Three dimensional simulation of negative-magnetophoretic filtration of non-magnetic nanoparticles

Seif-Eddeen K Fateen
Three dimensional simulation of negative-magnetophoretic filtration of non-magnetic nanoparticles

Seif-Eddeen K. Fateen\textsuperscript{a,b,*}, Mahmoud Magdy\textsuperscript{a}

\textsuperscript{a} Department of Chemical Engineering, Cairo University, Giza, Egypt
\textsuperscript{b} Department of Petroleum and Energy Engineering, American University in Cairo, New Cairo, Egypt

\begin{abstract}
Negative magnetophoresis of non-magnetic particles is the induced motion of non-magnetic particles suspended in magnetic media on the application of a magnetic field gradient. Negative magnetophoresis can be used to separate nanoparticles based on their size. An integrated finite-element model was developed using COMSOL to study the transport and separation of nonmagnetic particles in a negative magnetophoresis device. The model solves the magnetic field, fluid flow, and mass transfer equations in three dimensions. The model was used to successfully simulate an experimental separation device and was also used as a tool to develop modified designs that resulted in a substantial enhancement of the separation efficiency. In addition, the model successfully predicted the different phenomena that typically occur in a magnetophoretic device: trapping, focusing, and deflection.
\end{abstract}

\section{Introduction}

Magnetophoresis is successfully used to separate particles based on their magnetic properties. Current application of magnetophoresis for separation of nano-sized particulates include radioactive waste separation (Kaur et al., 2013), arsenate removal from water (Tuutijärvi, 2013), microfluidic bacterial separation from blood (Lee et al., 2014), and many other applications. If the nanoparticles to be separated do not have different magnetic properties, magnetophoresis can also be used by tagging magnetic nanoparticles to target species in the mixture through suitable handles. After tagging, the application of a magnetic field can be utilized for separation. Developing methods for magnetic tagging and separation continues to be an active area of research (Chen et al., 2013; Cerff et al., 2013; Tang et al., 2013; Pospiskova et al., 2013).

If the nanoparticles only differ in their size, their separation can be quite challenging. Negative magnetophoresis, which is the motion of non-magnetic particles immersed in a magnetic fluid resulting from the application of a magnetic field gradient, can be used to achieve such separation (Fateen, 2002). In negative magnetophoresis, the difference in magnetic susceptibility between the particles and the surrounding medium is negative as opposed to positive magnetophoresis in which the difference is positive. This phenomenon is analogous to buoyancy of objects that are less dense than the surrounding fluid. In the case of buoyancy, gravity acts on the more dense fluid stronger than it acts on the lighter object, causing this object to experience a hydrostatic force opposite to the usual direction of gravity. Similarly, in the case of negative magnetophoresis, the magnetic field acts on the magnetized fluid surrounding the non-magnetic particle, pushing the particle in the opposite direction. This concept can be implemented as a size classification tool that can be used to separate nano-sized particles that are difficult to separate otherwise (Kose et al., 2009).

