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HELIUM AND DEUTERIUM PERMEABILITY IN O-RINGS

J. F. Lakner

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MASTER

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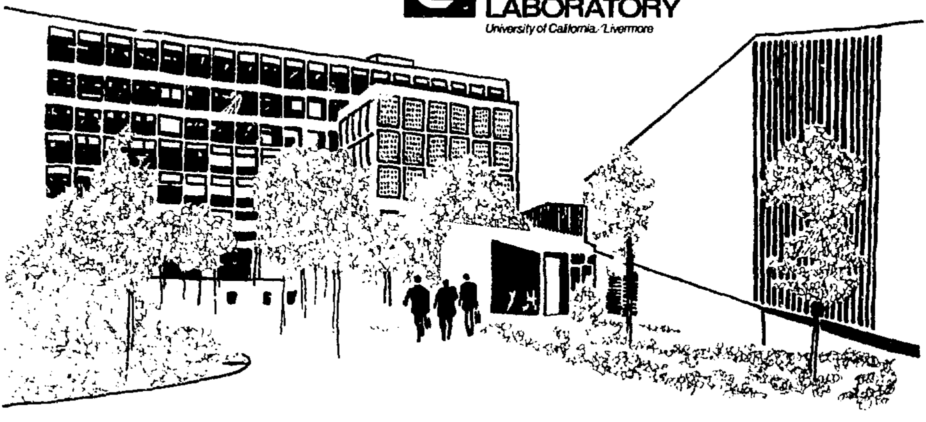

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Contents

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Abstract

To obtain more information on gas permeation through elastomeric O-rings, studies were performed on Parker Seal Company O-rings, Nos. 2-113, 2-006, 3-904, and 3-906, all made of a nitrile rubber. Also included in the tests was a valve packing (Autoclave Engineers) encased in AE Valve 20A-2142. Permeation experiments were run usually in duplicate to 82.7 MPa (12 000 psi) with helium and deuterium at room temperature. The data are extrapolated to give values for tritium.

Introduction

Elastomeric O-rings are used to close a passageway to prevent escape or loss of fluid. However, rubber membranes are permeable to gases, and the permeation rate differs for different gases. Furthermore, in recent years new types of synthetic rubbers have shown marked differences in behavior in the presence of specific gases. Tritium permeation through O-rings is of special concern for personnel engaged in tritium experiments. Therefore, it was decided to test a selected number of O-rings used in tritium handling apparatus. However, it was much easier to start the work with deuterium and helium and then add tritium experiments later.

Discussions on the phenomenon of gas permeation in O-rings are scarce.[1,2] Most of the literature deals with the classic permeation experiments through flat membranes, whereby the following permeation equation can be applied:

$$Q = K_p A \frac{\Delta P}{\ell} ,$$

where

Q = the amount of fluid permeating/unit time,

K_p = product of the diffusion coefficient and solubility coefficient of the permeating fluid in the barrier material,

A = area of flat membrane,

ΔP = pressure drop across the membrane,

ℓ = thickness of membrane.

It would be difficult to apply such a formula to an O-ring in a specific geometry because it involves such factors as percent squeeze and whether or not the O-ring is lubricated. Parker Seal Company has attempted to approximate leak rate with the following empirical formula for a specific gas:

$$L = 0.7\ FD\ PQ\ (1 - S)^2,$$

where

L = approximate leak rate,

F = He permeability rate of the elastomer (Parker tables[3]),

D = inside diameter of O-ring,

P = pressure differential,

Q = a factor which depends on the percent squeeze (Parker tables[3]),

S = % squeeze (e.g., 20% reductions in width in one direction = 0.2).

In view of the foregoing limitations, we decided to examine O-rings in the specific configurations used in the experimental systems. By performing work with deuterium, one could then roughly approximate the extent of tritium permeation by using the square root of the ratio of the atomic weights. However, tritium permeation experiments would still be required since we would incur the additional influence of radiation damage or possibly other factors. This report gives the test results with deuterium and also helium for comparison purposes. Both have mass 4, so it was quite easy to do the work with the same apparatus, a gas mass spectrometer.

Experimental

APPARATUS

Figure 1 shows a schematic diagram of the leak measuring system employed. The measuring instrument, a CEC-24-101A helium detector of Consolidated Electrodynamics Corporation, was set to detect mass 4 elements. To enable the rising leakage rate to be followed as a function of time, the helium leak detector was linked with a Hewlett-Packard recorder, Model 7100B. Although used, liquid nitrogen cold traps are not shown in the diagram.

