The Transport of Micron-Sized Particles Through Short Capillaries

J.P. Mitchell[1], D.A.V. Morton[1] and J. Higson[2]

[1] Aerosol Science Centre, AEA Environment and Energy, Winfrith Technology Centre, Dorchester, Dorset, DT2 8DH, UK.

[2] AEA Decommissioning and Radwaste, Chadwick House, Risley, Warrington, Cheshire, WA3 6AT, UK.

INTRODUCTION

The leak testing of containers for radioactive material is a time-consuming process, particularly since very low gas leakage rates have to be detected in order to demonstrate compliance with the regulations (IAEA 1985). The assessment of airborne particulate (aerosol) releases across seals is avoided by achieving such low leakage rates that the assumption can be made that particles do not pass through the widest conceivable pathway. In practical terms, a leakage rate of 10^{-6} Pa m^3s^{-1} is equivalent to a single orifice of 0.25 μm diameter (Burgess et al., 1989). It is desirable to test containers to an acceptance criterion that is equivalent to a leakage pathway at least ten times this size, since such leakage can be detected using simple-to-use pressure rise or fall methods, and corrections for temperature variations during the much shorter test duration can be reduced or even avoided altogether.

The penetration of 2 to 20 μm diameter glass microspheres has been investigated in a systematic study of model leak paths, consisting of straight glass capillaries with lengths ranging from 5 to 50 mm and bores varying from 2 to 80 μm. The glass particles were suspended as an aerosol using a pressurisable fluidised-bed and the particle penetration rate was measured through each capillary by means of either a single-particle counter or by counting the particles collected onto a membrane filter using a scanning electron microscope (Mitchell et al., 1990). Further tests were undertaken to investigate the penetration of compact, non-spherical cerium oxide particles of similar size to the glass microspheres. Every test was carried out with the upstream pressure fixed at 100 kPa and the outlet of the capillary at ambient pressure. The measurements were therefore made under conditions quite similar to those encountered during the leak-testing of type-B RAM transport containers.

AIR LEAKAGE BEHAVIOUR

All of the experiments were undertaken with short length borosilicate or quartz glass capillaries The leakage rate of particle-free air was established by measuring the rate of increase in pressure of a partly evacuated chamber located downstream of the capillary

(Mitchell et al., 1990). The equipment could detect air leakage rates as low as 10^{-7} Pa m^3 s^{-1} (Figure 1). The standard leakage rate (SLR Pa m^3 s^{-1}) was calculated from:

$$SLR = \frac{VT_0}{HT_1}[(P_1 - P_2)]\left[\frac{\mu_a\left(P_s^2 - P_0^2\right)}{\mu_s\left(P_u^2 - P_d^2\right)}\right] \qquad (1)$$

where V (m^3) is the volume of the evacuated chamber, T_0 and μ_s are the reference temperature (298 K) and air viscosity (1.85×10^{-5} kg $m^{-1}s^{-1}$) respectively, T_1 and μ_a are the measured air temperature and viscosity, P_1 and P_2 are the initial and final pressures of the chamber respectively (kPa), and H (s) is the duration of the test. The final term normalises the measured leakage rate to reference conditions, where P_s, P_0, P_u and P_d are the reference pressure (100 kPa), zero pressure, and measured capillary upstream and downstream pressures respectively.

Air leakage expressed as SLR values are summarised in Table 1; these results are the average of the leakage rates measured before and after aerosol testing. It is notable that with very few exceptions, the air leakage rate after exposure to aerosol agreed closely with the equivalent value before the test, indicating that plugging of the capillaries with either glass microspheres or cerium oxide particles did not occur to a significant extent. In most cases, SLR values agreed well with corresponding air leakage rates predicted assuming viscous laminar flow (Santeler, 1984). In a few cases, more notably with the larger bore capillaries, measured leakage rates were significantly greater than the predicted values. There is no obvious explanation for these discrepancies other than parasitic leakages within the test equipment, although care was taken to minimise such effects.

AEROSOL PENETRATION BEHAVIOUR

In the second part of the study, the aerosol penetration rate (particles h^{-1}) was measured for each capillary using a purpose-built pressure chamber-fluidised-bed aerosol generator (Figure 2), whose detailed operation is described elsewhere (Mitchell et al., 1990). Most of the measurements were made using non-porous glass microspheres ranging in size from about 0.7 to 15 µm aerodynamic diameter (mass median aerodynamic diameter of 7 µm (Figure 3a)). A few measurements were also made with cerium oxide particles ranging from 0.5 to 5 µm aerodynamic diameter (mass median aerodynamic diameter close to 3 µm (Figure 3b)).

