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Boundary conditions and growth of mean charges for radioactive aerosol particles near absorbing surfaces

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Abstract

Particle charge evolution equation is considered in the presence of diffusion and drifts combined with bipolar and radioactive charging in the vicinity of absorbing surfaces (walls). Boundary conditions to be prescribed for solving the mean charge evolution equation have been derived. It is shown that the mean charge $J(x)$ at a distance x from the surface ($x = 0$) satisfies the boundary condition

$$\left. \frac{\partial J}{\partial x} \right|_{x=0} = \frac{U(0)}{2D(0)} [\alpha J(0) + \sigma_0^2],$$

where $U(x)$ and $D(x)$ are the drift velocity and diffusion coefficients respectively, $\alpha = 1$ (0) for radioactive (non-radioactive) aerosols, and σ_0^2 is the variance of the Boltzmann charge distribution. Using this, specific cases of charge build-up of radioactive particles undergoing turbulent diffusion near walls is examined. Radioactive particle charges are found to increase both for zero and negative electric fields, although the increases are far smaller than those expected from stationary-state formulae. The results are further discussed. © 2002 Elsevier Science Ltd. All rights reserved.

1. Introduction

The evolution of charge distributions on aerosol particles suspended in stagnant and homogeneous ionic environments is commonly described by stochastic charging equations (Gunn, 1954; Hoppel & Frick, 1986) which account principally for charge fluctuations. In many situations involving particle charging in the presence of external forces or flows (Hoppel & Gathman, 1970; Stechkina, Kirsh, & Zagnit'ko, 1982; Romay, Pui, & Adachi, 1991) it becomes necessary to include effects of diffusion and drift in addition to charge fluctuations. These situations are likely to be further relevant for the cases of radioactive aerosol behaviour in reactor containment studies as pointed out

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by Clement and Harrison (2000). Mayya and Malvankar (1993) and Mayya (1994) discussed the process of electro-migration of non-radioactive particles in bipolar ionic environments and formulated a description based on charging induced diffusion. They, however, ignored molecular and turbulent diffusion processes and did not include radioactive particles.

Through a series of papers, Clement and Harrison (1992, 1995, 2000) and Clement, Calderbank, and Harrison (1994) have systematically examined the theory of self-charging of radioactive aerosols. In their recent work, Clement and Harrison (2000; referred to hereafter as CH2000) set up the most general form of particle charge evolution equation, by including self-charging, bipolar charging, diffusion and drift. From these, they derived the reduced equations for the evolution of the number concentration and the mean charge of the particles. Apart from being simpler than the full stochastic charging equation, the reduced equations provide direct descriptions for the quantities of practical interest, viz., the concentration profiles and the mean charges.

An important aspect discussed by CH2000 pertains to the increase of mean charges of radioactive particles in confined geometries. This is due to a suppression of negative ion concentrations resulting from the reduced ranges of beta particles as well as increased wall losses of ions. Gensdarmes, Boulaud, and Renoux (2000) appear to have found a qualitative evidence for the increased radioactive particle charges from their flow tube experiments. By analogous reasoning, one may deduce that radioactive particle charges would, in general, increase as they approach a surface, due to the lowering of ion concentrations in its vicinity. The increased particle charges will not only enhance their deposition rates through image-induced effects, but also cause space charge effects. This situation might be quite relevant in the context of reactor accident scenarios, wherein radioactive particles will be released into an ionized air space in the containment and will begin to migrate to the walls with increasing charges. As a first guess, if one employs the stationary-state formula (Clement & Harrison, 1992) for the charge build-up, viz.,

$$J = \frac{\eta \varepsilon_0}{e \mu^- n^-},$$

(where, J is the mean charge, ε_0 permittivity of free space, e -elementary charge, μ^- , n^- negative ion mobility and concentration, respectively), one obtains the unphysical result that the particle charges will be infinity at the walls since n^- vanishes at that point. This arises because of our neglecting the migration effects originating from particle diffusion and drift motions (if existing), which act as limiting factors in the growth of charges. The example considered above, thus, focuses on the necessity of having to include particle diffusion and other motions in order to arrive at reasonable estimates of charges of radioactive particles.

However, there exists a major difficulty in solving the mean charge evolution equation derived by CH2000. Their derivation does not specify the boundary condition for the charge at the surface, which is one of the two necessary conditions required for solving the boundary value problems associated with diffusion equation. While one of the boundary conditions may be specified as the value of the mean charge far away from the surface, the other boundary condition at the surface is not at all obvious. It may be added that in the case of the equation for the number concentration ($N(x)$), the two boundary conditions are well known, viz., the absorption boundary condition, $N(0) = 0$ and the asymptotic condition in the bulk, $N(x) \rightarrow N_\phi$ as $x \rightarrow \infty$. There is no physical basis for extending the absorption condition for the mean charge since (see Section 2) it is defined as the ratio of the charge density and the number concentrations both of which vanish at the surface, and their ratio

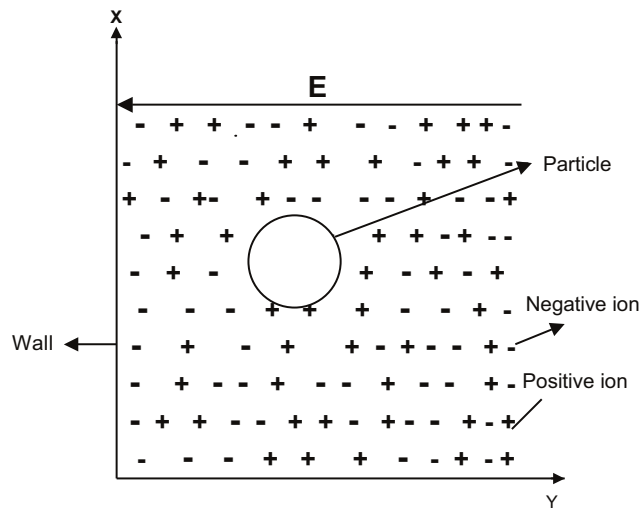


Fig. 1. Schematic diagram of particle-ion system near an absorbing wall.

might have a finite, nonzero limit. The situation thus calls for a separate analysis of the mean charge evolution equation starting from fundamental considerations.

