UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

ENERCORP SAND SOLUTIONS INC.
Petitioner,

v.

SPECIALIZED DESANDERS INC.,
Patent Owner

Patent No. 8,945,256

DECLARATION OF ANDRE L. DAVISON

Pursuant to 28 U.S.C. § 1746, I, Andre L. Davison, hereby declare and state as follows:

1. I am a Research Technology Manager in the Library and Research Services department of Blank Rome LLP. I hold a Master of Science in Library and Information Science from the University of North Texas.

2. One of the duties I perform in my position is locating and procuring physical copies of books, articles, and similar resources in libraries and other repositories.

ENERCORP EXHIBIT 1009

3. I was asked to locate a physical copy of the article "Sedimentation of Noncolloidal Particles at Low Reynolds Numbers," by Robert H. Davis and Andreas Acrivas, published in the Annual Review of Fluid Mechanics, Vol. 17, 1985 (hereafter, the "Davis article"). Therefore, I performed a search for the Davis article using the website of OCLC's WorldCat (www.worldcat.org). WorldCat is a well-known resource for locating libraries holding specific materials in their collections. The WorldCat website indicated that the Davis article could be found, among other libraries, at Rice University's Fondren Library in Houston, Texas. Therefore, I contacted Fondren Library and asked for a copy of the Davis article.

4. Exhibit 1005 to Enercorp Sand Solutions, Inc.'s Petition for *Inter Partes* Review of U.S. Patent No. 8,945,256 is a true and correct copy of the Davis article, as received from Ms. Elise McCutchen, a Senior Library Associate at Fondren Library.

5. I was also asked to determine if the Davis article had been publically cited by others since its publication. The Davis article's publisher, Annual Reviews Inc., maintains a website where copies of its publications may be purchased (www.annualreviews.org). The Annual Reviews website also uses the "Web of Science" citation network, which one may use to determine what works have cited a particular work. According to this functionality on the Annual Reviews website, the Davis article has been cited 250 times as of September 6, 2018. A true and

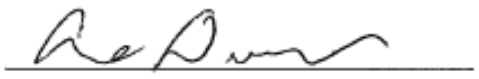
correct copy of the list of those citations, printed out from the Annual Review website, is attached hereto as Exhibit A.

6. The first citation by someone who was not one of the authors of the Davis article is "Magnetocentrifugal separation of particulates: Theory and principles of operation," by Yoram Zimmels, published in the Journal of Applied Physics in 1986 (the "Zimmels article"). To verify that this article indeed cited to the Davis article, I reviewed a copy of that article purchased from the Journal of Applied Physics. A true and correct copy of Zimmels article that I reviewed is attached hereto as Exhibit B.

7. The Zimmels article cites the Davis article at footnote 30. Footnote 30 states: "[30]R.E. Davis, Ann. Rev. Fluid Mech. 17, 91 (1985)." This appears to correctly reference the author, publication and volume number, first page, and publication date of the Davis Article.

I declare under penalty of perjury under the laws of the United States of America that the foregoing is true and correct.

Executed on the 14th day of September 2018 in Houston, Texas.



Andre L. Davison

EXHIBIT A

Sedimentation of Noncolloidal Particles at Low Reynolds Numbers

Annual Review of Fluid Mechanics

Vol. 17:91-118 (Volume publication date January 1985)
https://doi.org/10.1146/annurev.fl.17.010185.000515
(https://doi.org/10.1146/annurev.fl.17.010185.000515)

R H Davis, and A Acrivos

CITATION NETWORK

CITATION MANAGEMENT TOOLS

CITING ARTICLES

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EXHIBIT B

Magnetocentrifugal separation of particulates: Theory and principles of operation

Yoram Zimmels

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Magnetocentrifugal separation of particulates: Theory and principles of operation

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The theory and principles of operation of the new magnetocentrifugal separation are formulated. Magnetic and centrifugal g numbers for monodisperse and polydisperse particulates are defined. It is shown that the magnitude of electromagnetic and centrifugal forces can be matched in the full scale of centrifugal fields available to date. Magnetocentrifugal separation can be applied to separation of particulates down to colloidal and molecular sizes, and using a stationary fluid, it is independent of particle size, being controlled by density and magnetization of particles and fluid. Magnetic field configurations, different cases related to magnetization and density distributions of particulates, and specific modes of operations are discussed. Finally, diffusion controlled systems and potential applications of the new technology are considered.

I. INTRODUCTION

Centrifugal separation is used in many industrial and laboratory operations that involve processing of particles in a fluid.[1–4] This technique provides enhanced sedimentation velocities of particles relative to a fluid in which they are dispersed on one hand, and to enhanced flow of fluid through a bed formed from such particles[1–4] on the other hand. Enhanced sedimentation and flow velocities are particularly desirable in systems that involve dispersions of fine particles in a fluid. Fine particles are slow in moving through fluids and, once deposited on a supporting surface, are expected to form high resistance beds to the flow of fluids through them.[1–3] Here high driving body forces which are called for can be established by high acceleration fields. A wide spectrum of such fields is available today, i.e., from the earth gravitational acceleration (g) up to hundreds of thousands of g's.[4,5]

For example, standard commercial centrifuges provide accelerations equivalent to thousands and ten thousands of g's,[3–5] and standard commercial ultracentrifuges extend this range up to 350 000 g's.[5] For a given temperature and pressure, the required levels of acceleration field depend on the size of the dispersed particles, density difference between dispersed and continuous phases, concentration and distribution properties of the dispersed phase, and on viscosity of the fluid. Particle size is the most important parameter when considering sedimentation velocities and resistance of fixed beds to flow. Thus, for particles that are in the 1–1000-μm range, accelerations of one to thousands of g's are generally sufficient.

For smaller particles such as colloids, viruses, macromolecules, and molecules, in the 1–100-nm range accelerations of hundreds of thousands of g's are required to affect sedimentation in a reasonable (hours and days) period of time. Centrifugal separation is used for solid-liquid, liquid-liquid, and solid-solid separation.[1–5] Applications of this technique can be found in the chemical and mineral processing industries,[3–6] and in biological, pharmaceutical, and medically related processing testing and analysis procedures. To date centrifugal forces are the only body forces which can be generated at levels high enough to be effective in separation of ultrafine (colloid and molecular size) particles. However, centrifugal separation is expensive, and the cost per unit weight of the particles treated increases as their size decreases. Furthermore, single bowl centrifuges, or those that have a setting of tubes mounted within the bowl, are seldom designed (the density gradient method used in biochemistry being an exception) to yield simultaneously multifraction concentrates. Here costly multipath (possibly incorporating closed loops) processes may be required in order to produce the desirable concentrates. To date no adequate force field which can match, and hence also counteract, high centrifugal forces is commercially available. This means that whatever separation is carried out using high centrifugal fields, the outcome depends only on the response of the system being separated to this field. This sets limitations to separation efficiencies and maneuverability of the system. Availability of a matching force field is expected to extend significantly the scope of application of the present centrifugal separation technology.

Electromagnetic fields are currently used to separate particles from a fluid stream, by positive attraction towards deposition sites which are part of highly permeable matrices.[7–10] These matrices are energized by high external fields, and in turn generate high gradients in their vicinity.

The combined high-intensity, high-gradient field facilitates generation of relatively strong electromagnetic forces. Application of these forces to magnetizable particles, which results in deflection of their trajectories from the fluid stream, may subsequently lead to separation. Electromagnetic fields can also be used to counteract acceleration fields, e.g., gravity, by stresses induced in fluids which are sufficiently permeable to magnetic or electric fields. A related technique is known by the name of magnetohydrostatic separation.[11,12] Here pressure gradients are generated in a magnetizable liquid, by a nonhomogeneous magnetic field.

The pressure gradients generate levitation (expulsion) forces (directed away from the field) on immersed particles, the magnetization of which is lower compared to the fluid. As is the case in high-gradient magnetic separation (HGMS), high-gradient magnetohydrostatic systems[11] can

provide forces which can be made equivalent to high g forces. High-gradient electric separation[13-14] (HGES) is less widespread as compared to HGMS, but in principle it has similar potential applications. The incorporation of electromagnetic forces, which are comparable to centrifugal ones in a wide range of acceleration levels, is promising as a new means for extending the present capabilities of both centrifugal and magnetic separation techniques.

A new separation technology which combines magnetic and centrifugal fields is named here magnetocentrifugal separation (MCS), and the one involving electric and centrifugal fields is similarly named electrocentrifugal separation (ECS). The purpose of this paper is to establish the theory, principles, feasibility, and potential applications of the new MCS and ECS techniques and to investigate their expected capabilities.

