



# DIFFUSIVE AND CONVECTIVE DEPOSITION OF AEROSOLS IN RISING SPHERICAL BUBBLES WITH INTERNAL CIRCULATION

S. M. GHIAASIAAN and G. F. YAO

G. W. Woodruff School of Mechanical Engineering, Georgia Institute of Technology,  
Atlanta, GA 30332-0405, U.S.A.

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**Abstract**—Aerosol transport due to Brownian and convective mechanisms in variable-radius spherical bubbles rising in a stagnant liquid is modeled. The modeling is based on the extension of Kronig & Brink's analysis of mass transfer in a bubble or droplet, and accounts for the coupling between the two aerosol transport mechanisms.

Parametric calculations are performed for air and steam bubbles containing aqueous aerosols and rising in stagnant water, within parameter ranges where bubbles can remain spherical and non-oscillating, and Brownian and convective transport are the only dominant aerosol removal mechanisms. It is shown that, within the aforementioned parameter range, Fuch's model for Brownian aerosol removal is inadequate.

*Key Words:* aerosols, diffusion, convection, bubbles, internal circulation

## 1. INTRODUCTION

Aerosol generation and transport in bubbles rising in liquids is of interest in many industrial and natural processes. The removal of radioactive aerosols, suspended in gas bubbles rising in a liquid pool, by the surrounding liquid, a process commonly called wet scrubbing, is particularly important in the phenomenology of a number of hypothetical accident scenarios in nuclear reactors. Due to their occurrence in these hypothetical accidents, aerosol transport phenomena have recently been investigated rather extensively, leading to the development of several computer codes (Owczarski *et al.* 1985; Wassel *et al.* 1985; Jonas & Schütz 1988; Ghiaasiaan *et al.* 1990; Calvo *et al.* 1991).

Mechanistic modeling of aerosol transport in bubbles is difficult, however. The difficulties can be divided into two categories: those associated with bubble hydrodynamics, and those related to the multitude of the mechanisms contributing to aerosol removal.

Bubbles rising in liquids remain spherical when they are very small and are within limited ranges of geometric and property-related parameters (Clift *et al.* 1978). Analysis of deformed bubbles is significantly more complicated than spherical bubbles. Spherical bubbles, furthermore, undergo sustained oscillations when  $Re_b \geq 200$  to 1000 (Clift *et al.* 1978). Bubble oscillations can also occur during bubble generation, due to bubble coalescence or breakup, etc. These oscillations greatly enhance the transport processes in bubbles.

Several mechanisms can contribute to the removal of aerosols in rising bubbles. The most common mechanisms are diffusion (Brownian), inertial (due to bubble internal circulation) and sedimentation (Fuchs 1964). Other mechanisms include: convective deposition, which takes place when bubble size varies with time; thermophoresis, which occurs when the bubble and its surrounding liquid are at different temperatures; and diffusophoresis, which takes place when significant mass diffusion, e.g. due to evaporation or condensation in a multi-component gas bubble, occurs inside the bubble.

Fuchs (1964) modeled aerosol removal due to diffusion, inertial and sedimentation mechanisms, and represented their combined effect according to:

$$\frac{dN}{dz} = -\alpha_1 N \quad [1]$$

where  $N$  is the total number of aerosols in a bubble and  $z$  is the vertical, upwards co-ordinate. The total aerosol removal coefficient,  $\alpha_1$ , is found from:

$$\alpha_1 = \Sigma \alpha_j \quad [2]$$

where  $\alpha_j$  represent aerosol removal coefficients due to various mechanisms. Equations [1] and [2] assume that aerosol fluxes resulting from separate mechanisms can be added together in order to find the total aerosol flux. The removal coefficients, which were modeled separately for each mechanism by neglecting the possible coupling between them, are (Fuchs 1964):

$$\alpha_s = \frac{3g\tilde{t}_p}{4R_B U_B} \quad [3]$$

$$\alpha_i = \frac{9U_B \tilde{t}_p}{2R_B^2} \quad [4]$$

$$\alpha_d = 1.8 \left( \frac{\mathcal{D}}{R_B^3 U_B} \right)^{1.2} \quad [5]$$

In these equations,  $\alpha_s$ ,  $\alpha_i$  and  $\alpha_d$  represent the sedimentation, inertial and Brownian aerosol removal coefficients, respectively,  $R_B$  and  $U_B$  represent bubble radius and rise velocity, respectively,  $g$  is the gravitational acceleration and  $\mathcal{D}$  represents the aerosol diffusivity.

For spherical aerosols, the characteristic time,  $\tilde{t}_p$ , is governed by

$$\tilde{t}_p = m_p / f, \quad m_p = 4\pi\rho_p R_p^3 / 3 \quad [6]$$

where  $m_p$ ,  $\rho_p$  and  $R_p$  represent aerosol mass, density and radius, respectively, and  $f = 6\pi\mu_G R_p$  represents aerosol friction coefficient assuming Stokesian particles, where  $\mu_G$  represents the gas viscosity. When bubbles grow or shrink, e.g. due to evaporation or condensation, aerosol transport due to convection contributes to the overall aerosol removal. A convective aerosol removal coefficient,  $\alpha_c$ , defined according to the following equation, can then be included in the right hand side of [1] (Wassel *et al.* 1985; Ghiaasiaan *et al.* 1990):

$$\alpha_c = \frac{3\dot{m}''}{\rho_v R_B U_B} \quad [7]$$

where  $\rho_v$  represents the vapor density, and the mass flux at interphase,  $\dot{m}''$ , is assumed positive for condensation. Equation [7] can also be written as

$$\alpha_c = - \frac{3}{R_B U_B} \frac{dR_B}{dt} \quad [8]$$

where  $t$  represents time. Evidently  $\alpha_c < 0$  for a growing bubble, or when evaporation takes place at the bubble-liquid interphase. In any case the  $\alpha_1 \geq 0$  limit must be imposed on [2]. Aerosol removal due to other mechanisms has similarly been accounted for by including their corresponding removal coefficients in [1].

