NBS REPORT

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Quarterly Report

for

July 30, 1963 to September 30, 1963

on

ELASTOMERIC SEALS AND MATERIALS AT CRYOGENIC TEMPERATURES

by

P. R. Ludtke, D. H. Weitzel, and R. F. Robbins

NBS

U. S. DEPARTMENT OF COMMERCE
NATIONAL BUREAU OF STANDARDS
BOULDER LABORATORIES
Boulder, Colorado

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FOREWORD

This report was prepared by the National Bureau of Standards under USAF Contract No. 33(616)-61-04. The work was initiated under Project No. 7340 "Nonmetallic and Composite Materials", Task No. 734005, "Elastomeric and Compliant Materials". The contract was administered under the direction of the Air Force Materials Laboratory, Research and Technology Division, Air Force Systems Command, with Mr. Roger E. Headrick acting as Project Engineer.

Many of the items compared in this report were commercial items which were not developed or manufactured to meet any Government specification, to withstand the tests to which they were subjected, or to operate as applied during this study. Any failure to meet the objectives of this study is no reflection on any of the commercial items discussed herein or on any manufacturer.

This report covers work conducted from July 30, 1963 to September 30, 1963.

ABSTRACT

A new constant force experiment has been used to study the minimum force required to maintain a seal with five of the better elastomeric O-ring compounds. The most significant result is development of an indium-elastomer "sandwich" arrangement which takes advantage of the favorable properties of both materials and greatly reduces the force required to maintain a seal at cryogenic temperature.

Force decay data are given for three more groups of elastomeric O-rings and some general ideas on seal design are presented.

1. FORCE AND SEAL EVALUATION OF ELASTOMERIC O-RINGS

1.1 Continuation of Initial Screening

Previous work has included all the compounds except those of Group VIII, some of the Lithafrax® filled compounds, and some recently received experimental polymers. Additional results of the force and seal experiment will be discussed in this section.

1.1.1 Compounds of Group VIII

1F4(FBA), ASD compound VIII-21G, was tested at room temperature for material strength. Material failure began when the O-ring was compressed to 0.033 in. thickness with approximately 6100 pounds of force. Severe material failure occurred when the O-ring was compressed to 0.026 in. with 12,500 pounds force. No further testing was done on this compound because of its poor mechanical strength at room temperature.

Hycar® 4021, ASD compound VIII-28D, behaved much the same as EPR. It has a tendency to ooze out from under the compression disc of the test jig. It is very prone to creep or crawl and cannot be confined under the 2 in. diameter top compression disc. No cooldown test was made on this compound.

Hypalon,® ASD compound VIII-8C, was quite hard (90 durometer). 14,000 pounds of force would compress it to only 0.045 in. (68% compression). A cooldown test was made, figure 1. At this thickness the compound had considerable stress relaxation. 41% of the initial force decayed away during the initial relaxation period. However, in spite of the large thickness and excessive force decay, the O-ring leaked at relatively low temperatures: 125, 120, 112 K. For this application a softer compound (60 or 70 durometer) would probably be better.

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"Hycar" is a B. F. Goodrich Trademark
"Hypalon" is a DuPont Trademark

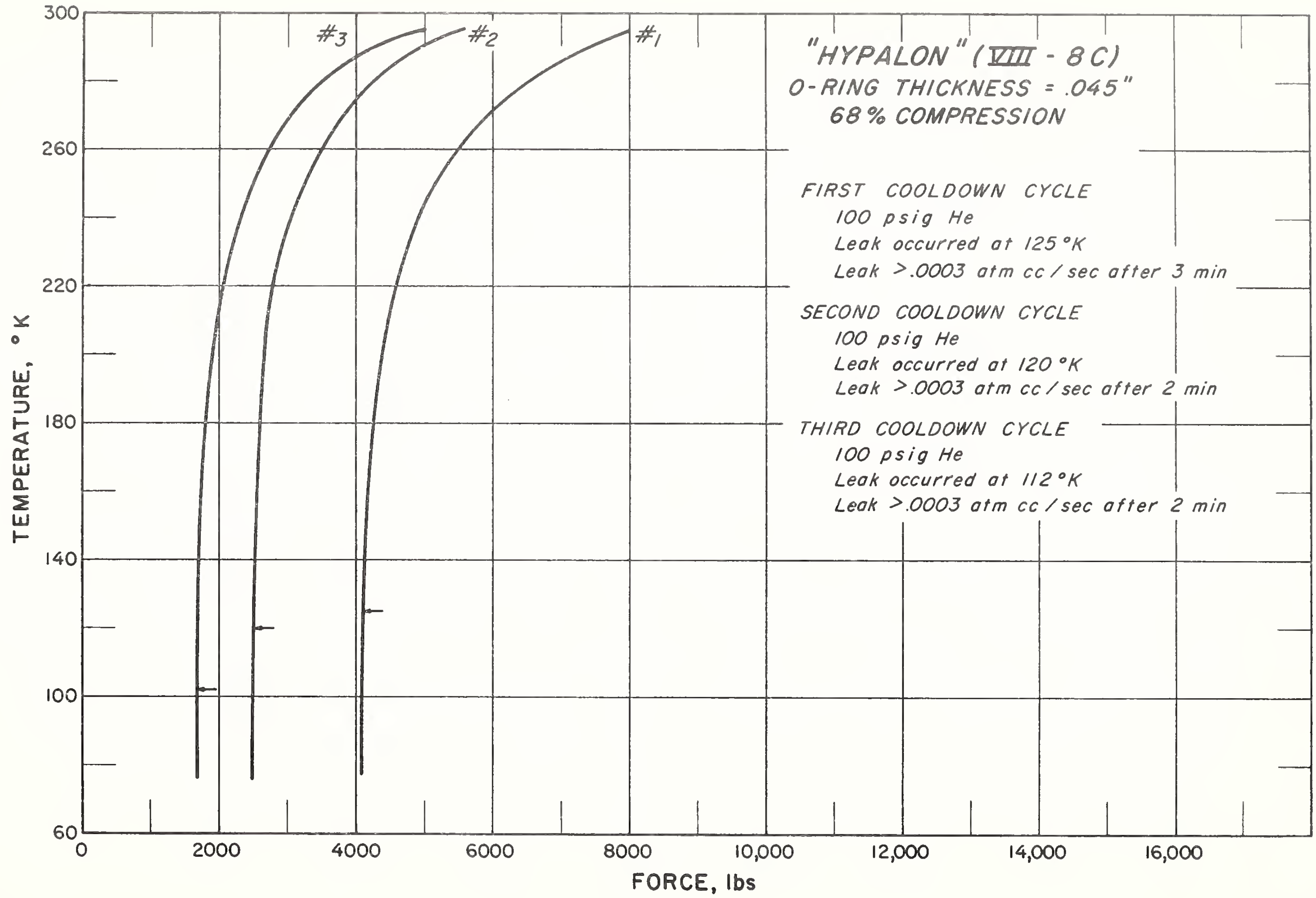

Figure 1 Force-Temperature Curve, "Hypalon"

Vinyl pyridine-acrylonitrile, ASD compound VIII-28C, is a terpolymer of 70 parts butadiene, 10 parts vinyl pyridine, and 20 parts acrylonitrile. This compound was tested twice, figure 2. The Diatron in the leak detector burned out during the first test so a second test was made to check the leakage and stress relaxation. In both tests the force decay was only 30% of the initial force during the initial relaxation period, and not too much after this. During the second test the O-ring did not leak during the first and second cooldowns but did leak during the third cooldown with 1000 psig helium pressure. The vacuum gage readings from the first test indicate the same pattern of leakage, which is very good seal performance in this test apparatus.

Compound VIII-28C, like natural rubber and neoprene, shows quite a bit of compression set (49%) under these test conditions. The stress relaxation of VP-A is similar to natural rubber and polybutadiene. These compounds have 20-30% initial relaxation and then show little more relaxation during successive cooldowns. There was no apparent mechanical failure.

VP-A shows good seal performance as well as favorable stress relaxation and mechanical properties and should definitely be included among the better elastomers for static cryogenic seals. Future testing will continue to include this compound.

1.1.2 Neoprene

We have noticed that seal and force decay results for neoprene were not as favorable as some previous seal tests of neoprene with other flanges. The data were checked and it was found that three different tests were started with neoprene and each showed 40-50% initial force lost, with leaks occurring at high temperatures.

We decided to make another test on a neoprene O-ring to double check the high stress relaxation. An O-ring from a new batch of samples was compressed to .027 inch thickness with 14,300 pounds force. This O-ring showed only 16% initial force loss and only moderate force decay after the first cooldown. This low force loss did not agree with the results from the three previous tests. Further checking revealed that the first force decay samples of neoprene were 85 durometer hardness, while the new batch were 75 durometer. This apparently explains most of the difference in stress relaxation. Harder compounds (as with "Hypalon") usually show higher initial stress relaxation, partly because the compressed O-ring remains thicker for a given applied force.

