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Spreading of a ferrofluid core in three-stream micromixer channels
Zhaomeng Wang, V. B. Varma, Huan Ming Xia, Z. P. Wang, and R. V. Ramanujan

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Spreading of a ferrofluid core in three-stream micromixer channels

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Spreading of a water based ferrofluid core, cladded by a diamagnetic fluid, in three-stream micromixer channels was studied. This spreading, induced by an external magnetic field, is known as magnetofluidic spreading (MFS). MFS is useful for various novel applications where control of fluid-fluid interface is desired, such as micromixers or micro-chemical reactors. However, fundamental aspects of MFS are still unclear, and a model without correction factors is lacking. Hence, in this work, both experimental and numerical analyses were undertaken to study MFS. We show that MFS increased for higher applied magnetic fields, slower flow speed of both fluids, smaller flow rate of ferrofluid relative to cladding, and higher initial magnetic particle concentration. Spreading, mainly due to connective diffusion, was observed mostly near the channel walls. Our multi-physics model, which combines magnetic and fluidic analyses, showed, for the first time, excellent agreement between theory and experiment. These results can be useful for lab-on-a-chip devices. © 2015 AIP Publishing LLC.

I. INTRODUCTION

Micro magnetofluidics (MMF), a newly established research area, combines magnetism and microfluidics. Ferrofluids consist of a stable colloidal dispersion of magnetic particles in a carrier fluid. In MMF, actuation and manipulation of ferrofluids by an external magnetic field can provide an inexpensive and convenient platform to wirelessly control and manipulate fluid flow at the microscale. These capabilities of MMF can be utilized in Lab-On-A-Chip (LOC) devices, e.g., in micro-fluid valves or micropumps,2,3 as well as the separation and extraction of biological particles.4,5 Other applications include separation of diamagnetic particles,6,7 rotating seals,8 loudspeakers,9 and microlens arrays.10,11 Usually, permanent magnets12–14 or electromagnets15,16 are placed adjacent to LOC devices to produce a magnetic field gradient. Alternatively, micro magnets are directly fabricated and embedded in LOC chips by micro fabrication techniques (e.g., sputter deposition and soft-lithography) to produce magnetic (e.g., Ni) thin films with the required geometry.17,18 If multiple magnets are required on a single chip, their interactions will result in complex magnetic fields. In contrast to such complex magnetic field distributions, it will be very advantageous if a simple uniform magnetic field can be used to modulate ferrofluid motion. This can be achieved by magnetofluidic spreading (MFS).

The spreading of an oil based ferrofluid into a diamagnetic solution (mineral oil) under uniform magnetic fields has been previously studied,19 but poor match between experiments and numerical results was obtained and the mechanisms governing MFS were not reported. Here, a three-stream flow system, with a water based ferrofluid core cladded by a diamagnetic buffer fluid, was used to experimentally and numerically study MFS of the ferrofluid core under the influence of a uniform...
magnetic field. Excellent agreement was obtained between the experiments and our modelling results without any correction factor. The factors that govern MFS were also identified for a deeper understanding of MFS.

A. Methodology

The microfluidic chip was made from poly(methyl methacrylate) (PMMA) by standard micro milling followed by thermal bonding. The test channel (Figure 1, top-left inset) inside the chip consists of three inlets and two exits, with length, width, and height of 2500, 200, and 30 µm, respectively. Here, the channel dimensions are for the channel where the three streams mix, starting from the junction of the three inlet channels to the outlet. The water based ferrofluid was delivered into the middle inlet as the core stream. The glycerol-water mixture was delivered into the channel through the two side inlets as cladding streams. An electromagnet (DEXING MAGNET, DXSB-178) was used to generate a uniform magnetic field within the 5 cm air gap where the chip was placed. A high speed camera (MIRO, M320S) was used to monitor MFS. The images were tuned to a gray color, and the width fraction (WF) was measured based on the gray level. The properties of the fluid will be presented later.

The modelling was performed using COMSOL Multiphysics (normal mesh, fluid dynamics). The mesh in the z direction was enhanced by 6 times compared to the other directions. The minimum and average element qualities were $1.163 \times 10^{-5}$ and $0.02544$, respectively.

