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# Dispersion of aerosol particles undergoing Brownian motion

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**Abstract**

The variance of the position distribution for a Brownian particle is derived in the general case where the particle is suspended in a flowing medium and, at the same time, is acted upon by an external field of force. It is shown that, for uniform force and flow fields, the variance is equal to that for a free particle. When the force field is not uniform but depends on spatial location, the variance can be larger or smaller than that for a free particle depending on whether the average motion of the particles takes place toward, respectively, increasing or decreasing absolute values of the field strength. A few examples concerning aerosol particles are discussed, with especial attention paid to the mobility classification of charged aerosols by a non-uniform electric field. As a practical application of these ideas, a new design of particle-size electrostatic classifier differential mobility analyser (DMA) is proposed in which the aerosol particles migrate between the electrodes in a direction opposite to that for a conventional DMA, thereby improving the resolution power of the instrument.

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## 1. Introduction

Brownian motion plays a key role in many processes involving submicron aerosol particles. In some instances, Brownian motion is favourable because it enhances the rate at which some processes occur, e.g. particle-size growth by coagulation and electrical charging of aerosols. In the case of extremely small particles, with diameters of a few nanometres, Brownian diffusion practically becomes the only mechanism by which they can be intercepted by filtering fibres. In other cases, however, Brownian motion is an unwanted process because it distorts the

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otherwise well-defined and deterministic particle trajectory. This occurs, for example, in the measurement of aerosol particle size using electrostatic methods [1–7], a topic that will be dealt with in great detail in the last part of this paper.

In the absence of external forces, the time derivative of the variance of the distribution of positions of a Brownian particle is twice the particle diffusion coefficient $D$ for each degree of freedom [8]. When the aerosol is acted upon by a non-uniform external force field, the variance differs from that found for a free particle, and can be smaller or larger than the latter. As will be seen, when the average particle motion takes place in the direction of increasing absolute values of the field, the particle trajectories spread about the mean trajectory to a greater extent than in the case of free particles, that is, the time derivative of the variance is larger than $2D$ per degree of freedom. Likewise, if particles on average move toward decreasing values of the force field, the rate at which the variance increases is smaller than for a free particle. This means that if, by a suitable means, we are able to select the direction of the average motion of the particles, we can control their spreading rate to some extent.

In this paper, after deriving the general equation relating the variance increase rate to the external force field, a few examples of interest to aerosol scientists are discussed. The main part of the paper is devoted to the application of the above idea to a specific and very important process, namely the size classification of charged aerosol particles by an electric field.

## 2. Variance of the distribution of particle positions

In the general case where the Brownian particle is suspended in a flowing gas medium, the Langevin equation of motion can be expressed as

$$m_p \frac{\mathrm{d}\boldsymbol{v}}{\mathrm{d}t} = \frac{1}{\mu}(\boldsymbol{u} - \boldsymbol{v}) + \boldsymbol{F} + m_p \boldsymbol{A}_B \tag{1}$$

where $m_p$ is the particle mass, $\boldsymbol{v}$ its velocity, $\mu$ its mechanical mobility, $\boldsymbol{u}$ the gas flow velocity, $\boldsymbol{F}$ the total external force acting on the particle, and $\boldsymbol{A}_B$ the random acceleration imparted to the particle by the fluid molecules. Equation (1) is only approximate because it does not include additional terms such as the force due to the pressure gradient in the fluid surrounding the particle, the force required to accelerate the apparent mass of the particle relative to the fluid, and the force arising from the deviation of the fluid velocity from the steady state (Basset history integral). However, in most practical cases of aerosol particles in air these three additional terms can be neglected [9].