This paper aims at such an analysis and obtains a derivation of the boundary conditions for the mean charge in a consistent manner. The key assumption is the validity of the Smoluchowski absorption condition for the charge distribution function, namely that it vanishes at the absorbing surface regardless of charge. This assumption essentially provides a simple framework and is not strictly valid from a kinetic point of view, which takes inertial effects (Crump & Seinfeld, 1981) and charge-velocity fluctuations in an electric field, into account. These aspects are not considered here.

2. Mean charge evolution equation

In order to obtain the boundary conditions, it is necessary to outline, in brief, the derivation of the equations proposed by CH2000 for the number concentration and the mean charge. While, the equations of CH2000 were presented in three-dimensional form, we restrict to the one-dimensional case of aerosol particles (radioactive or otherwise) undergoing bipolar charging in an air space bounded on one side by an absorbing wall (Fig. 1). In addition, let us impose diffusive and electric field-induced drift motions in the direction normal to the wall, and possibly a convective flow parallel to the wall. Let the origin be fixed at a point on the wall with X -axis normal to it and Y -axis along the direction of flow along the wall. Sufficiently far from the wall ($x \rightarrow \infty$), the particles as well as the ions are assumed to be well mixed having uniform and steady concentrations denoted by N_ϕ , n_∞^+ and n_∞^- , respectively. It is presumed that these quantities are sustained due to external sources coupled with turbulence in the bulk air space. The ions would set up differing profiles in the vicinity of the wall when electric fields are present.

Let $P(q, x, y, t)$ denote the concentration of particles having charge unit q , at the point x, y at time t . Let V be the convective flow velocity (assumed constant) in the Y -direction, $U(x)$ be the field induced particle drift velocity per unit charge along X and $D(x)$ be the diffusion coefficient involving both Brownian and turbulent components. If the local electric field is $E(x)$, $U(x) = B.E(x)$ where B is the particle mobility per unit charge. $P(q, x, y, t)$ satisfies the equation

$$\frac{\partial P}{\partial t} + V \frac{\partial P}{\partial y} + \frac{\partial}{\partial x} \left[-D(x) \frac{\partial P}{\partial x} + qU(x)P \right] = Q(q, x, y, t), \quad (1)$$

where

$$\begin{aligned} Q(q, x, y, t) = & [K^+(q-1)n^+(x, y) + \eta]P(q-1, x, y, t) \\ & + [K^-(q+1)n^-(x, y)]P(q+1, x, y, t) \\ & - [K^+(q)n^+(x, y) + \eta + K^-(q)n^-(x, y)]P(q, x, y, t). \end{aligned} \quad (1a)$$

The terms contained in $Q(x, y, t)$ account for bipolar charging due to ions {through the attachment coefficients, $K^\beta(q)$ } and due to beta emissions with a positive charge generation rate of η per particle. As the solution of Eq. (1) is quite formidable, and also since the knowledge of the full charge distribution near surfaces is generally not of practical interest, it suffices to obtain information on the number density $N(x, y, t)$ and the mean charge $J(x, y, t)$. To this end, we reduce Eq. (1) upon defining the following moments with respect to q :

$$\begin{aligned} N(x, y, t) \equiv P_0(x, y, t) &= \sum_q P(q, x, y, t), \\ P_1(x, t) &= \sum_q qP(q, x, y, t), \\ P_2(x, t) &= \sum_q q^2P(q, x, y, t). \end{aligned} \quad (2)$$

From Eq. (1), the first two moments defined above may be shown to satisfy the following equations:

$$\frac{\partial N}{\partial t} + V \frac{\partial N}{\partial y} + \frac{\partial}{\partial x} \left[-D(x) \frac{\partial N}{\partial x} + U(x)P_1 \right] = 0, \quad (3)$$

$$\frac{\partial P_1}{\partial t} + V \frac{\partial P_1}{\partial y} + \frac{\partial}{\partial x} \left[-D(x) \frac{\partial P_1}{\partial x} + U(x)P_2 \right] = \phi(x, y, t), \quad (4)$$

where

$$\phi(x, y, t) = \sum_q [K^+(q)n^+(x, y) + \eta - K^-(q)n^-(x, y)]P(q, x, y, t). \quad (5)$$

Eqs. (3) and (4) are not closed because of their dependence on P_2 and ϕ . If we set-up a moment equation for P_2 , it would depend upon higher moments and so on. Similarly the quantity ϕ , being a sum over the attachment coefficients weighted by the charge distribution, depends on all orders

of moments. Closure is achieved by relating P_2 and ϕ to the lower moments P_1 and N , through an approximation. For this, one sets up a differential form of the charging term Q valid in the continuum limit, (Mayya & Malvankar, 1993) and develops asymptotic solutions around the mean charge J . For a constant J , the asymptotic charge distribution has a shifted Gaussian form given by Mayya and Malvankar (1993) and Clement and Harrison (1995)

$$P_{\text{asy}}(q, x, t) = \frac{N(x, y, t)}{\sigma\sqrt{2\pi}} \exp\left[-\frac{(q - J)^2}{2\sigma^2}\right], \quad (6)$$

