Leak-Testing to Demonstrate Retention of Particulates

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INTRODUCTION

Packages designed to contain radioactive materials for transport or storage must be shown to be leak-tight to certain standards. Tests are performed to demonstrate compliance with these standards before acceptance for despatch or storage. The standards depend on the form of the radioactive contents and the potential hazards from the release of the isotopes concerned. Fluid (liquid or gas) leak rates can readily be assessed by evaluating flow rates from measured pressure drops. Solid matter will normally only leak in the form of particles entrained in the fluid stream but assessment of particle concentrations is not practical as an operational procedure.

This problem is avoided by achieving such low leak rates that solid particles cannot pass through the largest conceivable leak path. Further difficulties are encountered in following this route, however. It may be difficult to achieve the desired low leak rate; measurement to demonstrate achievement may require costly and sensitive equipment and may be time-consuming; the interpretation of hole size and particle size distributions may leave some uncertainty.

The usual assumption is that if the assessed particle size distribution includes a significant proportion of particles of size less than the maximum hole size, then particles can leak. Practical observations suggest that if there is a significant proportion of particles with size larger than this largest hole, then blockage mechanisms will prevent loss of particles (eg Curren and Bond, 1980).

Other assumptions are generally very pessimistic. The total leakage is attributed to a single leak path, maximising the size of the hole. An aerosol is assumed to exist within the container when there is rarely a mechanism to entrain particles in the air. An internal driving pressure is assumed for assessment purposes while, in the absence of a volatile liquid, there is little scope for such excess pressure. These pessimisms result in gross overestimates of particulate leakage even in accident conditions. It is therefore important to identify limiting features such as blockage mechanisms to reduce these pessimisms.

A programme of experimental measurements has started at Winfrith to determine the requirements for hole blockage. So far this has involved spherical particles and small capillaries to provide practical data. This will eventually be extended to orifices and non-spherical particles to provide an understanding of the mechanisms.

Previous experiments (eg Curren and Bond, 1980) have measured particle leak-rates through orifices but, where blockage occurred, the experiment was deemed to have failed. This programme of work is aimed at defining the regimes where blockage or other mechanisms will prevent leakage.

Practical Leak-Rate Measurements

Various methods of leak-testing are described in an Atomic Energy Code of Practice (AECP 1068, 1988). The commonly used interspace pressure measurement requires a double seal arrangement with access to the interspace (Figure 1). This volume is pressurised with a gas and connected to a sensitive pressure gauge. The rate of loss of pressure is related to the leak rate from the interspace volume.

A standardised leak rate (SLR) is defined by normalising the results to reference conditions of 25°C air leaking from an upstream pressure of 2.0 bar (200 kPa) to a downstream pressure of 1.0 bar (100 kPa). The interspace can be pressurised conveniently to one atmosphere gauge unless greater sensitivity is needed when higher pressures may be used. The SLR is measured in units of $bar.ml.s^{-1}$ (bar.cc/s) or 0.1 $Pa.m^3.s^{-1}$ in SI units.

The leak-rate is given by

$$L = \frac{VTo}{HT} (P_1 - P_2) \ Pa.m^3.s^{-1} \qquad (1)$$

where V is the interspace volume (m^3)

T_o is the reference temperature (K) (25°C = 298K)

T is the gas (or ambient) temperature (K)

P_1 is the initial pressure (Pa)

P_2 is the final pressure (Pa)

and H is the test duration (s)

If the initial differential pressure is 100kPa and the interspace volume is 10 ml (10^{-5} m^3), the rate of change of pressure for an SLR of 10^{-6} $Pa.m^3.s^{-1}$ is

$$\frac{dP}{dt} = 10^{-1} \ Pa.s^{-1} = 10^{-6} \ \delta P \ s^{-1} = 3.6 \times 10^{-3} \ \delta P \text{ per hour.}$$

The pressure thus declines at less than 0.5% per hour and sensitive and stable measurement techniques are required for this leak rate. The leak-rate chosen (10^{-6} $Pa.m^3.s^{-1}$) is generally accepted as sufficiently small to demonstrate that no hole size exists large enough to allow the passage of particles.

