum that frequently exceeds 149°C (300°F).

In response to a need for seal test data, a seal technology program, jointly funded by the U.S. Department of Energy Office of Environmental Restoration and Waste Management (EM) and the Office of Civilian Radioactive Waste Management (OCRWM), was initiated at Sandia National Laboratories. The program mission is to characterize the behavior of seals commonly used in RAM packages both under conditions experienced in service and during hypothetical accident transport conditions. Low- and high-temperature seal performance is currently being investigated. Axial and horizontal movement of the sealing surfaces, cyclic thermal loading, radiation, pressure, compression set, and aging data at operating temperatures may be explored in the future.

This paper documents initial experiments to characterize the low temperature performance of several elastomeric materials in a static face seal configuration. Helium leak rates are used as the performance measure. Because scaling laws have not been developed for seals (Chivers and Hunt, 1978), the leak rates are intended to be used in a qualitative manner.

BACKGROUND

Many factors influence the leakage rate of a seal system. The parameters that affect sealing capability include properties of the sealing surface, gasket material, closure design, and contents.

Sealing Surface

The ASM Metals Handbook (Boyer and Gall, 1985) define four elements of surface finish: roughness, waviness, lay, and flaws. Roughness consists of irregularities resulting from the manufacturing process and is measured by R_a , which is the arithmetic average deviation of the surface from the roughness center line, expressed in micrometers or microinches. Tests in this activity with surface finishes ranging from $0.40\ \mu\text{m}$ ($16\ \mu\text{in.}$) to $6.41\ \mu\text{m}$ ($250\ \mu\text{in.}$) showed little benefit from finishes finer than $1.62\ \mu\text{m}$ ($64\ \mu\text{in.}$). Therefore, the majority of the low-temperature tests were conducted with the $1.62\ \mu\text{m}$ ($64\ \mu\text{in.}$) finish.

Lay is the predominant surface pattern, ordinarily determined by the production process. Because tests with fixtures having circumferential and radial machining directions showed improved low-temperature performance with circumferential machining, the circumferential machining direction was used for all low-temperature tests.

Waviness includes all irregularities with spacing greater than the roughness sampling length. It may result from machine vibration or heat treatment. Flaws are irregularities that occur at widely varying intervals on the surface. They include cracks, inclusions, ridges, and scratches.

Seal Material

Elastomers and metals are the principal materials used for Type B RAM package seals. A catalog of closure seal materials used in packages certified by the U.S. Nuclear Regulatory Commission reveals that elastomers are more frequently used than metal (Warrant and Ottinger, 1989). Elastomer seals are usually less expensive, may be reused many times, and require less maintenance of the seal surface. An advantage of metal seals is better performance at high operating temperatures. Elastomer materials were selected for the initial tests; future plans include testing of metal seals.

Closure Design

The design of a seal system is determined by its intended application. Types of seals are separated into the general categories of dynamic and static. In dynamic seals, the sealed surfaces are in motion. Static seals with stationary sealing surfaces are used in RAM packages. A static face seal compresses an elastomer on the top and bottom; a static bore seal compresses an elastomer between the inner and outer diameters. The initial tests use a static face seal configuration that is frequently used in spent fuel cask designs.

Frequently the static face seal is held in place by a square or dovetail groove. The initial tests used the square groove with dimensions recommended by seal manufacturers that are more economical to fabricate.

Contents

Tests conducted on various seal materials for radioactive material transport indicate no elastomer can be expected to give lengthy service after a total dosage of 10^7 Gy (10^9 rad) of gamma radiation. Radiation and elevated temperatures produce compression set and physical degradation of the elastomer (Parker Seal Group, 1990; Wynn's-Precision Inc., 1989). Testing by Parker indicates most elastomers maintain sealing capabilities after exposure to 10^4 Gy (10^6 rad) of radiation at room temperature. The elastomers in the initial tests were not exposed to radiation and will provide baseline data in later tests.

During the design of spent fuel casks radiation dosages are evaluated to assure that levels are below those which would affect performance. Additionally, radiation exposure of seals is evaluated when length of service for this component is determined.