The measuring system also provides for attachment of standard leaks to calibrate the leak detector. However, when a standard leak is used, the pressure vessel housing is not in place. The standard leaks (D_2 and He) were purchased from Veeco, NRC,

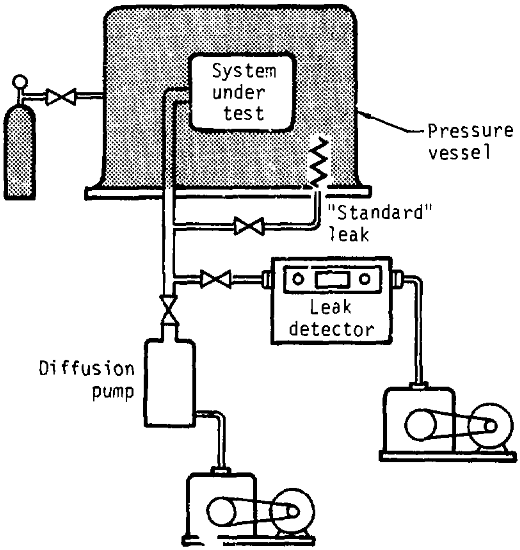


Fig. 1. Typical setup for permeation experiment.

and Hastings-Raydist. These leaks are within ±10% of the absolute accuracies in calibration.

O-Rings and Housings

Table 1 lists the O-ring seals, the fittings in which they were tested, and their application. Figure 2 is a photograph of the O-rings used in the permeation experiments, and Figs. 3 and 4 show the housings in which they were used. Appendix A gives detailed dimensions of each housing.

PROCEDURE

Standard Leaks

Calibrated gas leaks purchased from vendors were used to create a calibration curve showing scale reading of the leak detector vs leak rate obtained from standard leaks. The leak rates ranged between 5.4×10^{-8} to 7.69×10^{-4} for helium and 3.81×10^{-8} to 6.8×10^{-6} standard cm^3/sec for deuterium.

For calibration, a standard leak outlet was attached to the spectrometer

Table 1. O-rings examined; their high-pressure holding fixtures, and application for each O-ring.

O-Ring Parker Seal Co. Number	Dimensions			
	Cross-section width, mm	I.D., mm	Used in fixture number	Application
2-113 TVQ[a]	2.62	13.94	Conax-TX 30	Conax
(Compound N507-9)				Squib Valve TX 30
2-006 TVQ	1.78	2.9	Autoclave Engrs.	Conax
			10A 5986 Ferrule	Squib Valve TX 30
			10A 5983 Cap	
			10A 5984 Nut	
			Simulates Conax TX 30	
3-904 TVQ	1.83	8.92	Autoclave Engrs.	Pressure transducers
			10A 5918 Adapter	
			20A 1878 Blind cap	
3-906 TVQ	1.98	11.89	10A-5946 Adapter	Pressure transducers
			20A-1879 Blind cap	

[a]Top Value Quality.

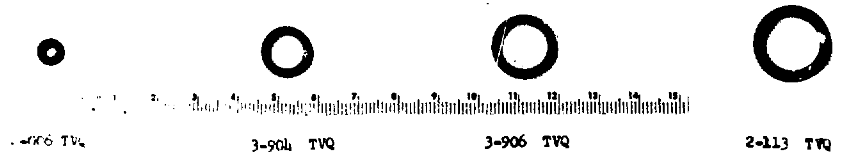


Fig. 2. O-rings used in permeation tests.

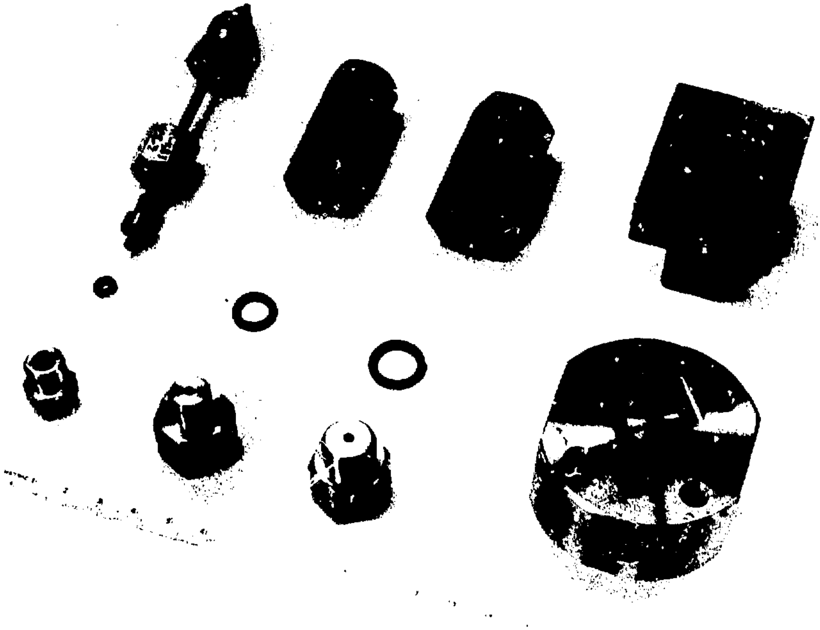
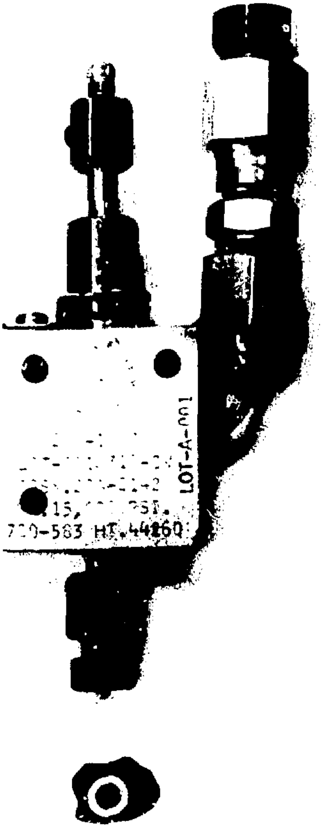


