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AEROSOL PENETRATION THROUGH CAPILLARIES AND LEAKS: THEORY

C. F. Clement

Intra Information Technologies, Chiltern House, 45 Station Road, Henley-on-Thames, Oxfordshire RG9 1AT, U.K.

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Abstract—A general outline theory is constructed of aerosol penetration through small capillaries in which the gas flow is mainly laminar. The rate of particle transfer through a capillary is linearly related to the standard leak rate (SLR) of the capillary when no particle sampling or deposition losses occur, but the experiments of Mitchell *et al.* show that particle penetration falls sharply at $SLR \approx 10^{-4} \text{ Pa m}^3 \text{ s}^{-1}$. The expected scaling of this possible cutoff in penetration with capillary dimensions and pressure changes is discussed, including the use of a laminar Stokes number to represent entrainment effects at the capillary entrance.

A rigorous proof is given that, provided aerosol deposits remain in position and do not break off, the total aerosol transmission through a leak should be independent of initial aerosol concentration as long as it is not high enough to change the flow. If deposition occurs by impaction in a limited region near the capillary entrance, a scaling relation is postulated for this behaviour which relates the capillary length and external pressures. Experimental evidence supports the relationship, but does not give decisive results on the scaling of a penetration cutoff with changes to the capillary length and radius.

Experimental results have been obtained by Burton *et al.* for aerosol penetration in capillary flows produced by varying the upstream pressure, and are shown to be consistent with an initial penetration efficiency close to unity in most cases. Exceptions occur where deposition is taking place at low pressure differences. The total mass deposited in a capillary is deduced for a case when aerosol penetration is cut off and the corresponding length of the deposit is 0.6 mm. Whilst considerable deposition in these capillaries is expected without any diminution in the gas flow, the very small reduction in flow in this case is difficult to reconcile with a deposit of this size.

NOMENCLATURE

A	area
c	concentration of aerosol (ρ_a/ρ_g)
d	capillary diameter
D_p	particle diffusivity
g	acceleration due to gravity
H	time interval
J_A	mass current
k	constant (equation (6))
K	constant (equation (23))
L	capillary length
L_d	deposition length
L_a	gas leakage rate
M_T	total mass transmitted
ΔM	mass transmitted in time interval
N	particle number concentration
ΔN	number of particles transmitted in time interval
p	pressure
p_1, p_2	final and initial values of downstream pressure, p_d
Δp	pressure difference ($p_u - p_d$)
Q_a	aerosol mass flux
Q_L	aerosol leakage flux
Q_m	gas mass flux
Q_v	gas volumetric flux
r	radial co-ordinate in capillary
R	radius of capillary
R_G	gas constant
Re	Reynolds number
R_p	particle radius

S	area
SLR	standard leak rate
St_L	laminar Stokes number
t	time
T	temperature
v_d	deposition velocity
\bar{v}_f	gas flow velocity
v_s	sedimentation velocity
\bar{v}	mean laminar flow velocity
z	distance along capillary

Greek letters

α	inverse particle response timescale
η	gas viscosity
μ_g	gas molecular weight
ρ	density
ρ_{dp}	density of aerosol deposit

Subscripts

a	pertaining to aerosol
d	pertaining to downstream
Dep	pertaining to deposit
En	pertaining to capillary entry
Ex	pertaining to capillary exit
g	pertaining to gas
s	pertaining to standard value
u	pertaining to upstream

1. INTRODUCTION

Containment buildings and transport flasks act as barriers to the airborne transmission of radioactivity. Small leaks via capillaries or other leak paths could traverse these barriers and are thus important safety issues. For reactor containments under accident conditions, the issues have been discussed by Morewitz (1982) and Van de Vate (1988). The related theory of particle deposition and plugging has recently been discussed by Williams (1994). For transport flasks, leakage is subject to international regulations (IAEA, 1985) and the U.K. Atomic Energy Code of Practice describes methods for the leakage testing of transport and storage packages (AEA Technology, 1992). The present time-consuming testing process extends to very low gas leakage rates in order to demonstrate compliance with the necessary regulations. If particle leakage did not occur at the lowest rates, simpler and cheaper tests might demonstrate compliance, and an experimental programme has been undertaken to measure aerosol leakage through small capillaries (Mitchell *et al.*, 1990; Morton *et al.*, 1991; Mitchell *et al.*, 1992; Burton *et al.*, 1993a, b). This paper describes associated theoretical work on gas and particle transport and the analysis of experimental results obtained by the programme, and accompanies a report of the experiments on the influence of pressure on aerosol penetration (Morton and Mitchell, 1995).

The experiments of Curren and Bond (1980) showed that critical orifices would block when some particles had dimensions greater than the leak aperture. Experiments summarized by Morewitz (1982) for leak paths up to large sizes showed that plugging occurred after a total aerosol mass leakage proportional to the cube of the leak path diameter. Exceptions have been found to this rule (Novick, 1990), and in most of the experimental work the leakage gas flow was turbulent and aerosol deposition occurred by impaction. The recent theory of Williams (1994) mainly describes the buildup of a plug by deposition from turbulent flow. The object of the present work is to examine theoretically gas and aerosol flows through capillaries in the laminar regime where different criteria could apply. The theory is restricted to general results which can immediately be applied to analyze experimental data. Stopford and Williams (1991, 1992) have performed numerical calculations for particle trajectories in laminar flow to investigate particle sampling by, and deposition in, capillaries, and their results provide a valuable guide to factors which could lead to lack of aerosol penetration.

In Section 2, we describe properties of gas and aerosol flows through a capillary. The leak rate used in the Code of Practice (AEA Technology, 1992) and the standard leak rate (SLR)

are related to the mass and volumetric gas leak rates, and thus to the aerosol particle penetration rate for no loss of particle penetration.

The initial aerosol sampling could possibly be limited by entrainment and a laminar Stokes number is defined to represent its characteristics. The final aerosol concentration emerging from a capillary is related to its initial value at the entrance by assuming a deposition velocity over a certain length of the tube, which gives a general expression in terms of the variables describing the flow as to where aerosol penetration would cut off.

In Section 3, aerosol penetration and plugging is treated in more detail, and we first show that, subject to a general limitation on the aerosol concentration, the total mass transmitted before plugging should be independent of the entrance concentration. Possible cutoffs in penetration from particle diffusion, gravitational settling and impaction are considered, and a general scaling law for cutoff by impaction is obtained which relates capillary length to the upstream and downstream pressures whose difference drives the flow. A number of possibilities are described for the scaling which would relate cutoff dimensions of the length and radius of a capillary.