SEAL TEST DESCRIPTION

The face seal fixture, shown in Figure 1, is constructed of 304 stainless steel. O-rings of 0.699-cm (0.275-in.) cross-sectional

diameter are positioned in grooves of nominal diameters of 30.5 cm (12 in.) and 33.0 cm (13 in.) in a bottom circular plate. The groove dimension was designed for a 25% compression. Upper circular plates with surface finishes of 1.62- μm (64- $\mu\text{in.}$) roughness are attached to the bottom plate, with 24 closure bolts torqued to 6.1 N·m (100 in.·lb). Temperature of the seal fixture is monitored by type K thermocouples embedded in the fixture adjacent to the seals.

Both the central cavity and the space between the two seals are evacuated to less than 1.0×10^3 Pa (.01 atm). Helium is released into the central cavity (approximately 30 s) until a pressure differential of approximately 1.0×10^5 Pa (1.0 atm) across the inner seal is reached. Next the leakage into the space between the seals is measured for 30 s with an Alcatel ASM 51 helium mass spectrometer. The helium detection response time of the leak detector is less than 1 s. Upper and lower detection limits of the leak detector are approximately 10^{-5} and 10^{-10} std cm^3/s helium, respectively.

Up to three fixtures are placed in the temperature-conditioning unit, as shown in Figure 2. The use of multiple fixtures allows investigation of material repeatability. The temperature range of the Tenney Six environmental chamber is -68°C (-90°F) to 199°C (390°F). A Hewlett Packard 9000-340 computer and a 3852 data acquisition unit control the testing, and record the leak rate and temperature data. Each fixture is first tested at an initial temperature of -7°C (20°F). The temperature is then decreased in 5.6°C (10°F) steps until the upper detection limit of the leak detector is exceeded at approximately 10^{-5} std cm^3/s helium or when the temperature reaches -68°C (-90°F). Following testing at each step, the helium is pumped from the fixture inner cavity to minimize permeation. The total time needed for a test varies from 4 to 6 h, depending on how low a temperature is required to exceed the upper detection level of the leak detector (loss of vacuum).

O-ring materials used in the test series include butyl, neoprene, ethylene propylene, fluorosilicone, silicone, Eypel, Kalrez, Teflon, and fluorocarbon. Two composite materials were also tested. One composite-material seal is constructed with a silicone core encapsulated in Teflon and the other utilizes a stainless steel spiral core encapsulated in Teflon. Lot numbers for each O-ring were recorded to study lot-to-lot variations.

TEST RESULTS

The low temperature data are summarized in Table 1. The first column lists the 18 materials tested. The number of tests on each material varies from 3 to 18. (As time permits, a larger sample of tests will be conducted for selected seals.) The entries in Column 3 give ranges of vacuum loss for the tests. Vacuum loss on the Alcatel leak detector occurs at approximately 1×10^{-5} std cm^3/s . All loss of vacuum values for a seal material are averaged in Column 4. Figure 3, a representative low-temperature leak plot, shows that loss of vacuum occurs abruptly. As the tests were conducted at 5°C (10°F) decrements, the leak-tight temperature could be as much as 5°C (10°F) above the loss of vacuum temperature.

Table 1

Low Temperature Seal Data With 25% Compression and 1.6 μm (64 $\mu\text{in.}$) Finish, Unless Footnoted

