Containment of Radioactive Powders by Seals

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INTRODUCTION

Safety regulations require that the possible loss of radioactive material from a packaging is restricted to very low levels for both normal and accident conditions. This can be achieved by using a robust steel containment vessel with double O-rings seals to validate access ports. The effectiveness of each seal can be demonstrated by pressurising the interspace and calculating any leak rate from the pressure drop. For packagings containing radioactive gas or liquid, it is then possible to quantify any resulting activity release rate but for fine particulate material the relationship of likely solid transport in a known gas flow is much harder to define. Because of this, more reliance has to be placed on experimental evidence of powder retention by seals and powder flow through capillaries and orifice plates, sometimes with unduly arduous assumptions made such as the existence of an aerosol within the container.

Relationship Between Regulatory Permitted Activity Release And Practical Engineering Methods of Measurements

The design of packages may be demonstrated as meeting the relevant requirements of the Regulations by subjecting prototype or actual vessels to physical and environmental tests.

The package containment requirements are laid down in the Regulations (IAEA) which quote A_1/A_2 values for individual nuclides. For normal operations the permitted activity release is $A_2 \times 10^{-6}$/hour, the A_2 value being calculated for individual nuclides or combination of nuclides.

It is generally impracticable to obtain direct measurement in these units therefore an alternative fluid leak rate method is normally used. Codes of Practice (ANSI 1987; UKAEA 1988) have been produced which recommend test methods. Normally gas leakage testing is used and the theoretical relationships involved in these can be used. A paper to this PATRAM conference (Higson et al 1989) reviews the relationship between leak geometry and flow regime and suggests flow equations.

Modes of Gas Flow Through Leaks

In order to calculate hole size from measured leak rates of gas the correct flow regime must be considered. Different basic laws relate leakage rates to pressure differential across the leak, the range of absolute pressure involved, and the nature of the gas passing through the leak. Gas flow through holes may be categorised as molecular, transitional, laminar, turbulent, or "choked" flow. Therefore it is essential to know the possible flow rates and flow regimes before calculating the hole size.

The American National Standards Institute Standard N14.5 "Leakage Tests on Packages for Radioactive Materials" indicates the range of leak rates for which the flow regime requires consideration. It states that a leak rate of 10^{-7} atmosphere $cm^3\ sec^{-1}$ is considered to be leak tight, whereas a leak rate greater than 10 atmosphere $cm^3\ sec^{-1}$ is large and does not require testing.

Capillaries

Within this range it can be shown (Higson et al 1989) that the relationship between leak rate and hole size for a capillary can be best described by the Knudsen equation for Transitional Flow (ASNT 1982)

$$\theta = \frac{\pi}{8}\left(\frac{d}{2}\right)^4 \frac{Pa}{\eta \ell}(P_1 - P_2) + \frac{1}{6}\sqrt{\frac{2RT}{M}}\,\frac{d^3}{\ell}(P_1 - P_2)\left[\frac{1 + \frac{M}{RT} Pa \, d/n}{1 + 1.24 \frac{M}{RT} Pa \, d/n}\right]$$

where θ	= gas flow	Pa m_3 sec^{-1}
d	= diameter of leak	m
ℓ	= length of leak	m
η	= viscosity of gas	Pa sec
P_1	= upstream pressure	Pa
P_2	= downstream pressure	Pa
Pa	= $\frac{P_1 + P_2}{2}$	Pa
T	= absolute temperature	
R	= gas constant	J/mole °K
M	= molecular weight of gas	Kg/mole

Knudsen used this equation to represent his experimental data. At low pressures the equation reduces to an equation for molecular flow whereas at high pressure the equation reduces to one of strictly laminar flow.

Orifices

The flow through orifices is dependent upon the pressure upstream and downstream of the orifice. Where the ratio of the downstream to the upstream pressure is less than a certain ratio choked (or sonic) flow occurs.

The critical ratio rc of downstream pressure to upstream pressure is given by

$$rc = \frac{Pd}{Pu} = \left(\frac{2}{\gamma - 1}\right)^{\left(\frac{\gamma}{\gamma - 1}\right)}$$

where γ = ratio of specific tests

rc for air = 0.528 so that conditions of 1 atmosphere to vacuum and 2 atmosphere to 1 atmosphere are in the sonic flow regime.

