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Diffusion charging of aerosol nanoparticles with an excess of bipolar ions

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Abstract

The equations of diffusion charging of aerosol particles in a bipolar ion environment have no analytical transient solution. Such a solution can, however, be obtained in the special but important case of a relatively high ion concentration and aerosols with a particle size of a few nanometres, for which the probability of multiple charging is vanishingly small. The analytical transient solution obtained has an application to nanoaerosol particle size distribution measurement by electric mobility analysis. It also allows determination of the mean aerosol residence time required to attain the stationary charge distribution as a function of the ion concentration and physical properties (mass, mobility), this result having a practical application to the design of charging devices for aerosol nanoparticles.

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1. Introduction

Aerosol particle sizing by electrostatic methods (e.g., differential mobility analyser (DMA) [1, 2]) requires knowledge of the distribution of the electric charges acquired by the particles. In particular, transformation of the measured mobility distribution into a particle size distribution is customarily carried out assuming that the aerosol has attained its stationary charging state before entering the DMA [3]. Particle charging is usually accomplished by passing the aerosol through a cloud of unipolar ions generated by electric discharge, or through a medium of bipolar ions generated by a radioactive source [4]. Whatever the technique employed, the design of the charging device and the selection of the operating conditions must be such that the product nt (ion concentration \times mean aerosol residence time in the charger) is enough for the particles to reach their stationary charge distribution. Generally, this requirement is satisfied by letting the particles spend a ‘sufficiently’ high residence time within the charger.

However, if the aerosol residence time in the charger is too high, particle losses by diffusion to the walls and by Brownian coagulation will also be large and, since the particle loss rate by both mechanisms depends on the particle size, the particle size distribution will be thereby modified and, as a consequence, the measured particle size distribution will differ from the actual, original size distribution [5].

The above-mentioned problems could be minimized if we had precise knowledge of the time required for attainment of the charging steady state, or better, if for any residence time of the aerosol in the charger we could know the particle charge distribution, for in the latter case one could reduce the aerosol residence time below that required for equilibrium, thereby reducing the extent of coagulation and diffusive losses, but still be able to convert the mobility distribution into the desired particle size distribution.

The temporal variation of the fraction of particles having a certain number of net charges of a given polarity can be known by solving the equations of charging. In these, the concentrations of the different species of particles (charged and neutral) and of the ions appear simultaneously in several equations, and the system of differential equations must be solved numerically. There is, however, a particular and very important case in which the transient equations can be solved analytically, namely, that of aerosols with particle size below about 20 nm in a cloud of bipolar ions with a typical concentration as that found in commercial chargers. For such small sizes, the probability that a particle acquires two or more net charges of either sign is practically zero [6], so that the aerosol consists of only three different species: neutral particles, and singly charged particles with positive or negative polarity. Besides, since the probability of singly charging is quite low for nanoparticles (less than 0.05 for 10 nm particles, and lower for smaller particles [6, 7]), the fraction of ions spent in charging is also quite small; hence, one can assume that the ion concentration remains constant throughout the process.

This particular case in which the transient charging equations admit an analytical solution is of great practical importance, because presently differential mobility analysis is the only reliable technique available to determine the particle size distribution of nanoaerosols [8–11].

2. The charging equations in the general case

The system to be considered consists of an aerosol stream flowing through a container in which monovalent bipolar air ions are continuously generated, by alpha or beta radiation emitted by a radioactive source, at a rate g (number of ion pairs per unit time and unit volume). The total concentration of aerosol particles, N , is assumed to remain constant during the charging process, though actually there are particle losses by diffusion to the container walls and by Brownian coagulation. The assumption of particle number concentration constancy may only be valid for very short residence times of the aerosol in the charging chamber, but after all, the main objective of this work is to find the analytical solution of the transient charging equations that will allow us to design a charger in which the aerosol spends, at most, a time equal to that needed for attainment of the stationary charging state. Denoting by $f_m^j(t)$ the fraction of particles that, at time t , have acquired m net charges of polarity j , the particle number conservation equation is simply

$$f_0 + \sum_{m=1}^{\infty} (f_m^+ + f_m^-) = 1 \quad (1)$$

where f_0 stands for the fraction of neutral particles.