Material	Number of Low Temp. Tests	Range °C (°F)		Aver. °C (°F)		Mfr. Rating °C (°F)
		Loss of Vacuum ¹		Loss of Vacuum		
1. Silicone S899-50	18	-40 to -68	(-40 to -90)	-65	(-86)	-73 to 206 (-100 to 400)
2. Silicone S613-60	15	-57	(-70)	-57	(-70)	-52 to 234 (-60 to 450)
3. Silicone S383-70	7	-35 to -68	(-30 to -90)	-49	(-55)	-116 to 206 (-175 to 400)
4. Silicone S604-70	5	-46 to -57	(-50 to -70)	-52	(-60)	-52 to 234 (-60 to 450)
5. Neoprene C1124-70	12	-40 to -52	(-40 to -60)	-47	(-52)	-54 to 122 (-65 to 250)
6. Fluorocarbon V835-75 ²	10	-29 to -40	(-20 to -40)	0	(-32)	-40 to 206 (-40 to 400)
7. Butyl B612-70	6	-23 to -63	(-10 to -80)	-52	(-60)	-60 to 108 (-75 to 225)
8. Fluorosilicone L677-70	6	-43 to -68	(-45 to -90)	-56	(-68)	-73 to 178 (-100 to 350)
9. Ethylene Propylene E740-75	6	-46 to -63	(-50 to -80)	-50	(-58)	-57 to 122 (-70 to 250)
10. Ethylene Propylene E540-80	6	-23 to -46	(-10 to -50)	-32	(-26)	-57 to 122 (-70 to 250)
11. Neoprene C873-70	6	-35 to -40	(-30 to -40)	-36	(-33)	-43 to 122 (-45 to 250)
12. Fluorocarbon V747-75	6	-12 to -29	(+10 to -20)	-19	(-2)	-26 to 206 (-15 to 400)
13. Eypel F0953-70	7	-46 to -63	(-50 to -80) ³	-57	(-70)	-65 to 178 (-85 to 350)
14. Eypel F-70-1801	3	-52 to -63	(-60 to -80)	-59	(-73)	-68 to 178 (-90 to 350)
15. Teflon PTFE	3	-29 to	(-20 to) ⁴	-53	(-63)	-40 to 234 (-40 to 450)
16. Teflon/Silicone	5	-35 to -68	(-30 to -90) ⁵	-55	(-66)	-63 to 206 (-80 to 400)
17. Teflon/Stainless Steel	5	-35 to -63	(-30 to -80)	-54	(-65)	-63 to 206 (-80 to 400)
18. Kalrez 4079 ⁶	5	-12	(+10)	-12	(+10)	-52 to 318 (-60 to 600)

¹Vacuum loss at $\sim 1 \times 10^{-5}$ std cm^3/s .²See text for results with 0.4 μm (16 $\mu\text{in.}$) finish.³Seals from 2 different batches.⁴No vacuum loss at -68°C (-90°F) (lower limit of environmental chamber).⁵Teflon joint is not uniform.⁶0.4 μm (16 $\mu\text{in.}$) finish, manufacturer specified.

Wide loss of vacuum ranges are observed for several seal materials. Physical examination of the O-rings did not reveal flaws or significant dimensional differences. A characteristic of elastomers is that the initial response to stress, whether static or dynamic, changes over the first 2 to 5 deformations and then approaches an equilibrium level. This phenomenon, known as the Mullens effect (Mullens and Tobin, 1965), was investigated as a possible cause of the wide loss of vacuum ranges. Following compression of the O-rings in the test fixture for 30 min, the fixture was unbolted. This procedure was repeated 6 times. Subsequent leak tests still showed similar wide loss of vacuum ranges. The basis for this behavior is unknown and will continue to be explored.

The temperature retraction at 10% (TR-10) low-temperature test procedure requires stretching a specimen 50% and determining the temperature at which the specimen retracts 10%. Manufacturer low-temperature ratings on seals frequently are based on temperatures approximately 8°C (15°F) below TR-10 values obtained by testing according to American Society for Testing Materials (1987) D1414-78, Standard Test Methods for Rubber O-Rings. Manufacturer high-temperature ratings often are based on continuous operating performance of 1000 hours in the media the compounds are designed to contain (Parker Seal Group, 1990). Column 5 of Table 1 records the manufacturer low- and high-temperature rating for each seal. A comparison of loss of vacuum values and low-temperature ratings shows that leak tightness has not been demonstrated at the low-temperature rating.

Seals from two different batches of Eypel F0953-70 were tested. One batch demonstrated a 5°C (10°F) loss of vacuum variation, while the second showed 17°C (30°F) variation.

The Teflon/silicone and Teflon/stainless steel seals use a fused joint. Some seals had a larger cross section at the joint. This could have produced the large loss of vacuum variations for these materials.

The fluorocarbon viton 835-75 is a candidate seal material for designers because of its high-temperature rating of 204°C (400°F) and low permeability. Following the tests with a 1.62µm (64 µin.) finish, tests were conducted with a 0.4 µm (16µin.) finish. Both finishes lost vacuum in the -29° to -40°C (-20° to -40°F) range.

