Rapid magnetofluidic mixing in a uniform magnetic field

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This paper reports the investigation of mixing phenomena caused by the interaction between a uniform magnetic field and a magnetic fluid in a microfluidic chamber. The flow system consists of a water-based ferrofluid and a mixture of DI water and glycerol. Under a uniform magnetic field, the mismatch in magnetization of the fluids leads to instability at the interface and subsequent rapid mixing. The mismatch of magnetization is determined by concentration of magnetic nanoparticles. Full mixing at a relatively low magnetic flux density up to 10 mT can be achieved. The paper discusses the impact of key parameters such as magnetic flux density, flow rate ratio and viscosity ratio on the mixing efficiency. Two main mixing regimes are observed. In the improved diffusive mixing regime under low field strength, magnetic particles of the ferrofluid migrate into the diamagnetic fluid. In the bulk transport regime under high field strength, the fluid system is mixed rapidly by magnetically induced secondary flow in the chamber. The mixing concept potentially provides a wireless solution for a lab-on-a-chip system that is low-cost, robust, free of induced heat and independent of pH level or ion concentration.

Introduction

In the last decade, the development of microfluidic devices increasingly attracts attention from industry and academia due to its advantages such as small sample volume, low cost and high efficiency. Micromixer is one of the most important microfluidic components. Micromixers can work as stand-alone devices or be integrated in a more complex microfluidic system such as a lab on a chip (LOC). Rapid mixing is important for many applications such as biological, chemical and biochemical analysis. Research efforts have been made to achieve rapid and homogenous mixing of multiple samples in the microfluidic environment. According to the review by Nguyen and Wu,¹ micromixers are categorized as passive and active types. Passive micromixers rely purely on the arrangement of the phases to be mixed, while active micromixers require external fields such as pressure, temperature, electric and acoustic fields. Most active mixing concepts need electrodes and a complex design for the induction of the external disturbance. The applied electric field, acoustic field or optical field often lead to an increase in temperature, which is not desirable for sensitive samples such as cells or deoxyribonucleic acids (DNA). There is a need for a new technology for mixing in microfluidic systems such as lab-on-a-chip (LOC) that are of low-cost, simple, wireless, free of induced heat and independent of pH level or ion concentration. The use of magnetism would provide a wireless solution for this need.

Micro magnetofluidic devices utilize the interaction between magnetism and fluid flow in the microscale to gain new functionalities and capabilities.² Magnetism has been used for manipulation, transport, positioning, separation and detection in microfluidic devices. In most cases, a permanent magnet was used as the source of the magnetic field. The permanent magnets need to be selected according to the requirement of a specific field pattern. Pamme reviewed the application of conventional magnets and the fabrication of integrated magnets for microfluidic applications.³

Magnetic force has been used for active mixing previously. Mixing was achieved by micro stirrers⁴⁻⁵ and magnetohydrodynamic instability.⁶⁻⁷ Magnetic forces were induced by introducing magnetic particles into the working fluid.

The capability of magnetic manipulation using an external field leads to additional applications such as tagging target entities to the magnetic particles in a bio separator. The feasibility of controlling magnetic particles using an external magnetic field offers the option of wireless manipulation, which leads to the possibility of drug delivery using magnetic particles. The utilization of magnetic particles and the underlying physical principles for medical and biomedical applications such as magnetic separation, drug delivery, hyperthermia treatments and magnetic resonance imaging (MRI) contrast enhancement were reviewed recently.⁸⁻⁹

According to the particle size, a magnetic fluid is categorized as ferrofluid, magnetorheological fluid and fluid with discrete magnetic particles.² The particles in a ferrofluid have a diameter of less than 10 nm and can be well dispersed in the carrier fluid due to the dominant thermal energy. A magnetorheological fluid has particles with larger diameters ranging from 10 nm to 10 μm . The magnetic particles of a ferrofluid are coated with surfactant to prevent agglomeration. Since the discovery of ferrofluid in the early 1960s, this material has been used extensively. Readers may refer to Rosensweig¹⁰ for further details on ferrofluid and ferro hydrodynamics. Vékás reviewed recent achievement on the synthesis of magnetic particles.¹¹ Numerous research works have been conducted on the behaviour of ferrofluid as functions of particle concentration,¹² magnetization,¹³⁻¹⁴ and viscosity.¹⁵⁻¹⁶