B. Geometry

A 3D model was developed, consisting of a long rectangular channel, with length, width, and height of 2500, 200, and 30 µm in the x, y, and z directions, respectively (Figure 1). The coordinate origin was set at the middle of its core stream inlet surface. The width of the inlet for the ferrofluid (middle section of the inlet surface in Figure 1) core stream was set to be 44 µm.

C. Materials

The water based ferrofluid (EMG707, FerroTec) has a density ($\rho_f$) of 1100 kg/m$^3$, black-brown appearance, dynamic viscosity ($\eta_f$) of 3 mPa s, saturation magnetization of 11 mT, initial volume concentration of $i = 2\%$, and magnetic susceptibility ($\chi_f$) of 0.12 in CGS units (1.51 in SI units). The magnetic particles have an average diameter $d_p = 2r_p = 10$ nm and a density of $\rho_p = 5240$ kg/m$^3$.

A glycerol-water mixture was used as the diamagnetic cladding fluid. This mixture has a glycerol volume concentration of 60%, density of $\rho_{diam} = 1169.4$ kg/m$^3$, and dynamic viscosity ($\eta_{diam}$) of 12.5 mPa s. The magnetic susceptibility of this glycerol-water mixture was calculated as $-9.5 \times 10^{-6}$, indicating a typical diamagnetic material.
The local density \( \rho_{\text{mix}} \) of the mixed flow is given by
\[
\rho_{\text{mix}} = c \rho_{\text{ff}} + (1 - c) \rho_{\text{diam}},
\]
where \( c \) is a local dimensionless concentration: \( c = l/i \), and \( l \) is the local volume concentration of magnetic particles in the ferrofluid and glycerol-water mixture. For initial inlet fluids, \( c \) is set to be 1 and 0 for ferrofluid and glycerol-water mixture, respectively.

The local viscosity \( \eta_{\text{mix}} \) of the mixed solutions was determined using a Grunberg-Nissan model for a binary mixture,
\[
\ln \eta_{\text{mix}} = c \cdot \ln \eta_{\text{ff}} + (1 - c) \cdot \ln \eta_{\text{diam}} + c \cdot (1 - c) \cdot G_{12},
\]
where \( G_{12} \) is an interaction parameter of two fluids, depending on the composition of the mixture and temperature. Based on experimental values, \( G_{12} \) was fit to the value of \(-1.44\).

At channel inlets, an inlet flow rate \( (FR) \) of ferrofluid core of 0.05 ml/h was selected. A flow rate ratio \( (Q_{\text{ratio}}) \) of the flow rate of the diamagnetic cladding fluid to the flow rate of the ferrofluid core of 4 was utilized.

II. GOVERNING EQUATIONS

A. Fluid flow equation

The magnetic particles are in the nanometric size range. A continuum approximation was utilized in our model, since particles undergo continuous motion within the length and time scales of the flow. The Navier-Stokes equation (Eq. (3)) describes the behavior of incompressible and viscous laminar fluids flowing inside the channel,
\[
\frac{\partial}{\partial t}(\rho_{\text{mix}}u) + u \cdot \nabla(\rho_{\text{mix}}u) = -\nabla p + \nabla \cdot [\eta_{\text{mix}}(\nabla u + \nabla u^T)] + F_f
\]
where \( \rho_{\text{mix}}, u, p, \eta_{\text{mix}}, \) and \( F_f \) denote the local density of the mixed flow, flow velocity vector \( (u, v, w) \), pressure, fluid viscosity, and external volume force vector within each mesh cell, respectively.

B. Convection-diffusion equation

The diffusion and migration of the magnetic particles within each mesh cell are described by the convection-diffusion equation,
\[
\frac{\partial c}{\partial t} + \nabla \cdot (u_p c) = D \nabla^2 c
\]
where particle velocity \( u_p = u + u_{\text{drift}} \) and diffusivity \( D = k_B T/3\pi \eta_{\text{mix}} d_p \), with \( k_B \) as Boltzmann’s constant and \( T \) as absolute temperature. Within each mesh cell, the local slip velocity of the particles relative to the surrounding flow \( (u) \) is described as the drift velocity \( u_{\text{drift}} = F_s/(6\pi\eta_{\text{mix}}r_p) \); it is due to the external force \( (F_s) \) applied on a single particle by magnetic or gravitational forces.