where, σ is the standard deviation of the charge distribution. For the purpose of achieving closure, we assume that Eq. (6) continues to hold in the sense of a local equilibrium approximation even when J is space–time dependent. With this, the reduced equations derived for J and N would represent diffusion approximations, as they would be accurate upto derivatives of second order in space and first order in time. From Eq. (6) it follows that

$$P_2(x, y, t) \approx (J^2 + \sigma^2)N(x, y, t), \quad (6a)$$

where $J \equiv J(x, y, t)$ is defined as the ratio of the particle charge density to the number concentration, i.e.,

$$J(x, y, t) = \frac{P_1(x, y, t)}{N(x, y, t)}. \quad (7)$$

In view of the discussions following Eq. (6), the quantity σ^2 may be prescribed as the variance of the local equilibrium charge distribution, generalised to include the case of radioactive aerosols by Clement and Harrison (1995), i.e.,

$$\sigma^2 = \alpha J + \sigma_0^2, \quad (8)$$

where, $\alpha = 0$ for nonradioactive particle,

$$\alpha = 1 \text{ for radioactive particles.} \quad (8a)$$

The quantity σ_0^2 is the variance of the Boltzmann charge distribution (for nonradioactive aerosols), i.e.,

$$\sigma_0^2 = \frac{r}{r_c}. \quad (8b)$$

where, r is the radius of aerosol particle and $r_c \equiv e^2/4\pi\epsilon_0k_bT \approx 56 \text{ nm}$ at 300 K, is the Coulomb radius.

Using Eqs. (6a) and (5), the K 's may be expanded around $q = J$ as a Taylor series and the sum in Eq. (5) may be replaced by an integral. With this, in the limit of $R \gg r_c$, the distribution-weighted sum of K 's may be replaced, to a good accuracy, by the values of K 's at the mean charge J multiplied by N . i.e.,

$$\phi(x, y, t) \approx \psi(x, y, J).N(x, y, t), \quad (9)$$

where, $\psi(x, y, J) \equiv [K^+(J)n^+(x, y) + \eta - K^-(J)n^-(x, y)]$.

Eq. (9) can be shown to be exact for the cases of symmetrical ion charging and also in the limit of strong positive charging due to beta emissions (CH2000). Substituting Eqs. (6a), (7) and (9)

into Eqs. (3) and (4), the following closed system of equations is obtained for N and J :

$$\frac{\partial N}{\partial t} + V \frac{\partial N}{\partial y} + \frac{\partial}{\partial x} \left[-D(x) \frac{\partial N}{\partial x} + U(x) J N \right] = 0, \quad (10)$$

$$\frac{\partial J}{\partial t} + V \frac{\partial J}{\partial y} + U(x) J \frac{\partial J}{\partial x} - \frac{\partial}{\partial x} \left[D(x) \frac{\partial J}{\partial x} \right] - 2D(x) \frac{1}{N} \frac{\partial N}{\partial x} \frac{\partial J}{\partial x} + \frac{1}{N} \frac{\partial}{\partial x} [U(x) \sigma^2 N] = \psi(x, y, J). \quad (11)$$

It may be added that Eqs. (10) and (11) may also be arrived at from an alternative, somewhat mathematically complicated approach that does not explicitly invoke the assumption of a shifted Gaussian (Eq. (6)), but involves recasting Eq. (1) in terms of a new set of co-ordinates, $q' = q - J(x, t)$, $x' = x$, $t' = t$. Upon applying closure to the moment hierarchy of this equation, one may arrive at Eqs. (10) and (11) through a series of systematic approximations.

3. Derivation of boundary conditions for $J(X)$

As discussed in the Introduction, it is necessary to prescribe boundary conditions (BCs) for solving Eq. (11) near surfaces. For simplicity of derivation, we consider a steady-state situation in which particles are being constantly driven to the wall from a steady concentration in the bulk. Under the assumption of steady-state, the moment equations (Eqs. (3) and (4)) reduce to

$$V \frac{\partial N}{\partial y} + \frac{\partial}{\partial x} \left[-D(x) \frac{\partial N}{\partial x} + U(x) P_1 \right] = 0, \quad (12)$$

$$V \frac{\partial P_1}{\partial y} + \frac{\partial}{\partial x} \left[-D(x) \frac{\partial P_1}{\partial x} + U(x) P_2 \right] = \phi(x, y). \quad (13)$$

Given n_{∞}^{\pm} , a corresponding steady-state charge distribution would exist on particles having a mean charge J_{ϕ} . On the wall, we invoke the Smoluchowski absorption condition for particle charge distribution, as well as for ion concentrations:

$$P(q, 0, y) = n^+(0, y) = n^-(0, y) = 0. \quad (14a)$$

This automatically implies, in view of Eqs. (2) and (5), that

$$N(0, y) = P_1(0, y) = P_2(0, y) = \phi(0, y) = 0. \quad (14b)$$

Next, we assume, quite justifiably, that the parameters $D(x)$, $U(x)$ and all the moments of the charge distribution function are Taylor expandable in x around $x=0$. The expansion coefficients may have y -dependencies. In view of Eq. (14b), the constant term of the Taylor expansion will be zero

for the moments and hence one may write

$$N(x) = a_1x + a_2x^2 + \dots, \quad (15a)$$

$$P_1(x) = b_1x + b_2x^2 + \dots, \quad (15b)$$

$$P_2(x) = c_1x + c_2x^2 + \dots, \quad (15c)$$

$$\phi(x) = \phi_1x + \phi_2x^2 + \dots, \quad (15d)$$

$$D(x) = d_0 + d_1x + d_2x^2 + \dots, \quad (15e)$$

$$U(x) = u_0 + u_1x + u_2x^2 + \dots. \quad (15f)$$

Upon substituting Eqs. (15a)–(15f) in Eq. (12), and noting that the coefficients are functions of y only, one may write

$$\frac{d}{dx}[-(d_0 + d_1x + \dots)(a_1 + 2a_2x + \dots) + (u_0 + u_1x + \dots)(b_1x + \dots)] + V[xa'_1 + \dots] = 0. \quad (16)$$

