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MULTIGRADIENT DIELECTROPHORESIS: THEORETICAL ASPECTS

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I. INTRODUCTION

Dielectrophoresis is a long-established process used in various areas of modern technology for separation of particles according to dielectric properties. These include biological[1,2] and petrochemical[3] as well as mineral processing[4] applications; a comprehensive review of the diverse possibilities of use is given by Lin and Benguigui[5]. The particles may be in any state (solid, liquid, gaseous) while the liquid may be a liquid or a gas, separation being possible only when the components display different polarization properties. There are three main ways of segregating particulates: deflection[4], trapping[5] and levitation[6]. The process may consist in solid/solid[4,7], liquid/liquid[8], solid/liquid[5] or gas/liquid[9] separation, and in biological applications may even differentiate between living and dead cells[10]. The desired split is obtained by judicious exploitation of geometric (particle size and shape), physical (permittivity, conductivity, density) and operational (flow rate, field intensity and frequency, temperature) parameters. Two main dielectrophoretic techniques are currently in use: open-gradient[2,4] and multigradient[3,5]. The latter is a new development initiated by the Gulf Co.[3] whereby glass beads are used to produce high gradients responsible for trapping of particulates and is

obviously most advantageous, since it allows high throughputs at relatively low capital and operational costs. However, this breakthrough has not been followed by a successful commercialization due to three main pitfalls:

1) The Gulf device is a batch system (limited capacity).
2) The matrix consists of glass beads (cleaning problems, low permittivity).
3) The power supply is only d.c. (no flexibility in dealing with conductive liquids).

Put in other words, this system is unable to deal properly with suspensions over a few percent solids, ineffective in trapping particulates in slightly conductive liquids, and inadequate for S-S separation.

In this context, the research group at the Technion investigated aspects likely to yield satisfactory answers to the problem listed above, as well as additional ideas:

1) Sophisticated matrices with a view to better yields, e.g. $BaTiO_3$ beads (high permittivity) and mixtures of metallic rods or beads with glass beads (high conductivity).
2) Grid electrodes serving simultaneously as matrix.
3) A two-dimensional interdigitized electrode system for qualitative analysis of liquids.
4) Sinusoidal a.c. or pulsed d.c. fields, permitting processing of conductive liquids.
5) A continuous carrousel device combining very high capacity and relatively small dimensions.
6) Stable dielectric suspensions of submicronic particulates ($BaTiO_3$) providing a high-permittivity medium for S-S fractionation.

The present paper offers an overview of the various theoretical aspects of matrix dielectrophoresis, with special emphasis on rod-type matrices, which have proved to be better suited to industrial operation, being easier to clean and permitting higher loadings.

II. DIELECTROPHORETIC FORCE

The simplest and most attractive approach is the one put forward by Pohl[1], Jones[11] and more recently by Benguigui and Lin[12], whereby the dielectrophoretic force is obtained by summing the forces on two equal and opposite electric charges ($\pm q$) located at a vector distance $\vec{d}$ apart in a non-uniform electric field as shown in Fig. 1, the net force on this small dipole being:

$$\vec{F} = q\vec{E}_o(\vec{r} + \vec{d}) - q\vec{E}_o(\vec{r}) \tag{1}$$

Using a form of Taylor's series:

$$\vec{E}_o(\vec{r} + \vec{d}) - \vec{E}_o(\vec{r}) = (\vec{d}\cdot\vec{\nabla})\vec{E}_o + \text{higher-order terms}, \tag{2}$$

retaining only the first term since $\vec{d} \to 0$, and interpreting the product $q\cdot\vec{d}$ as an electric dipole moment $\vec{p}$ - we have:

$$\vec{F} = (\vec{p}\cdot\vec{\nabla})\vec{E}_o \tag{3}$$

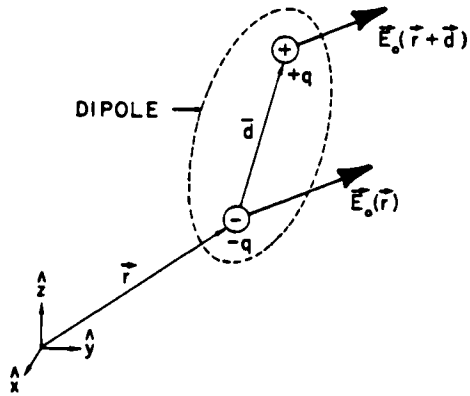

Figure 1 - Simple finite dipole model used to calculate dielectrophoretic force[11]

This equation is valid in practical applications, provided:

1) The particle is small compared to the scale of non-uniformities of the imposed electric field.
2) The macroscopic field remains unaffected, implying high dilution (large distances between individual particles).
3) Higher-order moments are negligible, which is necessarily true if $\vec{d} \to 0$.

The operator $(\vec{p}\cdot\vec{\nabla})$ can be put in a more familiar form using a vector transformation:

$$\vec{F} = (\vec{p}\cdot\vec{\nabla})\vec{E}_o = \vec{\nabla}(\vec{p}\cdot\vec{E}_o) - (\vec{E}_o\cdot\vec{\nabla})\vec{p} - \vec{p}\times(\vec{\nabla}\times\vec{E}_o) - \vec{E}_o\times(\vec{\nabla}\times\vec{p}) \quad (4)$$

Then, setting $\vec{p} = \alpha\vec{E}_o v$ where α is the polarizability tensor and v the particle volume, we obtain:

$$\vec{F} = \alpha v[\vec{\nabla}E_o^2 - (\vec{E}_o\cdot\vec{\nabla})\vec{E}_o - 2\vec{E}_o\times(\vec{\nabla}\times\vec{E}_o)] \quad (5)$$

where

$$(\vec{E}_o\cdot\vec{\nabla})\vec{E}_o = \vec{\nabla}E_o^2 - (\vec{E}_o\cdot\vec{\nabla})\vec{E}_o - 2\vec{E}_o\times(\vec{\nabla}\times\vec{E}_o)$$

thus

$$(\vec{E}_o\cdot\vec{\nabla})\vec{E}_o = \frac{\vec{\nabla}E_o^2}{2} - \vec{E}_o\times(\vec{\nabla}\times\vec{E}_o) \quad (6)$$

By Eqs. (5), (6), the final equation for the force is:

$$\vec{F} = \alpha v[\frac{\vec{\nabla}E_o^2}{2} - \vec{E}_o\times(\vec{\nabla}\times\vec{E}_o)] \quad (7)$$

The force $\vec{F}$ is thus seen to consist of two components:

$$\vec{F}_1 = \alpha v\frac{\vec{\nabla}E_o^2}{2} \quad (8)$$

$$\vec{F}_2 = -\alpha v\vec{E}_o\times(\vec{\nabla}\times\vec{E}_o) \quad (9)$$

where $\vec{F}_1$ is the dielectrophoretic force, while $\vec{F}_2$ is a levitation force which is a function of the associated magnetic field produced by the alternating electric field. $\vec{F}_2$ is a force not null in a.c. since $\vec{\nabla}\times\vec{E} = -\frac{\partial\vec{B}}{\partial t}$ and it can be shown to be proportional to a

term $\varepsilon_o\varepsilon_f\omega$ where ε_o is the absolute permittivity of free space, ε_f the fluid relative permittivity and ω the angular frequency. This term is zero for quasistatic fields and it becomes significant only for frequencies in the gigahertz range. However, dielectrophoresis in liquids is done at frequencies varying up to the megahertz level, thus $\vec{F}_2$ can be readily neglected.