Comparisons with experiment are described in Section 4. We show that particle penetration results (Mitchell *et al.*, 1992) indicate a cutoff in penetration at low gas leakage rates. Experimental information on the scaling of this cutoff is also examined and we find support for the pressure-length scaling from the experiments in which the driving pressure difference for the flow was varied (Burton *et al.*, 1993a, b; Morton and Mitchell, 1995). Absolute values are obtained for the particle penetration efficiencies in these experiments, and the amount of deposition which occurred in a case when the aerosol penetration efficiency reduced to zero is deduced. Its value is discussed in relation to the experimental observation that aerosol penetration could be reduced to zero with hardly any diminution in the gas flow.

Finally, in Section 5, we draw conclusions on the theory and information gained by comparison with experiment.

2. GAS AND AEROSOL FLOW

Consider the flow of gas containing aerosol through a capillary of length L and radius R . The variables describing the flow induced by a pressure difference ($p_u - p_d$) are shown in Fig. 1. The mass discharge of the gas is Q_m and mass fluxes of the aerosol of relative mass concentration (c) are denoted by Q_a . We now describe the flows in detail.

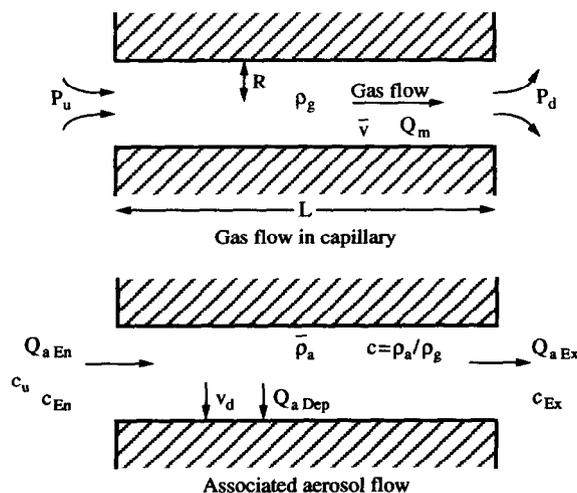


Fig. 1. Variables describing gas and aerosol flow through a capillary.

2.1. Laminar gas flow

For the laminar viscous flow of a perfect gas, the first order solution to the problem, which allows the gas to be compressible, is that the discharge (Q_m) is given by (Landau and Lifshitz, 1959)

$$Q_m = \pi R^2 \rho_g \bar{v} = \frac{\pi}{16} \frac{\mu_g R^4}{\eta L R_G T} (p_u^2 - p_d^2), \quad (1)$$

where μ_g is the gas molecular weight, η the gas viscosity, R_G the gas constant and T the temperature.

This result applies to a sufficiently long capillary ($L \gg R$), when a temperature fall due to the pressure drop can be ignored. In practice, such a temperature fall in laminar flow would only have a very small effect on the gas flow but, as discussed by Clement (1992), induced thermophoretic velocities could affect aerosol deposition. The gas stream velocity for the laminar flow has the well-known parabolic profile,

$$v(r) = \frac{2\bar{v}}{R^2}(R^2 - r^2). \quad (2)$$

The discharge gives the volumetric leak rate at the mean density given by the perfect gas law:

$$\rho_g = \mu_g \frac{\frac{1}{2}(p_u + p_d)}{R_G T}, \quad (3)$$

$$Q_v = \frac{Q_m}{\rho_g} = \frac{\pi R^4}{8 \eta L} (p_u - p_d). \quad (4)$$

When the pressure factor is restored, this gives the leak rate used in a Code of Practice (AEA Technology, 1992 (based on Burrows, 1961)) which is used to predict the measured leak rates by Morton and Mitchell (1995):

$$L_a = \frac{1}{2}(p_u + p_d)Q_v = k(p_u - p_d) \frac{d^4}{\eta L} \frac{p_u + p_d}{2}, \quad (5)$$

where the diameter $d = 2R$ is used to conform to the formula used in the Code and the constant is

$$k = \frac{\pi}{128} = 2.45 \times 10^{-23} (\text{Pa m}^3 \text{ s}^{-1}) (\text{Pa})^{-2} (\mu\text{m})^{-4} (\text{kg m}^{-1} \text{ s}^{-1}) (\text{mm}). \quad (6)$$

The final number is given to yield L_a in $\text{Pa m}^3 \text{ s}^{-1}$ when p_u, p_d are measured in Pa, d in μm , L in mm, and η in $\text{kg m}^{-1} \text{ s}^{-1}$.

The standard leak rate, SLR, used to represent measured rates (Mitchell *et al.*, 1990; Morton and Mitchell, 1995), is obtained from the experimentally measured rise in pressure of the downstream chamber, volume V , from $p_d = p_2$ to p_1 in a time H :

$$\text{SLR} = \frac{VT_s}{HT} (p_1 - p_2) \left[\frac{\eta(p_s^2 - p_d^2)}{\eta_s(p_u^2 - p_d^2)} \right], \quad (7)$$

where the result is normalized to reference conditions, $T_s = 298 \text{ K}$, $\eta = 1.85 \times 10^{-5} \text{ kg m}^{-1} \text{ s}^{-1}$, $p_s = 1 \text{ bar} = 100 \text{ kPa}$ and $p_0 = 0$.

In such a measurement, the mass rise in the downstream chamber would be

$$\Delta M = Q_m H = \frac{\mu_g (p_1 - p_2) V}{R_G T} \quad (8)$$

assuming a perfect gas, which results in

$$\frac{V(p_1 - p_2)}{H} = \frac{R_G T}{\mu_g} Q_m = L_a \quad (9)$$

directly, from equations (1) and (4).

The normalization of this leak rate to a standard pressure difference and viscosity is useful, but the temperature normalization in equation (7) is superfluous because L_a has no explicit temperature dependence. The omission of T_s/T would make no practical difference to the results obtained (Mitchell *et al.*, 1990).

Also in the above standard formulas, temperature differences between upstream and downstream chambers are neglected. Such differences could arise from adiabatic expansion, but, because of restoration to a uniform temperature by heat transfer in practice, it is best not to include this effect. Finally, experiments to measure the leak rate must, as has been the case in the experiments referred to here (Mitchell *et al.*, 1990; Morton and Mitchell, 1995), ensure that the change from p_2 to p_1 does not materially affect the pressure difference $p_u^2 - p_d^2$ in the definition (7). In practice, this is easy to ensure if $p_d \ll p_u$.