The total particle concentration in the pressure chamber was monitored by membrane filter collection. Particles that leaked through the capillary on test were either sampled using an APS33B Aerodynamic Particle Sizer (TSI Inc., St Paul, MN, USA) or collected on a membrane filter for microscopy-counting. Precautions were taken to ensure that particle leakage occurred only through the capillary, and that the particles penetrating the leak path were not contaminated with ambient airborne particles.

Glass microsphere penetration rates as a function of capillary bore (constant length of 50 mm) and capillary length (constant bore of 20 μm) are shown in Figures 4a and 4b respectively, and are summarised in Table 2, together with similar penetration rate data for the cerium oxide particles. All penetration rates are normalised assuming a fixed upstream aerosol concentration of 70 particles cm^{-3}. Particle penetration was strongly dependent on capillary bore; no penetration was detected with long capillaries when they were 20 μm bore or finer. It is notable that penetration rates determined by microscopy-counting agreed with data obtained directly using the APS33B. As expected, shorter capillaries showed greater particle leakage. Significant penetration rates were observed for 20 μm bore capillaries shorter than 30 mm in length. Surprisingly, size-selective sampling was seldom observed, except with capillaries that reduced particle leakage to near zero (Mitchell et al., 1990). Plugging of the capillaries rarely occurred, in contrast with previously reported work (Sutter et al., 1980).

Particle penetration rates for both glass and cerium oxide particles, were plotted as a function of air leakage rate (Figure 5), and showed a near linear dependence when these variables were scaled logarithmically. It is notable that the particle penetration rates measured for the cerium oxide particles were similar to equivalent data obtained with glass microspheres, despite the differences in the size distributions of the two types of aerosol and the shapes of the particles.

CONCLUSIONS

The process of aerosol penetration through ultra-fine leak paths takes place in two stages; the particles are initially 'sampled' at the entrance to the capillary, and are then transported through the capillary. Previous studies (Mitchell et al., 1990) have shown that all except the finest bore capillaries act in the same way as calm-air samplers. Preliminary theoretical assessments have indicated that some particle deposition should occur to the walls of the capillary during the transport phase of the leakage process. However, the rate of such deposition must be slow in view of the observed lack of plugging combined with the marked absence of size-selective leakage behaviour. This finding is further supported by the similar penetration behaviour of the spherical and compact non-spherical particles.

It is notable that the SLRs of capillaries with nominal entrance bores of 5 and 10 μm were close to the IAEA-recommended limiting leakage rate for type B radioactive packages designed to carry solid particulate material of 10^{-6} Pa m^3 s^{-1} (IAEA, 1985), and below which no particles would be expected to leak. Their lengths were comparable with the length across the current design of elastomer seal, assuming a single breach in seal integrity as a worst-case condition. It is therefore interesting to note that the glass particle penetration rates were always close to or lower than the lower detection limit of the APS33B (50 particles h^{-1}), supporting the viewpoint that aerosol penetration (at least for micron-sized particles) does not occur at the limit suggested by the IAEA. The penetration data for micron-sized cerium oxide particles are very similar to equivalent results for glass microspheres, suggesting that particle density and shape do not affect the penetration behaviour, within the size range studied.

The present experiments have confirmed that testing to the current standard leakage rate of

10^{-6} Pa m^3 s^{-1} is unlikely to result in the penetration of micron-sized aerosol particles. Nevertheless, reductions in the limit (by more than one-half an order of magnitude) may result in significant amounts of particle leakage. Work is under way to investigate the influence of a driving pressure on aerosol leakage (all data so far have been obtained with a pressure drop of 100 kPa across the capillary), as well as determining whether the behaviour of sub-micron particles is substantially different from that observed with the larger particles.

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Table 1 Standard Leakage Rates (SLRs) for Air Through Representative Capillaries