Fig. 3. O-ring housings.

inlet and pumped for at least 16 hours to insure a low residual gas content in the leak outlet and spectrometer inlet. An insulating mantle was put around the leak to reduce temperature variations during calibration runs. The temperature was held between 23 to 24°C. Before taking a leak rate reading for a particular standard leak, the mass spectrometer accelerator voltage was adjusted for maximum response. Separate calibration curves were established for He and D_2, since the ionization characteristics of the two gases are different. The calibration curves do not remain constant because of the drift in the leak detector. Therefore, calibration runs were made perodically.

O-Ring Permeation

O-rings were assembled in their fixtures as listed in Table 1. The sealing torque used for each O-ring is shown in Table 2. After the



fixture was assembled in the apparatus shown in Fig. 1, a vacuum was established on both sides of the O-ring. In addition, the manifold on the high-pressure side of the O-ring was purged five times with the test gas D_2 or He. The test gas was then admitted until the particular test pressure was reached. Subsequently, leak detector scale readings vs time were recorded so that with the use of the calibration curve, a leak rate vs time for a specific O-ring could be constructed.

We arbitrarily selected a pressure of 82.7 MPa (816 atm) for most of the tests, since this was approximately the pressure of the gas change in the storage cylinder.

Fig. 4. Valve housing and Teflon O-ring 10A-5965. Scale in mm.

Table 2. Torque applied to seal O-rings, and gap on low-pressure side.

Type of ring	Torque	Gap
2-113 TVQ[a]	35 in.-lb	6 mils — Compressed to 78.5% of original cross-sectional width.
2-006 TVQ	15 in.-lb	None
3-904 TVQ	Adapters contacts blind cap-metal to metal.	None — Gas leaks out through irregularities in contact surface.
3-906 TVQ	Adapters contacts blind cap-metal to metal.	None — Gas leaks out through irregularities in contact surface.

[a]Top Value Quality.

Results and Discussion

The permeation rates obtained in these experiments apply only to the geometry in question, the particular test pressure, the temperatures of 23 to 24°C, and the particular elastomer.

As is well known, gases permeate elastomers at a rate varying directly as the pressure, implying that here the molecules enter the solid undissociated. Figure 5 shows such a relationship with helium and Parker Seal Company's O-ring 2-113 TVQ installed in a Conax TX 30 valve assembly. This test was performed without removing the O-ring from the fixture, and by using several different pressures at 23 to 24°C.

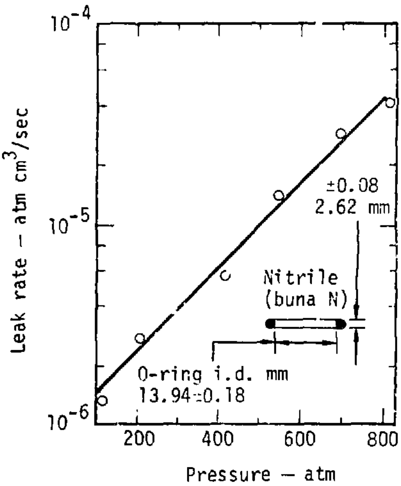


Fig. 5. Leak rate vs pressure for helium using Parker O-ring 2-113 TVQ for all points. Conax TX 30 valve geometry, 6-mil gap.

There would have been scatter in the data if different O-rings had been used for each test, since it would have been difficult to get the identical squeeze on the ring, and also a ring with identical characteristics.