A parameter of fundamental importance to aerosol particles is the so-called relaxation time, defined as [10]

$$\tau = m_p \mu = \frac{m_p C}{3\pi \eta D_p} \tag{2}$$

where $C$ is the slip correction factor, $\eta$ the fluid viscosity, and $D_p$ the particle diameter. Introducing the particle relaxation time, equation (1) becomes

$$\frac{\mathrm{d}\boldsymbol{v}}{\mathrm{d}t} = \frac{1}{\tau}(\boldsymbol{u} - \boldsymbol{v}) + \frac{1}{m_p}\boldsymbol{F} + \boldsymbol{A}_B. \tag{3}$$

Considering an ensemble of identical particles which are released from a given point, the ensemble averaging of (3) leads to

$$\tau \frac{\mathrm{d}\langle \boldsymbol{v} \rangle}{\mathrm{d}t} = \langle \boldsymbol{u} \rangle - \langle \boldsymbol{v} \rangle + \mu \langle \boldsymbol{F} \rangle \tag{4}$$

because, due to its random nature, $\langle \boldsymbol{A}_B \rangle = 0$. Note that the fluid velocity and the external force both appear as ensemble averages in the last equation, because in the general case they

might depend on the spatial coordinates, and the particle position (i.e. the specific point at which the vectors $\boldsymbol{u}$ and $\boldsymbol{F}$ must be evaluated) fluctuates about a certain average trajectory. On average (that is, leaving Brownian effects aside) the aerosol particle accelerates until the external forces balance the drag force, and hereafter the particle moves with a steady velocity given by

$$\langle \boldsymbol{v} \rangle = \langle \boldsymbol{u} \rangle + \mu \langle \boldsymbol{F} \rangle \qquad (\text{for } t \gg \tau). \tag{5}$$

Thus, $\tau$ represents a characteristic time for the particle to approach steady motion. The relaxation time is very small for typical aerosol particles. For unit-density spherical particles, $\tau$ is of the order of $10^{-8}$ s for $D_p = 0.05$ $\mu$m, and increases up to about $10^{-4}$ s for 10 $\mu$m particles. This means that in most cases of practical interest the aerosol particles, on the one hand, accommodate to the fluid motion almost instantaneously (this is actually why no additional force terms are needed in equations (1) or (3)) and, on the other hand, they soon acquire their steady terminal velocity given by (5).

In terms of the particle position vector $\boldsymbol{R}$, Langevin's equation can be rewritten as

$$\frac{\mathrm{d}^2 \boldsymbol{R}}{\mathrm{d}t^2} = \frac{1}{\tau}(\boldsymbol{u} - \boldsymbol{v}) + \frac{1}{m_p}\boldsymbol{F} + \boldsymbol{A}_B. \tag{6}$$

Starting from (6) one can easily derive the equation for the temporal variation of the variance by using the method that can be found in many textbooks (e.g. [11]) and which is briefly described here. First, forming the scalar product of (6) with $\boldsymbol{R}$, rearranging the resulting expression and averaging it over a large number of identical particles yields

$$\tau \frac{\mathrm{d}^2 \langle \boldsymbol{R}^2 \rangle}{\mathrm{d}t^2} + \frac{\mathrm{d}\langle \boldsymbol{R}^2 \rangle}{\mathrm{d}t} = 2\tau \langle \boldsymbol{v}^2 \rangle + 2\langle \boldsymbol{R} \cdot \boldsymbol{u} \rangle + 2\mu \langle \boldsymbol{R} \cdot \boldsymbol{F} \rangle. \tag{7}$$

To obtain (7) it has been necessary to use the fact that the random acceleration $\boldsymbol{A}_B$ and the position vector $\boldsymbol{R}$ are uncorrelated, so $\langle \boldsymbol{R} \cdot \boldsymbol{A}_B \rangle = 0$.