The aforementioned equations evidently apply to monodisperse aerosols. Polydisperse aerosols can also be treated using these equations by defining a finite number of aerosol size groups.

The summation of removal coefficients (or, equivalently, the summation of aerosol fluxes resulting from various removal mechanisms) according to [1] and [2] assumes no coupling between separate mechanisms, and is evidently acceptable in principle when only one of the mechanisms is dominant. However, aerosol removal due to the combined effect of two different removal mechanisms has been rigorously modeled in the past only for a few simple flow configurations (Yu *et al.* 1977; Taulbee 1978; Homsy *et al.* 1981; De la Mora & Rosner 1981; Mills *et al.* 1984). The latter authors addressed the coupling between thermophoresis and convection in laminar boundary layers, and showed that the method based on summation of the separately modeled two aerosol fluxes is inadequate (Mills *et al.* 1984). The combined effect of several mechanisms has received little attention.

Fuchs's model for Brownian removal is based on the application of the penetration theory, where the aerosol characteristic diffusion period is represented by the residence time of gas at the bubble surface during a bubble circulation period, and is estimated using the Hadamard-Rybczynski

solution for gas velocity inside the bubble (Hadamard 1911). The result, [5], can alternatively be represented as:

$$\text{Sh} = 0.85 \text{Pe}^{1/2} \quad [9]$$

where the Sherwood number,  $\text{Sh}$ , and the Peclet number,  $\text{Pe}$ , are defined as:

$$\text{Sh} = \frac{2R_B K}{\mathcal{D}} \quad [10]$$

$$\text{Pe} = \frac{2R_B U_B}{\mathcal{D}} \quad [11]$$

where  $K$  is the diffusive aerosol transfer coefficient.

Despite the above-mentioned idealizations, Fuchs's model has been widely applied to aerosol removal in bubbles, even when bubbles are large and likely to be non-spherical (Heinscheid & Schütz 1984; Wassel *et al.* 1985; Jonas & Schütz 1988; Ghiaasiaan *et al.* 1990; Pich & Schütz 1991).

Diffusion of mass in spherical, non-oscillating bubbles and droplets with internal circulation has been extensively studied in the past (Kronig & Brink 1950; Calderbank & Korchinski 1956; Brignell 1975; Tong & Sirignano 1986; Renksizbulut & Bussman 1993; Ghiaasiaan & Eghbali 1994). Kronig & Brink (1950) analyzed mass diffusion in a circulating spherical droplet with constant radius in creep flow by using the Hadamard–Rybczynski stream functions (Hadamard 1911), and casting the transferred species mass conservation equation in the orthogonal co-ordinate system  $(\xi, \zeta, \varphi)$ , where  $\varphi$  represents the azimuthal angle in polar spherical coordinates, and

$$\xi = 4\eta^2(1 - \eta^2)\sin^2 \theta \quad [12]$$

$$\zeta = \frac{\eta^4 \cos^4 \theta}{2\eta^2 - 1} \quad [13]$$

where  $\eta = r/R_B$  represents the dimensionless radial co-ordinate, and  $\theta$  is the tangential angle in polar spherical co-ordinates.

Since in most mass transfer problems the residence time of the fluid along the closed vortices is much shorter than the characteristic time for mass diffusion normal to the streamlines, the transient mass species conservation equation can be represented in a one-dimensional form, with  $\xi$  as the independent spatial variable, thus rendering the mass transfer process independent of the bubble internal vortex strength.

For the  $\text{Pe} \rightarrow \infty$  limit, which is representative of aerosol transport, the solution to the species conservation equation derived by Kronig & Brink can be represented as (Kronig & Brink 1950):

$$E = 1 - \frac{3}{8} \sum_{i=1}^{\infty} A_i^2 \exp(-16\epsilon_i \tau) \quad [14]$$

where the fractional approach to equilibrium,  $E$ , and the dimensionless time,  $\tau$ , are defined according to:

$$E = 1 - \frac{n}{n^0} \quad [15]$$

$$\tau = \frac{t\mathcal{D}}{(R_B^0)^2} \quad [16]$$

where  $n$  represents the number of aerosols, per unit bubble volume, and  $n^0$  and  $R_B^0$  represent initial values for  $n$  and  $R_B$ , respectively.

Kronig & Brink, who were interested in the solution for  $\tau \rightarrow \infty$  limit, only calculated the numerical values of coefficients  $A_i$  and  $\epsilon_i$  for the first two terms in the above series. Heetjes *et al.* (1954) subsequently calculated the numerical values of the aforementioned constants for the first seven terms in the series. Calderbank & Korchinski (1956) showed that the solution of Heetjes *et al.* can be approximately represented by:

$$E = [1 - \exp(-2.25\pi^2\tau)]^{1/2} \quad [17]$$

Kronig & Brink's method can evidently be applied to modeling the transport of aerosols in bubbles. Mills & Hoseyni (1988) thus suggested the application of [17] instead of Fuchs's model, [9], for Brownian removal of aerosols in bubbles, and derived:

$$Sh = \frac{3\pi^2 \exp(-2.25\pi^2\tau)}{4 E(1-E)} \tag{18}$$

It should be mentioned that, due to their transient nature, none of [14], [17] or [18] can be directly used for deriving a relation for  $\alpha_d$  to be incorporated along with other removal coefficients in [1] and [2].

Recently, Ghiaasiaan & Eghbali (1994) extended Kronig & Brink's method to the case of mass diffusion in internally-circulating spherical droplets with variable radius.