III. DETERMINATION OF POLARIZATION FOR A SPHERICAL PARTICLE IN A FLUID (QUASISTATIC FIELDS)

Laplace's equation in the spherical case yields the following solution for the potential outside and inside the sphere[13] (see Fig. 2):

$$\phi_f = \left(\frac{A}{r^2} + Br\right)\cos\theta \tag{10a}$$

$$\phi_p = \left(\frac{C}{r^2} + Dr\right)\cos\theta \tag{10b}$$

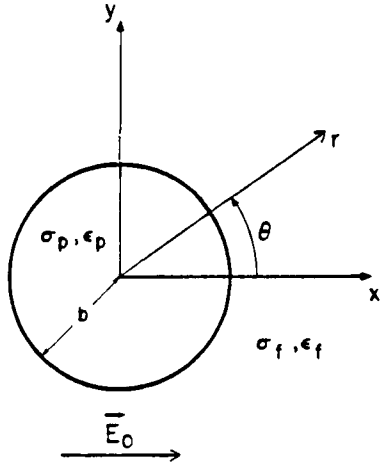

Figure 2 - Spherical particle in uniform applied field $\vec{E}_o$

the subscripts f, p denoting the fluid and particle respectively. With four coefficients to determine, four boundary conditions are needed. At the surface of the sphere (r = b, b being the sphere radius), we have:

$$\phi_f = \phi_p \Rightarrow E_{F\theta} = E_{p\theta} \tag{11}$$

$$\sigma_f E_{fr} - \sigma_p E_{pr} = - \frac{d\sigma_s}{dt} \tag{12a}$$

$$\sigma_s = \varepsilon_o(\varepsilon_f E_{fr} - \varepsilon_p E_{pr}) \tag{12b}$$

where σ_f and σ_p are respectively the fluid and particle conductivities, σ_s the superficial charge density at the fluid/particle interface, and ε_p the relative permittivity of the particle.

Equations (12a), (12b) express the continuity of the total current (conduction and displacement) and result in:

$$\sigma_f E_{fr} + \varepsilon_o \varepsilon_f \frac{dE_{fr}}{dt} = \sigma_p E_{pr} + \varepsilon_o \varepsilon_p \frac{dE_{pr}}{dt} \tag{12c}$$

This equation has been used by Pohl[2], Jones[11] as well as Benguigui and Lin[12] under various forms.

Two additional boundary conditions are that at large distances from the sphere the applied field remains undisturbed and at the center of the sphere the potential must stay finite:

$$\phi_{f\infty} = - E_o r \cos\theta \Rightarrow B = -E_o \tag{13a}$$

$$\phi_p \text{ finite} \Rightarrow C = 0 \tag{13b}$$

Using Eq. (13), the potential becomes:

$$\phi_f = \left(\frac{A}{r^2} - E_o r\right)\cos\theta \tag{14}$$

$$\phi_p = Dr \cos\theta \tag{15}$$

Combining Eqs. (11), (14) and (15), we have:

$$D = \frac{A}{b^3} = E_o \tag{16}$$

Using Eq. (16), we can write E_{fr} and E_{pr} for $r = b$ as:

$$E_{fr} = \left(E_o + 2\frac{A}{b^3}\right)\cos\theta \tag{17a}$$

$$E_{pr} = \left(E_o - \frac{A}{b^3}\right)\cos\theta \tag{17b}$$

Combining Equations (12c) and (17), we obtain:

$$(\sigma_p + 2\sigma_f)\frac{A}{b^3} + \frac{\varepsilon_o}{b^3}\frac{dA}{dt}(\varepsilon_p + 2\varepsilon_f) = E_o(\sigma_p - \sigma_f) + \varepsilon_o\frac{dE_o}{dt}(\varepsilon_p - \varepsilon_f) \tag{18}$$

Defining $\tau_p = \varepsilon_o\frac{\varepsilon_p + 2\varepsilon_f}{\sigma_p + 2\sigma_f}$ as the particle relaxation time, Eq. (18) becomes:

$$\frac{dA}{dt} + \frac{A}{\tau_p} = b^3\left(\frac{E_o}{\tau_p}\bar{\sigma}_p + \frac{dE_o}{dt}\bar{\varepsilon}_p\right) \tag{19}$$

where $\bar{\sigma}_p = \frac{\sigma_p - \sigma_f}{\sigma_p + 2\sigma_f}$ and $\bar{\varepsilon}_p = \frac{\varepsilon_p - \varepsilon_f}{\varepsilon_p + 2\varepsilon_f}$

With E_o suitably defined, Eq. (19) covers all situations — including the time-independent ones treated as a special subcase of the time-dependent case.

Investigating first the d.c. case, we have:

$$\begin{cases} t < 0 & E_o = 0 \\ t \geq 0 & E_o = \text{const} \end{cases} \tag{20}$$

which is the Heaviside unit-step function (14, p. 87). Thus, since for $t \geq 0$ E_o is time-independent Eq. (19) becomes:

$$\begin{cases} \frac{dA}{dt} + \frac{A}{\tau_p} = \frac{b^3 E_o}{\tau_p}\bar{\sigma}_p \\ t \geq 0 \end{cases} \tag{21}$$

This simple differential equation is readily solved (14, p.7) yielding:

$$A = b^3 E_o\,\bar{\sigma}_p + C\,e^{-t/\tau_p} \tag{22}$$

where C remains to be determined. For t = 0, the system behaves as if the conductivities were non-existent, thus A(0) should be a function of the permittivities alone:

$$A(0) = b^3 E_o \bar{\varepsilon}_p \tag{23}$$

which yields the final equation for A:

$$A = b^3 E_o [\bar{\sigma}_p (1-e^{-t/\tau_p}) + \bar{\varepsilon}_p e^{-t/\tau_p}] \tag{24}$$

In Eq. (24) t represents the time the particle has been exposed to the field.

This solution was first found by Jones[11]. It is readily seen that two cases are to be distinguished[15]:

$$t \ll \tau_p \quad , \quad A = b^3 E_o \bar{\varepsilon}_p \qquad \text{dielectric regime} \tag{25a}$$

$$t \gg \tau_p \quad , \quad A = b^3 E_o \bar{\sigma}_p \qquad \text{conductive regime} \tag{25b}$$

Now that A has been determined for d.c. fields, we proceed to find the dielectrophoretic force for the simple case of the dielectric regime. By Eqs. (14), (25a) we have:

$$\phi_f = E_o \left(\frac{b^3}{r^2} \bar{\varepsilon}_p - r \right) \cos\theta \tag{26a}$$

$$\phi_p = -3E_o \frac{\varepsilon_f r \cos\theta}{\varepsilon_p + 2\varepsilon_f} \tag{26b}$$

From Eq. (21b), the field inside the particle is found as:

$$E_p = 3 \frac{\varepsilon_f}{\varepsilon_p + 2\varepsilon_f} E_o \tag{27}$$

The polarization of the particle is, by definition:

$$P = \varepsilon_o (\varepsilon_p - \varepsilon_f) E_p = 3\varepsilon_o \varepsilon_f \bar{\varepsilon}_p E_o \tag{28}$$

Thus, the dipole moment is:

$$p = \frac{4}{3}\pi b^3 P = 4\pi b^3 \varepsilon_o \varepsilon_f \bar{\varepsilon}_p E_o \tag{29}$$