Second, the scalar product of (4) with $\boldsymbol{R}$ and subsequent averaging leads to

$$\tau \frac{\mathrm{d}^2 \langle \boldsymbol{R} \rangle^2}{\mathrm{d}t^2} + \frac{\mathrm{d}\langle \boldsymbol{R} \rangle^2}{\mathrm{d}t} = 2\tau \langle \boldsymbol{v} \rangle^2 + 2\langle \boldsymbol{R} \rangle \cdot \langle \boldsymbol{u} \rangle + 2\mu \langle \boldsymbol{R} \rangle \cdot \langle \boldsymbol{F} \rangle. \tag{8}$$

Subtracting (8) from (7) one obtains the second-order differential equation for the temporal variation of the variance of the particle position distribution ($\sigma^2 \equiv \langle \boldsymbol{R}^2 \rangle - \langle \boldsymbol{R} \rangle^2$):

$$\tau \frac{\mathrm{d}^2 \sigma^2}{\mathrm{d}t^2} + \frac{\mathrm{d}\sigma^2}{\mathrm{d}t} = 2\tau(\langle \boldsymbol{v}^2 \rangle - \langle \boldsymbol{v} \rangle^2) + 2(\langle \boldsymbol{R} \cdot \boldsymbol{u} \rangle - \langle \boldsymbol{R} \rangle \cdot \langle \boldsymbol{u} \rangle) + 2\mu(\langle \boldsymbol{R} \cdot \boldsymbol{F} \rangle - \langle \boldsymbol{R} \rangle \cdot \langle \boldsymbol{F} \rangle). \tag{9}$$

The first term in the right-hand side of (9) can be determined as follows. The particle velocity can be considered to consist of two contributions, a deterministic velocity $\boldsymbol{v}_F$ due to the external force $\boldsymbol{F}$, and a random velocity $\boldsymbol{v}_B$ arising from the bombardment of the surrounding fluid molecules:

$$\boldsymbol{v} = \boldsymbol{v}_F + \boldsymbol{v}_B. \tag{10}$$

Obviously, the ensemble average of $\boldsymbol{v}_B$ vanishes and, furthermore, since both velocity contributions are uncorrelated, $\langle \boldsymbol{v}_F \cdot \boldsymbol{v}_B \rangle = 0$. Hence,

$$\langle \boldsymbol{v}^2 \rangle - \langle \boldsymbol{v} \rangle^2 = \langle \boldsymbol{v}_B^2 \rangle = \frac{3kT}{m_p} \tag{11}$$

where the last equality comes from the assumption that the particles are in thermal equilibrium with the medium. Inserting (11) into (9) yields

$$\tau \frac{\mathrm{d}^2 \sigma^2}{\mathrm{d}t^2} + \frac{\mathrm{d}\sigma^2}{\mathrm{d}t} = 6kT\mu + 2(\langle \boldsymbol{R} \cdot \boldsymbol{u} \rangle - \langle \boldsymbol{R} \rangle \cdot \langle \boldsymbol{u} \rangle) + 2\mu(\langle \boldsymbol{R} \cdot \boldsymbol{F} \rangle - \langle \boldsymbol{R} \rangle \cdot \langle \boldsymbol{F} \rangle). \tag{12}$$

However, as pointed out above, the relaxation time $\tau$ for submicron aerosol particles (those for which Brownian motion is important in practice) is very small. Therefore, the first term on the left-hand side of (12) is negligible and the last equation can be simplified to

$$\frac{\mathrm{d}\sigma^2}{\mathrm{d}t} = 6D + 2\left(\langle \boldsymbol{R}\cdot\boldsymbol{u}\rangle - \langle \boldsymbol{R}\rangle\cdot\langle \boldsymbol{u}\rangle\right) + 2\mu\left(\langle \boldsymbol{R}\cdot\boldsymbol{F}\rangle - \langle \boldsymbol{R}\rangle\cdot\langle \boldsymbol{F}\rangle\right) \tag{13}$$

where

$$D = kT\mu \tag{14}$$

is the particle diffusion coefficient. Equation (13) is the general equation expressing the variance of the positions of particles suspended in a moving gas medium under an external field of force. In the rest of this paper, we will apply equation (13) to a few specific cases of relevance to aerosol processes.

It is important to point out that in the case of aerosol particles in an electric field and simultaneously undergoing electric charging or discharging by ions present in the medium, the random nature of the particle-charging process may lead to aerosol diffusion along field lines [12]. The general problem of a fluctuating force arising from charge changes has been dealt with recently [13]. However, we will not take into consideration this additional force, because the only example of charged particles in an electric field that will be discussed in this paper is that of the differential mobility analyser (DMA), an instrument in which aerosol particles are unipolarly charged and in which the concentration of free ions, if any, is negligible in comparison with the aerosol concentration. In the DMA, therefore, the particles do not suffer any modification of their charging state and thus there is no additional force arising from charge variability.

