Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Hournkumnuard, Kanok and Natenapit, Mayuree(2008) 'Diffusive Capture of Magnetic Particles by an Assemblage of Random Cylindrical Collectors', Separation Science and Technology, 43: 13, 3448 — 3460
To link to this Article: DOI: 10.1080/01496390802222566
URL: http://dx.doi.org/10.1080/01496390802222566

Separation Science and Technology, 43: 3448–3460, 2008

ISSN: 0149-6395 print/1520-5754 online
DOI: 10.1080/01496390802222566

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Diffusive Capture of Magnetic Particles by an Assemblage of Random Cylindrical Collectors

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Abstract: We develop an effective medium model describing the capture of ultra-fine magnetic particles (diameter much less than 1 μm) by an assemblage of parallel magnetic cylinders, randomly distributed in static fluid. The continuity equation describing the dynamics of concentration is solved numerically to obtain the concentration in various regions around the collector. The concentration contours are generated and the saturation, accumulation, and depletion regions are indicated. The effect of varying collector packing fractions appears significantly in regions close to the outer boundary of the representative cell where the build-up features of ultra-fine particles near the collector surface are similar for packing fractions in range 5–10%.

Keywords: Diffusive capture, effective medium, high gradient magnetic separation, nanoparticles

INTRODUCTION

High Gradient Magnetic Separation (HGMS) is a powerful method for the removal of ultra-fine weakly magnetic particles, that are much smaller than 1 μm, from suspensions. This method is applied in many fields of work, for example, chemical, blood separation in biochemical laboratories, and in the pharmaceutical industry. Recently, nano-level magnetic separation has received much attention (1–3). Theoretical investigation laying emphasis on the detailed dynamics of particle capture including the effect of neighbouring wire collectors was performed by Okada

Received 16 January 2008; accepted 10 March 2008.
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et al. (4). Microfluidic high gradient magnetic separation of blood cells have been discussed by Inglis et al. (5). The generalized theory describing the capture of ultra-fine magnetic particles by a single ferromagnetic cylindrical collector in one dimension and the simulation of the capture process have been reported by Gerber et al. (6,7). The simulation of ultra-fine particle capture by a single ferromagnetic cylindrical collector has been performed and reported by Davies and Gerber (8). Their concentration contours, showing areas of depletion, accumulation, and saturation, predict the behavior of particle buildup on the single collector.

Several types of devices which are feasible for separation of magnetic particles have been reported. These include a cell capture system using removable magnets (9), continuous flow separations in microfluidic devices (10) and a three-dimensional magnetic trap using permanent magnets (11). The theoretical studies of magnetic field distributions of some permanent magnets have been reported (12) which could be applied to determine the force acting on the magnetic particles. A practical magnetic cell sorting system for separation of large numbers of cells investigated by Miltenyi et al. (13) has been filled with ferromagnetic stainless steel wires coated with plastic by immersion into lacquer in order to avoid damage of cells trapped on the wire surface. The wires are magnetized by an external magnetic field creating high gradient magnetic field and extremely high forces on the magnetic cells (14). The structure of this device is similar to the type we aim to study. By using an electromagnet, the magnetic field is switched off to release the trapped particles. The adjustable field strength and collector packing fractions would enable us to design the filters suitable for different requirements. The devices are applicable to separate very weakly magnetic and nonmagnetic particles of dimensions down to nanometer size. The theoretical predictions of the device efficiency for varying flow rates, filter lengths and field strength have been reported previously (15–17).

In this research, the capture of ultra-fine magnetic particles by an assemblage of parallel cylindrical collectors randomly distributed in the static fluid is investigated. The only specified geometrical character of the system is the ratio of the total volume of collectors to the total volume of the system which is defined as the packing fraction of the collectors in the system. This research includes the theory of diffusive capture in high gradient magnetic fields, the effective medium treatment for prediction of the magnetic field around a representative cylindrical collector including the effect of neighboring collectors and the simulation results that show particle concentration distribution around a representative cylindrical collector. These results not only depict the buildup features of magnetic particles on the surface of the representative collector but also predict the percentage of magnetic particles captured on the surface of the collector from the total number of magnetic particles in the system. Finally, the conclusions of this work are given in the last section.

