DRIFT DIFFERENTIAL MOBILITY ANALYZER

Ignacio G. Loscertales
Dep. Ingeniería Mecánica y Energética, Universidad de Málaga 29013, Spain

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Abstract—This work addresses the problem of narrowing the peak diffusive broadening observed in the response of differential mobility analyzers (DMA) when dealing with nanoparticles and ions. Instead of achieving this task by solely increasing the sheath gas flowrate through the instrument (i.e. increasing the Peclet number, $Pe$), as done in recent studies, we show that the broadening can be further reduced by establishing an axial electric field upon the classical transverse one. This still nonexistent instrument would be what we call a drift-DMA (DDMA). The asymptotic analysis ($Pe \gg 1$) indicates that a well designed DDMA might triple the resolution of Rosell’s DMA (Rosell-Llompart et al., J. Aerosol Sci. 27, 695–719) when both instruments run at the same $Pe$, placing this hypothetic instrument in an excellent position to even compete with standard ion drift tubes. On the other hand, the DDMA would yield the same resolution of a Rosell’s DMA but running at a Reynolds number $Re$ near one order of magnitude smaller, with the corresponding saving on pumping needs. © 1998 Elsevier Science Ltd. All rights reserved

NOMENCLATURE

$a$ constant appearing in the description of the sheath gas velocity profile, equation (28)
$A, B, C$ functions defined in equation (42)
$c$ proportionality constant appearing in equations (24) and (26)
$D$ diffusion coefficient of particles or ions in the sheath gas
$d$ for "entrance" flow, the boundary layer thickness, equation (65)
$E(r)$ electric field vector in the DDMA, equation (15)
$E_r, E_z$ radial and axial electric field components in the DDMA, equations (15) and (16)
$G_r(y), G_z(y)$ integrals defined in equation (30). Given, for different flow types, in equations (48) and (53)
$g(y)$ function collecting the dependence of $u_r$ on $y$, equation (28)
$K$ constant of order one, appearing in equation (79)
$k$ Boltzmann’s constant.
$L$ streamwise position at which aerosol particles reach the inner electrode
$L_o$ axial distance between the midpoints of the aerosol entrance and exit slits
$L_{max}$ shift of $L$ if the axial electric field is absent, equation (20)
$L_i, L_o$ width of the aerosol input and output slits, respectively
$Ne$ number of elementary charges on the particle
$n(x, r)$ number density of aerosol particles or ions having a fixed mobility
$n_1$ value of $n$ at the output slit, equations (26) and (45)
$Pe$ diffusional Peclet number, equations (11) and (23)
$q$ sheath gas flowrate
$q$ dimensionless axial length of a DDMA, equation (31)
$q_{in}, q_{out}$ flowrate of aerosol entering and leaving the DDMA, respectively
$q^*$ optimum value of $q$, equation (50)
$r$ independent radial variable, or distance of a given point to the symmetry axis
$R_i, R_o$ radius of the inner and outer electrode, respectively
$Re$ Reynolds number, equations (11) and (23), based on $Q$
$Re^*$ value of $Re$ at which diffusive broadening equals "slits" broadening, equation (78)
$s(r)$ time-like variable defined in equation (25), with dimensions of $Dt$
$s_1$ value of $s$ at $r = R_i$, equation (29)
$Sc$ Schmidt number, equation (11)
$T$ absolute temperature of the gas
$t = (BR_z)^2/\gamma$ ratio between the characteristic axial and radial electric fields, equation (30)
$t^*$ value of $t$ that minimizes the diffusive broadening, equation (43)
$u_r(r)$ axial velocity of the sheath gas inside the DDMA
$u_{r/\gamma}$ velocity vector of non diffusing particles inside the DDMA. equation (17)
$U$ mean velocity of the axial flow in the DDMA, based on $Q$, equations (1) and (23)
$\nu$ potential difference between points of the inner and outer electrodes located at the same axial position

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\(V_p\) \text{ value of } V \text{ at the peak of the mobility distribution}\n\(x\) \text{ independent variable fixing the axial position; null at the midpoint of the entrance slit}\n\(x_0\) \text{ in "entrance" flow, axial distance from the point where the boundary layer starts to develop to the midpoint of the aerosol input slit, equation (70)}\n\(y, y_1\) \text{ dimensionless radial variable; } y_1 \text{ corresponds to } r = R_1\n\(Z\) \text{ particle's electrical mobility}\n
\text{Greek Letters}\n\(a\) \text{ angle between the nondiffusive particle's trajectory and the axial direction at the DDMA exit slit}\n\(\beta\) \text{ value of the axial electric field in the DDMA, equation (16)}\n\(\gamma\) \text{ voltage variable defined in equation (16)}\n\(\gamma_v\) \text{ value of } \gamma \text{ at the peak of the mobility distribution}\n\(\delta(x)\) \text{ Dirac's distribution function, vanishing everywhere except at } x = 0\n\(\Delta L = \Delta x / \sin a\) \text{ standard deviation of the sedimentation length on the inner electrode due to diffusion equation (56)}\n\(\Delta V\) \text{ full-width at half-maximum of the mobility distribution}\n\(\Delta x = (2D_1) \cdot I^{1/2}\) \text{ standard deviation of the particles from their nondiffusive trajectory due to diffusion}\n\(\epsilon\) \text{ dimensionless boundary layer thickness, equation (65)}\n\(\eta\) \text{ stream function variable, with dimensions of length, equation (19)}\n\(\lambda = x_0 / R_2\) \text{ axial dimensionless location of the entrance slit}\n\(v\) \text{ sheath gas kinematic viscosity}\n\(\sigma\) \text{ dimensionless width of the input plus the output slits, equation (74)}\n\(\tau\) \text{ time variable, equation (58)}\n\(\tau^*\) \text{ particle's residence time in the DDMA, equations (4) and (59)}\n\(\phi = \gamma / \gamma_v\) \text{ dimensionless voltage variable of the DDMA, equation (46)}\n\(\psi = L / L_o\) \text{ dimensionless length of a regular DMA; It also corresponds to } V / V_p \text{ in a DMA}\n
\text{1. INTRODUCTION}\n
Differential mobility analyzers (DMAs) are devices commonly used in the field of aerosols to measure size distributions of charged particles. These instruments are also employed to generate aerosol standards from polydisperse aerosols: select particles having a given (known) size. In the widely adopted axisymmetric design, that can be traced back at least to Zeleny (1929), a radial electric field is established between two coaxial cylinders by maintaining a potential difference \(V\) between them. Clean gas (called sheath gas) flows through the annular gap between the cylinders. If charged particles are continuously injected at a given axial position through an annular slit in the outer cylinder, they will drift in the radial field towards the inner cylinder, as they are carried axially downstream by the sheath gas. As a result, the stream-wise position \(L\) at which the particles reach the inner cylinder is a measure of the inverse of their electrical mobility \(Z\). If another slit is opened in the inner cylinder, charged species of a given mobility may be continuously extracted through it. There exist parallel-plates designs of DMAs, although the cylindrical ones avoid the end-effects associated to the former.

The response of these DMAs strongly deteriorates when dealing with particles within the nanometer size range or with ions (Tammet, 1970; Kousaka et al., 1986; Stolzenburg, 1988). Recently, Rosell-Llompart et al. (1996) (hereinafter referred to as RL) have studied both theoretically and experimentally how diffusivity affects the performance of a cylindrical DMA working at high Peclot numbers \(Pe\). First, they have theoretically shown that the broadening due to particle diffusion varies linearly with \(Pe^{-1/2}\), namely \([\Delta V / V_p]^2 \propto Pe^{-1}\), and arrived at the conclusion that the proportionality constant, which mostly depends on the geometry of the instrument, reaches a minimum when the axial separation between the inlet and outlet slits, \(L_{op}\), is of the order of the annular gap between cylinders. Second, these authors built a DMA having a (near) optimum axial length (it will be referred to as the optimum DMA), and experimentally demonstrated their predictions.