In the experiments with O-ring 2-113 TVQ installed in the Conax TX 30 fixture (Fig. 1), we saw droplets of plasticizer deposited on the metal surface on the exit side of the O-ring. This was the acylate and sebacate plasticizer from the O-ring. It was not noticeable in any of the other permeation experiments. In this experiment, there was an appreciable amount of "burping" — pressure pulses into the leak detector that take the form of repetitive pressure surges of very short duration. We believe this (see Appendix A) is because of the geometry of the O-ring seat (90-void corner in this case) where pressure builds up and then releases. This phenomenon has been observed by others and can be corrected by filling or eliminating the void.[4]

In Fig. 6 we have plotted the permeation obtained using a Parker O-ring 2-113 TVQ, and He at three pressures: 10.34, 41.37, and 82.74 MPa. There is also a plot for He at 41.37 MPa, but with a 4- to 5-mil flange gap instead of the previous 5- to 6-mil gap showing an appreciable effect of an additional 1% compression. The O-ring installed with a 6-mil flange gap was reduced by 21.5% in width (called squeeze).

The initial exponential rise of the curves points to a diffusion process through the sealing material. Gas losses are therefore not to be traced to defects in the O-ring or its groove, but rather to the diffusion of gas through the O-ring material and along the contact area. After complete saturation of the O-rings, the escape of gas should logically remain constant, and this is, in fact, confirmed by the final approximately horizontal path of the curves.

In Fig. 7 we show three curves for permeation of D_2 at 82.74 MPa through O-ring 2-113 installed in Conax fixture TX 30. The curves ● and o do not show a significant difference even though the gas exit gaps differed by a factor of 2, unlike the experiment with helium. Note the slow release of gas once the upstream pressure is released. Curve □ shows permeation through the same ring with D_2, but with the ring greased with D-C* hi-vacuum grease. Lubricating this O-ring with vacuum grease increased the leakage in this experiment. Parker literature states that lubrication with vacuum grease reduces the leakage of the light (15%) squeeze

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*Dow-Corning Company.

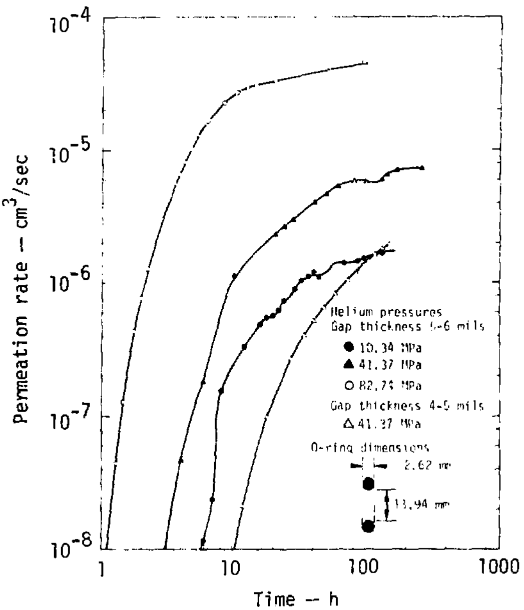

Fig. 6. Permeation rate vs time for helium using Parker O-ring 2-113 in Conax TX 30 housing.

rings, but the effect of grease is considerably less at 30% squeeze. At 50% squeeze the effect of the grease was not detectable. However, in Fig. 8 we agree with the Parker Co.

In Fig. 8 we show four curves for permeation of D_2 through Parker O-ring 3-906. Two curves are replicates, one is a greased ring and the remaining curve is for permeation using helium for comparison. Figure 9 shows the D_2 permeation through O-ring 3-904. Again the replication is good; the permeation in 3-906 and 3-904 is almost the same.

The pre-steady-state permeation in 2-006 (Fig. 10), a slightly smaller ring, is at a somewhat lower rate than in either 3-904 or 3-906 but the steady-state rate is approximately the same.

Figure 11 shows two curves for the permeation of D_2 through Autoclave Engineers' valve 20-A 1867, using Teflon as the seal Curve Δ was obtained with D_2 on a new valve with no previous treatment. Afterward, Curve o was obtained by pressurizing this valve with He to 22 500 psi for one-half hour. The pressure was dropped to 15 000, then leak-checked, and finally D_2 was allowed to permeate at an upstream pressure of 12 000 psi.

A peculiarity was noted in permeation of D_2 through the 3-904 and the 3-906 rings. These are rings which receive a twisting action when they are screwed into the female fittings. A peak value is reached, then it drops to a slightly smaller value