II. THEORY

A. General approach

The "theory" is divided into two parts. Part I deals with nondiffusing particulates, while diffusion-controlled systems are considered in Part II. In Part I we develop first the equation of motion for monodisperse particles in a fluid flow, centrifugal, and electromagnetic fields. This equation of motion provides the basis for analysis of magnetocentrifugal separation. Magnetic, electric, and acceleration g numbers are defined with a view to characterize the three fields as to the number of g's they can provide. These g numbers facilitate the use of a common basis for comparison of the expected role of each field. A clear distinction is drawn between g numbers, which are defined for monodisperse, and polydisperse, particle mixtures. The range of centrifugal fields in which magnetic and acceleration g numbers can be matched, and the magnetic fields that can be used to this end, are also considered. Upon completion of this introductory section, specific magnetocentrifugal separation systems of nondiffusing particles are defined, and their characteristic features are analyzed. An extension of the theory for magnetocentrifugal separation of diffusing mixtures of particles is given at the closure of this section. This completes the theoretical basis for the full range of particle sizes which are known to have been treated by centrifuges.

1. Part I: Nondiffusing particulate systems

a. Equation of motion of monodisperse particles suspended in a fluid in centrifugal and electromagnetic force fields. Assumptions:

(1) Particles are spherical and monodisperse.

(2) The dispersion is uniform (locally or throughout) with respect to particle concentration and distribution of particle properties (such as size, density, susceptibility, etc.)

(3) The fluid is incompressible.

(4) Flow is Newtonian.

(5) No significant interaction other than hydrodynamic exists between particles

(6) The fluid is contained within rigid walls at constant volume.

(7) Relaxation times associated with hydrodynamic drag forces are orders of magnitude smaller than the ones associated with the centrifugal and electromagnetic fields. Hence, given the particle instantaneous velocity, relative to the fluid, use of a fully developed drag force (for this particular velocity) introduces only a small error which can be neglected.

(8) Diffusion effects can be neglected.

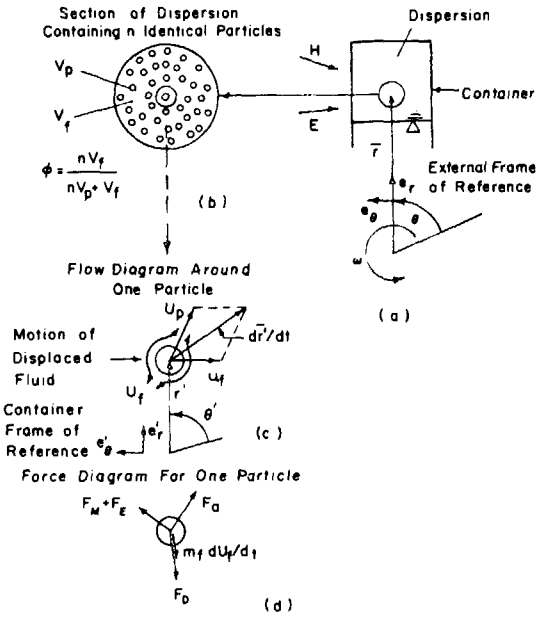

FIG. 1. Details of the separation system.

Figure 1 shows a diagram of the system under consideration. The external (unprimed) frame of reference is used to describe the motion of the container and the internal (primed) one for the motion of the suspension, its fluid and particles, relative to the container. The figure shows schematically (a) the container, (b) a section of the dispersion, (c) flow diagram relative to the container frame of reference in the vicinity of a particle, and (d) force diagram, in the container frame of reference, for one particle. (Note the two frames of reference are assumed two-dimensional for the sake of simplicity.)

The equation of motion of a particle moving in a fluid and centrifugal and electromagnetic fields at constant volume is given[15,16] by Eq. (1):

$$\frac{m_p dU_p}{dt} = F_A + F_M + F_E + F_D + \frac{m_f dU_f}{dt}, \qquad (1)$$

where F_A, F_M, F_E, and F_D are acceleration, magnetic, electrical, and drag forces, respectively. The last term on the right accounts for the average reaction force due to displaced fluid being accelerated by the moving particle.[15] U and m are velocity and mass, and subscripts p and f denote particles and fluid, respectively.

$$F_A = F_G + F_a, \qquad (2)$$

where the forces F_G (gravitation) and F_a (nongravitational two-dimensional acceleration) are given by

$$F_G = (\rho_p - \rho_\phi) V_p g = \rho(1-\phi) V_p g, \quad \rho = \rho_p - \rho_f, \qquad (3)$$

$$F_a = \int_{V_p} (\rho_p - \rho_\phi) a dV_p = \rho(1-\phi)\int_{V_p} a dV_p, \quad a = \frac{d\bar{v}}{dt}, \tag{4}$$

$$\frac{d\bar{v}}{dt} = \left(\frac{d^2 r}{dt^2} - r\omega^2\right) e_r + \left(\frac{r d^2\theta}{dt^2} + 2v\omega\right) e_\theta ,$$

$$\omega = \frac{d\theta}{dt}, \quad v = \frac{dr}{dt}.$$

ϕ is volume fraction occupied by particles in the dispersion. As a subscript it denotes a property pertaining to the whole dispersion at ϕ. V_p is volume of a particle, ρ density, and r and θ polar coordinates (see Fig. 1). e_r and e_θ are unit vectors in the radial and azimuthal directions, respectively. t is time, $\bar{v}$ vectorial velocity, and ω and v angular and radial velocities, respectively.

$$F_M = \int_{V_p} [(M_p - M_\phi)\cdot\nabla] H dV_p$$

$$= (1-\phi)\int_{V_p} (M\cdot\nabla) H dV_p , \tag{5}$$

$$M_\phi = \phi M_p + (1-\phi) M_f ,$$

$$M = M_p - M_f \text{(see Ref. 16)} .$$

M_p, M_f, and M_ϕ are magnetizations of particles, fluid, and of the dispersion, respectively. H is magnetic field intensity.

$$F_E = F_q + F_p , \tag{6}$$

where F_q and F_p are coulombic and electric polarization forces,

$$F_q = (1-\phi)\int_{V_p} E q_v dV_p + (1-\phi)\int_{S_p} E q_s dS_p , \tag{7}$$

and it is assumed that the fluid charge can be neglected.

E, q_v, and q_s are electric field intensity, and volume and surface charge densities, respectively. S_p is surface area of a particle.

$$F_p = \int_{V_p} [(P_p - P_\phi)\cdot\nabla] E dV_p$$

$$= (1-\phi)\int_{V_p} (P\cdot\nabla) E dV_p ,$$

$$P_\phi = \phi P_p + (1-\phi) P_f, \quad P = P_p - P_f . \tag{8}$$

P_p, P_f, and P_ϕ are electric polarization of particle, fluid, and dispersion, respectively. For the creeping (Stokes) flow regime:[15]

$$F_D = -3\pi d \mu'_\phi U_\phi, \quad \mu'_\phi = \mu_\phi (1 + \phi^{1/3}) , \tag{9}$$

and for the intermediate and turbulent regimes:[15]

$$F_D = -(\pi/8)[0.63(U_\phi d \rho_f)^{1/2} + 4.80\mu_\phi^{1/2}]^2 \times d(1+\phi^{1/3}) U_\phi , \tag{10}$$

$$\mu_\phi = \mu_f \exp\{\tfrac{5}{3}[\phi/(1-\phi)]\} , \tag{11}$$

$$U_\phi = U_p + U_f . \tag{12}$$

μ_f and d are fluid viscosity and particle diameter, respectively. U_p and U_f are average velocities of particles and fluid (which is displaced by particle motion), relative to the dispersion, respectively. U_ϕ is velocity of particles relative to the fluid they have displaced:

$$m_f \frac{dU_f}{dt} = -\alpha_m m_p \left(\frac{\rho_f}{\rho_p}\right)\frac{dU_p}{dt} , \tag{13}$$

where α_m is a factor accounting for the effect of the system geometry on the value of the virtual mass.[17–22]

For a sphere in an infinite inviscid fluid $\alpha_m = 1/2$.[17–22] By continuity we have[23]

$$U_p = U_\phi (1-\phi) , \tag{14}$$

and

$$U_f = U_\phi \phi . \tag{15}$$

Combining Eqs. (1–9), (13) and (14) yields

$$V_p(\rho_p + \alpha_m \rho_f)\frac{d^2\bar{r}'}{dt^2} = (1-\phi)\int_{V_p} (M\cdot\nabla) H dV_p + (1-\phi)\int_{V_p} (P\cdot\nabla) E dV_p$$
$$+ (1-\phi)\int_{V_p} E q_v dV_p$$
$$+ (1-\phi)\int_{S_p} E q_s dS_p + V_p \rho (1-\phi) g + \rho(1-\phi)\int_{V_p}\left(\frac{d\bar{v}}{dt}\right) dV_p$$
$$- 3\pi d \mu'_\phi \left(\frac{d\bar{r}'}{dt} - u_f\right) \Big/ (1-\phi) , \tag{16}$$

where u_f is the velocity of the suspension relative to the fixed frame of reference of its container and $d\bar{r}'/dt = U_p + u_f$, see Fig. 1.