Upon collecting the coefficients of x^0 in Eq. (16), one obtains

$$2a_2d_0 + d_1a_1 - u_0b_1 = 0$$

or

$$a_2 = \frac{u_0b_1 - a_1d_1}{2d_0}. \quad (17a)$$

Similarly, upon substituting Eq. (15a)–(15f) in Eq. (13), one obtains

$$b_2 = \frac{u_0c_1 - b_1d_1}{2d_0}. \quad (17b)$$

From Eqs. (7), (15a) and (15b), the mean charge J may be given by

$$J(x) = \frac{P_1}{N} = \frac{b_1 + b_2x + \dots}{a_1 + a_2x + \dots} \Rightarrow J(0) = \frac{b_1}{a_1}. \quad (18)$$

Therefore, $b_1 = J(0)a_1$.

Let us evaluate the gradient of the mean charge w.r.t. x at $x = 0$: Upon differentiating Eq. (18) w.r.t. x and setting $x = 0$, one obtains

$$\left. \frac{\partial J}{\partial x} \right|_{x=0} = \frac{b_2a_1 - a_2b_1}{a_1^2}. \quad (19a)$$

Upon substituting for b_2 and a_2 from Eqs. (17a) and (17b), and for b_1 from Eq. (18), Eq. (19a) yields

$$\left. \frac{\partial J}{\partial x} \right|_{x=0} = \frac{u_0}{2d_0a_1^2}[c_1a_1 - a_1^2J^2(0)]. \quad (19b)$$

It is required to eliminate c_1 from Eq. (19b) in terms of a_1 . For this, invoke the second moment closure approximation (Eq. (6a)) along with the expansion for P_2 given in Eq. (15c). It easily follows that

$$c_1 = a_1 \{J^2(0) + \alpha J(0) + \sigma_0^2\}. \quad (20a)$$

Upon substituting this in Eq. (19b) and simplifying, one obtains

$$\left. \frac{\partial J}{\partial x} \right|_{x=0} = \frac{u_0}{2d_0} [\alpha J(0) + \sigma_0^2]. \quad (20b)$$

Since u_0 and d_0 are the drift and the diffusion coefficients evaluated at the boundary, i.e., $U(0)$ and $D(0)$, we obtain the required boundary condition for the mean charge $J(x)$ in terms of the known parameters:

$$\left. \frac{\partial J}{\partial x} \right|_{x=0} = \frac{U(0)}{2D(0)} [\alpha J(0) + \sigma_0^2]. \quad (21a)$$

Since $\alpha = 0$ for nonradioactive particles, Eq. (21a) implies a “prescribed gradient” BC at $x = 0$. For radioactive particles, $\alpha = 1$ and the boundary condition is of the “mixed type”. When no field induced drift exists ($U(0) = 0$) both cases reduce to the Neumann type boundary condition, viz.,

$$\left. \frac{\partial J}{\partial x} \right|_{x=0} = 0 \quad \text{for } U(0) = 0. \quad (21b)$$

This completes our derivation of the boundary conditions for Eq. (11) under the assumption of a steady-state. One may go through the same arguments for time dependent cases as well and arrive at the BC in Eq. (21a).

4. Growth of radioactive particle charges

In order to illustrate the usefulness of the BCs derived above, typical case studies will now be taken up for the growth of mean charge of radioactive aerosols near absorbing surfaces by solving Eqs. (10) and (11) under steady-state conditions. For simplicity, situations without convective flows are only considered (i.e., $V = 0$). When no electric field is present, no net space charge exists; even then, radioactive particles would gain excess mean charges while approaching the boundary due to a suppression of negative ion concentration in its vicinity. However, when electric field is present, space charge effects have to be taken into account by coupling the Poisson’s equation to the ion balance equation. For the present, the aerosol-ion system is treated in the dilute limit, i.e., no space charge effects are included. Space-charge effects become significant when the parameter $s \equiv 4\pi r_c D_i n_\infty / k_e \gg 1$, where $D_i n_\infty$ are the molecular diffusion coefficient and bulk concentrations of ions and $k_e (s^{-1})$ is the parameter of quadratic turbulence (see below). For the parameters used in the foregoing calculations, $s \sim 1$ and hence one can expect small corrections due to space-charge considerations. These are ignored for the present and we focus mainly on illustrating the applicability of the BCs to the charging problem.

Eq. (11) now becomes one-dimensional dependent only on x . Following Crump and Seinfeld (1981), the turbulent diffusion coefficient is assumed to have a quadratic form, i.e.,

$$D(x) = D_0 + k_e x^2. \quad (22)$$

where k_e is the coefficient (s^{-1}) of eddy diffusion. One may write down similar form for the ion diffusion coefficients as well, by replacing the Brownian diffusion coefficient D_0 by the molecular diffusion coefficient for ions, D_i .

4.1. Case (i): No electric field ($E, U = 0$)

In the absence of external fields, Eq. (10) yields the following particle concentration profile $N(x)$ (Crump and Seinfeld, 1981):

$$N(x) = N_\infty \frac{2}{\pi} \arctan \left[\frac{x}{\delta_p} \right], \quad (23a)$$

where, $\delta_p = (D_0/k_e)^{1/2}$, is the particle boundary layer thickness.