The number concentration n^j of singly charged ions of polarity j evolves with time according to the expression (see, for instance, [6, 12] among many others)

$$\frac{dn^j}{dt} = g - \alpha n^j n^k - N n^j \left[\eta_0^{j0} f_0 + \sum_{m=1}^{\infty} (\eta_m^{jj} f_m^j + \eta_m^{jk} f_m^k) \right] \quad j, k = +, - \quad m \geq 0 \quad (2)$$

where α is the ion recombination rate constant and η_m^{jk} is the attachment rate coefficient of an ion of polarity j with a particle carrying m net charges of polarity k ($k = 0$ for neutral particles). Similarly, the temporal variation of the particle charge distribution is given by

$$\frac{df_m^j}{dt} = n^j (\eta_{m-1}^{jj} f_{m-1}^j - \eta_m^{jj} f_m^j) + n^k (\eta_{m+1}^{kj} f_{m+1}^k - \eta_m^{kj} f_m^j) \quad j, k = +, - \quad m \geq 0. \quad (3)$$

Equations (1)–(3) are supplemented with the charge conservation equation

$$n^+ - n^- + N \sum_{m=1}^{\infty} m (f_m^+ - f_m^-) = \frac{\rho}{e} \quad (4)$$

where ρ is the initial space charge (i.e. before aerosol particles are let into the chamber) and e is the elementary charge.

The general system of balance equations (1)–(4) has no analytical solution. The steady-state solution can be found by assuming a fixed ion ratio, and by application of the principle of detailed balancing. According to this principle [13, 14], the steady-state rate at which particles of charge m capture positive ions is equal to the rate at which particles of charge $m + 1$ capture negative ions, that is,

$$\eta_m^{jk} n^j f_m^{*k} = \eta_{m+1}^{kj} n^k f_{m+1}^{*j}. \quad (5)$$

(The asterisk indicates the steady-state condition.) From (1) and (5), one finds the steady-state fraction of particles carrying m net charges of polarity j :

$$f_m^{*j} = \frac{\left(\frac{n^j}{n^k}\right)^m \prod_{p=1}^m \frac{\eta_{p-1}^{jj}}{\eta_p^{kj}}}{1 + \sum_{q=1}^{\infty} \left[\left(\frac{n^j}{n^k}\right)^q \prod_{p=1}^q \frac{\eta_{p-1}^{jj}}{\eta_p^{kj}} + \left(\frac{n^k}{n^j}\right)^q \prod_{p=1}^q \frac{\eta_{p-1}^{kk}}{\eta_p^{jk}} \right]}. \quad (6)$$

For relatively large particles (continuum regime), there exist analytical expressions for the ion attachment rate coefficients η_m^{jk} as a function of particle size (see, for instance, [14, 15]); therefore, (6) gives the analytical expression for the steady-state distribution of charges on the aerosol particles as a function of the ionic ratio n^+/n^- and the particle size. This theoretical charge distribution is commonly used in differential mobility analysis to infer the original particle size distribution from the measured mobility distribution. In practical aerosol particle size measurements one employs a sufficiently large charging chamber (and/or sufficiently low aerosol flow rate) so that the required steady-state time is amply exceeded. This practice, as we have seen, may lead to erroneous results because, due to particle size distribution modification in the charger by diffusion losses and coagulation (the rates of which depend on particle size), the inferred particle size distribution may differ substantially from that of the original aerosol entering the charger.

3. The special case of nanometre-sized particles

The attachment rate coefficient between an ion of either polarity and a singly charged particle of the same polarity is practically zero for particle diameters below about 20 nm [6]. Therefore,

for this range of particle size, the number concentration of particles with two or more net charges is negligible or even zero.

Furthermore, the ion attachment rate coefficient between an ion and a neutral particle (charging coefficient) is extremely small, much smaller (between one and two orders of magnitude, depending on the particle size) than the corresponding attachment rate coefficient between a charged particle and an ion of opposite polarity (discharging or neutralization coefficient). As a consequence, the number concentration of singly charged particles at the steady state is but a very small fraction of the total aerosol number concentration, and this implies, first, that the fraction of ions spent in charging is negligible in comparison with ion losses by recombination and, second, that the equilibrium ion concentration existing in the charger before the aerosol is let in remains practically constant during the charging process. In this manner, the aerosol charging equations (3) decouple from the ion equations (2), and an analytical solution to the transient charging process is then possible. As already stated in the introduction, this particular case of aerosol charging is of great practical importance, especially since nowadays an increasing interest is being placed on the production and characterization of nanoparticles. Indeed, the most reliable method of aerosol particle size measurement presently available is electrical mobility analysis, which requires previous charging of the particles and precise knowledge of the resulting charge distribution.