The characteristic features of magnetocentrifugal and electrocentrifugal separation involve control of balance between centrifugal and electromagnetic forces, or alternatively between corresponding g numbers which are defined below.

b. Definition of magnetic electric and acceleration g numbers.

Monodisperse particles:

Let the magnetic force be defined in terms of an equivalent acceleration a'_m as per Eq. (17):

$$\rho_p V_p a'_m = (1-\phi)\int_{V_p} (M\cdot\nabla) H dV_p , \tag{17}$$

where a'_m is the "magnetic acceleration." Rearranging yields

$$a_m = (1/\rho_p)(1-\phi)|\overline{(M\cdot\nabla)H}|,$$

$$\overline{(M\cdot\nabla)H} = (1/V_p)\int_{V_p}(M\cdot\nabla)H dV_p, \quad (18)$$

where $a_m = |a'_m|$. The magnetic "g number" m_g is defined by

$$m_g = a_m/g = [1/(\rho_p g)](1-\phi)|\overline{(M\cdot\nabla)H}|. \quad (19)$$

The "electric polarization" g number e_g is defined in the same way:

$$e_g = [1/(\rho_p g)](1-\phi)|\overline{(P\cdot\nabla)E}|. \quad (20)$$

Note that here g is the modulus (i.e., number) of gravity acceleration.

Magnetic versus centrifugal g numbers:

The radial centrifugal g number C_{gr} is defined by

$$\rho_p V_p a_r = \rho(1-\phi)V_p\omega^2 r, \quad a_r = \rho(1-\phi)\omega^2 r/\rho_p, \quad (21)$$

where a_r is the equivalent centrifugal acceleration,

$$C_{gr} = a_r/g = \rho(1-\phi)\omega^2 r/(\rho_p g). \quad (22)$$

Taking the radial component of m_g and dividing Eqs. (19) and (22) yields the dimensionless ratio R_g of the magnetic to centrifugal g numbers:

$$R_g = m_g/C_{gr} = [\overline{(M\cdot\nabla)H}]_r/(\rho\omega^2 r). \quad (23)$$

Here subindex r denotes projection on the radial direction. Note that R_g is also the ratio of the respective magnetic and centrifugal forces.

It is of interest to identify under which conditions R_g may be equal or greater than one, in particular, in cases where C_{gr} is large, say 10^3–10^5. In the following equation it will be assumed that (1) M is colinear with H, and (2) particles are small compared to the field gradient, i.e., taking the field as constant throughout the particle volume does not introduce a significant error. In this case we have

$$R_g = (M\nabla H)_r/(\rho\omega^2 r). \quad (24)$$

Let

$$\rho = 2.0\ \text{g/cm}^3, \quad \omega^2 r = 3.0\times 10^8\ \text{cm/s}^2, \quad R_g = 1.0,$$

and assume that H and M are radial, hence

$$M\nabla H = 6.0\times 10^8\ \text{g cm}^{-2}\,\text{s}^{-2}.$$

If $4\pi M = 600$ G, then $\nabla H = 4\pi 10^6$ Oe/cm must be satisfied. This magnetization and higher ones can be obtained by ferromagnetic particles and ferrofluids. The required field gradient can be generated, for example, by a set of ferromagnetic wires 10-μm-thick which are placed in a 20 000-Oe (or higher) uniform field.

Thus it is seen that by using high gradient magnetic fields, the magnetic g number can be made comparable to the centrifugal g number in the full range of centrifugal fields currently used commercially. As shown below, the magnetic g number and R_g for particles moving in a dispersion comprising polydisperse particle mixture are different than those defined by Eqs. (19) and (23), respectively.

Polydisperse particles:

Let the polydisperse mixture comprise n distinct fractions. It is assumed that the mixture is uniformly dispersed in a continuous fluid, and the basic assumptions, excluding No. 1, listed at the beginning of Sec. II apply here as well. For this case the magnetic g number m_{gj} [Eq. (30)] of the particles pertaining to the jth fraction (of a mixture comprising n fractions) is defined using the same procedure described for monodisperse particles, as follows. Here the jth magnetic acceleration a_{mj} is defined by Eq. (25):

$$\rho_p V_p a_{mj} = \int_{V_p}[(M_{pj} - M_\phi)\cdot\nabla]H dV_p, \quad (25)$$

where the average magnetization of the dispersion M_ϕ is given by[16]

$$M_\phi = \sum_{i=1}^{n}\phi_i M_{pi} + (1-\phi)M_f, \quad (26)$$

$$\phi = \sum_{i=1}^{n}\phi_i, \quad i = 1,2,\ldots,j,\ldots,n, \quad (27)$$

$$M_{pj} - M_\phi = M_{pj} - \sum_{i=1}^{n}\phi_i M_{pi} - (1-\phi)M_f. \quad (28)$$

Combining Eqs. (27) and (28) and rearranging gives

$$M_{pj} - M_\phi = M_j(1-\phi) + \sum_{i=1}^{n}\phi_i M_{ji}, \quad (29)$$

where

$$M_j = M_{pj} - M_f \text{ and } M_{ji} = M_{pj} - M_{pi}.$$

Combining Eqs. (25) and (29) and dividing by $\rho_p V_p g$ yields the magnetic g number:

$$m_{gj} = [1/(\rho_p g)]\left|\left[\left(\bar{M}_j(1-\phi) + \sum_{i=1}^{n}\phi_i\bar{M}_{ji}\right)\cdot\nabla\right]H\right|, \quad (30)$$

where $\bar{M}_j$ and $\bar{M}_{ji}$ denote volume average in the same sense as in Eq. (19).

Equation (30) shows that for particles which are part of a polydisperse mixture the magnetic g number depends on the distributions of ϕ_i as well as of M_{ji} (i.e., of M_i). If M_{ji} includes positive as well as negative values, m_{gj} may have positive and negative values as well. The radial centrifugal g number of a particle which is part of a polydisperse mixture can be obtained in a similar way. Here the particle centrifugal acceleration a_r is defined by

$$\rho_p V_p a_r = \int_{V_p}(\rho_p - \rho_\phi)\omega^2 r dV_p, \quad (31)$$

which finally gives the radial centrifugal g number C_{grj} (and also R_{gj}), for particles pertaining to the jth fraction

$$C_{grj} = \left(\bar{\rho}_j(1-\phi) + \sum_{i=1}^{n}\phi_i\bar{\rho}_{ji}\right)\omega^2 r\Big/(\rho_p g), \quad (32)$$

where

$$\bar{\rho}_j = \overline{\rho_{pj} - \rho_f}, \quad \bar{\rho}_{ji} = \overline{\rho_j - \rho_i} .$$

$$R_{gj} = \frac{m_{gj}}{C_{grj}} = \left((\bar{M}_j(1-\phi) + \sum_{i=1}^{n} \phi_i \bar{M}_{ji}) \cdot \nabla \right) H \Big/ \left[\left(\bar{\rho}_j(1-\phi) + \sum_{i=1}^{n} \phi_i \bar{\rho}_{ji} \right) \omega^2 r \right] . \tag{33}$$

Equations (23) and (33) show that in cases where $\rho(=\rho_p - \rho_f)$ is small, or the mixture properties impose a small density difference between the particle and dispersion, R_g or R_{gj} may be further increased. In such cases the magnetic force may become dominant even at "high g" fields. In this context centrifugation of biological dispersions, where density differences are extremely small, may be included.

The magnetic force [Eq. (5)], equation of motion [Eq. (16)], and magnetic g numbers are all dependent on the magnetic field intensity as well as on its configuration. Here a nonhomogeneous magnetic field must be used for generation of magnetic forces which can be applied to magnetizable particles. Such fields can be designed using a nonhomogeneous field directly or, as often done, by combination of an external field and ferromagnetic bodies (e.g., arrays of wires or balls) which intensify and distort it in their vicinity. Properties of selected magnetic field configurations, which may be useful in design of magnetocentrifugal separation systems, are described in Appendix A. We proceed now to look at some implications of the equation of motion in magnetocentrifugal systems.

Magnetocentrifugal systems comprising magnetizable monodisperse particles moving in a fluid stream:

Consider the case where $E = P = 0$. Equation (16) reduces to Eq. (34):

$$\frac{d^2\bar{r}'}{dt^2} + A\frac{d\bar{r}'}{dt} + \bar{B} = 0 , \tag{34}$$

$$A = 18\pi\mu'_\phi / [(1-\phi)(\rho_p + \alpha_m \rho_f) d^2] ,$$

$$\bar{B} = -(1-\phi)[\overline{(M\cdot\nabla)H} + \rho(a+g) + \bar{f}] / (\rho_p + \alpha_m \rho_m \rho_f) ,$$

$$\bar{f} = 18\mu'_\phi \mu_f / [(1-\phi)d]^2 .$$

If the motion is two-dimensional, the derivatives in Eq. (34) are given by

$$\frac{d\bar{r}'}{dt} = \left(\frac{dr'}{dt}\right) e'_r + r'\left(\frac{d\theta'}{dt}\right) e'_\theta, \quad r' = |\bar{r}'| , \tag{35}$$

$$\frac{d^2\bar{r}'}{dt^2} = \left[\frac{d^2r'}{dt^2} - r'\left(\frac{d\theta'}{dt}\right)^2\right] e'_r + \left[r\frac{d^2\theta'}{dt^2} + 2\left(\frac{dr'}{dt}\right)\left(\frac{d\theta'}{dt}\right)\right] e'_\theta , \tag{36}$$

where e'_r and e'_θ are radial and azimuthal unit vectors, which are defined in the container frame of reference, see Fig. 1.

