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ATTACHMENT OF GASEOUS FISSION PRODUCTS TO AEROSOLS

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ABSTRACT

Accidents may occur in which the integrity of fuel cladding is breached and volatile fission products are released to the containment atmosphere. In order to assess the magnitude of the subsequent radiological hazard it is necessary to know the transport behaviour of such fission products. It is frequently assumed that the fission products remain in the gaseous phase. There is a possibility, however, that they may attach themselves to particles and hence substantially modify their transport properties. This paper provides a theoretical assessment of the conditions under which gaseous fission products may be attached to aerosol particles.

Specific topics discussed are:-

the mass transfer of a gaseous fission product to an isolated aerosol particle in an infinite medium; the rate at which the concentration of fission products in the gas phase diminishes within a container as a result of deposition on a population of particles; and the distribution of deposited fission product between different particle sizes in a log-normal distribution. It is shown that, for a given mass, small particles are more efficient for fission product attachment, and that only small concentrations of such particles may be necessary to achieve rapid attachment.

Conditions under which gaseous fission products are not attached to particles are also considered, viz, the competing processes of deposition onto the containment walls and onto aerosol particles, and the possibility of the removal of aerosols from the containment by various deposition processes, or agglomeration, before attachment takes place.

1. Introduction

Incidents in which the integrity of fuel cladding is breached and volatile fission products are released to the containment atmosphere are an important component in reactor Safety Cases. In particular the subsequent leakage of fission products from the containment has to be demonstrated to be within the constraints on release to the environment.

In order to assess the magnitude of the release it is necessary to know the mechanisms by which such volatile fission products deposit or plate-out on the containment surfaces. In assessments of fission product behaviour it is frequently assumed that the fission products remain in the gaseous phase. However, the fission products may attach themselves to aerosols present in the containment. For a given mass of particles the surface area is inversely proportional to the radius, R and hence increases with reducing particle size so that the surface area of small particles can be comparable to that of the containment. In addition the mass transfer coefficient for attachment of fission products is also proportional to R^{-1} , in general, and hence small particles are a particularly efficient sink for fission products. Should fission products attach to aerosols their behaviour with respect to deposition, filtration and release would be substantially different from their assumed behaviour as gases.

Experimental evidence for the attachment of fission products to aerosols has been obtained by several workers. The deposition of radon and thoron daughters in flow along a tube was found to be inhibited if the carrier gas was unfiltered (1). This was attributed to the reduced mobility of the fission products when they were attached to particles in the air. Others (2, 3, 4, 5, 6) have demonstrated and examined the attachment to a monodispersed aerosol, and measured (7) the uptake on a polydispersed aerosol. These experiments have covered a wide range of particle radii from $0.01 \mu\text{m}$ to $10 \mu\text{m}$ and have demonstrated reasonable agreement with theory for the case of unit sticking probability. The fission products used were $\text{RaA}(\text{}^{218}_{84}\text{Po})$ and $\text{ThB}(\text{}^{212}_{82}\text{Pb})$ which are relatively involatile.

A theory for the attachment of radioactive atoms to aerosols has been published (2), and extended (8, 9) to cover the case of a charged aerosol and a diffusing radioactive ion.

This paper examines the circumstances under which fission products may be attached to aerosol particles within a container. The analysis is carried out in terms of a sticking probability which is assumed to be independent of the gas concentration and particle size.

The mass transfer of a gaseous fission product to an isolated aerosol particle in an infinite medium is discussed in Section 2. The rate at which the concentration of fission product in the gas phase diminishes within a container as a result of deposition on a monodispersed and polydispersed population of aerosol particles is discussed in Sections 3 and 4 respectively. The distribution of fission products between different particle sizes in a log-normal distribution is also discussed in Section 4.

In order that the aerosol population has a significant effect on fission product behaviour deposition on aerosols must compete successfully with deposition on containment walls. Similarly deposition on aerosols must be rapid compared to the removal of the aerosols themselves by plate-out or agglomeration. These aspects are discussed in Section 5.