## 3. Uniform flow and force fields

If the fluid flow field $\boldsymbol{u}$ and the external force field $\boldsymbol{F}$ are both uniform—that is, independent of spatial location—the two last terms on the right-hand side of (13) vanish and

$$\sigma^2 = \sigma_B^2 = 6Dt \qquad (\boldsymbol{u} = \text{constant}, \boldsymbol{F} = \text{constant}). \tag{15}$$

Thus, in this case particle dispersion takes place at the same rate as for free particles suspended in a motionless fluid. Examples of this situation are the gravitational sedimentation of aerosols, and the electrostatic precipitation of charged aerosol particles in a parallel-plate condenser, provided that the velocity component of the fluid in the direction of the external field (gravitational or electric) is a constant independent of the particle position. In these two cases, since the particle motion takes place in only one direction, the variance is actually one third of that given by (15),that is, $2Dt$.

We see, therefore, that only when either the flow or the force field is not uniform might the variance of particle positions be different from that of a 'pure Brownian motion', $\sigma_B^2$.

## 4. Free particle in a flowing gas medium

In the absence of external forces, the situation of most practical interest where equation (13) can be applied is the transport of aerosol particles along a tube. The only non-zero component of the fluid velocity is in the axial direction $z$. This component, $u_z$, is at most a function of the radial coordinate $r$ but not of $z$. Hence, $\langle zu_z\rangle = \langle z\rangle\langle u_z\rangle$ and the variance (axial dispersion) is again equal to $2Dt$, that is, identical to that for a pure Brownian process.

## 5. Sedimentation of aerosol particles in a centrifugal force field

The situations where aerosol particles are driven by a non-uniform external force field are the most interesting because, as will be seen, the dispersion rate can be larger or smaller than that for a free particle depending on the direction of particle motion. For instance, for aerosol particles settling under the action of a centrifugal force field [10] the force is given by

$$F_r = m_p \omega^2 r \tag{16}$$

where $\omega$ is the angular velocity, and $r$ the radial coordinate. The solution of (13) in this case is

$$\sigma_r^2 = \sigma_{rB}^2 \frac{\exp\left(2\tau\omega^2 t\right) - 1}{2\tau\omega^2 t} \tag{17}$$

where $\sigma_{rB}^2 = 2Dt$ is the variance of the distribution of radial positions for a free Brownian particle. Therefore, we see that for any time $t > 0$ the variance of the distribution of particle positions along the radial direction is always larger than for a free particle and, furthermore, increases exponentially with time. As will be seen more clearly later, the actual reason for this increase in the variance is that aerosol particles move toward increasing values of the external force field. However, we must recall again that the relaxation time is very small, so this effect can be noticed only for quite large angular velocities of the aerosol centrifuge, much larger than those attainable in practice. Indeed, for the usual angular velocities of a few thousand r.p.m., the exponential term in (17) can be approximated as $1 + 2\tau\omega^2 t$ and, hence, $\sigma_r^2 \approx \sigma_{rB}^2$.

## 6. Size classification of aerosol particles by a non-uniform electric field

This is probably the most interesting example of an aerosol process to which equation (13) can be applied. Size classification of charged aerosol particles by an electric field is of fundamental importance to many fields of aerosol research. On the one hand, it permits the experimental determination of the particle-size distribution of aerosols down to the nanometre size range. On the other hand, it enables the separation and continuous extraction of a stream of monodisperse aerosol particles of the desired size from a polydisperse population; the thus-generated monodisperse aerosol can then be used to study aerosol processes where particle size plays a key role (e.g., particle-size growth by coagulation or condensation, aerosol deposition and filtration, charging). Indeed, the apparatus with which size classification is performed, the so-called DMA, is one of the most practical and useful instruments ever developed in the field of aerosol science.