With an instrument full range of 500 kPa, a test period of 30 minutes is needed to achieve 0.02% of this range, the limit of discrimination of a relatively sensitive instrument. Extending the measurement time is inconvenient and introduces

additional problems of temperature drift in the absence of a carefully controlled environment. Heat generated by the contents of a package further complicates the control of temperature.

Such measurements are achievable with small packages in a controlled environment and with a sensitive instrument. Practical applications to an industrial environment will reduce the guaranteed accuracy by at least an order of magnitude. Even with the acceptance that an SLR of 10^{-6} pa.m^3.s^{-1} (10^{-5} bar.cc/sec) will not allow particulate leakage, the measurement technique is only marginally acceptable. Numerical analyses of commercially available instruments shows that typical measurements of the type defined above are uncertain to about $\pm$25%. Larger interspace volumes reduce the accuracy further; to $\pm$40% at 20 ml and to $\pm$60% at 30 ml.

The interspace volume is a further source of uncertainty but can be calibrated at the time of measurement by introducing a standard volume into the system and measuring the step change in pressure. This technique has been integrated into a computer controlled instrument with multiple measurement channels. Highly sensitive and stable transducers are intercompared automatically, immediately before each measurement and the results analysed on-line with individual calibration curves.

Maximum Hole Size

The use of a differential pressure (to ambient air) of 100 kPa or more will ensure that sonic flow conditions exist in an orifice-type hole. Sonic, critical or choked flow will occur if the pressure ratio across an orifice is less than 0.53 (Mishima et al, 1980). It is not obvious that these conditions exist with a capillary as the pressure drop due to friction along the length could reduce the effective pressure ratio below this figure. The following analysis provides an estimate of equivalent orifice size however.

A gas leakage flow rate of V m^3.s^{-1} requires a sonic choke area of A m^3 given by:

$$A = \frac{V}{c} \text{ m}^2$$

where c is the sonic velocity (349 m.s^{-1} in nitrogen or 346 m.s^{-1} in dry air at 25°C). The diameter of a circular orifice of this area is:

$$D = 2\left\{\frac{A}{\pi}\right\}^{\frac{1}{2}} = 2\left\{\frac{V}{\pi c}\right\}^{\frac{1}{2}} \quad \text{m}$$

The choke area is generally smaller than the orifice area due to the vena contracta effect. A discharge coefficient, generally between 0.6 and 1.0, is applied to the flow area to allow for this. Choosing the smallest coefficient will yield the largest orifice area for a given flow, so the orifice diameter with air as the fluid becomes:

$$D = 2\left\{\frac{V}{0.6\pi c}\right\}^{\frac{1}{2}} = 0.0783\ V^{\frac{1}{2}} \text{ m}$$

The relationship of hole diameter to leak-rate is given by the following tabulation for air at 25°C (ie SLR conditions) with the assumption of sonic flow (Bomelburg, 1977).

(Pa.m^3.s^{-1})	10^{-7}	10^{-6}	10^{-5}	10^{-4}
(bar.cc.s^{-1})	10^{-6}	10^{-5}	10^{-4}	10^{-3}
(m^3.s^{-1})	10^{-12}	10^{-11}	10^{-10}	10^{-9}
Orifice Diameter (μm)	0.08	0.25	0.78	2.48

Thus, for a powder with little or no particles below 1 μm in diameter, a leak rate of up to 1.6 x 10^{-5} Pa.m^3.s^{-1} can be tolerated without particles leaking. Such a conclusion only applies to circular orifices and spherical particles. Non-spherical particles will have more difficulty passing through such a hole than a sphere with a similar aerodynamic diameter. Particles will have even more difficulty passing through capillaries of the same diameter as an orifice.