The mass flow rate under choked flow conditions is given (ANST 1982) by

$$\theta = \left[\frac{\pi d^2 P_1 Co}{4M}\right] \Big/ \left[RT\gamma\left(\frac{2}{\gamma - 1}\right)\right]^{\left[\frac{\gamma + 1}{\gamma - 1}\right]}$$

Figure 1 illustrates the range of hole size and leak rates for a 10mm capillary and an orifice at 1 bar to .01 bar upstream and downstream pressure conditions.

Experimental Leakage Investigation

A typical type B package would consist of a container with a seal, the seal probably being in the form of an O-ring. A typical leak across the seal could be caused by a scratch or a trapped "hair" which would produce a leak path with a relatively long length to diameter ratio (ie a fine capillary). A possible length could be of the order of 10 mm. From Figure 1 it can be seen that the range of hole diameter is 1 - 100 µm.

Other forms of seal might involve the use of a knife-edge. In these cases the hole would be in the form of an orifice rather than a long capillary. From Figure 1 it can be seen that the orifice diameter would be up to 100 µm.

A number of investigators have carried out experimental work to determine the practical flow of powders through small holes. Their investigations have tended to concentrate on particular material and with particular packages in mind. This paper reviews the experimental work to see if any general conclusions can be reached.

Curren and Bond (1980)

This study was directed at determining an appropriate standard of leak tightness for containment of plutonium oxide powder during shipment in wood/cadmium containers. They assessed the differential pressure that would be generated across a leak path in normal and accident conditions and, for the container being considered, arrived at a 1 bar differential. The experimental apparatus consisted of a steel vessel which could be pressurised. The vessel could be fitted with orifice plates. Orifice plates were manufactured with a range of sizes between 5 and 200 µm.

The experimental programme was carried out to try to establish a correlation between mass transfer and gas flow rate for a range of orifice sizes. UO_2 powder with a particle size range of 1 - 20 µm was used to simulate PuO_2. The experimental apparatus could be vibrated. The experiments were carried out with mainly 1 bar pressure inside the apparatus and vacuum on the outside of the orifice. In these conditions "choked" flow was achieved.

The experiments showed that:

the amount of powder transferred through the orifices of 25, 50, 100 and 200 µm is a function of differential pressure and time;

orifices of 5 µm and 10 µm became plugged; the 5 µm orifice even plugged during the "gas" flow tests.

when the hole was situated under the powder level the hole rapidly plugged;

the aerosol density increased when the hole was close to the powder surface, but bringing the hole closer to the powder resulted in more frequent blockage. Table 1 summarises the experimental data.

Yesso et al (1980)

This work was done to provide data for prediction of PuO_2 emissions from shipping containers. ThO_2 was used initially to set up the apparatus and validate the technique, then PuO_2 was used.

Experiments were designed around a four parameter matrix. (1) gas pressure (2) leak orientation (3) agitation and (4) leak size/type. Also considered for a few experiments was total run time. The data produced exhibited a high degree of non-reproducibility which severely limited the ability to draw any firm conclusions. There was no apparent dependency for any single parameter except for a weak dependency upon the interaction between orifice size and pressure.

The results of the experiments in which the run times were varied showed that a change in the run time did not result in a change of quantity of PuO_2 emitted. It was concluded that the majority of the PuO_2 was emitted during the initial pressurisation.

Experiments were carried out with capillaries 50 µm diameter and 44 mm long, at pressures of 0, 34 and 68 bar. The results are shown in Table 2. The maximum leakage of PuO_2 was 0.04 µm.

There was some indication that the mean particle size of the leaked PuO_2 was smaller than that of the original powder (2.5 µm).

The following tentative conclusions were made for the PuO_2 work for both orifices and capillaries.

(i) The greatest emission occurred at zero pressure differential with the leak under the powder level

(ii) The PuO_2 emission was slightly pressure dependent with the leak above the powder level

(iii) PuO_2 emission through a 50µm capillary was similar to that through a 5 µm orifice at equal pressure differentials.

Sutter et al (1980)

This work was carried out to provide data for prediction of leaks of PuO_2 shipping containers based on experiments with UO_2.

A parametric study was conducted to indicate relationships between

pressure, aperture diameter, aperture length, time and leak orientation.

Orifice diameters form 20µm to 200 µm and capillaries of length 0.76 cm and 2.54 cm with diameters 50 µm to 200 µm were used. Investigations concluded that there was no powder "leak rate" and that the duration of the run had no effect on the amount of powder transmitted. The significance of diameter and pressure parameters was confirmed and the influence of diameter was shown to be more important than pressure.