Consequently, the dielectrophoretic force is, using Eq. (8):

$$\vec{F}_{dc} = 2\pi b^3 \varepsilon_o \varepsilon_f \bar{\varepsilon}_p \vec{\nabla} E_o^2 \tag{30}$$

It is readily shown that for the conductive regime:

$$\vec{F}_{dc} = 2\pi b^3 \varepsilon_o \varepsilon_f \bar{\sigma}_p \vec{\nabla} E_o^2 \tag{31}$$

as in Jones[11]. For a mixed regime, with $t \simeq \tau_p$, we have:

$$\vec{F}_{dc} = 2\pi b^3 \varepsilon_o \varepsilon_f \beta_{dc} \vec{\nabla} E_o^2 \tag{32a}$$

where

$$\beta_{dc} = \bar{\sigma}_p (1 - e^{-t/\tau_p}) + \bar{\varepsilon}_p e^{-t/\tau_p} \tag{32b}$$

For the sinusoidal a.c. case a more suitable form of Eq. (12c) is obtained by using complex notation, thus we have:

$$\sigma_f^* E_{fr} = \sigma_p^* E_{pr} \tag{33a}$$

and

$$(\sigma_f + i\omega\varepsilon_o\varepsilon_f) E_{fr} = (\sigma_p + i\omega\varepsilon_o\varepsilon_p) E_{pr} \tag{33b}$$

or, expressing E_{fr} and E_{pr} explicitly with $E_o = E_m e^{i\omega t}$:

$$(\sigma_f + i\omega\varepsilon_o\varepsilon_f)(E_m e^{i\omega t} + \frac{2A}{b^3}) = (\sigma_p + i\omega\varepsilon_o\varepsilon_p)(E_m e^{i\omega t} - \frac{A}{b^3}) \tag{34}$$

Resorting to factorization, it is readily seen that the solution of this differential equation is:

$$A = \frac{\sigma_p - \sigma_f + i\omega(\varepsilon_p - \varepsilon_f)}{\sigma_p + 2\sigma_f + i\omega\varepsilon_o(\varepsilon_p + 2\varepsilon_f)} \; b^3 E_m e^{i\omega t} \tag{35}$$

Thus, the dipole moment is, as in Eq. (29):

$$p = 4\pi\varepsilon_o\varepsilon_f E_o b^3 \mathrm{Re}(A) \tag{36}$$

where Re stands for "real part of" and is determined by multi-

plying the numerator and denominator by the conjugate of the latter and expressing the field trigonometrically, the result being:†

$$\mathrm{Re}(A) = \frac{(\bar{\sigma}_p + \tau_p^2\omega^2\bar{\varepsilon}_p)\cos\omega t + \tau_p\omega\sin\omega t(\bar{\sigma}_p - \bar{\varepsilon}_p)}{1 + \tau_p^2\omega^2} b^3 E_m \tag{37}$$

An equivalent expression was obtained earlier by Benguigui and Lin[16]:

$$\mathrm{Re}(A) = [\bar{\varepsilon}_p\cos\omega t + 3\frac{(\varepsilon_f\sigma_p - \varepsilon_p\sigma_f)}{(\sigma_p + 2\sigma_f)^2} \cdot \frac{\omega\sin\omega t + (\cos\omega t)/\tau_p}{1 + \tau_p^2\omega^2}] b^3 E_m \tag{38}$$

Then the force being

$$\vec{F}_{ac} = (\vec{p}\cdot\vec{\nabla})\vec{E}_m\cos\omega t \tag{39a}$$

we obtain from Eqs.(36), (37) and (39a):

$$\vec{F}_{ac} = 2\pi\varepsilon_o\varepsilon_f b^3 \frac{(\bar{\sigma}_p + \tau_p^2\omega^2\bar{\varepsilon}_p)\cos^2\omega t + \tau_p\omega\frac{\sin 2\omega t}{2}(\bar{\sigma}_p - \bar{\varepsilon}_p)}{1 + \tau_p^2\omega^2} \vec{\nabla}E_m^2 \tag{39b}$$

Now, calling $\frac{\vec{F}_{ac}}{2\pi\varepsilon_o\varepsilon_f b^3\vec{\nabla}E_m^2}$ the <u>polarizability factor</u> and designating it by the letter β_{ac}, we can see that the three possible regimes of the d.c. case exist here as well:

$\tau_p\omega << 1$, $\beta_{ac} = \bar{\sigma}_p\cos^2\omega t$ conductive regime

$\tau_p\omega >> 1$, $\beta_{ac} = \bar{\varepsilon}_p\cos^2\omega t$ dielectric regime

$\tau_p\omega \approx 1$, $\beta_{ac} = \frac{(\bar{\sigma}_p + \tau_p^2\omega^2\bar{\varepsilon}_p)\cos^2\omega t + \tau_p\omega\frac{\sin 2\omega t}{2}(\bar{\sigma}_p - \bar{\varepsilon}_p)}{1 + \tau_p^2\omega^2}$ mixed regime

† In Appendix A a different approach is presented permitting inclusion of the transient effect and covering different field types. The case of a pulsed field is treated in Appendix B as an illustration.

In practical applications, only the average force is felt by the particle at sufficiently high frequencies, so that it is more useful in most cases:

$$\langle F_{ac} \rangle = \frac{1}{T} \int_0^T F_{ac}\, dt \tag{40}$$

The resulting average force is:

$$\langle F_{ac} \rangle = 2\pi\varepsilon_o\varepsilon_f b^3 \beta_{av} \vec{\nabla} E_{rms}^2 \tag{41}$$

where

$$\beta_{av} = \frac{\bar{\sigma}_p + \tau_p^2\omega^2\bar{\varepsilon}_p}{1 + \tau_p^2\omega^2}$$

The general expression for the force is:

$$\vec{F} = 2\pi\varepsilon_o\varepsilon_f b^3 \beta \vec{\nabla} E_o^2 \tag{42}$$

where

1) $\beta = \bar{\sigma}_p(1 - e^{-t/\tau_p}) + \bar{\varepsilon}_p e^{-t/\tau_p}$

in d.c. fields with $\vec{E}_o$ denoting the suddenly applied field.

2) $\beta = \dfrac{\bar{\sigma}_p + \tau_p^2\omega^2\bar{\varepsilon}_p}{1 + \tau_p^2\omega^2}$

for sinusoidal a.c. fields with $E_o = E_m/\sqrt{2}$ (rms value).

As the overall physical significance of all these equations is hard to visualize, it is worth trying to explain some of it at this stage. For a d.c. field and a dielectric regime, the meaning of the dielectrophoretic force with a polarization factor depending on the permittivities alone is obvious. The difficulty arises with conductive liquids, in which case (as can be predicted from the model, and was actually observed by Gherardi et al.[17]), the positive traction effect is eventually reversed owing to intervention

of an opposing force generated by current-induced polarization. For $\sigma_f >> \sigma_p$, charges arriving at the surface of the particle are not conducted through it as fast as through the fluid. Consequently, a net charge builds up on the particle surface, polarizing it along the lines of current flow with a polarity opposite to the dielectric one. This opposing force increases until it exceeds the positive one, causing negative dielectrophoresis. With a pulsed field it is readily understood that when the pulse time is shorter than τ_p, no charge accumulation can be expected and the dielectric regime can persist for a lengthy period of time (this obviously implies that in the dead intervals the charge is dissipated). In an alternating field, the net charge accumulated by the particle during the first half of the electric cycle is conducted back into the fluid during the second half. At low frequencies a half-cycle suffices for the current-induced polarization to overcome the dielectric one[4]. At sufficiently high frequencies this effect vanishes and experience has shown that at frequencies over a few thousand hertz the dielectric regime is reestablished — in accord with our theoretical model.