Their conclusions indicate that the resolution of a cylindrical optimum DMA can only be augmented by increasing the \(Pe\), which can only be achieved by increasing the Reynolds number \(Re\). For instance, by running their optimum DMA at \(Re \sim 1300\), RL managed to attain resolutions of about 4% with entities having electrical mobilities \(Z \approx 1 \text{cm}^2 / \text{Vs}\) (typical of atmospheric ions). However, this high resolution requires very large sheath flowrtes, with the consequent high pumping capacity needs.
Another way of measuring the mobility of nanoparticles and other highly mobile entities like ions is by resorting to special instruments called ion drift tubes (IDT), which can measure ion mobilities with resolutions as high as 2% (Siems et al., 1994). These IDT apparatus are of the time-of-flight (TOF) type, where a burst of ions is injected at the axis of a tube where an axial electric field is maintained. The tube is filled with quiescent gas. The group of ions drift in the axial electric field towards the end of the tube, where they arrive at different times depending on their mobilities. Thus, the drifting time provides a measure of the inverse of the mobility. As RL noticed, to compete with an IDT, the optimum DMA “should be run at a Re ~ 10^4, where it is conceivable to maintain a laminar flow through the short analyzing path of a well designed DMA”. However, this would need, for the case of a Reischls DMA, the somehow disproportionate sheath flowrate of about 1600 lmin^-1. Despite that, RL also pointed out that the main advantage of a DMA is that it supplies a continuous stream of ions, while the IDT disperses a pulsed ion stream in time.

What we propose here is an alternative way of increasing the resolution of existing DMAs, besides increasing the sheath flowrate, by superposing an axial electric field to the classical radial one. The resulting instrument would be a combination of a DMA and an IDT, that we name drift-DMA (DDMA). We shall see that the DDMA concept allows, theoretically, to increase the resolution of existing DMAs by a factor of 3 while working at the same Re. This new instrument, of the continuous feeding type, would be in an excellent position to simultaneously work as an aerosol instrument as well as an ion mobility analyzer.

It must be kept in mind, however, that the present study deals only with the theory of this DDMA, which does not exist. Accordingly, we shall not attempt to consider the numerous technical difficulties that might have to be overcome to successfully build such an instrument.

The structure of this paper is as follows. In Section 2 we describe an oversimplified model to show how the resolution of differential mobility analyzers can be improved by means of the addition of an axial electric field. In Section 3 the diffusion theory at large Pe developed by Rosell-Llompart et al. (1996) is applied to a cylindrical DMA in which an independent axial electric field may be superposed to the classical radial electric field. We shall arrive at the conclusion that, for a fixed Re, the diffusive broadening may be reduced to a minimum value, depending on the type of flow of the sheath gas, by properly tuning the axial and the radial electric fields. In Section 4 we compare the predicted performance of this DDMA against that of Rosell’s DMA, since this instrument provides the best resolution for nanoparticles and ions to date. The issue of the finite width of the input/output aerosol slits upon resolution is treated in Section 5. Finally, Section 6 collects the main conclusions.

2. MECHANISM OF INCREASING THE RESOLUTION IN THE DDMA

The motivation for the present work is based on the idea of increasing the axial separation between the mean trajectories followed by a cloud of particles bearing different mobilities inside an analyzer, by properly tuning the transversal and the (new) axial electric fields. If this can be done at a rate faster than what diffusion axially broadens the cloud of particles, the consequence would be an increase in resolution of such analyzer.

To explain this concept qualitatively, let us consider the case of a plane (bidimensional) mobility analyzer,* such as the one sketched in Fig. 1. This instrument consists of two infinite flat plates, parallel to each other, separated by a constant distance h. The sheath gas flows between the plates. For the sake of simplicity, we shall assume that this flow is of the plug type: constant gas velocity U between plates. The aerosol enters and leaves the analyzer through two slits located at an axial distance l from each other. The aerosol flowrate is assumed to be negligible when compared with the sheath gas flow rate.

*The author explains here the plane model suggested by Prof. Tammet (Tammet, 1997) to clarify the mechanism responsible for the improvement in resolution.
The innovation here is that the plates are not equipotential, and the electric field is not perpendicular to the sheath gas, as expected in classic differential mobility analyzers. Instead, the electric field between plates is inclined but uniform, and the voltage $V$ between any pair of facing points (see Fig. 1) is the same along the plates. Accordingly, the vertical field in the analyzer is $E_y = V/h$, and the horizontal field is $E_x = t(V/h)$, where $t$, the ratio between the axial and vertical fields, is a free parameter that can be selected. Figure 1 also shows the mean trajectory of a cloud of particles having a fixed mobility.

By comparing the vertical and horizontal evolution of a single particle, one can easily obtain the relationship between any particle’s mobility $Z$ and its corresponding precipitation length $L$

$$L = \frac{U h}{Z V} - t h.$$

When $L = L_0$ this particle is selected and leaves the analyzer.

Let us now inject a continuous stream of particles, all of them having the same mobility $Z$. Due to Brownian diffusion, the cloud will expand during the passage, as illustrated in Fig. 1. The one-dimensional standard deviation of particles from the center of the cloud will be given by

$$\Delta x = \sqrt{2 D \tau^*},$$

where $\tau^*$ is the particle’s residence time inside the analyzer, and $D$ is the diffusion coefficient of the particles in the sheath gas. $D$ may be related to the particle’s mobility through Einstein’s equation

$$D = k T Z / N e,$$

where $k$ is the Boltzmann’s constant, $T$ the absolute gas temperature, and $N e$ the number of elementary charges on the particle. From the vertical motion of the particle, $\tau^*$ may be estimated by

$$\tau^* \approx h^2 / Z V,$$

so that

$$\Delta x \approx \sqrt{\frac{2 k T}{N e V}} h.$$

If the average or mean trajectory of the cloud is not perpendicular to the plate, the standard deviation of the sedimentation length $\Delta L$ would be $\Delta x$ but increased by the ratio of the hypotenuse and the left leg of the small triangle drawn in Fig. 1 near the outlet exit:

$$\Delta L \approx \Delta x \sqrt{1 + \left(\frac{L_0}{h}\right)^2}.$$
For a given geometrical and flow condition \((U, h)\) and a given type of particle \((Z\) also fixed), equation (1) gives us the voltage \(V\) at which the particle's mean trajectory is selected

\[
V = \frac{U h}{Z [t + L_o/h]},
\]

which depends on \(t\). Accordingly, the precipitation length in equation (6) varies with \(t\) as

\[
\Delta L \sim (Z t)^{1/2},
\]

whereas the deposition length \(L\) varies as

\[
L \sim (Z t).
\]

Therefore, the mean cloud trajectory \(L\) varies with \(t\) faster than the diffusive broadening \(\Delta L\) does. In other words, if we had a cloud consisting of entities having two different but very close mobilities, since the separation between their mean deposition length may be increased faster than their precipitation length by increasing \(t\), they could eventually be resolved by this instrument.

On the other hand, we can have a look at the resolution of a differential analyzer, defined as the relative standard deviation \(\Delta Z/Z_o\), where \(Z_o\) is the mobility associated to the peak of the distribution or apparent mobility. The measured or apparent mobility and the precipitation length are related through equation (1), so that

\[
\frac{\Delta L}{\Delta Z} \approx \frac{dL}{dZ} = \frac{U h^2}{V Z_o^2}.
\]

The ratio \(\Delta Z/Z_o\) can be easily derived from equations (1), (6) and (7). The result can be split into three dimensionless factors

\[
\frac{\Delta Z}{Z_o} \approx Sc^{-1/2} Re^{-1/2} F,
\]

where \(Sc = v \, Ne/(k T Z_o) = v/D\) is the Schmidt number, \(Re = U h/v\) is the Reynolds number \((v\) is the sheath gas kinematic viscosity), and the third factor is given by

\[
F = \sqrt{2 \frac{1 + (L_o/h)^2}{t + (L_o/h)}}.
\]

The product \(Pe = Sc \, Re\) is called the diffusional Peclet number, which allows to write the equations in a more compact form (see RL). However, the different roles of \(Sc\) and \(Re\) in controlling the resolution has to be kept in mind.

The Schmidt number is fixed for a given type of particle and fixed flow condition. The Reynolds number, however, may be varied, and should be chosen as large as possible, provided the flow between plates remains laminar. Finally, the factor \(F\) depends on the relative length \(L_o/h\) and on \(t\). If \(t\) is fixed, minimization of \(F\) provides the optimum relative length of the analyzer

\[
\frac{L_o}{h} = \sqrt{t^2 + 1} - t,
\]

which yields the corresponding minimum value of \(F\)

\[
F = 2 \sqrt{(t^2 + 1)^{1/2} - t}.
\]

The resolution of a classic DMA, where \(t = 0\), can be improved by keeping its relative length \(L_o/h\) equal to one, as shown by RL. In the DDMA, where \(t > 0\), the resolution might be further improved by choosing a large value of \(t\), since \(F\) tends to zero as \(t\) increases. Furthermore, for large values of \(t\), the optimum relative length (equation (13)) also approaches zero, so that very short analyzer lengths would be required in this DDMA.
The qualitative analysis just developed embodies the physical reasons that might make this new approach to mobility analysis very attractive. However, the oversimplified model sketched above cannot be used to extract quantitative conclusions. Furthermore, the plane design is seldom used in present time, so we shall attempt to apply the same ideas to the case of an axisymmetric instrument, free of end effects, where the sheath gas velocity profile may depend on r, the radial coordinate.