It is also probable that particles will have more difficulty leaking through a capillary with the same leakage characteristics. This may have great length to diameter ratio and be very tortuous. The frictional pressure drop may reduce the gas flow below the critical flow rate, in which case the simple relationship between SLR flow and hole diameter is lost. The single hole diameter may be much larger than the particle size but blockage will still occur because of the tortuous nature of the leak path. No experimental evidence has been found for particle penetration through capillaries or orifices of less than 25 μm diameter. This experimental programme is aimed at demonstrating and investigating the blockage mechanisms.

Particle Size

The particles of most interest for container leakage assessments are derived from fuel and contain either fission products or plutonium. They are likely to be in oxide form. Assessments of PuO_2 and UO_2 particle sizes (unpublished) show that, although significant proportions by weight may have aerodynamic diameters less than 1 μm, larger size particles are always present in relatively high proportions. Even U_3O_8 particles with 6% by weight less than 1 μm in diameter (Iwasaki et al, 1968) have a large proportion of particles larger than 5 μm.

Median diameters are thus likely to be about 5 μm with a significant proportion above 10 μm and there will be plenty of particles available to block hole sizes significantly larger than 10 μm diameter. Experimental evidence is required to show that orifices larger than 25 μm will block and that SLR flows of 0.01 Pa.m^2.s^{-1} can be tolerated for test purposes.

Aerosol Experiments

The Aerosol Laboratory at Winfrith has embarked on a series of experiments aimed at demonstrating that blockage mechanisms prevent both gas and solid particulate leakage with holes of the order of 25 to 100 μm in diameter. Both orifices and capillaries will be tested to provide an understanding of the physical mechanisms. The work is scheduled for 1989/90 so only early experience can be reported.

The apparatus is illustrated in Figure 2. It comprises:

(a) a fluidised bed aerosol generator,

(b) an aerosol chamber, 9 litres capacity at up to 150 kPa above atmospheric pressure,

(c) a particle collection chamber at atmospheric pressure downstream of the test capillary, and

(d) an exhaust system.

The fluidising gas is filtered air and more air is supplied to the aerosol chamber to pressurise and dilute the aerosol and to the collection chamber to sweep the capillary leakage to the measuring devices. This 5 litre per minute flow to the collection chamber avoids particle fallout at slow gas flows downstream of the capillary. The exhaust vessel allows the pressure to be controlled and provides a means of measuring aerosol concentration from an up-stream sample.

An optical particle counter (OPC) is used to measure aerosol concentrations in the exhaust vessel. An aerosol particle size analyser (APS) measures aerosol characteristics downstream of the capillary and a membrane filter with 0.2 μm pores collects particles for off-line analysis.

The fluidised bed comprises about 200 g of 180 μm bronze spheres and is used to entrain glass microspheres as an aerosol, ensuring the break-up of natural agglomerations. The aerosol material comprises about 10g of glass microspheres the size distribution of which is illustrated in Figure 3. This distribution (unimodal and approximately log-normal) conforms with the supplier's specification and was measured by the APS without a capillary fitted. The 15μm maximum aero-dynamic diameter corresponds to a geometrical diameter of about 9.5μm.

The fluidised bed is supplied with between 8 litres per minute (at atmospheric pressure, 100kPa) and 32 litres per minute (at 200 kPa reduced to N.P.T.) to achieve the necessary "boiling" action.

The OPC is used to confirm that the aerosol concentration has stabilised but cannot measure concentrations in the chamber (it functions only at atmospheric pressure) due to losses in the exhaust system and the instrument. About 90 minutes settling time is required to obtain stability to within $\pm$20% of the average particle density after the initial high concentrations on start-up of the fluidised bed flow. Typical concentrations of 20 to 60 particles per cubic centimetre are used, but flow is allowed through the capillary only after stability has been achieved.

