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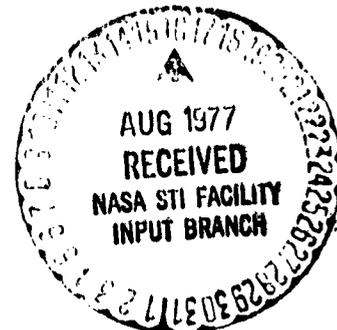
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HIGH-PRESSURE CRYOGENIC SEALS FOR PRESSURE VESSELS .

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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 (11.0-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 sealing, and to establish the necessary design philosophy and design criteria for the next generation of seals. Phase 3 was

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 sealing 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 (0.00054 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); Kal-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 (fluoroelastomer) 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(e) and (f) show seven- and ten-component seal configurations that were attempts to expand on Bridgman's unsupported-area sealing 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 prepressurization to stretch and set the double-oversized-cross-section O-rings and to precompress and seat 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 21.8 to 36.2 kg/cm²

(310 to 515 psi), with a thermal expansion coefficient nearly identical to 304 stainless steel and brass, enabled it to flow plastically and prevented the formation of any gaps by differential thermal contraction. Leakage rates of less than 50.0 std cm³/min (3.05 std in³/min) were attained at 1360 bars (20 000 psi) and 77.3 K. Depressurization to below 136 bars (2000 psi) did increase the seal leakage significantly. Depressurizations to ambient pressure caused seals to fail from O-ring "bends" (fig. 6(a)). A 40 000-std cm³/min leak (the maximum flow rate of the compressor) developed and limited the pressure attainable.

Indium is a commonly used material for cryogenic vacuum seals. Chua, et al. [5] used indium successfully for a small (28.6-mm o. d., 12.7-mm i. d.) high-pressure cryogenic seal. The primary disadvantage of using indium in a design similar to 5(a) for a larger pressure vessel may be its cost. Tests described in this phase prove that indium seal rings could be repeatedly reused, but with a doubling or tripling in leakage rate. Tin and lead were found to be unsuitable substitutes for indium; they do not extrude sufficiently to provide the supplementary sealing needed to achieve a high-pressure cryogenic seal. The seals shown in figures 5(a) and (b) successfully allowed numerous pressure cycles from 204 bars to 1360 bars with various depressurization rates and with no detectable leakages at liquid-helium temperature. A seal with no leakage is not realistic and suggests a blockage due to frozen helium. The equations of state of helium by Spain and others [6-8] indicate 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.

Phase 3B

Phase 3B concentrated on developing a seal capable of functioning at temperatures between 77.3 K and 4.2 K prior to pressurization. With seals 5(a) and 5(b), pressures of only about 50 bars (735 psi) 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(d) and 5(e) used no O-rings but required four to five times the quantity of indium used with seals

5(n) and 5(b). They were only able to achieve 500 to 1300 bars, with a corresponding seal leakage of 40 000 std cm³/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 15-cm-i. d. 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(f), 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-kg-m (500-ft-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 cm³/min seal leakage over a 16-hour pressure decay period. Seal leakage did increase to 283 sec · m³/min after four depressurization cycles to less than 3 bars. A leakage of 38 400 std cm³/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. Gaseous-helium samples were taken to determine the extent of contamination. The gas supplied ahead of the apparatus contained 619-ppm total impurities. Gaseous-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 (1818 ppm), nitrogen (72 ppm), oxygen (30 ppm), neon (3.0 ppm), argon (1.8 ppm), and carbon dioxide (1.4 ppm), totaling 1926.2 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 ~~9660~~ 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 134 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 I.

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 4.2 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.