Chaotic motion of magnetic particles can be induced to increase interfacial area of fluids and consequently their mixing efficiency. For instance, Suzuki and Ho numerically investigated mixing induced by magnetic beads in a serpentine channel.¹⁷ The chaotic motion of magnetic beads was realized by applying a locally time-varying magnetic force. The magnetic force was generated by embedding straight conductors driven by a periodic current. A similar numerical study was then carried out by Zolgharni et al. using straight channel with embedded serpentine conductors.¹⁸ Taking dipole interactions into account, the chain formation of particles were considered in the simulation. The resulting mixing efficiency was characterized by concentration distribution and chaotic advection of magnetic particles. Rong et al. proposed an active micromixer by combining rotational and vibrational force on magnetic beads.¹⁹ The sequential excitation of pairs of electromagnetic microtips leads to a rotating magnetic

Fig. 1 Magnetofluidic mixing in a uniform magnetic field: (a) Custom-made electromagnet; (b) Ferrofluid-core configuration; (c) Ferrofluid-cladding configuration.

field. To overcome the cost and complexity of the electromagnets, Grumann et al. used the lab-on-a-disk concept with a permanent magnet to improve the mixing process.²⁰

Micromixers utilizing ferrofluid were realized and reported previously. Mao and Koser reported mixing of ferrofluid with a fluorescein buffer using ferrohydrodynamics.²¹ Berthier and Ricoul investigated the ferrohydrodynamic instability in the vicinity of the magnet.²² Both experimental and numerical results show that the instability of ferrofluid would result in mixing in a microchannel. The instabilities of ferrofluid are caused by sudden velocity variation of the flow passing by a permanent magnet. Recently, Wen et al. reported the experimental and numerical investigation of an active micromixer based on fingering instability.²³⁻²⁴ In this work, ferrofluid and Rhodamine B solution were mixed. The fingering structures appear at the interface under an alternate magnetic field generated by an electromagnet driven by an AC current. With a magnetic field larger than 29.2 Oe and a frequency ranging from 45 to 300 Hz, the mixing efficiency of 95% can be achieved within 2 seconds.

There have been numerous reports on simulation of mass transport in ferrofluids including the effect of thermomagnetic convection.²⁵⁻²⁷ Furthermore, experimental results of particle transport in ferrofluids were also reported to support the corresponding theory.²⁸⁻²⁹ Finite element method was utilized to solve the governing equation of heat transfer in ferrofluids coupled with the thermomagnetic effect.³⁰ The numerical results

show the potential use of ferrofluids for cooling applications. The Galerkin finite element method was used for spatial discretization by Jue.³¹ The semi-implicit approach was employed including the explicit second-order Adams-Bashforth method for the advection terms and the implicit Euler method for the viscous terms. Besides, Ganguly et al. developed a numerical code based on finite-difference technique to solve the 2D problem that conforms to mass, momentum and energy conservation.³²⁻³³ The effect of the particles on flow field are all neglected in the above-mentioned work. Recently, the effect of the magnetic field was investigated on the particle concentration in the magnetic fluid.³⁴⁻³⁵ The effect of the particles are included. Finite volume method (FVM) was used in this study.

As mentioned above, magnetofluidic micromixers reported previously in the literature rely on the magnetic force caused by a field gradient. This field gradient is often generated by integrated electromagnet, which in turn induces a significant amount of heat and requires a complex fabrication process. Furthermore, time-dependent field gradients require synchronised external electromagnets or a moving permanent magnet leading to a complex overall mixer design. The present paper reports both experimental and numerical investigations of magnetically induced mixing based on the susceptibility gradient between a diamagnetic fluid and a ferrofluid. Instead of using a high magnetic field gradient and a high field strength, the concept reported here only needs a uniform magnetic field with a field strength of few milliteslas, two or three order of magnitudes lower than previously required by other magnetic mixing concepts. Our concept achieves efficient mixing by enhancing the migration of suspended magnetic nanoparticles and inducing the instability at the liquid-liquid interface. The mixing efficiency was evaluated for different flow rate ratios, which determine the mixing ratio between ferrofluid and the diamagnetic liquid. Furthermore, the relationship between mixing efficiency and viscosity ratio was also investigated. To adjust the viscosity of the diamagnetic fluid, water and glycerol was mixed in a corresponding ratio. In all experiments, the mixing efficiency was evaluated with varying magnetic flux densities. A numerical model based on finite volume method was developed to verify the mixing behavior observed in the experiment.

