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SPENT NUCLEAR FUEL TRANSPORTATION PACKAGE SEALS IN BEYOND DESIGN BASIS TEMPERATURE EXCURSIONS

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ABSTRACT

The US Nuclear Regulatory Commission (NRC) is studying the performance of seals in spent nuclear fuel (SNF) transportation packages exposed to fires that could exceed the hypothetical accident condition fire described in Title 10 of the Code of Federal Regulations, Part 71, such as the Baltimore Tunnel Fire that occurred in 2001, or the MacArthur Maze fire that occurred in 2007. The performance of package seals is important for determining the potential for release of radioactive material from a package during a beyond-design-basis accident. Seals generally have lower temperature limits than other package components and are the containment barrier between the environment and the radioactive package contents. The NRC Office of Nuclear Regulatory Research contracted the National Institute of Standards and Technology to conduct small-scale thermal testing to obtain experimental data of the performance of seals during extreme temperature exposures. The experimental testing consisted of several small-scale pressure vessels fabricated with a modified ASME flange design and tested metallic and polymeric seals, similar to those that might be used on an actual SNF transportation package. The vessels were heated in an electrical oven to temperatures as high as 800°C (1472°F), exceeding the rated temperatures of the seals in question. This paper will provide a summary of the testing conducted and present test results and conclusions.

BACKGROUND

The Nuclear Regulatory Commission (NRC) is collecting data to better characterize the performance envelope of seals used on spent nuclear fuel (SNF) transportation packages during fire exposures that exceed the hypothetical accident condition (HAC) fire described in 10CFR Part 71 Section 73 [2]. An example of an accident that could produce this type of exposure was the Baltimore tunnel fire that occurred in 2001. The performance of package seals is important for determining the potential for release of radioactive material from a package during a beyond-design-basis accident because the seals, in general, have lower temperature limits than other package components.

NUREG/CR-6886, “Spent Fuel Transportation Package Response to the Baltimore Tunnel Fire Scenario” [1], describes in detail an evaluation of the potential theoretical release of radioactive material from three different spent fuel transportation packages. This evaluation determined the temperatures of various components of the packages, including the seals, using temperatures resulting from models of the Baltimore tunnel fire (as boundary conditions) and finite element models of the SNF packages. For two of the packages evaluated, the model-estimated temperatures of the seals exceeded their continuous-use rated service temperature, meaning a release of radioactive material, such as Cobalt 60 (from CRUD) or Cesium 137 (from fission products), could not be ruled out with available information. However, for both of those packages, the analysis determined by a bounding calculation that the maximum expected release was well below the regulatory limits for a release during the HAC series of events in 10 CFR Part 71.

Testing of the types of seals used in SNF packages to determine their performance in beyond-design-basis thermal exposures can provide the physical data needed to understand the potential for a release of radioactive material during a severe fire accident. Previous work on the thermal performance of package seals has focused on temperatures well below 800°C (1472°F), [3][4]. The test fixture typically consisted of two flanges or two plates with two concentric O-ring grooves, one for the test seal and one for the containment seal, and a small cavity for helium tracer gas.

EXPERIMENTAL APPARATUS AND TEST METHOD

The test vessels were composed of a cylindrical shell and a flange fabricated from Stainless Steel 304 (SS304). The flange dimensions were made in conformity with the ASME Standard B16.5-2009, flange class 2500 with a design pressure rating up to 29.2 bar (423.5 psi) (Table 2-2.1, ASME Standard B16.5-2009) [5]. The vessel cavity had a nominal internal volume of 100 mL (6.1 in³). The seal was a metal O-ring made from

Inconel 718 and silver with an outer diameter of 6.35 cm (2.5 in) and a cross section of 0.32 cm (0.125 in).

The vessel body and the flange were joined together using four bolts (SS 304 1-1/8 in 7TPI), each tightened with a torque of 416 N·m (307 ft·lb) \pm 2 N·m using a micrometer torque wrench. A 24 cm (9.5 in) long SS tube with an inside diameter of 0.48 cm (0.189 in) and an outside diameter of 0.953 cm (0.375 in) was inserted into the bottom of the vessel body flush through a straight-hole with a bevel-groove and was all-around fillet welded to the vessel. The exposed end of the tubing was connected to a union cross equipped with two needle valves or bellow valves for filling and evacuating the test vessel and a tee connection for mounting a pressure transducer and a thermocouple to monitor the vessel pressure and temperature. A schematic of the experimental set up is provided in Figure 1. Photographs of the actual experimental setup are provided in Figure 2.