\* Corresponding author at: Department of Petroleum and Energy Engineering, American University in Cairo, PO Box 74, New Cairo 11835, Egypt. Tel.: +202 2615 1000.
E-mail addresses: sfateen@alum.mit.edu, Fateen@eng1.cu.edu.eg (S.-E.K. Fateen).
http://dx.doi.org/10.1016/j.cherd.2015.01.007
0263-8762 © 2015 The Institution of Chemical Engineers. Published by Elsevier B.V. All rights reserved.
### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Meaning</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a_1$</td>
<td>radius of the non-magnetic particle</td>
</tr>
<tr>
<td>$B$</td>
<td>magnetic flux density</td>
</tr>
<tr>
<td>$C$</td>
<td>total concentration of the fluid</td>
</tr>
<tr>
<td>$D_{12}$</td>
<td>diffusivity of the non-magnetic particles in the magnetic field</td>
</tr>
<tr>
<td>$D_p$</td>
<td>diameter of the spherical particle</td>
</tr>
<tr>
<td>$F$</td>
<td>applied force</td>
</tr>
<tr>
<td>$F_m$</td>
<td>magnetic force</td>
</tr>
<tr>
<td>$F_d$</td>
<td>drag force</td>
</tr>
<tr>
<td>$H$</td>
<td>magnetic field</td>
</tr>
<tr>
<td>$i$</td>
<td>index for the components in the mixture: 1 for non-magnetic particles and 2 for the magnetic fluid</td>
</tr>
<tr>
<td>$I_p$</td>
<td>molar flux of the non-magnetic particles relative to the mass average velocity $v$</td>
</tr>
<tr>
<td>$k$</td>
<td>Boltzmann constant</td>
</tr>
<tr>
<td>$k^{-1}$</td>
<td>Debye length</td>
</tr>
<tr>
<td>$M$</td>
<td>magnetization</td>
</tr>
<tr>
<td>$M_s$</td>
<td>saturation magnetization</td>
</tr>
<tr>
<td>$N_A$</td>
<td>Avogadro’s number</td>
</tr>
<tr>
<td>$p$</td>
<td>pressure field</td>
</tr>
<tr>
<td>$R$</td>
<td>universal gas constant</td>
</tr>
<tr>
<td>$R_p$</td>
<td>hydrodynamic radius of the non-magnetic particles</td>
</tr>
<tr>
<td>$u$</td>
<td>fluid velocity</td>
</tr>
<tr>
<td>$\mathbf{u}_f$</td>
<td>velocity vector of the magnetic fluid</td>
</tr>
<tr>
<td>$\mathbf{u}_p$</td>
<td>velocity vector of the non-magnetic particle</td>
</tr>
<tr>
<td>$\mathbf{v}$</td>
<td>mass average velocity</td>
</tr>
<tr>
<td>$V$</td>
<td>volume of the particle</td>
</tr>
<tr>
<td>$V_m$</td>
<td>magnetic scalar potential</td>
</tr>
<tr>
<td>$\rho_i$</td>
<td>weight fraction of component $i$</td>
</tr>
<tr>
<td>$\rho$</td>
<td>density of the fluid</td>
</tr>
<tr>
<td>$\rho_p$</td>
<td>density of the particle</td>
</tr>
</tbody>
</table>

### Greek Letters

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Meaning</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon$</td>
<td>electric permittivity of the fluid</td>
</tr>
<tr>
<td>$\psi_0$</td>
<td>electric potential</td>
</tr>
<tr>
<td>$\psi_0$</td>
<td>electric potential at the surface of the non-magnetic particle</td>
</tr>
<tr>
<td>$\mu$</td>
<td>fluid viscosity</td>
</tr>
<tr>
<td>$\mu_o$</td>
<td>permeability of vacuum</td>
</tr>
<tr>
<td>$\mu_r$</td>
<td>relative permeability</td>
</tr>
<tr>
<td>$\phi_i$</td>
<td>volume fraction of component $i$</td>
</tr>
<tr>
<td>$\eta$</td>
<td>fluid viscosity</td>
</tr>
<tr>
<td>$\rho$</td>
<td>density of the fluid</td>
</tr>
<tr>
<td>$\rho_p$</td>
<td>density of the particle</td>
</tr>
</tbody>
</table>

Annavarapu (Annavarapu, 2010) designed a separation system composed of a regular array of iron obstacle posts which utilized magnetic buoyancy forces to perform size-based separation.

Mao and his coworkers (Cheng et al., 2013; Zhu et al., 2010) and Xuan and his coworkers (Liang et al., 2011; Liang and Xuan, 2012a) experimentally studied the motion of non-magnetic particles immersed in magnetic fluids as they flow through a microchannel with the application of a non-uniform magnetic field. Negative magnetophoresis has been successfully used in multilaminar flow to manipulate particles suspended in magnetic fluids (Tarn et al., 2014). Recently, Zhu et al. (Zhu, in press) combined both positive and negative magnetophoresis to separate particles with different magnetic properties.

Different approaches can be used to model the phenomenon of negative magnetophoresis: continuum modeling, Brownian-dynamics simulation and particle trajectory modeling. Fateen (Fateen, 2002) has proposed a one-dimensional continuum transport model that contained no adjustable parameters to represent the concentration profile of the nonmagnetic particles suspended in the magnetic fluid. To represent the available experimental data, the model utilized one-dimensional conservation equation for mass and accounted for the magnetic and electrostatic repulsion forces. The magnetic field profile was measured, fitted to a polynomial and subsequently used in the model. Sharpe (Sharpe, 2004) used the continuum model approach to solve a two-dimensional transport equation for the particle concentration.