2.2. Aerosol flow and deposition

The problem of the passage of aerosol through capillaries divides naturally into two parts. The first part concerns entrance into the capillary and is closely related to the problem of calm air sampling. In the experiments (Mitchell *et al.*, 1990), the air in the chamber sampled by the capillary was only subject to small mixing velocities associated with aerosol suspension in the fluidized bed, and could be regarded as calm. The second part is whether deposition limits transit along the capillary. The discussion here is mainly confined to the second problem, but we first mention some aspects of the sampling problem.

The capillaries used in the experiments (Mitchell *et al.*, 1990) behaved as blunt samplers because their bores were very small ($< 100 \mu\text{m}$). Mitchell *et al.* (1990) discussed the sampling in terms of the criteria of Davies (1968) and Agarwal and Liu (1980) based on the effects of gravity and particle inertia. Gravity only has an effect when the entrance is vertically upwards from the sampling region, as in the experiments, but any bias on size-sampling from gravitational settling was calculated to be negligible (Mitchell *et al.*, 1990). According to the Davies (1968) criterion, sampling bias could have occurred from particle inertia, but, based on the size distributions observed (Mitchell *et al.*, 1990), this did not occur for capillaries for which most of the aerosol was transmitted. When cutoffs in penetration occurred, practically all of the size distribution was affected (Mitchell *et al.*, 1990). Numerical trajectory calculations have been performed by Stopford and Williams (1991), but the size selectivity found does not explain large reduction in aerosol penetration. On the other hand, numerical calculations including impaction and rebound inside the tube entrances (Stopford and Williams, 1992) show that considerable deposition can occur by impaction inside the capillaries not far from the entrance.

The conclusion is therefore that aerosol passage is not primarily being limited by aerosol entrance or sampling effects. However, because the amount of aerosol entrainment and impact into the walls appears to play a role in limiting penetration, it is possible that penetration could depend on a dimensionless quantity relating velocity changes within the capillary to the particle response time to acceleration. This quantity could be defined as a laminar Stokes number:

$$\text{St}_L = \frac{\bar{v}}{\alpha R} = \frac{1}{8} \frac{R}{\alpha L \eta} (p_u - p_d), \quad (10)$$

where \bar{v} is the laminar flow velocity and the second expression arises from the use of equation (1).

The quantity α is the usual inverse response timescale of an aerosol particle to acceleration, which, for a spherical particle radius, R_p , is

$$\alpha = \frac{9\eta}{2\rho_p R_p^2}, \quad (11)$$

where ρ_p is the particle density.

The scaling shown in equation (10) is examined in relation to the experimental data in Section 4.2.

We now describe the aerosol flow through the tube using the variables defined in Fig. 1. At any given time, the aerosol mass flow entering the tube is

$$Q_{a, \text{En}} = \int_{\text{area}} J_A dA = \pi R^2 \rho_a \bar{v}_{\text{En}}, \quad (12)$$

where ρ_a could be modified by entrance sampling effects.

This flux is clearly proportional to the gas flux (1) so that

$$Q_{a, \text{En}} = \frac{\rho_a}{\rho_g} Q_m. \quad (13)$$

However, unlike the gas flux which is conserved through the tube, the aerosol entering the tube can partly be deposited with a total rate

$$Q_{a, \text{Dep}} = \int_{\text{walls}} \rho_a v_d dS, \quad (14)$$

where v_d is a *net* deposition velocity.

If the deposition is assumed to be uniform over a length L_d of the tube surface, and $\bar{\rho}_a$ is the average density deposited.

$$Q_{a, \text{Dep}} = 2\pi R L_d \bar{\rho}_a v_d. \quad (15)$$

Suitable choices for L_d and v_d can be made for all deposition mechanisms, for example $L_d \approx L$ for gravity and $L_d \ll L$ for inertial deposition near the entrance. For these cases the uniformity assumption is expected to be fairly good, subject to only a small reduction in aerosol concentration for gravitational deposition. When the concentration is greatly reduced by diffusional deposition, there will be a non-uniform falloff in deposition with distance.

Clearly, from mass conservation, the aerosol flux leaving the tube is

$$Q_{a, \text{Ex}} = \pi R^2 \rho_{a, \text{Ex}} \bar{v}_{\text{Ex}} = Q_{a, \text{En}} - Q_{a, \text{Dep}}. \quad (16)$$

Since equation (13) applies at the exit with ρ_a replaced by $\rho_{a, \text{Ex}}$, we have

$$Q_{a, \text{Dep}} = (c_{\text{En}} - c_{\text{Ex}}) Q_m, \quad (17)$$

where the aerosol concentration is

$$c = \frac{\rho_a}{\rho_g}. \quad (18)$$

For uniform deposition, we obtain from equations (1) and (15),

$$c_{\text{Ex}} = c_{\text{En}} - \frac{\bar{\rho}_a 2L_d v_d}{\rho_g R \bar{v}} \quad (19a)$$

$$= c_{\text{En}} - \frac{\bar{\rho}_a}{\rho_g} \frac{16\eta L L_d v_d}{R^3 (p_u - p_d)}, \quad (19b)$$

where equations (1) and (3) have been used to obtain the second expression (19b).

Since $\bar{\rho}_a/\rho_g$ is the average concentration, we see that penetration becomes small when

$$\frac{16\eta L L_d v_d}{R^3 (p_u - p_d)} \approx 1. \quad (20)$$

The implications of this result are examined in Section 3.2.

If the initial aerosol density is expressed in terms of a particle number concentration (N particles per unit volume) we can find the number (ΔN) passing through the tube when there are *no* losses in transit. Clearly, in this case the number transmitted is proportional to the

mass of gas transmitted:

$$\frac{\Delta N}{N} = \frac{\Delta M}{\rho_u} = \frac{H}{\rho_u} L_a, \quad (21)$$

where ρ_u is the upstream density and we have used equations (8) and (9).

The rate of particle transfer is therefore

$$\frac{\Delta N}{H} = \frac{N}{\rho_u} L_a = \frac{NT}{\rho_u T_s} \left[\frac{\eta_s(p_u^2 - p_d^2)}{\eta_d(p_s^2 - p_0^2)} \right] \text{SLR}. \quad (22)$$

Comparison of a measured particle transmission rate to a measured leak rate can therefore be used to see whether deposition is taking place. This is done for the relevant experiments in question in Section 4.1.