3. TRANSPORT OF PARTICLES THROUGH A CYLINDRICAL DRIFT-DMA

We shall follow here the asymptotic theory developed by RL to describe the transport of particles through a cylindrical DMA. The author assumes that the reader has a good knowledge of RL’s work.

3.1. Non-diffusing particles

Let us assume we have an axisymmetric analyzer, in which we may vary the radial electric field independently of the axial electric field. A sketch of this DDMA is shown in Fig. 2. For the sake of simplicity, we further assume that the axial field is constant whereas the radial one depends only on r, the radial coordinate. Under these assumptions, the full electric field $E(r)$ between the coaxial cylinders is given in cylindrical polar coordinates by

$$E(r) = E_r e_r + E_\theta e_\theta, \quad R_1 < r < R_2,$$

where $R_1$ and $R_2$ are the radial location of the inner and outer cylinders, respectively, and $E_r$ and $E_\theta$ are given by

$$E_r = -\frac{\gamma}{r}, \quad \gamma = \frac{V}{\ln R_2/R_1}, \quad E_\theta = -\beta = \text{cte}, \quad 0 \leq \beta, \gamma.$$

In the absence of diffusion, the particle velocity $u_p(r)$ within the annular gap $R_1 < r < R_2$ is given by

$$u_p(r) = -\frac{\gamma Z}{r} e_r + (u_t(r) - \beta Z)e_\theta,$$

where $Z$ is the particle electrical mobility and $u_t(r)$ is the sheath gas velocity, taken here to be only $r$ dependant; this would be the case for a long instrument with a small annular gap.

As in RL, the particle concentration $n(x, r)$ in the gap is governed by the convection equation

$$u_r \nabla n = 0,$$

which may be solved as in RL to yield the trajectories along which $n$ is constant

$$\eta + \chi = \int_r^{R_2} \frac{(u_t(r) - \beta Z)}{\gamma Z} \, dr,$$

where $\eta$ is an integration constant. On the other hand $\eta = \eta(x, r)$ gives the particles trajectories. Since the particles are injected at $\eta = 0$ ($x = 0, r = R_2$), they are to be found nowhere in the flow region except at $\eta = 0$, so that $n(x, r) = \delta(\eta)$. Thus, particles of a given mobility will reach the inner cylinder at an axial position $L$ given by

$$L = \frac{Q}{2\pi \gamma Z} - \left[ \frac{\beta}{2\gamma} \right] R_2^2 (1 - y_1^2) = L_{DMA} - \bar{L}, \quad 0 < L$$

where $Q$ is the sheath gas flowrate given by

$$Q = 2\pi \int_{R_1}^{R_1} r u_t(r) \, dr.$$
Fig. 2. Sketch of an axisymmetric cylindrical DDMA. The outer and inner electrodes are defined by \( r = R_2 \) and \( r = R_1 \), respectively. The axial coordinate \( x \) is defined such that the aerosol enters the analyzer at \( x = 0 \) and leaves it at \( x = L \). The sheath flow rate \( Q \) flows from left to right. In this instrument it is possible to establish a constant axial electric field \( E_x \) along with the classical radial electric field \( E_r(r) \).

and \( y_1 \) is nothing but \( R_1 \) made dimensionless with \( R_2 \)

\[
y = \frac{r}{R_2} \rightarrow y_1 = \frac{R_1}{R_2}.
\]

Note from equation (20) that \( L \) is made up of two parts: \( L_{DMA} \), which corresponds to the axial location originated in a DMA (that is, if the axial field were absent), plus a new component \( \tilde{L} \) due to the axial field.

3.2. Transport when Brownian diffusion is present

In the case of diffusing particles, the equation governing the evolution of the particle concentration \( n \) in the annular gap is given by the standard diffusion-convection equation, namely

\[
u_r \nabla n = D \nabla^2 n,
\]

where \( D \) is the particle diffusion coefficient, related to the particle mobility \( Z \) through Einstein's equation (see equation (3))

\[
Z = \frac{NeD}{kT}.
\]

We shall consider the ideal situation when all the broadening is due to diffusion, so that \( n \) is infinitely narrow at \( r = R_2 \), and is described through Dirac’s distribution \( \delta \)

\[
n(R_2, x) = \delta(x).
\]

By following RL approach, under large Pe assumption, where

\[
Pe = Re \frac{v}{D} = \frac{UR_2(1 - y_1)}{D}, \quad U = \frac{Q}{\pi R_2^2(1 - y_1^2)},
\]

the solution of equation (21) subject to equation (22) is

\[
n = cs^{-1/2} \exp \left[ -\frac{\eta^2}{4s} \right],
\]

where \( c \) is an appropriate proportionality constant and \( s(r) \) is given by

\[
s(r) = D \int_r^{R_2} \left[ 1 + \frac{(u_r(r) - \beta Z)^2 r^2}{(\gamma Z)^2} \right] \frac{r}{(\gamma Z)} \, dr.
\]
At the DDMA exit, \( r = R_1 \) and \( x = L_\infty \), the particle concentration \( n_1 = n(R_1, L_\infty) \) will be given by

\[
n_1 = n(R_1, L_\infty) = c s_1 - 1/2 \exp \left( - \frac{\eta^2}{4 s_1} \right),
\]

where \( s_1 = s(r = R_1) \). The exponential term in equation (26) may be written in terms of the dimensionless DDMA length \( \psi \), defined as

\[
\psi = L_\infty / L,
\]

where \( L \) is defined in equation (20), so that

\[
- \frac{\eta^2}{4 s_1} = - L^2 \frac{(1 - \psi^2)^2}{4 s_1} = - \frac{L_\infty^2}{4 s_1} (1 - \psi^2)^2.
\]

The value of \( s_1 \) depends on the sheath gas flow, \( u(y) \). Let us describe such a flow in a general way by

\[
u_2 = L_\infty \psi (2 - \psi)
\]

\[
u_2 = L_\infty \psi \frac{d\psi}{y_{\infty}^3} = \frac{2 L_\infty}{\gamma}
\]

By substituting equation (28) into equation (25), and integrating between \( y_1 \) and \( 1 \), one gets a general expression for \( s_1 \)

\[
s_1 = \frac{D R_2}{\gamma Z} \frac{1}{2} (1 - \psi^2) + \frac{q}{\psi} + t \left( \psi \frac{d\psi}{y_{\infty}^3} \right) G_1 - 2 \left( \frac{U}{\beta Z} \right) G_2 + \left( \frac{1 - y_1^4}{4 t^2} \right),
\]

where \( G_1, G_2 \) and \( t \) are defined as

\[
G_1 = \int_{y_1}^{1} g(y) y^3 dy, \quad G_2 = \int_{y_1}^{1} g(y) y^3 dy, \quad t = \frac{\beta R_2}{\gamma}
\]

By means of equation (20), one may write

\[
\frac{U}{\beta Z} = \frac{1}{t} \left( \frac{q}{\psi} + t \right), \quad q = \frac{2 L_\infty}{R_\psi (1 - \psi^2)}.
\]

By plugging equation (31) into equation (29) one arrives at

\[
s_1 = \frac{D R_2}{U} \frac{1}{2} (1 - \psi^2) + \left( \frac{q}{\psi} + t \right) G_1 - 2 t \left( \frac{q}{\psi} + t \right) G_2 + \left( \frac{1 - y_1^4}{4 t^2} \right)
\]

so that

\[
\frac{\eta^2}{4 s_1} = \frac{1}{H(q, t, y_1, \psi)} \left( 1 - \psi^2 \right)^2,
\]

where \( H \) is given by

\[
H = \frac{16}{q^2 (1 - y_1^4)} \psi (q + t) \frac{1}{2} (1 - \psi^2) + \left( \frac{q}{\psi} + t \right) G_1 - 2 t \left( \frac{q}{\psi} + t \right) G_2 + \left( \frac{1 - y_1^4}{4 t^2} \right)
\]