For bipolar diffusion charging of nanoparticles, only four attachment rate coefficients are needed, η^{+0} and η^{-0} for charging of neutral particles, and η^{+-} and η^{-+} for neutralization of singly charged particles. (The subscripts denoting the number of charges in the particle will be omitted, since a particle can acquire at most one net charge.) The ion-pair equilibrium number concentration in the charging chamber at time $t = 0$, just before the aerosol stream is let in, is given by the steady-state solution of (2) without the charging–discharging terms:

$$n_{\text{eq}}^+ = n_{\text{eq}}^- = \sqrt{g/\alpha} = n. \quad (7)$$

Due to the different mobilities of positive and negative air ions, the attachment rate coefficients depend on the ion polarity and, as a consequence, for any time $t > 0$ the concentrations of positive and negative ions differ from each other. However, as explained above, the fraction of the equilibrium ion-pair concentration which is spent in charging or neutralization is so small that one can safely assume a constant ion-pair concentration throughout the process. In a sense, it is not that equation (3) decouples from (2), but simply that the latter is not needed to be considered at all to find the transient solution of the aerosol charging equations.

Under these special, but physically sound and of practical importance, circumstances, the aerosol charging equations become

$$\frac{df^j}{dt} = n\eta^{j0}f^0 - n\eta^{kj}f^j \quad j, k = +, -. \quad (8)$$

The particle number conservation equation is simply

$$1 = f^0 + f^+ + f^-. \quad (9)$$

The system of two coupled equations (8) can be decoupled by transforming them into second-order differential equations. Differentiating (8) with respect to time and using (9) yields

$$\frac{d^2f^j}{dt^2} = -n\eta^{j0}\frac{df^k}{dt} - n(\eta^{j0} + \eta^{kj})\frac{df^j}{dt} \quad j, k = +, -. \quad (10)$$

Setting $j = +$ in (8), solving for f^- and substituting the resulting expression into equation (8) with $j = -$ yields an expression for df^-/dt as a function of f^+ and df^+/dt . Inserting this expression into (10) finally results in an equation containing only f^+ and its first and second

time derivatives. A similar procedure can be carried out to obtain the corresponding second-order differential equation for the fraction of negative particles. The resulting equations are

$$\frac{d^2 f^j}{dt^2} + A \frac{df^j}{dt} + B f^j = n^2 \eta^{j0} \eta^{jk} \quad j, k = +, - \quad (11)$$

where

$$A = n(\eta^{+0} + \eta^{-0} + \eta^{+-} + \eta^{-+}) \quad (12)$$

$$B = n^2(\eta^{+0}\eta^{+-} + \eta^{-0}\eta^{-+} + \eta^{+-}\eta^{-+}). \quad (13)$$

(To avoid confusion, the superscripts of the attachment rate coefficients have been written down explicitly in the last two equations.)

Equation (11) must be solved with initial conditions

$$f^j = 0 \quad (j = +, -) \quad \text{at} \quad t = 0 \quad (14)$$

expressing that the aerosol particles are initially uncharged, and

$$\frac{df^j}{dt} = n\eta^{j0} \quad (j = +, -) \quad \text{at} \quad t = 0. \quad (15)$$

The last condition expresses the initial charging rates of neutral particles, the only existing species at time $t = 0$, as can be directly obtained from (8) and (9).

The solution of (11) with these initial conditions is

$$f^j = f^{*j} + \frac{(n\eta^{j0} + m_1 f^{*j}) \exp(m_2 t) - (n\eta^{j0} + m_2 f^{*j}) \exp(m_1 t)}{m_2 - m_1} \quad j = +, - \quad (16)$$

where m_1 and m_2 are the roots of the equation $m^2 + Am + B = 0$:

$$m_1 = -\frac{1}{2}A \left(1 + \sqrt{1 - 4B/A^2}\right) \quad m_2 = -\frac{1}{2}A \left(1 - \sqrt{1 - 4B/A^2}\right) \quad (17)$$