3. AEROSOL PENETRATION AND PLUGGING

For any capillary or leak path in which net deposition of aerosol takes place, the path will *always* eventually become plugged. The only way this could normally be prevented would be that deposits break off periodically and emerge from the path. Generally, plugging is observed to occur, as described by Morewitz (1982), but not all cases have the simple criteria that he described (Novick, 1990). For the large ducts and turbulent-flow mainly considered by Morewitz (1982) the data are consistent with the simple model of Vaughan (1978) which gives a cube law for the total amount of aerosol mass flow, before plugging,

$$M_T = \int Q_{a, \text{Ex}} dt = KR^3, \quad (23)$$

where K is a constant.

We first prove a general result applicable to both turbulent and laminar flow before discussing penetration cutoffs in detail.

3.1. Dependence on aerosol concentration

The proposed law (23) implies that the total aerosol transmission through a crack or tube would be almost independent of the upstream aerosol concentration (c_u). This result can be made rigorous under two assumptions which are not restrictive:

(A1) The gas flow is independent of aerosol concentration and only depends on the pressure difference and geometry of the tube.

(A2) At any time, the mean flow along the tube is uniform so that there is a steady-state discharge. This does not prevent the flow being turbulent in nature.

Both assumptions rely on the aerosol concentration being not too high. Assumption (A1) breaks down if the contribution of the aerosol to the total fluid density is considerable. Assumption (A2) fails if the aerosol deposition changes the wall geometry and the flow during the transit time of the flow through the tube.

Although the proof can be made more general, we consider a tube of radius ($R(z, t)$) for definiteness, where z is the distance along the tube. This is changing with time according to the local deposition rate, which in a continuum form may be written as

$$\frac{\partial R}{\partial t}(z, t) = -\frac{\rho}{\rho_{dp}} \left[c(R)v_d(R) - D_p \frac{\partial c(R)}{\partial r} \right], \quad (24)$$

where ρ_{dp} is the density of the deposit, and deposition may occur with a radial deposition velocity (v_d) or by diffusion.

This deposition rate is scaled with the upstream concentration by writing

$$\frac{\partial R}{\partial(c_u t)} = -\frac{\rho}{\rho_{dp}} \left[\frac{c}{c_u} v_d(R) - D_p \frac{\partial}{\partial r} \left(\frac{c(R)}{c_u} \right) \right]. \quad (25)$$

The dependence of the scaled concentration on r, z is determined by the steady-state flow equation

$$v_f \cdot \nabla \left(\frac{c}{c_u} \right) = - \left[v_d \left(\frac{c}{c_u} \right) - D \nabla^2 \left(\frac{c}{c_u} \right) \right], \tag{26}$$

where $v_f(r, z)$ is the local flow velocity.

By assumption (A2) the steady flow v_f only depends on time through R ;

$$v_f = v_f(r, R(z, t), \Delta p). \tag{27}$$

If we change the time variable to $u = c_u t$, equations (25) and (26) have a unique solution $R(z, u)$, $v_f(r, R(z, u), \Delta p)$ and $c(r, u)/c_0$ starting from the same state at $u = 0$. This proves that changing the concentration, c_u , just changes the timescale for wall deposition. The leak rate at time t may then be written as

$$Q_{a, Ex}(t) = c_u Q_L(u = c_u t) \tag{28}$$

since the fraction getting through is only a function of $R(u)$.

The total leak rate in time t is

$$\int_0^t Q_{a, Ex}(t') dt' = \frac{1}{c_u} \int_0^{u=c_u t} Q_{a, Ex}(u') du' = \int_0^{u=c_u t} Q_L(u') du'. \tag{29}$$

If plugging occurs and deposits do not break off, this integral tends to a constant value as $u \rightarrow \infty$ or $t \rightarrow \infty$ as shown in Fig. 2. At small times, however, the leak rate is proportional to c_u since

$$Q_L(u = c_u t = 0) = Q_L(R(z, t = 0)). \tag{30}$$

The above argument still holds when the flow in the tube is turbulent as long as R does not change significantly during the timescale of a turbulent fluctuation. The timescale for deposition must be long compared to the fluctuations. The same conclusion holds when a stochastic element is introduced into deposition via random trajectories for individual aerosol particles (Stopford and Williams, 1991, 1992). The random patterns that could result will just occur at different times when c_u , or the number of trajectories per unit time, is varied. The patterns themselves will just be functions of u .

3.2. Cutoffs in penetration

Aerosol penetration through a capillary is expected to cut off, in that most of the incident aerosol is deposited, when the criterion (20) is satisfied. We first consider the possibility that particle diffusion could cut off penetration with a particle diffusivity in terms of a radius, R_p , of

$$D_p = \frac{k_B T}{6\pi\eta R_p} \tag{31}$$

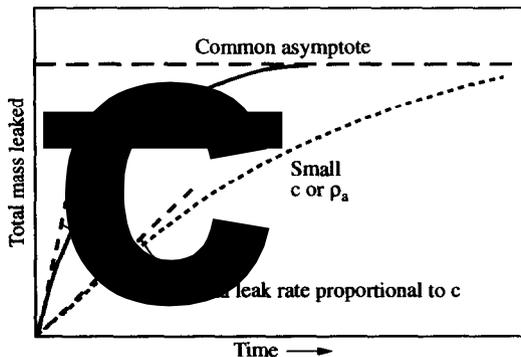


Fig. 2. Dependence of the total aerosol mass leaked on time for different aerosol concentrations.

One might guess that penetration would be cut off by diffusion with the choice in equation (20) of

$$L_d = L, \quad (32)$$

$$v_d = \frac{D_p}{R}. \quad (33)$$

The exact solution is known for particle penetration subject to diffusion alone in laminar flow (e.g. Williams and Loyalka, 1991 (7.35)), and is given in terms of the dimensionless group (20) with v_d specified by equation (33). The penetration decreases exponentially with the value of the group and is approximately 0.13 for the value 1, i.e.

$$\frac{16\eta L^2 D_p}{R^4(p_u - p_d)} = 1. \quad (34)$$

For particles in air with $\eta = 1.85 \times 10^{-5} \text{ kg m}^{-1} \text{ s}^{-1}$,

$$D_p = \frac{1.18 \times 10^{-11}}{R_p(\mu\text{m})} \text{ m}^2 \text{ s}^{-1}, \quad (35)$$

which gives numerical values in equation (34) much too small to account for the cutoffs observed in the experiments of Mitchell *et al.* (1990). However, it is clear that increasing L or reducing R or $p_u - p_d$ would eventually lead to a cutoff in penetration by diffusion deposition. In practice, a provision would be that any lift forces from thermophoresis (Clement, 1992) or the Saffman lift force (Stopford and Williams, 1991) were too small to prevent diffusive deposition.