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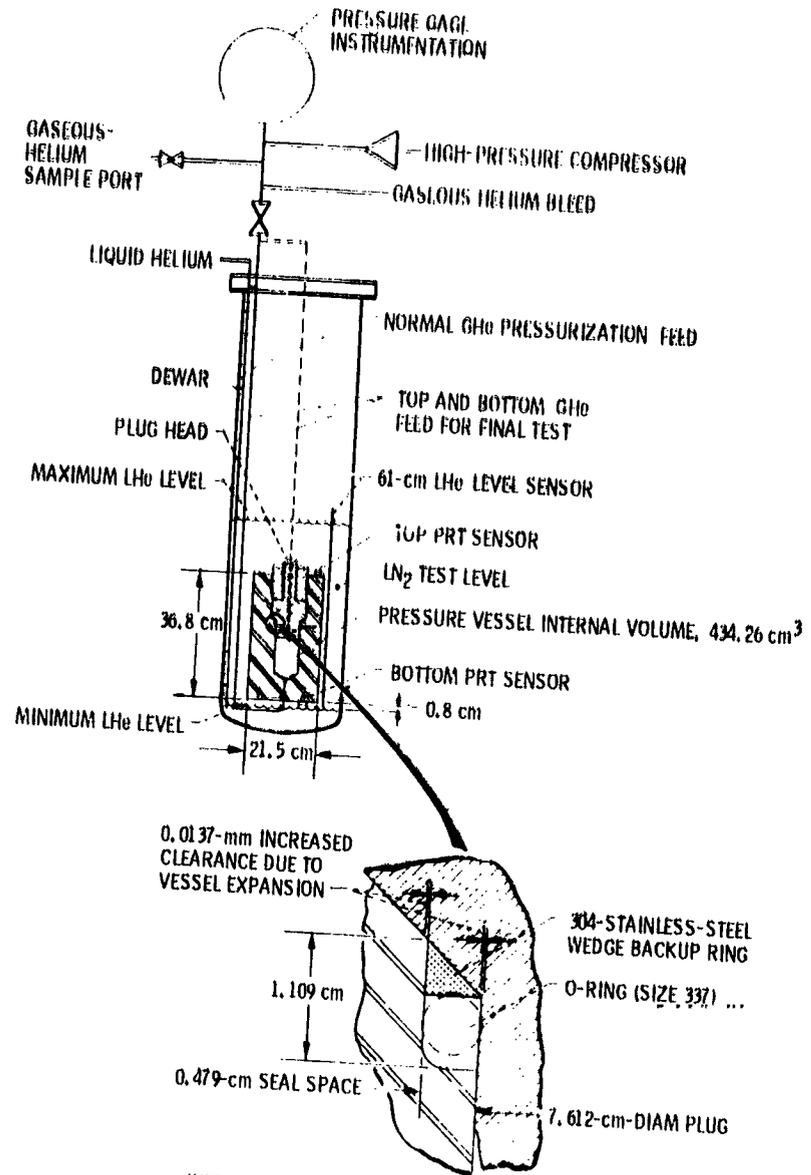
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TABLE I. SUMMARY OF RESULTS FOR IDEAL SYSTEMS TESTED AT CRYOGENIC TEMPERATURES

Development phase	Description of seal system	Number tests ¹	Seal performance at cryogenic temperature and 1300 to 1.30 bars ²	Observations and note
1	Single O-ring (fig. 4)	1	Unsuccessful	Dilucene rubber and PIRA 5 seals subject to blisters and "honds", Kel-F and Crevoxy seals subject to cold flow distortion
2	Three-part to 10-part seals (figs. 3(a) to (f))	12	Seals 3(a), 3(c), and 3(d) successful once, majority unsuccessful	Group unreliable, seal performance not confirmable, Felton distortion not uniform, O-ring blisters and fractures continue
3a	Indium, six- or seven-part seals (figs. 5(a) and (b))	11	Seal indium - successful, leakage rate, ± 60 std cm^3/min , Routed indium - successful, leakage rate, ± 151 std cm^3/min .	Indium extrusion ring or rings with double O-rings reliable at 77.3 and 4.2 K, subject to "honds" below 130 bars (2000 psi); requires ambient prepressurization
	Tin or lead, six- or seven-part seals (figs. 5(a) and (b))	2	Unsuccessful	Maximum attainable pressure, 1088 bars, tin and lead failed to extrude sufficiently
3b	Six- or seven-part seal (figs. 5(a) and (b))	9	Unsuccessful	Initial pressurization performed at 77.3 K
	Eight-part seal, one O-ring (fig. 5(e))	5	Successful once	Performance unreliable, not confirmable
	Nine-part seal, no O-rings (figs. 5(d) and (e))	2	Successful once	Maximum attainable pressure, ^b 1300 bars, unreliable and costly
	Seven-part seal, two O-rings (fig. 5(d))	3	Unsuccessful	
	Eight-part seal; two Kel-F O-rings; three indium, two brass anvil, and one 301 SS wedge ring	1	Successful at 77.3 K	Leakage rate, 7,371 std cm^3/min at liquid-nitrogen temperature; 16-hour decay, 1,4286 to 1,1122 kilobars; four depressurizations performed to 2 to 3 bars, failed on 11th
	2	Successful at 4.2 K	Results confirmable, seal capable of numerous pressurization/depressurization cycles, slow or rapid (fig. 7); 12 liquid-helium pressurization/depressurization cycles performed (six rapid), seal subject to failure during rapid cycling at warmer temperature; failed on 11th rapid pressurization at 116 K	