and

$$f^{*j} = \frac{n^2}{B} \eta^{j0} \eta^{jk} \quad j, k = +, - \quad (18)$$

is the stationary charge distribution. Indeed, since the roots m_1 and m_2 are both negative (at least, in the particle size range of interest, that is, below about 20 nm), the solution (16) leads to the correct result $f^j \rightarrow f^{*j}$ as $t \rightarrow \infty$. Using (13), the stationary distribution can also be written as

$$f^{*j} = \frac{R^j}{1 + R^j + R^k} \quad j, k = +, - \quad (19)$$

where

$$R^j = \frac{\eta^{j0}}{\eta^{kj}} \quad j, k = +, - \quad (20)$$

is the particle charging-to-discharging rate ratio. The same result (19) can be directly obtained by setting $df^j/dt = 0$ in (8).

Equation (16) gives the transient particle charge distribution and, as such, can be used in differential mobility analysis to transform the measured mobility distribution into the corresponding particle size distribution upon acquiring knowledge of the mean aerosol residence time in the charger. If wished, it can also be used to determine the time required to achieve the stationary distribution—strictly speaking, the stationary distribution is attained at $t \rightarrow \infty$, but an estimation, useful for practical purposes, can be done as explained later in section 6.

4. The ion attachment rate coefficients

In the continuum regime (aerosol particles much larger than the ionic mean free path), the theory of the diffusion of ions to particles is based on the solution of the diffusion equation for ions in the electric field of a charged particle [13, 16, 17], and analytical expressions for the attachment rate coefficients η^{jk} can be obtained. However, as we have seen, the solution of the transient charging equations must be found by numerical methods.

In contrast, for nanometre-sized aerosol particles (kinetic or free molecular regime) we have been able to obtain an analytical solution of the transient charging equations, but analytical expressions for the ion attachment rate coefficients are not available. The calculation of the attachment rate coefficients is based on the Fuchs theory [18], two recent good reviews of which can be found in [6, 19]. According to this theory, the space surrounding a particle is divided into two regions by a limiting sphere, concentric with the particle and having a radius in the order of one ionic mean free path larger than the particle radius. In the region outside the limiting sphere, ions move according to the continuum diffusion equation, whereas in the inner region they move as in a vacuum, without colliding with air molecules and following free-molecular transport trajectories. By matching the kinetic and continuum ion fluxes at the limiting sphere, the following equation is finally obtained:

$$\eta = \frac{\pi \delta^2 \bar{c} \beta \exp[-\phi(\delta)/kT]}{1 + \exp[-\phi(\delta)/kT][\delta^2 \bar{c} \beta / 4D] \int_0^1 \exp[\phi(D_p/2x)/kT] dx} \quad (21)$$

In this equation, δ is the radius of the limiting sphere, which depends on the particle size and on the ionic mean free path. $\bar{c} = \sqrt{8kT/\pi m}$ and $D = kTZ/e$ are, respectively, the mean thermal velocity and diffusion coefficient of an ion with mass m and electric mobility Z . β is the collision probability, which depends on the electrostatic potential energy ϕ of an ion in the field of the particle:

$$\phi(r) = \frac{e^2}{4\pi\epsilon_0} \left[\frac{p}{r} - \frac{\epsilon - 1}{\epsilon + 2} \frac{D_p^3}{16r^2 (r^2 - D_p^2/4)} \right] \quad (22)$$

where r is the distance measured from the centre of the particle of diameter D_p and dielectric constant ϵ , and ϵ_0 is the dielectric constant of a vacuum. In the above equation, $p = 0$ for a neutral particle, $p = 1$ if the ion and the singly charged particle have the same polarity and $p = -1$ if the charges on the ion and the particle are of opposite signs.

Upon acquiring knowledge of the ion properties (mass, mobility) one can determine numerically with equation (21) the values of the four different attachment coefficients as a function of particle diameter. For practical purposes, regression analysis can then be carried out to obtain fitted $\eta(D_p)$ functions.

Actually, in the absence of any reliable relationship between ion mass and mobility, it has been a common practice among aerosol researchers to leave these two ionic properties as sorts of fitting parameters whose values can, to a certain extent, be freely chosen so as to match the predictions of Fuchs' theory with the experimental results. In past experimental works on particle charging [6, 7, 20–26], ions were in all cases generated from the ionization of air molecules by alpha or beta radiation. The composition of the ions, and hence their physical properties, mainly depends on their age and the nature of the trace compounds present in the air [19]; hence, it is possible that the 'air ions' differed from work to work. However, it seems unlikely that ion masses and mobilities could be so different from each other in these works (see [19] for the whole collection of m and Z values assumed by a number of authors).