The exposure of the seal to high temperature environment was achieved using a programmable temperature-controlled electrical furnace with an internal capacity of 25.4 cm x 25.4 cm x 40.64 cm (10 in x 10 in x 16 in). Four Type K grounded thermocouples were used to monitor the transient temperature distribution of the test fixture.

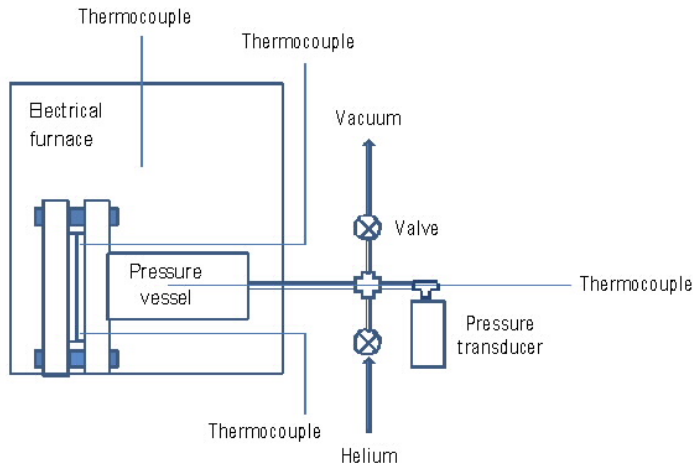


Figure 1: Schematic of experimental apparatus

The test series involved the thermal exposure of a test vessel with a metallic or a polymeric seal in an electric furnace to determine the seal performance at elevated temperatures. The test vessel with the test seal in place was first evacuated and then filled with helium at room temperature to nominal pressure of 5 bar (72.5 psi). The vessel was immediately tested for leaks using soapy water. The vessel pressure was then monitored for more than 48 hrs to further monitor for leaks. The vessel was placed in the electrical furnace and heated from room temperature to a variety of test temperatures. It typically took about 4 hours for the vessel to come up to temperature. Once the flange temperature had reached the pre-selected temperature, in most cases, the vessel was heated for an additional 9 hours at that temperature. For some of the testing, the temperatures were “ramped up” at selected time intervals. The furnace was then

turned off, and the vessel was allowed to cool to room temperature inside the furnace. The internal temperature and pressure of the vessel, the furnace temperature, and the flange temperatures were recorded during the heat-up and cool-down phases using a LabVIEW-based 16-bit DAQ (Data Acquisition) system with an input/output connector block. Table 1 provides a summary of the test conditions and parameters for all the tests conducted.

TABLE 1: TEST CONDITIONS AND PARAMETERS

Test #	Vessel #	Seal Type	Nominal Initial Vessel Conditions	Exposure duration
1*	1	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Heat-up + 30 min at 800 °C (1427°F) + cool-down
2	2	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Heat-up + 9 h at 800 °C (1427°F) + cool-down
3	3	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Heat-up + 9 h at 800 °C (1427°F) + cool-down
4	4	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Heat-up + 9 h at 800 °C (1427°F) + cool-down
5	5	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Heat-up + 9 h at 427 °C (800°F) + cool-down
6	2**	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Heat-up + 9 h at 427 °C (800°F) + cool-down
7	1**	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Heat-up + 9 h at 427 °C (800°F) + cool-down
8	6	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Heat-up + 9 h at 800 °C (1427°F) + cool-down
9	1***	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Heat-up to 427 °C (800°F) and then to 800°C (1427°F) for about 4 h + cool-down
10	7	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Incremental heating from 427°C (800°F) to 627°C (1160°F) with 100°C increment [§] + cool-down
11	3**	Ethylene-propylene	24°C (75°F) at 2 bar (72.5 psi)	Incremental heating from 150°C (302°F) to 300°C (572°F) with 50°C increment ^{§§} + cool-down
12	3**	TFE	24°C (75°F) at 2 bar (72.5 psi)	Incremental heating from 150°C (302°F) to 300°C (572°F) with 50°C increment ^{§§} + cool-down
13	8	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Incremental heating from 427°C (800°F) to 727°C (1340°F) with 100°C increment [§] + cool-down
14	9	Metallic	24°C (75°F) at 5 bar (72.5 psi)	Heat-up + 9 h at 800 °C (1427°F) + cool-down
15	3**	Ethylene-propylene	24°C (75°F) at 2 bar (72.5 psi)	Heat-up + more than 24 h at 450 °C (842°F) + cool-down

*Shakedown test; during this test DAQ malfunctioned and no temporal data was collected.