Materials and methods

Figure 1(a) shows the experiment setup with a customized electromagnet. The electromagnet was modified from a transformer, whose ferromagnetic core was cut to form an air gap of 5.5 mm. The uniform magnetic field in the air gap will apply across the entire microfluidic device. Demagnetization of the electromagnet is necessary to eliminate the remaining magnetization of the core after each round of the experiment. This measure was achieved by applying a reversed current for around 5-10 minutes with the highest current used in the previous experiment.

A circular chamber was employed for the mixing process between the ferrofluid and a diamagnetic mixture of water and glycerol. The device has three inlets and one outlet. The circular chamber has a depth of $H = 50 \mu\text{m}$ and a diameter of $D = 1 \text{ mm}$, Figure 1(b) and (c). The liquids are introduced into the inlets with a depth of $H = 50 \mu\text{m}$ and a width of $W = 50 \mu\text{m}$. The device was made of polydimethylsiloxane (PDMS) using standard soft lithography technique. Readers may refer to Song et al. for the

Fig. 2 Mixing efficiency as a function of magnetic flux density for ferrofluid-core configuration, the viscosity of ferrofluid and water/glycerol mixture are both fixed at $5 \text{ mPa} \cdot \text{s}$: (a) The flow rate of ferrofluid is fixed at 0.5 ml/h while the flow rate of mixture varies from 0.25 ml/h to 0.75 ml/h; (b) The flow rates of ferrofluid and mixture vary from 0.25 ml/h to 0.75 ml/h, but maintain a fixed ratio of 1.

detailed recipe for fabrication of the device.³⁶ The PDMS was peeled off and punched for access holes with a 0.75-mm diameter puncher. The substrate was then treated with oxygen plasma and bonded to another flat PDMS base. The PDMS device was further cut to fit into the air gap of the electromagnet. The fluids were delivered into the microfluidic device using two separated precision syringe pumps (KD Scientific Inc., USA). The whole setup was placed on a Nikon (Eclipse TE2000-E) inverted microscope equipped with a high-speed camera (Photron, APX RS). For generating the magnetic field, a current was supplied to the electromagnet by a DC power supply (GPS-3030D). The microfluidic device was slotted into the air gap for subsequent mixing experiments. A uniform magnetic field can be assumed as the mixing chamber is much smaller than the 5.5 mm×11 mm test section of the electromagnet. A magnetic flux density up to 50 mT can be generated by tuning the current up to 1.8 A. The magnetic flux generated at different currents was calibrated with a commercial gaussmeter (Hirst, GM05, UK).

Water-based ferrofluid (EMG707, Ferrotec) was used with super paramagnetic nanoparticles well dispersed in the carrier fluid. The ferrofluid has a saturation magnetization of 11 mT, a

density of $\rho_{FF} = 1.1 \times 10^3 \text{ kg/m}^3$, a viscosity at 27° of $\eta_{FF} = 5 \text{ mPa} \cdot \text{s}$ and a magnetic particle concentration of 2% vol and an initial susceptibility of $\chi_{FF} = 0.36$. The magnetization of this kind of ferrofluid was described in our previous work.³⁷ The diamagnetic liquid is a mixture of DI water and glycerol (G7757 Sigma-Aldrich). Glycerol has a density of $\rho_G = 1.26 \times 10^3 \text{ kg/m}^3$, and a viscosity at 20° of $\eta_G = 1410 \text{ mPa} \cdot \text{s}$. In order to characterize the effect of viscosity mismatch, different water/glycerol mixtures were used with viscosity at 25° of $\eta_{mix} = 2.5 \text{ mPa} \cdot \text{s}$ (65.6 wt% DI water and 34.4 wt% glycerol), $5 \text{ mPa} \cdot \text{s}$ (50 wt% DI water and 50 wt% glycerol), and $7.5 \text{ mPa} \cdot \text{s}$ (42.7 wt% DI water and 57.3 wt% glycerol). The corresponding density of the liquids at 25° are $\rho_{mix} = 1.09 \times 10^3 \text{ kg/m}^3$, $1.13 \times 10^3 \text{ kg/m}^3$ and $1.15 \times 10^3 \text{ kg/m}^3$. In the absence of the magnetic field, a clear interface was formed between the ferrofluid and the diamagnetic fluid, Figure 1(b) and (c). At a temperature of $T = 300 \text{ K}$, the diffusion coefficients of the magnetic nanoparticles ($d_p = 10 \text{ nm}$) into the different water/glycerol mixtures are estimated by Einstein's model as $D = k_B T / 3\pi\eta_{mix}d_p = 1.76 \times 10^{-11} \text{ m}^2/\text{s}$, $8.79 \times 10^{-12} \text{ m}^2/\text{s}$, $5.86 \times 10^{-12} \text{ m}^2/\text{s}$, respectively.