Several DMA designs have been developed and made commercially available. Of these, the most widely used is the concentric cylindrical DMA [14, 15], a sketch of which is shown in figure 1. It consists of two concentric cylindrical electrodes, one of which is grounded (normally the outer electrode), while the other one is connected to a dc power source. It has two flow inlets, one for a stream of filtered dry air (sheath), the other one for the test aerosol stream. Both streams flow laminarly within the annular gap. As soon as the particles enter the DMA near the outer electrode, the charged particles of a given polarity precipitate onto it; the particles of opposite polarity migrate toward the inner electrode while being transported axially by the flowing medium. The migration velocity of the particles depends on their electric mobility and, consequently, on their size, in such a manner that the smallest particles are the first to reach the inner electrode. The inner electrode contains a series of small orifices (*classification port*) near its bottom end, through which particles of a given size (mobility) are withdrawn from the instrument. Particles smaller than those classified precipitate onto the inner electrode upstream of the classification port, while the larger particles leave the DMA

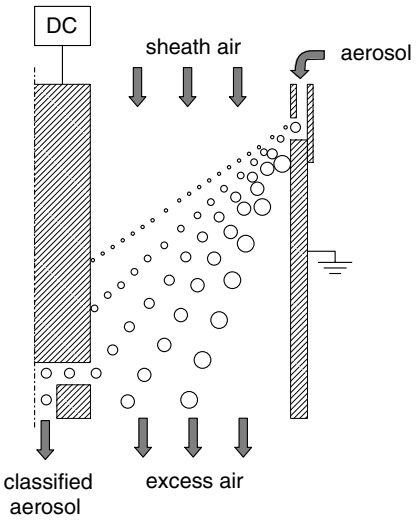


**Figure 1.** Sketch of the DMA.

without being precipitated or classified (*excess air*). By tuning the strength of the applied electric field one can classify particles of the required mobility (or size).

The DMA is usually used in combination with (i) a device where aerosol particles are electrically charged before entering the classification equipment [16,17], and (ii) an instrument to measure the number concentration of the classified particles. The latter can be either an electrometer or, more generally, a condensation nucleus counter [18,19].

The schematical drawing of figure 1 corresponds to what might be termed *conventional* DMA. In this apparatus (the only concentric cylindrical DMA currently available), the aerosol particles to be classified migrate from the outer electrode toward the inner one. As will be seen below, an alternative design in which the particles travel in the opposite direction—that is, from the inner electrode toward the outer one—should result in a drastically different performance of the DMA. This alternative design will be referred to as *reverse* DMA. The difference between these two apparently similar cases is that the diffusional spreading rate of the aerosol particles is much smaller for the reverse DMA because particles migrate in the direction of decreasing absolute values of the electric field. In turn, a smaller spreading rate of particles means a higher resolution power of the size-classification instrument.

### *6.1. Particles migrating in the direction of increasing field strength*

As pointed out above, particles in the conventional DMA travel in the direction of increasing absolute values of the field. The only non-vanishing component of the electric field is that along the radial direction, and is given by

$$E_r = -\frac{\phi}{r \ln(R_2/R_1)} \tag{18}$$

where $\phi$ is the potential difference between the electrodes, and $R_1$ and $R_2$ are the radii of the inner and outer electrodes, respectively. The corresponding radial component of the force is

$$F_r = peE_r \tag{19}$$

where $p$ is the number of charges in the particle, and $e$ is the elementary charge.

The gas flow takes place in the axial direction with a velocity $u_z(r)$. Since, as before, $\langle zu_z\rangle = \langle z\rangle\langle u_z\rangle$, and $F_z = 0$, from equation (13) one concludes that the axial contribution to the variance of the particle position distribution is exactly that of a pure Brownian process, $2Dt$. (The axial dispersion in the reverse DMA is also equal to $2Dt$; the difference between the two designs is in the spreading rate of the aerosol particles in the radial direction alone.)