In this paper, the combined Brownian and convective aerosol deposition in spherical rising bubbles is modeled based on the extension of Kronig & Brink's solution to bubbles with variable radii. Bubbles with constant radii as well as expanding and shrinking bubbles are thus treated. Illustrative parametric calculations are performed and the results are compared with predictions obtained using the widely-applied methods, thereby assessing the accuracy and validity of these methods.

## 2. MATHEMATICAL MODEL

### 2.1. Bubble aerosol transport

Figure 1 depicts the bubble and the internal circulatory stream lines. The bubble Reynolds number,  $Re$ , is assumed to be  $O(10-100)$ , therefore the bubble is assumed to remain spherical, without oscillations. The internal circulatory motion is represented by the stream function (Hadamard 1911; Batchelor 1956; Harper & Moore 1968):

$$\psi_G = 0.5Ar^2(R_B^2 - r^2)\sin^2 \theta \tag{19}$$

where  $\psi_G$  represents the Stokes stream function for the bubble interior,  $r$  represents the radial co-ordinate and  $A$  represents the Hill's vortex strength. The bubble is initially assumed to contain monodisperse, spherical aerosols with the number density  $n^0$ .

The characteristic time associated with the development of a steady-state droplet internal circulation,  $\tilde{t}_h$ , can be estimated from (Prakash & Sirignano 1978; Abramzon & Sirignano 1989):

$$\tilde{t}_h = O\left(\frac{\rho_L (R_B^0)^2}{\mu_L Re}\right) \tag{20}$$

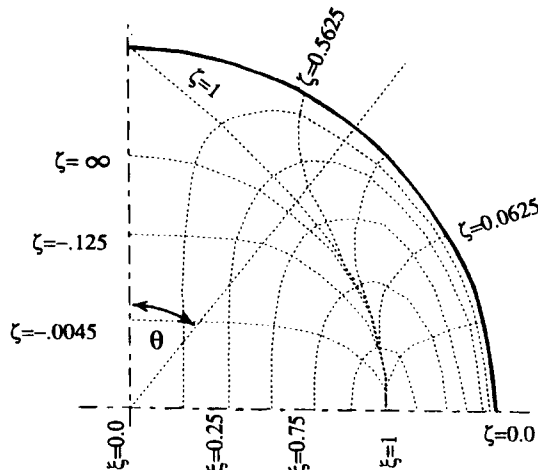


Figure 1. Bubble internal circulation.

where  $\rho_L$  and  $\mu_L$  represent the liquid density and viscosity, respectively, and  $Re = 2\rho_L R_B U_B / \mu_L$  represents the bubble Reynolds number. The characteristic time for aerosol diffusion in the bubble,  $\tilde{t}_d$ , can be estimated from (Abramzon & Sirignano 1989):

$$\tilde{t}_d = O\left[\frac{(R_B^0)^2}{\mathcal{D}}\right] \quad [21]$$

The aerosol diffusivity can be obtained from the Stokes–Einstein relation (Friedlander 1977; Reist 1993):

$$\mathcal{D} = \frac{kT_G C}{6\pi\mu_G R_p} \quad [22]$$

where  $T_G$  and  $\mu_G$  represent the gas temperature and viscosity, respectively, and  $k = 1.38 \times 10^{-23} \text{ J K}^{-1}$  represents Boltzmann's constant. The Cunningham correction factor,  $C$ , is found from the interpolation relation:

$$C = 1 + Kn \left[ a_1 + a_2 \exp\left(-\frac{a_3}{Kn}\right) \right] \quad [23]$$

where  $a_1 = 1.252$ ,  $a_2 = 0.399$  and  $a_3 = 1.10$  (Jennings 1988), and the Knudsen number,  $Kn$  is defined as:

$$Kn = l/R_p \quad [24]$$

The mean free path of the gas molecules,  $l$ , can be calculated from (Friedlander 1977):

$$l = v_G \left( \frac{\pi M_G}{2kN_A T_G} \right)^{1/2} \quad [25]$$

where  $v_G$  and  $M_G$  represent the gas kinematic viscosity and molar mass, respectively, and  $N_A = 6.02 \times 10^{26}$  molecules/k mol represents Avogadro's number. For conditions of interest here  $\tilde{t}_d \gg \tilde{t}_h$ , and quasi-steady state hydrodynamics can be assumed.

Assuming that all aerosol removal mechanisms except for convection and Brownian are negligible, the aerosol conservation equation in the bubble can be written as:

$$\frac{\partial \phi}{\partial t} + \mathbf{U}_G \cdot \nabla \phi = \mathcal{D} \nabla^2 \phi \quad [26]$$

where  $\phi = n/n^0$  is the normalized aerosol density. This equation can be recast in the  $(\xi, \zeta, \theta)$  co-ordinates. Since the residence time of the fluid along the closed vortices is much shorter than the characteristic time for aerosol diffusion normal to the streamlines in the parameter range of interest here, the transient aerosol conservation equation can be represented in a one-dimensional form with  $\xi$  as the spatial independent variable. Further manipulation of [26] then leads to (Ghiaasiaan & Eghbali 1994):

$$\gamma^2 \frac{\partial \phi}{\partial t} + 16 \frac{\mathcal{R}(\xi)}{\mathcal{Q}(\xi)} \gamma \left( \frac{d\gamma}{d\tau} \right) \frac{\partial \phi}{\partial \xi} = \frac{16}{\mathcal{Q}(\xi)} \frac{\partial}{\partial (\xi)} \left( \mathcal{P}(\xi) \frac{\partial \phi}{\partial \xi} \right) \quad [27]$$