Capillary Bore (μm)	Capillary Length (mm)	Aerosol Type	Predicted Leakage Rate (Pa m^3s^{-1})	Measured Leakage Rate (Pa m^3s^{-1})
3.5	6.1	G	1.7×10^{-7}	1.8×10^{-6}
11.3	4.7	G	2.3×10^{-5}	3.5×10^{-5}
11.3	10.6	G	1.0×10^{-5}	1.1×10^{-5}
15.0	15.8	C	2.2×10^{-5}	2.3×10^{-5}
19.9	4.5	G	2.3×10^{-4}	2.8×10^{-4}
19.9	7.2	G	1.4×10^{-4}	1.7×10^{-4}
20.4	10.1	G	1.0×10^{-4}	1.6×10^{-4}
19.9	20.7	G	4.8×10^{-5}	4.6×10^{-5}
19.7	25.2	G	3.8×10^{-5}	5.0×10^{-5}
25.4	25.4	C	1.1×10^{-4}	1.2×10^{-4}
30.1	20.1	C	2.7×10^{-4}	3.6×10^{-4}
45.0	19.9	C	1.4×10^{-3}	3.1×10^{-3}
50.2	15.8	C	2.7×10^{-3}	6.0×10^{-3}
59.8	19.8	C	4.4×10^{-3}	7.0×10^{-3}
69.0	20.4	C	7.5×10^{-3}	1.1×10^{-2}
80.6	20.4	C	1.4×10^{-2}	1.6×10^{-2}

G = glass microspheres C = cerium oxide particles

Table 2 Glass and Cerium Oxide Particle Penetration Rates for Selected Capillaries

Nominal Capillary Bore (μm)	Nominal Capillary Length (μm)	Aerosol Type	Particle Penetration Rate (particles h^{-1})	
			microscopy	APS33B
20	5	G	NM	4400
20	7	G	NM	275
20	10	G	NM	1900
20	25	G	NM	75
20	30	G	840	50
4	6	G	NM	<50
11	11	G	NM	<50
20	50	G	<10	<50
35	50	G	25000	11100
45	50	G	13400	18700
60	50	G	NM	92500
70	50	G	18700	174000
15	16	C	45	NM
25	25	C	110	NM
30	20	C	6700	NM
45	20	C	84000	NM
50	16	C	13500	NM
70	20	C	342000	NM
80	20	C	128000	NM