For a horizontal capillary, we again need to take $L_d = L$ but with the constant gravitational settling velocity, which for spherical particles is

$$v_d = v_s = \frac{2}{9} \frac{\rho_p g R_p^2}{\eta}. \quad (36)$$

This gives the dimensionless group (20) in the form

$$\frac{32}{9} \frac{\rho_p g R_p^2 L^2}{R^3(p_u - p_d)} = \frac{0.3488 \rho_p (10^3 \text{ kg m}^{-3}) R_p^2(\mu\text{m}) L^2(\text{mm})}{(p_u - p_d)(\text{bar}) R^3(\mu\text{m})}, \quad (37)$$

where dimensions appropriate to the experiments of Mitchell *et al.* (1990) have been used.

The capillaries were vertical in these experiments but, had they been horizontal, it is clear that a gravitational cutoff would have been seen in many cases, especially for larger particles.

For deposition by impaction near the capillary entrance, we certainly expect L_d to depend on R and not L and put

$$L_d \approx R. \quad (38)$$

From equation (1) and the gas law (3) for a point in the capillary where the pressure is p , the flow velocity is

$$\bar{v} = \frac{1}{16} \frac{R^2}{\eta L p} (p_u^2 - p_d^2). \quad (39)$$

This means that entrance flows must be identical for

$$\frac{(p_u^2 - p_d^2)}{L p_u} = \text{constant} \quad (40)$$

with a similar scaling for exit flows with p_d in the denominator.

Thus the entrance deposition velocity from impaction can only depend on this combined variable, i.e. $v_d(R, L p_u / (p_u^2 - p_d^2))$. From equation (19a), we see that the precise criterion for change in the aerosol concentration at the entrance depends on the same variables as v_d . This gives the following predicted scaling results.

Deposition behaviour and any cutoff to aerosol penetration through a capillary should, for constant p_u and R , scale as $Lp_u/(p_u^2 - p_d^2)$, if the main deposition mechanism is impaction near the entrance.

This general result is illustrated in Fig. 3 and is understandable from the point of view that, for entrance behaviour including deposition, it should not matter whether the velocities with the same \bar{v} arise from the capillary being long with a large pressure drop, or short with a smaller pressure drop.

Like the scaling for the concentration, the scaling represented by equation (40) extends to the build-up of deposits because it does not involve R directly.

Actual aerosol deposition will involve a sticking probability, S_a , which, in the formalism used here, must be a factor of v_d . The work of Stopford and Williams (1992), who calculated particle trajectories including the particle-wall rebound model of Tsai *et al.* (1990), shows that particles stick better when their velocity normal to the wall is relatively small. Particles with large normal velocities rebound. Experiments, for example Aylor and Ferrandino (1985) have shown that the tangential velocity also can be important in causing rebound. Deposition patterns found for particles of different sizes were concentrated in lengths L_d of order R centred at different points near the entrance of a capillary (50–250 μm inside capillaries of diameter 15 and 25 μm).

These results suggest that deposition depends on the particle velocity, as well as the degree of entrainment represented by the Stokes number, St_L (equation (10)). The basic scaling of the onset of large deposition rates and cutoff of penetration with R cannot therefore be simply deduced from equations (19) and (20), and some possibilities are listed in Table 1. In addition to scalings in terms of R and L already apparent in equation (10) and the laminar velocity of equation (1), we could postulate a dependence on this velocity

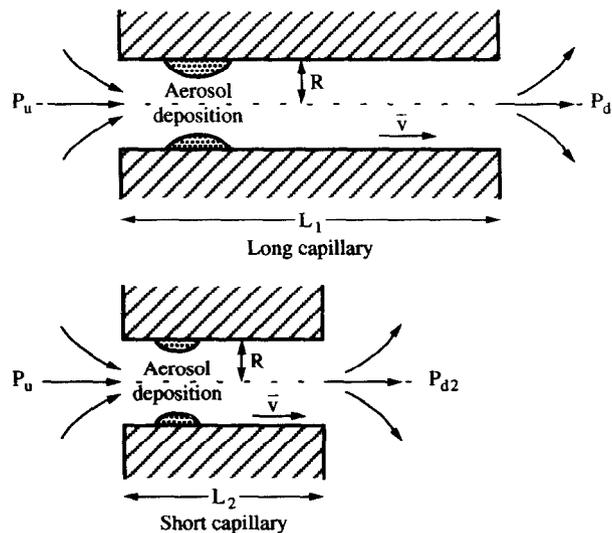


Fig. 3. Identical aerosol deposition near the entrance for capillaries with the same values of R and $(p_u^2 - p_d^2)/p_u L$.

Table 1. Possible scalings with capillary radius R and length L of a cutoff in particle penetration through capillaries

Scaling	Physical origin
R/L	Entrainment: Stokes number St_L (equation (10))
R^2/L	Flow velocity (equation (1))
R^3/L	Reynolds number for flow velocity, Re (equation (41))
R^3/L^2	Uniform deposition (e.g. gravity equation (37))

through a Reynolds number for the flow:

$$\text{Re} = \frac{2\rho_g \bar{v}R}{\eta} = \frac{1}{8} \frac{\mu_g}{R_G T} \frac{R^3}{\eta^2 L} (p_u^2 - p_d^2). \quad (41)$$

There is no guarantee that any scaling found from experimental information for aerosol of one composition would work for another composition, except where v_d is explicit such as for gravitational settling when the sticking probability would appear directly in equations (36) and (37). If sticking were the dominant influence on deposition, the probability would depend on the normal velocity times a function dependent on material properties.

4. COMPARISON WITH EXPERIMENT

Using the theoretical results obtained above, we now examine the experimental data from a series of experiments by Mitchell and collaborators on aerosol penetration through capillaries (Mitchell *et al.*, 1990; Morton *et al.*, 1991; Mitchell *et al.*, 1992; Burton *et al.*, 1993a, b; Morton and Mitchell, 1995).

4.1. Aerosol penetration dependence on leakage rate

Experiments were performed on the penetration through capillaries of aerosols consisting of glass microspheres and cerium oxide particles. The resulting penetration rates were summarized by Mitchell *et al.* (1992) in terms of the associated air leakage rates of the capillaries. Aerosols were generated using a fluidized bed system, and the aerosol penetration rate was measured by counting transmitted particles. The results can therefore be analyzed using equations (21) or (22) on the assumption of complete particle transmission with the gas, which gives

$$\frac{\Delta N}{H} (\text{particles h}^{-1}) = 5.4 \times 10^4 N (\text{particles cm}^{-3}) \text{SLR} (\text{Pa m}^{-3} \text{s}^{-1}). \quad (42)$$

Lines corresponding to $N = 70 \text{ particles cm}^{-3}$, the quoted number, and $N = 700 \text{ particles cm}^{-3}$, are shown in Fig. 4 compared to the experimental data. It is clear that the assumption that there was little or no deposition or holdup by the capillaries describes the data for $\text{SLR} > 10^{-4} \text{ Pa m}^3 \text{ s}^{-1}$, if the upstream number concentration varied by up to a factor of 10 above 70 cm^{-3} .