THEORY

In HGMS, magnetic collectors made from ferromagnetic or paramagnetic materials of cylindrical (or spherical) shape and a fluid with suspended magnetic particles are contained in a non-magnetic canister. In many practical cases, the magnetic collectors distribute randomly in the fluid. A uniform external magnetic field $\mathbf{H}_0$ is applied in the perpendicular direction to the axis of these collectors. There exist regions of high gradient magnetic fields around each collector. Any magnetic particles in or entering these regions are subjected to a large magnetic traction force. To capture these particles at the collectors, it is necessary that the magnetic traction force is directed towards the collectors and is large enough to prevail over the action of other forces and processes hence particles are brought to and retained at the collectors. The other forces and processes involved can be the viscous drag force of the fluid, the gravity force, thermal diffusion and inter-particles effects, etc. Not all of these forces and processes are significant in certain situations. In some situations, we can reasonably approximate that some forces or processes are more significant than others. The magnetic traction force acting on the magnetic particles can be expressed as (6)

$$\mathbf{F}_m = \frac{1}{2}\mu_0 \chi V_p \nabla(\mathbf{H}^2), \tag{1}$$

where μ_0, χ, V_p, $\mathbf{H}$ are the permeability of free space, the difference between the magnetic susceptibility of the particles and the fluid, the volume of the magnetic particles, and the magnetic field at the position of the magnetic particles, respectively. The HGMS theory describes, the dynamics of the capture of ultra-fine particles in terms of particle volume concentration and particle drift velocity denoted by c and $\mathbf{v}$, respectively. The particle volume concentration at a given point in the fluid is dimensionless and defined as a fraction of the volume of ultra-fine particles contained in an infinitesimal volume element of fluid at that point. The particle volume concentration is a function of the positions in fluid and time and also satisfies the continuity Eq. (6)

$$\frac{\partial c}{\partial t} + \nabla \cdot \mathbf{J} = 0, \tag{2}$$

where $\mathbf{J}$ at a given point in the fluid is considered to consist of two contributions as (6)

$$\mathbf{J} = \mathbf{J}_D + \mathbf{J}_F, \tag{3}$$

where $\mathbf{J}_D$ denotes the diffusion flux due to diffusion and $\mathbf{J}_F$ the particle volume flux due to the actions of external forces on the system of particles. Diffusion flux can be determined by Fick's law as (6)

$$\mathbf{J}_D = -D\nabla c, \tag{4}$$

where D is the diffusive coefficient of the ultra-fine particles in the fluid.

The particle volume flux due to the actions of external forces which imposes a drift velocity **v** on the system of ultra-fine particles is expressed as (6)

$$\mathbf{J}_F = c\mathbf{v} = cu\mathbf{F}, \tag{5}$$

where u is the mobility of the ultra-fine particles in the fluid and **F** is the total external force acting upon those particles composed of magnetic traction force, fluid viscous drag force, electric force and gravitational force.

When expressions of $\mathbf{J}_D$ and $\mathbf{J}_F$ in Eqs. (4) and (5), respectively, are substituted in Eq. (3), we obtain the continuity equation for the system of ultra-fine particles as

$$\frac{\partial c}{\partial t} = \nabla\cdot(D\nabla c) - \nabla\cdot(c\mathbf{v})\cdot \tag{6}$$

The diffusive coefficient of the ultra-fine particles in the fluid is determined, throughout this research, by Einstein's relation (6),

$$D = uk_BT, \tag{7}$$

where k_B and T are the Boltzmann's constant and the absolute temperature, respectively. The value of D is assumed to be independent of the position in fluid. With this assumption, Eq. (6) is rewritten as

$$\frac{\partial c}{\partial t} = D\nabla c^2 - \nabla\cdot(c\mathbf{v}), \tag{8}$$

which is the continuity equation describing the dynamics of the system of ultra-fine particles in high gradient magnetic separation for general cases.

It should be noted that the force Eq. (1) is inapplicable to superparamagnetic particles. Extensive investigations of superparamagnetic biotinylated-microparticles ($\sim$100 nm diameter) have been reported in Ref. 13.

CAPTURE OF ULTRA-FINE MAGNETIC PARTICLES BY AN ASSEMBLAGE OF RANDOM CYLINDRICAL COLLECTORS

We consider a system consisting of two parts. The first part is a static fluid with an assembly of monotype ultra-fine weakly magnetic particles as a suspension. Both fluid and particle are considered to be linear isotropic homogeneous magnetic media. The other part is an assemblage of paramagnetic cylindrical collectors randomly distributed in the fluid. These collectors are considered to have characteristic distributions of cylindrical radii and are very long compared with their diameters. In this research, the axes of these collectors are considered all parallel. The system of fluid and collectors are contained in a non-magnetic canister. A uniform magnetic field $\mathbf{H}_0$ is applied perpendicularly to the axes of these collectors. We study the dynamics of the capture of ultra-fine particles by these collectors. The only characteristic of the system we know is the packing fraction of the collectors in the fluid.