IV. DETERMINATION OF $\vec{\nabla}E_o^2$ AND ITS SUBSTITUTION INTO FORCE EQUATION

1) Bead-Type Matrix : Taking first the simplified case of a single bead, it can be easily deducted that the field distribution around the bead (Fig.3) is similar to the one for a spherical particle, thus:

$$\phi_f = E_o \cos\theta \left(\gamma \frac{a^3}{r^2} - r\right) \tag{43}$$

where a is the bead radius and γ the bead polarization factor, given as:

$$\gamma_{dc} = \bar{\sigma}_b^* \left(1 - e^{t_b/\tau_b}\right) + \bar{\varepsilon}_b e^{-t_b/\tau_b} \tag{44a}$$

$$\gamma_{ac} = \frac{\bar{\sigma}_b + \tau_b^2 \omega^2 \bar{\varepsilon}_b}{1 + \tau_b^2 \omega^2} \tag{44b}$$

where

$$\bar{\sigma}_b = \frac{\sigma_b - \sigma_f}{\sigma_b + 2\sigma_f} \quad , \qquad \bar{\varepsilon}_b = \frac{\varepsilon_b - \varepsilon_f}{\varepsilon_b + 2\varepsilon_f}$$

and

$$\tau_b = \varepsilon_o \frac{\varepsilon_b + 2\varepsilon_f}{\sigma_b + 2\sigma_f}$$

σ_b and ε_b represent the conductivity and relative permeability of the bead, t_b the time elapsed since the field was applied on the bead, and τ_b the relaxation time of the bead.

From Eq. (43) we have, in cylindrical coordinates:

$$\vec{E}_r = - \frac{\partial \phi_f}{\partial r} = \vec{E}_o \left(1 + 2 \frac{\gamma a^3}{r^3}\right) \cos\theta \tag{45a}$$

$$\vec{E}_\theta = - \frac{1}{r} \frac{\partial \phi_f}{\partial \theta} = \vec{E}_o \left(\frac{\gamma a^3}{r^3} - 1\right) \sin\theta \tag{45b}$$

Then, the resulting field is:

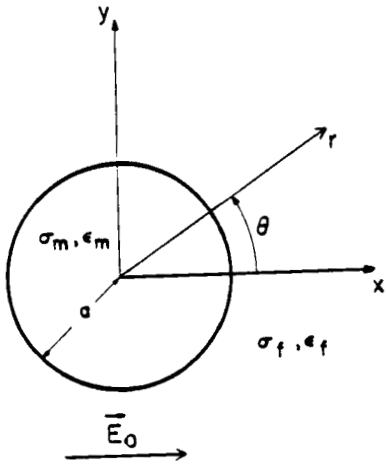

Figure 3 - Spherical bead or cylindrical rod in uniform applied field $\vec{E}_o$ (subscript m denotes matrix)

$$E = (E_r^2 + E_\theta^2)^{\frac{1}{2}} \tag{46}$$

And we can now determine the gradient of E^2 as:

$$\vec{\nabla}E^2 = \frac{\partial E^2}{\partial r}\hat{r} + \frac{1}{r}\frac{\partial E^2}{\partial \theta}\hat{\theta} \tag{47}$$

Combining Eqs.(45) and (47), we obtain the radial and azimuthal components:

$$(\vec{\nabla}E^2)_r = -3\gamma\frac{a^3}{r^4}E_o^2\left[\gamma\frac{a^3}{r^3}(3\cos 2\theta + 5) + 3\cos 2\theta + 1\right] \tag{48a}$$

$$(\vec{\nabla}E^2)_\theta = -3\gamma\frac{a^3}{r^4}E_o^2 \sin 2\theta \quad (\gamma\frac{a^3}{r^3} + 2) \tag{48b}$$

The force on a particle is obtained by combining Eqs. (42) and (48) using a dimensionless distance $r_a = r/a$:

$$F_r = K_b\left[\frac{\gamma}{r_a^3}(3\cos 2\theta + 5) + 3\cos 2\theta + 1\right] \tag{49a}$$

$$F_\theta = K_b\left(\frac{\gamma}{r_a^3} + 2\right)\sin 2\theta \tag{49b}$$

$$K_b = -\frac{6\pi\gamma\beta\epsilon_o\epsilon_f E_o^2 b^3}{a r_a^4} \tag{49c}$$

The force for a single bead does not tell much on what is really happening in the matrix where the packing density is around 0.65. Thus, a different approach is required. A direct one, allowing for the complexity of the bead matrix, does not appear possible. In an interesting model recently brought forward by Shapiro et al.[18,19], the space surrounding the collecting sphere is divided in two regions: a fluid envelope around the collector and an external matrix which has the filter porosity ϵ. The radius ℓ of the fluid envelope is simply

$$\ell = a(1-\epsilon)^{-1/3} \tag{50}$$

The potential ϕ can thus be calculated, using proper boundary conditions in the Laplace equation. First, however, it is required to determine ε_∞, the effective dielectric constant of the porous matrix. The latter may be calculated according to Hashin and Shtrikman[20]:

$$\varepsilon_\infty = \frac{\varepsilon_1+\varepsilon_2}{2} \tag{51a}$$

where

$$\varepsilon_1 = \varepsilon_f + \frac{1-\epsilon}{\frac{1}{\varepsilon_b-\varepsilon_f} + \frac{\epsilon}{3\varepsilon_f}} \; ; \; \varepsilon_2 = \varepsilon_b + \frac{\epsilon}{\frac{1}{\varepsilon_f-\varepsilon_b} + \frac{1-\epsilon}{3\varepsilon_b}} \tag{51b}$$

Now, the potential is expressed in the following form:

$$\ell < r < \infty \; , \qquad \phi_{f1} = E_\infty \cos\theta \; (K_1 \frac{\ell^3}{r^2} - r) \tag{52a}$$

$$a < r < \ell \; , \qquad \phi_{f2} = E_\infty \; K_2 \cos\theta \; (\gamma \frac{a^3}{r^2} - r) \tag{52b}$$

The values of K_1 and K_2 were calculated by Shapiro et al.[19] for the dielectric regime as functions of ε_b, ε_f and ϵ and are shown here after adaptation for any fluid (Shapiro et al dealt with aerosol separation):

$$K_1 = \frac{(\varepsilon_b+2\varepsilon_f)(\varepsilon_f-\varepsilon_\infty) - (1-\epsilon)(\varepsilon_f-\varepsilon_b)(\varepsilon_\infty+2\varepsilon_f)}{(\varepsilon_b+2\varepsilon_f)(\varepsilon_f+2\varepsilon_\infty) + 2(1-\epsilon)(\varepsilon_f-\varepsilon_b)(\varepsilon_\infty-\varepsilon_f)} \tag{53a}$$

$$K_2 = \frac{3\varepsilon_\infty(\varepsilon_b+2\varepsilon_f)}{(\varepsilon_b+2\varepsilon_f)(\varepsilon_f+2\varepsilon_\infty) + 2(1-\epsilon)(\varepsilon_f-\varepsilon_b)(\varepsilon_\infty-\varepsilon_f)} \tag{53b}$$