The magnetic field was generated by solving Maxwell equations for the magnetic field separately and fitting the results to a polynomial, which was then used as input for the transport model. The 2-D results successfully simulated the experimental separation results. Gonzalez (Gonzalez, 2009) used both the continuum model and the Brownian simulation approaches to model his experiments. The continuum model, which contained no adjustable parameters, was able to predict the experimental concentration profiles reasonably well over the range of experimental conditions used. The Brownian simulation was only able to semi-quantitatively predict the experimental results. Fateen and Magdy (Fateen and Magdy, 2010) used COMSOL to solve a two-dimensional continuum model for a negative magnetophoretic separation device.

The particle trajectories approach has been used by Annavarapu (Annavarapu, 2010) to describe the behavior of a single non-magnetic particle in regions of inhomogeneous magnetic field gradients. The trajectory simulations was used to conceptualize and built an experimental setup to perform the required separation. The particle trajectories approach has been also used by Mao and his co-workers (Cheng et al., 2013; Zhu et al., 2010; Liang et al., 2011; Zhu et al., 2011a) and by Zhu (Zhu and Nguyen, 2012) to develop a 2-D and 3-D model for transport of non-magnetic spherical microparticles in magnetic fluids in a microfluidic system that consists of a microchannel and a permanent magnet. The experimental results were compared to the results obtained by the analytical model and reasonable agreement was obtained.

In all the above modeling attempts, the magnetic field was obtained independent of the modeling environment. Any change in the configuration of the permanent magnets would require re-evaluation of the magnetic field through its measurement or through separate solution of Maxwell equation followed by polynomial fitting of the solution. Both approaches have limitations. The aim of this study is to develop an integrated continuum model that captures the multi-physics
associated with the negative magnetophoresis phenomena. To the best of authors’ knowledge, the published literature does not include any continuum model of the negative magnetophoresis phenomenon that solves the flow, transport and magnetic field equation together in three dimensions. In this study, COMSOL was the platform of choice for developing a simulation tool that takes into account the rigorous modeling of the fluid flow, diffusional, convective and magnetic transport, and the magnetic field. The developed tool was then used to simulate the experiment performed by Sharpe (Sharpe, 2004), explain its results, propose an improvement by changing the device configuration and simulate the separation capabilities of new devices.

The significance of this work is that the tool developed would allow the prediction of the performance of separation devices before manufacturing them. The simulation acts as a valuable design tool that can be used to optimize the configuration of permanent magnets and the flow pattern to yield the best possible separation of the nanoparticle mixture.

The remainder of this paper is organized as follows. Section 2 outlines the modeling framework used in this study. The theory of magnetophoresis and the different forces acting on the moving particle are introduced. The section also includes the governing equations used in the model, the implementation of the model to a separation device and the solution procedure. The results of the simulation are presented in Section 3 along with a discussion of the results. The conclusions of this work are summarized in Section 4.

2. Modeling framework

In this work, the systems under study are composed of magnetic elements, bulk magnetic fluid and nano-sized particles immersed in the bulk fluid. In this section, the theoretical framework used in modeling the system is presented and discussed.

2.1. Continuum modeling

The bulk fluid used in this work is magnetic fluid, which is a colloidal suspension of nanoparticles, each of which contains a magnetite core surrounded by a polymer shell, which acts as a stabilizer to keep the nanoparticles suspended in water. The average size of the core magnetic nanoparticles is 5–10 nm, while the size of the entire particle with the stabilizing polymer shell is up to 32 nm ( Fateen, 2002). Since the use of the continuum models is valid when the nonmagnetic particles are at least 2–3 times larger than the magnetic nanoparticles ( Erb and Yellen, 2008), the magnetic fluid was therefore treated as a continuum when compared to the nonmagnetic particles separated during the magnetophoresis process. In cases when the continuum model fails, particle trajectory modeling (Annavarapu, 2010; Cheng et al., 2013; Liang et al., 2011; Berthier et al., 2001) can be pursued. This study uses the continuum model approach.

