HIGH-PRESSURE CRYOGENIC SEALS FOR PRESSURE VESSELS

By: Buggele

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INTRODUCTION

Large high-pressure vessels designed for ambient or high temperatures are not presently capable of containing gaseous helium at equivalent pressures when at cryogenic temperatures. Problems associated with achieving and maintaining high pressures at cryogenic temperatures as they involve seal designs were studied. The purposes of this study were

1. To identify suitable materials and develop a seal for cryogenic applications at pressures to 4080 bars (60 000 psi) (The pressure vessel design limited this portion of the study to 1530 bars (22 500 psi).)

2. To identify some factors that affect seal performance and reliability for cryogenic pressure vessels.

To accomplish this, the seal study was divided into three phases; 51 seal configurations were tested.

METHOD

This seal study used a conventional monoblock pressure vessel made of 304 stainless steel in an annealed condition (fig. 1). The vessel has a single threaded closure containing an O-ring seal with a 304 stainless-steel, wedge-shaped backing ring (fig. 1(a)). The apparatus was supplied with gaseous helium at pressures to 1530 bars, the elastic safety limit. The assembled high-pressure-vessel apparatus (fig. 2) was hung in a nominal 27.94-cm-inside-diameter (11 in.-i.d.) dewar for tests in liquid nitrogen (77.3 K) or liquid helium (4.2 K).

A series of experimental seal systems were designed and tested in this typical pressure vessel that had a representative closure. In phase 1 a common O-ring design of various materials was studied. In phase 2 attempts were made to improve the seal design, to identify reasons for not scaling, and to establish the necessary design philosophy and design criteria for the next generation of seals. Phase 3 was

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used to refine the design and to identify operational idiosyncrasies, limitations, and failure mechanisms for two developed groups of high-pressure cryogenic sealing configurations. Group A consists of those configurations that required high pressures initially at ambient temperature to set the seal and to maintain pressure for later cryogenic operation. Group B consists of those configurations that were capable of sealing without an initial scaling pressure. These seals can be pressurized and depressurized repeatedly at 4.2 and 77.3 K.

APPARATUS AND INSTRUMENTATION

Figures 1 and 2 show the equipment and the numerous components used in conducting this study. The pressure vessel is 21.5 cm by 36.8 cm, with a mass of 106 kg (235 lb). Two platinum resistance thermometers were attached to the pressure vessel at its top and bottom and were used in addition to Chromel-Constantan thermocouples to confirm temperature stabilization. Pressure was monitored by a 0- to 40,000 psi (2720 bar) laboratory calibration-type gage that was located 1 meter from the vessel. This gage was fitted with a potentiometer that provided an analog signal to a strip-chart recorder. For liquid-nitrogen tests, the liquid level was automatically maintained 2.54 cm (1.0 in.) below the top of the vessel. For liquid-helium tests, the levels were controlled manually between the extremes illustrated in figure 1.

A pressure of 1530 bars caused the seal width to increase 0.0137 mm (0.00051 in.) at ambient temperature, 0.0096 mm (0.00038 in.) at liquid-nitrogen temperature, and 0.0074 mm (0.0003 in.) at liquid-helium temperature. A high-pressure cryogenic seal must retain sufficient flexibility to contain the pressure regardless of dimensional changes from either pressure or temperature effects.

Analyses of gaseous-helium samples taken prior to phase 3 testing were within specification [1] which requires a 99.995-percent purity and allows 50-ppm total impurities.

RESULTS AND DISCUSSION

Current design criteria for common pressure-vessel closure systems have been documented [2, 3]. The original closure seal system of the pressure vessel used in
this study (fig. 1(a)) follows typical design practice.

Phase 1

Phase 1 was undertaken to evaluate O-rings made of several common materials. The materials were BUNA-N, a nitrile rubber (70 durometer), silicone rubber (~50 durometer); Kel-F, a fluorocarbon rubber (~90 durometer); and Creavey seals consisting of a Teflon (fluorocarbon resin) tubular torus having a stainless-steel resilient spring inside with a Viton (fluoroclasticomer) core in its center. The high-durometer-number O-rings, or materials with a greater rubber hardness, tended to leak and to distort by plastic flow and had poor pressure-cycle life at ambient temperatures. All the seals in phase 1 failed at or above liquid-nitrogen temperature. The pressures that were achieved in this first phase of tests did not exceed several hundred bars. The unsuitability of the seal design shown in figure 1(a) for cryogenic operation was apparent.

