# INTERSPACE LEAK-TESTING
## A COMPARISON WITH HELIUM MASS SPECTROMETRY

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## INTRODUCTION

Experience with transport packages for radioactive materials has revealed unexpected discrepancies between different techniques used to measure leakage.

Turn-round or dispatch measurements frequently involve the Interspace Pressure Decay (IPD) technique. This requires a double seal arrangement for each penetration of the containment boundary as illustrated in Figure 1. The space between the seals (generally elastomer O-rings) is pressurised via a port and the pressure monitored over about 30 minutes. The gas leakage rate is derived from the rate of fall of pressure.

The evaluation of leak rate is described in various standards and codes of practice AECP 1068. It involves the knowledge of the pressurised volume which may vary with the exact disposition of the elastomer seals. The volume can be determined at each measurement by connecting a calibrated volume at ambient pressure to the interspace. The gas laws can be used to obtain the interspace volume from the step change in pressure.

The results of this measurement are frequently reduced to standard conditions of temperature, pressure and a common gas to facilitate comparison with leakage tolerances for a particular application. This Standardised Leak Rate (SLR) may be compared with the results of other techniques used for the same application provided both are presented in SLR format. This is described in AECP 1068 and programmed into instruments such as the FLITE meter for automatic treatment of these procedures (Eaves 1992).

Alternatively, the more sensitive Helium Mass Spectrometry (HMS) method may be used. This is generally reserved for total leak-rate determinations following major maintenance and during manufacture when small leak paths through faulty welds can be detected. Total leakage measurements require access to the load cavity and may not be convenient for regular use.

The technique is particularly sensitive because helium leakage can be distinguished from other gases at the atomic level. Quantitative measurements require the collection of the leaking gas via a high vacuum system which is not often convenient for turn-round tests. The HMS method relies on calibration with well-defined leaks supplied with the instrument. These calibrated leaks are traceable to national standards.

The IPD technique relies on the calibration of sensitive transducers and correction for temperature changes if high accuracy is demanded. Automatic instruments like the FLITE meter intercalibrate several transducers before each measurement, identifying deviations from internally-stored calibrations and rejecting doubtful signals.

Inevitably, the IPD and HMS techniques have been applied to the same seals under nominally similar conditions and significant discrepancies observed. Additional, better controlled comparisons have been performed and the discrepancy confirmed. This has produced the better understanding of leak-rate measurements described below. This will enable the most appropriate method to be selected for each application and valid conclusions to be drawn from the results.

## RESULTS OF INTERCOMPARISONS

Measurements of standardised leak rates from several pairs of Viton O-ring seals using the IPD and HMS methods have revealed significant differences between the results. The introduction of the sophisticated FLITE meter, capable of accurate measurements to an accuracy of $10^{-7}$ Pa.m$^3$/s ($10^{-6}$ bar.cm$^3$/s) compared with previous simpler methods capable of only $10^{-4}$ bar.cm$^3$/s, has made this discrepancy more obvious. It can no longer be attributed to the large tolerances of the IPD method. As a result, a programme of measurements under laboratory conditions was mounted to confirm the magnitude of the discrepancy and to investigate the source.

A pair of steel plates with three concentric Viton O-rings (Figure 2) were bolted together to form three sealed volumes; the "cavity", an inner interspace and an outer interspace. This enables leak-rates across the two inner O-rings to be measured with flow in either direction, i.e. with either an internal or external overpressure. The cavity and interspace volumes are respectively about 10 and 20 milli litres but the exact values varied with O-ring position.

The results of standard tests presented in Table 1 confirm that the IPD method yields "leak-rates" some two orders of magnitude higher than from the HMS method in this arrangement where bypass leakage is small or zero (see below). Leak-rates from the cavity using air, nitrogen and helium are consistent within the tolerance of the FLITE meter. The SLR value of $(4\pm1)\times10^{-6}$ Pa.m$^3$/s does not change significantly with the gas used, so the different diffusion properties of helium used with the HMS method is not the source of the discrepancy. The higher mobility of helium would, in any case, suggest higher leak rates from the HMS method, not as found.