In a recent experimental work on nanoaerosol charging with bipolar air ions generated by an ^{241}Am source [7], the measured ion mobilities were $Z^+ = 1.15$ and $Z^- = 1.65 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.

Charging experiments were carried out at several aerosol mean residence times with the aim of finding the time required to attain the stationary charge distribution. The measured steady-state fractions of positive and negative particles were compared with the theoretical ones as given by equation (19), using the charging-to-discharging rate ratios (20) calculated with equation (21). We found that Fuchs' theory yielded theoretical predictions in reasonable agreement with experiments for assumed ionic masses of $m^+ = 150$ and $m^- = 80$ amu. These values were coherent with those used by other authors [6, 20–26].

Using the above values for the ionic masses and mobilities, we have recalculated the ion attachment rate coefficients with equation (21) for the particle diameter range between 2 and 20 nm. The numerically calculated attachment rate coefficients are well reproduced by the following expressions (correlation coefficients above 0.9965 in all cases):

$$\eta^{+0} = 2.19 \times 10^{-9} D_p^{1.51} \quad (23a)$$

$$\eta^{-0} = 3.02 \times 10^{-9} D_p^{1.51} \quad (23b)$$

$$\eta^{+-} = (5.68 + 3.38 D_p - 0.522 D_p^2 + 0.042 D_p^3 - 0.0017 D_p^4 + 0.000027 D_p^5) \times 10^{-7} \quad (23c)$$

$$\eta^{-+} = (2.15 + 4.20 D_p - 0.526 D_p^2 + 0.038 D_p^3 - 0.0014 D_p^4 + 0.000020 D_p^5) \times 10^{-7} \quad (23d)$$

where η is expressed in $\text{cm}^3 \text{s}^{-1}$ and D_p in nm.

Alternative theories to calculate the ion attachment coefficients have been proposed, all of them based on the concept of limiting sphere, but removing some of the simplifications assumed by Fuchs. Thus, Marlow and Brock [27] took into account the Maxwellian velocity distribution of approaching ions far from the particle; Hoppel and Frick [6] included three-body trapping as a further mechanism of ion capture; and, more recently, Filippov [28] considered collisions between ions and neutral molecules inside the limiting sphere. These alternative charging theories give practically the same predictions as those of Fuchs, in spite of the latter being much simpler, in the particle diameter range above about 2 nm. Large discrepancies between Fuchs' theory and those of Marlow–Brock and Filippov appear for particle diameters below 2 nm, but, unfortunately, in this extremely small particle size range there are no experimental data available which permit us to decide which of the proposed theories is correct. Therefore, since Fuchs' theory agrees fairly well with the available experimental data, and is easier to use than the others, we have preferred to adopt it in the present work.

5. Comparison between the analytical transient solution and former experimental results

As commented above, in a former experimental work on nanoparticle diffusion charging with bipolar ions [7], we measured the fractions of positively and negatively charged particles as a function of the mean residence time spent by the aerosol in the charger. The experimental method will not be described here, as it has already been explained in detail in the original publication. However, it is important to point out that the experimentally measured steady-state fraction of charged particles as a function of particle size was in quite reasonable agreement with the results of other authors. Furthermore, we performed two sets of experiments, one for charging of initially uncharged particles, the other for neutralization of initially charged particles. The resulting steady-state fractions of charged particles were equal in both cases, as they should be. This fact, along with the above-mentioned agreement between our steady-state results and those of other groups, should serve to demonstrate the reliability of our former experimentation.