2.2. Forces acting on the non-magnetic particles

The particle transport in magnetic fluids under an external magnetic field is governed by various forces and interactions including magnetic buoyancy force, hydrodynamic viscous drag force, gravity, buoyancy force, particle–particle interaction (van der Waals force, electrostatic force) and, diffusion due to Brownian motion. These forces depend on the size of the system, strength of the permanent magnets, the concentration of the particles in the bulk fluids, and size of the particles immersed on the bulk fluid. Gravitational and buoyancy forces are insignificant for nanoparticles and can be ignored as they are usually much weaker than the magnetic force.

2.2.1. Magnetic force

The magnetic force on non-magnetic objects located in a magnetic fluid under the application of an external magnetic field can be written as

\[ F_n = \mu_0 \nabla \times \mathbf{M} \]  

(1)

The product \( \nabla \times \mathbf{M} \) can be interpreted as the magnetic moment of the medium volume displaced by the non-magnetic particle. The above equation is similar to the force on a magnetic point dipole located in a non-magnetic medium, with the exception of the negative sign.

The field distribution and the magnetic response of the particle (i.e., \( \mathbf{M} \) vs \( \mathbf{H} \)) must be known to calculate the magnetic force. The magnetic field distribution is discussed later. There are rigorous models for the magnetization of the particle that take into account self-demagnetization and magnetic saturation. In this study, since the linear relation between the particle magnetization and the magnetic field strength is a valid assumption, the magnetization was simply described by

\[ \mathbf{M} = \left( \frac{M_s}{H_s + H} \right) \mathbf{H} \]  

(2)

2.2.2. Hydrodynamic viscous drag force

When a non-magnetic particle is driven by the negative magnetophoresis force and moves in magnetic fluids, it is affected by flow resistance from the magnetic fluids, which is the hydrodynamic drag force. The simplest form of this force can be obtained for a spherical particle and is called the Stokes’ drag formula,

\[ F_d = 3\pi \eta D_p (u_t - u_p) \]  

(3)

This formula applies to a single isolated particle in an infinite uniform flow field.

The drag force can be evaluated if the fluid velocity, \( u_t \) is obtained via the solution of the flow equation according to the configuration of the separation device, the fluid properties and the flow regime.

2.2.3. Particle–particle interaction

The particle–particle interaction (inter-particle interaction) consists of mainly three forms (a) van der Waals force, (b) electrostatic repulsion, and (c) magnetic dipole–dipole interactions.

Van der Waals forces are the strongest interaction forces affecting colloidal particles at short ranges. They are mostly attractive forces that arise due to thermal fluctuations of dipoles, and lead to strong aggregation when particles approach each other to a fraction of their diameter. The van der Waals forces were not considered in this model due to the relatively large size of the non-magnetic particles, upon which electrostatic repulsion applies.

Electrostatic repulsion prevents the colloidal particles from forming stable aggregates due the attractive van der Waals
forces, and renders the colloidal suspensions stable. When colloidal species are suspended in water, they generally acquire a net surface charge, which depends on the chemical nature of their surface. Electrostatic effects are not sufficient to make a system thermodynamically stable, as they cannot fully counteract van der Waals forces at very short particle separations, but they do slow down aggregation. The electrostatic force in this work was based on a DLVO-type model (Derjaguin–Landau–Verwey–Overbeek) and was derived following the work of others (Fateen, 2002; Sharpe, 2004) to obtain

$$\psi^2 = 16\pi^2 e^2 R^2 k_0^2 (1 - 2k_0 R) N_A^2$$

(4)

In this study, a term was added a term in the molar flux equation to account for the electrostatic repulsion. The flux due to the electrostatic force is related to the particle concentration, the particle concentration gradient and the electrostatic force described in Eq. (4) (Fateen, 2002; Sharpe, 2004).

Magnetic particles suspended in magnetic fluids exhibit magnetic dipole–dipole interactions. Since the size of the magnetic particles (~5–10 nm) in a typical magnetic fluid are much smaller than the size of non-magnetic particles (>100 nm), the magnetic fluid can be treated as a continuum. This type of interaction is not applicable to the non-magnetic particles.