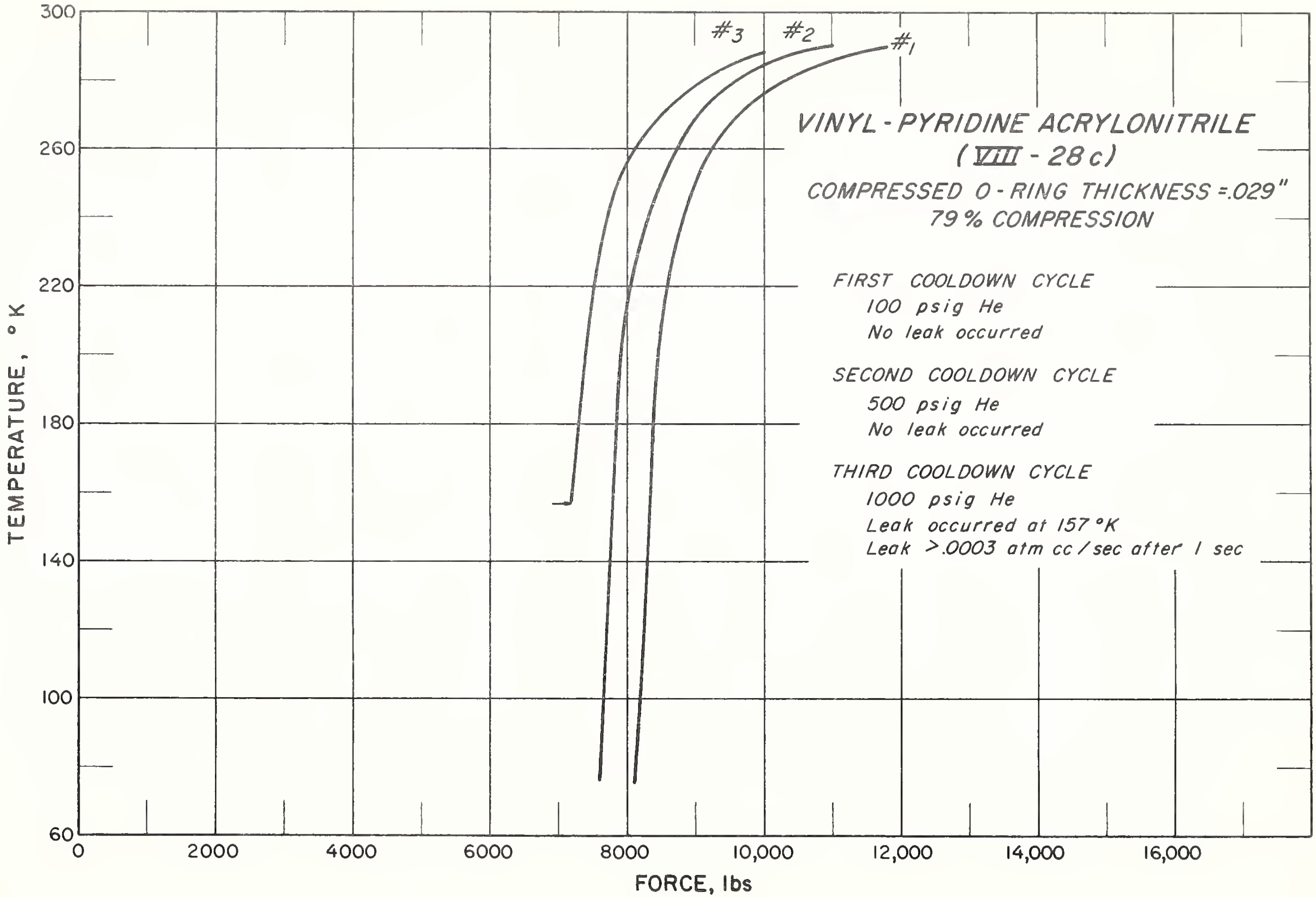

Figure 2 Force-Temperature Curve, Vinyl-Pyridine Acrylonitrile

1.1.3 Experimental Elastomers

Philprene® VP-15, ASD compound 61-20-B, was tested for comparison with the VP-A of group VIII, ASD compound VIII-28C. "Philprene" VP-15 is an 85/15 copolymer of butadiene and 2 methyl-5 vinylpyridine. An O-ring of "Philprene", figure 4, was compressed to 0.024 in. with 13,500 pounds force. The initial force relaxation was low but leaks occurred during every cooldown at fairly high temperatures. The compression set (65%) was about the same as the other compounds of group VIII. "Philprene" was softer (70 durometer) than the VP-A of group VIII (80 durometer) and did not show the good seal performance of the VP-A compound.

A sample sheet of nitroso rubber was obtained. This is an experimental polymer which has no carbon atoms in the main chain or polymer backbone. An O-ring was cut from the 0.080 in. sheet and compressed to 0.015 in. with approximately 2000 pounds force. This O-ring was soft (30 durometer) and tended to ooze and creep very easily. A cooldown test was made. The force at start of cooldown had decayed to almost zero, so no force measurement was possible during cooldown. The O-ring leaked at 169 K, and only one cooldown was made. This soft compound is not suitable for static cryogenic seals, but there was no sign of mechanical failure as with some of the silicones. The compression set was also small (15%). A harder nitroso compound, if available, should be tested.

Three compounds of Dow Corning sil-phenylene elastomer were obtained for evaluation. All the compounds were relatively hard (90 - 100+). Compound E-41-34-1 was tested first. The shore A hardness was 95 durometer. An O-ring was compressed to 0.029 in. with 8200 pounds force. The force at start of cooldown was 5700 pounds and the force after one cooldown was only 2600 pounds. The O-ring leaked at the fairly low temperature of 116 K. The O-ring was removed after one cooldown because of the high stress relaxation. The compression set was high and the surface looked very rough under a microscope. Also, there were small bubbles or strain marks on the surface and inside the compound.

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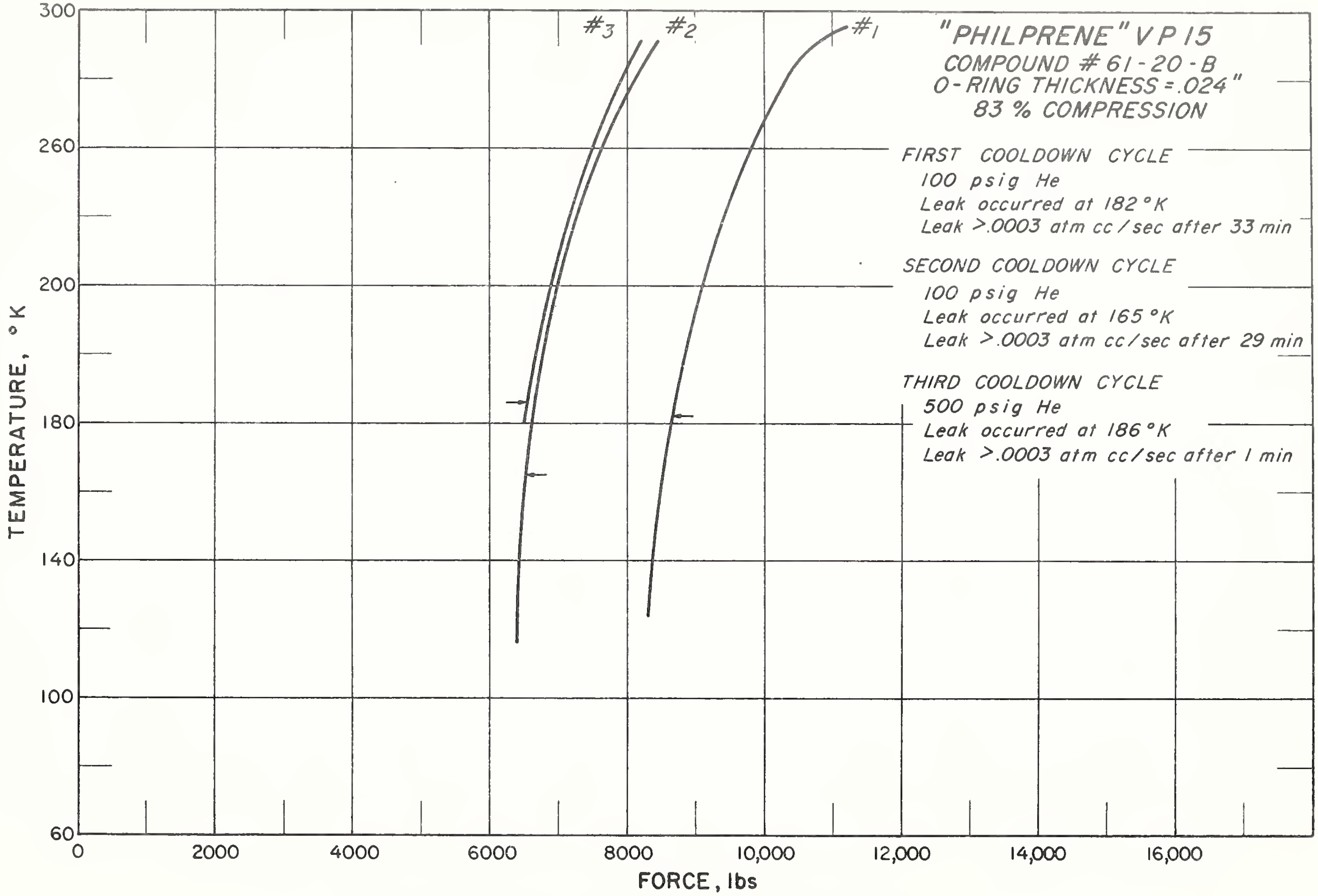

Figure 3 Force-Temperature Curve, "Philprene"

A second sil-phenylene O-ring, ASD compound E-41-34-3 was tested at room temperature. The compound was very hard (100+) and showed the same internal failure marks as the previous sil-phenylene compound. The O-ring required high force for compression and no cooldown was made.

A third compound of sil-phenylene ASD compound E-41-34-2 was tested, figure 4. This compound was the softest (90 durometer) of the three sil-phenylenes and did not have the small bubbles or strain marks when compressed at room temperature.

An O-ring was compressed to 0.025 in. with 12,800 pounds force. The initial force decay was extremely small (8%). Three cooldowns were made with 100, 500, and 1,000 psig helium. No leak occurred during the first two cooldowns but the O-ring finally leaked at 104 K during the last cooldown with 1000 psig helium pressure.

Because the first test looked so good, a second test was made with a little more force and compression, figure 5. The second O-ring was compressed to 0.023 in. with 15,500 pounds force. The initial force decay was average (31%). Again the O-ring did not leak during the first two cooldown cycles but finally leaked during the third cooldown with 1000 psig helium pressure. This compound tends to keep losing force during successive cooldowns, unlike natural rubber and polybutadiene. It also shows considerable force decay during the cooldowns. It seems most promising of the three sil-phenylenes, and does not show the material failure common to other tested silicones under high compression. This appears to be the most promising silicone compound to date and should probably be tested for thermal expansion.

ASD compound 61-19-A, of natural rubber filled with 50 parts lithium-aluminum silicate ("Lithafrax") was tested for comparison with the regular carbon-black filled natural rubber and the previously tested cis-4 polybutadiene compound filled with "Lithafrax". "Lithafrax" is an experimental filler with a very low coefficient of thermal expansion. In previous thermal expansion tests[1] of "Lithafrax" filled polybutadiene, the $\Delta L/L$ from 76 K to 297 K was approximately 25% less than the carbon-black filled compound.