Then $\vec{\nabla}E^2$ can be determined in the same way as for the single bead, with a slightly different result. For $a < r < \ell$, E_o in the expression for K_b (Eq. 49c) is replaced by $E_\infty \; K_2$ while for $\ell < r < \infty$ we have $K_1/1-\epsilon$ instead of γ and E_∞ instead of E_o. The field E_∞ is defined as:

$$\vec{E}_\infty = \frac{\vec{E}_o}{\varepsilon_\infty}\varepsilon_f \tag{54}$$

It is readily seen that these equations for ϕ_f reduce to the ones derived for the single bead when $\epsilon \to 1$, as then $\ell \to \infty$, $\varepsilon_\infty \to \varepsilon_f$ and $E_\infty \to E_o$ resulting in $K_1 \to 0$ ($\phi_{fl} \to \phi_{f\infty}$) and $K_2 \to 1$. This approach, previously used for the flow field[21], seems very attractive and could be applied for various matrix types. It was also successfully used in magnetic separation with a ball matrix[22] to analyze the flow field.

2) Rod-Type Matrix: Here the potential around a single rod in a uniform applied field is given by (23, p. 128):

$$\phi_f = E_o\left(\frac{\gamma a^2}{r} - r\right)\cos\theta \tag{55}$$

where a is the rod radius and γ the rod polarization factor as in Eq. (44), but here we have:

$$\bar{\sigma}_r = \frac{\sigma_r - \sigma_f}{\sigma_r + \sigma_f} \qquad \bar{\varepsilon}_r = \frac{\varepsilon_r - \varepsilon_f}{\varepsilon_r + \varepsilon_f}$$

and

$$\tau_r = \varepsilon_o \frac{\varepsilon_r + \varepsilon_f}{\sigma_r + \sigma_f}$$

Here σ_r and ε_r are the conductivity and relative permittivity of the rod while τ_r is its relaxation time. Eq. (55) yields:

$$E_r = E_o\left(1 + \frac{\gamma a^2}{r^2}\right)\cos\theta \tag{56a}$$

$$E_\theta = E_o\left(\frac{\gamma a^2}{r^2} - 1\right)\sin\theta \tag{56b}$$

whence, as for the bead:

$$(\vec{\nabla}E^2)_r = -4\frac{\gamma a^2}{r^3}E_o^2\left(\frac{\gamma a^2}{r^2} + \cos 2\theta\right) \tag{57a}$$

$$(\vec{\nabla}E^2)_\theta = -4\frac{\gamma a^2}{r^3}E_o^2 \sin 2\theta \tag{57b}$$

The force is obtained by combining Eqs. (42) and (57), using the dimensionless distance $r_a = r/a$:

$$F_r = K_r\left(\frac{\gamma}{r_a^2} + \cos 2\theta\right) \tag{58a}$$

$$F_\theta = K_r \sin 2\theta \tag{58b}$$

where

$$K_r = -\frac{8\pi\gamma\beta\varepsilon_o\varepsilon_f E_o^2 b^3}{a r_a^3} \tag{58c}$$

3. Rod Matrix with Applied Field in Arbitrary Direction:

Taking E_o at an arbitrary angle as shown in Fig. 4, the potential is given by[24]:

$$\phi_f = E_o\left[\left(\frac{\gamma a^2}{r} - r\right)\cos\theta\cos\delta + \left(r - \frac{\gamma a^2}{r}\right)\sin\theta\sin\delta\right] \tag{59}$$

whence

$$E_r = E_o\left(1 + \frac{\gamma a^2}{r^2}\right)\cos(\theta+\delta) \tag{60a}$$

$$E_\theta = E_o\left(\frac{\gamma a^2}{r^2} - 1\right)\sin(\theta+\delta) \tag{60b}$$

Then:

$$(\vec{\nabla}E^2)_r = -4\gamma\frac{a^2}{r^3}E_o^2\left[\frac{\gamma a^2}{r^2} + \cos 2(\theta+\delta)\right] \tag{61a}$$

$$(\vec{\nabla}E^2)_\theta = -4\gamma\frac{a^2}{r^3}E_o^2\sin 2(\theta+\delta) \tag{61b}$$

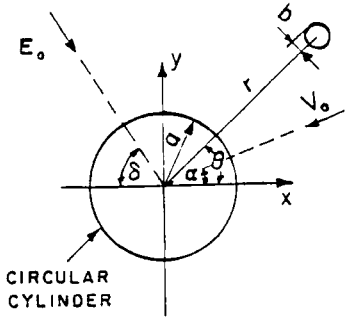

Figure 4 - Scheme of spherical particle/ cylindrical rod system

Thus, the force equation becomes:

$$F_r = K_r \left[\frac{\gamma}{r_a^2} + \cos 2(\theta+\delta) \right] \tag{62a}$$

$$F_\theta = K_r \sin 2(\theta+\delta) \tag{62b}$$

Unlike the bead, the above calculation for a single rod is applicable for an array of rods with a **low** packing density since then rods are widely spaced.

V. PARAMETRIC DISCUSSION

1) Common Features of Bead and Rod Matrices

In both cases the force is seen to be directly proportional to b^3, E_o^2, ε_f, γ and β and inversely proportional to a[25,26]. Thus better yields are expected for strong fields, large particles and a fine matrix, γ and β exert more influence through their sign than through their absolute value, the variation interval of γ being $(-\frac{1}{2}, +1)$ for the bead and $(-1,+1)$ for the rod, while that of β is $(-\frac{1}{2}, +1)$.

β is critical in determining the capture ability of the filter in so far as the particle electrical properties are concerned. For d.c. fields some interesting cases have been described[25], for instance:

(a) In most practical applications $\varepsilon_p > \varepsilon_f$ while $\sigma_p < \sigma_f$; under such conditions β is positive for $t = 0$, becomes zero at $t_o = \tau_p \ell n \frac{\bar{\sigma}_p - \bar{\varepsilon}_p}{\bar{\sigma}_p}$ and is negative for larger values of t, implying "negative" dielectrophoresis or in other words no trapping.

(b) For metallic particles $\tau_p \to 0$, thus $\beta_{dc} = 1$; in this case β_{dc} is seen to be independent of the liquid permittivity and conductivity, and always positive.

(c) For perfectly insulating particles $\sigma_p << \sigma_f$ and $t >> \tau_p$ we have $\beta_{dc} \to -\frac{1}{2}$; this case can be seen as the opposite of the preceding, now β_{dc} is again independent of the liquid properties and always negative.

In a.c. fields the critical time t_o has its equivalent in the form of a critical frequency $\omega_o = \left[\frac{\sigma_p - \sigma_f}{\tau_p(\varepsilon_p - \varepsilon_f)}\right]^{\frac{1}{2}}$. Here as ω increases for the case $\varepsilon_p > \varepsilon_f$ and $\sigma_p < \sigma_f$, β passes from a negative value through zero at ω_o to a positive one for $\omega > \omega_o$. This means that the conductive regime is ruling at low frequencies and the dielectric one is obtainable by increasing the frequency beyond ω_o.