2.3. Governing equations

2.3.1. Fluid flow

The velocity profile of the magnetic fluid inside the flow conduit has a major impact on the transport of the non-magnetic particles. The flow profile itself can be one of the driving forces of the separation or the manipulation of the particles. For simple flow configurations, the flow profile can be obtained analytically and used as input in the solution of the particle transport equation. When the analytical solution is not possible and when the purpose of the simulation is to allow for studying the effect of the configuration on the efficiency of the separation process, the solution of the complete flow equation together with the solution of the particle transport and magnetic field equations provides flexibility and versatility. The governing equation for the steady-state flow and the continuity equation are

$$\mu (\nabla u) = \nabla \left[ -p I + \mu (\nabla u + (\nabla u)^T) - \frac{2}{3} \mu (\nabla u) I \right] + F$$

(5)

$$\nabla \cdot (\rho u) = 0$$

(6)

2.3.2. Particle transport

The governing equation for the steady-state concentration profile, $C_p$, of the nonmagnetic particles immersed in magnetic fluid continuum is

$$\nabla C_p + \nabla \rho = 0$$

(7)

Different approaches exist in the literature (Fateen, 2002; Gonzalez, 2009) to derive the molar flux; Balancing the forces acting on the particles of interest yields

$$J_p = \frac{D_p C_p}{kT} \left[ \frac{R T \nabla C_p + C_p \nabla T C_p + C_p \nabla \rho (1 - \frac{\rho P}{\rho}) + C_p \mu c_0 \nabla \nabla \rho} \right]$$

(8)

The above equation represents the molar flux $J_p$ of the non-magnetic particles immersed in magnetic fluid suspension. The first term represents the diffusion driven by the concentration gradient. The second term represents the flux imposed by the electrostatic repulsion. The third term represents gravitational force, while the last term represents the magnetic force. $D_{12}$ can be calculated from the Stokes-Einstein relationship for the diffusion coefficient

$$D_{12} = \frac{k T}{6 \pi \eta A}$$

(9)

2.3.3. Magnetic field

The magnetic field can be generated by solving the Maxwell equation for the permanent magnet configuration. In general, numerical techniques are used to solve for the magnetic fields of permanent magnets. Analytical solutions can be used for simple setups of the permanent magnets. In this case the analytical solution is preferred to avoid the numerical noise generated by the numerical solutions. These numerical noises can have sensible effects on the calculation of the magnetic force that depends on the magnetic field gradient.

However, analytical solutions cannot be obtained for complex magnets configuration designs. The problem of the numerical noise can be solved by adjusting the grid size in the simulation software. The two equations that govern the magnetic field are

$$\nabla B = 0$$

(10)

$$H = -\nabla \rho$$

(11)

The Magnetization relation $B = \mu_0 (H + M)$ was used for the four magnets blocks. The relative permeability relation $B = \mu_i H$ was used otherwise.

2.4. Model assumptions

The assumptions used to develop the model described in the previous sections are valid for the simulation cases in this study. These assumptions are summarized below:

- The magnetic fluid is a continuum when compared to the non-magnetic particles
- Constant magnetic fluid density, viscosity and concentration
- Constant temperature, or temperature far from the Curie temperature
- The magnetization of the magnetic fluid is collinear with the magnetic field
- Negligible electrical conductivity of the magnetic fluid
- A total magnetization of the magnetic fluid that is much less than the magnetic field
- Low concentration of the non-magnetic particles in the fluid
- The non-magnetic particles are spherical
- The non-magnetic particles have nearly zero magnetization in the presence of a magnetic field
- Nearly constant $H$ and $VH$ through the non-magnetic particle volume
- The presence of the non-magnetic particles does not distort the magnetic field lines
- Migration of the charged non-magnetic particles is slow enough not to induce any significant fields in the magnetic fluid
Table 1 – Boundary conditions for the base case.