C. Magnetic field equations

Equations (5) and (6) can be used to describe the magnetic fields,
\[
\text{Gauss equation : } \nabla \cdot \mathbf{B} = 0 \tag{5}
\]
\[
\text{Constitutive equation : } \mathbf{B} = \mu_0(h + M) = \mu_0(1 + \chi)\mathbf{H} = \mu_r\mathbf{H} \tag{6}
\]
where \( \chi = \chi_{\text{ff}}c \) is the local susceptibility of the ferrofluid diluted by the cladding diamagnetic fluids. The vectors \( \mathbf{B}, \mathbf{H}, \) and \( M \) indicate the magnetic flux density, magnetic field strength, and magnetization, respectively. The \( \mu_0 \) and \( \mu_r \) present the vacuum permeability and relative permeability, respectively.
D. Force calculations

The volume force term \( F_f \) (N/m³) in the Navier-Stokes equation is the sum of the gravitational force vector \( F_g \) (in \(-z\) direction only) and the magnetic force vector \( F_m \).

\[
F_f = F_g + F_m.
\]  (7)

The gravitational force vector, which was usually ignored in the previous literature,\(^{19}\) is considered here,

\[
F_g = \rho_{mix} g.
\]  (8)

The volume force vector term \( F_m \) (N/m³) is expressed as\(^{21,22}\)

\[
F_m = \frac{\chi_f c \mu_0}{\mu_0} (B \cdot \nabla B)
\]  (9)

where \( c = l/i \), has the same meaning through the whole work, e.g., in Eqs. (1), (2), (4), (6), and (9).

The uniform magnetic field refers to the uniformity of the applied external magnetic field. However, this field is not uniform within the three stream system, because of the differences in susceptibility between the cladding diamagnetic streams and the magnetic ferrofluid core stream. The magnetic field lines would bend towards materials with higher magnetic susceptibility.

The volume magnetic susceptibilities of pure glycerol and water are \(-9.8 \times 10^{-6}\) and \(-9.0 \times 10^{-6}\), respectively, indicating two typical diamagnetic materials. The magnetic susceptibility of the glycerol-water mixture (with 60 vol.% glycerol) can be calculated as \(-9.5 \times 10^{-6}\), which is negative and does not differ substantially from that of air.

Since the magnetic susceptibility of ferrofluid (1.51) is much higher than that of the diamagnetic glycerol-water mixture (\(-9.5 \times 10^{-6}\)), the magnetic flux densities inside the ferrofluid core is much higher than the cladding diamagnetic streams, as indicated by the magnetic field lines in Figure 2. This non-uniform flux density arrangement leads to gradients of the magnetic field, resulting in magnetic forces on the ferrofluid (Figure 2).

FIG. 2. Cross-sectional views (z-y plane) of the channel show the concentration, the magnitude of magnetic flux density, and the \( y \) and \( z \) components of magnetic forces at time of 0 s and 0.6 s. The simulated magnetic field lines were also shown in the concentration images. The simulation was carried out using a 2D model with applied magnetic flux density of 5 mT in the \( y \) direction, without consideration of the effect due to flow velocities along the \( x \) direction.
FIG. 3. The (a) experimental and (b) simulated particle concentration profiles (depth averaged along the z direction, in the x-y plane), and (c) the simulated volume concentration profile at \( x = 2.25 \) mm (in the y-z plane). All with \( H_a = 3.98 \) kA/m and \( Q_{ratio} = 4 \).

### III. RESULTS AND DISCUSSION

#### A. Concentration profile

Magnetofluidic spreading was studied in the channel using a ferrofluid core cladded by a glycerol-water mixture. The experiments (Figure 3(a)) were conducted with an applied magnetic field \( (H_a) \) of 3.98 kA/m and \( Q_{ratio} \) of 4. With a uniform magnetic field applied in the \( y \) direction, it was found that the width of the ferrofluid core (dark area) becomes wider (spreads) as the fluid flows through the channel. The experimental results and numerical simulations were compared (Figures 3(a) and 3(b)). These experimental results were in good agreement with the simulated magnetic particle concentration profile for the same parameters as those used in the experiments. The simulated particle concentration profile at the outlet was used to compare with the experimental results of the width of the ferrofluid core. The \( WF \) was defined as the ratio of ferrofluid width, measured at the outlet, divided by the channel width. This value of width fraction is used to quantify the magnetofluidic spreading. The images obtained from the high speed camera were tuned to gray color; the \( WF \) was measured based on the gray level.