Since all of the collectors in the system are randomly distributed, when an arbitrary collector is considered, all residual collectors locate randomly with respect to it. This situation is the same for any collectors in the system. Since the assemblage of collectors have characteristic distributions of cylindrical radii, the distribution of collectors' radii surrounding an arbitrary collector is random. From this, we can reasonably approximate that the capture operation of an arbitrary collector is affected by the existence of other collectors equally.

The magnetic field around the assemblage of cylindrical paramagnetic collectors randomly distributed in a fluid has been determined by Natenapit and Sanglek (18) by using the effective medium model originally conceived by Hashin (19). In the effective medium model, the system of magnetic cylinders (permeability μ_c) and surrounding fluid (permeability μ_f) is considered to be composed of cylindrical composite cells, each containing exactly one of the cylinders. In this model, only a representative cell is considered, while the neighbor cells are replaced by a homogeneous medium with effective permeability μ^* to be determined. Figure 1 shows a representative cell in the effective medium

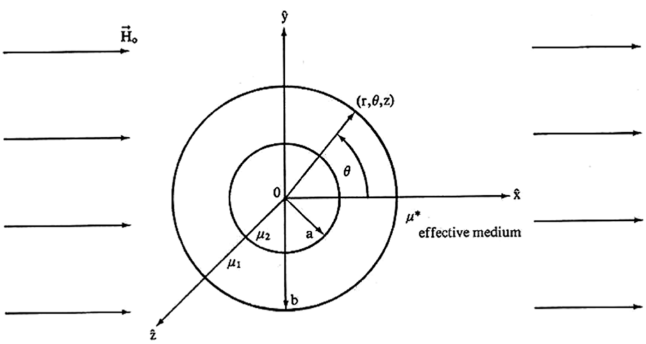

Figure 1. A representative cylindrical cell.

model with a and b being the radii of the collector and the representative cell, respectively. Since the ratio of the collector to the cell volume is set equal to the packing fraction (P) of collectors in the fluid then we obtain $P = a^2/b^2$. The z-axis of cylindrical coordinate system is set along the cylinder axis.

To determine the magnetic fields in the cell and the effective medium, the boundary value problem of coaxial magnetic cylinders subject to the boundary condition of the uniform magnetic field far away from the cell was solved. According to the effective medium model, a composite cell was chosen to be the representative cell and the residual cells were considered to be a homogeneous effective medium. A self-consistency had to be satisfied that the magnetic induction averaged over the representative cell (cylindrical collector plus surrounding fluid) had to equal the volume average of the magnetic induction over the effective medium (19). According to Natenapit and Sanglek (18), the magnetic field in the fluid surrounding the collector in a representative cell was determined as

$$\mathbf{H} = AH_0\left[\left(1+\frac{K_c}{r_a^2}\right)\cos\theta\,\hat{r} - \left(1-\frac{K_c}{r_a^2}\right)\sin\theta\,\hat{\theta}\right], 1 < r_a < b/a, \qquad (9)$$

where $r_a = r/a$, $A = (1 - PK_c)^{-1}$, $K_c = (\nu - 1)/(\nu + 1)$ and $\nu = \mu_c/\mu_f$.

The magnetic field in the effective medium outside the representative cell was determined as

$$\mathbf{H} = \mathbf{H_0}, \quad b/a < r_a < \infty. \qquad (10)$$

We can see from Eq. (10) that, according to the effective medium model, the gradient $\nabla\mathbf{H}^2$ outside the representative cell is equal to zero hence the capture of ultra-fine particles can be considered only in the representative cell. From Eq. (9), it is observed that the effects of the existence of other collectors on the magnetic field around an arbitrary collector are contained in the factor A. In the limit of dilute packing fraction (P approach to zero), the factor A approach to unity and the problem was reduced to the case of single collector as expected.

By using the effective medium model, the problem of HGMS capture of ultra-fine magnetic particles by an assemblage of random cylindrical paramagnetic collectors can be transformed to the problem of single cylindrical paramagnetic collector in the representative cell.