2. Attachment of Gaseous Fission Products to a Single Particle in an Infinite Medium

Consider a particle of radius R in an infinite gaseous medium containing a fission product gas with initial uniform concentration C_0 . For times $t > 0$ fission product molecules will strike the particle surface and a fraction α , the sticking probability, will stick. It is assumed that molecules remain on the surface once they have stuck, and that there is no surface saturation which would inhibit other molecules sticking. Thus there will be a net flux of fission product gas to the particle by diffusion.

The variation in the concentration of gaseous fission product molecules can be obtained from the solution of the appropriate diffusion equations. The diffusion equations, however, cannot be used within a radial distance $r < \lambda$ of the particle surface, where λ is the mean free path of the gas molecules. Thus the diffusion equation will be solved for the region $r = a = (R+\lambda)$ to infinity.

The boundary equation at a may be shown to be

$$\left. \frac{\partial C}{\partial r} \right|_{r=a} = h C_a, \quad (1)$$

where $h = \frac{\bar{v}\alpha}{4D(1+\lambda/R)^2}$,

D = diffusion coefficient of the gas molecules,
and \bar{v} = mean molecular speed of the gas molecules.

Outside the imaginary surface at a, the motion of the gas molecules may be described by the diffusion equation

$$\frac{\partial C}{\partial t} = D \left(\frac{\partial^2 C}{\partial r^2} + \frac{2}{r} \frac{\partial C}{\partial r} \right) \quad (2)$$

with initial condition

$$C = C_\infty \text{ for } a < r < \infty \text{ at } t = 0$$

and boundary condition

$$C = C_\infty \text{ at } r = \infty \text{ for all } t$$

and equation (1) at a for all t.

The solution to these equations is (10)

$$C = C_\infty \left\{ 1 - \frac{ha^2}{r(ah+1)} \left[\operatorname{erfc} \left(\frac{r-a}{2\sqrt{Dt}} \right) - e^{(h'(r-a)+h'^2Dt)} \operatorname{erfc} \left(\frac{r-a}{2\sqrt{Dt}} + h'\sqrt{Dt} \right) \right] \right\} \quad (3)$$

where $h' = h + 1/a$

For times t such that

$$t \gg \tau_0 = (h'^2 D)^{-1} \quad (4)$$

a steady state is reached in which

$$C_a = \frac{C_\infty}{(1+ah)} \quad (5)$$

Thus the net flux of molecules which stick to the particle, F , is

$$F = \pi R^2 \bar{v} \alpha \frac{C_\infty}{(1+ah)} \quad (6)$$

Equation (6) corresponds to that derived by Lassen and Rau (1) for the special case of unit sticking probability.

For the cases of practical interest the time constant for attainment of steady state, τ_0 , equation (4), is extremely short (microseconds or less). Thus for practical timescales equation (6) may be used to describe the flux of molecules to an isolated particle.

For $(ah) \gg 1$ equation (6) may be simplified to

$$F = 4\pi R D C_\infty \quad (7)$$

where the net flux of molecules which stick to the particle is diffusion controlled, is independent of the sticking probability and is proportional to the particle radius.

For $(ah) \ll 1$ we have

$$F = \pi R^2 \bar{v} \alpha C_\infty \quad (8)$$

in which case the net flux is controlled by the surface interaction rate and is proportional to the square of the particle radius i.e. its surface area.

3. Attachment of Gaseous Fission Products to a Monodispersed Population of Particles

The analysis of Section 2 assumed a single particle in an infinite medium. In practice there will be an assembly of particles and hence the boundary condition $C = C_0$ at $r = \infty$ will not hold. In this section we shall assume an idealised condition of a population of particles each with radius R_0 arranged in a regular array. If there are N particles per unit volume then we may define a parameter b , the radius of the sphere of influence of each particle, by the equation