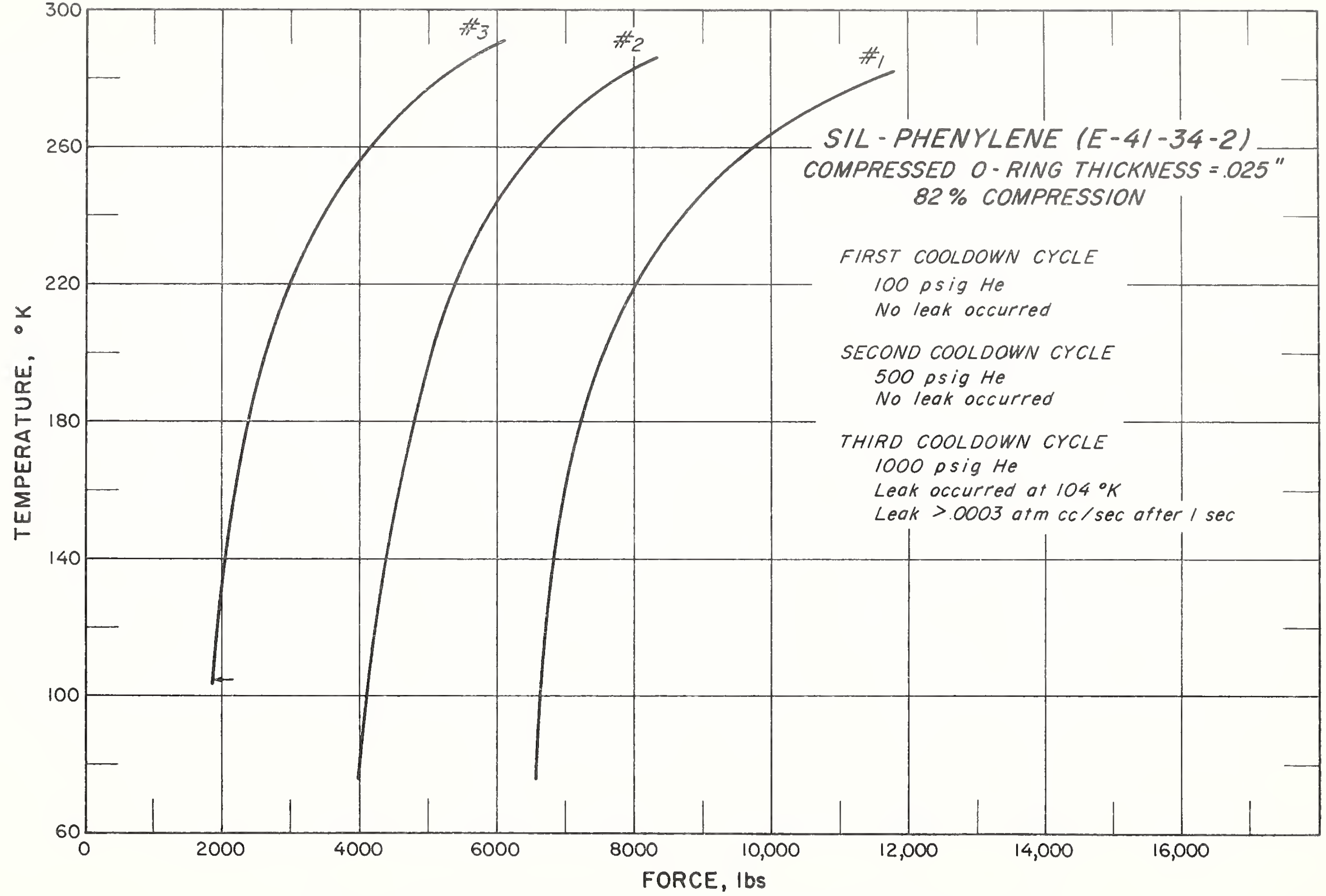

Figure 4 Force-Temperature Curve, Sil-Phenylene (E-41-34-2).

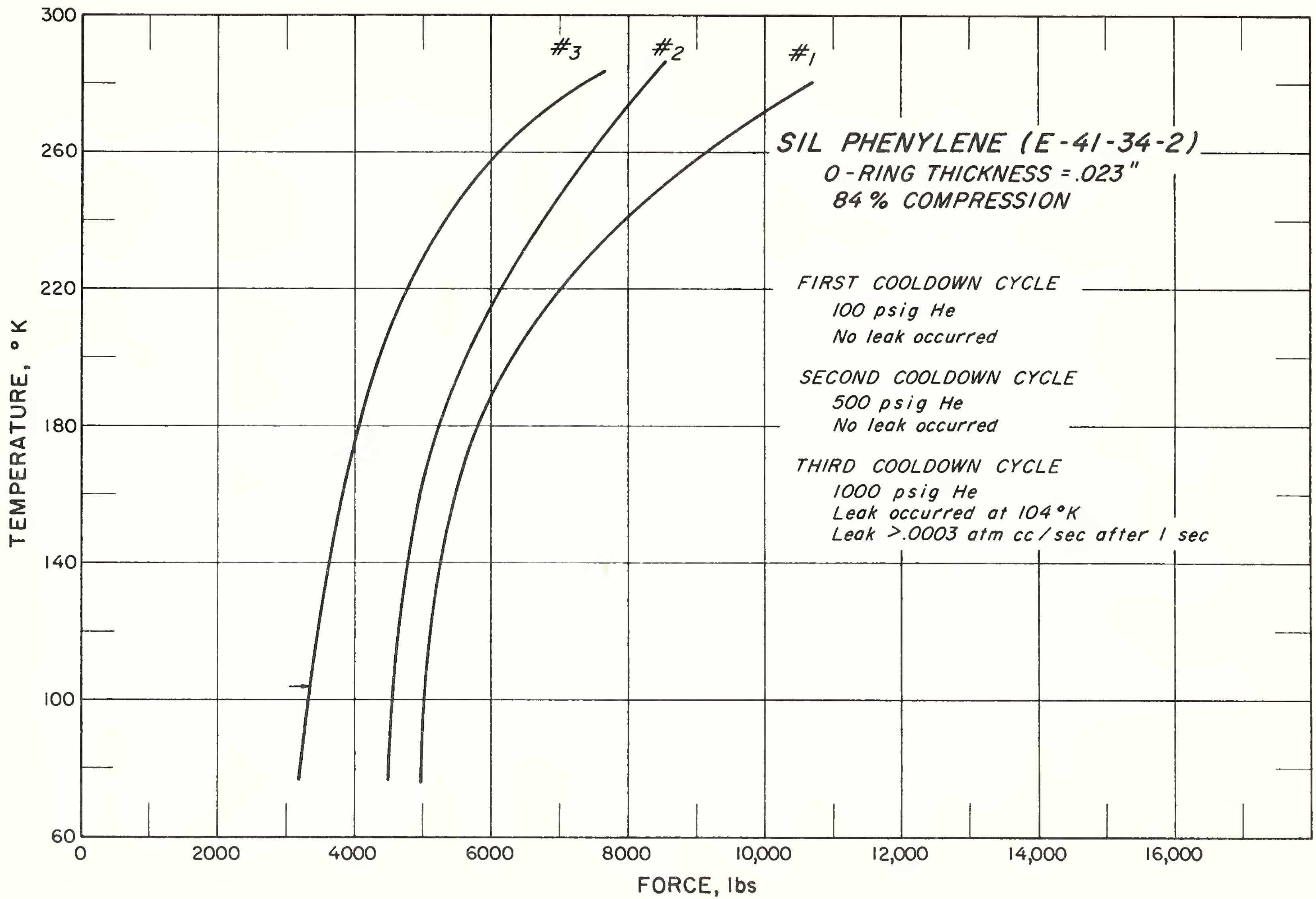

Figure 5 Force-Temperature Curve, Sil-Phenylene (E-41-34-2).

In previous tests with the polybutadiene O-ring it was found that the abrasive nature of "Lithafrax" causes radial scratch marks on the flange faces. This increased surface friction tends to decrease the force decay but does not improve the seal performance of polybutadiene.

A natural rubber O-ring filled with "Lithafrax" was compressed to 0.024 in. thickness with 14,000 pounds force, figure 6. The O-ring leaked during the first cooldown with 100 psig helium pressure. The leak occurred at 88 K and the constant leak rate at 76 K was 80 x 10^{-6} atm cc/sec. There was no leak during the second and third cooldown at 500 and 1000 psig helium respectively. The O-ring was cooled for the fourth time with 1175 psig helium pressure. A leak occurred at 94 K. The compression set was high (71%), which is not unusual for natural rubber in this type test. There was no material failure and the stress relaxation was low. The seal performance was commensurate with that of a regular carbon-black filled O-ring.

From the two tests performed with "Lithafrax" filled polybutadiene and natural rubber it appears that this filler has little or no effect on compression set or material failure of the compounds. "Lithafrax" does not significantly improve the seal performance of either compound (in spite of the apprecaibly decreased thermal expansion), and it has the disadvantage of scratching the flange sealing faces.

2. SEAL INTERFACE PROBLEM

2.1 Introduction

Previous tests with the force decay test jig have shown leaks occurring while there is still considerable force (four to five thousand pounds) on the O-ring sealing surface. This is hard to visualize but evidently small passages are developing at the elastomer-flange interface, allowing small leaks to occur. These leak paths are probably formed by some sort of interface "unseating", most likely due to radial differential contraction.