Results of Initial Trials

Only scoping tests have so far been completed with glass capillaries of 20 μm bore and 50 mm in length. Each run is performed with a new capillary after microscopic examination of the entrance aperture. Used capillaries are similarly examined for deposited microspheres but no evidence of deposition has been found so far.

A method of characterising the capillary, by measuring air flow in the absence of an aerosol, is under development but has yet to be applied to the test capillaries. This will be performed in-situ by evacuating the collection chamber and measuring the rate of increase in pressure from flow through the orifice. This will yield the leak-rate as a function of differential pressure in much the same way as leak-testing is performed.

The anticipated null result means that the sensitive detection equipment must be free of dust and other particles which could be entrained into the air flow. In spite of returning the APS to the suppliers for cleaning, a 60 minute test with airflow directly from a 0.2 μm HEPA filter yielded a count of 80 particles. This corresponds to a very low concentration of less than 10^{-3} particles per cubic centimetre (approximating to clean room conditions) and is expected to decline as particle sources within the APS become depleted by extended use.

Figure 4 shows the particle size distribution measured by the APS downstream of the 20 μm capillary with a 100 kPa differential pressure. The distribution is quite different from the source aerosol (Figure 3) but similar to the "background" distribution of Figure 5.

This suggests strongly that none of the aerosol particles pass through the capillary, a conclusion confirmed by examination of the 0.2 μm membrane filter replacing the APS. This examination uses a scanning electron microscope (SEM) coupled to an automated image analyser to examine the whole surface of the filter at low magnification. No glass particles have been seen following two 60 minute sampling periods. This off-line technique is more time consuming than the on-line APS method but it provides unambiguous results. If and when particles are detected (eg with larger capillaries), energy dispersive X-ray analysis (EDS) can be used to confirm that the composition of the particles corresponds with that of the glass microsphere source.

Conclusions

The preliminary results from the experiment confirm the expectation that no particles will pass through a 20 μm capillary even when the majority of spherical particles are less than 2 μm in diameter and virtually all less than the hole diameter. This suggests that blockage mechanisms are effective in preventing solid particulate flow and possibly reducing fluid flow. However, no particles have yet been observed at the entrance to the capillary or otherwise blocking the hole. Alternative mechanisms may be helping avoid particle loss even in the early transients before "blockage" is effective.

These conclusions must be confirmed by more extensive testing with larger capillaries and orifices and by characterising the holes with gas flow measurements. However, if confirmed, the results will provide the basis for a much simplified leak-test procedure where the release of solids from a container is unacceptable or must be restricted to low release rates.

An understanding of blockage mechanisms, with spherical and other shapes of particle, will provide the basis for simplified test procedures. Significant savings of operational time and test instrumentation will result.

References

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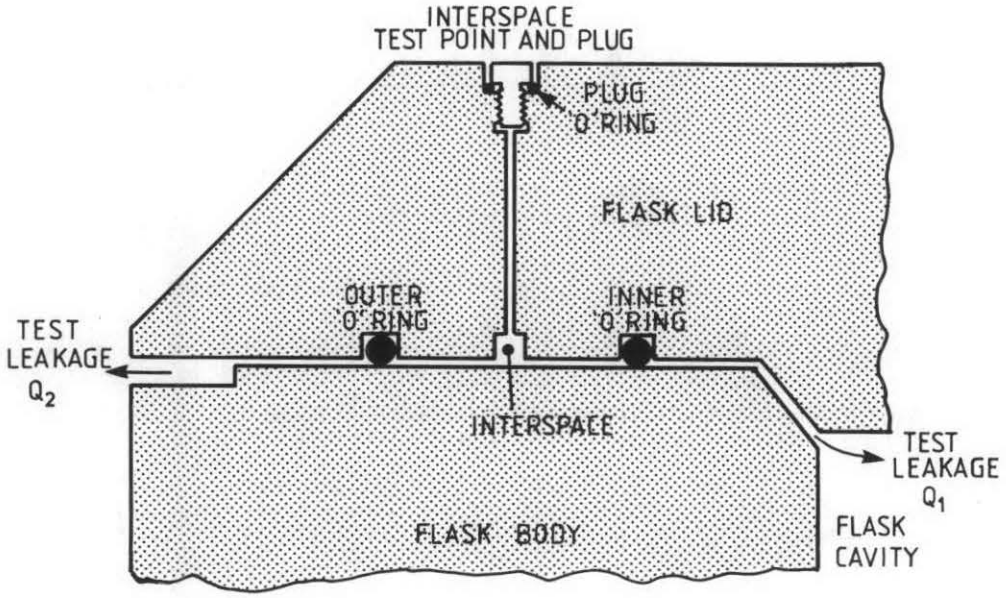