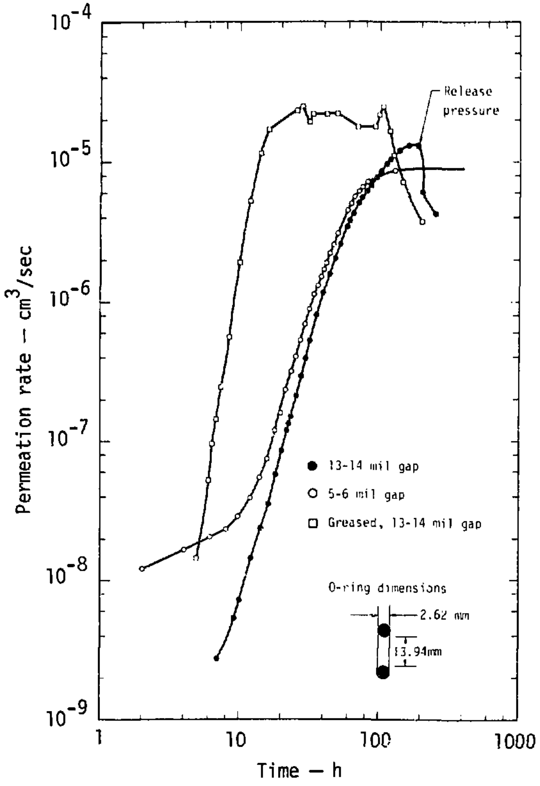

Fig. 7. Permeation rate vs time for D_2 through Parker O-ring 2-113 in Conax TX 30 housing. Note gap variation and effect of grease. Upstream pressure, 12 000 psi.

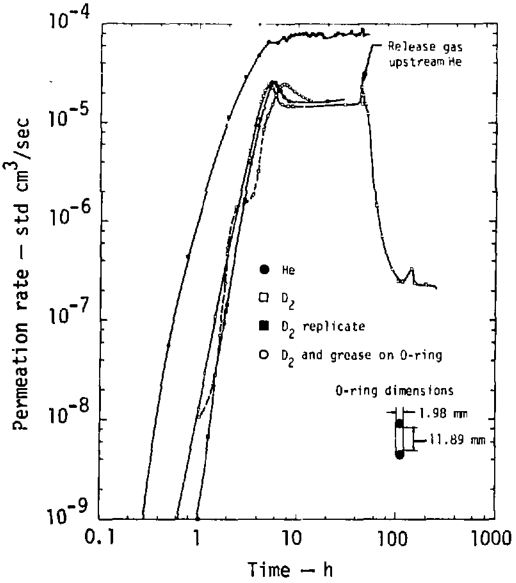

Fig. 8. Permation rate vs time of He and D_2 through Parker O-ring 3-906. Note erratic effect of vacuum grease. Upstream pressure, 12 000 psi.

as the steady-state portion is approached. This is in contrast to the behavior of other O-rings (compressed in a vertical direction without twisting), where the ascending curve gradually enters the horizontal steady-state portion. We have no good explanation now for the existence of the peak when D_2 permeates twisted O-rings.

In Table 3 we have listed the time that elapses before an arbitrarily fixed permeation rate of 1×10^{-8} cm^3/sec is noticed. The steady-state permeation rate for each ring is also shown for the maximum pressures at temperatures of 23 to 24°C. The time to reach this steady-state is also listed.

Note that the smaller O-rings (2-006, 3-904, 3-906) in Table 3 have the larger permeation rates. This is most likely due to the larger exposure area available in their respective fixtures. If the ratio of the square root relationship $\sqrt{D}|\sqrt{T}$ holds true, then tritium permeation should be less than deuterium unless the ring suffered radiation damage and subsequent catastrophic failure resulted. This, however, should be confirmed in future experiments, but with apparatus designed specifically for tritium.

From Table 3 the total D_2 permeating (we have added in the amount permeating in a valve, V-3, AE No. 20A-2142) at steady-state amounts to 7.8 $\times 10^{-5}$ std cm^3/sec at 82.74 MPa (12 000 psi). Tritium permeation

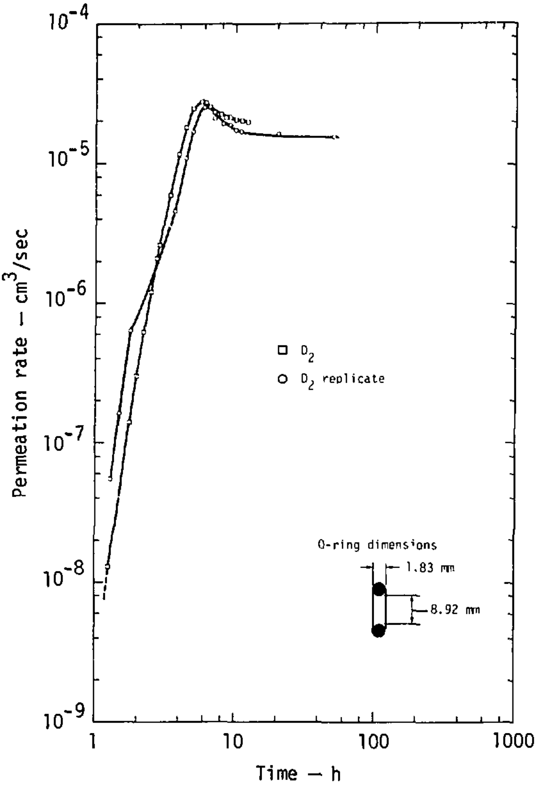

Fig. 9. Permeation rate vs time for D_2 through Parker O-ring 3-904. Curves are replicates on different rings. Upstream pressure, 12 000 psi.