Since $U = 0$, the ion profiles will be the same for both positive and negative ions assuming that their mobility differences are negligible. They have the same form as $N(x)$, with δ_p being replaced with the ion boundary layer thickness, $\delta_i = (D_i/k_e)^{1/2}$:

$$n^\pm(x) = n_\infty \frac{2}{\pi} \arctan \left[\frac{x}{\delta_i} \right]. \quad (23b)$$

It may be noted from the arguments of CH2000, that for radioactive aerosols, with considerable positive charge build-up, positive ion concentration is not of serious relevance and hence $K^+(J)n^+$ term may be neglected in ψ defined in Eq. (9). Also, in the continuum regime, one may approximate, $K^-(J) \approx e\mu J/\epsilon_0$. With these, Eq. (11) reduces to

$$-\frac{\partial}{\partial x} \left[D(x) \frac{\partial J}{\partial x} \right] - 2D(x) \frac{1}{N} \frac{\partial N}{\partial x} \frac{\partial J}{\partial x} = \eta - \frac{e\mu n^-(x)}{\epsilon_0} J \quad (24)$$

subject to the BCs

$$J(\infty) = J_\infty = \frac{\eta\epsilon_0}{e\mu n^-(\infty)} \quad (24a)$$

and

$$J'(0) = 0 \quad (\text{from Eq. (21b)}).$$

Since solutions to Eq. (24) are not analytically tractable, we have integrated it numerically. The parameters chosen are $\eta = 1 \text{ s}^{-1}$, $k_e = 1 \text{ s}^{-1}$, $r = 0.1 \text{ }\mu\text{m}$ and $n_\infty^\pm = 4 \times 10^5 \text{ cm}^{-3}$, representing moderate radioactivity, ionization levels and weak turbulence. The corresponding particle mean charge in the bulk can be obtained as $J_\phi = 1$. Also the boundary layer parameters turn out to be $\delta_p = 15 \text{ }\mu\text{m}$ for particles and $\delta_i = 1.8 \text{ mm}$ for ions. Information on N_ϕ is not required since only $N'(x)/N(x)$ enters into Eq. (24).

The variations of the particle and the ion profiles as a function of a dimensionless distance, $x^* = x/\delta_p$, are shown in Fig. 2. While the particle profiles are nearly uniform except within distances of a few δ_p 's near the wall, ions attain uniform values at much larger distances, i.e., beyond about 1 cm.

The numerically computed variation of $J(x)$ as a function of x^* is presented in Fig. 3. From this, it may be seen that the particle charge increases monotonically as one approached the wall from

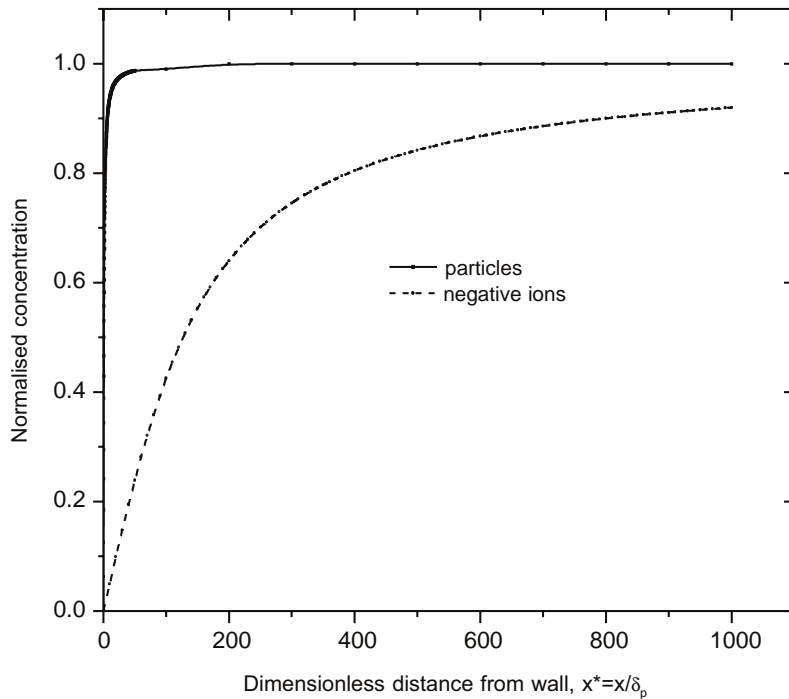


Fig. 2. Variation of the normalized concentrations of the particles and negative ions with respect to the dimensionless distance $x^*(=x/\delta_p)$ from the wall. The parameters are: $E=0$, $k_e=1\text{ s}^{-1}$, $r=0.1\text{ }\mu\text{m}$, $D_i=0.035\text{ cm}^2\text{ s}^{-1}$, and $\delta_p=15\text{ }\mu\text{m}$.

the bulk. At the wall, the charge attained is more than four times its asymptotic value. Although this is considerable, the charge build up does not proceed indefinitely, in spite of the negative ion concentration being zero at the boundary (see Introduction). This is mainly because of the nonzero migration velocity of particles brought about by the diffusive flux at the wall. The charge decays quite rapidly with distance initially, but approaches its asymptotic value far away from the wall in a reciprocal fashion with respect to the negative ion profile.