As pointed out by RL, if \( Pe \) is large enough, the exponential term of \( n_1(\psi) \) in equation (26) will drop rapidly to zero when \( \psi \) departs slightly from 1, so that \( n_1(\psi) \) might be well

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1RL calls this variable \( \psi \) the dimensionless DMA voltage. However, as it will be shown, \( \psi \) does not play the same role in our DDMA.
approximated by
\[ n_1 \simeq \exp \left[-\frac{Pe (1 - \psi)^2}{H(\psi = 1)}\right] \], \quad \frac{Pe}{H(\psi = 1)} \gg 1. \] (34)

Under these assumptions, the dimensionless width of the distribution \( n_1 \) in terms of \( \psi \) is simply given by
\[ \left( \frac{\Delta \psi}{\psi = 1} \right)^2 = \frac{4 \ln 2}{Pe} \frac{1}{H(\psi = 1)}. \] (35)

In order to minimize \((\Delta \psi/(\psi - 1))^2\) while keeping \( Pe \) constant, one must simply minimize \( H(\psi = 1) \),
\[ H(\psi = 1) = \frac{16}{q^2(1 - \gamma_1^2)} (q + t) \left\{ \frac{1}{2} (1 - \gamma_1^2) + (q + t)^2 \left( G_1 - 2t(q + t)G_2 + \frac{1}{4}t^2 \right) \right\}. \] (36)

However, it is clear from equation (36) that \( H(\psi = 1) \) increases with \( t \) (remember that \( 0 < t, 0 < G_1, G_2 \)), so that the function \( H(\psi = 1) \) has a global minimum for \( t = 0 \) and \( q = -(1 - \gamma_1^2)/2G_1^{1/2} \), which is exactly the result obtained by RL.

Thus, it appears that the effect of an axial field \( (t > 0) \) would be no other than decreasing the resolution of the instrument, in apparent contradiction with the results sketched in Section 2.

What really happens, however, is that the formulation in terms of \( \psi \) is not appropriate to describe the performance of our DDMA. The reason is that the physical spectral variable of the device is the voltage \( \gamma \); for RL optimum DMA, the dimensionless DMA voltage \( \gamma/(\gamma_p) \), where \( \gamma_p \) is the voltage at which the peak of \( n_1 \) appears, is exactly equal to \( \psi \):
\[ \psi = \frac{L_0}{L} = \frac{Q}{2\pi Z\gamma_0} = \frac{\gamma}{\gamma_p}, \] (37)

so that minimizing the width in terms of \( \psi \) is nothing but minimizing the broadening, since
\[ (1 - \psi)^2 = \left( \frac{\gamma_p - \gamma}{\gamma_p} \right)^2 \approx \frac{1}{4} \left( \frac{\Delta \gamma}{\gamma_p} \right)^2. \]

However, for our DDMA one has
\[ \psi = \frac{L_0}{L} = \frac{R_2(1 - \gamma_1^2)}{2} \left[ \frac{U R_2}{\gamma_p Z} - \frac{t}{2} \right] = \left[ \frac{U R_2}{\gamma_p Z} - \frac{t}{2} \right] = -\frac{\gamma}{\gamma_p}. \] (38)

In fact, if one writes \( \gamma \) as a function of \( \psi \), one obtains
\[ \gamma = \frac{U R_2}{Z} \left( \frac{1}{t + \frac{q}{\psi}} \right), \quad \gamma_p = \gamma(\psi - 1) = \frac{1}{t + \frac{q}{\psi}}, \] (39)

so that the broadening \((\Delta \gamma/\gamma_p)^2\) may be written as
\[ \frac{1}{4} \left( \frac{\Delta \gamma}{\gamma_p} \right)^2 \approx \left( \frac{\gamma_p - \gamma}{\gamma_p} \right)^2 \approx \left( 1 - \frac{\gamma}{\gamma_p} \right)^2 \approx \frac{(1 - \psi)^2}{(1 + \frac{t}{q} \psi)^2} \neq (1 - \psi)^2. \] (40)

Nevertheless, since one knows the width of the distribution in terms of \( \psi \) (equations (35) and (36)) and the relationship between \( \psi \) and \( \gamma \) (equation (39)), the real width in terms of \( \gamma \) can be
readily obtained
\[
\left( \frac{\Delta y}{\gamma_p} \right)^2 = \frac{64 \ln 2}{\text{Pe}(1 - y_r^2)(q + t)} \left\{ \frac{1}{2} (1 - y_r^2) + G_q(q + t)^2 - 2tG_q(q + t) + \frac{1 - y_r^4}{4} t^2 \right\},
\]
which, after reordering in powers of \( t \), becomes
\[
\left( \frac{\Delta y}{\gamma_p} \right)^2 = \frac{64 \ln 2}{\text{Pe}(1 - y_r^2)(q + t)} \left\{ At^2 + Bt + C \right\},
\]
where
\[
A = G_1 \quad 2G_2 \frac{1 - y_r^4}{4},
\]
\[
B = 2qG_1 - 2qG_2 = 2q(G_1 - G_2),
\]
\[
C = \frac{1}{2} (1 - y_r^4) + G_1q^2.
\]
According to equation (42), the way \( (\Delta y/\gamma_p)^2 \) depends on \( t \) suggests the possible existence of an optimum value \( t = t^* \), which depends on the type of flow, that minimizes the broadening.

In fact, by doing
\[
\frac{\partial}{\partial t} \left[ \left( \frac{\Delta y}{\gamma_p} \right)^2 \right] = 0,
\]
together with the condition \( t^* \geq 0 \), one obtains
\[
t^* = \left( \frac{1 - y_r^4}{2A} \right)^{1/2} \left( \frac{1 + y_r^4}{q^2 + 1} \right)^{1/2} - q.
\]
Thus, the optimum performance that can be expected from the DDMA is given by
\[
\left( \frac{\Delta y}{\gamma_p} \right)^2 = \frac{64 \ln 2}{\text{Pe}(1 - y_r^2)(q + t^*)} \left\{ At^{*2} + Bt^* + C \right\},
\]
the particle distribution at the DDMA exit being given by
\[
n_1 = c \exp \left\{ -\frac{16}{(1 - y_r^2)(q + t^*)} \phi \left\{ \frac{1}{2} (1 - y_r^2) + G_q \left( \frac{q + t^*}{\phi} \right)^2 - 2t^*G_q \left( \frac{q + t^*}{\phi} \right) + \frac{1 - y_r^4}{4} t^{*2} \right\} \right\},
\]
where
\[
\phi = \gamma_p
\]
The relative broadening given in equation (44) is to be compared to that given by the Rosell’s DMA which, written in our variables, is given by \(^4\)
\[
\left( \frac{\Delta y}{\gamma_p} \right)^2_R = \frac{64 \ln 2}{\text{Pe}} \frac{1}{2},
\]
where the subindex R stands for Rosell’s DMA. However, since the value of \( t^* \) (and so the optimum resolution) depends on the type of flow through \( G_1 \) and \( G_2 \), it seems convenient to

\(^1\)In equation (32) of RL, there also appears the factor \( G(y_r) \), but since for most type of flows \( G(y_r) \approx 1 \), we have used \( G(y_r) = 1 \) on writing equation (47). We have also taken \( q = 2(1 + y_r) \), since this value optimizes the performance (see RL).
particularize the comparison for various types of flows. In particular, we shall look at the
two limiting situations: plug flow and Hagen–Poiseuille flow. As we shall see, it will also
allow to identify the flow conditions in which the performance of the DDMA overcome by
far that of existing DMAs, as well as to choose the appropriate value of \( q \).

3.2.1. Plug-flow

For this type of flow one has

\[
a = 1, \quad g(y) = 1 = \text{cte,} \quad G_1 = G_2 = \frac{1}{2} (1 - y_1^2),
\]

\[
A = B = 0, \quad C = \frac{1}{2} (1 - y_1^2) [1 + \frac{1}{2} (1 + y_1^2) q^2].
\]

(48)

Since \( A = 0 \), equation (43) implies that \( \tau^* \to \infty \). Equation (44) becomes

\[
\left( \frac{\Delta \gamma}{\gamma^*_p} \right)^2 = \frac{64 \ln 2}{\text{Pe}} \frac{1}{2(q + \tau)} \left[ 1 + \frac{1}{2} (1 + y_1^2) q^2 \right],
\]

(49)

where the subindex \( P \) stands for plug flow. Noteworthy, the numerator of equation (49)
becomes independent of \( \tau \), whereas the denominator increases linearly with \( \tau \). This behavior
suggests that \( (\Delta \gamma_{y, P}, \gamma^*_P)^2 \) can be made as small as desired by simply increasing the value of \( \tau \). In
other words, the resolution tends to infinity with \( \tau \).