$$N \cdot \frac{4}{3} \pi b^3 = 1 \quad (9)$$

The boundary condition for the diffusion equation would then be

$$D \frac{\partial C}{\partial r} \Big|_{r=b} = 0 \quad (10)$$

with equation (1) at a , and initial condition

$$C = C_{\infty} \text{ at } t = 0 \text{ in } a < r < b \quad (11)$$

The solution to the diffusion equation, equation (2), with boundary conditions (10) and (2) and initial condition (11) may be found in Carslaw and Jaeger (10). For times t such that

$$t > \frac{b^2}{D(3\pi/2)^2} \quad (12)$$

the average concentration of gaseous fission product is given approximately by

$$\bar{C} = C_{\infty} e^{-t/\tau_N} \quad (13)$$

where the time constant τ_N is given by

$$\tau_N = \frac{(1+ah)}{3a^2 h} \frac{R^3}{D} \frac{\rho_p}{\rho_{pg}} \quad (14)$$

where ρ_p is the particle density, and ρ_{pg} is the mass of particles per unit volume.

Values of τ_N for the case of deposition onto graphite particles of fission product molecules of atomic mass $M=130$ in carbon dioxide at 100°C and pressure 20 atmospheres are shown in figure (1). Except for large particles and low particle mass concentrations it may be seen that for unit sticking probability the magnitude of the time constant is very small, of the order of seconds or less.

4. Attachment of Gaseous Fission Products to a Polydispersed Population of Particles

Section 2 described the flux of gaseous fission products to an isolated particle and showed that the flux is greater to larger particles. However, because the number of particles for a given total mass varies inversely as the cube of the particle radius, section 3 showed that for a given total mass small particles are much more efficient in absorbing fission products than larger particles. In practice, since we seldom have a monodispersed population of particles, it is useful to know how the fission products are distributed amongst a polydispersed population.

Assuming a log-normal particle size distribution (11) the fraction by mass of particles having radii between R and $R + dR$ is

$$g(R)dR = \frac{1}{\sqrt{2\pi} \log \beta_g} \cdot \exp\left(-\frac{(\log R - \log R_g)^2}{2(\log \beta_g)^2}\right) \frac{dR}{R}$$

where R_g is the mass geometric mean of the particle radii, and β_g is the standard geometric deviation.

For spherical particles of constant density, ρ_p the number distribution is

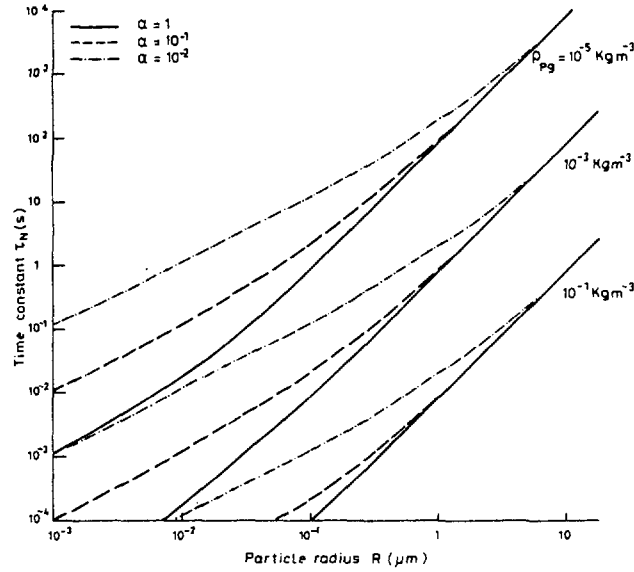


FIG.1. Time Constant τ_N for Reduction in Fission Product Concentration due to Absorption by Monodispersed Particle Population with Radius R and Concentration ρ_{pg} .

$$f(R)dR = \frac{3\bar{m}}{4\pi\rho_p} \frac{g(R)dR}{R^3} \quad (15)$$

where \bar{m} is the arithmetic mean of the masses of the particles.

Assuming that the particles are separated by distances which are large compared with their radii, for times greater than τ_0 (equation 4), the flux of fission product F to a particle of radius R is given approximately by equation (5), where $C_\infty = C$ the concentration at, strictly speaking $r = \infty$, but for this approximation $r \gg R$.