Although Eq. (24) cannot be solved analytically, we tried an approximate approach by linearizing the $\arctan[x]$ function around $x=0$, where the charge build up is expected to occur. Eq. (24) can then be solved in terms of Airy functions, $\text{Ai}(z)$ (Abramowitz & Stegun, 1968) leading to the following formula for charge at the boundary:

$$\frac{J(0)}{J_\infty} = \left(\frac{\pi\delta_i}{2\delta_p} \right)^{2/3} \cdot \left(\frac{e\mu n_\infty^-}{\varepsilon_0 k_e} \right)^{1/3} \left| \frac{\text{Ai}'(0)}{\text{Ai}(0)} \right|. \quad (24b)$$

The obtained charge build up was too large ($J(0)/J_\phi \sim 25$) as compared to the numerical result ($J(0)/J_\phi \sim 4.4$) for the parameters chosen. Nevertheless, the analytical formula in Eq. (24b) indicates parametric dependencies, viz., $J(0)/J_\phi$ increases with n_ϕ and particle radius r (since

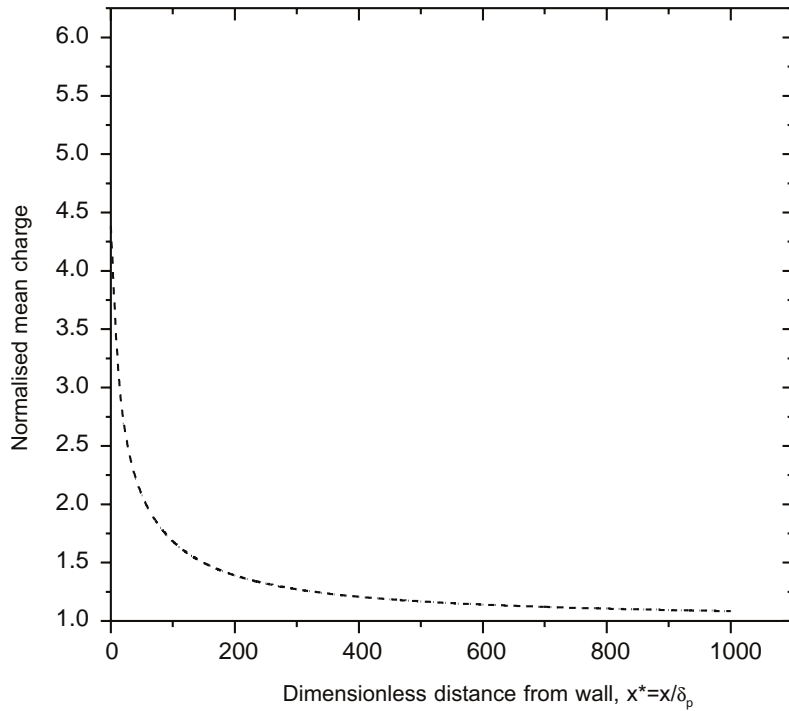


Fig. 3. Variation of the normalized mean charge $J(x)/J(\infty)$ of the particle with respect to x^* corresponding to the case in Fig. 2. $\eta = 1 \text{ s}^{-1}$, $n_{\infty}^{\pm} = 4 \times 10^5 \text{ cm}^{-3}$ and $E = 0$.

$\delta_p \sim D_0^{1/2} \sim r^{-1/2}$) and decreases with k_e . These trends have been found valid in numerical computations as well (not shown).

4.2. Case (ii): Charging with electric fields

The situations with electric fields are far more complicated, since strictly speaking, ion and the electric field profiles have to be solved self-consistently using ion migration and Poisson’s equations to account for space charge effects (Hoppel & Gathman, 1970; Stechkina et al., 1982). For reasons mentioned at the beginning of this section, we ignore this and consider only constant electric field, along the X -direction. The Crump and Seinfeld (1981) solution for a uniform force field leads to the following positive and negative ion profiles:

$$n^{\pm}(x) = \frac{\exp[\pm eE\delta_i/k_bT \arctan(x/\delta_i)] - 1}{\exp[\pm \pi/2eE\delta_i/k_bT] - 1}. \tag{25}$$

When E is positive (directed away from the wall), $n^-(x)$ is nearly constant except within a small distance at the wall, and $n^+(x)$ increases slowly as one moves away from it. This case is not expected to yield significant increase in the charges of radioactive aerosols. On the other hand, when E is

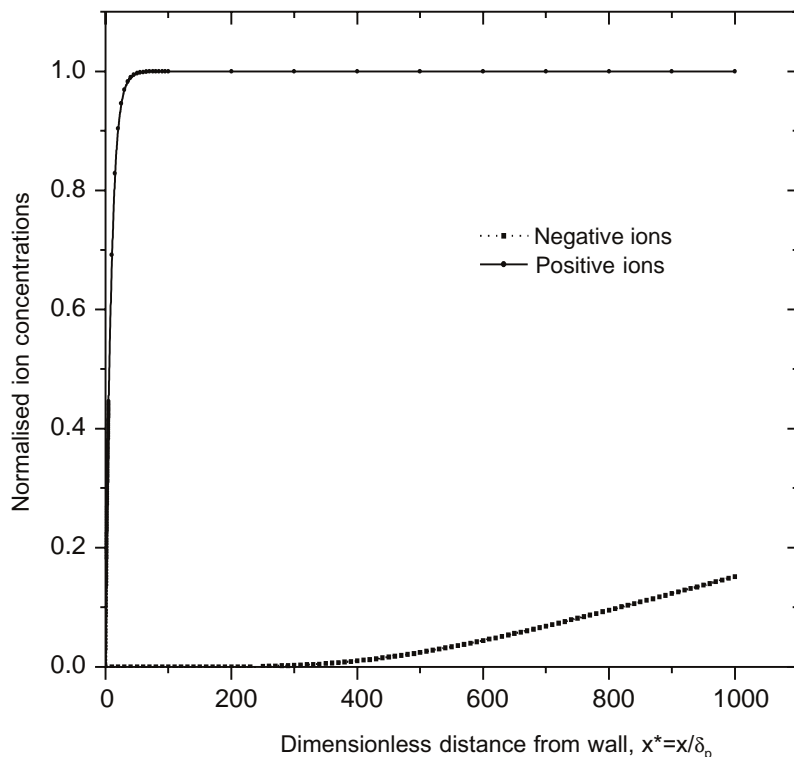


Fig. 4. Variation of the normalized negative ion and positive ion concentrations with respect to the dimensionless distance x^* for an electric field directed towards wall. The parameters are: $k_e = 1 \text{ s}^{-1}$, $r = 0.1 \text{ }\mu\text{m}$ and $D_i = 0.035 \text{ cm}^2 \text{ s}^{-1}$ and $E = -2 \text{ V cm}^{-1}$.

negative, negative ions will be pushed away from the wall and considerable self-charging may be expected. Hence we treat the negative E case, first.