<table>
<thead>
<tr>
<th>Equation</th>
<th>Boundary conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Magnetic flux</td>
<td>The magnetic insulation boundary condition was used for all the boundary to represent the zero magnetic flux density at all boundaries; ( n \mathbf{B} = 0 ) M was the same and assumed to equal 499,905 A/m because this is the magnetization value of the Neodymium Iron Boron 40 MGoe permanent magnets used in the experiments.</td>
</tr>
<tr>
<td>Fluid flow</td>
<td>Constant velocity at the inlet ( u = u_{\text{inlet}} = 4.423 \times 10^{-5} ) m/s. Constant pressure at the outlet ( P = 1 ) atm; No slip conditions at the walls of the Cylinder ( V = 0 ).</td>
</tr>
<tr>
<td>Particle transport</td>
<td>At the inlet boundary ( z = 0 ), ( C_p = C_{p0} ). At the outlet boundary ( z = L ), Convective flux boundary condition was selected. Symmetry boundary condition was used at the axis ( (r = 0, \frac{\partial}{\partial r} \mathbf{v} = 0) ) and insulation at the wall ( (r = R_0, \frac{\partial}{\partial r} \mathbf{v} = 0) ).</td>
</tr>
</tbody>
</table>

- The volume exclusion contribution is considerably smaller than the electrostatic contribution for the charged nonmagnetic particles.
- The double-layer theory is valid for the charged nonmagnetic particles.

2.5. Model implementation

2.5.1. Base case configuration

The set of governing equations presented in the previous section was solved for an actual case study, for which the experimental data were available. The simulation was performed using COMSOL 4.1, which numerically solved the partial differential equations system using finite elements. The base case used was that of Sharpe’s separation experiments (Sharpe, 2004). Four magnets blocks were arranged in a cross shape equidistant from one another, with the cylindrical column placed in the middle of the four magnets. The size of each magnet was 18 cm long \( \times \) 18 cm wide \( \times \) 18 cm thick. The overall configuration is shown schematically in Fig. 1. The purpose of the small inner cylinder was to collect the nonmagnetic particles after they were focused at the axis of the flow. The diameter of the small cylinder is 0.25 cm and the height of the inner cylinder is 2 cm. The configuration of the device as drawn to scale in COMSOL is shown in Fig. 2.

The entire device was inserted inside a large cylinder, in which the magnetic flux equation was discretized over a small enough mesh to result in a smooth magnetic flux around the magnets and inside the cylinders.

2.5.2. Boundary conditions

The boundary conditions for the three governing equations are given in Table 1.

2.5.3. COMSOL implementation

The mesh size used for the magnetic field model was large for the big cylinder and small for the magnets configuration and the magnetophoresis cylinder. The solver used was GMRES. A stationary analysis was made since the flow is at steady state. Meshing was adjusted to the Fine option after a sensitivity study was performed to understand the impact of the mesh size on the results. The adaptive mesh refinement option was chosen. Quadratic discretization was used for the magnetic scalar potential, while linear discretization was used for the velocity and the concentration.

The selected COMSOL physics models were Magnetic Fields (No Currents), Laminar Flow, and Transport of Diluted Species. The equations used for the Transport of Diluted Species model in COMSOL were modified to account for the inclusion of the magnetic and electrostatic repulsion terms. The output of the magnetic field simulation was taken as input for the transport simulation through the parameters \( (H, M, \frac{\partial H}{\partial t}, \mathbf{V}, \mathbf{F}) \); a one-way coupling was established between the two physics interfaces. The magnetic field simulation was performed first and then the results obtained from this simulation were used by the particles transport simulation to solve for the nonmagnetic particles concentration profile. The output of the laminar flow model was also used as input for the transport model. The crosswind diffusion parameter was adjusted to the value of 0.001 below which the solver did not converge.
3. Results and discussion

3.1. Base case

The magnetic flux density was generated for the quadruple magnetic configuration via the solution of the magnetic flux governing equations on a large cylindrically-shaped “air box”, with the separation device placed at its centerline. The resultant profile of the magnetic flux close to the permanent magnets is shown in Fig. 3. The solution of the magnetic flux equation allows the calculation of the magnetic field gradient inside the flow tube. This calculation was performed in each of the three dimensions using special differentiation function in COMSOL. With this analysis, three-dimensional effects, such as the entrance of the tube, can be better understood. Such understanding would not have been possible with a polynomial fitting of the magnetic field in a typical flow cross section with the axial symmetry assumption.