For SLR between 10^{-5} – $10^{-4} \text{ Pa m}^3 \text{ s}^{-1}$, although experimental results may not be reliable below particle numbers of about 50 h^{-1} , the limit of the particle detector efficiency, there are strong indications of a cutoff in particle penetration through the capillaries.

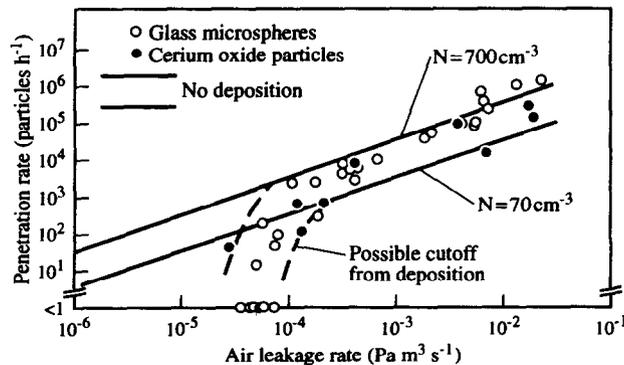


Fig. 4. Observed (Mitchell *et al.*, 1992) and theoretical dependence of aerosol penetration rates through capillaries on the air leakage rate.

4.2. Scaling of penetration cutoff

From equations (19) and (20) and the results of the last section, we can generally understand why penetration could be cut off with changes of the capillary variables, specifically increase in L and decrease in R . Originally, experiments were performed in which one of these dimensions was varied whilst the other was kept fixed (Mitchell *et al.*, 1990). Sharp cutoffs were indeed seen in particle penetration, and their observed values are compared in Table 2 with the possible scalings suggested in Table 1. For a given scaling law to explain the observed cutoffs, the numbers in the two columns should be the same. In addition, for the validity of the possible scaling with R_p discussed below, the numbers in the two rows for $R_p = 1 \mu\text{m}$ and $R_p = 0.5 \mu\text{m}$ should also be identical.

The results appear to favour the R/L scaling associated with the inertial or postulated Stokes number (10) discussed earlier which, in this case, is given by

$$\text{St}_L = 8.11 \frac{R(\mu\text{m})}{L(\text{mm})} (p_u - p_d) (\text{bar}) \rho_p (10^3 \text{ kg m}^{-3}) R_p^2 (\mu\text{m}) \quad (43)$$

$$= 21 \frac{R(\mu\text{m})}{L(\text{mm})} R_p^2 (\mu\text{m}) \quad (44)$$

for the glass spheres of density $2.60 \times 10^3 \text{ kg m}^{-3}$ and $p_u - p_d = 1 \text{ bar}$ (Mitchell *et al.*, 1990).

This correlation would predict a strong dependence on aerosol size. There is little evidence of a strong size selective value of the cutoff in the size distributions of the penetrating particles (Figs 11 and 12 of Mitchell *et al.*, 1990), although small particles of $R_p < 0.5 \mu\text{m}$ appear to be cut off at $L = 10 \text{ mm}$ for the $10 \mu\text{m}$ capillaries and $R = 22.5 \mu\text{m}$ for the 50 mm capillaries. In only the first case does the scaling shown in Table 2 support the Stokes number description, and, for particles larger than $R_p = 1 \mu\text{m}$, there appears to be no evident scaling with R_p .

In the experiments of Morton *et al.* (1991) on short capillaries, the experiments appeared to be divided between cases where penetration was seen and cases where it was not. The data and possible scalings are shown in Table 3 where we see that penetration generally occurred when the ratios were large, but was cut off at small ratios (small R or large L). Although most of the data are consistent with any of the possible scalings, in that the values of the ratios for penetration are always greater than those for no penetration, there are some anomalies (underlined) for the R/L and R^3/L^2 scalings. Thus these data would tend to support the velocity-based R^2/L or R^3/L scalings.

In the pressure variation experiments (Burton *et al.*, 1993a, b; Morton and Mitchell, 1995), penetration of glass microsphere aerosols through capillaries of length $L = 20 \text{ mm}$ and radius $R = 15 \pm 0.5 \mu\text{m}$ was measured as a function of the pressure difference, $p_u - p_d$, with $p_d = 100 \text{ kPa}$.

Much higher aerosol concentrations were used than the earlier experiments (Mitchell *et al.*, 1990), and, after an initial period, a total lack of penetration and plugging from deposition occurred at the lower pressure differences.

Table 2. Observed cutoffs in L and R in the experiments of Mitchell *et al.* (1990) and scaling ratios obtained from values of R , L and R_p

Fixed capillary dimension	$R = 10 \mu\text{m}$	$L = 50 \text{ mm}$
Experimental penetration cutoff in variable dimension for $R_p = 1 \mu\text{m}$	$L = 30 \text{ mm}$	$R = 15 \pm 5 \mu\text{m}$
Scaling ratios		
R/L	0.33	0.3
R^2/L	3.3	4.5
R^3/L	33	67.5
R^3/L^2	1.11	1.35
$R_p^2 R/L$	0.33	0.3
Experimental penetration cutoff in variable dimension for $R_p = 0.5 \mu\text{m}$	$L = 10 \text{ mm}$	$R = 22.5 \mu\text{m}$
Scaling ratio		
$R_p^2 R/L$	0.25	0.11

Table 3. Data for short capillaries from Morton *et al.* (1991)

Radius R (μm)	Length L (mm)	Penetration	R/L	R^2/L	R^3/L	R^3/L^2
9.93	4.54	Yes	2.19	21.7	216	47.5
9.73	5.58	Yes	1.74	17.0	165	29.6
9.94	7.22	Yes	1.38	13.7	136	18.8
9.69	10.05	Yes	0.96	9.34	90.5	9.01
9.87	19.82	No	0.50	4.92	48.5	2.45
9.94	20.69	No	0.48	4.78	47.5	2.29
9.88	20.38	No	0.48	4.72	46.6	2.26
9.87	25.18	No	0.39	3.87	38.2	1.52
6.28	4.60	No	1.37	8.57	53.8	11.7
5.64	4.70	No	1.20	6.77	38.2	8.12
5.64	10.78	No	0.52	2.95	16.6	1.54
5.64	10.60	No	0.53	3.00	16.9	1.60
1.75	6.08	No	0.29	0.50	0.88	0.14