4.2.1. E negative

In what follows, only weak field situations are considered so that while the ion profiles are polarized, the field adds negligible drift velocity to the particle (since their mobilities are small). Eq. (10) may now be decoupled from Eq. (11) so that $N(x)$ has essentially the same form as that given in Eq. (23a).

We examine the numerical solution to the equation for J (Eq. (11) under steady state and $V = 0$). The particle charging term, $\psi(x, J)$ (Eq. (9)) is given by the rhs of Eq. (24). The mixed boundary condition (Eq. (21a)) has been used. Electric fields of -2 and -4 V/cm are considered (wall acts as negative electrode). Other parameters are kept the same as in the zero field case. Fig. 4 shows the positive and the negative ion concentration profiles (for $E = -2 \text{ V/cm}$) as a function of normalized distance $x^*(=x/\delta_p)$. As expected, considerable ion polarization is seen even at large values of $x^*(\sim 1000)$. The variation obtained in $J(x)$ as a function of x^* for the cases with $E = -2$ and

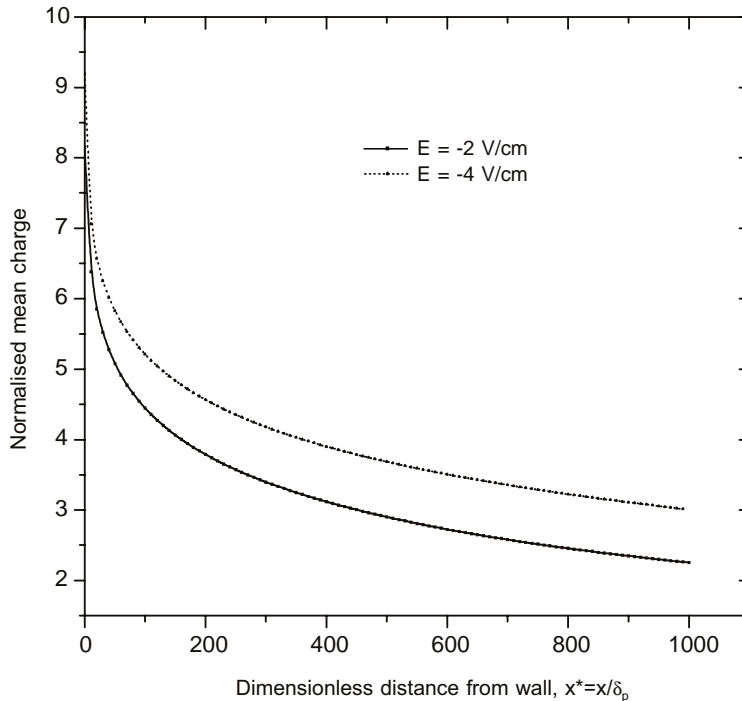


Fig. 5. Variation of the normalized mean charge, $J(x)/J(\infty)$ with respect to x^* when the electric field is directed towards the wall. The parameters $k_e, r, D, \eta, n_\infty^\pm$ are the same as in Figs. 3 and 4.

-4 V/cm are shown in Fig. 5. In the former case, the value of $J(0)/J_\phi$ is about 8.2 which is a factor about 2 times larger than that in the zero field case. This increases slightly, i.e., to about 9.7 at $E = -4$ V/cm. Thus, beyond some value, increases in E lead to marginal increases in charges. It may also be noted from Fig. 5 that the charges fall off more slowly with x^* as compared to the zero field case.

4.2.2. E positive

As the negative ions are now closer to the wall, one does not expect any significant radioactive particle charging. Computations of J have been made with fields of $+2$ and $+4$ V/cm and are shown in Fig. 6. The ion profiles would appear similar to Fig. 4 excepting that the positive and the negative ion profiles would be interchanged. Interestingly, as may be seen in Fig. 6, the $J(x)/J_\phi$ curves show peaks for positive fields. The peak values are 1.80 and 1.35 at $E = +2$ and $+4$ V/cm occurring at around $x^* \sim 2$, respectively. The corresponding values at $x = 0$ are 1.7 and 1.2, respectively. The peak is a result of the boundary condition (Eq. (21a)), which prescribes that $J'(0)$ is positive since $U(0)$ and $J(0)$ are positive. Hence $J(x)$ should increase as x^* increases from zero. However, the increase in $n^-(x)$ rapidly counteracts the increasing charge, leading to the peak for the latter and a steady decrease thereafter.