2) Differences between Bead and Rod Matrices

In a bead matrix with $\gamma > 0$, there is attraction at the contact points between the beads for $\beta > 0$[26]; in a widely-spaced rod matrix there are regions of attraction and repulsion aroung the rod as seen in Fig. 5. Thus at least in theory simultaneous positive and

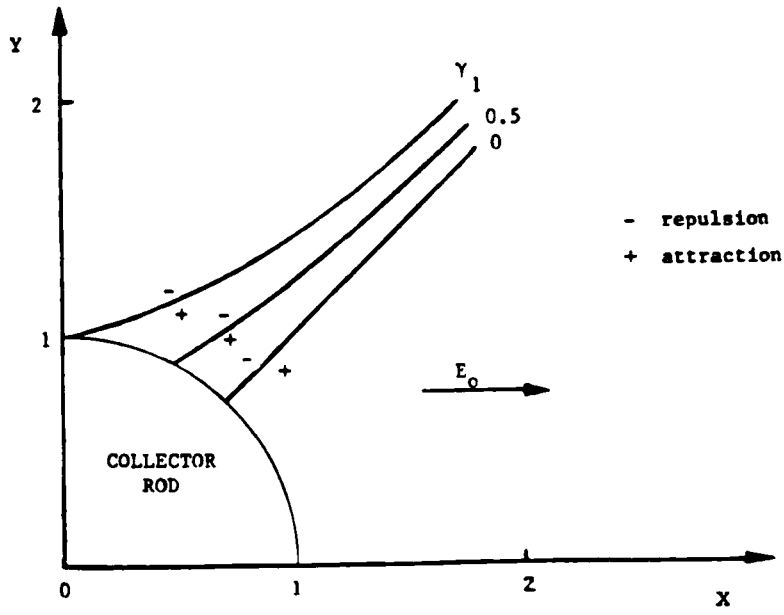

Figure 5 - Mapping of attraction and repulsion zones in first quadrant for different values of under static conditions and $\beta > 0$[32].

negative dielectrophoresis is feasible in a rod matrix but not in a bead matrix. In fact, negative dielectrophoresis is very hard to obtain since the force is weak. The efficiency of a bead matrix can be greatly improved by replacing part of the glass beads by metallic or ferroelectric ones, the cause of the improvement being not a sizable change of γ but a jump in a conductive liquid from a negative value of γ to a positive one (the metallic beads are efficient in the conductive regime; the ferroelectric one in the dielectric regime). Moreover, a matrix of metallic rods can be so designed as to act as electrodes of alternate polarity; with the spacing D kept sufficiently small ($D/2a < 5$), the whole energized space displays a highly non-uniform field, yielding good separation characteristics at lower voltages[27].

VI. EQUATIONS OF MOTION FOR ROD MATRIX

Here only the case of a single rod will be examined. Generalizing Newton's second law of motion, we have:

$$(m_p + km_f) \frac{d\vec{v}_p}{dt} = \vec{F}_G + \vec{F}_D + \vec{F}_E \tag{63}$$

where (m_p+km_f) is the virtual mass of the particle, m_p being its actual mass and m_f the fluid mass displaced by it; k is a coefficient depending on its geometry and on the nature of its motion[28] (a conservative estimate for k is 0.5 for a spherical particle). v_p is the particle velocity. $\vec{F}_G$, the gravity force, is given by:

$$\vec{F}_G = -\frac{4}{3}\pi b^3 g\,(\rho_p-\rho_f)(\sin\theta\vec{i}_r + \cos\theta\vec{i}_\theta) \tag{64}$$

where $\vec{i}_r$ and $\vec{i}_\theta$ are unit vectors in the radial and azimuthal directions respectively, g is the gravity acceleration; ρ_p and ρ_f the particle and fluid densities. $\vec{F}_D$ is the drag force. A Stokesian drag law is applicable for particle Reynolds numbers much smaller than 1, i.e. $Re_p = 2V_o b\rho_f/\eta \ll 1$ (V_o is the superficial velocity in the separator). Thus we have:

$$\vec{F}_D = 6\pi\eta(\vec{v}_f - \vec{v}_p)b \tag{65}$$

where η is the fluid viscosity. The flow regime around the rod is governed by the rod Reynolds number $Re_r = 2V_o a\rho_f/\eta$. Two cases have to be distinguished as reported by Gerber and Birss[29]:

a) $Re_r >> 1$ potential flow

b) $Re_r << 1$ creeping flow

These two cases fall under laminar flow. In the first case, the flow potential is given as (referring to Fig. 4):

$$\phi = V_o(r + \frac{a^2}{r})\cos(\theta-\alpha) \tag{66}$$

then

$$\vec{v}_f = -\vec{\nabla}\phi = -V_o[(1 - \frac{a^2}{r^2})\cos(\theta-\alpha)\vec{i}_R - (1 + \frac{a^2}{r^2})\sin(\theta-\alpha)\vec{i}_\theta] \tag{67}$$

The creeping flow equations were derived by Zebel[30] from the theoretical work of Lamb[31]:

$$\vec{v}_f = -V_o C_L[(2\ell n\frac{r}{a} - 1 + \frac{a^2}{r^2})\cos(\theta-\alpha)\vec{i}_R - \left(2\ell n\frac{r}{a} + 1 - \frac{a^2}{r^2}\right)\sin(\theta-\alpha)\vec{i}_\theta] \tag{68a}$$

$$C_L = 1/\ell n\,(7.4/Re_r)^2 \tag{68b}$$

The equations of motion for a particle in potential flow are given below, similar ones can be written for creeping flow. For a more convenient treatment we introduce the dimensionless variables[32]:

$$t_a = V_o t/a \quad , \qquad r_a = r/a$$

and the dimensionless groups:

$$B = 6E_o^2\varepsilon_o\varepsilon_f\gamma\beta/V_o^2(\rho_p+\rho_f/2) \quad \text{dielectrophoretic/inertial} \tag{69a}$$

$$G = (\rho_p-\rho_f)ag/V_o^2(\rho_p+\rho_f/2) \quad \text{gravitational/inertial} \tag{69b}$$

$$S = 2V_o(\rho_p+\rho_f/2)b^2/9\eta a \quad \text{inertial/viscous} \tag{69c}$$

where G and S represent respectively variants of the Froude and Stokes numbers.

The value of E_o is function of the field type. For d.c., E_o = const.; for sinusoidal a.c. fields two cases must be distinguished:

1) Low frequencies, for which $E_o = E_m \cos(\omega t+\phi)$, where ϕ is the phase associated with the instantaneous value of E_o at the moment of entry of the particle into the separation cell (t=0), so $\phi = \cos^{-1}[E_o(t=0)/E_m]$. Obviously, when dealing with a suspension, ϕ takes on a different value for each particle; thus at low frequencies the trajectory of a particle does not depend only on ω but also on ϕ[33,34].

2) High frequencies, for which $E_o = E_m/\sqrt{2}$; in this case the particle "feels" an average force with only the rms value of the field to be taken into account.

For pulsed fields the situation is very similar to sinusoidal fields; the same two cases must be distinguished, but here $E_o = E_m \sum_{n=0}^{\infty} a_n \cos(n\omega t+\phi)$ at low frequencies while at high frequencies E_o is found from the applicable rms value.