Since the air flow does not affect the radial dispersion, equation (13) becomes

$$\frac{\mathrm{d}\sigma_r^2}{\mathrm{d}t} = 2D - \frac{2\mu_e\phi}{\ln(R_2/R_1)}(1 - \langle r\rangle\langle r^{-1}\rangle) = \frac{\mathrm{d}\sigma_{rB}^2}{\mathrm{d}t} - \frac{2\mu_e\phi}{\ln(R_2/R_1)}(1 - \langle r\rangle\langle r^{-1}\rangle) \tag{20}$$

where $\mu_e = pe\mu$ is the particle electric mobility, and $\sigma_{rB}^2 = 2Dt$ the variance of the distribution of particle radial positions for a free particle. The last equality in (20) clearly shows that the variance is made up of two contributions, one due to a pure Brownian motion (free particle), and another due to the existence of a non-uniform external force field.

A first important conclusion can be drawn from (20) without the need for solving the equation. It is known that given a collection of $r_i$ values, the arithmetic and harmonic means satisfy the inequality [20]

$$\langle r\rangle\langle r^{-1}\rangle \geqslant 1. \tag{21}$$

The equality only holds in the case where all the $r_i$ values are equal. Therefore, if particles migrate in the direction of increasing absolute values of the field strength, the variance of the distribution of particle positions is always larger than that for the free-particle case:

$$\frac{\mathrm{d}\sigma_r^2}{\mathrm{d}t} > \frac{\mathrm{d}\sigma_{rB}^2}{\mathrm{d}t} \qquad \text{(migration toward increasing field strength)} \tag{22a}$$

or

$$\sigma_r^2 > \sigma_{rB}^2 \qquad \text{for } t > 0 \text{ (migration toward increasing field strength).} \tag{22b}$$

This has important implications concerning the resolution power of the DMA. In the case where the Brownian motion of the particles is negligible (particle diameter larger than about 50 nm), the classified aerosol stream contains particles having electric mobilities (or size) distributed within a relatively narrow range of values. As the particle size decreases, the particle Brownian dispersion causes an increasing deterioration of the resolution power of the instrument, because in the classified stream there also appear particles with mobilities outside the nominal mobility range corresponding to the specific applied voltage $\phi$. The smaller the particle size, the larger the particle diffusion coefficient and, consequently, the broader the mobility interval of the classified particles. The monodispersity of the classified aerosol deteriorates as the particle size decreases. And this deterioration is larger than that expected for particles undergoing a pure Brownian motion because of the non-uniformity of the field or, better, because the particles in the conventional instrument are made to migrate from the outer electrode toward the inner one, that is, in the direction of increasing field strength.

We have not been able to solve (20) because of the presence of $\langle r^{-1}\rangle$. An approximate solution can be found as follows. First, $1/r$ is linearized about the starting radial coordinate of the particle, $r_0$ (in the present case, $r_0$ corresponds to the radius of the outer electrode, $r_0 = R_2$). The radial component of the force then becomes

$$F_r \approx -\frac{pe\phi}{r_0\ln(R_2/R_1)}\left(2 - \frac{r}{r_0}\right). \tag{23}$$

With this approximation, equation (13) is then simplified to

$$\frac{\mathrm{d}\sigma_r^2}{\mathrm{d}t} \approx 2D + \frac{2\mu_e\phi}{r_0^2\ln(R_2/R_1)}\sigma_r^2 \tag{24}$$

the solution of which is

$$\sigma_r^2 = \sigma_{rB}^2 \frac{\exp(\gamma t) - 1}{\gamma t} \tag{25}$$

where

$$\gamma = \frac{2\mu_e \phi}{r_0^2 \ln(R_2/R_1)}. \tag{26}$$

The variance of the distribution of particle radial positions is thus larger than that for a free particle, thereby diminishing the resolution power of the instrument. Furthermore, the difference between $\sigma_r^2$ and $\sigma_{rB}^2$ increases steadily with time. Therefore, a way to minimize the particle beam spreading within the DMA is simply to shorten the aerosol mean residence time, as has been experimentally demonstrated by de Juan and Fernández de la Mora [21]. However, the variance for a free particle, $\sigma_{rB}^2$, constitutes an insuperable limit for the resolution of the conventional DMA because the particle spreading can never become smaller than $2Dt$. A drastic improvement can be achieved if the particles to be classified are made to migrate in the opposite direction, that is, from the inner electrode toward the outer one. This case is discussed in the next subsection.