**Flange and groove surfaces refurbished

***Flange and groove surfaces refurbished again

§Vessel was heated at each set temperature for 9 hours or more

§§Vessel was heated 150°C (302°F) for 1 hour, 200°C (392°F) for 1 hour, 250°C (482°F) for 1 hour and 300°C (572°F) for more than 20 hours

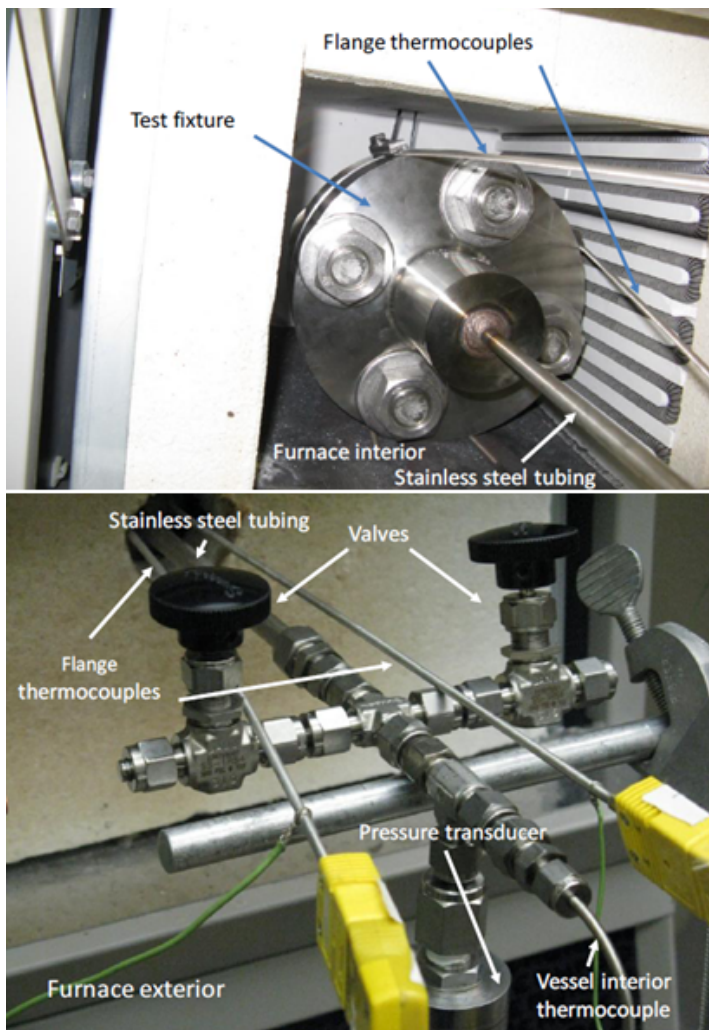


Figure 2: Test rig: test vessel inside the furnace with thermocouple (top, [Note: Furnace was insulated while test was ongoing]) and pressure transducer in place (bottom)

Test Results and Discussion

During the heat-up phase of the vessel, the temporal variation of vessel pressure could not readily be used to determine if there was a potential leak unless a catastrophic seal failure occurred, causing a significant drop in pressure. As the vessel was heated, the vessel pressure and temperature increased. If there was a very small leak, the reduction in pressure due to the reduction in helium in the vessel from the leak could easily be compensated for by the increase in pressure due to increasing temperature. The net effect would still indicate an increase in pressure, thus masking the leak. The use of the temporal variation of vessel pressure as a means to detect potential leakage is best applied to conditions where the vessel is at a constant temperature.