Denoting the radial and azimuthal components of $\bar{B}$ by B_r, B_θ and introducing the dimensionless variables $r_a = r'/b$ and $\tau = (u_0/b)t$, Eq. (34) takes the following equivalent form:

$$\frac{d^2r_a}{d\tau^2} - r_a\left(\frac{d\theta'}{d\tau}\right)^2 + A'\left(\frac{dr_a}{d\tau}\right) + B'_r = 0 , \tag{37}$$

$$r_a\frac{d^2\theta'}{d\tau^2} + 2\left(\frac{dr_a}{d\tau}\right)\left(\frac{d\theta'}{d\tau}\right) + A' r_a\left(\frac{d\theta'}{d\tau}\right) + B'_\theta = 0 , \tag{38}$$

$$A' = \frac{b}{u_0}A, \quad B'_r = \frac{b}{u_0^2}B_r, \quad B'_\theta = \frac{b}{u_0^2}B_\theta ,$$

where b is a characteristic length such as the radius of a magnetized wire[16] and u_0 is a reference velocity of the particle outside the magnetic field. If $M \| H$, and the variation of H within the particle is negligible, we have

$$B'_r = -(1-\phi)b M_r\left(\frac{\partial H}{\partial r'}\right)\left\{1 + [\rho(a_r + g_r) + f_r] \Big/ \left[M_r\left(\frac{\partial H}{\partial r'}\right)\right]\right\} \Big/ [u_0^2(\rho_p + \alpha_m\rho_f)] , \tag{39}$$

$$B'_\theta = -(1-\phi)b M_\theta\left(r'\frac{\partial H}{\partial \theta'}\right)\left\{1 + \left[\rho(a_\theta + g_\theta) + f_\theta\right] \Big/ \left[M_\theta\left(r'\frac{\partial H}{\partial \theta'}\right)\right]\right\} \Big/ [u_0^2(\rho_p + \alpha_m\rho_f)] ,$$

$$f_r = \bar{f}\cdot e'_r, \quad f_\theta = \bar{f}\cdot e'_\theta .$$

Subscripts r and θ (which, for the sake of simplicity, are not primed) denote here radial and azimuthal components in the container frame of reference.

Let the following dimensionless numbers characterize the magnetocentrifugal separation system:

$$N_n = B'_n/A' = \frac{-(1-\phi)^2 M_n (r^\delta \partial H/\partial n)\{1 + [\rho(a_n + g_n) + f_n]/[M_n(r^\delta \partial H/\partial n)]\} d^2}{18\pi\mu'_\phi u_0} , \tag{40}$$

where

$$n = r' \text{ or } \theta' \quad \text{and} \quad \delta = |n - r'|/|\theta' - r'| .$$

$N_n u_0$ has the dimension of velocity comprising magnetic and acceleration components. If magnetic forces prevail, it is related to the magnetic velocity defined by Watson.[24] If acceleration force prevails, it may be related to an "acceleration velocity," which can be defined similar to the magnetic velocity. If both centrifugal (i.e., acceleration) and magnetic forces are significant, $N_n u_0$ yields the magnetocentrifugal velocity. Note that replacing M by P and H by E in Eq. (40) yields the electric polarization counterpart of the magnetic velocity. In this case the polarization electrocentrifugal velocity is obtained from $N_n u_0$.

Different cases of MCS can be obtained by variation of fluid density and magnetization relative to those of the particles. Figure 2 summarizes relations between directions of magnetic and centrifugal forces in relation to M and ρ and the directions of magnetic field gradient, acceleration field,

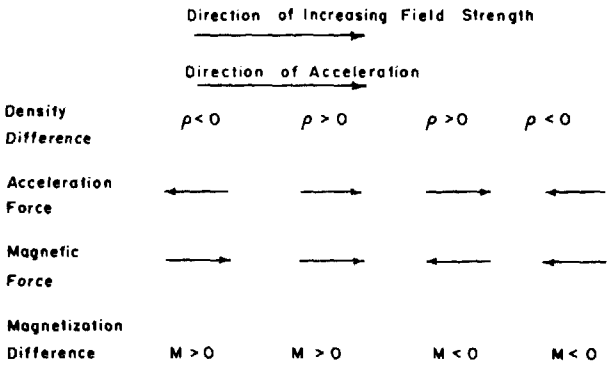

FIG. 2. Directions of magnetic and centrifugal forces in relation to the sign of M and ρ, and the directions of magnetic field gradient and acceleration field.

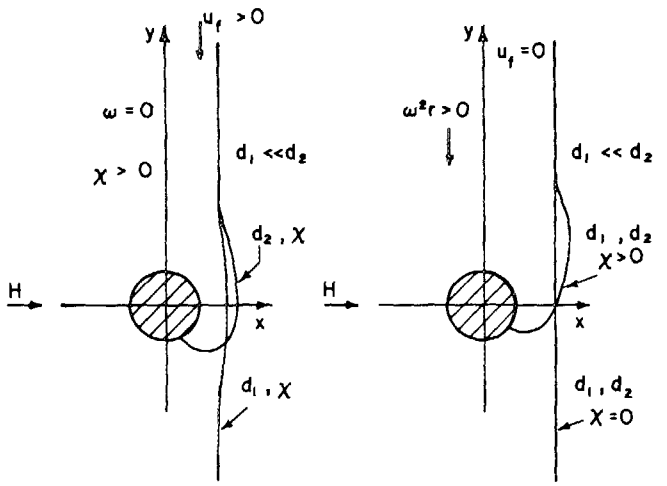

FIG. 3. Schematic trajectories of two particles, of different sizes and the same susceptibility, in HGMS: $u_f > 0$, $\omega^2 r = 0$, and MCS: $u_f = 0$, $\omega^2 r > 0$, systems.

and motion of particles. Choosing an appropriate combination from Fig. 2 and setting the relative intensities of the magnetic and acceleration forces is expected to yield an effective magnetocentrifugal separation system. We proceed to study special cases of magnetocentrifugal separation where the fluid velocity vanishes $u_f = 0$.

d. Magnetocentrifugal separation of magnetizable monodisperse particles in a stationary fluid of lower magnetization and lower density.

Magnetocentrifugal separation of magnetizable particles ($\chi_p > \chi_f$) for which $M > 0$ and $\rho > 0$, is an extension of the high-gradient magnetic separation (HGMS) technique. Here, in addition to the magnetic and fluid drag forces, the centrifugal force is available for control of the system. If the fluid is stationary, then the driving forces are proportional to the volume or mass of the particles. Rearranging Eqs. (37) and (38) followed by their division yields

$$\frac{1}{r_a}\left(\frac{dr_a}{d\theta}\right) = \frac{B'_r + d^2 r_a/d\tau^2 - r_a(d\theta'/d\tau)^2}{B'_\theta + r_a d^2\theta'/d\tau^2 + 2(dr_a/d\tau)(d\theta'/d\tau)}. \tag{41}$$

If $u_f = 0$, then [see Eq. (34)] $f_r = f_\theta = 0$, and B'_r, B'_θ and hence $dr_a/d\theta'$, which determines the shape of the particle trajectories, are independent of particle size d. Recall that the particles were assumed small enough so that variation of the field and its gradient across them can be neglected.

Figure 3 depicts schematically a basic difference, regarding expected trajectories, between a magnetocentrifugal ($u_f = 0$) and a conventional single wire HGMS ($u_f \neq 0$) systems. The figure shows that in the HGMS system the larger (d_2) and smaller (d_1) particles follow different trajectories. d_2 is captured while d_1 does not deviate sufficiently from the fluid streamlines and hence is carried downstream. Both particles are expected to follow the same trajectory, in the magnetocentrifugal system, if their upstream starting points are identical. In this case if d_2 is expected to be captured so is d_1, and if no capture is expected, both particles will flow downstream. It is seen that magnetocentrifugal separation is expected to render the system more selective with respect to susceptibilities and densities of particles. HGMS systems are versatile for a wide range of particle sizes and susceptibilities but are less known as an efficient separator that functions as a concentrator.