Various methods have been considered for dealing with this problem. One possibility is an elastomeric O-ring coated with Teflon.® "Teflon" is soft and, more important, has a tendency to flow,

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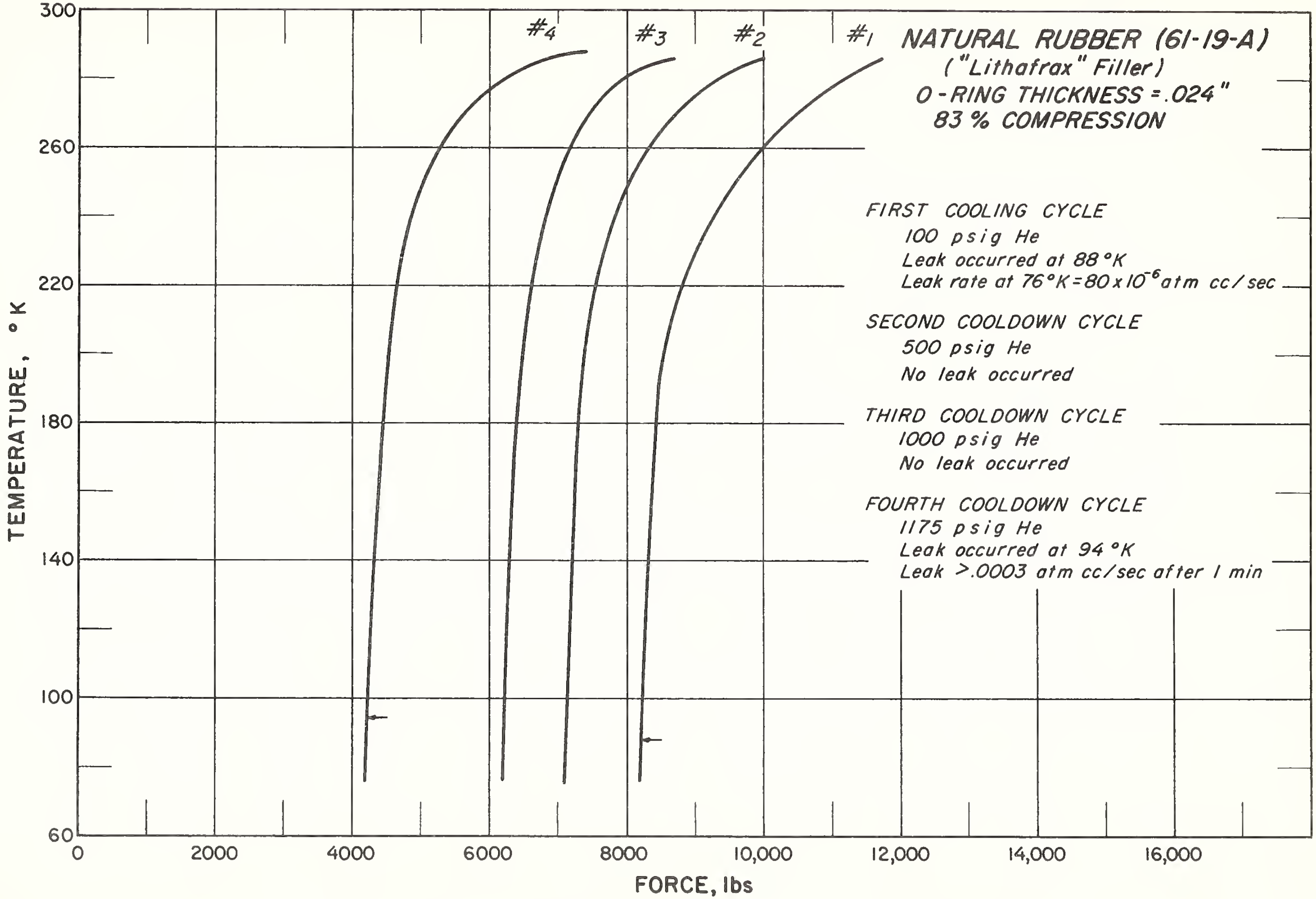

Figure 6 Force-Temperature Curve, Natural Rubber (61-19-A)

even at cryogenic temperatures. A thin coating of "Teflon" might flow at the interface and prevent these leak passages from forming.

Another possibility might be to treat the O-ring surface with the "slippery rubber" process[2] developed by Quantum, Inc., of Wallingford, Connecticut. This is not a "Teflon" coating, but a graft polymerization process wherein the CH groups on the elastomer surface are converted to CF groups. This treatment gives rubber stick-slip characteristics similar to those of "Teflon." The "rub" is absent from the rubber surface, while the elasticity and other bulk characteristics remain essentially unchanged. The final outer layer is 2 to 5 mils thick and might help to seal the small leak passages.

Indium is another possibility for solving the interface problem. Indium is soft and flows readily at cryogenic temperatures. It is used successfully on the large bubble chamber window seals because of it's flow characteristics. It seems reasonable that a thin film between the elastomer and flange surface would fill the interface irregularities, and flow as differential contraction occurs, thus preventing the formation of leak paths.

2.2 New Test Apparatus

To test and possibly solve the interface problem, a test apparatus was developed to do the following:

(a) Determine the amount of constant force required to maintain a seal to 76 K with several of the elastomers judged better for cryogenic sealing.

(b) If it is confirmed that leaks are occurring at high forces, investigate the possibility of remedying this by one or more of the methods mentioned above.

The new test apparatus, shown in figure 7, was designed to test a seal while applying a constant force on the O-ring as it is cooled to 76 K.

A hydraulic Tinius-Olsen testing machine is used to apply a constant force during cooldown. The entire test apparatus is contained inside a dewar for cooling. Inside the dewar is a solid support which is connected by means of a stainless steel post to a heavy base plate which forms the bottom of the dewar. The dewar insulation is evacuated powder; the support post which passes through this insulation is designed to minimize additional heat leak via solid conduction.

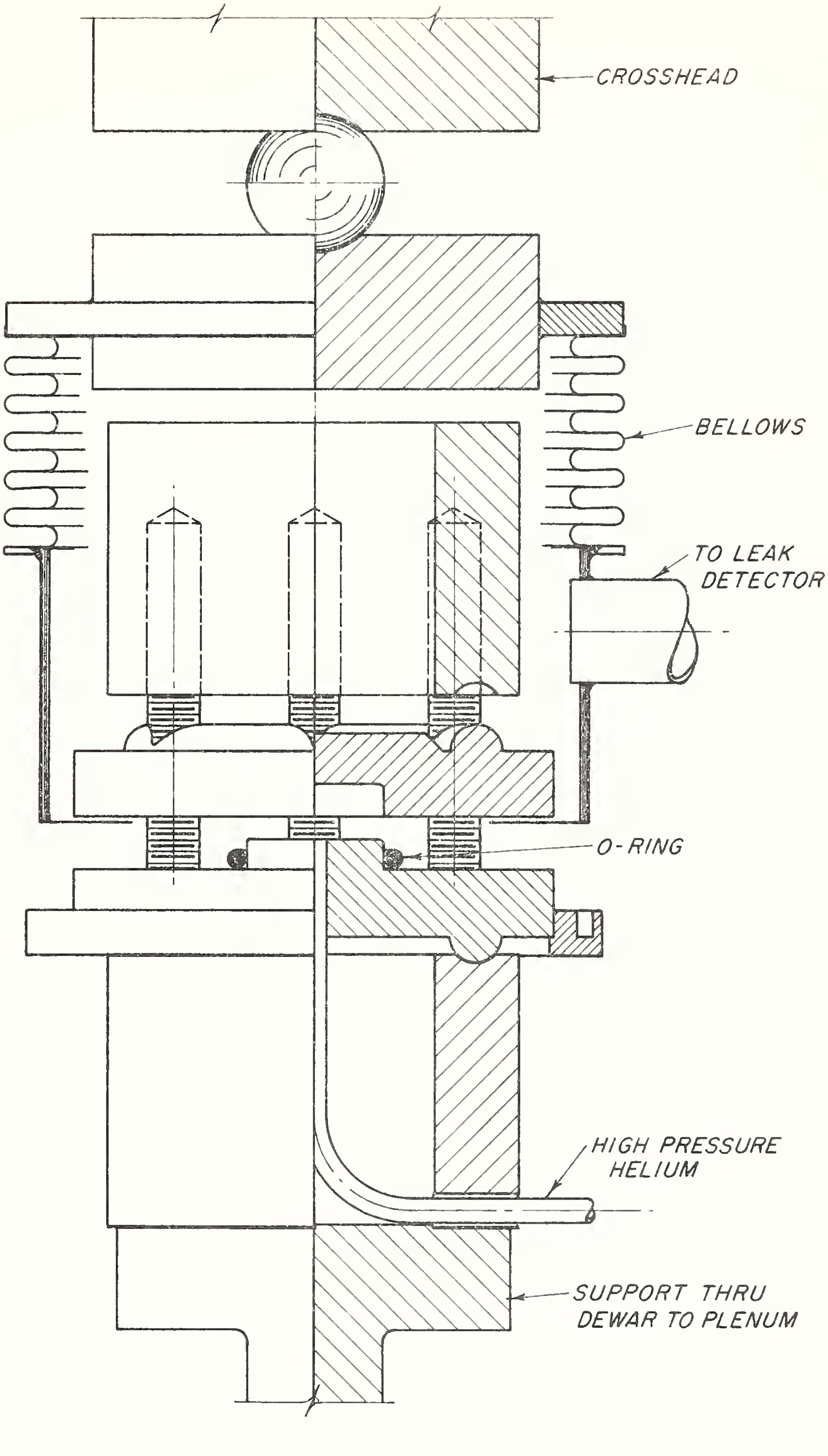

FIGURE 7
CONSTANT FORCE TEST APPARATUS

The test fixture inside the dewar is loaded through rings which apply the force to the flanges at the bolt circle.

An O-ring slips over a 1.000 in. diameter machined step on the bottom flange and a high pressure helium line connects to a hole in the center of the flange. The top flange has a circular recess having a diameter of 1.002 in. which fits down over the machined step. Studs are provided to compress the O-ring when the Tinius-Olsen is not used. The O-ring extrudes radially outward when compressed.

A thermocouple is placed against the under side of the top plate at the bolt circle to record the temperature of the O-ring as it is cooled. The surface finish of the bottom sealing surface is 18 microinches; the finish of the top sealing surface is 15 microinches. These finishes were measured radially with a Taylor-Hobson profilometer. Small spacer tabs of soft solder are placed 120° apart at the bolt circle to record the average minimum compressed thickness of the O-ring.

The vacuum cover incorporates a flexible bellows and heavy top which contacts the upper loading cylinder of the test fixture. A steel ball is placed between the cover top and crosshead to correct any misalignment. The cover is soft soldered to the bottom flange. The space inside the cover is evacuated by a mass spectrometer leak detector which monitors any helium leakage past the O-ring seal.

2.3 Test Procedure

The O-ring to be tested was compressed in the test fixture for one half hour before cooldown started. The applied force was held constant during this initial relaxation period as well as during cooldown. During the initial relaxation period 1000 psig helium pressure was applied to the inside of the O-ring. This gas exerts pressure on $\frac{\pi}{4}\ in^2$ inside the O-ring, and this opposing force must subtracted from the applied force to arrive at the net force on the O-ring.

The 1000 psig helium pressure was held constant during cooldown for all tests except where noted. The applied force was monitored during cooldown and not allowed to vary more than ± 100 pounds. The entire test jig was cooled by slowly adding LN_2 to the compression dewar. Cooling the test jig to 76 K required approximately 90 minutes. Any leak detector reading greater than 10^{-7} atm cc/sec was called a leak.

2.4 Seal Materials Tested

Three groups of O-ring seals were tested.