Figure 3(c) shows the cross-sectional view of volume concentration profile of magnetic particles near outlet. It was found, for the first time, that spreading of the ferrofluid was parallel to the magnetic field direction (along the \( y \) direction), but the spreading was not uniform. Spreading was more obvious near the channel walls compared to the core regions of the channel. This effect is due to the slower flow velocities near the channel walls. The ferrofluid near the walls is exposed to the magnetic field for a greater time period, resulting in increased spreading. Also, examining the cross section of the channel, the maximum magnetic flux densities are at the top and bottom boundaries (that parallel to the initially applied magnetic field direction) of the ferrofluid core (Figure 2), the particles will prefer to be located at these positions. Figure 3(c) considers both the effects mentioned above, but Figure 2 only includes the magnetic force effect. Comparing the concentration profile images in Figures 2 and 3(c), spreading near the top and bottom channel walls in Figure 2 is somewhat smaller than the one in Figure 3(c). The spreading of the ferrofluid near the channel walls could increase the interfacial area of the ferrofluid core and the cladding fluids, which will be useful for applications such as particle mixing.

The gravitational force was ignored in the previous literature. The ferrofluid with slightly lower density rises and the heavier cladding diamagnetic sinks, due to the gravitational force (along the \( -z \) direction). Here, with gravitational force considered, it was found that the spreading width of the ferrofluid was greater near the top boundary compared to the bottom boundary (Figure 3(c)).
In this work, the simulated width fraction of the ferrofluid core near the outlet was quantified by the following expression: \( WF = 0.375WF_{\text{surface}} + 0.625WF_{\text{ave}} \), where \( WF_{\text{surface}} \) and \( WF_{\text{ave}} \) are the maximum width fraction value of the ferrofluid core measured at the outlet surface (Figure 3(c), in the y-z plane) and the depth-averaged volume concentration profile (Figure 3(b), at outlet), respectively. The values of 0.375 and 0.625 are fitting parameters between the experimental results and the above relationship. \( WF_{\text{surface}} \) and \( WF_{\text{ave}} \) were recorded when the particle volume concentration was higher than a critical value of 0.33.

The magnitude of the magnetofluidic spreading depends on the magnitude of the applied magnetic field, the magnetic properties of the ferrofluid, initial fluid velocities, and the material properties of both fluids. These parameters will be studied in Secs. III B–III F.

B. Factors governing MFS

The spreading of the ferrofluid was due to the combined effect of the following factors: (1) diffusion of particles, (2) drift velocity of magnetic particles \((u_{\text{drift}})\) due to the magnetic force applied on each particle, and (3) convective diffusion due to the fluid velocities \(u\).

1. Particle diffusion

The effect of particle diffusion was related to the diffusivity \(D = k_B T / 3\pi \eta_{\text{mix}} d_p\). Higher temperature of the fluid, lower fluid dynamic viscosity, and/or smaller particle size lead to higher particle diffusivity and increased spreading and mixing. Here, since the magnitude of \(D\) is small (less than \(1.5 \times 10^{-11} \text{ m}^2/\text{s}\)) and the flow duration is short (~0.22 s), the effect of particle diffusion is small.

2. Drift velocity

The particle drift velocity \((u_{\text{drift}})\) depends on the dynamic viscosity of the fluid, particle size, and the magnetic force applied on each particle. Since the magnetic force \((F_{\text{ms}})\) applied on each particle is linearly related to the particle volume, larger particle size leads to higher drift velocity. Figure 4(a) shows the drift velocity profile for a cross-sectional plane of the channel. High drift velocities are observed near the boundary of the ferrofluid and the cladding glycerol-water mixture, where relatively high magnetic forces \((F_{\text{ms}})\) are applied on the particles. Since the duration of the ferrofluid flow through the channel is short (~0.22 s) and particle drift velocity is very small (~50 nm/s) for the small particle sizes used in this study, the effect of particle drift velocity is negligible.