Magnetocentrifugal separation can be applied to ferromagnetic, paramagnetic, or diamagnetic particles, and for each category of particles a different field design (i.e., matrices) must be implemented. Figure 4 shows three alternative systems of separation using arrays of ferromagnetic wires. In the first system, particles which are not captured must cross the array plane; in the second and third systems they flow parallel to it, and those that are captured deviate from the fluid stream towards the wires where they are finally deposited. Intermediate design involves an array set at an angle $0 < \alpha < 90°$ towards $\omega^2 r$, and also at an angle (different from $\pi/2$) to H. A similar arrangement, using a matrix of random wires or packed beds of ferromagnetic particles, may be effective. Figure 5 shows schematically a cell packed with a bed of ferromagnetic matrix (which, for example, can be steel balls or wire mesh) supported by a screen and nonmagnetic material, e.g., particles such as glass beads. Part of the magnetic particles may be captured if the right balance between the magnetic and acceleration force is maintained. The nonmagnetic particles will penetrate the top bed of ferromagnetic particles and be collected at the bottom.

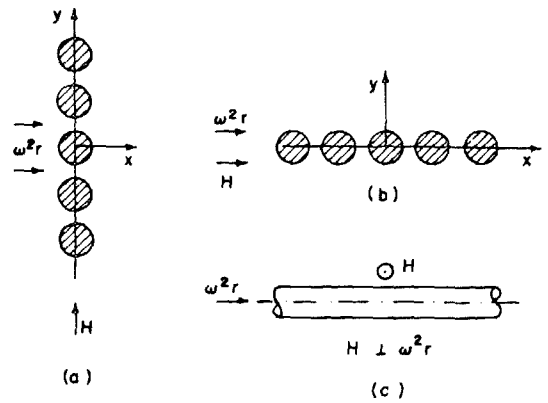

FIG. 4. Three alternative MCS systems using arrays of ferromagnetic wires.

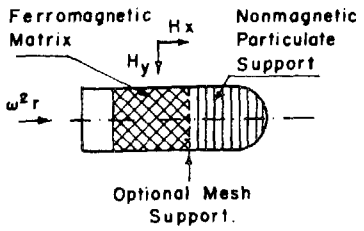

FIG. 5. MCS separation cell packed with ferromagnetic matrix on top of a nonmagnetic support.

e. Magnetocentrifugal separation of particles moving in a stationary fluid of intermediate and higher magnetization, but lower density.

This case bridges magnetocentrifugal separation (MCS) which is related to high-gradient magnetic separation (HGMS) and MCS which relates to the magnetohydrostatic separation (MHS). In HGMS the dominant separation variable is M while in MHS it is ρ. Six different cases, which relate magnetic properties of the particle population to the field gradient, can be defined as follows:

In the first two cases $M>0$ for the whole particle population, in the third and fourth cases the particle population splits as regards magnetization and $M>0$ and $M<0$ coexist, and in the fifth and sixth cases $M<0$ for the whole particle population.

(1) $M_i>0$, $\nabla H>0$ for all particle fractions i.

(2) $M_i>0$, $\nabla H<0$ for all particle fractions i.

(3) $M_i<0$, for $i<i_l$, and $M_i\geqslant 0$ for $i\geqslant i_l$, $\nabla H>0$, i, and i_l denote the ith particle fraction and division between two parts of the particle population, respectively.

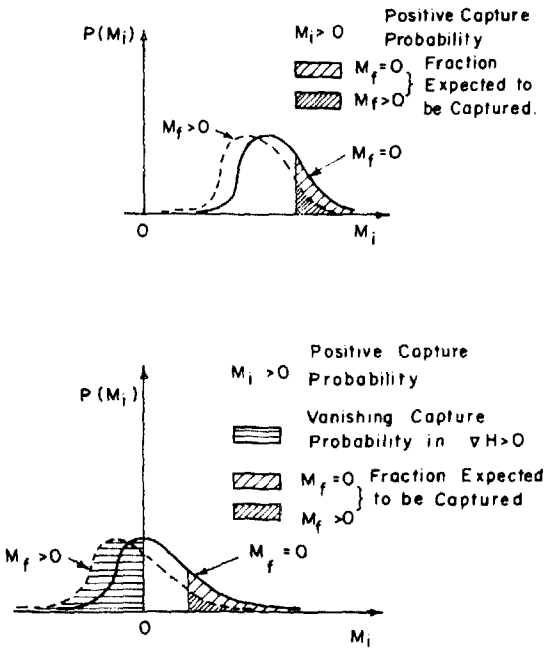

FIG. 6. (a) Effect of raising fluid magnetization on capture probability of $M_i>0$ particles (schematic). (b) Effect of raising fluid magnetization on capture probability of a particle distribution characterized by $M_i>0$ and $M_i<0$ fractions.

(4) $M_i<0$ *for* $i<i_l$, and $M_i>0$ for $i\geqslant i_l$, $\nabla H<0$.

(5) $M_i<0$, $\nabla H>0$, for all particle fractions i.

(6) $M_i<0$, $\nabla H<0$, for all particle fractions i.

Figure 6(a) depicts schematically the effect of raising the fluid magnetization, i.e., decreasing M_i, on capture of particles for which $M_i>0$. The figure shows that the whole population can be captured if the right combinations of magnetic and centrifugal forces exist. Raising M_f from $M_f=0$ to $M_f>0$ decreases the fraction which is expected to be captured. This may have an effect of improving grade, i.e., purity, of the separated particles. In the second case no particle capture due to the magnetic field is expected. Figure 6(b) illustrates schematically the third case. Here part of the distribution characterized by $M_i<0$ has a vanishing probability of being captured by the $\nabla H>0$ magnetic field. As the fluid magnetization is raised, the distribution shifts in the $-M_i$ direction, leaving a smaller fraction which can be expected to be captured. In the fourth case only part of the $M_i<0$ can be expected to be captured. The fifth case involves the phenomenon of levitation.

Figure 7 shows two types of M_i distributions. If the first distribution denoted I applies, then both M_i and ρ_i control the MCS separation. If, however, the second one denoted II (for which $M_p\rightarrow 0$, $M_i\rightarrow -M_f$) prevails, then ρ alone becomes the controlling separation variable. This case provides a new separation technology for fine particles. This technology is named magnetocentrifugal density separation (MCDS). Here particles are not separated by being captured, but rather by crossing (or not crossing) an apparent density barrier existing within the fluid. Figure 8 depicts schematically a possible MCDS separation cell. Particles which cross the density barrier (generated by the wire array) will settle to the cell bottom and hence separate from the "lighter" ones, which will be collected in the region marked A in Fig. 8. The setup shown in Fig. 5, which was considered before, can also be used to separate $M_i<0, \rho>0$ particles according to their density by attraction into demagnetization zones (generated near each ferromagnetic matrix element).

2. Part II: Diffusion-controlled magnetocentrifugal separation

Diffusion-controlled magnetocentrifugal separation involves a combined effect of concentration gradients and drift velocity imposed on the diffusing particles, by centrifugal

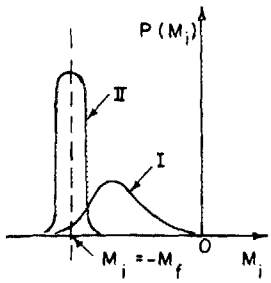

FIG. 7. Narrow and wide distribution of the difference (M_i) between particle and fluid magnetizations. $P(M_i)$ is probability density function.

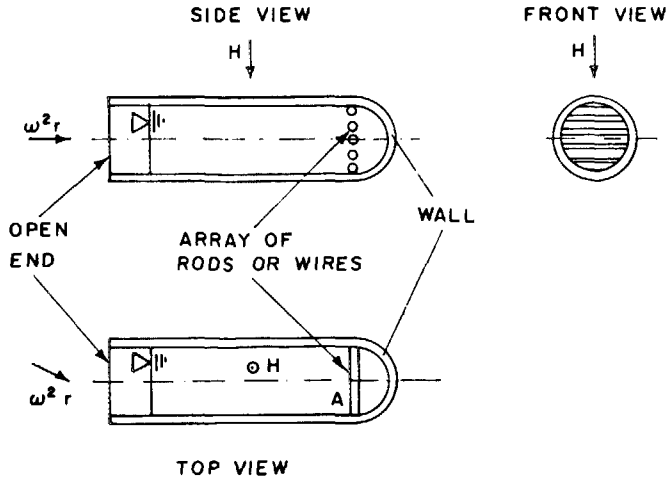

FIG. 8. A MCDS separation cell.

and magnetic forces. Related systems can be described by a magnetocentrifugal diffusion and sedimentation equation, which is developed here for polydisperse diffusing mixtures.