The Control Group consisted of non-treated, 1/8 in. thick x 1 in. I. D. O-rings made from ASD compounds of natural rubber, neoprene, polybutadiene, "Hycar" 1002 and Viton® A. The recipes for these compounds are given in Section 6.

The second group is the "Slippery Rubber" Group. The O-rings in this group were made from the same compounds as the O-rings of the control group, except they were given the "slippery rubber" treatment as described in section 2.1.

The third group is the Sandwich Group. The "sandwich" seals of this group were made with elastomeric O-rings of the control group and thin washers placed on both sides of the O-ring. A sketch of the sandwich seal configuration is shown in figure 8. The washers were cut from indium, aluminum, "Teflon", and lead, ranging from 1/2 to 7 mils thick.

2.5 Test Results

The test results are shown in table 1. Some clarification of the table headings should be made. The "applied constant force during cooldown" is the force read on the dial of the testing machine. This is not the net force on the O-ring. The "compressed O-ring thickness" is the minimum thickness of the elastomeric O-ring and not the thickness of washers plus O-ring. These values were obtained by averaging the thicknesses of the three soft solder spacer tabs and then subtracting the film thickness (in case of the sandwich seals).

The net force is the force applied by the testing machine plus atmospheric pressure less the force exerted by the helium pressure inside the O-ring. For existing conditions, the net force was equal to the applied force less a correction of 644 pounds. The "net force per linear inch of 1/8 in. O-ring" is the net force divided by the mean circumference of the O-ring.

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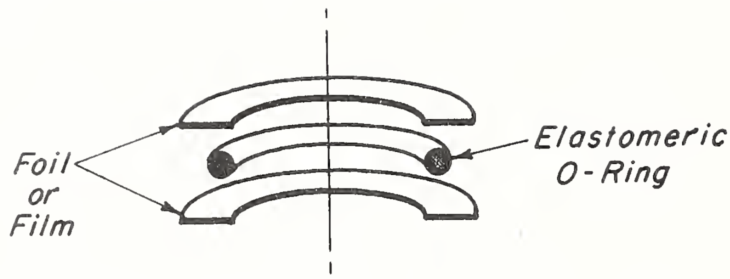

FIGURE 8

SANDWICH SEAL CONFIGURATION

TABLE I
CONSTANT FORCE TEST DATA

Group	Seal Material	Compressed O-Ring Thickness	Net Force per Linear Inch of 1/8" O-Ring (lbs)	Net Pressure on Sealing Surface (lbs/in²)	A. S. D. Compound Number	Shore A Hardness
Control Group	Natural Rubber	.047"	940	2900	IV-8A	62
		.046"	1210	3640		
		.038"	1500	3700		
	Neoprene	.066"	940	4000	IV-8B	80
		.063"	1210	4980		
		.056"	1500	5450		
	Poly-Butadiene	.043"	940	2620	IV-29B	67
		.041"	1210	3240		
	Viton ® A	.045"	1210	3560	I-80	80
		.043"	1500	4200		
		.043"	1780	4960		
	Hycar ® 1002	.050"	1210	3960	II-21B	70
		.048"	1500	4670		
"Slippery Rubber" Group	Natural Rubber	.053"	940	3230	IV-8A	66
		.052"	1210	4120		
	Neoprene	.066"	940	4030	IV-8B	80
		.063"	1210	5000		
	Poly-Butadiene	.060"	940	3660	IV-29B	72
		.058"	1210	4600		
	Hycar ® 1002	.066"	1210	5240	II-21B	76
		.065"	1500	6350		
Sandwich Group	Natural Rubber-Indium	.049"	380	1210	IV-8A	62
		.047"	520	1580		
	Neoprene-Indium	NO TEST			IV-8B	80
		.069"	380	1700		
	Poly-Butadiene-Indium	NO TEST			IV-29B	67
		.045"	520	1520		
	Hycar ®-Indium	.063"	380	1550	II-21B	70
	Viton ® A-Indium	.055"	380	1356	I-8D	80
		.049"	520	1650		
	Hypalon ®-Indium	.081"	240	1260	VIII-8C	90
	Neoprene-Teflon ® (1/2 MIL)	.048"	1210	3800	IV-8B	80
	Neoprene-Teflon ® (3 MIL)	.049"	1210	3880		
	Neoprene-Lead	.063"	1210	5000	IV-8B	80
	Neoprene-Aluminum	NO TEST	10,000 lbs. FORCE WOULD NOT SEAL 100 psig HELIUM AT ROOM TEMP.		IV-8B	80

Legend: Leak; No Leak; Insufficient Force to Confine O-Ring

GENERAL NOTES:

Helium pressure = 1000 psig

® Viton, Teflon, and Hypalon are Du Pont Trade-names

® Hycar is a B. F. Goodrich Trade-name

The "net pressure on sealing surface" is the net force divided by the area of the sealing surface of the compressed O-ring. The area of the sealing surface is easily calculated if no volume compression is assumed, and this assumption was made for the calculation.

2.5.1 Control Group

O-rings of natural rubber, ASD compound IV-8A, were tested with applied constant forces of 4000, 5000 and 6000 pounds. The O-rings tested with 4000 and 5000 pounds force leaked during cooldown. The O-ring tested with 6000 pounds force maintained a seal to 76 K.

O-rings of neoprene, ASD compound IV-8B, were also tested with 4000, 5000, and 6000 pounds force. The O-ring with 4000 pounds force leaked during cooldown and the O-rings tested with 5000 and 6000 pounds force maintained a seal to 76 K.

O-rings of polybutadiene, ASD compound IV-29B, were tested with 4000 and 5000 pounds force. The O-ring tested with 4000 pounds force leaked and the O-ring with 5000 pounds force maintained a seal to 76 K.

O-rings of "Viton" A, ASD compound I-8D, were tested with 5000, 6000 and 7000 pounds force. The O-rings tested with 5000 and 6000 pounds force leaked during cooldown and the O-ring tested with 7000 pounds force maintained a seal to 76 K. "Viton" was included in the control group for comparative purposes because it is a well known fluorocarbon elastomer, and because it has in some test programs been considered LOX compatible(3).

O-rings of "Hycar" 1002, ASD compound II-21B, were tested with 5000 and 6000 pounds force. The O-ring tested with 5000 pounds force leaked during cooldown and the O-ring tested with 6000 pounds force maintained a seal to 76 K.

Some compounds of the group appear to seal a little better than others. Polybutadiene and neoprene maintained seals with the least amount of force. "Viton" is somewhat worse, requiring 7000 pounds force to maintain a seal. Between these are "Hycar" and natural rubber. The most significant result is that none of the O-rings would maintain a seal with 4000 pounds force. This agrees with our force decay test data and substantiates our statement that small leak passages develop along the interface while there is still considerable force on the sealing surface.

Considering the force required to seal the better compounds (all but "Viton" A), it appears that the threshold for maintaining a seal for 1000 psig helium is between 5000 and 6000 pounds constant force for this size of O-ring. This is not initial force, but is the amount of force which must be maintained during cooldown. More initial force would have to be applied to a flange assembly to allow for stress relaxation, creep, etc.

2.5.2 "Slippery Rubber" Group

O-rings of natural rubber, ASD compound IV-8A, with a "slippery" surface treatment were tested with 4000 and 5000 pounds force. The O-ring tested with 4000 pounds force leaked during cooldown and the O-ring tested with 5000 pounds force maintained a seal to 76 K. Apparently the "slippery" treatment reduced the required force from 6000 to 5000 pounds in the case of natural rubber.

O-rings of neoprene, ASD compound IV-8B, with a "slippery" surface treatment were also tested with 4000 and 5000 pounds force. Again, the O-ring tested with 4000 pounds force leaked during cooldown and the O-ring tested with 5000 pounds force maintained a seal to 76 K. The seal performance here was the same as for the untreated material.

O-rings of polybutadiene, ASD compound IV-29B, with a "slippery" surface treatment were tested with 4000 and 5000 pounds force. The O-ring with 4000 pounds force leaked during cooldown and the O-ring with 5000 pounds force maintained a seal to 76 K. Again, the seal performance was the same as for the untreated elastomer.

O-rings of "Hycar" 1002, ASD compound II-21B, with a "slippery" surface treatment were tested with 4000 and 5000 pounds force. The O-ring tested with 4000 pounds force leaked during cooldown and the O-ring tested with 5000 pounds force maintained a seal to 76 K. Here the "slippery" O-ring maintained a seal with 5000 pounds force and the control O-ring did not.

Comparing the performance of the "slippery" compounds to the control group, there were two tests in which the "slippery" compound sealed at 1000 pounds less force than the control compound and in no test did the "slippery" compound require more force to maintain a seal. On the basis of the few tests performed, there appears to be a tendency toward slightly better seal performance with the "slippery rubber" O-rings.

There was no indication of cracking or peeling of the surface due to the "slippery rubber" treatment. Of the four control compounds treated, three were harder after the graft-polymerization treatment. There was no change in the hardness of neoprene. Natural rubber increased from 62 to 66 durometer. Polybutadiene increased from 67 to 72, and "Hycar" 1002 increased from 70 to 76. C. M. Doede of Quantum, Inc., states that this is to be expected and is probably due to at least two factors: "a post cure resulting from the thermal cycle of the process and a possible additional vulcanization caused by the introduction of small amounts of sulfur by certain of the chemical reagents."(2)

2.5.3 Sandwich Group

The sandwich group consists of seals made by placing an elastomeric O-ring from the control group between two thin washers of foil or film. The dimensions of the washers were 1 in. I.D. x 1-5/8 in. O.D.

2.5.3.1 Indium Foil Sandwich

Three mil thick pure indium was the first film tested in a sandwich. All sandwich seals were tested with 1000 psig helium pressure.

An O-ring of natural rubber, ASD compound IV-8A, was placed between two indium washers. The sandwich was placed over the machined step of the test jig the same as a plain O-ring. This sandwich was tested with 2000 pounds force but leaked during cooldown. This is the only indium sandwich seal that leaked during cooldown. Another natural rubber-indium sandwich was tested with 2500 pounds force and a seal was maintained to 76 K.

An O-ring of neoprene, ASD compound IV-8B, was tested between two indium washers in the sandwich configuration. This neoprene compound was considerably harder (80) than the natural rubber (62) and somewhat stiffer. Because of these properties, we thought it might be possible to maintain a seal with only 1500 pounds applied force. A test was started but the helium pressure stretched the O-ring outward beyond the indium washer and a leak occurred, i.e., 1500 pounds was not sufficient force to confine the O-ring. The net force on the O-ring was 1500 - 644 = 856 pounds. This is extremely low for an elastomeric cryogenic seal. An identical neoprene-indium sandwich was tested with 2000 pounds applied force, and maintained a seal to 76 K.