On the other hand, from equation (49) one might also obtain the most appropriate value
of \( q = q^* \), for a given \( \tau \), that further minimizes the broadening, which turns out to be

\[
q^* = \tau \left[ \left( 1 + \frac{2}{(1 + y_1^2) \tau^2} \right)^{1/2} - 1 \right].
\]

(50)

Since for this type of flow one would operate at large values of \( \tau \), for which \( q \sim \tau^{-1} \to 0 \), the
best performance would be obtained for \( q = 0 \):

\[
\left( \frac{\Delta \gamma}{\gamma^*_p} \right)^2 = \frac{64 \ln 2}{\text{Pe}} \frac{1}{2 \tau}.
\]

(51)

This situation, \( q = 0 \), corresponds to the case where the aerosol input and output slits are
located at the same axial position.

3.2.2. Hagen-Poiseuille flow

This type of flow is represented by

\[
g(y) = \frac{1 - y^2}{1 - y_1^2} = \frac{\ln y}{\ln y_1}, \quad y = \frac{r}{R_2}, \quad y_1 = \frac{R_1}{R_2},
\]

\[
a = \frac{1 - y_1^2}{2F(y_1)}, \quad F(y_1) = \int_{y_1}^{y_1} y g(y) \, dy.
\]

(52)

The factors \( G_1 \) and \( G_2 \) are given by

\[
G_1 = \frac{(1 - y_1^2)^4}{2 \ln y_1} + \frac{1 + y_1^2}{2} \left[ \frac{12 (y_1^6 + y_1^4 + y_1^4 + 1) \ln^2 y_1}{288 (1 - y_1^2) \ln^2 y_1}ight.
\]

\[+ \left. \frac{20 (1 - y_1^2) (y_1^4 + y_1^2 + 1) \ln y_1 + 9 (1 + y_1^2) (y_1^2 - 1)^2}{288 (1 - y_1^2) \ln^2 y_1} \right],
\]

\[
G_2 = \frac{1 - y_1^2}{2 \ln y_1} + \frac{1 + y_1^2}{2} \left[ \frac{1}{16 \ln y_1} + \frac{y_1^4 + y_1^4 + 1}{12} \right].
\]

(53)

In this case, \( A \neq 0 \) for \( 0 < y_1 < 1 \), so the expressions in (43) and (44) have their full meaning.
4. PREDICTED PERFORMANCE: DDMA AGAINST ROSELL'S DMA

To assess the convenience of using a DDMA when dealing with high mobility entities (i.e. nanoparticles, ions), we shall compare its theoretically predicted performance against that of an optimum Rosell's DMA. The reason for choosing Rosell's instrument as a reference is because, to date, this DMA has yielded the best resolution for nanoparticles and ions.

The resolution of Rosell's DMA is given in equation (47), whereas that of the DDMA is given explicitly in equation (51) for the plug flow and, implicitly, in equation (44) for the Hagen–Poiseuille flow. To compare, it seems convenient to get rid of the common factor $(64 \ln 2) / Pe$. Thus, we define

$$R = \left( \frac{\Delta y}{\gamma_p} \right)^2 \frac{Pe}{64 \ln 2}$$

so that $R_R$, $R_P$ and $R_{HP}$ represent the Rosell's DMA, the DDMA (plug flow) and the DDMA (Hagen–Poiseuille flow) "normalized" resolutions, respectively.

4.1. Plug flow

In this case

$$R_P / R_R = 1 / t$$

indicating that the DDMA, operated at the same Re than a Rosell's DMA, can reduce the diffusive broadening to almost any desired value by simply increasing $t$.

As an example, Fig. 3 shows the response of both instruments when $Pe = 1000$ (e.g. $Re = 200, Z = 1 \, \text{cm}^2 \, (\text{vs})^{-1}$). The vertical axis represents $n_1$, but normalized such that $n_1(\phi = 1) = 1$. The horizontal one is the dimensionless voltage $\phi$ defined in equation (46). The curved labeled as "Rosell" indicates the output expected from the optimum Rosell's DMA. The other two curves, labeled as "$q = 0, t = 10$" and "$q = 0, t = 1000"$, correspond to the expected response of the DDMA, equation (45) ($q = 0$), operated at $t = 10$ and $t = 1000$, respectively. For the case of $t = 10$, the resolution of the DDMA is about 3 times better than that of Rosell's DMA, and it goes up to a factor near 15 when $t = 1000$. However, for such a large value of $t$, the response of the DDMA deteriorates in the sense that $n_1$ loses its Gaussian shape. Despite this drawback, it seems clear that, for the case of
plug flow, the DDMA is far more appropriate than Rosell's DMA to deal with nanoparticles and ions.

4.2. *Hagen–Poiseuille flow*

Before one proceeds with the comparison, one must look for the optimum value of \( q = q^* \) that further minimizes the resolution of our DDMA. Although an analytic expression of \( q^* \) may be obtained from equations (44), (52) and (53), it is much more straightforward to simply plot \( \pi \) vs \( q \), taking \( y_1 \) as a parameter. Figure 4 shows such a plot. The horizontal axis is the value of \( q \) as defined in equation (31), and the different curves correspond to different values of \( y_1 \). Except, perhaps, for the case of small \( y_1 \), taking \( q^* = 0 \) seems to be the proper choice. Accordingly, we will constrain ourselves to a DDMA having an interslit length \( q = 0 \). Note that, in this case, \( \pi = \pi(y_1) \).

In Fig. 5 we represent the dependence of \( \pi \) (vertical axis) upon \( y_1 \) (horizontal axis). Clearly, only an increase of resolution of about 25% is, at most, possible in this situation.

4.3. *Effect of flow type on DDMA's resolution*

An explanation for the later result may be based on the type of trajectories the selected particles follow within the analyzer, as we did in Section 2. Figure 6 shows the paths travelled by the particles when the sheath gas flow is of the HP type and diffusion is neglected. When brownian motion is present and in the limit of \( Pe \gg 1 \), diffusion primarily occurs across the \( q \) coordinate (see equation (19)), that is, "perpendicularly" to the non-diffusive paths, as sketched in Fig. 7. According to this, the axial length \( \Delta L \) spanned by the selected particles at \( y = y_1 \) due to diffusion may be approximated by

\[
\Delta L \approx \Delta x / \sin \alpha,
\]

where \( \alpha \) is the angle formed by the non-diffusive path at \( y = y_1 \) and the axial direction, and \( \Delta x \) is the diffusive broadening perpendicular to the non-diffusive path. By using equation (19) it is easy to compute \( \sin \alpha \) from \((dx/dr)_{r=R_1}\), which for the HP flow yields

\[
\left( \frac{dx}{dr} \right)_{r=R_1} = ty_1 = tg(\beta) \rightarrow \sin \alpha = (1 + y_1^2 t^2)^{-1/2}.
\]

On the other hand, \( \Delta x \) may be estimated from the diffusion coefficient \( D \) and the residence time \( r^* \) the particles spend within the analyzer

\[
\Delta x \approx (2D t^*)^{1/2}.
\]
Fig. 5. Comparison of the response of an optimum Rosell’s DMA and a DDMA when the sheath flow is of the Hagen–Poiseuille type. The DDMA is chosen such that $q = 0$. The plot shows how $R_{HP}/R_R$ (vertical axis) varies with $y_1$ (horizontal axis).