where  $\gamma = R_B/R_B^0$ , is the dimensionless bubble radius, and

$$\mathcal{P}(\xi) = - \int_{\eta_1^*}^{\eta_2^*} \frac{(2\eta^2 - 1)^2 \sin^2 \theta}{\eta \cos^3 \theta} d\zeta \quad [28]$$

$$\mathcal{Q}(\xi) = - \int_{\eta_1^*}^{\eta_2^*} \frac{(2\eta^2 - 1)^2}{4\eta^3 \cos^3 \theta \Delta} d\zeta \quad [29]$$

$$\mathcal{R}(\xi) = - \int_{\eta_1^*}^{\eta_2^*} \frac{(2\eta^2 - 1)^3 \sin^2 \theta}{8\eta \cos^3 \theta \Delta} d\zeta \quad [30]$$

$$\eta_1^*, \eta_2^* = [\frac{1}{2}(1 \mp (1 - \xi)^{1/2})]^2 \quad [31]$$

$$\Delta = (1 - \eta^2)^2 \cos^2 \theta + (2\eta^2 - 1)^2 \sin^2 \theta \quad [32]$$

As noted, [27] is independent of the vortex strength  $A$ . The second term on the left hand side of [27] represents the effect of time-dependent bubble radius. Numerical values of the functions  $\mathcal{P}(\xi)$  and  $\mathcal{Q}(\xi)$  can be found in Kronig & Brink (1951). Values of  $\mathcal{R}(\xi)$  are provided in Ghiaasiaan & Eghbali (1994).

The initial condition for [27] is

$$\phi = 1, \quad \text{for } \tau \leq 0 \quad [33]$$

At the center of the vortices  $\phi$  is assumed to be a regular function of  $\xi$  (Brignell 1975). Therefore, at  $\xi = 1$ ,

$$\gamma^2 \frac{\partial \phi}{\partial \tau} + 16 \frac{\mathcal{R}(\xi)}{\mathcal{Q}(\xi)} \gamma \left( \frac{d\gamma}{d\tau} \right) \frac{\partial \phi}{\partial \xi} = \left( \frac{16}{\mathcal{Q}(\xi)} \frac{d\mathcal{P}(\xi)}{d\xi} \right) \frac{\partial \phi}{\partial \xi} \quad [34]$$

At  $\xi = 0$ , representing the bubble surface,  $\phi = 0$ . The average normalized aerosol number density in the bubble,  $\bar{\phi}$ , can be shown to be

$$\bar{\phi} = \frac{3}{8} \int_0^1 \mathcal{Q}(\xi) \phi(\xi) d\xi \quad [35]$$

It can also be shown that

$$\text{Sh} = \frac{32}{3\bar{\phi}} \left. \frac{\partial \phi}{\partial \xi} \right|_{\xi=0} \quad [36]$$

## 2.2. Bubble size variation and rise velocity

Bubble size variations can occur due to condensation, evaporation and pressure change. For parametric calculations, the removal of aerosols in bubbles of various sizes is considered, where the bubble radii are assumed to vary with time at constant rates. Growing ( $d\gamma/dt > 0$ ) and shrinking ( $d\gamma/dt < 0$ ) bubbles are both considered. It is emphasized, however, that the aforementioned formulation is general and can be applied to bubble size variations, as long as  $\tilde{t}_R \gg \tilde{t}_h$ , where  $\tilde{t}_R = R_B/(dR_B/dt)$  represents the characteristic bubble radius variation period.

In the forthcoming calculations, to be presented in the next section, bubble size variation is assumed to be slow, and inertial effects are neglected. The bubble is assumed to be single-component. It can be easily shown that the convective removal coefficient,  $\alpha_c$ , presented earlier in [7] and [8] for condensation, can be represented in terms of dimensionless parameters as:

$$\alpha_c = - \left[ \frac{3\mathcal{D}}{(R_B^0)^2 U_B \gamma} \right] \frac{d\gamma}{d\tau} \quad [37]$$

## 3. METHOD OF SOLUTION

Equation [27] was numerically solved using the fully-implicit finite-difference technique with equally-spaced mesh points in the  $\xi$  co-ordinate, and applying central differencing to the spatial derivatives. A small time step size, typically representing  $\Delta\tau = 10^{-6}$ , was used at the beginning of each calculation, and was increased during each numerical run, typically to  $\Delta\tau = 5 \times 10^{-4}$  near the end of each calculation. The number of mesh points was 500.

Numerical values of functions  $\mathcal{P}(\xi)$ ,  $\mathcal{Q}(\xi)$  and  $\mathcal{R}(\xi)$  were separately calculated by numerical integration of [28]–[30], by the trapezoidal rule, using  $\Delta\xi = 10^{-5}$  as the step size. Values of the function  $\mathcal{R}(\xi)$  can be found in Ghiaasiaan & Eghbali (1994).

Bubble rise velocity,  $U_B$ , is obtained everywhere using the correlations of Peebles & Garber (Peebles & Garber 1953; Wallis 1969).

## 4. RESULTS AND DISCUSSION

## 4.1. Constant bubble radius

Air and water at atmospheric pressure and room temperature (300 K) were chosen for parametric calculations. Thus,  $\rho_L = 996 \text{ kg/m}^3$ ,  $\mu_L = 2.79 \times 10^{-4} \text{ kg m/s}$ ,  $\sigma = 0.071 \text{ N/m}$ ,  $\rho_G = 1.18 \text{ kg/m}^3$  and  $\mu_G = 1.84 \times 10^{-5} \text{ kg m/s}$  were assumed. The air bubble is assumed to contain monodisperse aqueous aerosols, also with  $\rho_p = 996 \text{ kg/m}^3$ .