Thus in the general case, the following set of second-order differential equations is obtained:

$$\frac{d^2 r_a}{dt_a^2} - r_a \left(\frac{d\theta}{dt_a}\right)^2 = -\frac{\beta}{r_a^3}\left[\frac{\gamma}{r_a^2} + \cos 2(\theta+\delta)\right] - G\sin\theta - \frac{1}{S}\left[\left(1 - \frac{1}{r_a^2}\right)\cos(\theta-\alpha) + \frac{dr_a}{dt_a}\right] \tag{70a}$$

$$r_a \frac{d^2\theta}{dt_a^2} + 2\left(\frac{dr_a}{dt_a}\right)\left(\frac{d\theta}{dt_a}\right) = -\frac{\beta}{r_a^3}\sin 2(\theta+\delta) - G\cos\theta + \frac{1}{S}\left[\left(1 + \frac{1}{r_a^2}\right)\sin(\theta-\alpha) - r_a\frac{d\theta}{dt_a}\right] \tag{70b}$$

More details are provided in Ref.31. For small Stokes and Froude numbers the inertial and gravity terms may be neglected and Eq. (70)

simplifies to a set of first-order differential equations:

$$\frac{dr_a}{dt_a} = -\frac{BS}{r_a^3}\left[\frac{\gamma}{r_a^2} + \cos 2(\theta+\delta)\right] - \left(1 - \frac{1}{r_a^2}\right)\cos(\theta-\alpha) \qquad (71a)$$

$$r_a \frac{d\theta}{dt_a} = -\frac{BS}{r_a^3}\sin 2(\theta+\delta) + \left(1 + \frac{1}{r_a^2}\right)\sin(\theta-\alpha) \qquad (71b)$$

where

$$BS = \frac{4b^2 E_o^2 \varepsilon_o \varepsilon_f \gamma \beta}{3\eta a V_o} \qquad \text{dielectrophoretic/viscous} \qquad (71c)$$

Equations (71) have no analytical solution except for $\gamma = 0$[29,30]; this solution has been studied in detail for viscous flow by Shalom and Lin[35]. The general case requires numerical analysis, and examples of trajectories in d.c. fields were given for potential flow[32] and viscous flow[35] in recent publications. Low-frequency effects on the trajectories have been also investigated for sinusoidal fields[33,34].

Determination of the trajectories is useful in investigating the mechanism of particle trapping but is also helpful in finding the filter efficiency and we shall see how. In tracing the particle trajectories for particles at various positions, a critical trajectory can be identified (Fig. 6) corresponding to a capture radius,

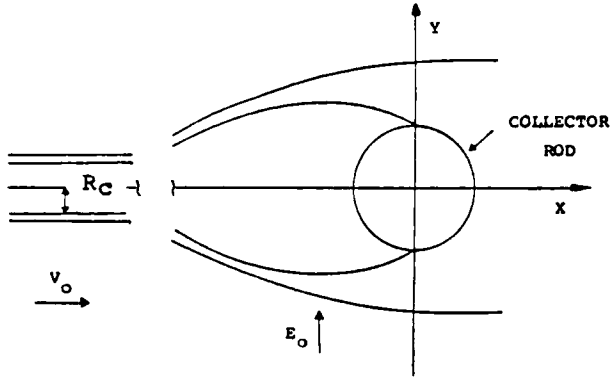

Figure 6 - Schematic explanation of critical trajectory[32].

R_c, equal to the coordinate of the particle at the point of departure. Then R_c can be plotted against the dimensionless group BS (see Eq.(71c)) which is most characteristic of the separation process[32,35]. Then a capture area per unit length can be defined as $2R_c a$ (Fig. 7) which is in fact a measure of the probability of capture in a single rod-particle interaction. This probability can then be easily incorporated in a simple statistical expression for the extraction efficiency of the particles, E_f, per filter length and the volume packing of the rods $1 - \epsilon$. Zebel[30] obtained this efficiency as:

$$E_f = 1 - (1 - R_c \frac{2a}{D})^m \tag{72}$$

where m is the number of layers of rods. In terms of the volume packing of the rods, Eq. (72) becomes:

$$E_f = 1 - \left[1 - 2R_c \left(\frac{1-\epsilon}{\pi}\right)^{\frac{1}{2}}\right]^m \tag{73}$$

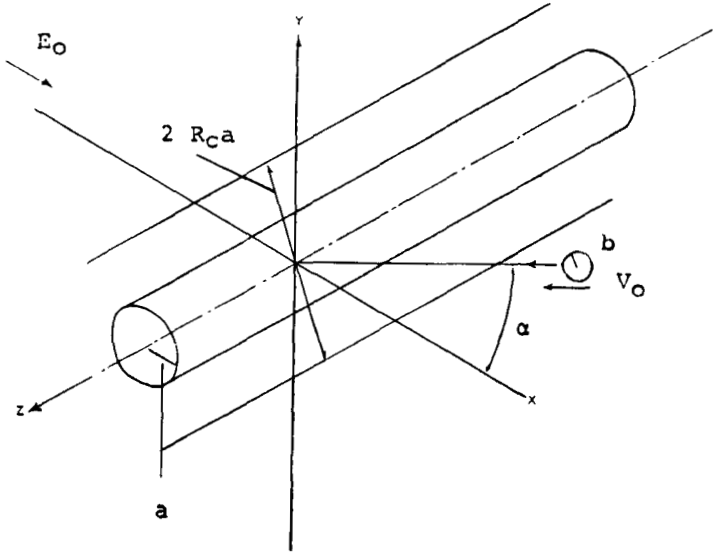

Figure 7 - Capture area per unit length of rod

Other expressions are found in literature[19,37,38] with an exponential term, for instance[38]:

$$E_f = 1 - \exp\left[-4(1-\epsilon)R_c L/3\pi a\right] \quad (74)$$

where L is filter length.

Anyhow, it should be borne in mind that Eqs.(73), (74) are approximately correct only for a separation matrix in its early stages of separation. The build-up of particles on the rod surface distorts both the electric and the flow fields in its vicinity gradually decreasing the capture radius down to zero for which saturation is obtained. A more sophisticated model dealing with this equilibrium situation is necessary to take loading factors into consideration and this is the object of future work.

VII. CONCLUSION

Various theoretical aspects of matrix dielectrophoresis have been reviewed in an attempt at describing the complex situation created by the large number of parameters: physical, operating and geometric. Perhaps more questions have been raised than answers provided throughout this work clearly indicating that multigradient dielectrophoresis is still in its infancy.

Specialists familiar with high gradient magnetic separation can easily note the close similarity existing between the magnetic and dielectric traction forces (for the dielectric regime) with H replaced by E and μ by ε. Obviously, what makes dielectric separation much more intricate is the existence of conductivity parameters, transient effects and the fact that it is possible to use a.c. and pulsed electric fields. As for the influence of parameters common to both processes (e.g. V_o, η, a, b, γ, β) it may be safely stated that the same effects are to be expected in both areas thus the experience gathered in magnetic separation can be applied in the dielectric area.