Thus the rate of fission product deposition onto those particles whose radii lie between R and $R + dR$ is

$$I(R)dR = F \cdot f(R)dR \cdot N_0 \quad (16)$$

where N_0 is the total number of particles.

In a restricted volume C will not be constant but will diminish as deposition continues. Since C reduces slowly in comparison with the time constant for attainment of equilibrium, τ_0 , the rate of reduction of fission product concentration in volume V may be approximated by

$$V \frac{dC}{dt} = - \int_0^\infty I(R)dR \quad (17)$$

Substituting for $I(R)dR$ from equation (16) and F from equation (5) we have

$$\frac{dC}{dt} = - \frac{\pi \bar{v} \alpha}{V} N_0 C \int_0^{\infty} \frac{R^2 f(R) dR}{(1+ah)} \quad (18)$$

Integrating equation (18) with respect to t , and in this case taking $C = C_0$ at $t=0$

$$C = C_0 \exp \left\{ - \frac{\pi \bar{v} \alpha}{V} N_0 t \int_0^{\infty} \frac{R^2 f(R) dR}{(1+ah)} \right\}. \quad (19)$$

Equation (19) is an approximate description of the reduction in the concentration of fission product gas away from the particle surfaces.

The total amount of fission product gas absorbed on a particular size range with radii between R and $R+dR$ over all time is

$$J(R)dR = \int_{t=0}^{\infty} (I(R)dR) dt \quad (20)$$

and the fraction of fission product gas absorbed on particles less than a given radius R is defined as

$$Q(R) = \int_{R=0}^R \frac{J(R)dR}{VC_0}. \quad (21)$$

Substituting (15), (16) and (20) into equation (21) and integrating gives

$$Q(R) = 0.5 [1 + \text{erf } \phi_0], \quad (22)$$

$$\text{where } \phi_0 = \frac{[\log R - (\log R_g - (3-m) \ln 10 (\log \beta_g)^2)]}{\sqrt{2} \cdot \log \beta_g},$$

$$m = 1 \text{ when } ah \gg 1,$$

$$\text{and } m = 2 \text{ when } ah \ll 1.$$

An illustration of the distribution of fission products amongst a log-normal population of particles is shown in figure 2 for the limiting case of $ah \gg 1$ and $ah \ll 1$.

For the case of $ah \gg 1$, i.e. when the product of sticking probability and particle radius is large, we see that the fission product activity is highly concentrated in the small size component of the distribution. Even though only 6% of the mass is less than 0.5 μm radius these particles contain 64% of the activity.

For $ah \ll 1$, i.e. when the product of sticking probability and particle radius is small, the fission product activity is still relatively concentrated in the small size range, but less so than the case for $ah \gg 1$, with only 28% in the 0 to 0.5 μm radius range and the peak occurring in the 0.5 to 1 μm size range.

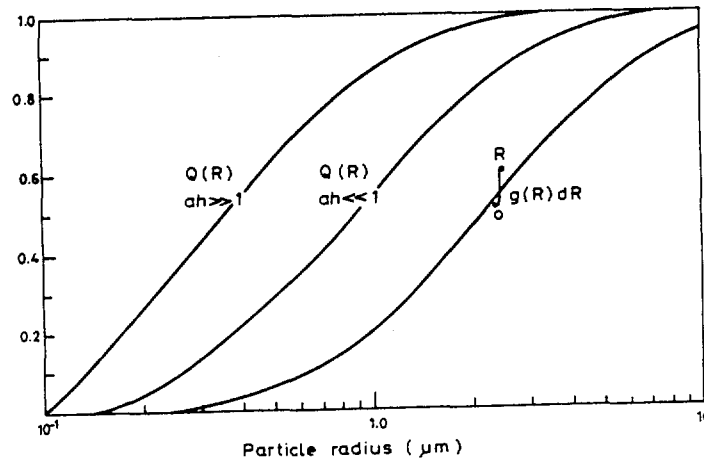


FIG. 2. Distribution of Fission Products between a Log-Normal Distribution of Particles with $R_g = 2.2 \mu\text{m}$ and $\beta_g = 2.6 \mu\text{m}$.