The experiments were carried out with ferrofluid-core and ferrofluid-cladding configuration, Figures 1(b) and 1(c). In the ferrofluid-core configuration, the ferrofluid acts as the core stream while the diamagnetic liquid works as the cladding stream. In the ferrofluid-cladding configuration, ferrofluid serves as the cladding stream while the diamagnetic liquid works as the core stream. The distribution of magnetic particles determines the magnetic force, and thus the induced secondary flow which would affect the particle distribution in return. In the case of the ferrofluid-core configuration, ferrofluid is delivered into the middle inlet at a constant flow rate of 0.5 ml/h. The water/glycerol mixture serves as cladding streams with a flow rate varying from 0.25 to 0.75 ml/h. Based on the properties of the water/glycerol mixture, the ranges of the Reynolds number $\text{Re} = \rho_{mix} U D_h / \eta_{mix}$ are $1.73 \times 10^{-1} - 2.88 \times 10^{-1}$, $0.896 \times 10^{-1} - 1.49 \times 10^{-1}$ and $6.07 \times 10^{-2} - 1.01 \times 10^{-1}$. Using the estimated diffusion coefficient of the magnetic particles in water/glycerol mixture, the ranges of the Peclet number $\text{Pe} = WU/D$ are $2.37 \times 10^5 - 3.95 \times 10^5$, $4.74 \times 10^5 - 7.90 \times 10^5$ and $7.11 \times 10^5 - 1.18 \times 10^6$.

In order to further investigate the effect of flow rate on the mixing behaviour, experiments were also carried out with flow rates of core and cladding stream varying from 0.25 to 0.75 ml/h but maintaining a fixed flow rate ratio of 1. The corresponding ranges of the Reynolds number are $1.15 \times 10^{-1} - 3.45 \times 10^{-1}$, $0.597 \times 10^{-1} - 1.79 \times 10^{-1}$ and $4.05 \times 10^{-2} - 1.21 \times 10^{-1}$. The ranges if the the Peclet number are $1.58 \times 10^5 - 4.74 \times 10^5$, $3.16 \times 10^5 - 9.48 \times 10^5$ and $4.74 \times 10^5 - 1.42 \times 10^6$.

In the case of ferrofluid-cladding configuration, the ferrofluid is delivered into the sheath inlets at a constant flow rate of 0.5 ml/h. The water/glycerol mixture served as the core stream with flow rate varying from 0.25 to 0.75 ml/h. The same flow rate was used to investigate the effect of magnetofluidic instabilities. The corresponding Reynolds number and Peclet number are the same

Fig. 3 Mixing efficiency as a function of magnetic flux density for ferrofluid-cladding configuration, the viscosity of ferrofluid and mixture are fixed at $5 \text{ mPa}\cdot\text{s}$ and $5 \text{ mPa}\cdot\text{s}$ respectively. (a) The flow rate of ferrofluid is fixed at 0.5 ml/h while the flow rate of mixture varies from 0.25 ml/h to 0.75 ml/h ; (b) The flow rates of ferrofluid and mixture vary from 0.25 ml/h to 0.75 ml/h , but maintain at a fixed ratio of 1.

as in the case of ferrofluid-core experiment. The above conditions imply a laminar flow in the channel and negligible diffusive mixing due to the high Peclet number.

Evaluating the quality of mixing inside the mixing chamber is difficult because of the circular shape of the chamber, and the nonlinear correlation between grey scale of the ferrofluid image and its concentration. We selected an interrogation window inside the chamber covering all three streams for mixing evaluation. The homogeneity of the recorded greyscale was used as a measure for the mixing efficiency. First, the mean value of the grey scale $\bar{I}$ of the window was determined. Next the standard deviation of the grey scale values in the interrogation window was evaluated:

$$\sigma = \sqrt{\frac{1}{N} \sum_{i=1}^N (I_i - \bar{I})^2} \quad (1)$$

If the window is homogeneous, the value of the standard deviation will be $\sigma = 0$. The mixing efficiency is subsequently determined as $\varepsilon = 1 - \sigma$.