The fluid flow equations and particle transport model were solved and the velocity profile (not shown) was typical for the flow inside a cylinder. The results of the particle transport model are shown in Fig. 4, which shows the normalized concentration profile obtained by dividing the actual concentration by the initial concentration \( C_{po} \). The Concentration profile showed a peak concentration at the center of the column due to the induced movement of the particles towards the center of the column. The concentration profile is similar in profile, but slightly higher than the concentration profile obtained from a two-dimensional model solved previously (Fateen and Magdy, 2010), as shown in Fig. 5, which is due to a slight difference in the magnetic field gradients along the cylinder radial direction.
The effect of changing model parameters was investigated in order to gain a better understanding of the negative magnetophoresis process. The effect of changing the feed mixture flow rate is plotted in Fig. 6, which shows the change of the separation efficiency in the magnetophoresis device in response to the change of the feed mixture flow rate. The separation efficiency, calculated as the ratio of the nonmagnetic particles collected in the central outlet to the inlet flow rates of the nonmagnetic particles, decreases with increasing the flow rate of the feed mixture for both 1 and 2 μm nonmagnetic particles diameters. At higher flow rates, the hydrodynamic drag force overcomes the magnetophoresis process resulting in poor separation. Higher magnetic force increases the separation efficiency, while higher drag force decreases the separation efficiency. In addition, the retention time of the particles in the column is higher at low flow rates resulting in better separation.

The model results for 1-micron particle matches the experimental data obtained by Sharpe (Sharpe, 2004), which validate the effect of the flow rate on the separation efficiency. For the 2-micron particles, the same effect was also observed both experimentally and from simulation. At constant flow rate, the separation efficiency of the 2-micron particles is higher than the efficiency of the 1-micron particles. Also the peak concentration value for the 2-micron particles is higher than for the 1-micron particles. The magnetophoresis force is proportional to the cube of the particle diameter, whereas the hydrodynamic drag force is proportional to the square of the diameter. Thus, larger particles will always be separated more efficiently using this separation device.

The computer simulations predicted higher value of magnetic separation for the 2-micron particles, which suggests that the model fails to offer predictions that match the experimental data at this relatively larger particle size. The failure of the model to accurately predict the relatively low separation efficiency for the larger particles is due to the over-estimation of the particle concentration at the center line. Particle–particle interaction, including magnetic dipole moments, play a larger role at this level of high particle concentration. Better prediction requires the inclusion of this type of particle–particle interaction. However, the purpose of the model is to predict the separation of submicron particles, the current model formulation provides good predictions of the experimental results in size range of 1 μm or less without the use of any adjustable parameters.

The effect of initial nonmagnetic particle concentration in the feed mixture was studied by performing the simulation at 1 and 2 wt% initial particle concentration. All simulations were performed for 1 μm particles diameter. The results of simulation showed, as presented in Fig. 7, the negative effect of increasing the nonmagnetic particles concentration at low flow rate. This negative effect can be attributed to the increase of the electrostatic repulsion force at higher particles concentrations, which reduced the separation efficiency. At higher flow rates, the balance between the repulsion, magnetic, and drag forces shifts towards the drag force resulting for the same separation efficiency for the two concentrations.

3.2. Entrance Effect

The investigation of the entrance effect in the magnetophoretic device was made possible by the inclusion of the three dimensional magnetic field flux and its coupling with the particle transport model. A close graphical view at the entrance of the cylinder is shown in Fig. 8. The concentration increased at the edges but upstream of the cylinder due to magnetic force resulting from the shape of the magnetic field.
gradient upstream of the separation cylinder. The increase in concentration was weak (about 1.5), but had a substantial effect at low flow rates. The effect of the axial force concentrated the particles before entering the separation zone of the cylinder and hindered them from flowing into the separation zone. This entrance effect was observed experimentally (Sharpe, 2004) and is a further validation of the model.