Table 3 shows the results for the capillary tests with the capillary above and below powder level. This demonstrates the relationship of pressure and capillary size. As the pressure is increased the powder throughput is increased, but even at 68 bar differential pressure the 100µm capillary powder transmission is only 87.8 µg. At low pressure differentials (2 bar) the throughput of powder for all sizes is quite small. 100 µm is the smallest capillary size reported in the table of results, although 50 µm capillaries were manufactured albeit with great difficulty. The inference is that these small capillaries frequently blocked.

Leisher et al (1983)

This work, a co-operative effort between Sandia Laboratories and PNC Japan, was carried out to determine the correlation between solid powder and helium leakage through O-ring seals. The experimental work consisted of assembling a flanged joint with double O-ring seals, testing the seal to a known standard, and then introducing a powder (UO_2). The powder was pressurised and the leak of powder measured. The mean powder diameter was less than 1µm. Tests were carried out from -40°C to + 130°C with pressures up to 8 bar. The apparatus was vibrated throughout the test. The results are given in Table 4.

From the leakage flows of helium in the first part of the experiment equivalent capillary diameters were calculated. The test results given in Table 4 are the only results carried out. The very limited conclusions that can be drawn from these tests indicate:

There is no correlation between powder leakage and equivalent tube size. There is no correlation between powder leakage and helium leak rate.

Bild et al (1984)

This work is a continuation of the experiments carried out by Leisher et al using the same apparatus. To improve the technique Bild irradiated the UO_2 powder to convert a small fraction of the ^{238}U to ^{239}Np. The experiments used depleted UO_2 with 79.5% of the powder in the range 0.2 - 2.0 µm diameter. The results shown are shown in Table 5. These indicate that there is some leakage past the O-ring. This occurred mainly at low temperature (-40°C).

Anderson (1986)

During the development and licencing process for the Plutonium Air Transportable Model 2 (PAT-2) package, several different plutonium containment vessels were considered with varying seal types each representing a containment vessel in the PAT-2 package.

These consisted of:

A primary vessel with copper joint rings contained within a groove equipped with a knife-edge.

A secondary containment vessel sealed by means of a PTFE - sealed shouldered screw thread.

A laboratory type container sealed by means of a screw thread and PVC gasket in compression.

The containers were leak tested and then filled with UO_2 powder with a mean particle size of 5.5 µm. The results of the tests are given in Table 6. Anderson attempts to argue for long apertures rather than single holes. This could be a logical extension of the information but as the single capillary is the most pessimistic assumption I have not quoted Anderson's aperture data.

DISCUSSION

Parametric Survey

(i) **Pressure**

A review of US work (Owzarski 1980) using statistical analysis indicated that the particle transmission was proportional to $\sqrt{P}$ where P was the internal vessel pressure. Work in the UK (Curren and Bond 1980) indicated that the rate of powder transmission was a function of differential pressure. The review of Owzarski also found that rapid pressurisation transmitted significantly more powder than slow pressurisation.

(ii) **Time**

Working only with orifice plates Curren and Bond (1980) found that with a constant pressure differential across the plate the powder transmission was a function of time. The very limited work using actual O-ring (Leisher) indicated a very poor correlation between powder transmission and time. The extensive work on UO_2 and PuO_2 leakage in the US (Sutter 1980; Yesso 1980) indicated that the length of run had no effect on the transmission of powder.

(iii) **Orifice/Capillary Size**

All experiments found a correlation between orifice/capillary size and powder leakage. The larger the orifice/capillary the large the powder leakage. The review of US data (Owzarski 1980) using statistical methods found a strong dependence upon leak path cross-sectional area. One surprising conclusion of the work by Sutter (1980) was that capillaries transmitted slightly more powder than did orifices of the same diameter, but plugging occurred in 17% of capillary runs compared with 6% of the orifice runs.

(iv) **Leak Orientation**

Most work found that orifices and capillaries under the powder level blocked rapidly indicated by loss of gas flow. This must be considered along with the fact that the larger leaks of powder occurred with orifices and capillaries situated under the powder level. This can be explained by the fact that most of the US work indicated no relationship between powder loss and time, with the powder loss occurring upon initial pressurisation.

Capillary Information

A larger number of experiments have been conducted using orifices rather than capillaries because of the easier handling involved. The sizes used both in capillaries and orifices have been much larger in diameter compared with the size of interest, this is because at the smaller diameter leakage paths become blocked and results become erratic.