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3. Convective diffusion

The flow direction and velocities of the fluids can greatly influence particle spreading. Convective diffusion is influenced by two factors: (1) initial fluid velocities, which are controlled by $Q_{\text{ratio}}$ and $FR$ of the ferrofluid. For laminar flows, cross-sectional convection of the species is a challenge. (2) The influence on fluid velocities due to the magnetic volume force ($F_m$) applied on the ferrofluid. When the magnetic field was applied, the ferrofluid can be driven by the magnetic volume force to work like a liquid piston, resulting in a change in the fluid cross-sectional velocities along the magnetic force directions. As shown in Figures 4(b) and 4(c), the y component of velocity ($v_y$) without applied magnetic field is much less than the velocity in the presence of the magnetic field ($H_a = 3183 \text{ A/m}$). Fluid velocities can increase spreading by greater convective diffusion.

Comparing Figures 4(a) and 4(b), the particle drift velocity ($v_{\text{drift}}$) can be $\sim 1000$ times smaller than the y component of fluid velocity ($v_y$), thus drift velocity is considered to be negligible in this case.

Thus, magnetofluidic spreading is mainly a result of convective diffusion driven by the magnetic volume force applied on the ferrofluid. The magnitude of spreading mainly depends on initial flow conditions (e.g., $Q_{\text{ratio}}$ and $FR$) and magnetic volume forces applied to the ferrofluid (e.g., $H_a$ and $i$).

C. Applied magnetic field ($H_a$) effect

Both magnetic volume force on the ferrofluid and the particle drift velocity increase with increasing applied magnetic field, leading to greater MFS, i.e., larger $WF$. As shown in Figure 5, for constant flow rate ratio, width fraction of ferrofluid core increases with higher applied magnetic field. The simulated results match the experimental results, demonstrating good accuracy of the model.

The $Q_{\text{ratio}}$ was also varied (Figure 5) to study the extent to which the $H_a$-$WF$ curve shifts with changing $Q_{\text{ratio}}$. It is clear that the curves shift towards lower width fraction values for larger $Q_{\text{ratio}}$ value. Very good agreement was observed between the experimental and modelling results.

D. Flow rate ratio ($Q_{\text{ratio}}$) effect

The influence of fluid velocities can be studied in the terms of two factors: (a) flow rate ratio $Q_{\text{ratio}}$, the flow rate of the diamagnetic cladding fluid divided by the flow rate of the ferrofluid core and (b) the inlet flow rate of ferrofluid core, $FR$.

The $Q_{\text{ratio}}$-$WF$ curves are shown in Figure 6. Width fraction decreases with increasing $Q_{\text{ratio}}$ for fixed $H_a$ and initial flow rate of ferrofluid core, indicating less spreading of the ferrofluid core.

![Simulation results with $Q_{\text{ratio}}$ = 4, 6, 8, 10, 12. Experimental results with $Q_{\text{ratio}}$ = 4, 6.](image)

**FIG. 5.** Simulation and experimental results, the $WF$ of the ferrofluid core increases with increasing $H_a$ and $Q_{\text{ratio}}$. 

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This can be explained by the hydrodynamic focusing effect. Although the inlet flow rate of the ferrofluid core is fixed, the flow rate of the diamagnetic cladding fluid is higher for larger $Q_{ratio}$, leading to higher pressure inside the diamagnetic cladding fluid compared to the ferrofluid core in the y-z cross section view of channel. The pressure force (the negative value of pressure gradient, $-\nabla p$) lies mainly along the boundary of the two fluids and would suppress spreading by acting against the magnetic volume forces. Thus, higher $Q_{ratio}$ leads to higher pressure difference and lower WF of ferrofluid.

The effect of applied magnetic field $H_a$ on the width fraction of the ferrofluid core is shown in Figure 6. $Q_{ratio}$-WF curves shift to higher width fraction values for higher $H_a$ values, which can be explained by the greater magnetic volume force of the ferrofluid and higher particle drift velocities.