The thermal exposure test using Vessel #1 was intended to be a shakedown experiment. The entire heating process took about 4.5 hrs for the flange temperature to reach the equilibrium furnace temperature of 800°C (1472 °F). The vessel was then maintained at this temperature for an additional 30 min before turning off the electrical furnace. During this shakedown test the DAQ readings from the pressure transducer and

thermocouples became erratic, but the DAQ was diagnosed and repaired for subsequent tests. The readings from the pressure transducer and the thermocouples were recorded manually using a voltmeter and a thermocouple reader, respectively. At 800°C (1472 °F), the vessel pressure reached 14.6 bar (211.8 psi) and was holding at 14.5 bar (210.3 psi) for an additional 30 min of heating. After the vessel was cooled to room temperature, the vessel pressure recovered to its initial pressure of 5.0 bar (72.5 psi), which indicated the absence of a leak. A post test inspection of the tested vessel revealed that the metallic seal was soldered to the flange of the vessel body and a silver-colored coating was imprinted on the surface of the O-ring groove. The high-temperature exposure also discolored the test fixture.

Test #2 included a heat-up to a flange temperature of 800°C (1472°F) and exposure at that temperature for 9 hours. For this test (although the results are not presented here) the vessel pressure starts to decrease shortly after the vessel temperature reached 800°C (1472 °F) and continues to decrease during the rest of the 9 hour constant-temperature heating phase. Although the decrease in pressure, which is within the expanded measurement uncertainty of the pressure transducer, is not significant in this test, the continuous downward trend implied a very small leak.

In order to demonstrate repeatability of the results, Tests 3 and 4 repeat Test #2. Figures 3 and 4 present flange and furnace temperatures during the heat-up, the constant-temperature heating (isothermal) exposure, and the cool-down phases for Tests 3 and 4, respectively. The occurrence of a leak is noted during the 9 hour constant-temperature heating phase at 800°C (1472 °F). In Test #3, the vessel pressure decreases slowly initially and then significantly after about 25000 sec at 800°C (1472 °F). Since the leakage rate is directly proportional to the time rate of change of pressure, this two-stage decrease in pressure indicates a slower leak rate at first and then a faster leak rate at the end. In Test #4, the pressure remains relatively constant for about 10000 sec initially during the 9-hour constant-temperature heating phase, begins to drop significantly for about 15000 sec, and decreases slowly for the remaining duration.