¹1300 bar (22,800 psi) apparatus pressure corresponds to ambient elastic limit of vessel, confirmed by instrumented test.

²Leakage rate greater than 10,000 std cm^3/min corresponds to maximum capacity of high pressure compression.



INITIAL SEAL CONFIGURATION - PHASE I
 Figure 1. - High-pressure-cryogenic-seal development system.

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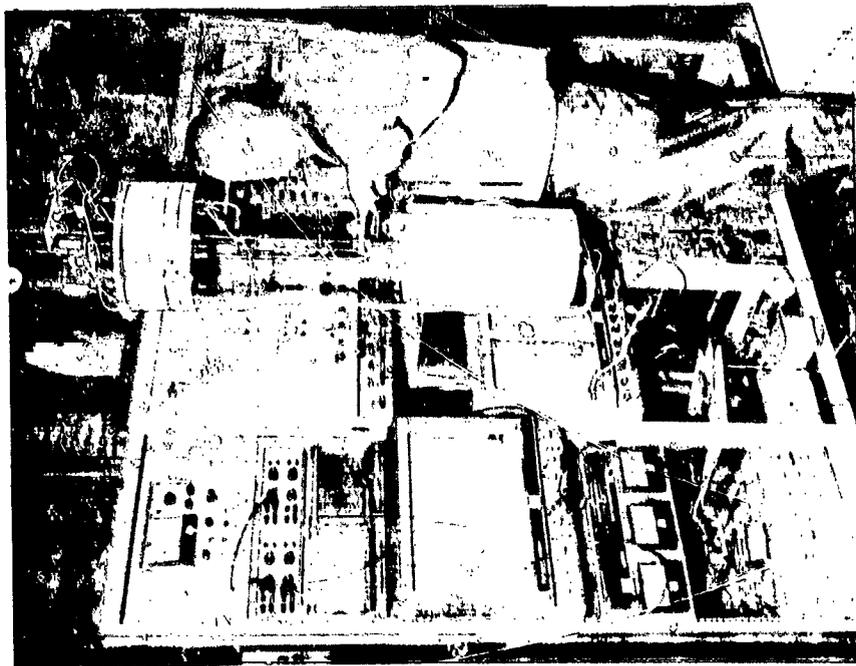
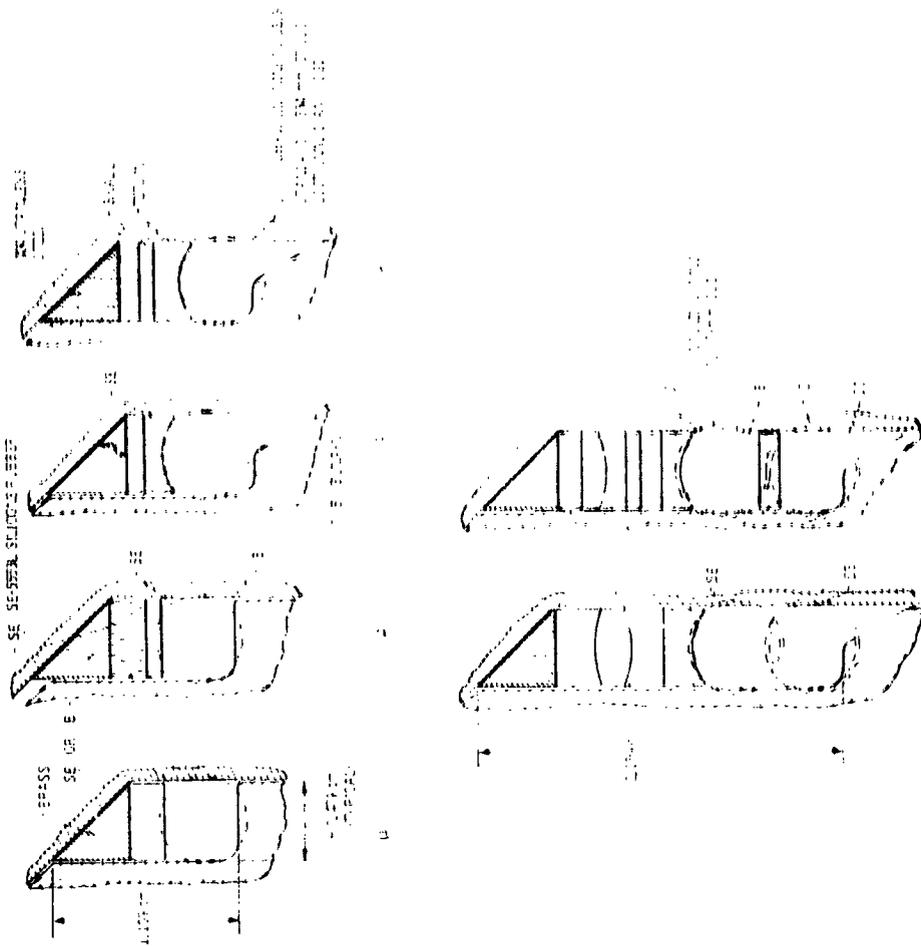