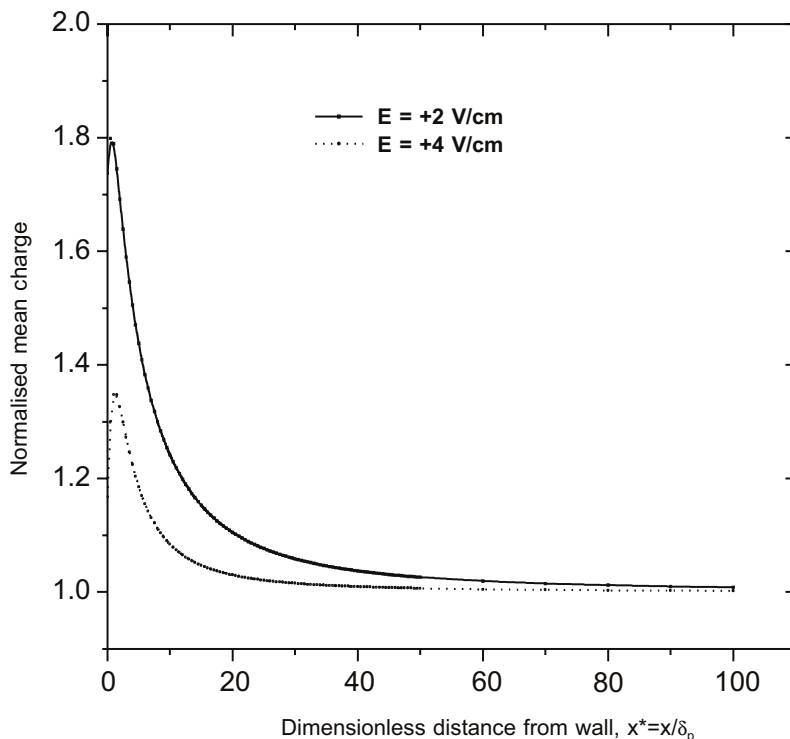


Fig. 6. Variation of the normalized mean charge, $J(x)/J(\infty)$, with respect to x^* when the electric field is directed away from the wall. The parameters $k_e, r, D_i, \eta, n_{\infty}^{\pm}$ are the same as in earlier figures.

5. Conclusions

In this paper, the appropriate boundary conditions to the equation of Clement and Harrison (2000) describing the mean charge evolution of aerosol particles undergoing bipolar charging have been derived. These conditions are mathematical necessities for computing charge build-up on aerosol particles in the presence of diffusion onto absorbing surfaces. The derivation includes the cases of both radioactive and nonradioactive particles. The application of the boundary conditions is demonstrated by examining the build-up of charges on radioactive particles (beta emitters) through numerical solution of the equations at zero and weak electric fields originating from plane surfaces. Their charges show up to about ten-fold increase near the surfaces maintained at negative potentials. These charges can enhance the wall deposition rates due to image forces and possibly due to self-generated electric fields proposed by CH2000, for radioactive particles. In fact, in the recent PHEBUS experimental studies, Jones and Kissane (2000) reported an increased deposition of radioactive particle on heated surface (thermophoretic suppression of deposition), and electrical effects were conjectured as possible explanations. On the whole, these studies support the observations of Clement and Harrison (2000) that charge effects might be important in governing radioactive aerosol removal at surfaces. In the context of nuclear aerosols, the self-generated fields proposed by them will not be uniform in space, but will increase towards the boundary from a zero value in the bulk region. It is quite

important to carry out a detailed investigation of these fields by considering charge generation due to radioactive emissions and space-charge effects due to ions along with the particle charging equations presented in this work.

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References

- Abramowitz, M., & Stegun, I. A. (1968). *Handbook of mathematical functions*, Vol. VIII. NBS.
- Clement, C. F., & Harrison, R. G. (1992). The charging of radioactive aerosols. *Journal of Aerosol Science*, 23, 481–504.
- Clement, C. F., & Harrison, R. G. (1995). Charge distribution and coagulation of radioactive aerosols. *Journal of Aerosol Science*, 26, 1207–1225.
- Clement, C. F., & Harrison, R. G. (2000). Enhanced localized charging of radioactive aerosols. *Journal of Aerosol Science*, 31, 363–378.
- Clement, C. F., Calderbank, D., & Harrison, R. G. (1994). Radioactive aerosol charging with spatially varying ion concentrations. *Journal of Aerosol Science*, 25, 623–637.
- Crump, J. G., & Seinfeld, J. H. (1981). Turbulent deposition and gravitational sedimentation of an aerosol in a vessel of arbitrary shape. *Journal of Aerosol Science*, 12, 405.
- Gensdarmes, F., Boulaud, D., & Renoux, A. (2000). Experimental study of the electrical charging of radioactive aerosols. *Journal of Aerosol Science*, 31, S1025–S1026.
- Gunn, R. (1954). Diffusion charging of atmospheric droplets by ions and the resulting combination coefficients. *Journal of Meteorology*, 11, 329–347.
- Hoppel, W. A., & Frick, G. M. (1986). Ion-aerosol attachment coefficients and steady-state charge distributions on aerosols in a bipolar ion environment. *Aerosol Science Technology*, 5, 1–21.
- Jones, A. V., & Kissane, M. (2000). State of understanding of fission product transport in the circuit and of aerosol in the containment behaviour of PHEBUS-FP. *Journal of Aerosol Science*, 31, S37–S38.
- Mayya, Y. S. (1994). Charging induced drift and diffusion of aerosol particles in oscillating electric fields. *Journal of Aerosol Science*, 25, 277–288.
- Mayya, Y. S., & Malvankar, S. V. (1993). Charging-induced diffusion of aerosol particles moving under external fields in ionized gases. *Journal of Colloid and Interface Science*, 156, 78–84.
- Romay, F. J., Pui, D. Y. H., & Adachi, M. (1991). Unipolar diffusion charging of aerosol particles at low pressures. *Aerosol Science and Technology*, 15, 60–68.
- Stechkina, I. B., Kirsh, A. A., & Zagrit'ko, A. V. (1982). The charge on aerosol particles in an inhomogeneous bipolar ionic atmosphere. *Russian Journal of Physics Chemistry*, 56, 98–101.