Fig. 4 Mixing efficiency as a function of magnetic flux density for the same flow rate ratio of 1: (a) Ferrofluid-cladding configuration with viscosity of ferrofluid fixed at $5 \text{ mPa}\cdot\text{s}$ while the viscosity of mixture varies from $2.5 \text{ mPa}\cdot\text{s}$ to $7.5 \text{ mPa}\cdot\text{s}$; (b) Ferrofluid-core configuration with viscosity of ferrofluid fixed at $5 \text{ mPa}\cdot\text{s}$ while the viscosity of mixture varies from $2.5 \text{ mPa}\cdot\text{s}$ to $7.5 \text{ mPa}\cdot\text{s}$.

Numerical simulation

Because of the shallow circular chamber, the numerical simulation was realized with a two-dimensional (2D) model to gain more insights into the physics of the mixing process. The dimensions of the circular chamber used in the simulation are the same as those of the experiments. A fully developed velocity profile was assumed for the inlets of both the core flow and the sheath flow:

$$u = -\frac{3}{2} \frac{Q}{WH} \left(\frac{y^2 - b^2}{b^2} \right) \quad (2)$$

where Q is the volumetric flow rate of the flows, W and H are the width and height of the channel, $-H/2 \leq y \leq H/2$ is the coordinate along the channel height and $b = H/2$. No-slip condition was employed at the walls. The exit was set at outflow condition.

In Cartesian notation, the continuity and momentum

Fig. 5 Simulated velocity and concentration fields of ferrofluid-core configuration: (a) no magnetic field; (b) with magnetic field.

equations for the unsteady, viscous, and incompressible system of our problem are expressed as:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \bar{u}) = 0 \quad (3)$$

$$\frac{\partial}{\partial t} (\rho \bar{u}) + \nabla \cdot (\rho \bar{u} \bar{u}) = -\nabla P + \nabla \cdot [\eta (\nabla \bar{u} + \nabla \bar{u}^T)] + \bar{F} \quad (4)$$

where ρ indicates the density of the liquid sample, $\bar{u}$ is the velocity vector, and P represents the pressure. The concentration distribution in the whole computational domain is determined by the mass transfer of magnetic particles. The non-dimensional advection-diffusion equation for the concentration distribution is given as,

$$\frac{\partial C}{\partial t} + (\bar{u}_p \cdot \nabla) C = D \nabla^2 C \quad (5)$$

where C is the non-dimensional volume concentration and $\bar{u}_p$ represents the velocity of the particles. The magnetic force would induce an additional migration velocity to the magnetic particles and affect their diffusive behavior inside the circular chamber. By balancing the hydrodynamic force and magnetic force on each particle, the velocity is given by Stokes drag:

$$\bar{u}_p = \bar{u} + \bar{u}_{mag} = \bar{u} + \bar{F}_{mag} / 6\pi\eta r_p \quad (6)$$

where r_p is the radius of the magnetic nanoparticles, $\bar{F}_{mag}$ is the magnetic force on each particle:

$$\bar{F}_{mag} = \frac{1}{2} V_p \mu_0 \nabla \chi \bar{H}^2 \quad (7)$$

where V_p is the volume of each magnetic nanoparticle, χ is the susceptibility of the resulting liquid after mixing, μ_0 is the permeability of the free space which has a constant value of $4\pi \times 10^{-7} \text{ NA}^{-2}$ and $\bar{H}$ is the strength of the external uniform magnetic field. Without the applied magnetic field,

Fig. 6 Simulated velocity and concentration fields of ferrofluid-cladding configuration: (a) no magnetic field; (b) with magnetic field.

diffusive mixing is supposed to be negligible because of the high Peclet number. However, the diffusion coefficient in the simulation was affected by the so-called false numerical diffusion coefficient. Even with the 2nd order upwind scheme, numerical diffusion still exists in the simulation. As a result, the magnetic force which is proportional to the gradient of the susceptibility is much lower in the simulation than the actual experimental value of a much larger gradient. Because of the difficulty to get rid of numerical diffusion, the magnetic force in our simulation was adjusted accordingly to achieve a magnitude comparable to that of the experiment. In ferrofluid-core configuration, the magnetic force was corrected by a coefficient with the value of around 100. However, the numerical diffusion of magnetic liquid is stronger in ferrofluid-cladding configuration. Consequently, a coefficient of 1000 was used for the compensation of numerical diffusion.