CONCLUSION

Most elastomer seal applications are in the automotive and aircraft industries as evidenced by manufacturer low-temperature ratings. The helium leak test commonly used in RAM transportation is very stringent and apparently different from the manufacturer low-temperature basis. The seal materials tested in this program with a fixture similar to a RAM cask closure, with the exception of silicone S613-60, are not leak tight (1.0×10^{-7} std cm³/s) at manufacturer low-temperature ratings.

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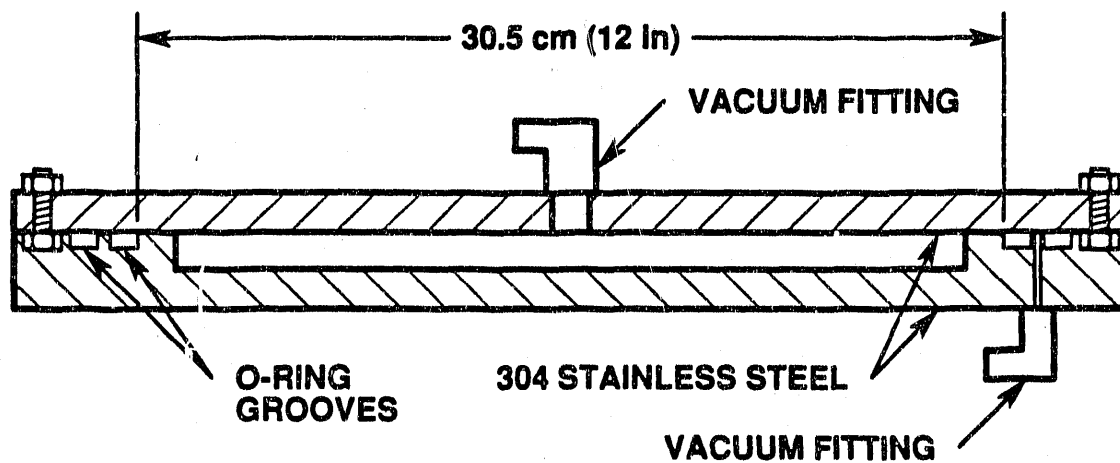