FIG.1 DOUBLE 'O' RING SEAL GEOMETRY

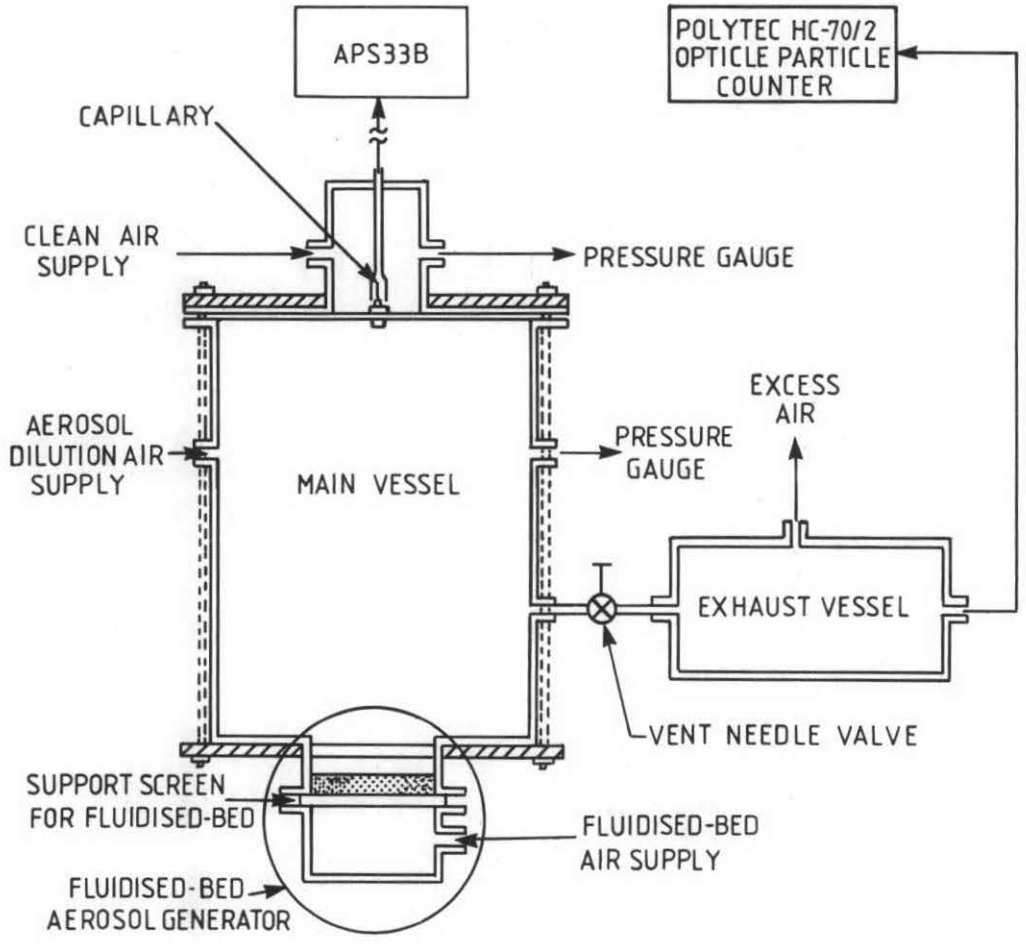

FIG.2 CAPILLARY LEAK TEST APPARATUS

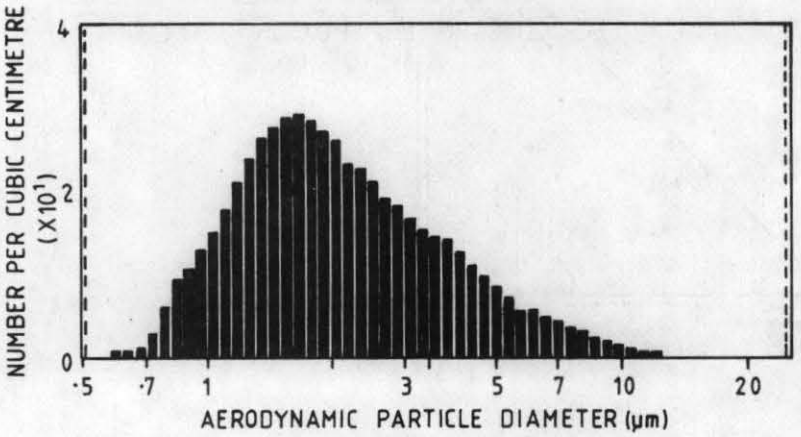

FIG.3. GLASS MICROSPHERE SIZE DISTRIBUTION