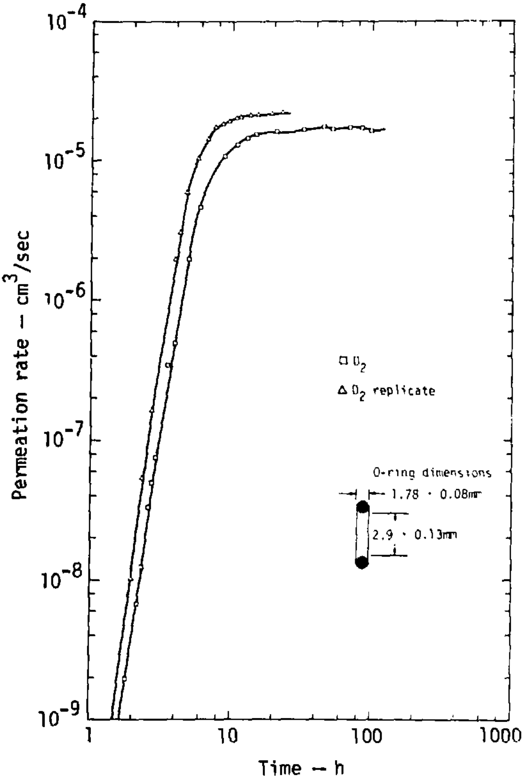

Fig. 10. Permeation rate vs time for D_2 through Parker O-ring 2-006. Curves are replicates on different rings. Upstream pressure, 12 000 psi.

would be about 0.8 of the above value, or 6.2×10^{-5} cm^3/sec, barring radiation or other effects as yet undetermined. This permeation rate corresponds to about 5 Ci/da for pure tritium.

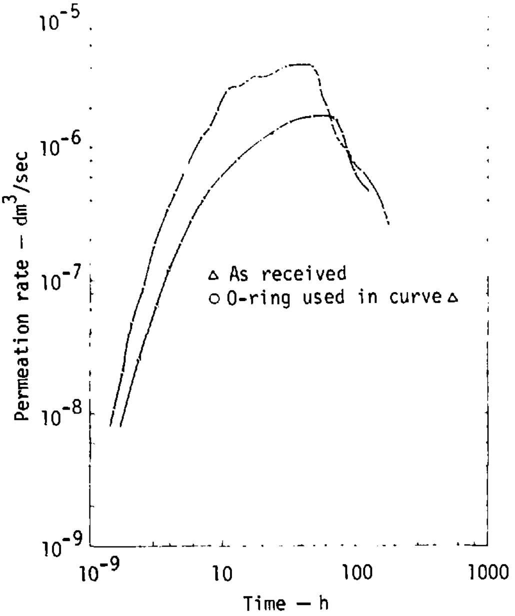

Fig. 11. Permeation rate vs time for D_2 through Autoclave Engineers Valve 20-A-1867 with Teflon ring 10A-5965. Upstream pressure, 12 000 psi.

Table 3. Time required to reach permeation rate of $1 \times 10^{-8} cm^3$/sec, and maximum permeation rate and time to reach steady-state using D_2.

O-ring type	Pressure, MPa	Time to reach 1×10^{-8} std cm^3/sec, hr	Steady-state permeation at max presure, std cm^3/sec	Time to reach steady-state, hr
Parker No.				
2-113 TVQ[a]	82.74	8.5 ("t_d" in Fig. 4)	9.0×10^{-6}	130
2-006 TVQ	82.74	2.0	2.2×10^{-5}	16
3-904 TVQ	82.74	1.2	2×10^{-5}	10
3-906 TVQ	82.74	1.35	1.65×10^{-5}	10
(Refer to Figs. 4 and 5)				

[a]Top Value Quality.

Summary

Four O-ring sizes were examined to determine rates of deuterium and helium permeation. The sizes were Parker Seal Company Nos. 2-113, 2-006, 3-904, and 3-906. Totaling flow rates through the two O-rings in a Conax TX 30 valve (2-113 and 2-006), pressure transducer O-rings (3-904 and 3-906), and valve V-3, AE 20A-2142, results in 7.8×10^{-5} std cm^3/sec for D_2, or 6.2 std cm^3/sec $\times 10^{-5}$ for T_2, barring radiation and other possible effects. Since this is only an approximation for tritium permeation, accurate numbers will be obtained when actual experiments are performed with T_2 gas.