The two-dimensional continuity equation for this case can be expressed as (8)

$$\frac{\partial c}{\partial \tau} = \left\{\frac{\partial^2 c}{\partial r_a^2} + \frac{1}{r_a}\frac{\partial c}{\partial r_a} + \frac{1}{r_a^2}\frac{\partial^2 c}{\partial \theta^2}\right\} - \left\{\frac{G_r c}{r_a} + \frac{\partial (G_r c)}{\partial r_a} + \frac{1}{r_a}\frac{\partial (G_\theta c)}{\partial \theta}\right\}, \qquad (11)$$

where

$$G_r(r_a,\theta) = G_0\left(\frac{\cos(2\theta)}{r_a^3} + \frac{K_c}{r_a^5}\right), \tag{12}$$

$$G_\theta(r_a,\theta) = G_0\left(\frac{\sin(2\theta)}{r_a^3}\right), \tag{13}$$

$$G_0 = -\frac{8\pi\mu_0\chi A^2 H_0^2 K_c b_p^3}{3k_B T}, \tag{14}$$

and $\tau = Dt/a^2$ is a dimensionless normalized time.

BOUNDARY CONDITIONS OF SIMULATIONS

We impose two boundary conditions in the simulation. The first is the boundary condition at the surface of the collector and the surface of static buildup of ultra-fine particles. Both surfaces are treated as impervious surfaces where the particle volume flux in the radial direction at any point on these surfaces is considered equal to zero. The second boundary condition is imposed on the outer boundary of the representative cell. All simulations in this research are performed for the capture of ultra-fine particles in static fluid. The situation can be compared as the same as the capture of ultra-fine particles in a closed canister where the net particle volume flux flow through the overall system equals zero. Since the representative cell is the representation of the system, we then impose a boundary condition that the net particle volume flux in all directions perpendicular to the outer boundary of the representative cell must equal zero.

RESULTS OF SIMULATIONS

In this research, we simulate the capture of paramagnetic $Mn_2P_2O_7$ particles (radius $b_p = 1.2 \times 10^{-8}$ m) in static water. The difference between the magnetic susceptibility of the particles and water ($\chi_p - \chi_f = \chi$) is $+4.73 \times 10^{-3}$ (the paramagnetic mode of the capture). The uniform external magnetic field has its magnitude $H_0 = 2.0 \times 10^6$ A/m (the magnetic flux density = 2.5 Tesla) which is perpendicular to the axes of all cylindrical collectors. The parameter K_c is equal to 0.20. The absolute temperature is set equal to 300 K. The value of initial concentration at every points in the representative cell is set equal to 1.0×10^{-3} and the saturation concentration is set equal to 0.10 (8). The value of the packing fractions of cylindrical collectors in fluid is varied from P = 5%, 8% to 10%. For all values of packing fractions, simulations are performed at the same interval of normalized time as $0 \leq \tau \leq 0.10$. From the results of the simulations, concentration contours are generated around the representative collector and the feature of magnetic particle buildup on the surface of the collector is depicted. The concentration contours are shown only in the first quadrant, $(0 \leq \theta \leq \pi/2)$, since the distribution of concentration is symmetrical about X and Y axes.

Figure 2 shows the concentration contours around the collector for the case of P = 5%, $G_0 = -16.95$ and $\tau = 0.10$. The figure shows the build-up features of magnetic particles on the surface of the collector and the feature of concentration distribution around the collector in the representative cell. The symbols S, A, and D refer to the saturation, accumulation, and depletion region, respectively. In Fig. 2, we see that at $\tau = 0.10$ saturation concentration has already take place on the surface of the collector. Figure 3 shows the concentration contours around the collector for the case of P = 8%, $G_0 = -17.16$ and $\tau = 0.10$ and Fig. 4 shows the concentration contours around the collector for the case of P = 10%, $G_0 = -17.30$ and $\tau = 0.10$. The symbols of

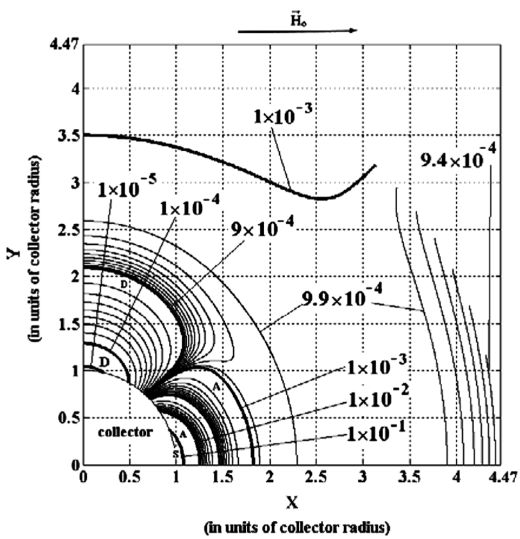

Figure 2. A family of concentration contours around the paramagnetic collector in the representative cell, P = 5%, $G_0 = -16.95$, $\tau = 0.10$.