The experiments using actual capillaries were carried out by Yesso et al and Sutter et al.

Sutter et al used UO_2 powder with a mean particle diameter of 3.3µm and a range of capillaries from 50 µm to 250 µm diameter. The experimental results found no relationship between flow rate, length of run and transmission of powder. It was argued that powder transmission occurred at time of pressurisation. The smallest capillary used was 100 μm, and with this size the maximum powder leakage detected was 4 μg at a differential pressure of 7.0 bar.

Yesso et al used PuO_2 powder with a mean particle diameter of 2.5 μm with a capillary of 50 μm diameter. The length of capillary used was 44 mm. The experiments indicated a slight dependency between flow of powder and pressure, but no correlation with length of run. The maximum powder leakage detected was .04 μg. It was postulated that this powder leaked at pressurisation and no further powder flow occurred. If this figure is compared with the allowable Normal Conditions Of Transport limit ($A^2 \times 10^{-6}$ per hour) for plutonium oxide of .032 μg it can be deduced that ~ 50 μm diameter capillary will almost meet the requirements for Normal Conditions of Transport. Therefore a smaller capillary, say 25 μm, would certainly meet the requirements of Normal Transport.

Orifice Information

The experiments using orifices were carried out by Curren and Bond, Yesso et al and Sutter et al.

Curren and Bond used UO_2 powder with particles generally in the range 0.5 - 5.0 μm with a mass peak in the 1 -2 μm range. The 5 μm diameter orifice suffered blockage during the gas flow tests prior to the use of UO_2 powder. The 5 μm and 10 μm orifices blocked during the powder tests and blockage was encountered during some tests using the 25 μm diameter orifice. Sutter et al used UO_2 powder with a mean particle size of 3.3 μm. The minimum orifice diameter used was 20 μm at this condition the powder transmission varied from 8.0 to 60 μg at pressure differentials up to 68 bar. There was no correlation between powder transmission and time and it was assumed that most powder was transmitted at original pressurisation. Yesso et al used orifice down to 5 μm in diameter. They found no correlation with time and concluded that powder was transmitted at original pressurisation. At orifice diameter of 5 μm powder transmission was .01 μg at pressure differentials up to 68 bar.

If these figures are compared with the allowable Normal Conditions of Transport limit ($A^2 \times 10^{-6}$) for PuO_2 of .032 μg it can be seen that a 5 μm diameter orifice will meet the requirements for Normal Conditions of Transport.

Conclusions

Although there is not an overwhelming amount of experimentation the work that has been carried out indicated that micron size powders will not be transmitted through orifices less than 5 - 10 μ diameter and capillaries of 25 - 50 μ diameter. If the leak rates are calculated at Standardised Conditions (1 bar upstream .01 bar downstream and for capillaries a length of 1 cm) we get leak rates of approximately 5×10^{-3} bar $cm^3\ sec^{-1}$ for capillaries of 25 μm diameter and 4×10^{-3} bar $cm^3\ sec^{-1}$ for orifices of 5 μm diameter.

Allowing for a degree of pessimism it could be logical to state that powder would not pass through holes of any type with leak rates less than 1×10^{-3} bar $cm^3\ sec^{-1}$ standard leak rate.

References

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Table 1 Transmission of Powder Through Orifices (Curren and Bond 1980)

Orifice Size µm	Release of UO_2 g/hr x 10^{-3}			
	Max	Min	Mean	Std. Dev
200	1.408	0.894	1.13	0.16
100	0.321	0.055	0.16	0.08
50	0.156	0.028	0.096	0.050
25	0.060	0.002	0.018	0.016
10	PLUGGING			
5	PLUGGING			

Table 2A Transmission of PuO_2 Powder Through a 50μ Diameter Capillary (Yesso et al 1989)

Pressure	Release of PuO_2 gas x 10^{-6}	
bar	Max	Min
68	0.0129	0.001
34	0.009	0.001
0	0.04	0.000

Table 2B Transmission of PuO_2 Powder Through Orifices (Yesso et al 1980)

Orifice Size μm	Average Release of PuO_2 g x 10^{-6}
5	0.011
8	0.011
10	0.003
20	0.008

Table 3A Transmission of UO_2 Powder Through Capillaries (Sutter et al 1980)