Phase 2

Phase 2 consisted of a series of improved seals. Twelve different types evolved, and the design criteria were established for the next phase. Six partially successful, but mostly unsuccessful, seal configurations that were developed and tested are shown in figures 3. Configurations 3(b), 3(c), and 3(d) showed some promise initially but later proved to be unreliable. Figure 4 illustrates typical examples of seal failure modes revealed during this phase of tests. The Teflon extrusion ring was partially successful in filling the voids created by the shrinking methyl-phenyl RTV silicone rubber [4] extrusion compression ring of seal design 3(a). The primary problem with Teflon was its uncontrollable distortion as shown in figure 4(a). As figures 4(b) and (c) illustrate, decompression breathing blisters (analogous to the biological condition known as the "the bends") caused by gas absorption within the elastomer was a common failure mode for both BUNA-N and silicone rubber O-rings. Gaseous helium diffuses into the compressed O-ring elastomer and causes the blistering when it cannot escape upon rapid depressurization. Figure 4(c) shows typical numerous small fracture blisters just below the surface as well as larger fractures that cut through the entire cross section of the O-ring. The "bends" were not observed for material
samples that were subjected only to high pressure but did occur when the same materials were used as an O-ring in a seal application.

A primary cause for seal leakage and resultant failure at liquid-nitrogen temperatures is thermal shrinkage. Silicone rubber, for example, shrinks 10 times faster than 304 stainless steel. If the coefficients of expansion for 304 stainless steel and silicone rubber are assumed to remain constant from room temperature to liquid-nitrogen temperature, the calculated shrinkage of the pressure-vessel diameter is 0.273 cm less than that of the O-ring's major diameter. This reduces the O-ring's cross section by 0.0094 cm. As the temperature is reduced, the O-ring shrinks, becomes brittle and inflexible, loses compressibility, and is apt to fracture. Pressurization or repressurization of the vessel at cryogenic temperatures is affected (1) by the inability of the O-ring to provide sufficient compression to maintain a seal; (2) by lack of resilience; and (3) by thermal contraction mismatch, which causes a change in the common surface contact, alters the set of the seal, and produces leakage paths. Increasing compression by using oversize-cross-section and oversize-mean-diameter silicone rubber O-rings did tend to improve seal performance. Installation problems causing O-ring twist, excessive squeeze, and compressive loadings result if too large an oversize O-ring is used in an attempt to compensate for the expected shrinkage. Lubricants, if used, caused extensive O-ring rupture.

Figures 3(c) and (f) show seven- and ten-component seal configurations that were attempts to expand on Bridgman's unsupported-area scaling principle [2]. A more effective O-ring seal was also used to provide increased side-wall compressive forces. The double O-rings worked sufficiently well to warrant further investigation despite continuation of O-ring "bends," such as those shown in figure 4(b).

Phase 3A

Phase 3A required ambient-temperature pressurization to stretch and set the double-oversized-cross-section O-rings and to precompress and seal the indium extrusion seal. In phase 3A, a total of 13 six- and seven-part seal configurations were tested (figs. 5(a) and (b)). Teflon extrusion seal-ring material was replaced with indium. Indium's low tensile and compressive strength of 24.8 to 36.2 kg/cm²
Phase 3B concentrated on developing a seal capable of functioning at temperatures between 77.3 K and 1.2 K. Prior to pressurization, with seals 5(a) and 5(b), pressures of only about 50 bars (725 psig) could be achieved due to the limited capacity of the high-pressure compressor. An eight-part seal having a single O-ring (Fig. 5(c)) performed considerably better, but the results were not repeatable. Seals 5(c) and 5(d) used un-O-rings but required four to five times the quantity of helium used with seals 5(a) and 5(b). Test results were not repeatable.

Further testing in phase 3B circumvented the frozen helium condition by raising the pressure-vessel temperature slightly.

Phase 3C

Leakage rates of about 0.0001 m³/min were attained at 1500 bars (20 000 psig) and 77.3 K. Depressurization to below 150 bars (2000 psig) did not increase the seal leakage significantly. Depressurization to about 50 bars (725 psig) may be adequate for cold-seal applications. Indium is a commonly used material for cryogenic vacuum seals. Chung, et al. (9) used indium successfully for a small (28 R-R-m-o-d. 12.7-mm-I-d.) high-pressure cryogenic seal. The primary disadvantage of using indium is that solid helium would be created inside the pressure vessel at these conditions.