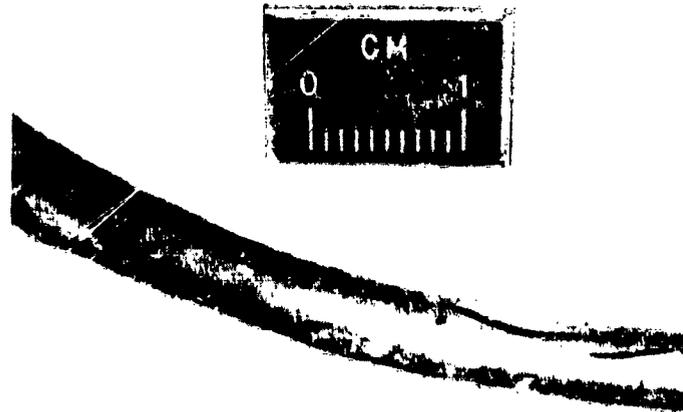
Figure 2 - Pressure vessel, instrumentation, and controls assembly.



(a) NONUNIFORM DISTORTION OF TEFLON EXTRUSION RING.



(b) DECOMPRESSION BLISTERS (BENDS) - COMMON FAILURE OF O-RING ELASTOMERIC.



(c) COMMON SILICONE RUBBER O-RING DETERIORATION NUMEROUS BREATHING BLISTERS AND FRACTURE.

Figure 4. - Typical examples of seal failure modes.