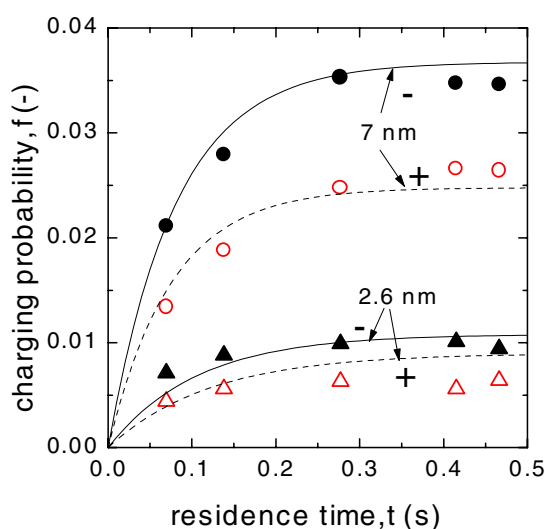


Figure 1. Evolution of the fraction of charged particles in a diffusion-type charger for initially discharged aerosols. Points: experimental results obtained in the course of a former work [7]. Curves: calculations done with equation (16) using the ion attachment rate coefficients calculated with the fitting expressions (23).

In addition, as commented before, we also performed measurements for times shorter than that required to attain the steady-state condition; as far as we know, such measurements have not been done before. It means that ours are likely to be the only experimental data on the transient charging of nanoparticles available to date. The comparison between these data and the theoretical curves given by equation (16) (using the fitted ion attachment rate coefficients (23)) is presented in figure 1 for two different particle sizes. The agreement is excellent for both particle sizes, and also for both ion polarities. It must be recalled, however, that the values of ionic mass used to calculate the attachment rate coefficients (80 and 150 amu for negative and positive ions, respectively), though coherent with the values assumed by other authors, may not be correct. The most one can say is that Fuchs' theory seems to be correct provided that 'adequate' values are selected for the ionic masses.

6. Timescale for charging

The timescale to reach the steady state can be found directly from the transient solution (16). Since two exponential terms appear in this equation, actually there are two possible values (quite close to each other) for the timescale τ , namely, $-1/m_1$ and $-1/m_2$. However, in the particle size range of interest (2–20 nm), $|m_1| > |m_2|$, so that it is the root m_2 that controls the approach to the stationary state. The timescale

$$\tau = -1/m_2 \quad (24)$$

with m_2 given by (17), is plotted in figure 2 as a function of particle size. For comparison, the timescale proposed by Mayya and Saira [29] is also included, which is claimed to be valid for the entire particle size range (free-molecular, transition and continuum regimes). These authors also employed Fuchs' theory for the calculation of the ion attachment coefficients. The results compare favourably for particle diameters above about 5 nm, but the curves clearly diverge for smaller particles.

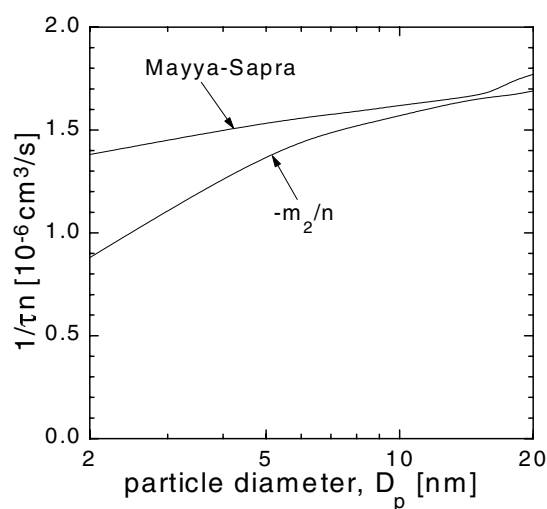


Figure 2. Timescale to attain the stationary charging state as a function of particle size. The upper curve is that given in the work of Mayya and Sapra [29]; the lower one has been obtained using equation (24).

In whatever case, the time required to attain the stationary charge distribution depends on the particle size. This implies that in the practical case of charging of polydisperse aerosols, the smaller particles require longer residence times to attain their stationary charging state, so that by the time they have reached equilibrium, the larger particles (and, of course, the smaller ones too) have had enough time to undergo concentration changes by coagulation and diffusion losses. This problem has already been commented on above. The practical importance of the present work is that one is no longer in any need to over-design the charging device, because for any given mean aerosol residence time in the charger, equation (16) allows determination of the charge distribution independently of whether the stationary state has been achieved or not.

7. Conclusions

In summary, an analytical expression for the temporal variation of the particle charge distribution has been found for the special case of aerosol particles with diameters below about 20 nm. This result has an important practical application to aerosol research, particularly in the measurement of particle size by differential mobility analysis, because it permits the particle mobility distribution to be transformed into the corresponding particle size distribution for whatever value of the mean residence time spent by the aerosol in the charger.

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