### *6.2. Particles migrating in the direction of decreasing field strength*

This case is identical to the one considered above except for just one point: the aerosol particles to be classified travel in the opposite direction. The appropriate instrument where this can be accomplished (not available yet, and hardly thought of so far) must be designed such that the aerosol stream enters close to the inner electrode, while the sheath air must occupy the external part of the annular gap. Likewise, the classification port must now be located in the outer electrode. Since the particle average motion is reversed, the last term in (20) changes its sign:

$$\frac{\mathrm{d}\sigma_r'^2}{\mathrm{d}t} = \frac{\mathrm{d}\sigma_{rB}^2}{\mathrm{d}t} + \frac{2\mu_e \phi}{\ln(R_2/R_1)}(1 - \langle r \rangle \langle r^{-1} \rangle). \tag{27}$$

A prime has been attached to $\sigma_r^2$ in order to distinguish it from the former case. Because of the inequality (21), we now conclude that the variance increases at a slower rate than that for a free particle:

$$\frac{\mathrm{d}\sigma_r'^2}{\mathrm{d}t} < \frac{\mathrm{d}\sigma_{rB}^2}{\mathrm{d}t} \qquad \text{(migration toward decreasing field strength)} \tag{28a}$$

or

$$\sigma_r'^2 < \sigma_{rB}^2 \qquad \text{for } t > 0 \text{ (migration toward decreasing field strength).} \tag{28b}$$

As before, an approximate solution for the variance of the distribution of particle radial positions can be obtained by linearizing $1/r$ about $r_0$. The result is

$$\sigma_r'^2 \approx \sigma_{rB}^2 \frac{1 - \exp(-\gamma t)}{\gamma t}. \tag{29}$$

Therefore, the variance is always kept below $\sigma_{rB}^2$. We have thus found the interesting result that the formerly accepted limit for the resolution power of the size-classification instrument can be exceeded by simply reversing the direction of motion of the aerosol particles. However, since the variance still increases steadily with time, improved resolution powers for the reverse DMA are also achieved at as short residence times as are practicable.

At this point, it is important to have an idea of the order of magnitude of the resolution improvement attainable in practical situations. Also using $r_0 = R_2$ for the reverse DMA, from (25) and (29) one finds

$$\frac{\sigma_r'^2}{\sigma_r^2} \approx \exp(-\gamma t). \tag{30}$$

As is well known in the theory of the operation of DMAs [14, 15], the electrical mobility of the classified particles is given by

$$\mu_e = \frac{q_c \ln(R_2/R_1)}{2\pi L \phi} \tag{31}$$

in the usual case in which the flow rate of inlet clean air, $q_c$, equals the flow rate of the outlet excess air. In the above equation, $L$ is the effective length of the DMA, that is, the distance measured from the inlet aerosol port to the classification port. Thus, for a specific DMA geometry ($R_1$, $R_2$, $L$) and clean air flow rate ($q_c$), the product of the applied voltage and electrical mobility of the classified aerosol is a constant:

$$\mu_e \phi = \text{constant} = \frac{q_c \ln(R_2/R_1)}{2\pi L}. \tag{32}$$

Inserting this result in (30), and taking $r_0 = R_2$, gives

$$\frac{\sigma_r'^2}{\sigma_r^2} \approx \exp\left(-\frac{q_c}{\pi L R_2^2} t\right). \tag{33}$$

In practice, one is fundamentally interested in the degree of particle dispersion at the classification port, that is, after the particles have travelled a time equal to the mean aerosol residence time in the apparatus. The latter is given by $t_R = \pi L(R_2^2 - R_1^2)/(q_c + q_a)$, where $q_a$ is the inlet aerosol flow rate. Therefore,

$$\left(\frac{\sigma_r'^2}{\sigma_r^2}\right)_{t=t_R} \approx \exp\left[-\frac{1-(R_1/R_2)^2}{1+(q_a/q_c)}\right]. \tag{34}$$