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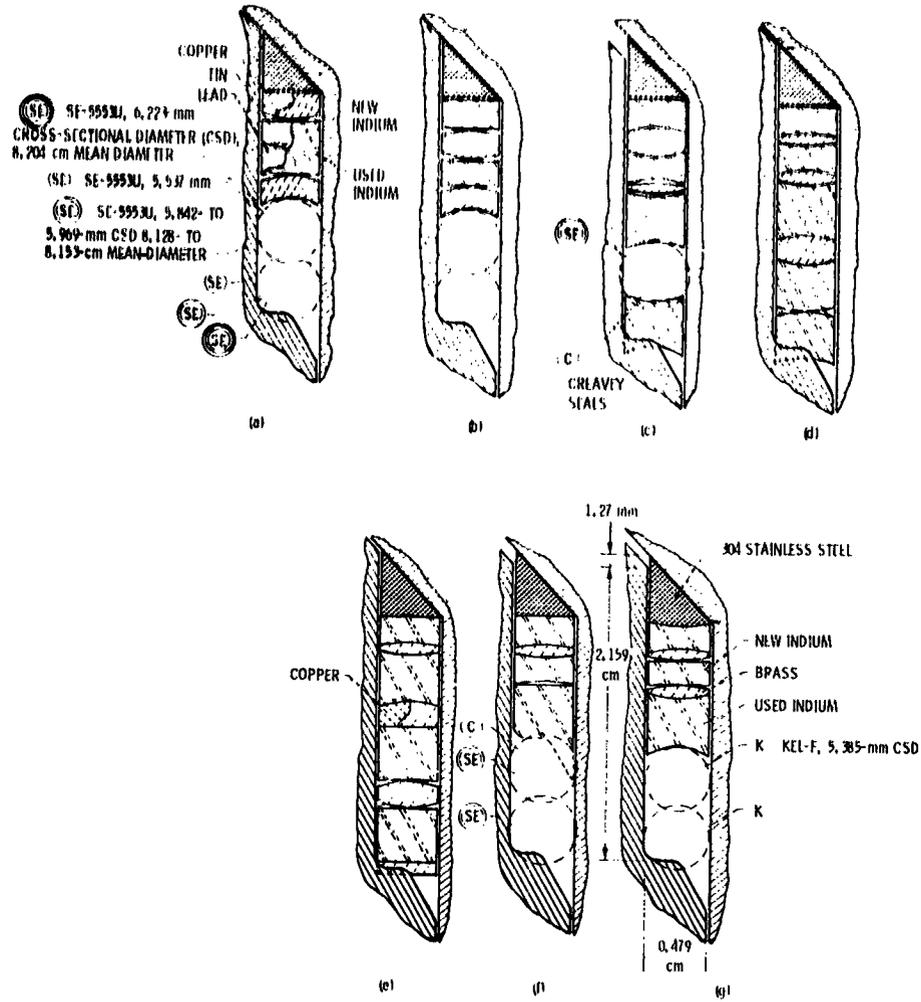
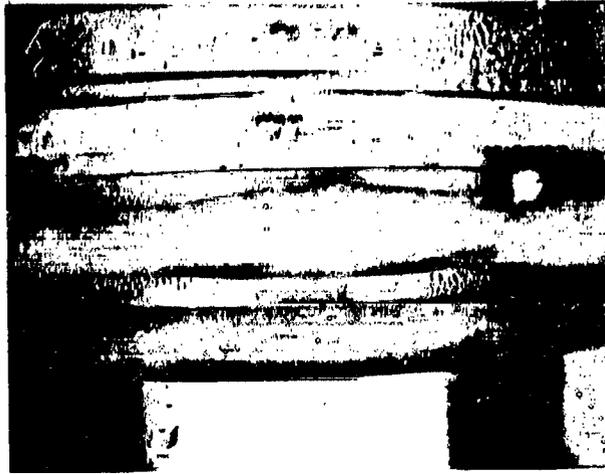
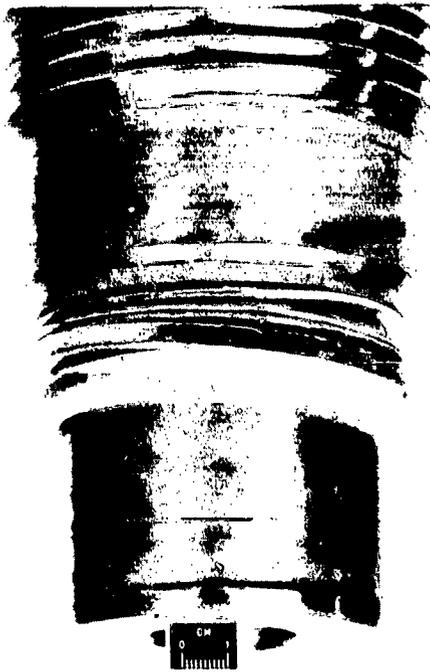


Figure 5. - High-pressure cryogenic seal configurations - phase 3.