To compute $\tau^*$ let us use the radial motion of the selected particles

$$\frac{dr}{dt} = -\frac{yZ}{r} \frac{r^2}{2} + cte = -\gamma Z \tau.$$

Imposing the initial condition $r = R_2$ when $\tau = 0$ yields

$$\tau = \frac{R_2^2}{2} (1 - y_1^2) \frac{1}{\gamma Z}.$$

(58)

The residence time $\tau^*$ will be obtained by doing $y = y_1$

$$\tau^* = \frac{R_2^2}{2} (1 - y_1^2) \frac{1}{\gamma Z}.$$

(59)

Accordingly, one is led to

$$\Delta L \approx \frac{\Delta x}{\sin \alpha_0} \approx \left[2D\tau^*(1 + y_1^2\tau^2)\right]^{1/2}.$$

(60)
Fig. 7. Detail of the particle arrival at the output slit. In the limit $Pe \gg 1$, particles mostly diffuse perpendicularly from the nondiffusive path, producing a broadening $\Delta x$ of the particle distribution. This broadening is translated to a sedimentation length broadening $\Delta L$ through the angle $\alpha_p$ formed by the non-diffusive path at $y = y_1$ with the axial direction.

which may be written in terms of $\tau$, $Pe$ and $q$ by means of equation (20) and noticing that, for selected particles $L = L_{\text{on}}$ to yield

$$\Delta L \approx R_d[1 - y_1]\left\langle \frac{1 + y_1}{\pi Pe} \right\rangle^{1/2}(q + t)^{1/2}(1 + y_1^2 t^2)^{1/2}. \tag{61}$$

To envisage how the resolution varies with $\tau$, one must compare $\Delta L$ with the axial separation experienced by particles other than the selected ones. Let $Z'$ be the mobility of such entities. The axial position $L$ at which they strike the inner electrode will be given by equation (20) if $Z$ is replaced by $Z'$. Let

$$Z' = Z + \Delta Z, \quad \Delta Z/Z \ll 1, \quad L = L_{\text{on}} + l.$$

In this situation, $l$ is given by

$$l = \frac{R_d^2}{2}(1 - y_1^2)(q + t)\left(\frac{\Delta Z}{Z}\right). \tag{62}$$

The limiting resolving power of the instrument will be determined by the conditions at which $l \sim \Delta L$, that is, when the diffusive broadening spans up to the axial position where the particles having mobilities $Z$ reach the inner rod. In particular, by equating $l$ and $\Delta L$ one obtains

$$\frac{\Delta Z}{Z} \approx [\pi(1 + y_1) Pe]^{1/2}\left\langle \frac{1 + y_1^2 t^2}{q + t} \right\rangle^{1/2}. \tag{63}$$

Clearly the expression above resembles equation (42), and also points out that the resolution in terms of mobility has a maximum at a certain value of $\tau$, as obtained previously. If one computes the same quantity $\Delta Z/Z$ for the case of plug flow, one ends up with

$$\frac{\Delta Z}{Z} \approx [\pi(1 + y_1) Pe]^{-1/2}\left\langle \frac{1 + y_1^2 q^2}{q + t} \right\rangle^{1/2}, \tag{64}$$

similar to that in equation (51). As expected, the term $t^2$ in the numerator of equation (63) has disappeared; instead, one finds the constant factor $q^2$. Therefore, the resolution of the DDMA when the sheath flow is of the plug-flow type increases with $\tau$, as previously obtained in Section 3.2.1.
In conclusion, it seems to be the bend suffered by the non-diffusive path that causes the resolution to go down. One must keep in mind, however, that the bend itself is caused by the different \( r \) dependences exhibited by \( u_r \) and \( E_r \). The way of withdrawing the best possible resolution out of a DDMA would then be to manipulate either \( E_r \) or \( u_r \), such that both fields be subject to the same \( r \) dependence, a very difficult task.

Thus, once one has chosen or fixed \( E_r \), the type of flow that the sheath gas creates between the electrodes becomes a critical issue from the point of view of a DDMA.

In many practical situations, the existing DMAs run at a moderate or high Reynolds number \( Re \), so that the flow between electrodes actually resembles something close to a plug-flow, except for the boundary layers that develop on the solid walls of the instrument. This is even more so, since most of the actual DMAs bear a wind tunnel type contraction prior to the analyzer to decrease the possible fluctuating components of the gas velocity.

4.4. Entrance flow

In this section we shall analyze the response of a DDMA when the sheath flow is of the entrance flow type: a flat velocity profile with two thin boundary layers developed on the electrodes. For the sake of simplicity, the Blausius type of boundary layer profile will be substituted by a linear interpolation. We expect this simplification not to affect seriously the performance of the DDMA compared to that with the real Blausius profile, since we will assume both boundary layers to be confined within a very thin region near the walls \((Re \gg 1)\). Accordingly, we shall assume that the sheath gas axial velocity \( u_d(y) = Uag(y) \), between electrodes is described by

\[
\begin{align*}
\alpha &= 1 + \frac{\varepsilon}{1 - y_1}, \quad \varepsilon = \frac{d}{R_2} \ll 1, \\
g(y) &= \frac{1}{\varepsilon} (y - y_1), \quad y_1 < y < y_1 + \varepsilon, \\
g(y) &= 1, \quad y_1 + \varepsilon < y < 1 - \varepsilon, \\
g(y) &= \frac{1}{\varepsilon} (1 - y), \quad 1 - \varepsilon < y < 1,
\end{align*}
\]

where \( U \) is defined in equation (23) and \( d \) represents the boundary layer thickness. In this case, the geometric factors \( G_1 \) and \( G_2 \) in equation (42) take the form

\[
\begin{align*}
G_1 &= \frac{1}{4} (1 - y_1) + \left\{ \frac{y_1 y_1 + 1}{2} = - \frac{1}{6} (1 + y_1^2) \right\} \varepsilon + O(\varepsilon^2), \\
G_2 &= \frac{1}{4} (1 - y_1^2) + \frac{1}{4} \left\{ y_1 (y_1 + 1) - (1 + y_1^2) \right\} \varepsilon + O(\varepsilon^2),
\end{align*}
\]

where only the linear terms in \( \varepsilon \) are retained, under the hypothesis that \( \varepsilon \ll 1 \). Since \( \lambda \neq 0 \) for \( 0 < y_1 < 1 \), equations (43) and (44) can be used, taking now the form

\[
\begin{align*}
t^* &= \frac{M}{\varepsilon^{1/2}} - q, \quad M = \left[ \frac{3 (1 - y_1^2)}{2 (1 + y_1^2)} \left\{ 1 + \frac{1}{2} (1 + y_1^2) \right\} q^2 \right]^{1/2} \sim O(1), \\
\left( \frac{\Delta y}{y_1} \right)^2 &= \frac{32 \ln 2 \varepsilon^{1/2}}{Pe} \left( \frac{2 (1 + y_1^2)}{3 (1 - y_1^2)} \left[ 1 + \frac{1}{2} (1 + y_1^2) \right] q^2 \right)^{1/2},
\end{align*}
\]

where the subscript \( E \) stands for entrance flow. Note that, in the last equation, only the smallest power of \( \varepsilon \) has been retained.

Again, the resolution depends on \( q \) and \( y_1 \). However, it is clear from equation (67) that \( q = 0 \) minimizes \( \Delta y / y_1 \), similarly to what we obtained in previous sections. This suggests that a well designed DDMA should have the input and output slits placed at the same axial
Fig. 8. Comparison of the response of an optimum Rosell's DMA and a DDMA when the sheath flow is of the entrance type. The vertical axis represents $\mathcal{R}_E/\mathcal{R}_R$, and the horizontal one represents $\log(Re)$. There are two groups of curves, corresponding to $y_1 = 0.7$ and $y_1 = 0.3$. Each group consists of three different curves, associated to the values $\lambda = 1$, $\lambda = 0.5$ and $\lambda = 0.1$.

position, so we shall constrain ourselves to the case $q = 0$. In this situation the resolution will then be expressed by

$$ (q = 0), \left( \frac{\Delta y}{y_p} \right)^{1/2} = \frac{32\ln 2}{\pi} \frac{2}{\epsilon^{1/2}} \left\{ \frac{2(1 + y_1^3)}{3(1 - y_1^2)} \right\}^{1/2} = \frac{64\ln 2}{\pi} \frac{2}{\epsilon^{1/2}} \left\{ \frac{1 + y_1^3}{6(1 - y_1^2)} \right\}^{1/2}. \quad (68) $$

Accordingly, one has

$$ \frac{\mathcal{R}_E}{\mathcal{R}_R} = 2\epsilon^{1/2} \left\{ \frac{1 + y_1^3}{6(1 - y_1^2)} \right\}^{1/2} = \frac{\mathcal{R}_E}{\mathcal{R}_R}(\epsilon, y_1). \quad (69) $$

From the boundary layer point of view, $\epsilon$ may be taken, in a first approximation, as

$$ \epsilon = \frac{d}{R_2} = \frac{d}{x_0} \frac{x_0}{R_2} \approx \frac{1}{\text{Re}^{1/2}} \lambda, \quad \lambda = \frac{x_0}{R_2}, \quad (70) $$

where $\text{Re}$ is the Reynolds number and $x_0$ represents the axial distance from the point where the boundary layer starts to develop to the input/output aerosol slits (remember that $q = 0$).