A polybutadiene O-ring, ASD compound IV-29B, was tested between two indium washers. A test was started with 2000 pounds force but this was not sufficient to confine the O-ring. Another polybutadiene-indium sandwich was tested with 2500 pounds force, and maintained a seal to 76 K.

A "Hycar" O-ring, ASD compound II-21B, was tested between indium washers. The "Hycar"-indium sandwich was tested with 2000 pounds force and maintained a seal to 76 K.

A "Viton" A O-ring, ASD compound I-8D, was tested between indium washers. The first sandwich was tested with 2500 pounds force and maintained a seal to 76 K. A second "Viton"-indium sandwich was tested with 2000 pounds force; this sandwich also maintained a seal to 76 K.

A "Hypalon" O-ring, ASD compound VIII-8C, was tested between indium washers. This compound is not one of the control group and has not been one of the better elastomers for cryogenic sealing, but it is hard (90 durometer) and fairly stiff. Because of this, we thought a "Hypalon"-indium sandwich might be capable of maintaining a seal with only 1500 pounds force.

The "Hypalon"-indium sandwich was tested with the usual 1000 psig helium pressure and did maintain a seal to 76 K. The net force on the O-ring was 1500 - 644 = 856 pounds, or 240 pounds per linear inch of O-ring. This test was made primarily to investigate the minimum amount of force required to maintain a seal to 76 K with the elastomer-indium sandwich.

Indium foil seems to be one good solution to the interface problem. Plain indium is a popular cryogenic seal material but it has the one disadvantage of cold flowing easily. This constant flowing tends to open up a seal after a few temperature cycles. One remedy for this flow problem is to keep the gasket so thin that frictional and adhesive forces prevent bulk flow of the indium. But, in order to minimize the flow, the gasket must be so thin that flange tolerances become critical.

With the elastomer-indium sandwich, indium's ability to cold flow is used effectively to maintain the seal. A very thin layer of indium remains compressed between the elastomer and the flange surface and continuously fills any openings which tend to form as the surfaces move relative to one another. In this respect the indium functions as an O-ring lubricant or grease, but one which does not become brittle at cryogenic temperatures.

The elastomer is capable of conforming to seal thickness variations of considerable magnitude. Each time the elastomer warms up to its brittle point, the inherent elasticity comes to life like a compressed spring and reseats the seal interface. There is no continuous flow problem as with plain indium.

The elastomer-indium sandwich is not a high temperature seal. Indium has a melting point of 310 F and most elastomers, too, begin degradation at this temperature. The foil does not tend to separate or split and it is easily removed from the flanges. The indium washer reduces the force required to maintain a cryogenic seal to 1/3 that of the plain O-ring. Indium is soft and expensive but it seals well. For a large seal, arc-shaped sections could be formed and lapped over one another to form a large washer. There would be no need to cut and handle large fragile washers. Indium might also be shaped to shield the inner portion and sides of an O-ring from a non-compatible fluid.

3.5.3.2 Other Foil or Film Materials

Since indium foil is expensive, fragile, and not readily available, the possibility of using other film or foil materials in a similar manner is an obvious consideration. "Teflon", lead, aluminum, copper, tin, gold, and silver, for example, might all be considered as possible substitutes for indium. Sandwiches using the first three of these were immediately investigated using neoprene, ASD compound IV-8B, as the "filling" of the sandwich.

A neoprene-"Teflon" sandwich was tested first. The TFE film thickness was 1/2 mil. With 4000 pounds compressive force, the seal leaked at room temperature. The force was increased to 5000 pounds and the leak stopped. The sandwich was tested with 5000 pounds force and leaked at 240 K shortly after cooldown began. A plain neoprene O-ring with no interface film will give better seal performance than this. Upon disassembly, it was noted that the "Teflon" separated near the center of the sealing surface.

A neoprene-"Teflon" sandwich made with 3 mil film was tested next. With 4000 pounds compressive force, the sandwich leaked at room temperature. The force was increased to 5000 pounds and the leak sealed. The sandwich was tested with 5000 pounds force but leaked at 255 K shortly after cooldown began. There was no separation or tearing of the 3 mil "Teflon" washers, but the seal performance was still not as good as a plain neoprene O-ring.

A neoprene-lead sandwich was tested. The lead washers were 7 mils thick and circumferentially polished to remove any surface contamination. With 3000 pounds force and only 100 psig helium the sandwich leaked at room temperature. With 4000 pounds force and 1000 psig helium the sandwich again leaked at room temperature. 5000 pounds force was required to seal the sandwich at room temperature. A test was made with 5000 pounds force and 1000 psig helium pressure but a leak occurred at 200 K during cooldown. Again, a plain neoprene O-ring makes a better seal.

A neoprene-aluminum sandwich was tested. The aluminum foil was 1/2 mil thick. This sandwich gave extremely poor seal performance. The compressive force was increased to 10,000 pounds but the sandwich would still not seal even 100 psig helium pressure at room temperature. This was the worst of the sandwich seals, undoubtedly because the aluminum was the hardest of the film materials tested.

To summarize the testing of these three film materials, it appears that a plain elastomeric O-ring will maintain a seal better than a sandwich seal using lead, aluminum, or "Teflon." Although these film materials are cheaper and more readily obtainable their performance does not approach that of indium when used with elastomers for high vacuum sandwich seals. Evidently the softness and ductility of indium enable it to flow and maintain a continuous conformable interface in a manner analogous to that of conventional lubricants at ordinary temperatures.

2.5.3.3 Elastomer-Indium Sandwich in Heavy Test Jig

The elastomer-indium sandwich seal was also tested in the heavy (force decay) test jig. No indium foil was on hand for the very first sandwich seal so thin brass discs were "tinned" with a layer of indium metal. These discs were placed on both sides of the O-ring. Neoprene was chosen as an elastomer because this material has been tested in the force decay jig many times and has never maintained a seal to 76 K, even with only 100 psig helium pressure.

The neoprene-indium sandwich was tested in the usual manner; the results are shown in figure 9. The sandwich was given 14,800 pounds initial force. There was more than average force decay during initial flow of the indium. There were three cooldowns at 100, 500, and 1175 psig helium pressure. The seal did not leak during any of these cooldowns. At the end of the last cooldown, while the O-ring was at 76 K, the test jig was removed from the dewar and jarred sharply nine or ten times on the concrete floor as a vibration test. The seal would still not leak. No previous seal has withstood such treatment in the force decay test jig. This was actually our first test of the indium sandwich idea, and led to the testing program which has been described above.

A second test, figure 10, was made with the heavy test jig using 0.003 in. thick indium foil, instead of the indium coated brass plates, and a neoprene O-ring for the sandwich. Three cooldowns were made as before and the test jig was again dropped sharply on the floor as a vibration test. The seal did not leak.

In previous tests with this heavy jig, elastomeric O-rings at 76 K have been sensitive to extreme shock, such as that incurred when the test jig is dropped on the concrete floor from a height of about 4 inches. Indium foil at the interface makes an O-ring much less sensitive to leakage from this type of mechanical shock.

In order to compare the seal performance of the elastomer-indium sandwich with plain indium foil, two 0.003 in. thick indium washers were placed in the test jig, without an O-ring, and tested in the usual manner, figure 11. The plain indium seal was cooled three times with 100, 500 and 1000 psig helium pressure, and given the same strenuous vibration test as the elastomer-indium sandwich.