(a) SEAL 5 (a) AND TROUBLESOME O-RING "BENDS".



(b) SEAL 5 (g).

Figure 6. - Successful high-pressure cryogenic seals.

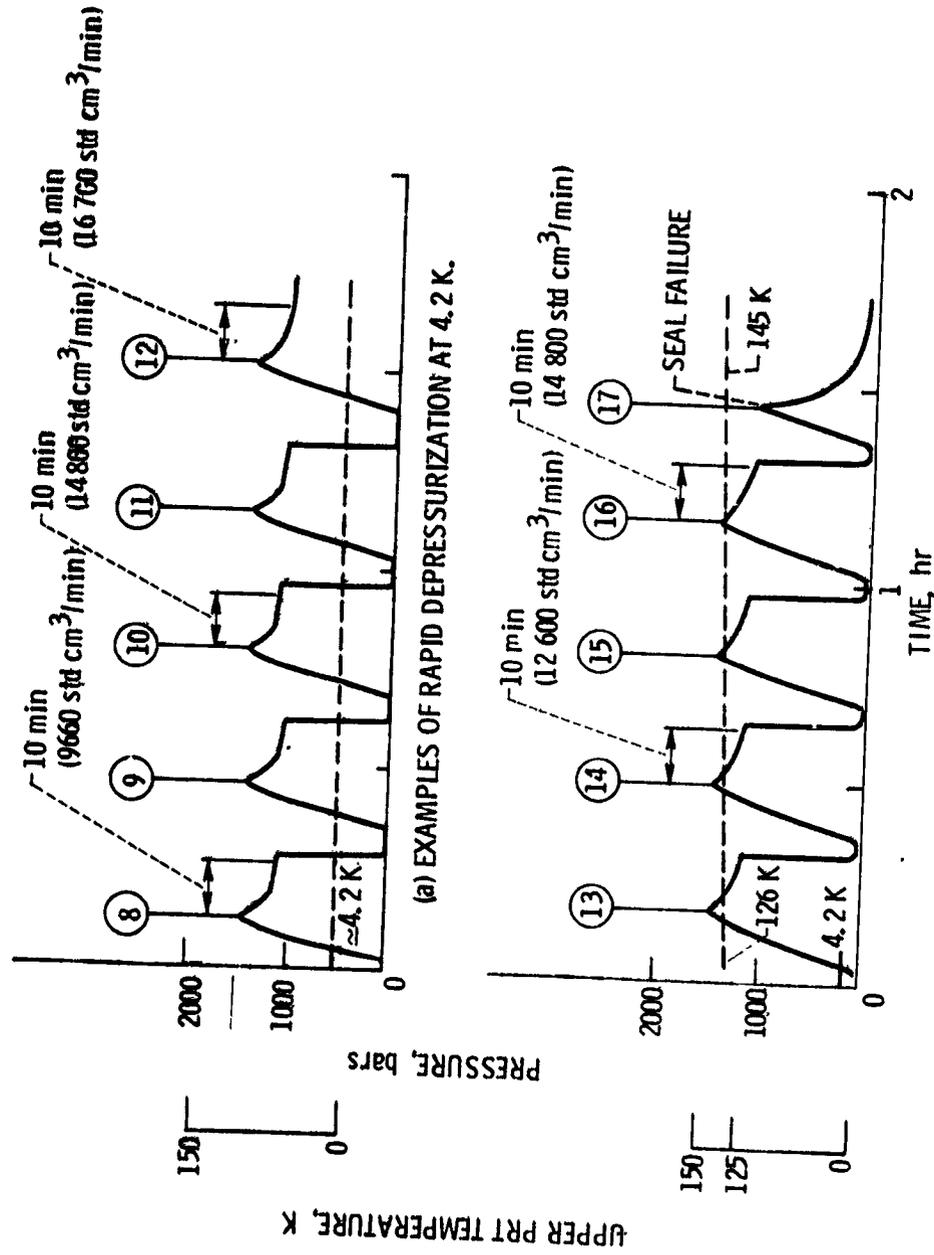


Figure 7. - High-pressure cryogenic performance of seal 5 (g).