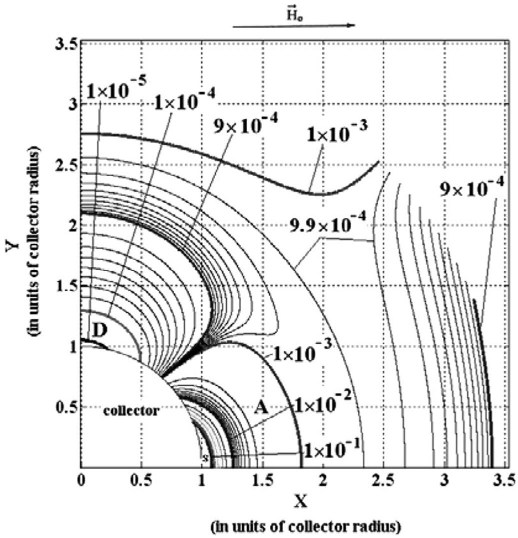

Figure 3. A family of concentration contours around the paramagnetic collector in the representative cell, $P = 8\%$, $G_0 = -17.16$, $\tau = 0.10$.

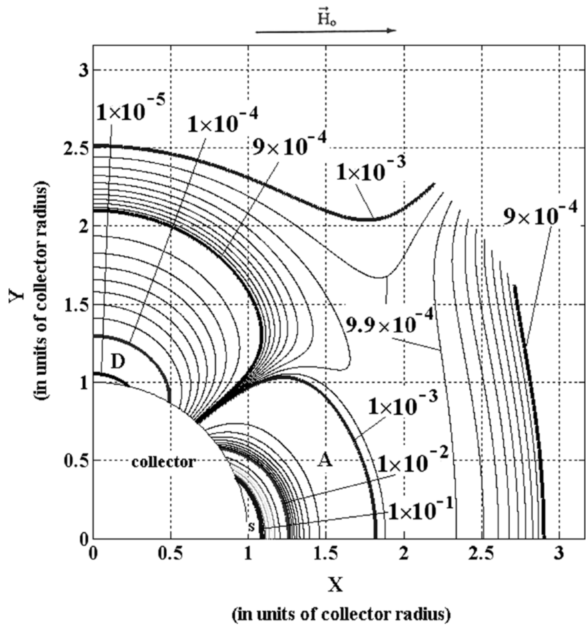

Figure 4. A family of concentration contours around the paramagnetic collector in the representative cell, $P = 10\%$, $G_0 = -17.30$, $\tau = 0.10$.

the saturation, accumulation, and depletion region in these figures are the same.

The concentration contours describing the feature of ultra-fine particle buildup on the single ferromagnetic cylindrical collector have been reported by Davies and Gerber (8). In their simulation, the outer boundary was treated very far from the surface of the collector and the concentration on the outer boundary was fixed equal to the initial value. Then the concentration contour on the outer boundary was like a circle. The concentration contours generated by Davies and Gerber (8) have the same feature as ours in the case of an assemblage of random cylinders with the packing fraction $P = 1\%$ which is the very dilute case. Our contours in Figs. 2, 3, and 4 are compared with the results of Davies and Gerber (8). We find that, in our case of an assemblage of random cylinders, the features of the concentration distribution close to the surface of the representative cylinder in a representative cell are similar to the case of single collector. This is because the value of the packing fraction $P = 5\%$, 8%, and 10% can be considered to be dilute cases. The effect of the neighboring collectors in other composite cells to the features of concentration contours around the collector in a representative cell appears significantly in regions close to the outer boundary of the cell. Unlike the case of the single collector, the contours near the outer boundary are not circular. This is because they are disturbed by the existence of other collectors around the representative collector. This effect is stronger when the packing fraction is larger. In order to investigate the effect of the applied field strength (H_0), we defined a variable, denoted by P_{sat}, as the percent of the volume of particles captured in saturation regions:

$$P_{sat} = \frac{\text{sum of volumes of ultra-fine particles in all saturation regions}}{\text{total volume of ultra-fine particles in the representative cell}} \times 100. \tag{15}$$