The aerosol-to-sheath flow rate ratio, $q_a/q_c$, is a DMA operation parameter of great importance because it is related to the width of the mobility interval of the classified particles. Indeed, in the absence of diffusional effects, the half-width of the classified mobility band is $\Delta\mu_e = \mu_e q_a/q_c$ [15]. In order to produce an aerosol as monodisperse as is practicable, the $q_a/q_c$ ratio is usually kept low, about 0.1. On the other hand, a widely used DMA for aerosol nanoparticles at present is the short column of TSI Inc., for which $L = 11.11$ cm, $R_1 = 0.937$ cm, and $R_2 = 1.958$ cm; for this column, thus, $R_1/R_2 = 0.479$. For these values, the variance ratio becomes $\left(\sigma_r'^2/\sigma_r^2\right)_{t=t_R} \approx 0.5$. Thus, by simply reversing the direction of particle motion in the DMA one can halve the extent of particle dispersion. This is certainly a great improvement. Obviously, it is desirable to demonstrate the validity of this prediction experimentally, but this will only be possible after the new instrument, the reverse DMA, is constructed.

All the conclusions arrived at in this section are valid for other processes in which charged particles move driven by a non-uniform electric field. Thus, another example of great industrial importance is the electrostatic precipitator [22] used to remove particulate matter from a gas exhaust previous to its emission to the atmosphere. However, although the operating physical principle is the same, for electrostatic precipitators one is interested not in producing a narrow beam of monodisperse particles, but rather in precipitating them all on the collector electrode. Therefore, in spite of the fact that in this case too one can manipulate the degree of particle dispersion by selection of the particle migration direction, it is of no practical use because this would not affect the particle collection efficiency of the equipment.

## 7. Qualitative explanation of the effect of the direction of particle motion on the variance

In the previous section we have seen that the variance of the distribution of particle positions can be larger or smaller than that for a free particle depending on whether the aerosol moves toward increasing or decreasing (absolute) values of the field strength, respectively. A qualitative explanation of this effect can be given as follows. Consider two particles separated by a distance $s$ at time $t$. During the subsequent time interval $\Delta t$, the particles undergo two types of displacement, namely, a deterministic displacement due to the external field, and a random displacement due to molecular bombardment of the fluid medium. The latter, on average, has the same magnitude for both particles. But the magnitude of the deterministic displacement is different for the two particles. Consider the DMA case discussed above, and suppose that particle A is closer to the inner electrode than particle B. Hence, particle A feels an electric force larger than that felt by particle B. Now, if both particles on average are migrating toward the inner electrode (i.e. in the direction of increasing field strength), the deterministic displacement during the time interval $\Delta t$ is larger for A than for B and, consequently, the separation distance between them increases—the dispersion increases. But if they migrate toward the outer electrode, i.e. in the direction of decreasing absolute values of the field strength, A advances toward B at a speed higher than the speed at which B tries to 'escape' from A. In this second case, thus, the separation distance between A and B decreases and particle dispersion is reduced. In the first case, the advanced particle steadily escapes from the lagging one because the difference in their deterministic velocities increases as they approach the inner electrode. In the second case, in contrast, the lagging particle steadily approaches the advanced one as they move toward the outer electrode because the deterministic velocity of the former is always larger.

## 8. Conclusions

The variance of the position distribution of particles undergoing Brownian motion has been derived for the general case in which the particles are suspended in a flowing medium and are acted upon by an external field of force. It has been shown that for uniform flow and force fields, the variance is exactly the same as for a free particle in a stagnant medium. For non-uniform external fields, the variance can be larger or smaller than that for a free particle depending on whether the particles, on average, move toward increasing or decreasing absolute values of the field strength. This interesting result may find a useful application in the development of a new type of DMA with higher resolution power.

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