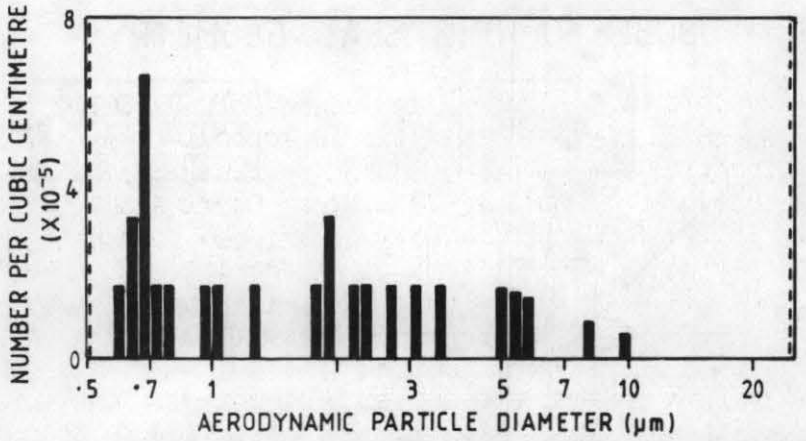

FIG.4 PARTICLE SIZE DISTRIBUTION DOWNSTREAM OF A 20 μm CAPILLARY

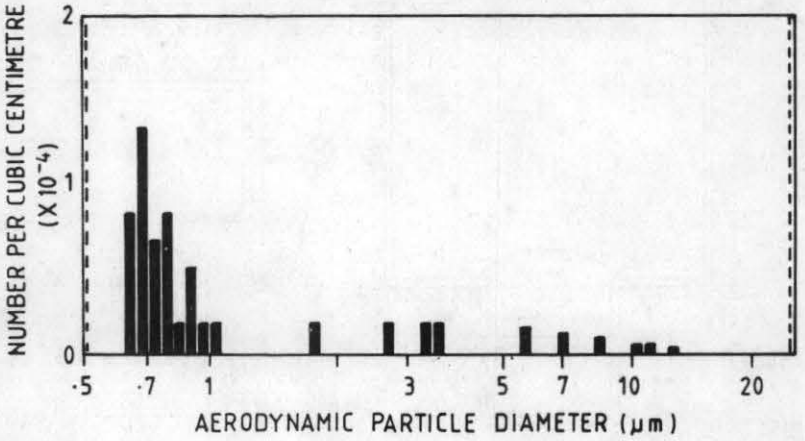

FIG.5 BACKGROUND PARTICLE SIZE DISTRIBUTION