The results are comparable to the experiments (Mitchell *et al.*, 1990) with capillaries of length $L = 50$ mm, where particle penetration was cut off at $R \approx 15 \mu\text{m}$ (see Table 2). The pressure were $p_u = 200$ kPa and $p_d = 100$ kPa, and we can use the proposed scaling relation (40) to see where the cutoff would be predicted to be in the pressure variation experiments (Burton *et al.*, 1993a, b; Morton and Mitchell, 1995):

$$\frac{(p_u^2 - 100^2)}{20p_u} = \frac{(200^2 - 100^2)}{50 \times 200} \quad (45)$$

The solution to the quadratic is $p_u = 134$ kPa, and the value $p_u - p_d = 34$ kPa is precisely in the region between 40 and 20 kPa where rapid deposition begins to take place (Burton *et al.*, 1993a, b). In the constant pressure tests at $\Delta p = 20$ kPa, aerosol penetration ceased after a 10 min period (Burton *et al.*, 1993a). Thus the scaling relation (40) is supported by the combined results of the experiments (Mitchell *et al.*, 1990; Burton *et al.*, 1993a, b; Morton and Mitchell, 1995).

4.3. Aerosol and gas penetration

In the pressure variation experiments (Burton *et al.*, 1993a, b; Morton and Mitchell, 1995), values of the upstream aerosol concentration (ρ_a in mg l^{-1}) and particle penetration rates (particles h^{-1}) were obtained at 10 min intervals (Burton *et al.*, 1993a). For each separate experiment, an initial value of the efficiency of the aerosol penetration was obtained by normalizing the ratio of the penetration rate to the aerosol concentration such that its maximum value was equal to 1. Here, we obtain absolute values of the aerosol penetration efficiency by normalizing the rates between experiments.

The actual aerosol penetration is proportional to the actual gas flow rate as well as the concentration and is given by equation (22), which in the present case is numerically:

$$\Delta N(\text{particles h}^{-1}) = \frac{60^2 \rho_a (\text{mg l}^{-1}) [(p_u(\text{kPa})/100)^2 - 1] \text{SLR} (\text{Pa m}^3 \text{s}^{-1})}{m_p(\text{mg}) p_u(\text{kPa})}, \quad (46)$$

where m_p is the mean particle mass used to transform the observed aerosol mass concentration ρ_a into the equivalent number concentration.

An absolute transmission efficiency can therefore be defined as

$$E = \frac{\Delta N(\text{particles h}^{-1}) m_p(\text{mg}) p_u(\text{kPa})}{3600 \rho_a (\text{mg l}^{-1}) [(p_u(\text{kPa})/100)^2 - 1] \text{SLR} (\text{Pa m}^3 \text{s}^{-1})}. \quad (47)$$

To evaluate this quantity, the experimental information for the 10 min interval with maximum relative transmission was used together with the initial value of SLR for the capillary and the pressure, $p_u(\text{kPa}) = 100 + \Delta p(\text{kPa})$. The mean particle mass was taken from the count median geometric diameter of $3.0 \mu\text{m}$ ($= 2R_p$) based on the volume

equivalent diameter (Ball *et al.*, 1989) and a density of 2.4 g cm^{-3} :

$$m_p = \frac{4}{3}\pi R_p^3 \rho_p = 10.8\pi 10^{-9} \text{ mg.} \quad (48)$$

The results are shown in Table 4 and, considering the large variation in concentrations and the averaging over a 10 min interval, show a remarkable consistency with the hypothesis that the transmission efficiency was close to unity. However, in two of the low pressure difference cases when the aerosol penetration ceased in the following interval, the low values of the efficiency obtained indicate that there must have been considerable deposition in the interval in question.

To calculate the amount of material that deposited and caused lack of penetration is not possible in the cases of sudden plugging ($\Delta p = 20 \text{ kPa}$). However, we can perform the calculation in the case of $\Delta p = 40 \text{ kPa}$ where particle penetration gradually decreased to zero over seven 10 min periods, under the assumption that the transmission efficiency started at unity during the first period. The total mass deposited is obtained by multiplying the mass transmission (equation (22)) by (1-efficiency) for each 10 min period and summing:

$$M_d = \frac{600((p_u(\text{kPa})/100)^2 - 1)\text{SLR}(\text{Pa m}^3 \text{ s}^{-1})}{p_u(\text{kPa})} \sum_n \rho_a(\text{mg l}^{-1})(1 - E_n)\text{mg.} \quad (49)$$

The data are reproduced in Table 5 from which we find $M_d = 1.09 \times 10^{-3} \text{ mg}$. At a density of 2.4 g cm^{-3} this corresponds to a volume of $4.54 \times 10^{-4} \text{ mm}^3$. For a capillary of diameter $31 \mu\text{m}$ (Test 3, see Burton *et al.*, 1993a), the length plugged would be 0.60 mm . This length is much less than that of the capillary (20 mm), confirming the supposition and experimental observation that aerosol deposition was localized near the entrance. However, if all the deposition occurred inside the capillary, it does make it difficult to explain why the final observed gas leak rate ($2.83 \times 10^{-4} \text{ Pa m}^3 \text{ s}^{-1}$) is so little reduced from the initial value ($2.86 \times 10^{-4} \text{ Pa m}^3 \text{ s}^{-1}$) unless the deposit was dislodged (Morton and Mitchell, 1995) as discussed below.

A qualitative explanation can be given as to why considerable deposition in the capillary does not significantly reduce the gas flow rate. From equations (1), (7) and (9) the average

Table 4. Aerosol penetration efficiencies in the pressure variation experiments of Burton *et al.* (1993a, b)

Test no.	Δp (kPa)	Conc. ρ_a (mg l^{-1})	SLR ($\text{Pa m}^3 \text{ s}^{-1}$)	Pen. rate ΔN (particle h^{-1})	Efficiency E
1	80	0.67	3.64×10^{-4}	270,000	1.95
2	60	0.71	3.26×10^{-4}	330,000	2.57
3	40	1.33	2.86×10^{-4}	273,000	1.48
4*	20	3.22	1.24×10^{-4}	6000	0.048
5*	20	0.14	2.00×10^{-4}	12,000	1.35
6*	20	4.24	2.57×10^{-4}	119,400	0.35
7*	20	0.33	2.19×10^{-4}	45,000	1.96
8	60	0.04	8.88×10^{-4}	60,000	3.03
9	80	1.18	1.33×10^{-4}	360,000	4.05
10	100	4.3	4.10×10^{-4}	1,830,000	1.96

*Cases for which no penetration was observed for the next 10 min interval.