Further testing in phase 3B circumvented the frozen helium condition by raising the pressure-vessel temperature slightly. Further testing in phase 3B circumvented the frozen helium condition by raising the pressure-vessel temperature slightly.
5(a) and 5(b). They were only able to achieve 500 to 1300 bars, with a corresponding seal leakage of 40 000 std cm$^3$/min. These seals, besides being costly, required large amounts of gland-nut closure torque in order to generate sufficient compressive force to adequately extrude the indium. These high values of compressive force may be difficult to attain for vessels in the 45-cm-1.4 class. Also, no sustaining compressive force exists to afford continual extrusion to compensate for pressure-vessel expansion upon pressurization, as in the case of the double O-ring supplementary seal.

To be most effective, a seal must not suffer from the "bends" during rapid decompression. Design 5(l), an elastomer-indium combination was unsuccessful. The successful 5(g) seal used two Kel-F O-rings, which have low gas absorption characteristics, combined with a 304 stainless steel convex-wedge-shaped anvil backup ring. A 69-km (500-lb) torque was applied to the gland nut, resulting in a plug-to-body gap of 1.27 mm. This efficient seal compressed the indium more, causing it to extrude further past the convex wedge, thereby increasing the total indium seal surface in contact with the pressure vessel. This seal is shown in figure 6(b).

Pressurization to 1430 bars (21 000 psi) at 77.3 K revealed an average 7,374-std cm$^3$/min seal leakage over a 16-hour pressure decay period. Seal leakage did increase to 283 std cm$^3$/min after four depressurization cycles to less than 3 bars. A leakage of 38 400 std cm$^3$/min occurred on the fifth pressurization cycle at 1326 bars.

The last series of liquid helium, high-pressure-seal tests was performed with a redundant pressurization feed tube to minimize the effect of solid helium or contaminant blockages. The factors that have the greatest effect on the sealing ability of configuration 5(g) when the vessel is submerged in liquid helium have not been established. However, a series of pressure cycles related to the liquid-helium level did indicate that frozen helium or contaminants can contribute to the seal effectiveness. Gasous-helium samples were taken to determine the extent of contamination. The gas supplied ahead of the apparatus contained 619 ppm total impurities. Gasous-helium samples from the inside of the pressurized vessel were taken during high-
pressure, liquid-helium tests. Analysis with a modified analytical mass spectroscopy unit revealed excessive quantities of hydrogen (181 ppm), nitrogen (72 ppm), oxygen (30 ppm), neon (3.6 ppm), argon (1.8 ppm), and carbon dioxide (1.4 ppm), totaling 1929.2 ppm. The repetitive pressurizations caused further concentration of the impurities due to cryotrapping condensables at liquid-helium temperature.

Examples of repetitive pressurizations of configuration 5(g) at approximately liquid-helium temperature are shown in figure 7. To minimize the effects of freezing helium, the liquid-helium level was reduced and maintained just below the pressure vessel for the seal tests shown in figure 7(a), and the liquid-helium supply was stopped to allow warmup for the tests shown in figure 7(b). A series of rapid pressurization-depressurization cycles between 1500 bars and 3 bars were performed at 4.2 K to 145 K. Figure 7(a) shows that seal leakage did tend to increase with the number of cycles.

The 8600-std cm³/min rate for cycle 8 increased to 16,700 std cm³/min for cycle 12 at liquid-helium temperature. A 12,600-std cm³/min leak for cycle 14 at 131 K increased to 14,800 std cm³/min for cycle 16, with failure occurring on cycle 17 at 145 K (fig. 7(b)).

Results of the tests described herein are summarized in table 1.

Upon concluding these operational life tests, inspection of the internal walls of the vessel revealed an oily substance on the upper portion and a brown clear liquid on the bottom portion. Infrared spectroscopic analysis identified the oily substance as a common pump oil, with some rust particles. Most of the brown liquid evaporated before analysis, with rust particles remaining. Careful adjustment of temperature above the 4.2 K level, in spite of the frozen helium and the contaminant phenomena, permitted the successful development of the seals.

CONCLUSIONS

This investigation of the problems associated with reliably containing gaseous helium pressurized to 1530 bars (22,500 psi) between 1.2 K and 150 K led to the following conclusions.
1. Common seal designs used in existing elevated-temperature pressure vessels are unsuitable for high-pressure cryogenic operation.

2. Extrusion seal-ring materials such as Teflon, tin, and lead are not good seal materials for cryogenic high-pressure operation.