Thus, equation (69) may be written as

$$ \frac{\mathcal{R}_E}{\mathcal{R}_R} = 2\epsilon^{1/2} \left\{ \frac{1 + y_1^3}{6(1 - y_1^2)} \right\}^{1/2} = \frac{\mathcal{R}_E}{\mathcal{R}_R}(\text{Re}, y_1, \lambda). \quad (71) $$

Figure 8 collects the results of comparing the DDMA (entrance flow, $q = 0$) and Rosell's DMA. The vertical axis represents $\mathcal{R}_E/\mathcal{R}_R$, and the horizontal one represents $\log(\text{Re})$. There are two groups of curves, corresponding to $y_1 = 0.7$ and $y_1 = 0.3$. Each group consists of three different curves, associated to the values $\lambda = 1$, $\lambda = 0.5$ and $\lambda = 0.1$. For each group ($y_1 = \text{constant}$), one can see that increasing $\text{Re}$ and/or decreasing $\lambda$ produces a decrease on $\mathcal{R}_E/\mathcal{R}_R$. On the other hand, smaller values of $y_1$ at a constant $\text{Re}$ also reduces the value of $\mathcal{R}_E/\mathcal{R}_R$. Note that in a typical situation where $100 \leq \text{Re} \leq 1000$, a DDMA having $y_1 \approx 0.5$ and $\lambda \approx 0.5$ can double the resolution of a Rosell's DMA. On the other hand, to ensure the presence of a well defined electric field along the analyzer length, it is probably desirable to design a DDMA having values of $\lambda$ of order one or even larger. In this frame, a good performance would require small values of $y_1$.

In Fig. 9 we particularize this comparison to a case where both instruments are fed with particles bearing $Z = 1 \text{cm}^2 (\text{vs})^{-1}$, and both operate at $\text{Re} = 1500$ ($\text{Pe} = 7500$ in air). The
vertical and horizontal axes represent the normalized aerosol output $n_1$ and the dimensionless voltage $\phi$, respectively. The DDMA is chosen such that $y_1 = 1/3$ and $\lambda = 1$. Under these circumstances, the DDMA yields a peak which is about 2.6 times narrower than that produced by the Rosell's DMA.

It is also interesting to analyze the situation where both instruments yield the same resolution. From equations (47) and (68), this condition is described by

$$\text{Re}_E = \text{Re}_E^{0.5} \left\{ \frac{\lambda (1 + y_1^2)}{6(1 - y_1^2)} \right\}^{2/5},$$

where $\text{Re}_r$ and $\text{Re}_E$ are the Rosell's DMA and DDMA Reynolds number, respectively. The result above indicates that a DDMA might attain the same resolution as Rosell's DMA, but operating at smaller $\text{Re}$. This would imply a reduction of the pumping needs. As an example, let us consider the case of a DDMA having $\lambda = 1$ and $y_1 = 1/3$. In this particular situation one has

$$\text{Re}_E = 0.519 \text{Re}_r^{0.5}.$$  

Thus, the resolutions that one might obtain operating the Rosell's DMA at $\text{Re}_r \approx 3000$ would be the one obtained with this DDMA running at $\text{Re}_E \approx 314 (t^* = 4.8)$: near one order of magnitude smaller. In terms of flowrate, if one takes $R_2 = 3$ cm, the corresponding flowrate through the DDMA would be $Q = 35 \text{ min}^{-1}$ (air), and the resolution yielded for particles having $Z = 1 \text{ cm}^2 \text{ (vs)}^{-1}$ would be 5.4%.

5. BROADENING DUE TO OTHER EFFECTS

The analysis in Section 3.2 must be modified to include other factors that affect the broadening of the trajectories in addition to Brownian motion. As explained in Section 3.2 of RL, in a real experiment particles are fed with a finite spatial width associated with a finite aerosol flowrate $q_a$, so that $n_1$ would be given by a convolution of the initial distribution with the corresponding fundamental solution given in Section 3. Something similar happens at the aerosol output slit, where certain amount of gas $q_s$ is drawn, carrying with it particles from a finite set of streamlines. However, the fundamental solutions given in Section 3 may still hold in the limit when $q_a = q_s$ or $q_a, q_s \ll Q$. 
5.1. **Effect of the width of the input/output slits**

We address here the possible limiting effect of the small, but finite, width of the aerosol input and output slits whose lengths are denoted by \( l_i \) and \( l_o \) respectively. Figure 10 shows a sketch of a DDMA having \( q = 0 \). It includes two different aerosol non-diffusive trajectories, labeled as \( a \) and \( b \), corresponding to the path followed by the same aerosol when the DDMA is operated at two different values of \( \gamma \) while keeping \( t = \text{cte} \).

![Sketch of the input and output aerosol slits of a DDMA having \( q = 0 \). The length of the slits are \( l_i \) and \( l_o \) respectively. It also shows two different aerosol nondiffusive trajectories, labeled as \( a \) and \( b \), corresponding to the path followed by the same aerosol when the DDMA is operated at two different values of \( \gamma \) while keeping \( t = \text{cte} \).](image)

According to this, the maximum resolution given by the DDMA would be

\[
\frac{\Delta \gamma}{\gamma_p} \approx \frac{2|\gamma' - \gamma_p|}{\gamma_p}.
\]

Thus, one must write the equation above in terms of known quantities to, finally, compare this resolution to that due to diffusion. From equation (20), one has

\[
\text{trajectory } a: \gamma = \gamma_p, L = 0 \Rightarrow \gamma_p = \frac{Q}{2 \pi Z tR_2(1 - \gamma_1^2)}.
\]

\[
\text{trajectory } b: \gamma = \gamma', L \approx \frac{l_i + l_o}{2} \Rightarrow \gamma_p = \frac{Q}{2 \pi Z tR_2(1 - \gamma_1^2)} - \frac{Q}{2 \pi Z \gamma'} tR_2(1 - \gamma_1^2).
\]

By combining the expressions above, one is led to

\[
\left(\frac{\Delta \gamma}{\gamma_p}\right)_s \approx \frac{2}{1 + \frac{t(1 - \gamma_1^2)}{\sigma}}, \quad \sigma = \frac{l_i + l_o}{R_2},
\]

where the subscript \( s \) stands for \( \text{"slits"} \).

Interestingly enough, this \( \text{"slits"} \) resolution depends on \( \sigma \) and \( t \), whereas for a DMA depends only on \( \sigma \). Furthermore, equations (74) indicates that the \( \text{"slits"} \) broadening decreases as \( t \) gets larger, exactly as happened with the diffusive broadening.

For the case of plug flow (equation (51)), operating the DDMA at a sufficiently large value of \( t \) ensures that the limiting resolution is fixed by diffusion, since \( (\Delta \gamma/\gamma_p)_p^2 \approx 1/t \), whereas \( (\Delta \gamma/\gamma_p)_s^2 \approx 1/t^2 \). But most important, both resolutions tend to infinity as \( t \) does.