Figure 5 shows the comparison of variations of P_{sat} (in paramagnetic mode with all same parameters) with τ in the interval $0 \le \tau \le 0.10$ for two values of H_0. In Fig. 5, we see that the evolution of P_{sat} with τ is increased rapidly with increased H_0. The curve of $H_0 = 1.0 \times 10^6$ A/m is not shown since the value of P_{sat} is equal to zero for total range of τ. The obtained result can be understood by considering Eq. (14). We can see that the factor G_0 is proportional to H_0^2. Consequently, when H_0 is increased, the number of particles captured in saturation regions on the surface of the collector increases rapidly.

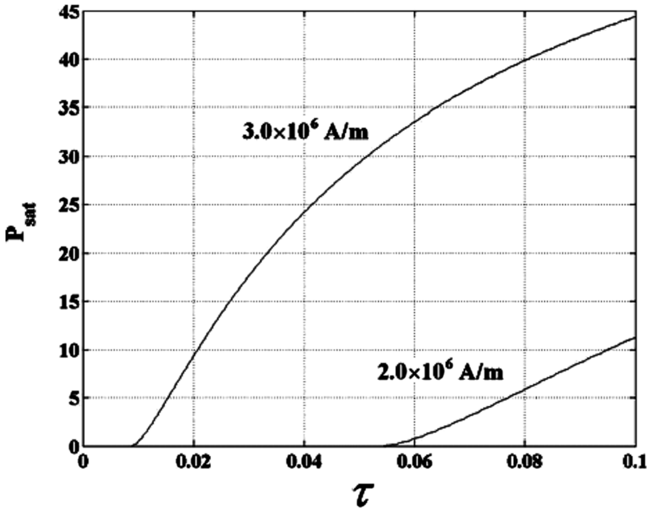

Figure 5. Comparison of variations of P_{sat} with τ between $H_0 = 2.0 \times 10^6$ A/m and 3.0×10^6 A/m.

CONCLUSIONS AND FUTURE RESEARCH

We develop a two-dimensional computational model describing the capture of ultra-fine magnetic particles by an assemblage of randomly distributed parallel cylindrical collectors. We study the effect of collector packing fractions to the features of the build-up of ultra-fine particles on the collector and to the features of particle concentration distribution around the collector.

The results of the simulations show that the effect of the existence of neighboring collectors appears significantly in regions close to the outer boundary of the representative cell. The build-up features of ultra-fine particles in regions close to the collector in a representative cell are similar for three values of packing fractions (5%, 8%, and 10%) and also similar to the case of a single collector in the same mode of the capture (8). The study of ultra-fine particle captured by an assemblage of random cylinders using an effective medium treatment also allow us to predict the fraction of particles captured in the saturation regions on the surface of the collector from the total number of particles in the system. Finally, we study the effect of the strength of the uniform external magnetic field to the number of magnetic particles captured in saturation regions on the surface of the collector at the same intervals of normalized time. The results of the simulation also show that the number of particles captured in saturation regions on the surface of the collector increases rapidly when the strength of the external magnetic field is increased. The model developed in this research is for the capture of ultra-fine particles in a static fluid. In practice, the fluid containing the ultra-fine particles flows with small velocity. Then, the boundary conditions at the outer boundary of the representative cell must be modified. In the case of flowing fluid, the particle volume flux in a perpendicular direction to the outer boundary of the representative cell is not equal zero. The outer boundary conditions can be separated into two types at the upstream and downstream side (8). At the upstream side where ultra-fine particles flow into the representative cell continuously, we can approximate that the concentration on the outer boundary on the upstream side is fixed equal to the initial concentration. At the downstream side, where some of ultra-fine particles flow out of the representative cell while others are captured in the representative cell, we set the boundary condition that the component of the particle flux in the direction parallel to the direction of the flow at that point is constant while the component of particle flux in the direction perpendicular to the direction of the flow can vary. We note that an experiment visualizing the contour lines would be useful to confirm our theoretical model.

ACKNOWLEDGEMENTS

One of the authors (K. H.) would like to thank the University Development Commission on Higher Education, Ministry of Education, Thailand, for the financial support during his graduate study.

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