3. Several high-pressure cryogenic seal systems suitable for large-pressure-vessel applications were developed; two seals required prepressurization, and one seal functioned repeatedly without any prepressurization. These designs used indium seal rings, brass or 304 stainless-steel anvil rings, and two O-rings of silicone rubber or Kel-F.

REFERENCES

<table>
<thead>
<tr>
<th>Development phase</th>
<th>Description of seal system</th>
<th>Number</th>
<th>Leak performance at cryogenic temperature and 1000 to 15,000 psig¹</th>
<th>Observations and note</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Single O ring (Fig. 1)</td>
<td>1</td>
<td>Unsuccessful</td>
<td>Silicone rubber and PURA E seals subject to blisters and &quot;weeping&quot;. Leak 1 and Gasket seals subject to cold flow distortion.</td>
</tr>
<tr>
<td>2</td>
<td>Three-part to 6-part seal (figs. 3a to 6a)</td>
<td>12</td>
<td>Modular, satisfactory, successful, and uniform. Seals were satisfactory.</td>
<td>Should be used in conjunction with other systems.</td>
</tr>
<tr>
<td>3a</td>
<td>Ruby material, six or nine part seals (Figs. 10 and 23)</td>
<td>11</td>
<td>Leak at the seal, leakage rate: 50 to 100 cm²/min. Increased modulus successful, leakage rate: 1.5 to 50 cm²/min.</td>
<td>Helium extraction ring or rings with double O-rings reliable at 77 °C and 1.7 kbar. Subject to &quot;weeping&quot; below 100 bars (2000 psig) and require ambient pressurization.</td>
</tr>
<tr>
<td>3b</td>
<td>Tin or lead, six or nine-part seal (Figs. 4 and 10)</td>
<td>2</td>
<td>Unsuccessful</td>
<td>Maximum attainable pressure, 1000 bars, but leak and lead failed to extrude sufficiently.</td>
</tr>
<tr>
<td>3c</td>
<td>Six or seven-part seal (Figs. 6 and 7)</td>
<td>9</td>
<td>Unsuccessful</td>
<td>Initial pressurization performed at 77 °C.</td>
</tr>
<tr>
<td>3d</td>
<td>Eight-part seal, one O-ring (Figs. 8 and 10)</td>
<td>5</td>
<td>Successful once</td>
<td>Performance questionable, not confirmed.</td>
</tr>
<tr>
<td>3e</td>
<td>Nine-part seal, no O-ring (Figs. 9 and 10)</td>
<td>2</td>
<td>Successful once</td>
<td>Maximum attainable pressure, 2000 bars, unreliable and needed.</td>
</tr>
<tr>
<td>3f</td>
<td>Seven-part seal, two O-rings (Fig. 11)</td>
<td>3</td>
<td>Unsuccessful</td>
<td></td>
</tr>
<tr>
<td>3g</td>
<td>Eight-part seal, two seals O-rings (Figs. 12 and 13)</td>
<td>1</td>
<td>Successful at 77 °C, 3 kbar.</td>
<td>Results confirmed, seal capable of numerous pressurization-depressurization cycles, slow or rapid dia, 12 L liquid-helium pressurization-depressurization cycles performed six rapid, seal subjected to thermal cycling; rapid cycling at warmer temperature, failed on fifth cycle pressurization at 120 kbar.</td>
</tr>
<tr>
<td>3h</td>
<td>Eight-part seal, two seals O-rings, three helium, two brass and, and one 304 SS wedge ring (Fig. 14)</td>
<td>1</td>
<td>Successful at 77 °C, 3 kbar.</td>
<td>Results confirmed, seal capable of numerous pressurization-depressurization cycles, slow or rapid dia, 12 L liquid-helium pressurization-depressurization cycles performed six rapid, seal subjected to thermal cycling; rapid cycling at warmer temperature, failed on fifth cycle pressurization at 120 kbar.</td>
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</tbody>
</table>

¹Leak rate greater than 500 cm²/min correspond to ambient-class limit of vessel, confirmed by instrumental test.
²Leak rate greater than 500 cm²/min correspond to ambient-class limit of vessel, confirmed by instrumental test.
Figure 1. - High-pressure-cryogenic seal development system.
Figure 4. - Typical examples of seal failure modes.
Figure 3. - High pressure cryogenic seal configurations - phase 3.
Figure 6. - Successful high-pressure cryogenic seals.
Figure 7. - High-pressure cryogenic performance of seal 5 (g).