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*We have seen that this choice, \( q = 0 \), is the most appropriate for a DDMA.*
For the case of entrance flow (equation (67)), since the optimum value of \( t = t^* \) is given as a function of \( \varepsilon \) and \( y_1 \) (or more precisely, as a function of \( \text{Re}, \lambda \) and \( y_1 \)), there might be regimes where the “slits” resolution is larger than the diffusive one and vice versa. To find out these regimes, let us compare both resolutions

\[
\frac{(\Delta y)^2_{p}}{(\Delta y)^2_{E}} = \left[ \frac{4}{1 + \frac{t^*(1 - y_1^2)}{\sigma}} \right]^{1/2} \frac{1}{64 \ln 2 \left( \frac{6(1 - y_1^2)}{(1 + y_1^2)} \right)^{1/2}},
\]

where \( t^* \) is defined in equation (67) with \( q = 0 \). Note that

\[
t^*(1 - y_1^2) = \frac{1}{\sigma^{1/2}} \left[ \frac{3(1 - y_1^2)^3}{2(1 + y_1^2)} \right]^{1/2},
\]

which by virtue of equation (70) reads

\[
\frac{(\Delta y)^2_{p}}{(\Delta y)^2_{E}} = \sigma^{1/2} \left( \frac{v}{D} \right)^{1/2} \left[ \frac{6(1 + y_1^2)}{(1 - y_1^2)^3} \right]^{1/2} = f \left( \lambda, \sigma, y_1, \text{Re}, \left( \frac{v}{D} \right) \right),
\]

where \( v \) is the carrier gas kinematic viscosity and \( D \) the particle diffusivity coefficient. Thus, for a fixed geometry (\( \lambda, \sigma \) and \( y_1 \)), the different regimes will be determined by the Reynolds number \( \text{Re} \). In fact, by equating

\[
(\Delta y)^2_{p}/(\Delta y)^2_{E} = 1,
\]

one obtains the value of the \( \text{Re} = \text{Re}^* \) at which one of these resolutions takes over the other

\[
\text{Re}^* = \frac{1}{\lambda^{4/3} \sigma^{8/3} \left( \frac{D}{v} \right)^{1/3} \left[ \frac{(1 - y_1^2)^5}{6(1 + y_1^2)} \right]^{2/3}}.
\]

In Fig. 11 we plot the values of \( \log(\text{Re}^*) \) (vertical axis) as a function of \( \sigma \) (horizontal axis), taking \( y_1 \) and \( \lambda \) as parameters. In this particular case we have taken \( D/v = 0.168 \), which would correspond to ions having one charge, \( Z = 1 \text{ cm}^2 \text{ (vs)}^{-1} \), moving through air at room conditions. There are two groups of curves corresponding to the values of \( y_1 = 0.7 \) and \( y_1 = 0.3 \), respectively. Each of these groups consist of three different curves associated to the values of \( \lambda = 1 \), \( \lambda = 0.5 \) and \( \lambda = 0.2 \), as indicated in the chart. Note that for \( \varepsilon < 0.15 \), a DDMA may be run at \( \text{Re} \) well above 1000 before the “slits” resolution starts limiting the response. However, for less diffusive particles the corresponding \( \text{Re}^* \) would be smaller, so that the maximum resolution would be fixed by this “slits” effect.

In Fig. 12 we show an example of the resolution of a DDMA compared to that of a Rosell’s DMA, but now taking into account the “slits” resolution. The vertical axis represents the ratio \( (\Delta y)^2_{p}/(\Delta y)^2_{E} \), where \( * \) stands for either diffusion limited resolution, E, or “slits” limited resolution, s; \( R \) stands for Rosell. The horizontal axis represents \( \log(\text{Re}) \).

*We will not consider the HP regime since the 25% gain in resolution makes the DDMA uninteresting in this case.
Fig. 11. Comparison between diffusive broadening and "slits" broadening. Both become equal at $Re = Re^*$, where $Re^*$ is given in equation (64). The plot shows the values of $\log(Re^*)$ (vertical axis) as a function of $\sigma$ (horizontal axis), taking $y_1$ and $\lambda$ as parameters. In this particular case we have taken $D/v = 0.168$, which would correspond to ions having one charge, $Z = 1 \text{ cm}^2/(\text{vs})^{-1}$, moving through air at room conditions.

Fig. 12. Comparison of the response of an optimum Rosell's DMA and a DDMA when the sheath flow is of the entrance type, but taking into account the "slits" effect. The vertical axis represents the ratio $(\Delta y/\gamma_p)^2/(\Delta y/\gamma_p)^2 R^2$, where * stands for either diffusion limited resolution, E, or "slits" limited resolution, s; R stands for Rosell. The horizontal axis represents $\log(Re)$, where Re is the Reynolds number. We have chosen $\lambda = 0.5$, $\sigma = 0.05$ and $D/v = 0.168$.

where Re is the Reynolds number. There are two groups of curves having $y_1 = 0.7$ and $y_1 = 0.3$, respectively. Each group consists of two curves: * = E, where diffusion limits the resolution, and * = s, where the "slits" effect does the same. For this example, we have chosen $\lambda = 0.5$, $\sigma = 0.05$ and $D/v = 0.168$ as before. The intersection of each pair of curves is marked with an arrow, which indicates the corresponding value of $\log(Re^*)$. One may see that for the less favorable case of large $y_1 = 0.7$, it is possible to double the resolution attained by Rosell's DMA, even if the DDMA is run at $Re = 1000 > Re^* (t^* = 5)$. For the case where $y_1 = 0.3$, one might easily triple Rosell's DMA resolution still operating at $Re < Re^* (t^* = 12.8)$. 
5.2. Aerosol flowrate

As mentioned at the beginning of this section, the former theory discussed in Section 3 may hold in the limit when \( q_a = q_s \) or \( q_a, q_s \ll Q \). Still, as pointed out by RL, it seems convenient to introduce an approximated correction to account for the effect of \( q_a \) (assumed \( q_s = q_a \)). It seems consistent to take

\[
\left( \frac{\Delta y}{r_p} \right)^2 \approx \left( \frac{K q_s}{Q} \right)^2,
\tag{79}
\]

where \( K \) being a constant of order one, as in RL. In order for this correction to be of minor relevance, one must simply operate at \( q_a/Q \) as small as needed, the limit probably being set by particle losses and detector sensitivity.

6. CONCLUSIONS

The theory of transport of the particles through a cylindrical DMA operated at \( \text{Pe} \gg 1 \) developed by RL has been used to theoretically study the effect that a constant axial electric field, superposed to the classical radial electric field present in the DMAs, has in the instrument resolution when dealing with high mobility particles. This new instrument has been termed drift-DMA (DDMA).

- For a fixed \( \text{Pe} \), the maximum resolution depends on \( u_I = u_d(r) \), that is, the type of flow the sheath gas experiences inside the analyzer.
- When the flow is of the plug type \( (u_I = U) \), the resolution of the DDMA increases with \( t \), the ratio between the axial field \( \beta \) and a characteristic radial field \( \gamma/R_2 \). In this case, the limiting resolution would be fixed by other factors (probably the ratio between aerosol flow rate \( q_a \) and sheath flow rate \( Q \)). For a fixed value of \( t > 1 \), the optimum analyzer length seems to be \( L_0 = 0 \).
- The other limiting situation considered is when \( u_I \) is of the Hagen–Poiseuille type. In this case, there exists an optimum value \( t = t^* \) that, for a fixed \( \text{Pe} \), minimizes the diffusive broadening. However, the gain in resolution, compared with a Rosell's DMA, would be at the most 25%, making the DDMA unattractive.
- Since the flow type strongly influences the response of the DDMA, we study a more realistic flow consisting of a flat velocity profile everywhere except at the solid electrodes, where a thin boundary layer exists \( (\text{Re} \gg 1) \). We call this flow type an entrance flow. As before, there exists a certain \( t = t^* \) that minimizes the diffusive broadening. For \( t = t^* \), the optimum analyzer length turns out to be \( L_0 = 0 \). \( t^* \) is found to depend on \( \text{Re} \), and on two geometric factors: \( y_1 = R_1/R_2 \) and \( \lambda = x_0/R_2 \), where \( x_0 \) is the axial location of the input/output slits with respect to the point where the boundary layer starts to develop. For particles having \( Z = 1 \text{ cm}^2 \text{ vs}^{-1} \), the resolution yielded by the DDMA can be easily made a factor of 3 better than that of a Rosell's DMA when both instruments operate at the same \( \text{Re} \). On the other hand, the DDMA can yield the same resolution as the Rosell's DMA but operating at a much smaller \( \text{Re} \) (near one order of magnitude), with the consequent saving on pumping expenses.
- We have considered the effect of the aerosol input/output slits on the resolution. The analysis, although qualitative, shows that, for plug flow, the limit on the resolution is being set by diffusion. However, as mentioned before, it is of no importance since this resolution can be made as good as desired by increasing \( t \). In the case of entrance flow, there exists a Reynolds number \( \text{Re} = \text{Re}^* \) which depends on geometric factors and on the type of particles at which the broadening due to diffusion equals that due to the finiteness of the slits. For \( \text{Re} < \text{Re}^* \), diffusion limits the resolution, and vice versa. When dealing with high mobility entities, however, by properly choosing the geometry of the DDMA (small values of \( y_1, \lambda \approx 1 \)), \( \text{Re}^* \) can be made of order 3000, so that the effect of finite slits would not be the limiting issue.
In brief, the analysis here described, although incomplete, indicates that this new approach increase to the resolution of existing differential mobility analyzers, when dealing with nanoparticles and ions, might be worth of some experimental effort.

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REFERENCES