The HMS method yields values of SLR lower by a factor of about 200, i.e. $(1.9\pm1.0)\times10^{-8}$ Pa.m$^3$/s. This is confirmed by the cavity leakage results (across a single O-ring) where the flow is in the opposite direction to that from the interspace (leakage across two O-rings). These results are at the limit of measurement for the two techniques and therefore represent a zero bypass leakage situation. The so-called "leaks" are a simple interpretation of the signals from the instruments.

The two orders of magnitude discrepancy, observed during manufacture and turn-round operations, has therefore been confirmed under conditions allowing direct comparison of the techniques. Further tests have therefore been performed, looking for a time dependence as described below.

## TIME DEPENDENCE

Both the IPD and HMS techniques have been used in a time-dependent mode. The HMS method has been applied with a continuous vacuum but with spot measurements over a seven-hour period. The IPD procedure, as used with the FLITE meter, requires repressurisation of the interspace to allow checks on transducer calibration prior to each measurement. Measurements were derived from the transducer output, avoiding the need for depressurisation, over a six-hour period with the gas pressure maintained between measurements.

The time-dependent HMS results are shown in Figure 3 with the equivalent IPD results in Figure 4. The HMS values show a steady increase with time, rising from less than $10^{-9}$ to about $7 \times 10^{-8}$ Pa.m$^3$/s. Care was taken to ensure that the seals were not contaminated with helium prior to the tests, so very low initial leak rates were obtained. The values quoted are raw leak rates, not corrected to SLR format, a difference of about 5%. The SLR conversion process programmed into the FLITE Meter assumes laminar gas flow through a capillary when these results suggest the process is permeation through the seal volume. Such conversions are therefore invalid.

The IPD values show an initial high level ($2 \times 10^{-6}$) which rapidly declines to less than $5 \times 10^{-7}$ Pa.m$^3$/s during the first two hours. There is some evidence that a seal moved at one hour giving rise to a high reading in one case. The results are re-plotted as Figure 5 with this one point reduced by a factor of 10 to illustrate the changes better. The ultimate values (about $(1 \pm 2) \times 10^{-7}$ Pa.m$^3$/s) correspond to residual permeation leakage, not inconsistent with the HMS values of 5 to $10 \times 10^{-8}$ Pa. m$^3$/s.

Earlier measurements of helium leakage were performed with a seal which had been used with helium with the IPD technique. The results, shown in Figure 6, are consistent with the release of helium from the seal followed by an increase representing both permeation and the release of dissolved gas. In this case the helium flow appears to saturate at about four hours, significantly before any evidence of saturation in Figure 3. The difference is attributed to helium previously dissolved in the seal.

## THEORY

Interspace leakage may be analysed by reference to the basic gas laws:

$$PV = MRT \qquad (1)$$

where P is the gas pressure (Pa)
V is the gas volume (m$^3$)
M is the mass of gas in volume V (kg)
R is the gas constant (J.kg$^{-1}$.K$^{-1}$)
T is the absolute temperature (K)

Thus the rate of change of pressure (with t the time in seconds) is:

$$\frac{dP}{dt} = \frac{d(MRT)}{V.dt} = \frac{R}{V} . \frac{d(MT)}{dt} \qquad (2)$$

$$\frac{dP}{dT} = R.\left\{\frac{T}{V} . \frac{dM}{dt} + \frac{M}{V} . \frac{dT}{dt} - \frac{MT}{V^2} . \frac{dV}{dt}\right\} \qquad (3)$$

$$\frac{1}{P} . \frac{dP}{dt} = \frac{1}{M} . \frac{dM}{dt} + \frac{1}{T} . \frac{dT}{dt} - \frac{1}{V} . \frac{dV}{dt} \qquad (4)$$

The rate of change in pressure may be caused by a change in gas content (i.e. mass, resulting from leakage for example), a change in temperature or a change in volume, due to the movement or compression of the elastomer seals for example.

The Standardised Leak Rate is given by AECP 1068:

$$SLR = \frac{VT_o(P_1 - P_2)}{tT} \qquad (5)$$

where $T_o$ is the standard temperature (298K)
and $P_1$-$P_2$ is corrected for the temperature change during the measurement period t seconds

Substituting $\frac{dP}{dt}$ for $(P_1-P_2)/t$

$$SLR = VPT_o \left\{ \frac{1}{M} \cdot \frac{dM}{dt} + \frac{1}{T} \cdot \frac{dT}{dt} - \frac{1}{V} \cdot \frac{dV}{dt} \right\} \qquad (6)$$

Thus the measured leak rate may be influenced by temperature and/or volume changes unless these are corrected. The FLITE meter incorporates a sensitive gas temperature measurement and automatically corrects for changes during a measurement. Volume changes cannot readily be separated from the gas leakage (M) term and therefore cannot be corrected.