Bubble initial radii of 0.1–0.4 mm, and aerosol diameters of 0.01 and 0.1  $\mu\text{m}$  are used in parametric calculations. For these bubbles  $\text{Re} \lesssim 600$ , and the bubbles can be assumed to remain spherical and non-oscillating. In this parameter range, furthermore, sedimentation and inertial removal are negligibly small compared with the Brownian removal. Brownian and convective mechanisms are thus the only significant aerosol removal mechanisms, rendering the present theory applicable.

Figure 2 depicts the removal of 0.01- $\mu\text{m}$  diameter aerosols in rising bubbles, where the bubble radius remains constant. The effect of bubble expansion due to reduction of hydrostatic head in these and other calculations reported in this paper is negligibly small, and is neglected everywhere. With a constant bubble radius, convective deposition is evidently absent, and only Brownian aerosol transport takes place.

As noted in figure 2, the method due to Mills & Hoseyni (1988) is close to the present model for  $\tau \ll 1$ . For large  $\tau$ , however, their method underpredicts the quantity  $(1 - E)$  by a factor of 2–5. The discrepancy, furthermore, increases with increasing  $\tau$ . Fuchs's model (1964) on the other hand, shows significant disagreement, by as much as three orders of magnitude, with the other two methods. Fuchs's model predicts a strong dependence of  $E$  on bubble radius, which is evidently not predicted by the present theory. According to Fuchs, for a given  $\tau$ , the fractional approach

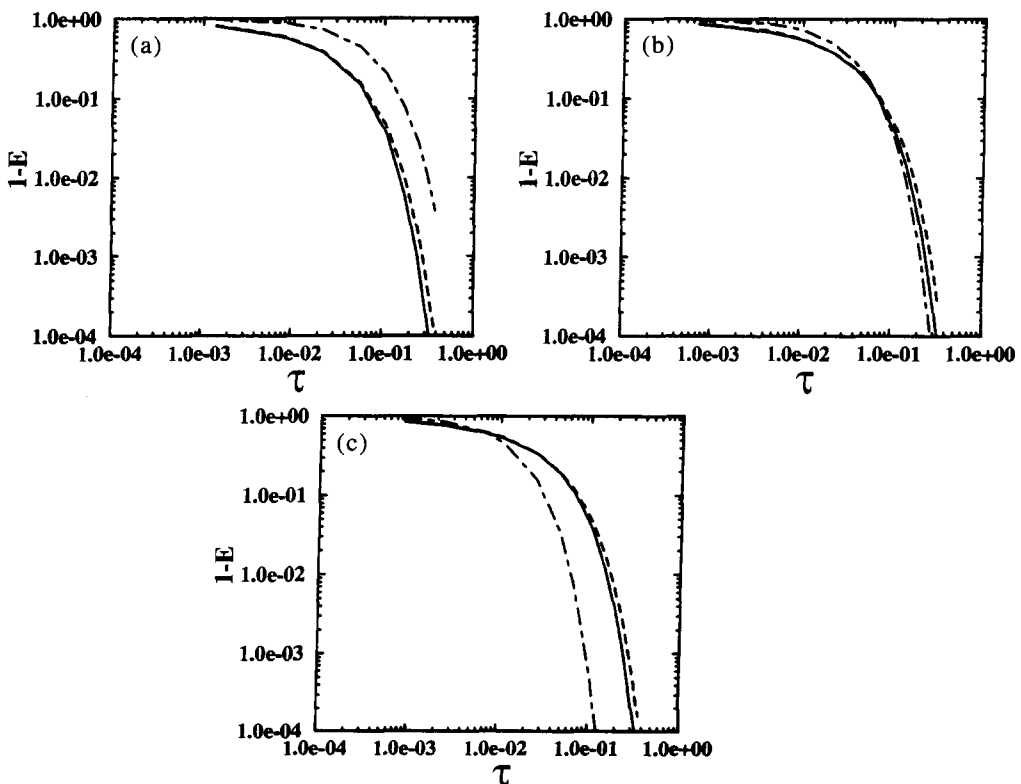


Figure 2. Transport of 0.01  $\mu\text{m}$  diameter aqueous aerosols in constant-radius air bubbles rising in water. —, Present theory; ----, Fuchs (1964); - · - ·, Mills & Hoseyni (1988). (a)  $R_B = 0.1 \text{ mm}$ ; (b)  $R_B = 0.2 \text{ mm}$ ; and (c)  $R_B = 0.4 \text{ mm}$ .

to equilibrium significantly and monotonically increases with increasing bubble radius,  $R_B$ . Thus, while underpredicting aerosol removal for  $R_B = 0.1$  mm, Fuchs's model coincidentally agrees with the present model for  $R_B = 0.2$  mm, and overpredicts aerosol removal for  $R_B = 0.4$  mm.

Figure 3 depicts the removal of  $0.1 \mu\text{m}$ -diameter aqueous aerosols in rising air bubbles with constant radii. Relatively close agreement between the present theory and the approximate method of Mills & Hoseyni (1988) can be noted. Fuchs's model, however, grossly overpredicts the aerosol removal, in particular for the  $R_B = 0.4$  mm case where the overprediction is by several orders of magnitude for  $\tau > 0.01$ .

4.2. Variable bubble radius

Variations in bubble radius, as mentioned earlier, can occur due to condensation and evaporation. With the bubble radius varying, convective and Brownian aerosol removal should both be considered.