VIII. ACKNOWLEDGMENT

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APPENDIX A: DETERMINATION OF POLARIZABILITY FOR A SUDDENLY APPLIED SINUSOIDAL FIELD (TRANSIENT EFFECT)

Defining $E_o = E_m \cos\omega t$ and using Eq. (19), we have:

$$\frac{dA}{dt} + \frac{A}{\tau_p} = b^3 E_m\left(\frac{\bar{\sigma}_p}{\tau_p}\cos\omega t - \omega\bar{\varepsilon}_p\sin\omega t\right) \tag{A1}$$

Solving this differential equation, we obtain:

$$A = \frac{b^3 E_m}{e^{t/\tau_p}}\int e^{t/\tau_p}\left(\frac{\bar{\sigma}_p}{\tau_p}\cos\omega t - \bar{\varepsilon}_p\omega\sin\omega t\right)dt + c\,e^{-t/\tau_p} \tag{A2}$$

and

$$A = b^3 E_m \frac{(\bar{\sigma}_p + \tau_p^2\omega^2\bar{\varepsilon}_p)\cos\omega t + \tau_p\omega\sin\omega t\ (\bar{\sigma}_p - \bar{\varepsilon}_p)}{1 + \tau^2\omega^2} + c\,e^{-t/\tau_p} \tag{A3}$$

As for the d.c. case we have:

$$A(0) = b^3 E_m \bar{\varepsilon}_p$$

Thus:

$$A = \frac{b^3 E_m}{1+\tau^2\omega^2}\left[(\bar{\sigma}_p + \tau_p^2\omega^2\bar{\varepsilon}_p)\cos\omega t + (\bar{\sigma}_p - \bar{\varepsilon}_p)(\tau_p\omega\sin\omega t - e^{-t/\tau_p})\right] \tag{A4}$$

For $t \gg \tau_p$ Eq. (A4) is seen to be identical to Eq. (37). An equivalent expression was obtained earlier by Benguigui and Lin[16]:

$$A = b^3 E_m\left[\bar{\varepsilon}_p\cos\omega t + 3\,\frac{\varepsilon_f\sigma_p - \varepsilon_p\sigma_f}{(\sigma_p+2\sigma_f)^2}\cdot\frac{\omega\sin\omega t + \frac{\cos\omega t_p}{\tau_p} - \frac{e^{-t/\tau_p}}{\tau_p}}{1 + \tau_p^2\omega^2}\right] \tag{A5}$$

APPENDIX B: DETERMINATION OF POLARIZABILITY FOR PULSED FIELD INCLUDING TRANSIENT EFFECT

This type of field is especially difficult to deal with in view of the need for a suitable function describing its time pattern. As an example, let us examine the step function shown in Fig. 8. This simple case is readily treated with the aid of a Fourier series (35, p.142):

$$E_o = E_m\left[\frac{1}{2} + \frac{2}{\pi}\sum_{n=1}^{\infty}\frac{(-1)^{n+1}\cos k\omega t}{k}\right] \tag{B1}$$

where $k = 2n-1$.

Using Eq. (19), we obtain

$$\frac{dA}{dt} + \frac{A}{\tau_p} = b^3 E_m \left\{ \frac{\bar{\sigma}_p}{\tau_p} \left[\frac{1}{2} + \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{k} \cos k\omega t \right] + \frac{2\bar{\varepsilon}_p \omega}{\pi} \sum_{n=1}^{\infty} (-1)^n \sin k\omega t \right\} \tag{B2}$$

Then, as in Appendix A:

$$A = \frac{b^3 E_m}{e^{t/\tau_p}} \int e^{t/\tau_p} \left\{ \frac{\bar{\sigma}_p}{\tau_p} \left[\frac{1}{2} + \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{k} \cos k\omega t \right] + \right.$$

$$\left. + \frac{2\bar{\varepsilon}_p \omega}{\pi} \sum_{n=1}^{\infty} (-1)^n \sin k\omega t \right\} dt + c\, e^{-t/\tau_p} \tag{B3}$$

Whence

$$A = b^3 E_m \left\{ \frac{\bar{\sigma}_p}{2} + \frac{2}{\pi} \left[\bar{\sigma}_p \sum_{n=1}^{\infty} (-1)^{n+1} \frac{\cos k\omega t + k\tau_p \omega \sin k\omega t}{k(1+k^2 \tau_p^2 \omega^2)} + \right. \right.$$

$$\left. \left. + \tau_p \omega \bar{\varepsilon}_p \sum_{n=1}^{\infty} (-1)^n \frac{\sin k\omega t - k\tau_p \omega \cos k\omega t}{1 + k^2 \tau_p^2 \omega^2} \right] \right\} + c\, e^{-t/\tau_p} \tag{B4}$$

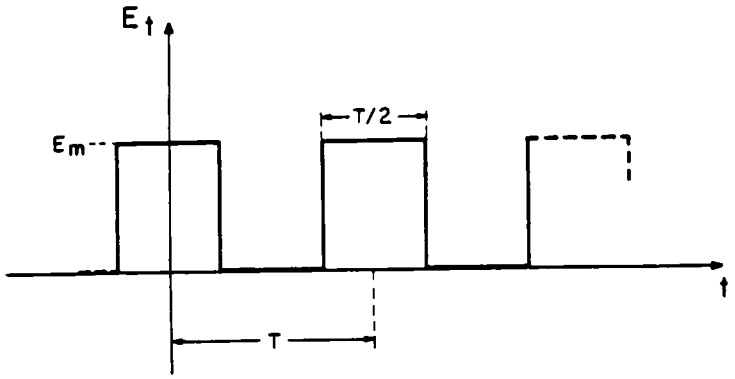

Figure 8 - Example of pulsed field

or

$$A = b^3 E_m \left\{ \frac{\bar{\sigma}_p}{2} + \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{(-1)^n (\bar{\varepsilon}_p - \bar{\sigma}_p) k\tau_p \omega \sin k\omega t + (-1)^{n+1} \cos k\omega t (\bar{\varepsilon}_p k^2 \tau_p^2 \omega^2 + \bar{\sigma}_p)}{k(1 + k^2 \tau_p^2 \omega^2)} \right\} + c\, e^{-t/\tau_p} \tag{B5}$$

Using the same initial condition as for the other cases, we have:

$$c = b^3 E_m \left[\bar{\varepsilon}_p - \frac{\bar{\sigma}_p}{2} - \frac{2}{\pi} \sum_{n=1}^{\infty} (-1)^{n+1} \frac{\bar{\varepsilon}_p k^2 \tau_p^2 \omega^2 + \bar{\sigma}_p}{k(1+k^2 \tau_p^2 \omega^2)} \right] \tag{B6}$$

As in the sinusoidal case, a much simpler expression is obtained for the conductive and dielectric regimes for $t >> \tau_p$:

1) $\tau_p \omega << 1$ (conductive regime):

$$A = b^3 E_m \bar{\sigma}_p \left[\frac{1}{2} + \frac{2}{\pi} \sum_{n=1}^{\infty} (-1)^{n+1} \frac{\cos k\omega t}{k} \right] \tag{B7}$$

Comparing with Eq. (B1), we see that:

$$A = b^3 E_o \bar{\sigma}_p \tag{B8}$$

2) $\tau_p \omega >> 1$ (dielectric regime):

$$A = b^3 E_m \left[\frac{\bar{\sigma}_p}{2} + \frac{2\bar{\varepsilon}_p}{\pi} \sum_{n=1}^{\infty} (-1)^{n+1} \frac{\cos k\omega t}{k} \right] \tag{B9}$$

or

$$A = b^3 E_o \bar{\varepsilon}_p + \frac{b^3 E_m}{2} (\bar{\sigma}_p - \bar{\varepsilon}_p) \tag{B10}$$

Whence

$$A = b^3 E_o \bar{\varepsilon}_p + \text{const.} \tag{B11}$$

Eq. (B7) is analogous to its counterpart of a sinusoidal field, but Eq. (B11) differs by the additive constant.