The change in leak-rate with test duration, as measured by the FLITE meter, may be the result of either a variable rate of loss of gas or a variable rate of change of volume or a combination of both, assuming that any effects of temperature changes are compensated by corrections. In this test series, pressures are resolved to 0.1 mbar (10 Pascals) equivalent to about 0.02°C.

## ANALYSIS

The two methods of leak-rate measurement yield different results because they measure different parameters. The HMS technique detects gas flowing from the outside of the seals while the IPD technique detects gas leaving the interspace volume. These will not be the same if gas can be absorbed in and released from the seals under the influence of gas pressure. Evidence from the series of tests reported here demonstrates that gas dissolves in the elastomer seal material but is not released immediately from the outer surfaces of the seal. No other significant leak-paths exist to provide this gas flow.

A model of gas dissolving in the seal material, under increased interspace pressure, is consistent with the time-variation of the two measurements. Initial gas loss by solution will decline as the internal face of the seals reaches saturation. An equilibrium leak-rate will result when the gas solution distribution through the seal becomes linear and a constant rate of diffusion (permeation) is achieved Weise et al. 1990.

Similarly, the HMS method initially detects only bypass leakage but will eventually see diffusion/permeation flow as shown in Figure 6.

The importance of permeation for gas leakage determinations has been recognised in the past George and Williams 1984 but the interpretation of short-term measurements by the pressure decay method has not been widely understood. Pressure-rise techniques, where a vacuum is established in the interspace, will also be influenced by gas permeation. The time taken to establish the initial vacuum will determine what contribution gas leaving solution in the elastomer will have.

## CONCLUSIONS

Gas leakage determination methods will generally yield results with contributions from bypass leakage through holes (capillaries and orifices) and permeation (diffusion through the body of the seal material). Where elastomers, such as Viton or Silicone Rubber, are used as seal materials, gas will dissolve in the bulk material under pressure or will be released from the material under reduced pressure. After a period of time, depending on the seal materials, the size and shape of the seals and on the gas used, a steady rate of flow of gas by permeation will result.

The Interspace Pressure Decay method will initially overestimate gas leakage for two reasons. Gas leakage through two seals is measured and attributed to the inner seal as flow from the cavity through the inner seal only is of importance for practical purposes. The method cannot distinguish the potentially dominant loss of gas by solution in the seals from bypass leakage. The ultimate loss of gas by permeation leakage from the outer surface will be slower than the initial loss by dissolution.

Evidence presented here suggests that the technique will overestimate bypass leakage by up to two orders of magnitude. This conclusion is clearly geometry and material dependent and will be influenced by the magnitude of any real bypass leak rate.

The use of the Interspace Pressure Drop method with several hours of interspace pressurisation (the time depending on material, geometry and gas) will ultimately measure the sum of bypass and permeation leakage.

The Helium Mass Spectrometry method will initially measure only bypass leakage. Only after several hours (depending on geometry etc) will the technique measure the sum of bypass and permeation leak rates. The permeation contribution will be that for helium and differ from that for air and for any gases which require containment for safety reasons.

Gas leak-rate measurements are frequently used to demonstrate that there are no holes (capillaries or orifices) through the containment capable of transmitting solid matter in the form of aerosols. This is generally based on the assumption that a small gas leak rate implies the presence of only very small holes not capable of passing micron-sized particles.

The use of the IPD method within about two hours of pressurising the interspace will exaggerate gas leak rates. The technique will clearly be adequate to demonstrate compliance with the agreed limits, provided of course the results are acceptable. The use of the HMS method will also be adequate to demonstrate compliance, but without the large margins inherent in IPD methods. HMS measurements within about one hour of introducing helium will not detect significant permeation flow, if the data presented here are typical.

Where the results of IPD measurements are higher than the aerosol tolerance limit and low bypass leak rates are expected, improved results may be obtained by extending the test duration, provided the seal is maintained under gas pressure. This will ensure equilibrium gas solution in the seal, minimising gas flow by permeation from the interspace.