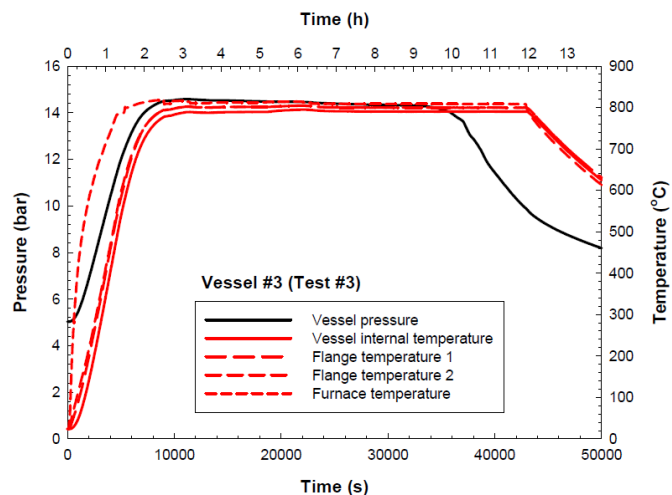


Figure 3: Temporal variations of vessel pressure and temperature in Test #3

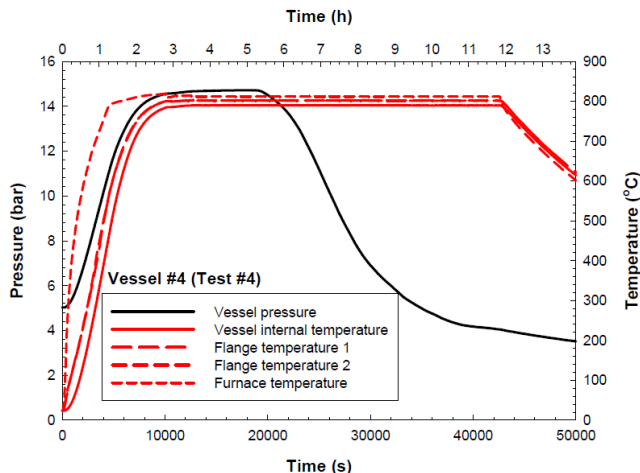


Figure 4: Temporal variations of vessel pressure and temperature in Test #4

It is interesting to note that despite the consistent occurrence of a leak in these three tests, the vessel pressure remained above atmospheric pressure at the end of the cool-down-phase, which took several days. This seems to indicate the possibility that during the heat-up phase, the silver coating on the metallic seal (with a melting point of 962°C) softened and “flowed” in the O-ring groove. At that point it may not have been able to hold the system pressure and a slow leak commenced. During the cool-down phase, the softened silver hardened, bonded to the flange surface, and left a silver coating on the O-ring groove surface. The hardening of silver might have re-sealed the vessel. While it cannot be verified experimentally, the test results discussed below appear to indicate the possibility of re-sealing. The ability of the vessel to maintain pressure following the exposures provides further support, in addition to the photographic evidence described above, to the possibility that re-sealing could potentially occur as the silver in the seal cools.

For Test #5, the vessel pressure remains constant during the entire 9-hour constant-temperature heating period at 427°C (800°F). The seal held vessel pressure. This was further confirmed by the fact that the initial pressure of 5 bar (72.5 psi) was recovered after the cool-down phase. A postmortem inspection revealed that the metallic O-ring seal was not soldered to the flange surface and no silver coating from the seal was transferred to the O-ring groove.

Test #6 was performed using the refurbished Vessel #2. The refurbishment only involved the re-facing of the surfaces of the flanges and the O-ring groove to the specified tolerances. For this test, the vessel pressure started to decrease very slowly during the constant-temperature heating period. The vessel experienced a very small continuous decrease in pressure which seems to imply that a very small leak might have occurred during the test.

Test #7 was performed using the refurbished Vessel #1. The same refurbishing process used in Vessel #2 was applied to Vessel #1. The vessel pressure remained unchanged during the 9-hour constant-temperature heating period. No leak was observed during Test #7.

It is not known if the leak in the refurbished Vessel #2 was caused by a different thermal response of the vessel, which had previously been exposed at 800°C (1472°F) for 9 hours. Although Vessel #1 was also refurbished, the previous exposure time at 800°C (1472°F) was much shorter than that of Vessel #2 (30 min vs. 9 hrs).

Test #8 was performed using Vessel #6 and in similar conditions to Tests #2, 3, and 4. Figure 5 shows the thermal exposure results for Vessel #6. The pressure trace indicates that leakage occurs after 3 h exposure to 800°C (1472°F). For this test, the pressure initially decreases very rapidly, followed by a gradual decrease in vessel pressure; however, the pressure in the test never dropped below 2 bar (29 psi), indicating that the seal had some residual sealing capacity after the exposure.

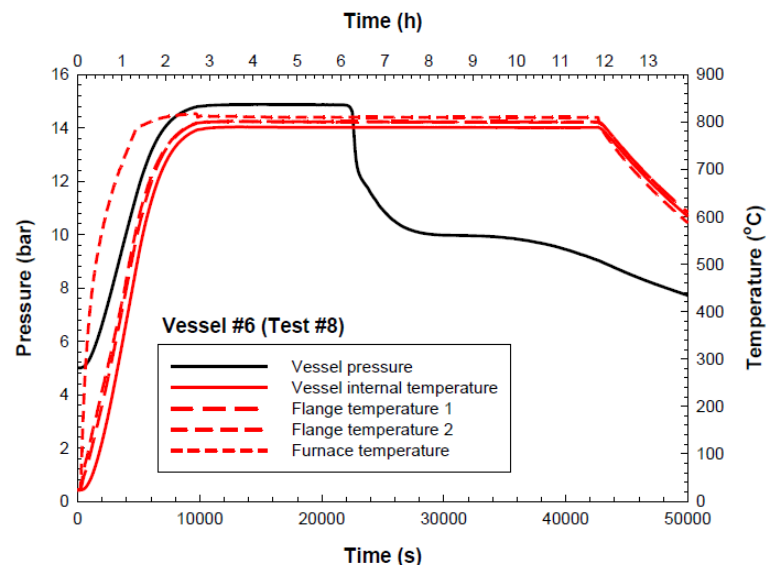


Figure 5: Temporal variations of vessel pressure and temperature in Test #8

Test #9 was performed using Vessel #1, which had been used once at 800°C (1472°F) for 30 min and once at 427°C (800°F) for 9 h and refurbished twice. The sole intent of this test was to check the performance of the pressure transducer against a pressure gauge (WIKA model with a range of 0 psig to 200 psig and a resolution of 2 psig) to ensure that the pressure transducer functioned properly. The only difference between this arrangement and the original set-up was the addition of a pressure gauge. The pressure transducer clearly functioned properly, and the transducer readings corresponded, within the measurement uncertainties, to those obtained from the pressure gauge.