In the case of a continuous ferrofluid flow in a circular chamber with an applied magnetic field perpendicular to the stream, the term $\bar{F}$ in Eq.(4) consists of gravitational force $\bar{F}_g$, interfacial tension $\bar{F}_s$ and magnetic force $\bar{F}_m$. The gravitational force is negligible as the experiment was carried out in a microscale. The interfacial tension can also be omitted as the fluid system is miscible. The magnetic force is calculated by solving the magnetic scalar potential which is introduced in the form of $\bar{H} = -\nabla \psi$. Starting with Maxwell equations, Liu et al. reported the governing equation for magnetic scalar potential and the corresponding magnetic force for an immiscible two-phase flow.³⁸ In our present work with miscible flow, the susceptibility is a function of particle concentration $\chi = \chi(C)$.³⁹ In the calculation, the susceptibility is assumed to vary linearly with the concentration. According to the review by Pankhurst et al.,⁹ the physical principles behind the bulk magnetic force is the response of magnetic particles to the applied magnetic field. In a uniform magnetic field, the magnetic potential equation and the corresponding magnetic force can be expressed as:

$$\nabla \cdot [(1 + \chi(C)) \nabla \psi] = 0 \quad (8)$$

$$\vec{F}_m = -\frac{1}{2}\mu_0|\vec{H}|^2 C_v \nabla \chi(C) \quad (9)$$

where C_v is the volume concentration of the particles which is the product of the initial volume concentration value C_{v0} and the non-dimensional concentration C calculated by the advection-diffusion equation (5). For the ferrofluid used in our study, the initial volume concentration is 0.02. The boundary condition for solving the magnetic potential inside the chamber is,

$$\vec{H} \cdot \hat{n} = -\frac{\partial \psi}{\partial n}, \quad \forall \vec{x} \in \partial \Omega \quad (10)$$

In the calculation, the variation of density and viscosity need to be considered as functions of concentration as well. By using the concept of volume fraction, the density and viscosity can be determined by the following expression²⁴

$$\rho = C\rho_m + (1-C)\rho_0 \quad (11)$$

$$\eta = \eta_m e^{R(1-C)} \quad (12)$$

where ρ_m and ρ_0 are the densities of water-based ferrofluid and water/glycerol mixture respectively. In Eq. (12), the parameter R is expressed as $R = \ln(\eta_0 / \eta_m)$ with η_m and η_0 representing the viscosities of water-based ferrofluid and water/glycerol mixture, respectively. In the concentration equation (5), the diffusion coefficient is taken to be constant as the field strength used in the experiment is relatively low.

The finite volume method (FVM) was used to solve the governing equations in a Cartesian staggered grid. The pressure/velocity coupling was achieved with SIMPLER algorithm which is helpful in improving convergent efficiency of the calculation.⁴⁰ The 2nd order upwind scheme was employed for discretizing the advection-diffusion equation. The higher the order of the upwind method, the less is the false numerical diffusion. The time integration is proceeded with the fully implicit scheme.

Results and discussion

The results of mixing with ferrofluid-core configuration for different flow rate ratios and flow rates are shown in Figures 2. The mixing efficiency was determined from a rectangular interrogation window in the mixing chamber. The discrete experimental points are fitted with the Boltzmann function. Without a magnetic field, a regular lens shape was formed in the circular chamber with negligible diffusion. In the early stage of increasing magnetic flux, the magnetic force causes the ferrofluid core to spread due to the migration of magnetic particles into the diamagnetic fluid. In addition to the hydrodynamic velocity, the magnetic particles are subjected to secondary flow caused by the magnetic force. The secondary flow and thus the mixing efficiency increase as the magnetic flux increases. As the magnetic flux continues to increase, instability occurs at the liquid/liquid interface. Mixing is faster due to instability and the

subsequent chaotic flow behaviour in the chamber induced by the magnetic force. Complete mixing between the core and cladding flow occurs almost immediately at a high magnetic flux. The value of mixing efficiency remains constant at its maximum when a critical magnetic flux density is reached. The inserts in Figure 2 show the images of the mixing chamber at the initial condition, the inception of instability and the completely mixed state.

The amount of ferrofluid in the mixing chamber can be tuned by adjusting the flow rate ratio between the core and the cladding streams. Figure 2(a) shows that faster mixing and higher efficiency can be achieved with a lower flow rate ratio between cladding and core streams. Because ferrofluid is the active component of the miscible fluid system, the more ferrofluid in the system the more efficient is the mixing process. Due to the initially homogenous distribution of magnetic particles in the ferrofluid, the maximum mixing efficiency is higher with a broader stream of the core flow. Figure 2(b) shows how the total flow rate affects the mixing behaviour in the chamber. Faster mixing and a higher mixing efficiency can be achieved at a lower flow rate. Since the mixing concept relies on the competition between magnetically induced secondary flow and the pressure-driven convective flow, the lower the total flow rate the more dominant is the induced secondary flow, and consequently the more efficient is the mixing process. In the case of ferrofluid-core configuration, complete mixing was achieved with mixing efficiency approaching 0.9.