Alternatively, helium mass spectrometry can be used to demonstrate low bypass flow. This technique is generally less convenient, however.

Where gas leakage from a containment is important, the interspace pressure decay method is the most appropriate. Short-term tests will exaggerate leakage as two seals are under test and the method will include the gas solubility effect. Long-term tests will also overestimate leakage because of the double seal effect.

Helium mass spectrometry will yield low results for gas flow unless time is allowed for equilibrium helium dissolution in the seal. Initially the results represent only bypass leakage when permeation losses may eventually dominate. Care must also be taken when interpreting helium leakage in terms of other gas losses. While helium is likely to diffuse faster through discrete holes, permeation flow rates are influenced by gas solubilities which are not generally lower for helium than for other gases Weise et al 1990.

## ACKNOWLEDGEMENTS

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## REFERENCES

AECP 1068 - Atomic Energy Code of Practice, February 1992, AEA Engineering, Harwell, UK.

A R Eaves (AEA Technology) - Improved Techniques for Leak Testing of Flasks, Nuclear Engineering International, February 1993

Weise, H-P; Ecker,K-H; Kowalewsky, H and Wolk, Th. - Gas Permeation Through Common Elastomer Sealing Materials. - Vuoto, Vol. XX, No 2, April 1990

George, AF and Williams, D. - An Investigation of Leakage Past Static Elastomer Seals. - 10th International Conference on Fluid Sealing, Innsbruck, Austria, April 1984.

TABLE 1

COMPARISON OF MEASUREMENT TECHNIQUES

| Volume Pressurised | Gas | IPD (Pa.m$^3$/s) | HMS (Pa.m$^3$/s) |
|---|---|---|---|
| Interspace | Air | $3.0\times10^{-6}$ | - |
| Interspace | Nitrogen | $3.4\times10^{-6}$ | - |
| Interspace | Helium | $5.0\times10^{-6}$ | $1.9\times10^{-8}$ |
| Cavity | Helium | $7.0\times10^{-6}$ | $1.3\times10^{-8}$ |

Notes

1. IPD - Interspace Pressure Drop method using the FLITE meter.
2. HMS - Helium Mass Spectrometer method.
3. Interspace - Volume between inner and middle seals (Figure 2); involves two seals.
4. Cavity - Space within inner seal (Figure 2) - Leakage across one seal.
5. Accuracies - IPD (FLITE meter) (± 10%) or (± $1.0\times10^{-6}$) Pa.m$^3$/s.
   HMS (± 10%)