5. Non-Attachment of Gaseous Fission Products to Aerosols

The previous sections have dealt with quantifying the degree to which gaseous fission products attach to a population of aerosol particles. There may be circumstances, however, when the co-existence of an aerosol population and gaseous fission products does not necessarily result in the deposition of one on the other. Two such circumstances will be examined in this section. Firstly, where the aerosol is confined in an enclosure so that the walls of the enclosure compete for the fission product. Secondly, where the attachment of fission product to particles is slower than removal of the particles by various mechanisms, e.g. agglomeration, plate-out.

5.1 Attachment of Fission Products to Enclosure Walls

As an indication of the relative attachment to particles and container walls we shall evaluate the time constant for attachment to the latter and compare this with that for the former, equation (14) and figure 1.

Assuming the fission product gas is in a well mixed container of volume V and surface area A , and that the mass transfer coefficient to the wall is K , then conservation of mass gives

$$V \frac{dC}{dt} = -KA(C - C_w) \quad (23)$$

where C_w = concentration of the fission product at the wall.

Integrating equation (23) and taking $C_w = 0$ and $C = C_0$ at $t = 0$

$$C = C_0 \exp(-t/\tau_w) \quad (24)$$

where
$$\tau_w = \frac{V}{AK} \quad (25)$$

τ_w is the time constant for attachment to the container walls.

In the case of molecular transport K is governed by Brownian diffusion of the molecules through a relatively stagnant region adjacent to the walls (the boundary layer) and the subsequent sticking to the wall. For a boundary layer thickness δ and sticking probability to the wall α_w the steady state mass transfer coefficient is

$$K = \frac{Dh_w}{(1+h_w\delta)} \quad (26)$$

where

$$h_w = \frac{\bar{v} \alpha_w}{4D}$$

Thus

$$\tau_w = \frac{V}{A} \frac{(1+h_w\delta)}{Dh_w} \quad (27)$$

Evaluation of equation (27) shows that for $\delta > 10^{-4}$ m τ_w is proportional to δ and that the sticking probability has negligible effect on the time constant for attachment to the walls (in contrast to attachment of particles). Taking typical values of $\delta = 10^{-3}$ m and $V/A = 10^{-1}$ m τ_w is 10^2 seconds.

Comparing this value with figure 1 it may be concluded that for a significant concentration of submicron particles in a large container deposition of gaseous fission products onto particles is far more rapid than onto the container walls.

5.2 Time Constant for Removal of Particles

Particle removal mechanisms may be divided into two categories: firstly, agglomeration, in which the particles are not actually removed from the gas phase but the quantity in a given size distribution changes; and secondly, plate-out onto container surfaces where the particles are removed from the gas phase.

(a) Agglomeration

There are several mechanisms by which particles agglomerate, e.g. Brownian diffusion, gravitational and turbulent agglomeration. For small particles Brownian diffusion is the dominant mechanism, and since small particles have greatest efficiency for the attachment of fission products we shall consider only this mechanism.

For a monodispersed population of particles with radius R_o it may be shown (12) that the number concentration, n , varies with time as

$$n = \frac{n_o}{[1 + t/\tau_a]} \quad (28)$$

where $\tau_a = \frac{2}{A(R_o)n_o}$,

n_o = number concentration of particles at $t = 0$,
and $A(R_o)$ = agglomeration coefficient.

From the definition of $A(R_o)$ (12) the time constant for the reduction in the concentration of particles, τ_a , may be written

$$\tau_a = \frac{\sqrt{2}}{3} \frac{R_o}{\bar{v}_o} \frac{\rho_p}{\rho_{pg}} [1 + \sqrt{2} R_o h_o] \quad (29)$$

where $h_o = \frac{\bar{v}_o}{4D_o}$

$$\bar{v}_o = \sqrt{\frac{8kT}{\pi m_o}} = \text{particle mean velocity}$$

and D_o = diffusion coefficient of the particle.