Capillary Diameter (μm)	Maximum Powder Transmitted g x 10^{-6}			
	68 bar	34 bar	6.8 bar	2 bar
250	15,800	11,500	188	217
200	2,690	3,340	1.04	13.0
150	1,280	959	320	18.0
100	87.8	4.07	-	4.96

Table 3B Transmission of Uo_2 Powder Through Orifices (Sutter et al 1980)

Orifice Diameter (μm)	Maximum Powder Transmitted g x 10^{-6}			
	68 bar	34 bar	6.8 bar	2 bar
200	11,500	3,480	109	722
110	1,330	602	26	105
36	1,350	32	9	44
20	36	60	8	11

Table 4 Transmission Of UO_2 Powder Through O-ring Seals (Leisher et al 1983)

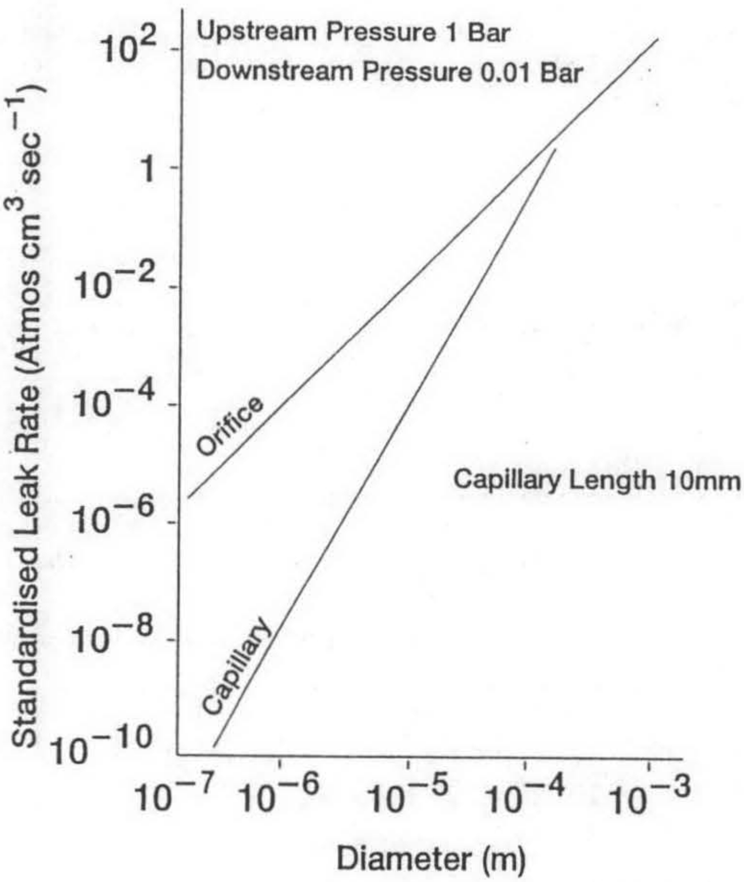
Temperature (°C)	Pressure Range (bar)	Time (hours)	Maximum Powder Transmitted g x 10^{-6}
-40	5	3	0.25
27	1 - 8	3	0.16
130	1 - 8	100 - 180	0.13

Table 5 Transmission Of UO_2 Powder Through O-ring Seals (Bild et al 1984)

Temperature (°C)	Pressure Range (bar)	Time (hours)	Maximum Powder Transmitted g x 10^{-6}
-40	130	168	0.03
26	0 - 1	3	0.065
130	1 - 7	3	0.074

Table 6 Transmission Of UO_2 Powder Through Various Seals (Anderson 1986)

Seal Design	Gas Leakage Rate (bar cm^3/sec)	Calculated Capillary dia (μm)	Powder Transmitted g x 10^{-6}
Bolted Copper Gasket	2×10^{-5}	6.9	nil ($<10^{-2}$)
Shouldered Screw Thread	2×10^{-9}	0.5	nil ($<10^{-2}$)
PVC Compression	1×10^{-3}	11.0	nil ($<10^{-2}$)



Upstream Pressure 1 Bar
Downstream Pressure 0.01 Bar
Orifice
Capillary
Capillary Length 10mm
Standardised Leak Rate (Atmos cm^3 sec^{-1})
10^2
1
10^{-2}
10^{-4}
10^{-6}
10^{-8}
10^{-10}
10^{-7}
10^{-6}
10^{-5}
10^{-4}
10^{-3}
Diameter (m)

FIGURE 1