Consider a mixture comprising N components, the fluid (denoted by $i=1$) being included. The diffusion equation for the ith component of this mixture can be derived from continuity as follows:

$$\frac{\partial c_i}{\partial t}+\nabla J_i=0, \quad J_i=J_{iD}+J_{iF}, \tag{42}$$

$$J_{iD}=-\sum_{k=2}^{N} D_{ik}\,\nabla c_k, \quad i=2,3,\ldots,N, \tag{43}$$

$$J_{iF}=U_i c_i, \tag{44}$$

where c_i is molar concentration given in moles of the dispersed or dissolved species per unit volume; J_{iD}, J_{iF}, and J_i are molar flux densities, due to diffusion, drift velocity, and their sum, respectively. D_{ik} is diffusion coefficient of the ith component with the respect to the k th component of the mixture, and U_i is the drift velocity relative to the dispersion. Combining Eqs. (42)–(44) gives

$$\frac{\partial c_i}{\partial t}=\sum_{k=2}^{N}\nabla\cdot(D_{ik}\nabla c_k)-\nabla\cdot(U_i c_i). \tag{45}$$

Different separation methods have been or can be developed, by application of different external forces on the diffusion system.

A good example is the versatile ultracentrifugal analysis currently used today for separation of a diffusing system. This method has become standard practice in biochemistry.[25,26] In ultracentrifugal analysis separation occurs due to differential sedimentation velocities (of the diffusing system) which are generated in high acceleration fields. Recently it was shown by Takayasu *et al.*[27] and others[28] that high-gradient magnetic separation (HGMS) can be applied successfully for separation of submicron particles dispersed in a motionless fluid. This method was effective in treating a wide range of particle susceptibilities, e.g., diamagnetic, paramagnetic, and ferromagnetic. Combining the ultracentrifugal and HGMS techniques for diffusion systems is the essence of the new diffusion-controlled magnetocentrifugal separation method. In the following we consider general aspects of diffusion sedimentation systems, and proceed towards the development of the magnetocentrifugal diffusion and sedimentation equation [Eq. (60)]. Drift velocities are usually related to particle mobilities (u_i), or in ultracentrifugal analysis to the so-called sedimentation coefficient (S_i). In the latter case we find[25,26]

$$J_{iF}=c_i S_i\omega^2 r, \quad U_i=S_i\omega^2 r. \tag{46}$$

Combining Eqs. (45) and (46) yields the differential equation governing the behavior of a multicomponent diffusion-sedimentation system. This equation takes the following form in the radial (r) direction:[25,26]

$$\frac{\partial c_i}{\partial t}=\frac{1}{r}\frac{\partial}{\partial r}\left[\left(\sum_{k=2}^{N} D_{ik}\frac{\partial c_k}{\partial r}-\omega^2 r\,S_i c_i\right)r\right]. \tag{47}$$

Equation (47) reduces to Eq. (48) for a two-component system, where D and S (subscripts dropped for simplicity) are independent of c (which is often not the case) and therefore of r:

$$\frac{\partial c}{\partial t}=D\left(\frac{\partial^2 c}{\partial r^2}+\frac{1}{r}\frac{\partial c}{\partial r}\right)-S\omega^2\left(r\frac{\partial c}{\partial r}+2c\right). \tag{48}$$

In ideal two-component monodisperse particle dispersions, which, for example, are well approximated by dilute solutions, D can be estimated using the Nernst–Einstein relation

$$D=ukT, \tag{49}$$

where u satisfies the Stokes hydrodynamic drag law

$$u=1/(3\pi\mu_f d). \tag{50}$$

k,T,μ_f, and d are Boltzmann's constant, absolute temperature, fluid viscosity, and particle diameter, respectively. If, however, the effect of concentration cannot be neglected, a concentration dependent correction factor $f(c)$ must be applied:

$$u=f(c)/(3\pi\mu_f d). \tag{51}$$

Many forms of $f(c)$ can be derived from literature concerned with sedimentation.[29–32] For example, Fujita[33] used Eq. (52) to calculate diffusion sedimentation profiles:

$$f(c)=1-Kc, \quad K=\text{const}. \tag{52}$$

We shall use here (see Appendix B) an advanced form of $f(c)$ following the results of Barnea and Mizrahi,[29] which were obtained for sedimentation of monodisperse particles in the creeping flow regime:

$$f(c)=(1-\phi)^2/\{(1+\phi^{1/3})\exp[\tfrac{5}{3}\phi/(1-\phi)]\}, \tag{53}$$

$$\phi=cV, \tag{54}$$

where ϕ is total volume fraction occupied by the dispersed particles (or solute) and V is partial molar volume. In cases where polydisperse, i.e., multicomponent, systems are involved, each fraction (component) i has a different concentration dependent correction factor $f_i(c)$, see Appendix B.

$$f_i(c)=\left(\xi_i(1-\phi)+\sum_{k=1}^{N}c_kV_k\xi_{ki}\right)\Big/\left[\xi_i\left(U_{pi}+\sum_{i=2}^{N}c_iV_iU_{\phi i}\right)(1+c_i^{1/3}V_i^{1/3})\times\exp\left(\{\tfrac{5}{3}[c_iV_i/(1-c_iV_i)]\}\right)\right], \quad (55)$$

$$\xi_i=\xi_{pi}-\xi_f,\quad \xi_{ki}=\xi_{pk}-\xi_{pi},$$

where ξ is ρ or M in the case of centrifugal or magnetic fields, respectively.

In cases involving polydisperse, i.e., multicomponent, systems different concentration correction factors apply to each fraction i of the mixture and to each type of driving force. Thus, if there are N fractions and m types of driving force ($F_{mo}, m=1,2,...,m'$), then mN correction factors should be applied. Let $f_{mi}(c)$ denote this array. If only two types of driving forces exist, namely centrifugal and magnetic, then

$$f_{mi}(c)=\left(\xi_{mi}(1-\phi)+\sum_{k=1}^{N}c_kV_k\xi_{kmi}\right)\Big/\left[\xi_{mi}\left(U_{pi}+\sum_{i=2}^{N}c_iV_iU_{\phi i}\right)(1+c_i^{1/3}V_i^{1/3})\times\exp\{\tfrac{5}{3}[c_iV_i/(1-c_iV_i)]\}\right], \quad (56)$$

$$\xi_{mi}=\xi_{pi}-\xi_f,\quad \xi_{kmi}=\xi_{pk}-\xi_{pi}.$$

Here ξ_m is ρ for $m=1$, and M for $m=2$, which stand for the cases of centrifugal and magnetic forces, giving $f_{1i}(c)$ and $f_{2i}(c)$, respectively. The drift velocity of the ith fraction (or disolved component) becomes

$$U_{pi}=\sum_{m=1}^{m'}F_{moi}u_{mci}, \quad (57)$$

$$U_{mci}=f_{mi}(c)/(3\pi\mu_f d). \quad (58)$$

Combining Eqs. (45), (57), and (58) yields a generalized diffusion and sedimentation equation:

$$\frac{\partial c_i}{\partial t}=\sum_{k=2}^{N}\nabla\cdot(D_{ik}\nabla c_k)-\sum_{m=1}^{m'}\nabla\cdot\left(F_{moi}f_{mi}(c)\frac{c_i}{(3\pi\mu_f d)}\right),\quad i=2,3,...,N,\quad m=1,2,...,m'. \quad (59)$$

Applying Eq. (59) to the case of magnetocentrifugal separation (MCS) yields the MCS diffusion and sedimentation equation.

$$\frac{\partial c_i}{\partial t}=\sum_{k=2}^{N}\nabla\cdot(D_{ik}\nabla c_k)-\nabla\cdot[(V_{pi}\rho_i\omega^2rf_{1i}(c)+V_{pi}(M\cdot\nabla)Hf_{2i}(c)c_i]/(3\pi\mu_f d). \quad (60)$$

In a two-component MCS system, and dropping the subscript $i=2$, Eq. (60) reduces to Eq. (61):

$$\frac{\partial c}{\partial t}=\nabla\cdot(D\nabla c)-\nabla\cdot\left(\frac{(V_p\rho\omega^2r+V_p(M\cdot\nabla)H)(1-cV)^2c}{3\pi\mu_f d(1+c^{1/3}V^{1/3})\exp\{\tfrac{5}{3}[cV/(1-cV)]\}}\right). \quad (61)$$

Note that in this case, for monodisperse single solute particles suspended in a fluid solvent, the concentration correction factors for the centrifugal and magnetic forces are identical. Presentation of solutions to the magnetocentrifugal diffusion and sedimentation equation is outside the scope of this work. We shall, however, discuss the significance of the theoretical results. Eqs. (60) and (61) indicate that diffusion-controlled magnetocentrifugal systems facilitate a significant extension of the separation capabilities of the conventional ultracentrifugal method. In view of the fact that different magnetic field configurations can be designed, the magnetic term in Eqs. (60) and (61) facilitates formation of the concentration gradient in different directions, which are otherwise limited to the radial direction only. This opens new opportunities, which are not available today by other techniques, for separation of diffusing particulates according to their magnetization and density levels. Consider a case in which high magnetic and centrifugal forces are comparable in magnitude. Here the magnetic force can counteract the centrifugal and diffusion drift of magnetizable particles, thus separating them from the drifting (less magnetizable) ones. In other cases the magnetic force can be used to vary the drift velocities of diffusing particles by deflecting, to different degrees, their trajectories. In this way separation according to particle magnetization, density, and diffusion coefficient can be achieved.