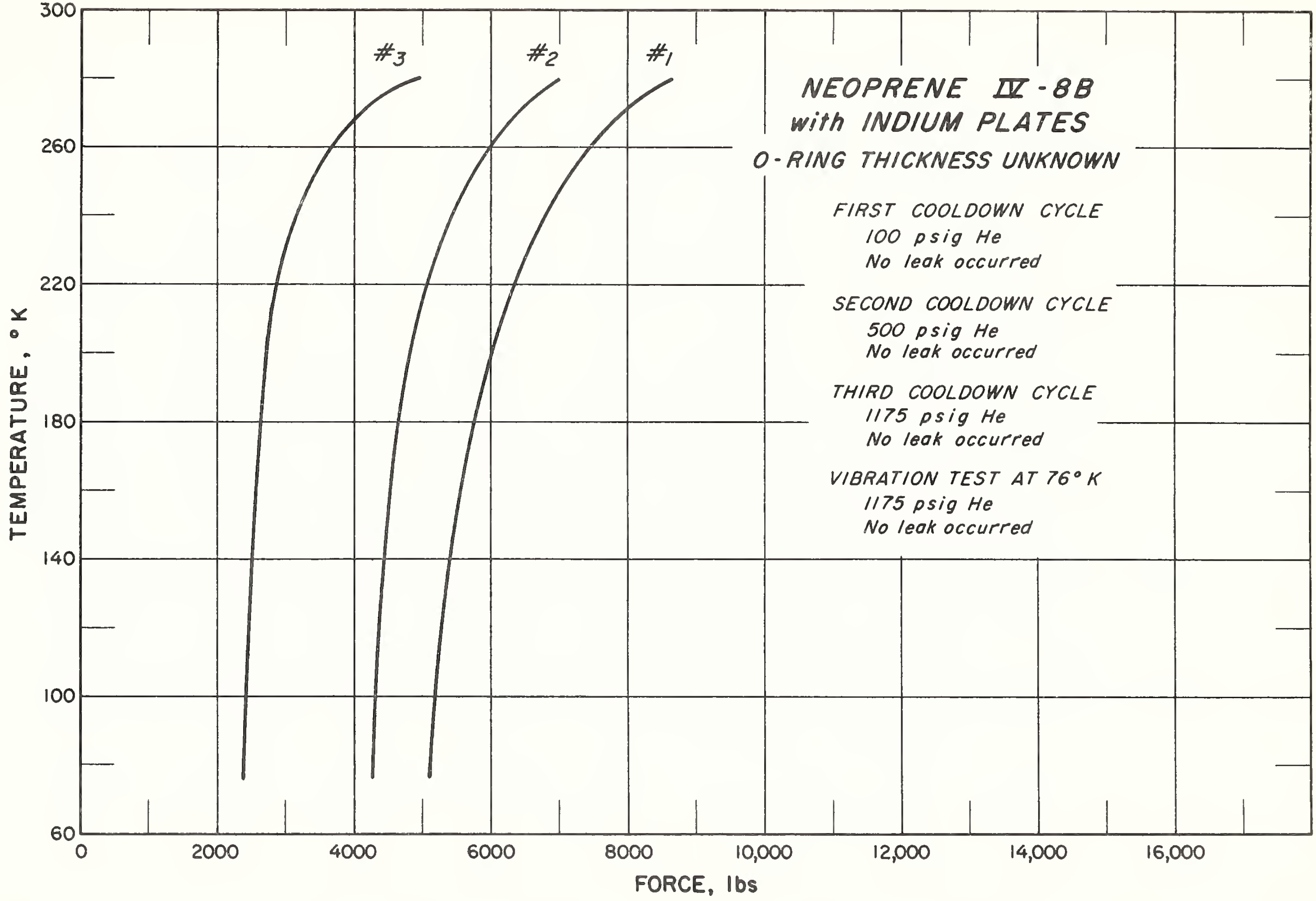

Figure 9 Force-Temperature Curve, Neoprene with Indium Plates

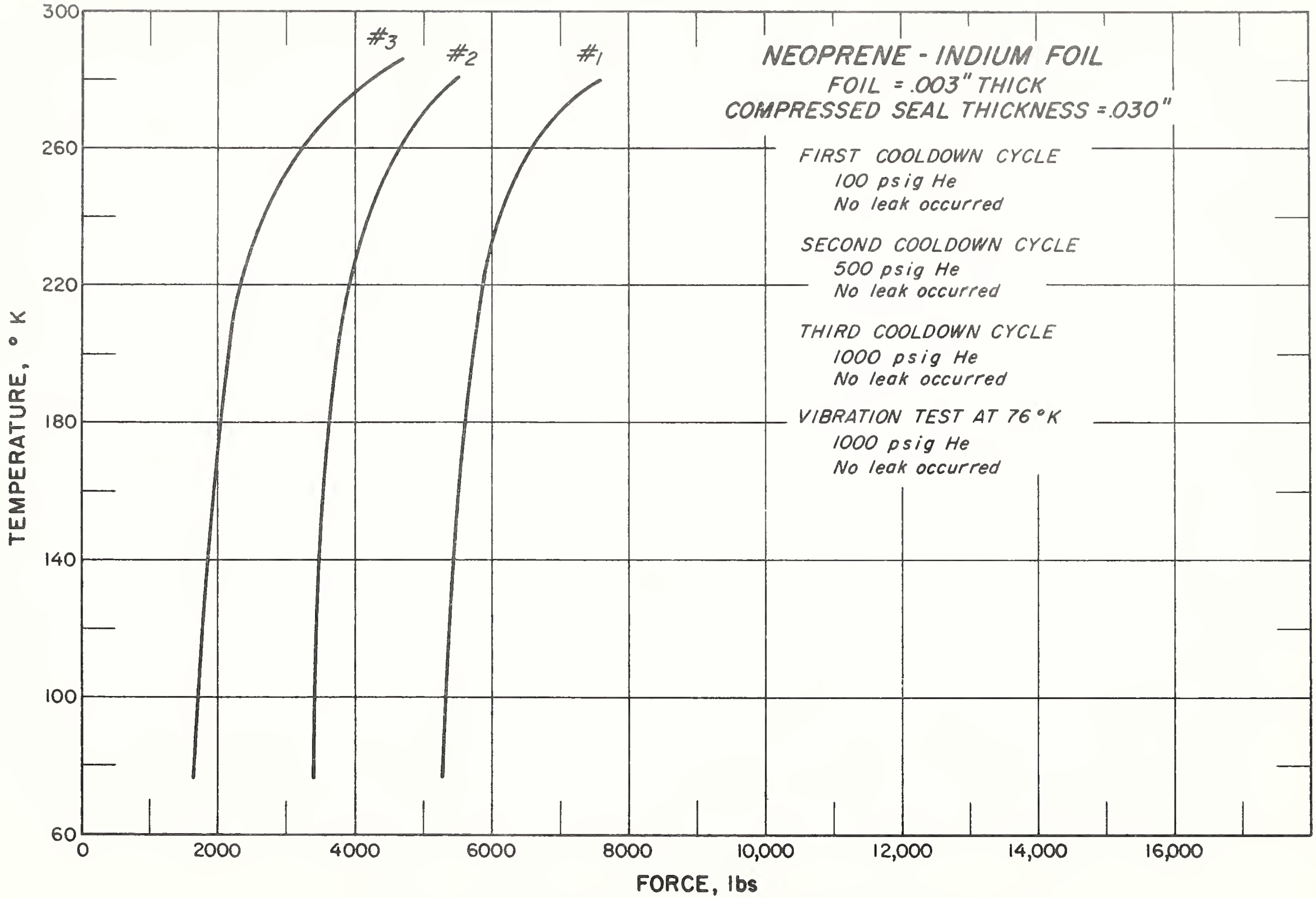

Figure 10 Force-Temperature Curve, Neoprene-Indium Foil

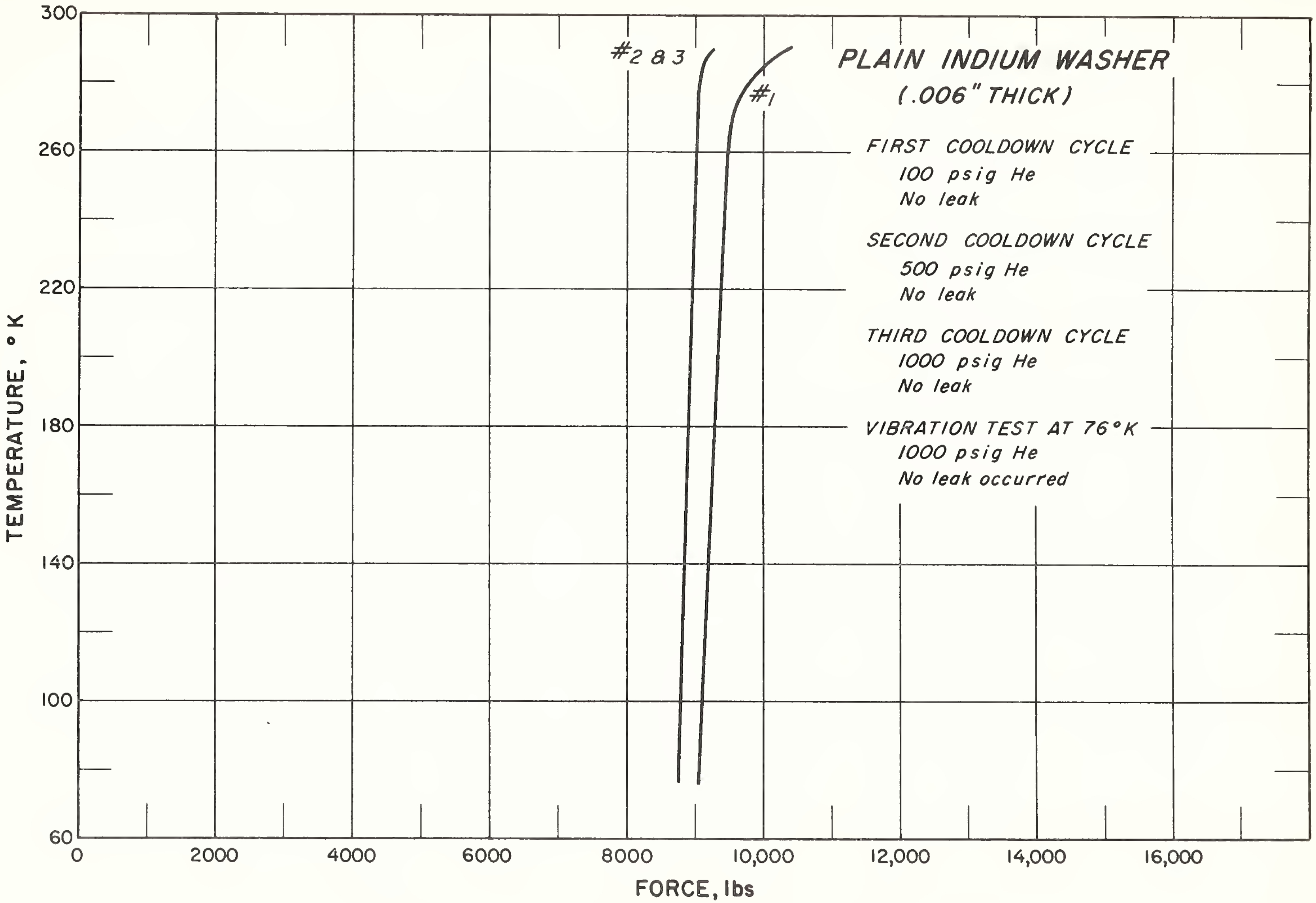

Figure 11 Force-Temperature Curve, Plain Indium Foil

No leak developed. The seal performance was as good as the elastomer-indium seal. However, we feel that this type of plain indium seal has the limitations which have been described in section 2.5.3.1. We plan to make further comparisons between the "sandwich" seal and plain indium, using flanges of larger diameter and flange surfaces which have been given various degrees of irregularity.

3. CONCLUSIONS

Vinyl pyridine-acrylonitrile, ASD compound VIII-28C, has low stress relaxation and excellent seal performance. This compound should be tested under constant force in the new test apparatus for further evaluation, and should be included among the compounds considered better for cryogenic sealing.

"Lithafrax" filler is abrasive and causes small radial scratches on the flange faces. This is not desireable. Thermal expansion tests[1] on "Lithafrax" filled polybutadiene showed a decrease in $\Delta L/L$ from 297 to 76 K of 25%. However, force decay tests on "Lithafrax" filled natural rubber and polybutadiene show no significant improvement in seal performance. More tests should be made, but at present there appears to be no advantage in using "Lithafrax" filler for this application.

The minimum applied force required to seal 1000 psig helium pressure to 76 K with one of the better elastomers appears to be around five or six thousand pounds for this size O-ring (see table 1). This is the minimum force required to maintain a seal and not the <u>initial</u> force. More initial force must be applied to allow for stress relaxation and creep of the elastomer. There should be adequate springloading in a flange assembly to maintain more than this minimum force at any temperature.

Small leak passages do occur at the elastomer-flange interface during cooldown even though there is a force of four or five thousand pounds on the O-ring. The mechanism of this "unseating" is not known but is probably due to radial differential contraction.