For simplicity, calculations are performed using saturated steam bubbles rising in near-saturated water at one atmosphere. Thus,  $\rho_L = 996 \text{ kg/m}^3$ ,  $\mu_L = 2.79 \times 10^{-4} \text{ kg m/s}$ ,  $\sigma = 0.071 \text{ N/m}$  ( $\sigma$  represents the surface tension),  $\rho_G = 0.6 \text{ kg/m}^3$  and  $\mu_G = 1.2 \times 10^{-5} \text{ kg m/s}$ , were assumed. The bubbles are assumed to contain monodisperse aqueous aerosols, where  $\rho_p = 996 \text{ kg/m}^3$ . The bubbles are assumed to have initial radii equal to 0.1, 0.2 and 0.4 mm. Their normalized radii are assumed to vary at constant rates, however. Thus, with  $dy/dt = 0.5 \text{ s}^{-1}$ , the bubble radius is assumed to reduce by one half over one second of bubble rise time period.

Figures 4 and 5 compare predictions by the present theory with results obtained by using [1] and [2], where the Brownian and convective removal coefficients were obtained using [5] (Fuchs's model) and [37], respectively. The aqueous aerosols are assumed to be  $0.1 \mu\text{m}$  in diameter. The latter

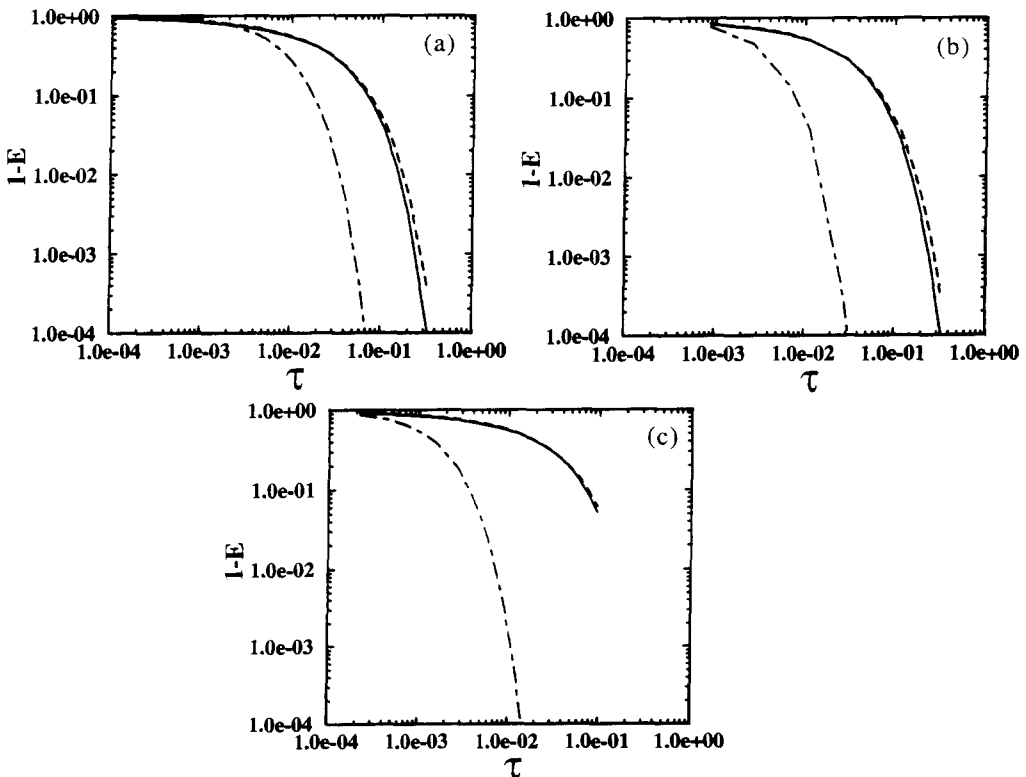


Figure 3. Transport of  $0.1 \mu\text{m}$  diameter aqueous aerosols in constant-radius air bubbles rising in water. —, Present theory; ----, Fuchs (1964); - · - ·, Mills & Hoseyni (1988). (a)  $R_B = 0.1$  mm; (b)  $R_B = 0.2$  mm; and (c)  $R_B = 0.4$  mm.