### 3.3. Design modification

The separation efficiency of the quadruple magnetophoretic separator simulated was less than 50% for the one micron particle at lower mixture flow rates and was even lower for higher flow rates. The separation efficiency is lower for smaller particles. A small modification of the device configuration can lead to higher separation efficiency. The simulation of the base case described above showed that higher separation efficiency can be obtained by increasing the magnetic force or decreasing the drag force on the particles or both. The option of using stronger magnets is always available. The developed model allows for the investigation of more refined options. In this work, we used the model to investigate the division of the separation cylinder into two separation stages.

The original cylinder was divided into three smaller cylinders: the primary separation cylinder, the medium cylinder and the secondary separation cylinder. Several simulation runs were implemented to optimize the diameter of the medium cylinder and the height of the primary and secondary separation cylinders. The final dimensions of the optimized device are represented in Fig. 9.

The modified design was able to achieve an increase of separation efficiency of from 60% to 150% for range of flow rate 10–80 ml/h, 200% for 100 ml/h and more than 150% for flow rates up to 200 ml/h. In fact, at high flow rates, the separation efficiency of the original design was quite low, while it exceeded 40% for the modified design. Thus, the model validated the idea that the mere division of the separation device into two stages significantly improved the separation.

### 3.4. Simulation capabilities

The integrated model was developed in order to facilitate the design and simulation of the magnetophoresis devices. In this section, the results of the application of the model on three possible magnetophoresis processes are presented. The three applications, trapping, focusing, and deflection, were simulated using the integrated model to test the model capabilities to predict the behavior of the nonmagnetic particles in each method. These applications were chosen due to the availability of their experimental validation. For example, Xuan and his co-workers (Liang et al., 2011; Liang and Xuan, 2012a; Liang and Xuan, 2012b; Zhu et al., 2012) used deflection to focus particles in microchannels. Pamme and her co-workers used deflection (Tarn et al., 2014; Vojtisek et al., 2012) and trapping (Tarn et al., 2015) to separate particles. Zhu et al. (Zhu et al., 2011b) demonstrated focusing of particles using two permanent magnets.

The three magnetophoretic devices are illustrated in Fig. 10. The flow cylinder had a 1-cm radius in all three simulations. The device geometry used for simulating the trapping of non-magnetic particles consisted of two magnet blocks ($18 \times 6 \times 8$ cm) with the cylinder inserted between them. The
device configuration for the focusing experiment consisted of two magnet blocks (4 × 4 × 4 cm) and the flow cylinder inserted between them. The device configuration for the deflection experiment consisted of one magnet block (2 × 4 × 4 cm) placed next to the flow cylinder.

The results of the three simulations are compiled in Fig. 11, which shows the profile for the normalized concentration. For the trapping simulation, the peak concentration of the nonmagnetic particles occurred upstream of the magnets. The peak concentration of the nonmagnetic particles for the focusing setup occurred in the centerline of the cylinder. For the deflection setup, the peak concentration of the nonmagnetic particles occurred at the bottom of the cylinder. The integrated model was able to simulate the physical phenomena that were expected theoretically and observed experimentally (Peyman et al., 2009).

4. Conclusions

The overall goal of this work was to develop a three-dimensional integrated model able to simulate the magnetophoresis of the nonmagnetic particles suspended in magnetic fluid solutions. The integrated model was intended to include magnetic field, fluid flow, and particle transport physics in one simulation tool. The model was implemented using COMSOL to solve the magnetic field, fluid flow, and particle transport partial differential equations. A quadruple magnetophoretic separation device was simulated with this mode and the separation efficiencies obtained simulation results were comparable to those obtained experimentally for the 1-micron particle. The model failed to predict the separation efficiency for larger particle sizes due to the questionable validity of some of the model assumptions at larger particle sizes range.

The model was used to study the effect of the operating parameters on the performance of the magnetophoretic device to define the best operating range for the device. It was used to perform design modifications on the magnetophoretic device in order to enhance the separation efficiency of the nonmagnetic particles. A change was made to the dimensions of the device to lower the drag force to increase the effect of the magnetic force and magnetophoresis increased the separation efficiency by 200% for certain flow rate and by 100% for the most of the operating flow rate range.

The developed model was also successfully used to predict the different phenomena that typically occur in a magnetophoretic device: trapping, focusing, and deflection. The model can now be used as a design tool before actual magnetophoresis experiments are to be performed, which would lead in savings in research time and cost.

References