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RMD/jn/vt/lmc

Appendix A

Dimensions of Housings

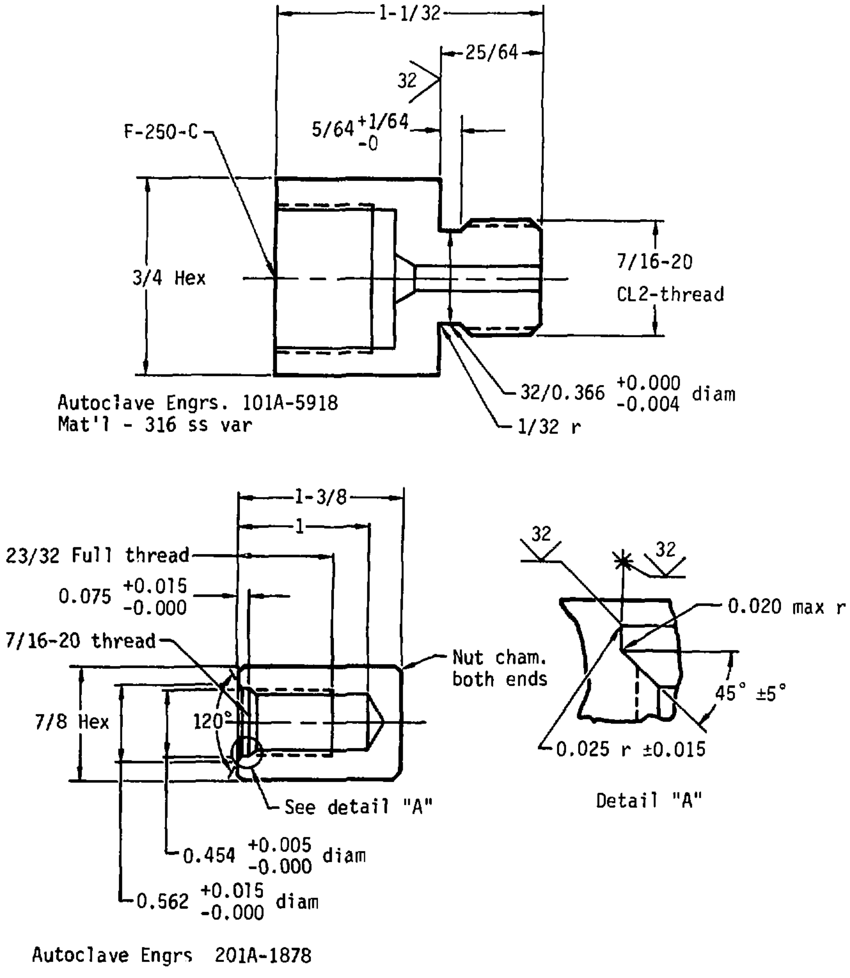


Fig. A-1. Subassembly O-ring 3-904.

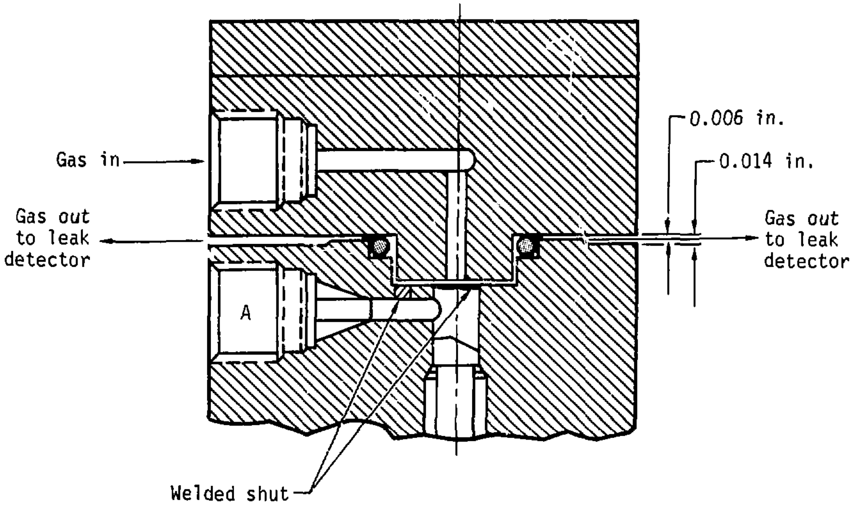
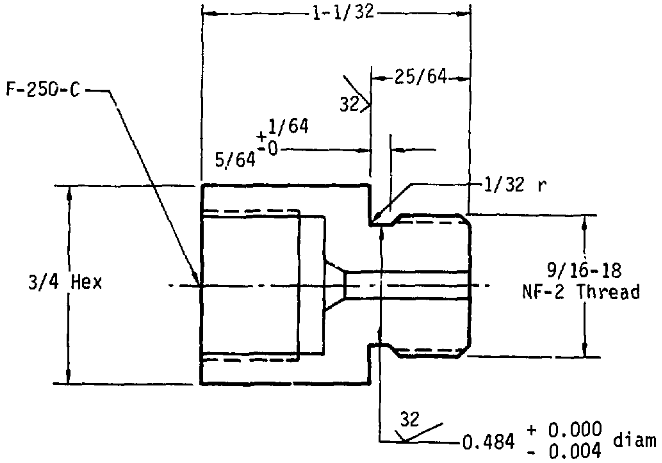


Fig. A-2. Conax TX 30 Valve modified for permeation experiments on O-ring 2-113 TVQ.