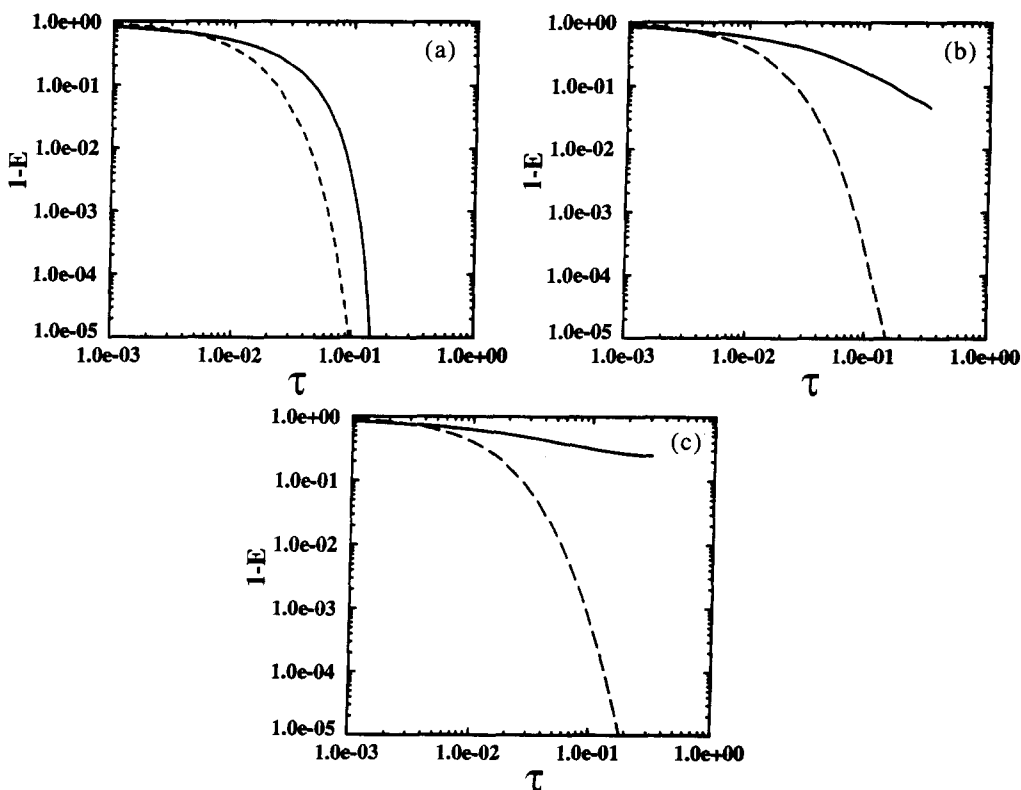


Figure 4. Transport of  $0.1 \mu\text{m}$  aqueous aerosols in variable-radius steam bubbles rising in water. Initial bubble radius =  $0.1 \text{ mm}$ . —, Present theory; ----, [1] along with [5] and [37]. (a)  $d\gamma/dt = -0.5 \text{ s}^{-1}$  ( $d\gamma/d\tau = -4.0486$ ); (b)  $d\gamma/dt = 0.5 \text{ s}^{-1}$  ( $d\gamma/d\tau = 4.0486$ ); and (c)  $d\gamma/dt = 1.0 \text{ s}^{-1}$  ( $d\gamma/d\tau = 8.097$ ).

approach, as noted, overpredicts the aerosol removal. This overprediction is particularly significant for bubbles undergoing growth. For the  $0.2 \text{ mm}$  diameter bubble with  $d\gamma/dt = 1.0 \text{ s}^{-1}$ , for example, the present theory indicates that the Brownian and convective mechanisms, which induce and retard aerosol removal, respectively, are approximately at equilibrium. The application of [1], along with [5] and [37], on the other hand, results in the prediction of very rapid removal of aerosols.

Figures 6 and 7 depict aerosol removal in steam bubbles similar to those represented in figures 4 and 5, this time containing  $0.01 \mu\text{m}$ -diameter aqueous aerosols. For aerosols this small, aerosol transport due to Brownian motion is quite rapid because of the very large aerosol diffusion coefficient (see [22]), and thus the Brownian mechanism dominates the aerosol removal process, rendering the effect of convective removal mechanism insignificant. The dominance of the Brownian mechanism is predicted by the present theory, as well as by using [1], [5] and [37]. As a result of the relative insignificance of convective aerosol removal, the results are insensitive to the rate of change of the bubble radius. The two methods, nevertheless, are in disagreement with respect to the effect of bubble radius. For  $R_B = 0.1 \text{ mm}$ , the latter approach represented by [1], [5] and [37] underpredicts aerosol removal. This is consistent with figure 2(a). For  $R_B = 0.2 \text{ mm}$ , by coincidence, it agrees with the present theory, in consistency with figure 2(b). For  $R_B = 0.4 \text{ mm}$  (not shown here for brevity), the application of [1], [5] and [37] overpredicts aerosol removal, consistent with figure 2(c).

The analysis presented in this paper shows the inadequacy of Fuchs's model for predicting the Brownian aerosol removal in bubbles. In the selected parameter range for parametric calculations in this paper sedimentation and inertial aerosol removal were both negligibly small; therefore as a result of assuming a small temperature difference between the gas and the liquid, only Brownian and convective aerosol removal mechanisms were significant. Modeling of the aerosol removal due to the combined effect of all important mechanisms, including in particular sedimentation and inertial mechanisms, is desirable.