Test #10 was performed using Vessel #7 and was an incremental heating protocol was used to assess the metallic seal

performance. The vessel was initially heated to 427°C (800°F) and held for 9 h at this temperature. Then, the furnace temperature was raised to 527°C (980°F), and the vessel was heated at 527°C (980°F) overnight (more than 9 h). The vessel was then heated to 627°C (1160°F) for 9 h. No leak was observed based on the pressure traces at the test temperatures (427°C [800°F], 527°C [980°F], and 627°C [1160°F]) and the recovery of the initial pressure at room temperature. Post-test removal of the O-ring from the groove was very easy. A postmortem inspection of the vessel indicated that the metallic seal did not solder onto the O-ring groove.

Test #11 was performed using refurbished Vessel #3 and with an ethylene-propylene compound O-ring. Vessel #3 was refurbished by re-facing the flange to remove the O-ring groove originally machined for the metallic seal and re-machining a new O-ring groove for the ethylene-propylene compound O-ring which has different dimensions from its metallic counterpart. During the first 1 h heating at 150°C (302°F), the vessel pressure remains constant. The subsequent 1 h heating at 200°C (392°F) and the following 1 h heating at 250°C (482°F) also indicate the maintenance of vessel pressure. The final heating at 300°C (572°F) for more than 20 hours shows a slow decrease in pressure, albeit within the pressure measurement uncertainty. However, the vessel pressure is restored to its initial value after the thermal exposure test. Post-mortem inspection of the exposed vessel revealed that the O-ring was still pliable and could be easily removed from the O-ring groove.

Test #12 was performed using refurbished Vessel #3 and with a tetrafluoroethylene (TFE) O-ring. The refurbishment of the vessel simply consisted of cleaning of the O-ring groove previously used for the ethylene propylene seal test (Test #11). During the 22 h heating at 300°C (572°F), there is a slight drop in vessel pressure at the end of the isothermal heating phase; however, the pressure decrease was within the measurement uncertainty of the pressure transducer. The calculated reference helium leakage rates during the 22 h heating period, indicates a time-averaged reference leakage rate of 4.5×10^{-5} ref-cm³/s-He. Leakage was observed in this test during the cooling phase. The original pressure (~ 2 bars [29 psi]) was never recovered, and the vessel pressure dropped below the original pressure at room temperature.

Test #13 used a metallic seal and new Vessel #8 was used. This test was performed using incremental heating from 427°C (800°F) to 727°C (1340°F) with a 100°C heat-up increment. This vessel was evacuated (for 1 min) and filled with helium (to 5 bar [72.5 psi]) and then evacuated and filled again twice. Contrary to the previous vessel filling process used in Tests #1 to #12, two additional evacuating and filling cycles were applied in order to determine if the evacuation process had any effect on the results. This test had no leak and no difference could be noted in the pressure vs. temperatures due to the purging of helium. Test #14 and #15 were also evacuated and filled with helium following Test #13 protocol.

Test #14 used a metallic seal and new Vessel #9 was used. The conditions of this test were similar to Tests 2 – 5, with heat-up to 800°C (1427°F) for 9 hours. No leak was detected on this test.

Test #15 was performed using refurbished Vessel #3 with an ethylene-propylene compound O-ring. The refurbishing involved simply cleaning the O-ring groove and the cavity of the used vessel with ethanol. The conditions of this test were similar to Test #11, except that the temperature used for the thermal exposure test was 450°C (842°F) for an extended period of time (24+ hrs) instead of 300°C (572°F). The pressure trace indicates a leak occurred soon after the vessel had attained the nominal target temperature of 450°C (842°F). The calculated reference helium leakage rates over the 25 h isothermal heating period at 450°C (842°F) was a time-averaged reference leakage rate of 9.2×10^{-4} ref-cm³/s-He. From Figure 6 a peculiarity is noted. The measured attainable pressure (~ 8.5 bars [123 psi]) at 450°C (842°F) before the leak occurred is noticeably higher than the pressure calculated using the ideal gas law (~ 4.9 bars [71psi]), even assuming no thermal expansion of the vessel volume. The addition of vapor mass from the potential thermal decomposition and off-gas of the O-ring and the lubricant used at 450°C (842°F) is conjectured to be the cause for this high pressure. Another abnormality is that after the vessel was cooled down to room temperature, the pressure never recovered to its original value (~ 2 bar [29 psi]) but remained relatively constant at ~ 2.16 bar (31.3psi) over a duration of more than 100 h, implying the O-ring still possessed some residual sealing capability.