Autoclave Engrs. 101A-5946
Mat'l - 316 ss var

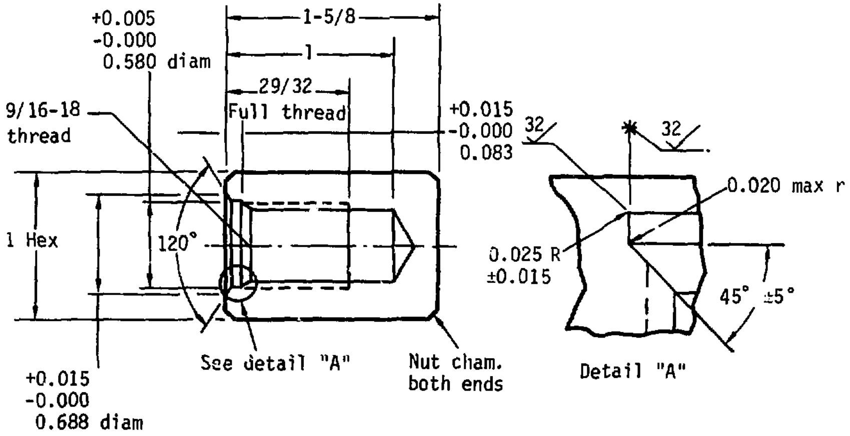


Fig. A-3. Subassembly O-ring 3-906.

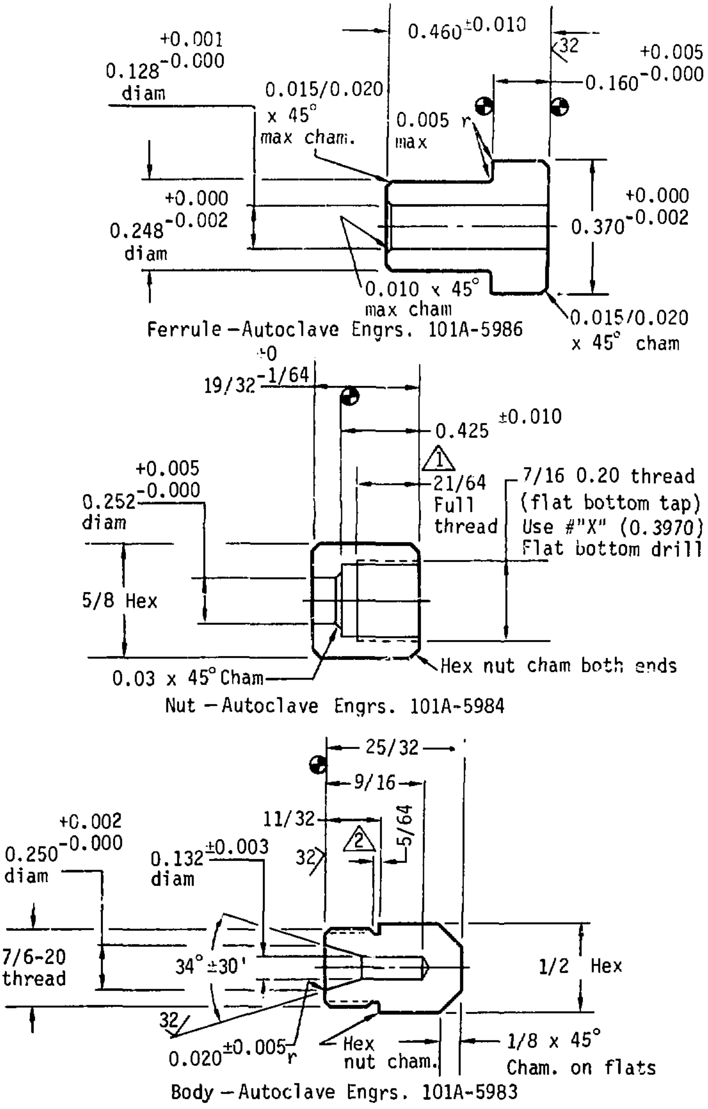


Fig. A-4. Subassembly O-ring 2-006 TVQ.