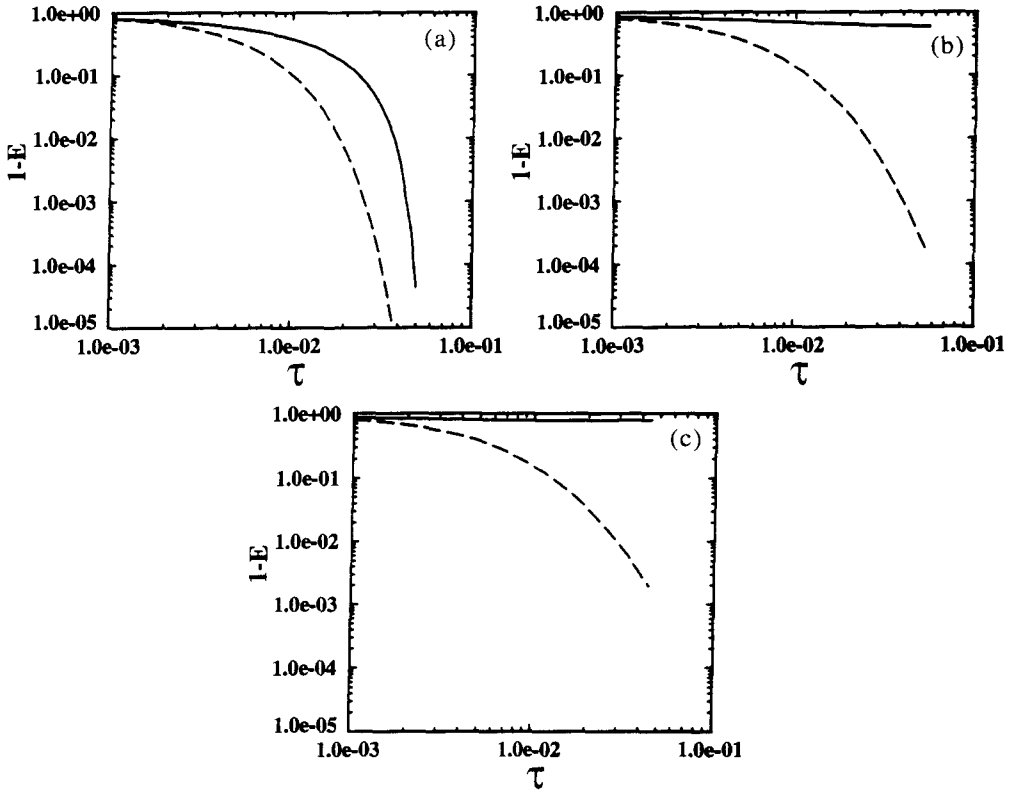


Figure 5. Transport of  $0.1 \mu\text{m}$ -diameter aqueous aerosols in variable-radius steam bubbles rising in water. Initial bubble radius =  $0.2 \text{ mm}$ . —, Present theory; ----, [1] along with [5] and [37]. (a)  $d\gamma/dt = -0.5 \text{ s}^{-1}$  ( $d\gamma/d\tau = -16.195$ ); (b)  $d\gamma/dt = 0.5 \text{ s}^{-1}$  ( $d\gamma/d\tau = 16.195$ ); and (c)  $d\gamma/dt = 1.0 \text{ s}^{-1}$  ( $d\gamma/d\tau = 32.39$ ).

5. CONCLUDING REMARKS

Aerosol removal due to the combined effects of Brownian and convective mechanisms in spherical bubbles rising in a stagnant liquid and undergoing growth and shrinkage was modeled in this paper. Modeling is based on the extension of the classical Kronig & Brink's method to bubbles with variable radii, and rigorously accounts for coupling between Brownian and convective aerosol mechanisms.

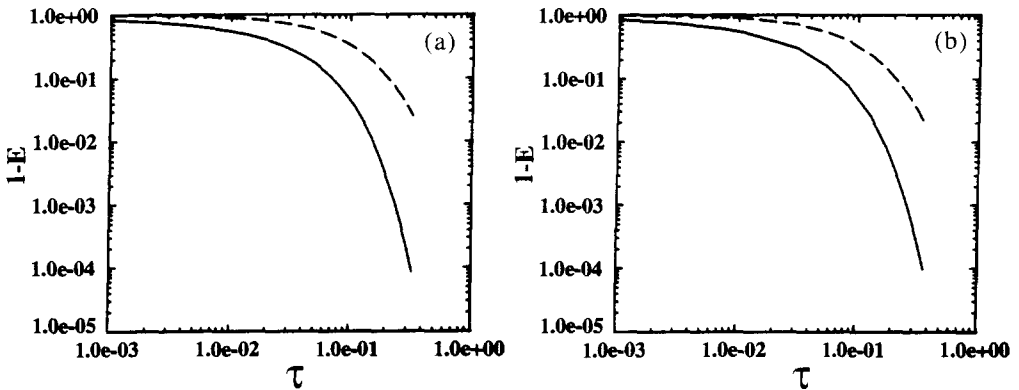


Figure 6. Transport of  $0.01 \mu\text{m}$ -diameter aqueous aerosols in variable-radius steam bubbles rising in water. Initial bubble radius =  $0.1 \text{ mm}$ . —, Present theory; ----, [1] along with [5] and [37]. (a)  $d\gamma/dt = -0.5 \text{ s}^{-1}$  ( $d\gamma/d\tau = -5.33 \times 10^{-2}$ ); and (b)  $d\gamma/dt = +1.0 \text{ s}^{-1}$  ( $d\gamma/d\tau = 0.1066$ ).

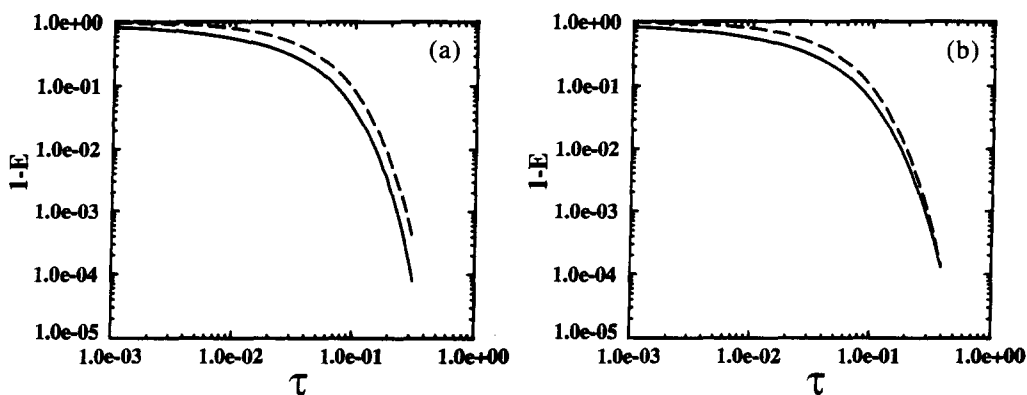


Figure 7. Transport of  $0.01\ \mu\text{m}$ -diameter aqueous aerosols in variable-radius steam bubbles rising in water. Initial bubble radius =  $0.2\ \text{mm}$ . —, Present theory; ----, [1] along with [5] and [37]. (a)  $d\gamma/dt = -0.5\ \text{s}^{-1}$  ( $d\gamma/d\tau = -0.2133$ ); and (b)  $d\gamma/dt = +1.0\ \text{s}^{-1}$  ( $d\gamma/d\tau = 0.426$ ).

Parametric calculations were performed for spherical, non-oscillating bubbles (with bubble Reynolds numbers smaller than about 600), rising in stagnant atmospheric water and carrying aqueous aerosols with diameters smaller than or equal to  $0.1\ \mu\text{m}$ . Results indicate that Fuchs's model for Brownian aerosol removal in bubbles is inadequate and can lead to significant errors.

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