Table 5. Deposited mass in Test 3 with $\Delta p = 40 \text{ kPa}$

End of time interval minutes	Conc. (ρ_a) (mg l^{-1})	Efficiency E_n	Deposited mass (mg)
10	1.33	1	—
20	0.51	0.886	6.82×10^{-5}
30	0.52	0.436	3.45×10^{-4}
40	0.28	0.262	2.43×10^{-4}
50	0.19	0.154	1.89×10^{-4}
60	0.13	0.163	1.28×10^{-4}
70	0.10	0	1.18×10^{-4}

gas velocity is related to the leak rate and tube radius by

$$\bar{v} = \frac{p_u^2 - p_d^2}{p_s^2 - p_0^2} \frac{2SLR}{p_u + p_d} \frac{1}{\pi R^2} = \frac{728}{R^2(\mu\text{m})} \text{ m s}^{-1} \quad (50)$$

for a capillary at $p_u = 140 \text{ kPa}$, $p_d = 100 \text{ kPa}$, and $SLR = 2.86 \times 10^{-4} \text{ Pa m}^3 \text{ s}^{-1}$.

With the initial radius of $15.5 \mu\text{m}$, $\bar{v} = 3.03 \text{ m s}^{-1}$ which is over two orders of magnitude less than the limiting sound and choked velocities for the capillary. Local deposits will just cause local acceleration and a higher velocity for the gas so that the total drag on the flow will hardly be increased in a long capillary. There is an absolute limit to the radius of an aperture which would maintain the same flow rate, and this would correspond to $R \approx 1.5 \mu\text{m}$ in this case. An aperture of this size or, more likely, a set of equivalent small apertures in a loose deposit structure would prevent aerosol penetration but allow a similar gas flow. Deposits were observed to have a loose dendritic structure (Morton and Mitchell, 1995).

This explanation of little reduction in gas leak rates will certainly work up to a certain level of deposition, but the quantitative difficulty with the capillary in question is why it should still hold for a deposit which almost fills the capillary for a distance of $600 \mu\text{m}$. It is possible, however, that the deposit in this capillary was dislodged before the final gas leak rate was observed (Morton and Mitchell, 1995).

The gas acceleration near a deposit also provides a qualitative explanation of why the deposition rate should rapidly increase with time once deposition reaches a certain level. This is shown in Table 5 where, after small initial deposition, the rate increases very rapidly after 20 min.

5. CONCLUSIONS

An outline theory on the transport of aerosol through small capillaries has been constructed and used to examine the experimental results obtained by Mitchell *et al.* (1990, 1992), Morton *et al.* (1991) and Burton *et al.* (1993a, b), Morton and Mitchell (1995). The theory is outline in nature because it obtains general results based on a postulated deposition velocity and distance, and does not attempt to solve the detailed transport equations. The principal conclusions of this work are the following.

(a) A cutoff in aerosol penetration through capillaries is expected to occur (equations (19) and (20)) at low laminar flow rates corresponding to small capillary radius, long capillary length, or small pressure difference. The experiments support the existence of this cutoff (Fig. 4) at air leakage rates between 10^{-5} and $10^{-4} \text{ Pa m}^3 \text{ s}^{-1}$.

(b) If any net deposition occurs in a capillary or leak path, the path will always plug and aerosol penetration will be reduced to zero. The total aerosol mass transmitted before plugging is predicted to be independent of initial aerosol concentration. The experiments (Burton *et al.*, 1993a, b) show that plugging does occur, but that deposits can break off, generally when the gas flow changes. Quantitative experimental information on the dependence of plugging on aerosol concentration is lacking. Concentrations in the experiments of Burton *et al.* (1993a, b) spanned a suitable range, but varied during the course of each experiment.

(c) The experimental results support the proposed general scaling relation (40) relating capillary length and upstream and downstream pressures at which aerosol penetration cuts off rapidly. This relation follows if aerosol deposition occurs over a limited capillary length by impaction near the entrance, a supposition which is backed up by direct experimental observation (Burton *et al.*, 1993a, b; Morton and Mitchell, 1995) and by calculations of particle trajectories (Stopford and Williams, 1992). These calculations describe strongly convergent streamlines for gas flow into a narrow entrance in a flat wall with the particle trajectories causing deposition meeting the wall at distances inside the capillary from three to over 10 times the capillary radius, depending on particle size.

(d) The scaling of the penetration cutoff with capillary radius depends on details of aerosol entrainment and deposition (including bounce), near the capillary entrance. Various

physical processes would give differing scaling dependences on capillary radius (R) and length (L) about which the available experimental evidence is not decisive. A general dependence on R^n/L with $n = 1-3$ is supported, but a rigorous scaling as $R^3/(L^2 R_p^2)$ is expected (where R_p is the particle radius) for gravitational settling, which would occur in horizontal capillary or leaks (not in the current experiments).

(e) The absolute penetration efficiencies deduced for the maximum penetrations in the experiments of Burton *et al.* (1993a, b), Morton and Mitchell (1995) are consistently close to unity, showing that, in most cases, periods occurred when practically all the aerosol particles penetrated the capillaries. Once deposition begins, the rapid cutoff in penetration is attributed to acceleration in the gas flows near deposits and an increase in the aerosol impaction rate.

(f) For initially slow laminar gas flows, considerable localized deposition is possible before the total gas leakage rate is reduced. However, it is difficult to reconcile a deposit of length 0.6 mm, deduced for one of the plugged cases seen by Burton *et al.* (1993a, b), with practically no observed reduction in the rate unless the deposit was dislodged before final gas leak rate testing (Morton and Mitchell, 1995).

Impaction near the entrance appears to play the major role in cutting off aerosol penetration through capillaries in the experiments performed. This might not be the case universally: in horizontal capillaries sedimentation and even diffusion for fine particles in very long capillaries could be important. Also, thermophoresis and vapour condensation, not discussed here but previously considered (Clement, 1982), could effect deposition velocities, especially for cases of large pressure drops.

In conclusion, we see that, for laminar gas flow through leak paths or capillaries, aerosol behaviour is not as uniform as it probably is in turbulent flows where impaction will always take place. Under some conditions for laminar flow practically no aerosol will be deposited, but sharp changes to complete lack of penetration can occur when small changes take place in the flow and when gas flows accelerate past existing deposits. Complete explanations of these changes are still lacking, but the theory given here provides a framework in which to examine experimental evidence on the subject.

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