The results of ferrofluid-cladding configuration with different flow rate ratios and flow rates are shown in Figure 3. The mixing process of the ferrofluid-cladding configuration also underwent the transition from spreading to interface instability to complete mixing. However, the final mixing efficiency is higher than that of the ferrofluid-core case. As shown in the gradient of the efficiency curve, mixing of ferrofluid-cladding configuration is faster and requires a lower magnetic flux density. Similar to the ferrofluid-core case, the same conclusion can be drawn from the results shown in Figure 3(a). The more ferrofluid in the system or the lower is the flow rate ratio between the diamagnetic fluid and the ferrofluid, the more efficient is the mixing process. The effect of pressure-driven convective transport over magnetically induced transport is also apparent in Figure 3(b). A higher magnetic field strength is needed for mixing at a higher total flow rate.

The effect of viscosity ratio on mixing efficiency is analyzed and illustrated in Figures 4. The results are presented with the same flow rate of core and cladding streams. Figures 4(a) and 4(b) compare the effect of viscosity in ferrofluid-core and ferrofluid-cladding experiment, respectively. In both cases, the initial mixing efficiency is lower with smaller viscosity ratio between the diamagnetic fluid and the ferrofluid. However, a lower viscosity of the diamagnetic fluid allows faster mixing and approaches a higher final mixing efficiency. The viscosity of the diamagnetic fluid plays two roles in the mixing process. First, the viscosity affects the migration of magnetic nanoparticles into the diamagnetic fluid. According to equation (6) the higher the viscosity, the lower is the migration velocity. Second, the viscosity also affects the bulk secondary flow as governed by the Navier-Stokes equation (4). A higher viscosity means a weaker magnetically induced secondary flow. The combined effect of these two factors leads to the reduction of mixing efficiency with highly viscous diamagnetic fluid.

Fig. 7 Numerically simulated mixing efficiency at the different magnetic flux densities for both the ferrofluid-core and ferrofluid-cladding configurations.

As described in the previous section, numerical simulation was implemented for both ferrofluid-core and ferrofluid-cladding configurations. The viscosity ratio between the diamagnetic fluid and the ferrofluid was fixed at 0.5. The flow rates of both fluids were fixed at 0.5 ml/h. The present numerical simulation aims at providing a qualitative understanding of the mixing behaviour. Thus, the effects of viscosity and flow rate variation on the mixing behaviour were not investigated numerically. In the simulation, the ferrofluid was assumed to be linearly magnetized. The grid independent study was first performed to find a compromise between grid size and accuracy. Calculation error can be reduced with further grid refinement as the numerical diffusion is significantly affected by the grid size. A grid size of 200×182 was finally selected in the simulation, because numerical diffusion could not be considerably reduced with further grid refinement. Figures 5 and 6 show the representative results of the velocity and concentration fields of the ferrofluid-core and ferrofluid-cladding configurations, respectively. Without a magnetic field, the pressure-driven flow follows stable paths from the inlet to the exit of the chamber. Mixing is caused purely by molecular diffusion. As mentioned in the previous section, the excessive diffusion observed in Figures 5(a) and 6(a) is actually caused by the numerical calculation. A sharper interface was observed in the experiments due to the high Peclet numbers involved. The existence of a magnetically induced secondary flow is apparent in the simulated results of the velocity field shown in Figures 5(b) and 6(b). Fast mixing is achieved by the chaotic secondary flow induced by the mismatch of magnetic susceptibilities. The numerical results of mixing efficiency are presented in Figure 7 with the same Boltzmann fitting function used for experimental data. The mixing efficiency curves show a similar trend as the experimental results. The mixing process can basically be divided into two regimes. At low magnetic field strengths, mixing is caused by improved migration of magnetic particles into the diamagnetic fluid. The applied uniform magnetic field leads to a higher apparent diffusion coefficient of the particle. The mixing process behaves similar to purely molecular diffusive mixing.⁴¹ At high magnetic field strengths, the effect of magnetically induced secondary flow becomes apparent. Mixing occurs rapidly due to the more effective transport of the bulk fluid. In agreement with the experiment, the

Fig. 8 Mixing of a three-fluids system consisting of fluorescent diamagnetic fluid, ferrofluid and non-fluorescent diamagnetic fluid: (a) Intensity profile across the mixing chamber in a bright field image with epi-fluorescent filter; (b) Intensity profile in a fluorescent image with epi-fluorescent filter only.

ferrofluid-cladding configuration leads to a higher final mixing efficiency.