G = glass microspheres C = cerium oxide particles

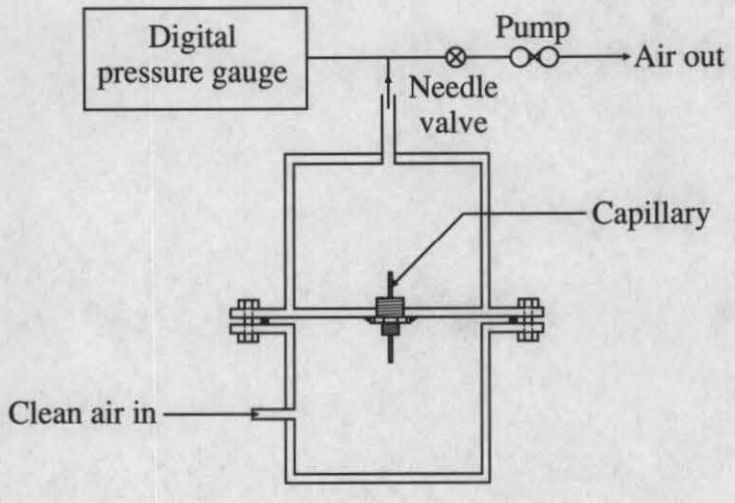


Figure 1 Apparatus for the Determination of Capillary Leak Rates

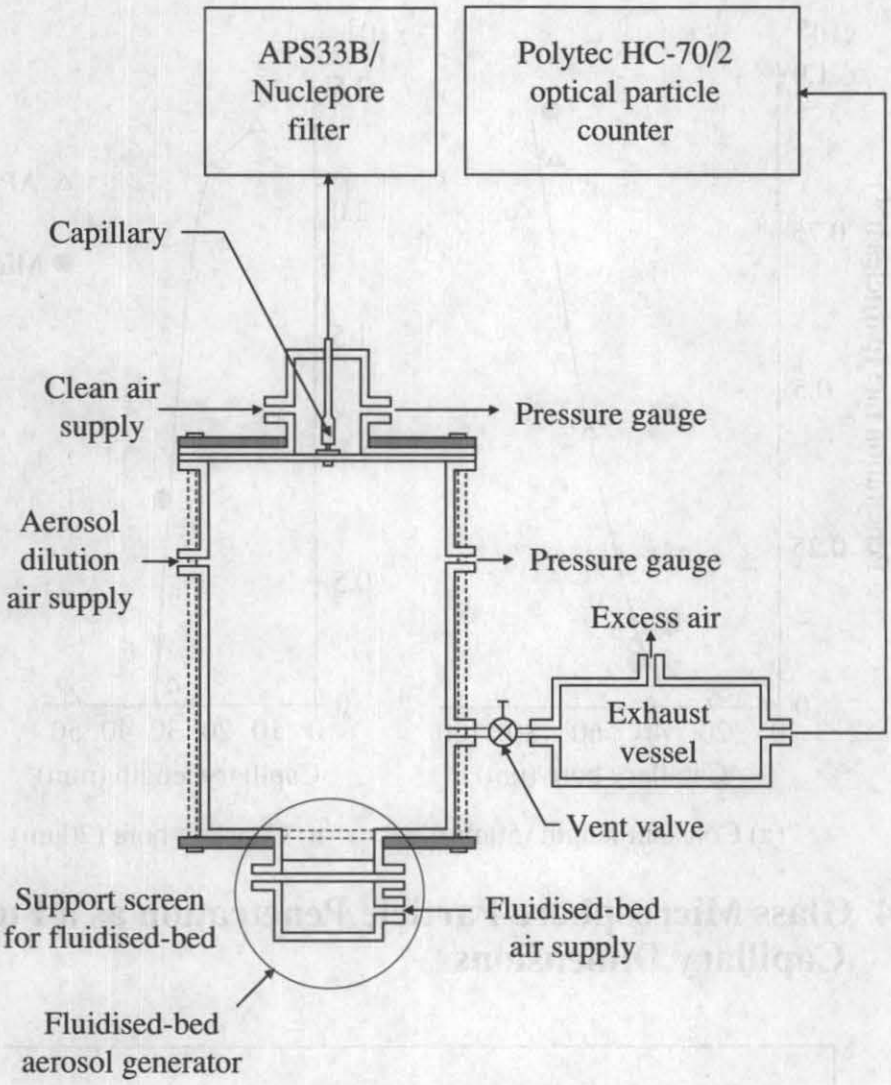
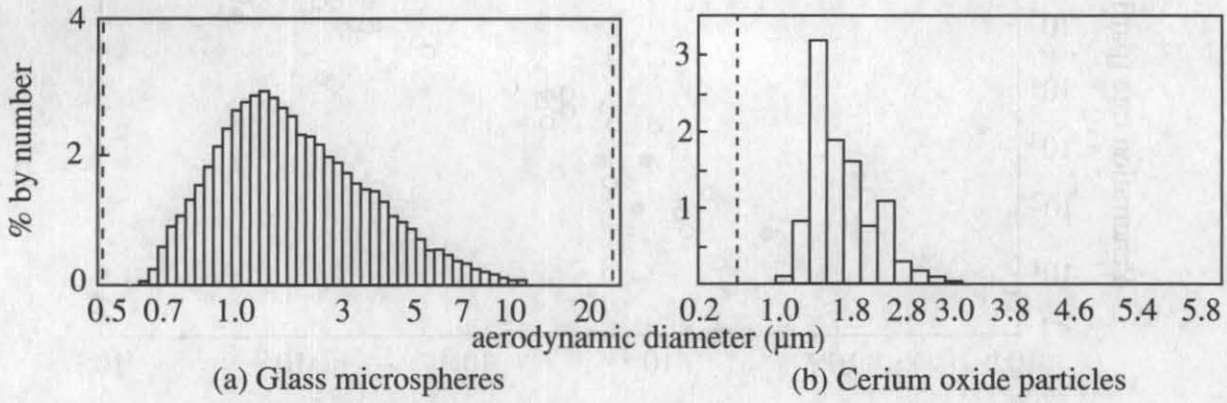


Figure 2 Aerosol Generator and Capillary Test Apparatus



(a) Glass microspheres (b) Cerium oxide particles

Figure 3 APS33B-Measured Size Distributions for Glass Microspheres and Cerium Oxide Particles Presented to the Capillaries