τ_a , equation (29), may be compared with the time constant for attachment of gaseous fission products to a monodispersed population of particles of radius R_o , τ_N , equation (14). Substituting typical values it may be shown that τ_N is very much shorter than τ_a (except for very small particles and very low sticking probabilities). Thus we may conclude that agglomeration of particles will not inhibit deposition of fission products onto them.

(b) Plate-Out

There are several mechanisms which can contribute to the plate-out of particles in a container and evaluation of their magnitude depends upon specific conditions, e.g. turbulence intensity, heat flux, etc. Therefore it is not possible to generalise on the relative rates of particle plate-out and fission product attachment to particles, and this section in no way sets out to be comprehensive. However, the method of evaluating particle plate-out time constants is set out together with some examples for typical conditions.

If the transfer coefficient of the particles to the wall is K_p , then, following section 5.1, the time constant for particle plate-out is

$$\tau_p = \frac{V}{AK_p} \quad (30)$$

For gravitational settling K_p may be taken as the terminal settling velocity of the particle v_g , where (11)

$$v_g = \frac{2}{9} \frac{R_p^2 \rho_p g}{\mu}$$

The resultant plate-out time constants τ_p are shown in figure 3. [For the purpose of illustration the case of graphite particles, $\rho = 2.3 \cdot 10^3 \text{ kg m}^{-3}$, in CO_2 at 20 atmospheres pressure and 100°C is taken throughout this section.]

For turbulent deposition the situation is considerably more complicated. However, an upper bound to the mass transfer coefficient may be taken as (13)

$$K_p \approx 0.1 U_\tau$$

where $U_\tau = U\sqrt{f/2}$ = friction velocity

U = mean flow velocity

f = friction factor for the turbulence (14).

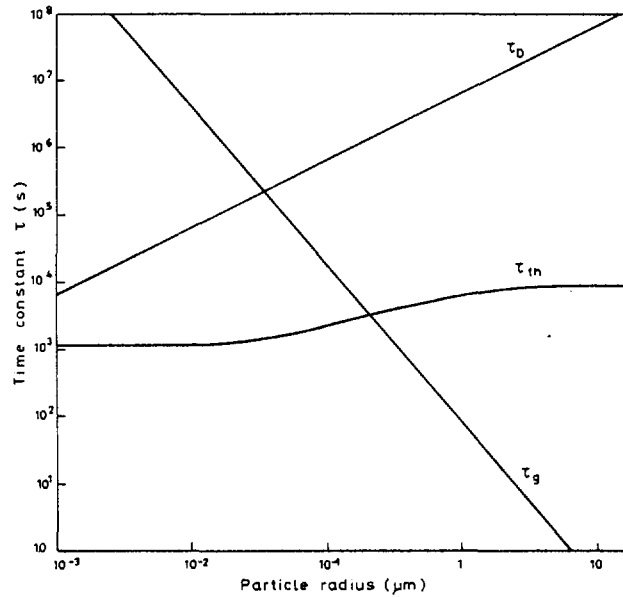


FIG.3. Time Constant for Particle Plate-out Due to Gravity (τ_g), Brownian Diffusion (τ_D) and Thermophoresis (τ_{th}). $V/A = 0.1 \text{ m}$, $\delta = 10^{-3} \text{ m}$.

Typically the friction velocity is much less than the mean flow velocity. Taking a typical value for U_τ of $0.1 U$ and using a value of 10 m s^{-2} for U gives a plate-out time constant of 10^3 s .

For Brownian deposition the particle diffuses through a stagnant boundary layer of thickness δ . The mass transfer coefficient in these circumstances is

$$K_p = \frac{D}{\delta}$$

Values of the time constant for Brownian deposition τ_D are shown in figure 3 as a function of particle size for a boundary layer thickness of 10^{-3} m .