The "slippery rubber" treatment does not significantly improve the seal performance of elastomeric O-rings or offer any apparent advantage over the plain non-treated O-rings for a static seal application.

One of the disadvantages of elastomer O-ring seals for cryogenic applications has been the high initial compressive force required. The elastomer-indium sandwich essentially eliminates this adverse feature. The sandwich seal requires only 1/3 as much force as the plain elastomer O-ring. At cryogenic temperatures elastomer-indium sandwich seals are much less sensitive to shock and vibration than the plain elastomer O-ring.

Indium is expensive and not readily available but tests indicate that "Teflon", aluminum and lead (which are available and less expensive) will not do the job. In our tests these did not perform as well as a plain elastomer O-ring.

4. FUTURE WORK

4.1 Force Decay

Preliminary screening of all elastomer groups has been completed. This test jig will be used as we see fit for further work.

4.2 New Large Test Jig

Large diameter cryogenic seals are much more difficult to attain and maintain, especially if the internal pressure is high. A new large test jig is being considered. The test fixture would have medium weight flanges and would be at least three inches in diameter. The jig would be used to compare the seal performance of elastomer-indium sandwich seals with plain indium wire or foil seals and plain elastomeric O-ring seals.

One flange would be machined with a built in warp so that the thickness of a compressed gasket would vary at different points on the circumference. A large fixture such as this, with known irregularity, should make possible a more critical comparison between the elastomer-indium sandwich (for example) and other types of seals. As more refinements in the use of elastomeric O-rings are worked out, it becomes increasingly important to provide convincing evidence that these simple devices are not laboratory curiosities, but are concepts capable of solving most of the more difficult cryogenic static seal problems.

4.3 LOX Compatibility

Four of the better compounds have been coated with TFE "Teflon." The coating was not successful on all of the compounds, but tests of seal performance and coating behavior at cryogenic temperature will be conducted.

It is possible that elastomers could be made acceptable for LOX systems by using indium as a shield between the oxygen and the O-ring. This might be combined with the sandwich idea by coating or wrapping indium around part of the O-ring. Other methods of placing a "Teflon" or indium barrier between the elastomer and the confined fluid can be visualized; some of these configurations should be given careful thought and possibly tested.

4.4 New Seal and Flange Designs

It was shown in table 1 and section 2.5.3.1 that the elastomer-indium "sandwich" seal is capable of sealing 1000 psig helium gas at cryogenic temperatures with flange loads so low that gasket blowout becomes a problem. The flange of the test fixture was designed with an "internal" step, i.e., the O-ring was initially placed around the O. D. of the step, as shown in figure 12A. This configuration provides back-up support for pressure outside the O-ring as, for example, a flange on an evacuated vessel. When the pressure inside the sealed vessel exceeds that on the outside, it is better design to place the O-ring on the I. D. of the step, as shown in figure 12B. If the elastomer-indium "sandwich" seal tests had been carried out with the back up step outside the O-ring there is little doubt that a seal could have been maintained to 76 K with even lower flange loading.

Figure 12B is in general better design than figure 12A, so far as control of pressure is concerned, because pressure inside the seal will usually exceed that on the outside. Evacuated vessels are an exception, but in this case the pressure outside the seal will normally exceed that on the inside by only one atmosphere, a pressure differential which is not great enough to constitute a "blowin" hazard even with lightly loaded gaskets.

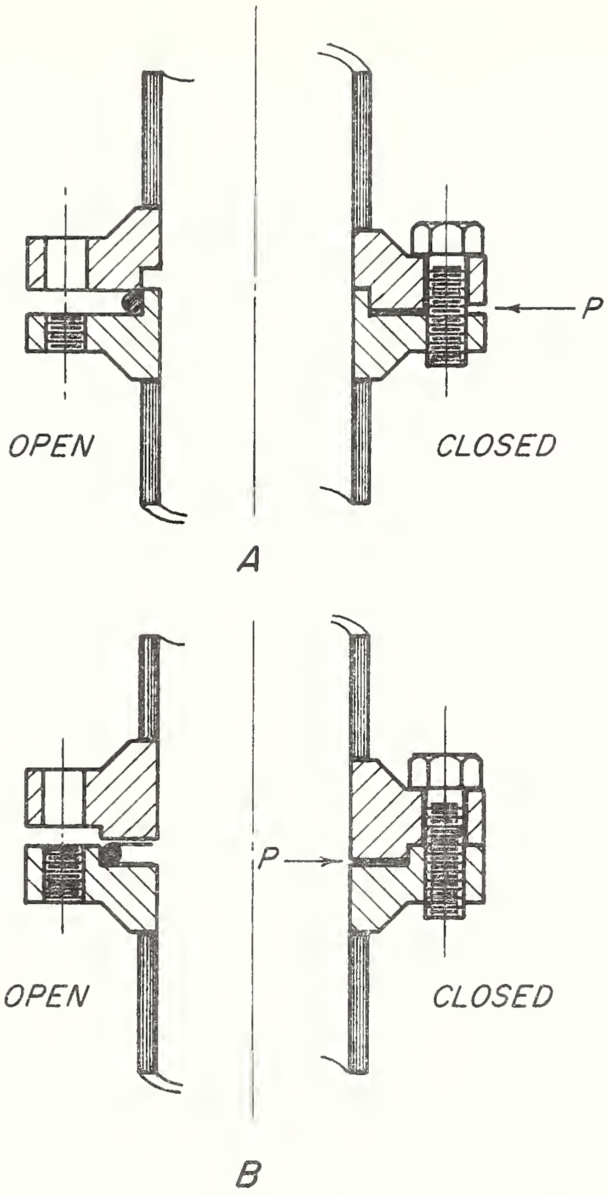

FIGURE 12: STEPPED FLANGES

The "internal" step of figure 12A has been favored because of the convenience it offers for positioning and holding the O-ring before compression. This is an advantage worth considering, especially for seals of large diameter, but an "external" step is much more logical for confinement of high pressure fluids.

The simple stepped flanges shown in figure 12 obviously are not end products of careful analytical design. Shaped flanges and inserts offer the seal designer many additional possibilities which are not immediately apparent. Among these are the control of bolt loads, optimum spring loading, pressure actuation, temperature actuation, and elimination of extraneous load transmission through the O-ring. It will be shown in our next report that these and other advantages can be achieved with elastomeric O-rings using flanges which are both simple in shape and light in weight.

5. REFERENCES

1. Weitzel, D. H., Robbins, R. F., Ludtke, P. R., Ohori, Y., Elastomeric Seals and Materials at Cryogenic Temperatures, ASD-TDR-62-31, Pt. II (May 1963).

2. Seventh Joint Army-Navy-Air Force Conference on Elastomer Research and Development. Vol. 1, p. 153 (1962).

3. Curry, J. E., Riehl, W. A., Compatibility of Engineering Materials With Liquid Oxygen. NASA Internal Report MTP-M-S&M-M-61-7 (1961).

6. COMPOUNDING RECIPES OF ASD MATERIALS

ASD No.	Polymer	Estimated Monomer Ratio	Recipe		Hardness (Shore A)
I-8D	Vinylidene Fluoride & Perfluoropropylene (Viton A, DuPont)	70/30	Polymer	100	80
			Magnesium Oxide	20	
			Hexamethylene Diamine Carbamate	1.3	
			M T Carbon Black	25	
			Cure 20 min at 280 F		
			Post cure 16 hr at 400 F		
II-21B	Butadiene & Acrylonitrile (Paracril 18-80, Naugatuck Chem. Co.)	70/30	Polymer	100	70
			Zinc Oxide	5	
			Benzothiazyl Disulfide	1.5	
			Stearic Acid	1.5	
			Sulfur	1.5	
			FEF Black	50	
			Cure 20 min at 310 F		
IV-8A	Natural Rubber (Smoked Sheet)		Polymer	100	62
			Stearic Acid	3	
			Zinc Oxide	5	
			N-Cyclohexyl-2-Benzothiazole Sulfonamide	.6	
			Sulfur	2.75	
			HAF Black	50	
			Polymerized trimethyldi hydroquinoline (Resin D)	1	
			Cure 15 min at 310 F		

ASD No.	Polymer	Estimated Monomer Ratio	Recipe		Hardness (Shore A)
IV-8B	Chloroprene (Neoprene , WRT, Du Pont)		Polymer	100	80
			Stearic Acid	5	
			Zinc Oxide	5	
			Magnesium Oxide	4	
			HMF Black	50	
			Na 22	. 5	
			Cure 20 min at 310 F		
IV-29B	Cis 4 Polybutadiene		Polymer	100	67
			Zinc Oxide	5	
			Stearic Acid	. 5	
			Sulfur	2. 5	
			HAF Black	50	
			Tetra Methyl Thiuram Disulfide	. 4	
			Cure 30 min at 310 F		
VIII-21G	1, I-dihydroper-fluorobutyl acrylate [1F4(FBA)]		Polymer	100	75
			Sulfur	1	
			FEF Black	40	
			Paraffin	1	
			Triethylene Tetramine	1. 25	
			Cure 30 min at 310 F		
VIII-28C	2-Methyl 5-Vinyl pyridine-acrylonitrile-butadiene (VP-A)	10/20/70	Polymer	100	85
			MT Black	150	
			Zinc Oxide	5	
			Tetra Methyl Thiuram Disulfide	3	
			HAF Black	2	
			Cure 45 min at 310 F		

ASD No.	Polymer	Estimated Monomer Ratio	Recipe		Hardness (Shore A)
VIII-28D	Ethyl-acrylate ("Hycar" 4021, B. F. Goodrich Chem Co.)		Polymer	100	85
			Stearic Acid	1	
			Sulfur	.5	
			FEF Black	50	
			Trimine Base	3	
			Cure 45 min at 310 F		
VIII-8C	Chlorosulfonated Polyethylene ("Hypalon" 5-20, Du Pont)		Polymer	100	90
			Magnesium Oxide	3	
			Hexamethylene Diamine Carbomate	1	
			Pentaerythritol	5	
			Magcarb W	10	
			HMF Carbon Black	5	
			Cure 25 min at 310 F		
61-20-B	2 Methyl, 5-Vinyl pyridine acrylonitrile-butadiene (Philprene VP-15)	15/85	Polymer	100	70
			Zinc Oxide	3	
			Stearic Acid	1	
			Santo Cure	1.1	
			Sulfur	1.75	
			M. T. Black	150	

Compounding Recipes for the remaining compounds are not available at this time.

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