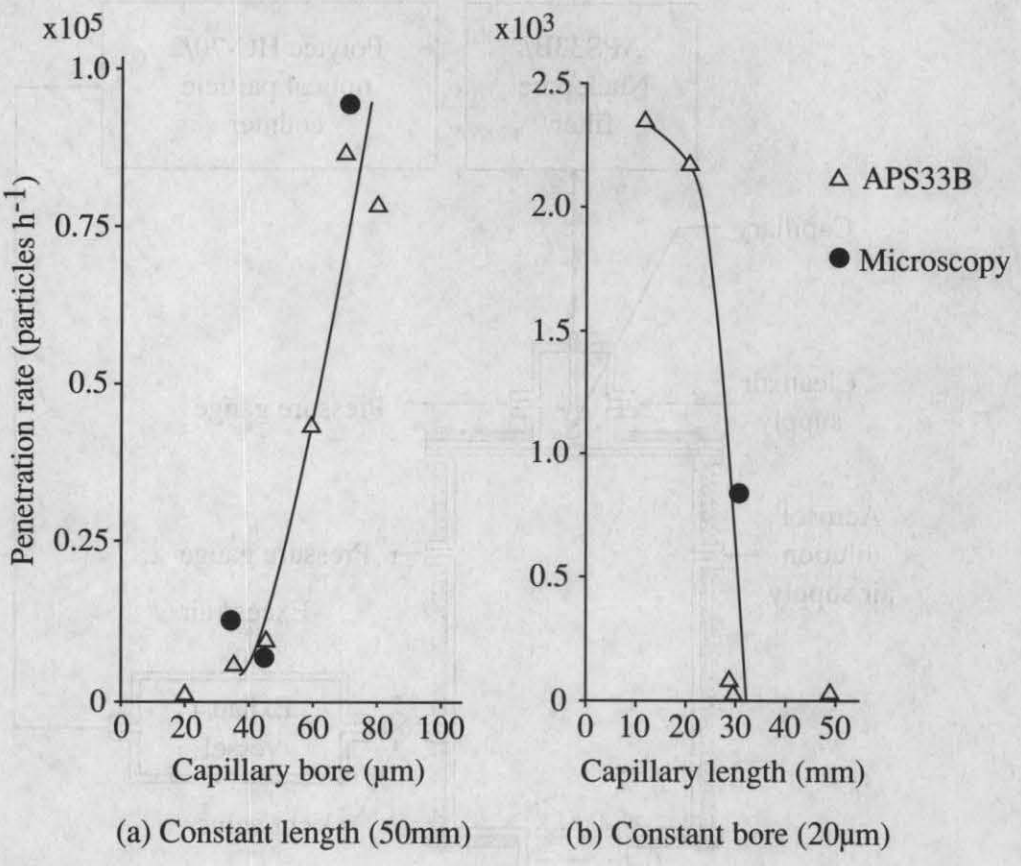


Figure 4 Glass Microsphere Particle Penetration as a Function of Capillary Dimensions

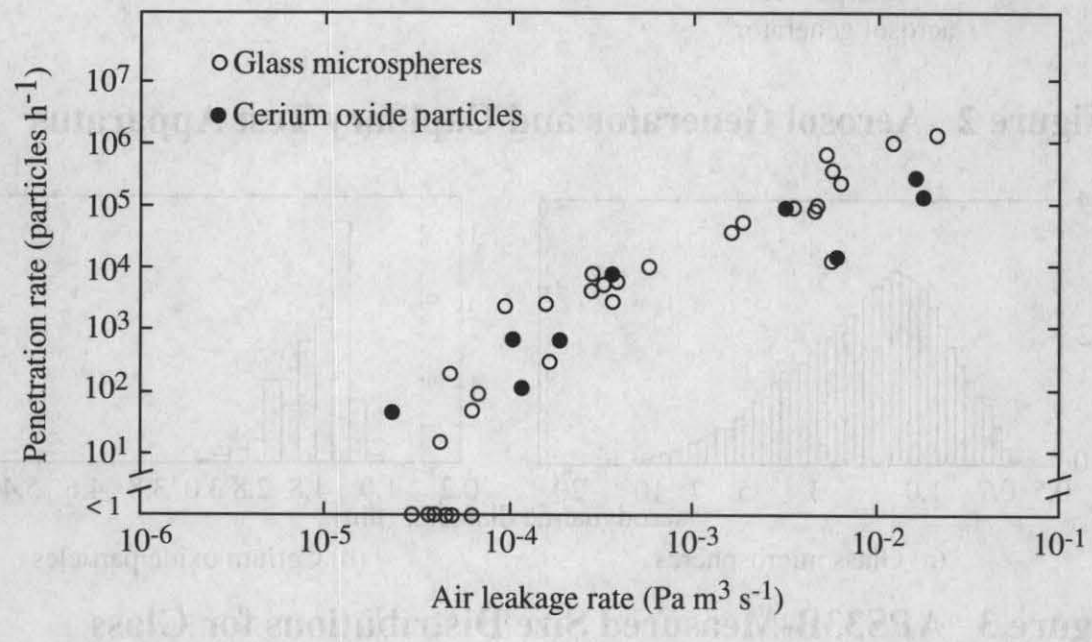


Figure 5 Particle Penetration as a Function of Air Leakage Rate

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