Figure 1. Seal Test Fixture Configuration

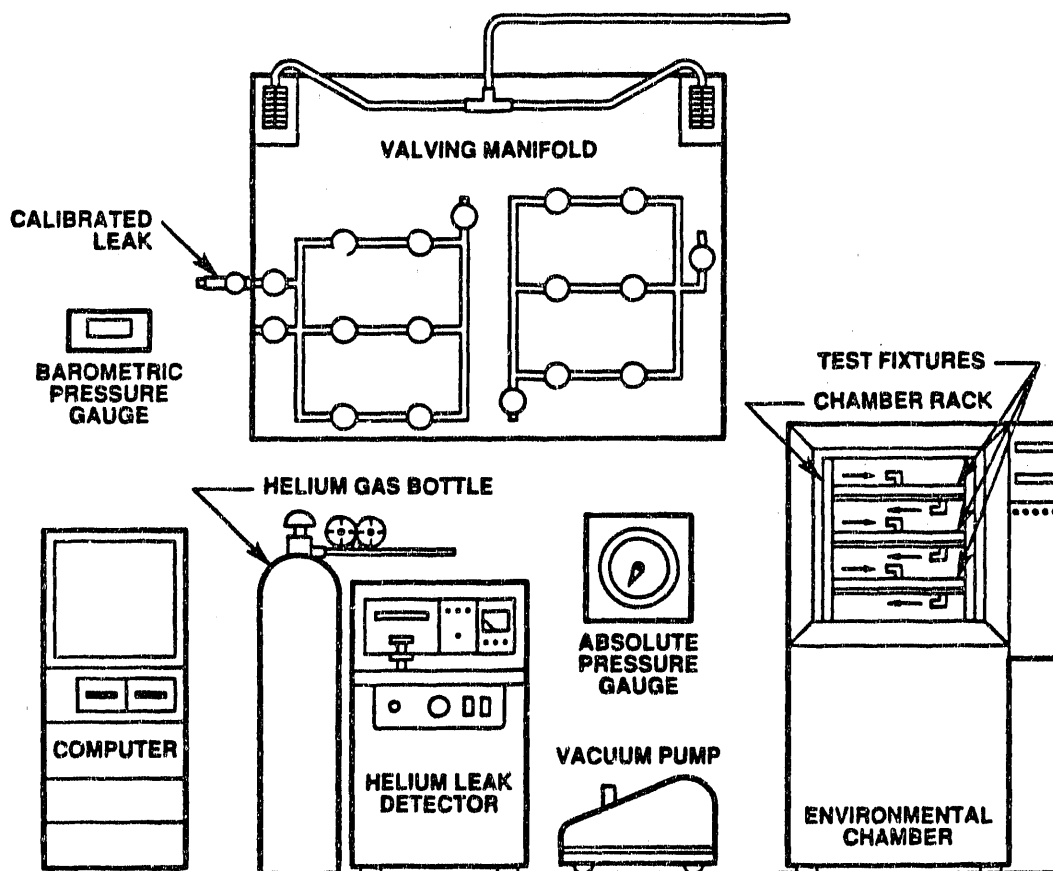


Figure 2. Schematic of Automated Seal Test System Components

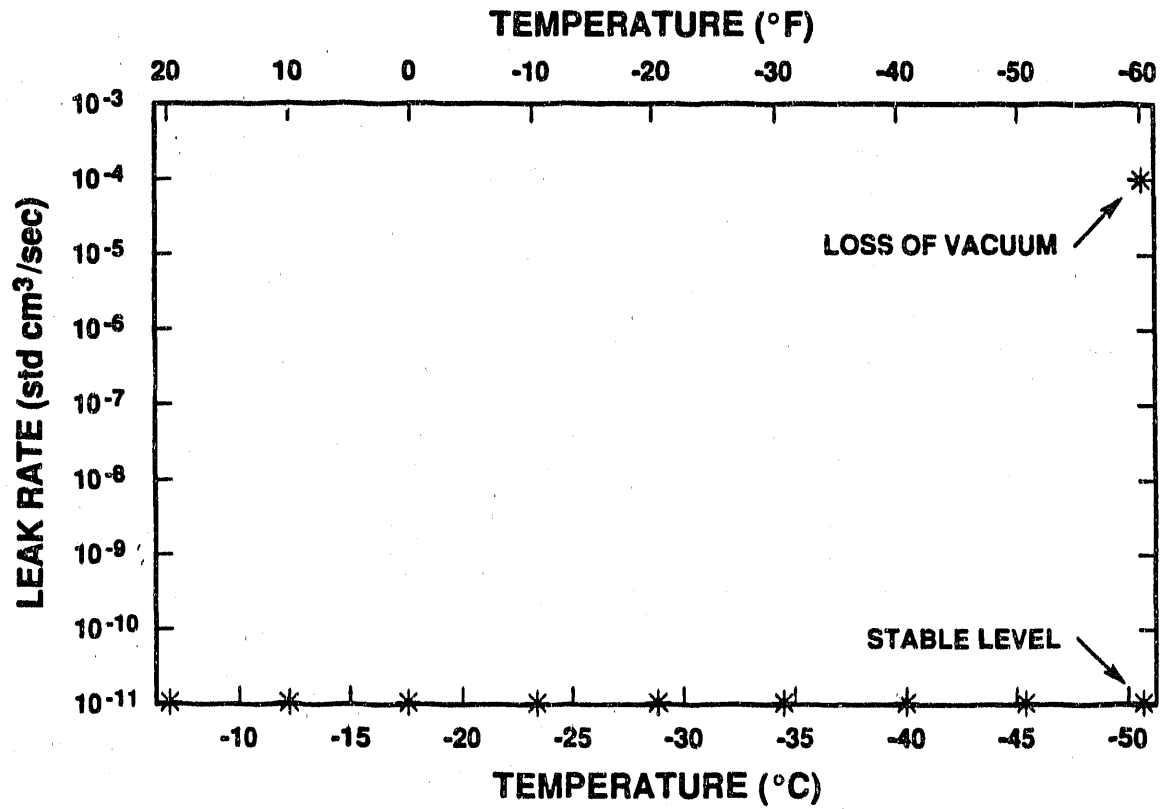


Figure 3. Representative Low Temperature Leak Test

List of Captions

Figure 1. Seal Test Fixture Configuration

Figure 2. Schematic of Automated Seal Test System Components

Figure 3. Representative Low Temperature Leak Test

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