B. Potential applications of magnetocentrifugal separation (MCS)

In a general sense MCS has promising potential for application in cases spanning from magnetic to density separation of particulates. A vanishing effect of particle size obtained by use of stationary fluids, or else the elimination of the effect of their flow, facilitates better selectivity in MCS systems. In such systems magnetic susceptibility and density become the prevailing separation variables. Thus, MCS can be applied to suspensions involving fine and ultrafine (colloidal) particles which are characterized by magnetic and density distributions. The distribution may be natural or artificially prepared. Magnetic tagging of particles can be used to change their magnetic properties, and attachment or impregnation of materials with different densities will change their density distribution.

Such techniques in conjuction with MCS may find applications in biochemistry chemical and biomedical engineering, i.e., in processing and separation of cells, bacteria, and viruses. MCS can be used to separate colloidal matter extracted by or mixed with ferrofluids. Here the 5–15-nm ferromagnetic (and surfactant coated) particles will be captured in the magnetic field while the nonmagnetic ones will be driven away by the centrifugal force.

Multicomponent and high-resolution density separa-

tion can be achieved by control of density and magnetization of the fluid. MCS is advantageous for fine and ultrafine particles made of materials which are heavier than available fluids. Furthermore, it is promising in cases characterized by variable "near density" particles which exhibit narrow density differences. Here sensitive adjustment of the fluid's apparent density can be done by variation of the field intensity, without having to change its composition. It should be noted that if magnetizable dispersions such as ferrofluids are used as medium of separation, the particle size of this dispersion should be significantly smaller compared to the size of the particles being separated. Thus, paramagnetic solutions may be required (instead of ferrofluids) for separation of molecular size particulates.

Finally, MCS may be designed to function as susceptibility or density chromatographic systems. This area is yet to be explored as regards its potential uses and design features. It is clear nevertheless that in such systems a distribution of susceptibility or density may be "fanned out" along the axis of separation, thus yielding a continuous chromatographic separation.

C. Summary and conclusions

(1) Magnetocentrifugal separation (MCS) incorporates forces which are exerted on particulates by the simulataneous action of magnetic and centrifugal force fields.

(2) The magnitude of electromagnetic and centrifugal g numbers and related forces, available from technologies currently used today, can be matched throughout the acceleration range of commercial centrifuges and ultracentrifuges.

(3) Magnetic and acceleration g numbers of particles which are part of a magnetization and density distributions depend on the latter's properties such as average and standard deviation. The corresponding g numbers may span positive as well as negative values.

(4) Centrifugal fields combined with magnetic field gradients, generated by ordinary and superconducting magnets in conjunction with harmonic pole pieces or around ferromagnetic elements such as wires, spheres, metal fragments, or generally particulates, are essential parts of MCS systems.

(5) The intensity of the acceleration and magnetic fields and the magnetic field gradient of a MCS system should be adjusted according to size of the particulates involved. The smaller their size, the higher the required intensities and field gradients.

(6) MCS systems in which particle concentration is low and the fluid is stationary are practically independent of the size of particles which are being separated. Such systems are dependent on density and magnetization of particles and fluid.

(7) MCS systems can be applied in the full range of magnetizations known to date, i.e., for diamagnetic, paramagnetic, ferromagnetic, ferrimagnetic, and antiferromagnetic particles. The combination between values of M_i and ∇H determines probability of particle capture. For a given acceleration field, higher values of M_i and ∇H increase probability of capture.

(8) Magnetocentrifugal density separation (MCDS) can be applied to nonmagnetic particles by using magnetizables fluids. If the fluid is a dispersion, then the sizes of its particles must be significantly smaller than the ones being separated.

(9) Diffusion-controlled MCS systems involve balance between diffusion and centrifugal and magnetic drift forces. A generalized definition of particle mobilities, which applies to concentrated dispersions, is applied for mono and polydisperse diffusion-controlled MCS systems.

(10) A generalized diffusion and sedimentation equation for polydisperse particles in a system involving several types of drift forces has been formulated. Here a generalized concentration-dependent correction factor, to the mobility of each fraction under each type of drift force, has been incorporated.

(11) MCS and MCDS have potential application in chemical, biochemical, and biomedical engineering, in particular for ultrafine colloid and "near density" particulate systems. Successful applications require engineering of separation cells, ferromagnetic matrix, and the centrifugal and magnetic fields. Suggestions for cell design have been presented.

APPENDIX A: CHARACTERISTICS OF SOME USEFUL NONHOMOGENEOUS MAGNETIC FIELDS

The nonhomogeneous fields considered here consist of the following types:

(1) Nonhomogeneous fields which are obtained by shaping the field-generating device. In this case we say that the primary field is nonhomogeneous.

(2) Nonhomogeneous fields which are obtained by distortion of an external (i.e., primary) field, which may be uniform or nonuniform. In this case the required degree of field distortion is obtained by placing ferromagnetic bodies in the external field.

Figure 9 shows schematically a field generated between two-dimensional curved pole pieces, N and S, of a magnet. For this configuration the primary field is nonuniform, and its intensity increases as the gap and field gradient diminish.

Figure 10 shows schematically a two-dimensional array of ferromagnetic cylinders (e.g., wires) in a uniform magnetic field. In this case the highly permeable cylinders distort the uniform field, thus generating localized strong nonuniform field. The wire arrangement facilitates increase of field

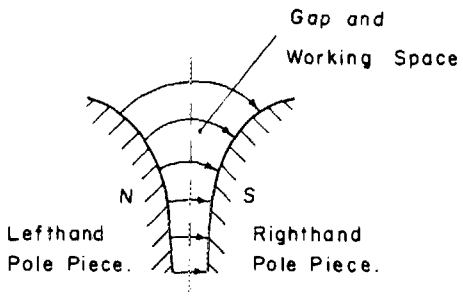

FIG. 9. Magnetized ferromagnetic pole pieces which produce nonuniform magnetic field (schematic).

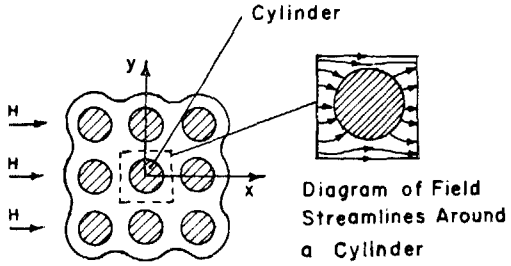

FIG. 10. A two-dimensional array of ferromagnetic wires in a uniform magnetic field.

gradient (and hence of the magnetic force) in the vicinity of each wire, by decreasing their diameter. Thus, the setup shown in Fig. 9 may be used for relatively low-gradient fields, while the type represented by Fig. 10 can extend the range to very high gradients (for example, 10^6–10^7 G/cm).

A. Primary harmonic fields

In the absence of electric currents, and assuming that the ferromagnetic metal surface can be matched with an equipotential surface, then the field in the working space can be described by harmonic functions which satisfy Eq. (A1), and boundary conditions $\Phi(x,y) = \text{cons.}$ set by the surface of the pole pieces:

$$\nabla^2\Phi = 0 . \tag{A1}$$

Equation (A1) is satisfied by two-dimensional complex functions[12] of the type given by Eq. (A2),

$$W_n = \sum_{n=1}^{n} A_n (Z - Z_0)^n, \quad Z = x + jy, \quad j = \sqrt{-1} . \tag{A2}$$

Since W_n is a complex function, we also have

$$W_n = U' + jV', \quad U' = \mathrm{Re}(W), \quad V' = \mathrm{Im}(W) . \tag{A3}$$

1. Simple harmonic field

If $A_j = 0$ for all $j \neq n$, and $z_0 = x_0 + jy_0 = 0$, then U and V are given by[12]

$$U' = -A_n r^n \sin n\theta, \quad \theta = \theta'' - \pi/(2n), \tag{A4}$$

$$V' = A_n r^n \cos n\theta , \tag{A5}$$

where r and θ'' are polar coordinates. Equation (A4) can be used to describe magnetic fields generated between pole pieces of the type shown in Fig. 9. The magnetic forces generated by such fields are considered next.

If particle and fluid magnetizations are colinear with H, Eq. (A6) is satisfied:

$$M_p = \chi_p H, \quad M_f = \chi_f H . \tag{A6}$$

In this case the net magnetic force exerted on monodisperse particles which are immersed in a fluid is given by Eq. (A7).

$$f_m = (1/2) V_p \chi (1 - \phi) \nabla H^2, \quad \chi = \chi_p - \chi_f . \tag{A7}$$

Recalling that

$$H = -\nabla U' \tag{A8}$$

and combining Eqs. (A4), (A7), and (A8) yields the x and y components of the magnetic force:

$$f_{mx} = V_p (\chi_p - \chi_f)(1 - \phi)(n - 1) n^2 A_n^2 r^{2n-3} \cos\theta , \tag{A9}$$

$$f_{my} = f_{mx} \tan\theta . \tag{A10}$$

Equation (A9) shows that if $n = 3/2$, and provided ϕ is held constant, f_{mx} and f_{my} are independent of the radial distance. In this case the magnetic field is identified as isodynamic, and particles moving on the symmetry axis ($\theta = 0$) are subjected to a constant magnetic force.