The experimental and numerical results of $Q_{ratio}$-WF curves were also compared in Figure 6, very good agreement between the two curves was obtained, confirming the validity of our model.

E. Flow speed of the ferrofluid core effect

For a fixed $Q_{ratio}$, the velocities of the fluids inside the channel depend linearly on the inlet flow rate of the ferrofluid core. Thus, for higher inlet FR value of ferrofluid core, the initial flow speeds of both ferrofluid core stream and diamagnetic cladding fluid, as well as the average flow velocity of the fluids inside the channel, are correspondingly higher.

FIG. 6. Simulation and experimental results, WF of ferrofluid core increased with increasing $Q_{ratio}$ and $H_a$.

FIG. 7. (a) WF of ferrofluid core increased with increasing inlet FR of ferrofluid core and $Q_{ratio}$ and (b) comparison of experimental and simulation results with increasing $H_a$ and FR of ferrofluid core.
The effect of flow rate of ferrofluid core on width fraction is shown in Figure 7(a). For fixed $Q_{ratio}$, width fraction of the ferrofluid core is reduced with increasing inlet flow rate of the ferrofluid core. This is because, for larger flow rate, the fluids spend less time within the channel due to the higher flow speed of the fluids inside the channel. For higher $Q_{ratio}$, the FR-WF curves in Figure 7(a) shift to higher width fraction values, similar to the results observed in Figure 6.

Figure 7(b) shows both experimental and numerical results of the influence of core flow rate on the $H_a$-WF curves. The curves shifted to higher width fraction values for lower inlet flow rate of the ferrofluid core. An excellent match was observed between the experimental results and the numerical predictions.

**F. Initial particle concentration of ferrofluid ($i$) effect**

For fixed $H_a$ and magnetic particle properties, the magnetic volume force applied on the ferrofluid depends on its initial particle volume concentration ($i$). The width fraction of the ferrofluid core is higher for larger $i$ (Figure 8). For higher $i$, the magnetic volume force on the ferrofluid is larger, facilitating ferrofluid spreading.

The contact area of the core and cladding streams was greatly enhanced by the hourglass shaped cross-sectional concentration profile (Figure 3(c)). This geometry can be very useful for microfluidic applications like micro mixing and micro chemical reactors, where the mixing level or reaction speeds of the two fluids depend on their contact area. Our numerical simulation includes various tunable parameters, e.g., inlet velocities of the three streams, flow rate ratios, diameter and magnetic susceptibility of magnetic particles, dynamic viscosity and density of the ferrofluid, and magnitude of external magnetic field. The influence of these parameters was investigated. These results provide understanding as well as practical guidance for novel wireless technologies for important microfluidic applications, such as micromixers, cytometers or cell sorters, biological analysis and catalysis with magnetic beads, optofluidics devices, and microlenses.

**IV. CONCLUSIONS**

The spreading of a water based ferrofluid core cladded by a glycerol-water mixture fluid in a three stream flow system was investigated. A uniform external magnetic field was applied perpendicular to the flow direction. Both experiments and modelling results were obtained and excellent agreement was found between the experimental results and modelling output. The mechanisms governing this spreading were determined. It was found that (1) the magnetic susceptibility differences caused the distortion of the applied uniform magnetic field, leading to the field gradients and magnetic forces applied on ferrofluids, (2) spreading of ferrofluid is mainly near the channel.
walls rather than the channel core sections, due to the slow flow velocities near the walls, and also the maximum field strength at top and bottom boundaries (parallel to the initially applied magnetic field direction) of the magnetized ferrofluid core in the cross-sectional plane view, (3) spreading of ferrofluid is mainly due to convective diffusion, driven by the magnetic volume force applied on the ferrofluid, (4) higher initial volume concentration of ferrofluid and/or a higher external magnetic field strength can increase magnetic forces and increase the width of the ferrofluid core, (5) higher flow rate ratio and/or a higher flow speed reduces the duration of fluid flow through the channel and suppresses convective diffusion in cross-sectional planes, reducing spreading. These provide methods to develop novel wireless technologies for microfluidic applications, such as micromixers, cytometers, cell sorters, and tuneable optofluidics.

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