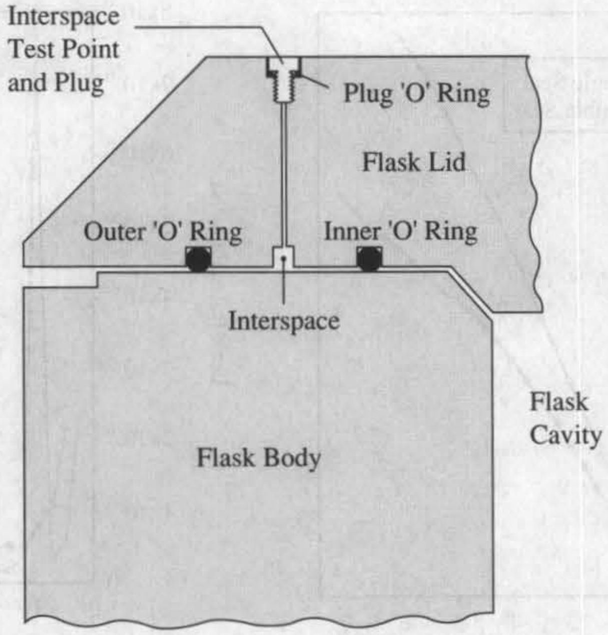

Figure 1 Double 'O'-Ring Seal Geometry

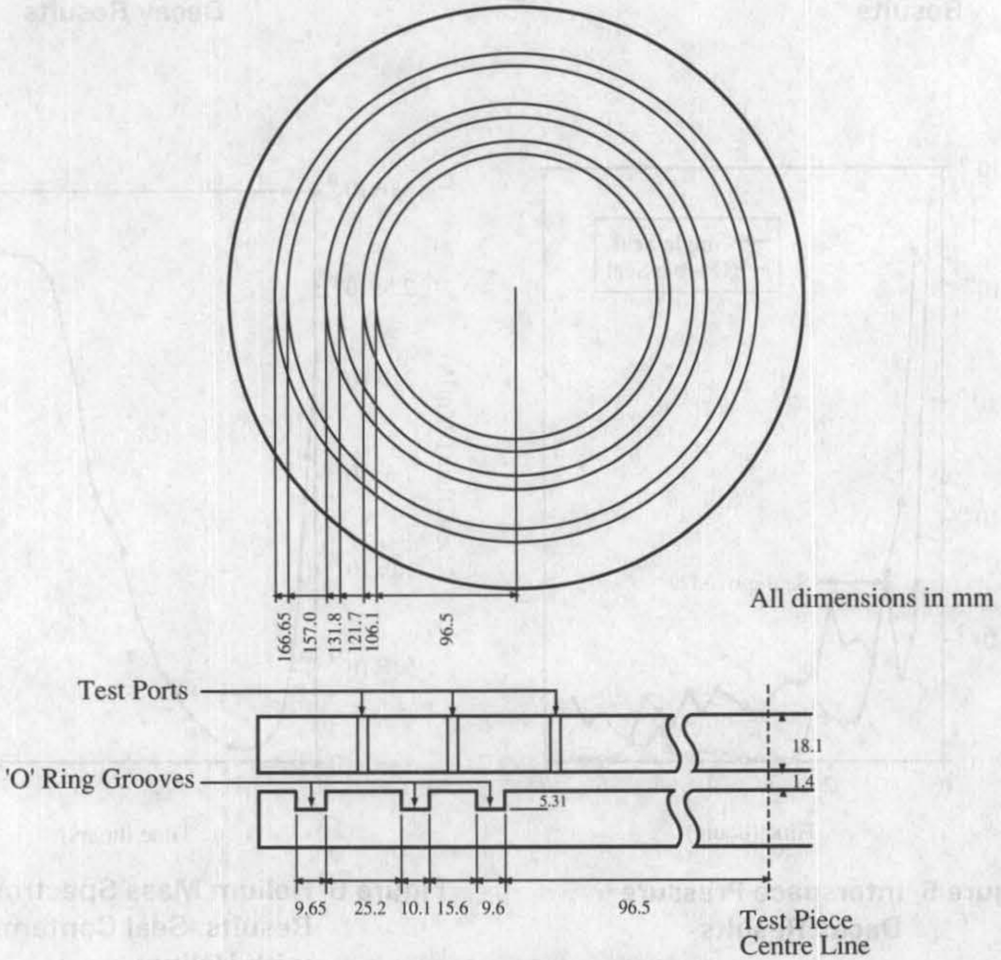

Figure 2 Test Piece

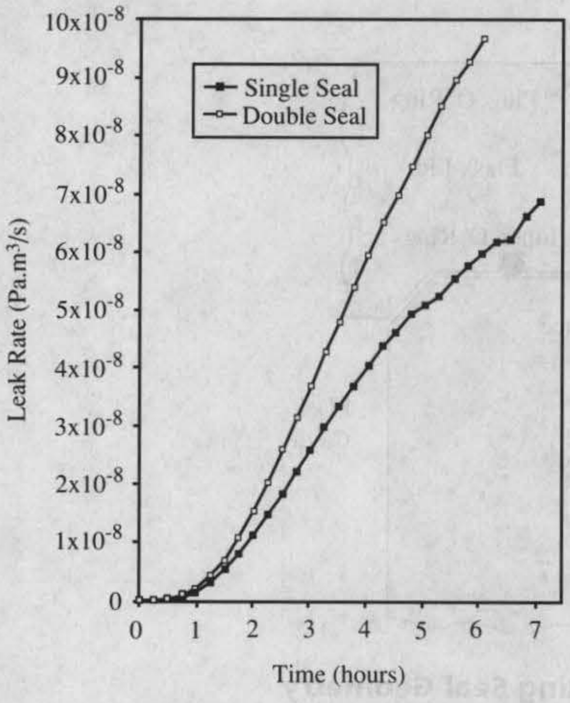

**Figure 3 Helium Mass Spectrometry Results**

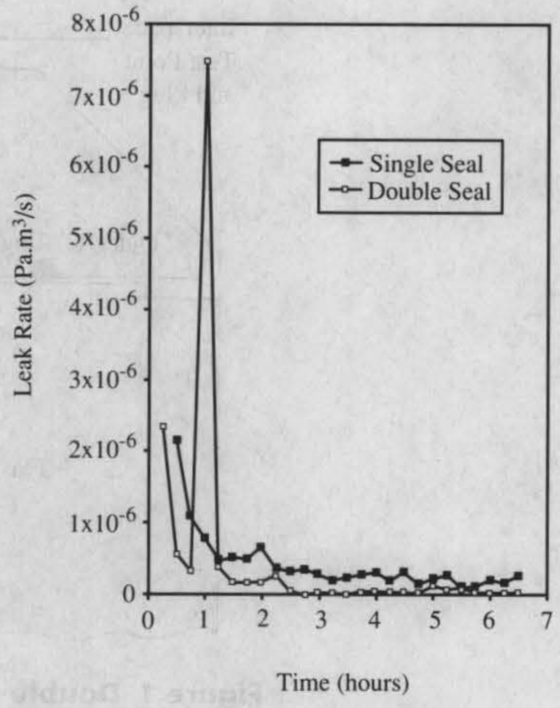