B. Field around arrays of wires

Consider the field around an infinite two-dimensional array of ferromagnetic wires shown in Fig. 10. Note that one-dimensional arrays are special cases of two-dimensional ones. They are obtained by increasing infinitely the y and x spacing of the wires, in Fig. 10, respectively. The following cases are of interest: (a) The wire surfaces are equipotentials. (b) The wires are saturated. For both cases the field outside the wires can be approximated using a generalized complex function given by Eq. (A11):

$$W = \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} A_{kl} \sum_{i=1}^{i'} f_i [Z - Z_i(k,l)] - H_e Z , \tag{A11}$$

where A_{kl} (which are real), i', and $Z_i(k,l)$ are determined by boundary conditions, $f_i[Z - Z_i(k,l)]$ are analytic complex functions, and H_e a uniform external field. i' denotes the number of functions used per wire, and k, l the wire position.

1. Wire surfaces are equipotentials[34]

In this case $A_{kl} = -K$, where K is a positive constant, $i' = 2$, $f_1 = \ln$, and $f_2 = -f_1$. Thus, Eq. (A11) takes the following form:

$$W = -K \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} \{\ln[Z - Z_1(k,l)] - \ln[Z - Z_2(k,l)]\} - H_e Z , \tag{A12}$$

where

$$\left.\begin{aligned} Z_1(k,l) &= kx_0 + ly_0 j + a \\ Z_2(k,l) &= kx_0 + ly_0 j - a \end{aligned}\right\} k,l = -\infty,\ldots,-1,0,1,\ldots,\infty . \tag{A13}$$

x_0 and y_0 are the x and y spacing of the wires, respectively. The parameter a accounts for effects of wire interactions, hence, given the wire spacing, it is a function of the wire radius R. For a square grid:[34]

$$a = 1.5113 (R/B)^{2.975} , \tag{A14}$$

for a one-dimensional array parallel to the field:

$$a = 1.4234 (R/B)^{3.092} , \tag{A15}$$

and for the one set across the field:

$$a = 1.0997 (R/B)^{2.8686} , \tag{A16}$$

where B is half the interwire spacing. Figure (11) shows values of H/K vs a plotted for square grids, and the parallel and across-the-field one-dimensional array. Substituting a [Eqs. (A14)–(A16)] in Eq. (A13) and using the data of $H/$

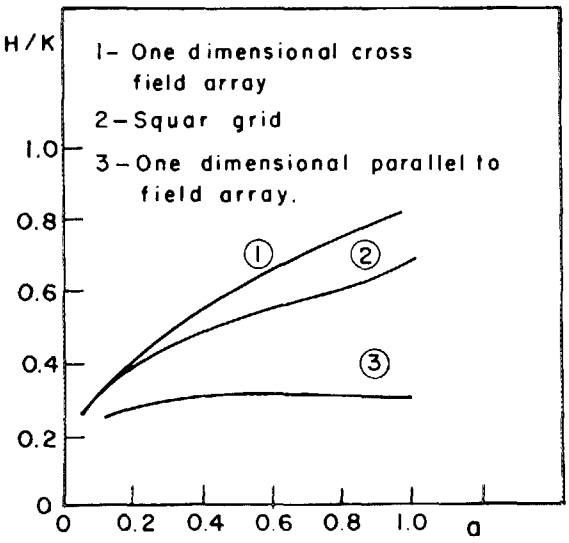

FIG. 11. A plot of H/k vs a for square grids.

K from Fig. (11) yields, via Eq. (A12), a complex potential around the array.

2. Saturated wires

In this case the array can be simulated by replacing each wire with a doublet set in its center parallel to the field. Here $a \to 0$ and K [Eq. (A12)] must be set so as to maintain a finite field to dipole moment ratio and the boundary conditions. The field intensity modulus $|H|$ may be evaluated using Eqs. (A12) and (A17):

$$|H| = \left|\frac{dW}{dZ}\right| \tag{A17}$$

$$|H| = K\left|\sum_{k=-\infty}^{\infty}\sum_{l=-\infty}^{\infty}\frac{1}{Z-Z_1(k,l)} - \frac{1}{Z-Z_2(k,l)} + H_e\right| . \tag{A18}$$

Hence the magnetic force around the arrays can be evaluated via Eq. (A7).

Other types of arrays, with different geometrical arrangements and wire cross sections, may be effective in generating large forces (i.e., high-intensity high-gradient fields) which are comparable to the full range of acceleration available from centrifuges today. These include triangular array, which facilitates higher packing densities of the wires, and two dimensional array of rectangular slabs, which can provide high-intensity high-gradient fields in the vicinity of their edges. The choice of an appropriate array depends, however, on the objectives of the MCS system being considered.

APPENDIX B

The hydrodynamic creeping flow drag force exerted on monodisperse particles [see Eqs. (9), (11), and (14)] is given by

$$F_D = -3\pi\mu_f d(1+\phi^{1/3}) \times \exp\{\tfrac{5}{3}[\phi/(1-\phi)]\}U_p/(1-\phi) . \tag{B1}$$

The centrifugal [Eq. (4)] and the magnetic force [Eq. (5)] are also related to ϕ:

$$F_\phi = F_0(1-\phi) , \tag{B2}$$

where F_ϕ and F_0 imply the value of the force (centrifugal or magnetic) at $\phi \geqslant 0$ and at $\phi = 0$, respectively. Dividing Eqs. (B2) by (B1), dropping the minus sign and setting $F_0 = 1$ yields the generalized particle mobility u_c:

$$u_c = [1/(3\pi\mu_f d)]f(\phi) , \tag{B3}$$

$$f(\phi) = (1-\phi)^2/((1+\phi^{1/3})\exp\{\tfrac{5}{3}[\phi/(1-\phi)]\}) . \tag{B4}$$

expressing ϕ as a function of c yields $f(c) = f[\phi(c)]$, see Eqs. (53) and (54).

$f(\phi)$, and hence $f(c)$, are defined here so that u_c is the correct particle mobility which is obtained per unit force, calculated at $\phi = 0$. For example, if $a_c = \omega^2 r$, then,

$$F_0 = \rho V_p \omega^2 r . \tag{B5}$$

The expected particle drift velocity at $c > 0$ would be

$$U_p = F_0 u_c = [\rho\omega^2 r d^2/(18\mu_f)]f(c) . \tag{B6}$$

Consider now the case of a polydisperse mixture, i.e., a multicomponent system. The mixture comprises N fractions of monodisperse particles. Here the drift velocity of the ith fraction with respect to the dispersion or solution (in case the mixture comprises solvent and solutes) is given by

$$U_{pi} = U_{\phi i} - U_f, \quad i = 2,3,\ldots,N , \tag{B7}$$

$$U_f = \sum_{i=2}^{N} \phi_i U_{\phi i} , \tag{B8}$$

where

$$\phi = \sum_{i=2}^{N} \phi_i, \quad \phi_i = c_i V_i . \tag{B9}$$

$i = 1$ denotes the fluid (or solvent), and V_i is molar volume of the ith component:

$$\sum_{i=1}^{N} \phi_i = \sum_{i=1}^{N} c_i V_i = 1 , \tag{B10}$$

hence

$$F_{Di} = 3\pi\mu_f d(1+\phi^{1/3})\exp\{\tfrac{5}{3}[\phi/(1-\phi)]\} \times\left(U_{pi} + \sum_{i=2}^{N} c_i V_i U_{\phi i}\right) . \tag{B11}$$

The driving force, whether centrifugal or magnetic, must now be calculated taking into account the average density or magnetization of the mixture,

$$F_{\phi i} = F_{0i}\frac{\xi_{pi} - \xi_\phi}{\xi_{pi} - \xi_f}, \quad \xi_\phi = \sum_{i=2}^{N} \xi_{pi}\phi_i + \xi_f(1-\phi) , \tag{B12}$$

where ξ denotes ρ and M for centrifugal and magnetic force, respectively. Rearranging Eq. (B12) gives

$$F_{\phi i} = F_{0i}\left(\xi_i(1-\phi) + \sum_{k=1}^{N} c_k V_k \xi_{ki}\right)\Big/\xi_i , \tag{B13}$$

$$\xi_i = \xi_{pi} - \xi_f, \quad \xi_{ki} = \xi_k - \xi_i .$$

Equation (55) is obtained by dividing Eq. (B13) by (B11) and setting $F_{0i} = 3\pi\mu_f d$.

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