A post-test inspection of the tested vessel revealed that the interior vessel wall and the thermal couple inserted into the vessel interior were coated with a thin layer of black substance. Although the O-ring was still properly seated in the groove (see Figure 7 [top]), it had turned into a packed layer of powdered char material and could not be removed from the groove without destroying its structural integrity (see Figure 7 [bottom]). Additional tests would be needed to further examine this finding.

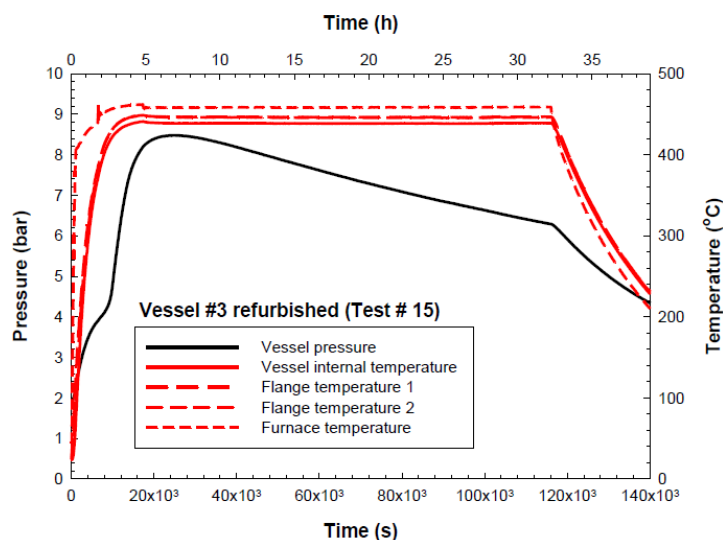


Figure 6: Temporal variations of vessel pressure and temperature in Test #15.



Figure 7: Photograph of Test #15 ethylene-propylene O-ring after heat up showing packed layer of powdery charred material (top) and destroyed seal after the attempt to remove it (bottom)

CONCLUSION

Fifteen metallic and elastomeric seal performance tests including two shakedown tests were performed under beyond-design-basis thermal exposure conditions; twelve tests used metallic seals, two used ethylene-propylene seals, and one used a Teflon seal.

Of the five repeat metallic-seal tests (Tests #2, #3, #4, #8, and #9), leakage (decreasing vessel pressure) was observed in three of the tests (Tests #3, #4, and #8) during the 9 h 800°C (1472°F) exposure. The times when the leakage occurred (the vessel pressure started to decrease) varied in the three tests performed. The two shakedown tests were conducted using a 30 min and 4 h exposure to 800°C (1472°F), respectively, and the seal appeared to hold vessel pressure. No leakage (unchanged vessel pressure within the pressure measurement uncertainty) was also observed in the two metallic seal tests that used 100°C incremental heating from 427°C (800°F) to 627°C (1160°F) and from 427°C (800°F) to 727°C (1340°F), respectively, with at least 9 h exposure to each temperature increment. Three repeat metallic seal tests were also conducted at the seal maximum operating temperature of 427°C (800°F) for 9 h. The seal maintained vessel pressure within the measurement of the pressure transducer in all three tests.

No leakage was observed in one ethylene-propylene seal tested at 300°C (572°F) for more than 20 hours; however, leakage was found in another ethylene-propylene seal tested at 450°C (842°F) for more than 20 hours (Test #15). Leakage was also observed in the test (Test #12) that used a TFE seal subject to 300°C (572°F) exposure.

In these fifteen tests the temperature and time exposure variables were changed to map the performance of the metallic and polymeric seals at elevated temperatures beyond the rated operating temperatures. Both the metallic and polymeric seals were able to hold pressure even after leaks were detected and no catastrophic seal failure (i.e., a rapid release of pressure to atmospheric) occurred. Further testing on the polymeric seals would need to be performed to confirm the results in these tests.

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