**Figure 4 Interspace Pressure Decay Results**

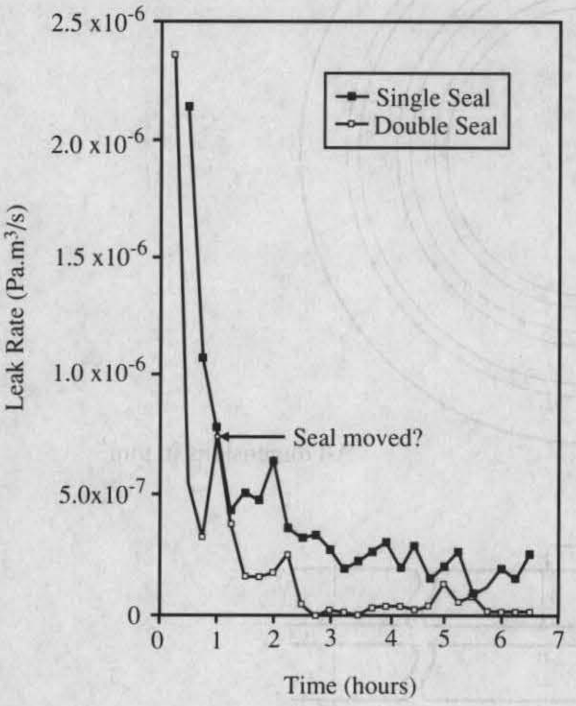

**Figure 5 Interspace Pressure Decay Results**

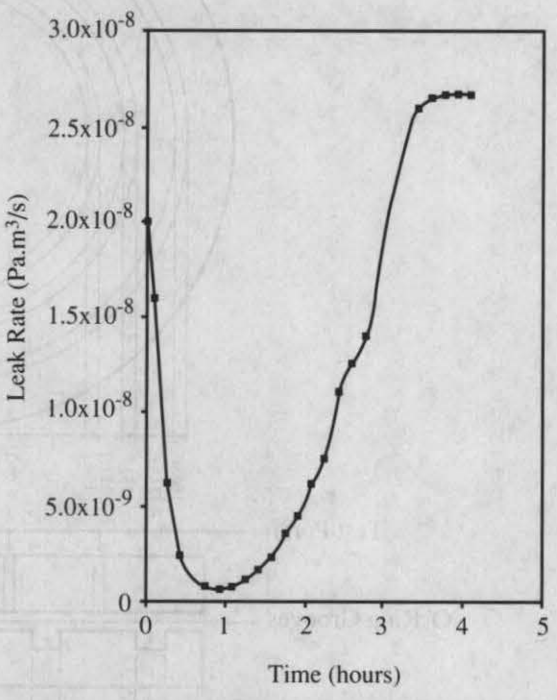

**Figure 6 Helium Mass Spectrometry Results. Seal Contaminated with Helium**