Thermophoretic deposition results from the motion of a particle in a temperature gradient. The mass transfer coefficient in such circumstances may be taken as the equilibrium drift velocity, v_{th} (15), given by

$$v_{th} = \frac{\mu}{\rho} \cdot \frac{\nabla T}{T} \cdot G(\chi_p/\chi_g, R)$$

where ∇T is the temperature gradient, and G is a function of the relative thermal conductivities of particle and gas (χ_p/χ_g) and the particle radius, R .

The resultant time constants for thermophoretic deposition as a function of particle radius for a thermal gradient of 10^5 K m^{-1} and (χ_p/χ_g) of 36 are shown in figure 3.

Comparison of the time constants for particle deposition evaluated in this section, figure 3, with those for attachment of fission products to particles, figure 1, shows that particles are unlikely to plate-out before fission products have time to attach to them.

6. Discussion

Lassen and Rau's theory for the attachment of gaseous fission products to an isolated particle has been extended to the case of non-steady state and non-unit sticking probability. The results confirm that equilibrium is achieved exceedingly rapidly and show that the equilibrium flux is directly proportional to R for

particles where $R > \lambda$ and unit sticking probability, and is proportional to R^2 particles whose radii $R < \lambda$ and when the sticking probability is sufficiently less than unity (even for $R > \lambda$).

Therefore for a given mass of particles smaller particles are more efficient than larger in absorbing fission products. These results have been confirmed experimentally for the case of unit sticking probability (2, 3).

The above results have been used to derive the time constant τ_N with which the concentration of fission products attaches to a population of particles. It is found that the magnitude of τ_N can be very short even for relatively modest concentrations of particles e.g. for $10^{-3} \text{ kg m}^{-3}$ of $1 \text{ }\mu\text{m}$ radius particles with unit sticking probability τ_N is ~ 1 second. τ_N is predicted to be inversely proportional to the concentration of particles and to depend on particle radius in a similar fashion to that discussed above.

Direct measurement has been made (4, 5) of τ_N for attachment of ThB to a monodispersed aerosol. For particles with radii between $0.6 \text{ }\mu\text{m}$ and $2.9 \text{ }\mu\text{m}$ (4) confirmed that τ_N was inversely proportional to the particle number density (the authors quote a sticking probability of 0.08 but careful examination of the data shows that it is consistent with unity), and (5) confirmed the theoretical equation with unit sticking probability for particles with radii between $0.0045 \text{ }\mu\text{m}$ and $0.015 \text{ }\mu\text{m}$.

Experiments on iodine vapour release in a container (16, 17, 18) showed that considerable quantities (50%-80%) attached to adventitious particles, with attachment predominantly on particles with sizes similar to that of Aitken nuclei (0.01 - $0.4 \text{ }\mu\text{m}$). The average sticking probability was estimated to be $\sim 10^{-3}$ to 10^{-2} ; however, iodine has been found to attach to certain specific aerosols, e.g. Ag, MgO much more readily than to adventitious atmospheric aerosols (19).

Equations have also been derived for the distribution of fission products between a log-normal size distribution of particles. An example for a typical distribution shows that whereas only 6% of the mass is below $0.5 \text{ }\mu\text{m}$ radius these particles contain 64% of the fission products.

Although fission products could attach rapidly to a population of particles circumstances may arise where they may attach preferentially to the container surfaces, or the particles may be removed by agglomeration or plate-out before the fission products have time to deposit on them. It is shown that:-

- (i) preferential attachment to container walls is only likely in the case of very low particle concentrations and/or low sticking probability to the particle;
- (ii) agglomeration of particles is a much slower process than attachment of fission products and so does not hinder attachment;
- (iii) particle removal by plate-out is unlikely to be faster than attachment of fission products to the particles.

Overall it may be concluded that when gaseous fission products are released in the presence of a population of particles, particularly small particles, there is a strong possibility that they will attach rapidly to those particles. In such circumstances gaseous fission product transport would be governed by the transport properties of the particles.

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