The present work only focuses on mixing and the concentration distribution of magnetic particles in the ferrofluid. The above insights gained from the numerical simulation show that in the first regime of “improved diffusive mixing” only magnetic particles migrate into the diamagnetic fluid. Solutes in the diamagnetic fluid will not be affected in this regime. Only in the second regime, effective mixing of solutes in the diamagnetic fluid can be achieved due to the bulk transport of the magnetically induced secondary flow. However, it’s difficult to evaluate the mixing efficiency of a solute optically, because magnetic particles and the ferrofluid are opaque and block both excitation and emission light to and from a fluorescent solute. To verify this point, we carried out the mixing experiment with the ferrofluid-core configuration using a three fluid systems: fluorescent diamagnetic fluid, ferrofluid and non-fluorescent diamagnetic fluid. All fluids have the same viscosity. The flow rates of the cladding diamagnetic fluids are half of that of the ferrofluid core, so that the total flow rate ratio between diamagnetic fluid and ferrofluid is unity. As the fluorescent fluid, the mixture prepared for the previous experiments with a viscosity matched that if the ferrofluid was mixed with a fluorescent dye (fluorescein disodium salt $C_{20}H_{10}Na_2O_5$).⁴² This dye is also called Acid Yellow 73 or C.I. 45350. Figure 8(b) shows the intensity profile across the mixing chamber for both

Fig. 9 Intensity profile across the mixing chamber with a two-fluids system consisting of a ferrofluid and a fluorescent diamagnetic fluid.

cases with and without magnetic field. The images were recorded with both bright through light and an epi-fluorescent filter (Nikon B-2A, excitation filter for 450–490 nm, dichroic mirror for 505 nm and an emission filter for 520 nm), so that the fluorescent and the non-fluorescent diamagnetic fluids can be discriminated. Figure 8(a) shows a similar intensity profile across both fluid indicating that magnetic particles has migrated into the diamagnetic fluids. However, it cannot be concluded that dye molecules is transported to the other non-fluorescent side of the chamber. Figure 8(b) shows the same profiles of the images using the epi-fluorescent filter only. A slight increase in intensity value on the non-fluorescent side can be observed. However, the blocking effect of the magnetic particles is still dominant.

The effectiveness of mixing the fluorescent solute in the experiment of Figure 8 is not apparent, and may be caused by the symmetric arrangement of the streams. The fluorescent dye may mix well with the ferrofluid in the upper half of the chamber but does not cross to the lower half due to symmetry. To verify this point, we designed another experiment with an asymmetric arrangement of a two-fluids system: ferrofluid and fluorescent diamagnetic fluid. The results of the intensity profile and the corresponding fluorescence images are depicted in Figure 9. Similar to the experiment of Figure 8, a slight increase in intensity of the ferrofluid side can be observed. At a much higher flux density of 56.15 mT with an applied current of 2 A, clear secondary flow pattern can be observed and the magnetic particles agglomerate and form large clusters due to dipole interactions. The reduction of the concentration magnetic particles elsewhere, allowing excitation light to reach the dye and emission light to escape. A relatively uniform intensity profile with increased intensity values is the proof that the fluorescent solute molecules spread across the entire mixing chamber.

Conclusions

Mixing in a circular chamber was realized with a ferrofluid under a uniform external magnetic field. The microfluidic device used in the experiment offers a laminar flow condition with negligible molecular diffusion. The uniform magnetic field was employed to generate instability at the interface of the liquids with mismatched magnetic susceptibilities and consequently a gradient in magnetization. Mixing is instantaneous at a relatively low magnetic field of less than 10 mT. The maximum mixing efficiency is around 90%. The experimental results show that the mixing process is affected

by factors such as flow rate ratio, flow rate and viscosity. The mixing process consists of two basic regimes. The first regime under low applied field strength is similar to molecular diffusive mixing caused by the improved migration of magnetic particles into the diamagnetic fluid. The second regime is bulk transport mixing caused by magnetically induced secondary flow. Between these two regimes is a transition region of unstable interface between the two fluids. The numerical simulation incorporates the underlying physics of the mixing process and can qualitatively explain the experimental data. However, the numerical diffusion considerably affects the simulation results. To obtain more accurate and quantitative simulation results, the problem of numerical diffusion needs to be addressed in the future works.

Notes and References

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