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Gravity-induced swirl of nanoparticles in microfluidics

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Abstract

Parallel flows of two fluids in microfluidic devices are used for miniaturized chemistry, physics, biology and bioengineering studies, and the streams are often considered to remain parallel. However, as the two fluids do not always have the same density, interface reorientation induced by density stratification is unavoidable. In this paper, flow characteristics of an aqueous polystyrene nanofluid and a sucrose-densified aqueous solution flowing parallel in microchannels are examined. Nanoparticles 100 nm in diameter are used in the study. The motion of the nanoparticles is simulated using the Lagrangian description and directly observed by a confocal microscope. Matched results are obtained from computational and empirical analysis. Although solution density homogenizes rapidly resulting from a fast diffusion of sucrose in water, the nanofluid is observed to rotate for an extended period. Angular displacement of the nanofluid depends on the ratio of gravitational force to viscous force, Re/Fr^2 , where Re is the Reynolds number and Fr is the Froude number. In the developing region at the steady state, the angular displacement is related to y/D_h , the ratio between distance from the inlet and the hydraulic diameter of the microfluidic channel. The development of nanofluid flow feature also depends on h/w , the ratio of microfluidic channel's height to width. The quantitative description of the angular displacement of nanofluid will aid rational designs of microfluidic devices utilizing multistream, multiphase flows.

Keywords

Microfluidics; Density stratification; Nanoparticles; Angular displacement; Miscible fluids

Introduction

Lab-on-a-Chip type of devices utilizing parallel streams of two or more fluids have been developed for a wide range of applications (Weigl and Yager 1999; Kamholz et al. 1999; Tokeshi et al. 2002; Hessel et al. 2003; Chen and Meiners 2004; Kenis et al. 1999; Takayama et al. 2001b; Jeong et al. 2004; Zhao et al. 2001; Takayama et al. 1999; Takayama et al. 2003; Kane et al. 1999; Paek et al. 2010, 2011; Zheng et al. 2003, 2004; Pollack et al. 1999, 2001; Yamashita et al. 2004; Takayama et al. 2001a; Ferrigno et al. 2002; Chohan et al. 2004; Cohen et al. 2005; Bazylak et al. 2005). In the past few years,

multistream flow containing nanoparticles has attracted interest for nanoparticle separation, concentration (Yamada et al. 2004; Huh et al. 2007; Zhao and Cheng 2011; Abecassis et al. 2008), assembly (Lin et al. 2003; Bresme and Oettel 2007), enhancement of heat transfer and fluid flow (Zhao et al. 2011; Yang et al. 2012) as well as thermal insulation (Zhao et al. 2012). In these applications, adjacent streams from separate inlets are usually assumed to remain parallel, while mass transport across streams happens exclusively through diffusion. The transport phenomena in such systems have been carefully studied employing both computational and experimental approaches (Kamholz and Yager 2001; Ismagilov et al. 2000; Hatch et al. 2001; Kamholz et al. 2001).

However, parallel streams introduced from the inlets do not always have matched density. The resulting density stratification is expected to drive reorientation of the streams. Interfacial reorientation induced by gravity has been observed in microfluidic flows (Stiles and Fletcher 2003; Yoon et al. 2005; Yamaguchi et al. 2006, 2007). For example, Yoon et al. (2005) found empirically that the interface between two miscible and density-stratified fluids reoriented when diffusion was negligible. The tilting angle depends on several dimensionless parameters including the dimensionless distance in the flow direction, y/D_h , and the ratio of gravitational force to viscous force, Re/Fr^2 . In particular, Re/Fr^2 was found to dominate the flow characteristics. Later, Yamaguchi et al. (2007) performed fluid dynamics simulation and demonstrated that the reorientation took place in a creeping shear flow of miscible fluids even considering diffusion. In addition to the two parameters mentioned above, microchannel geometry, wall property, interfacial tension between two fluids and interstream mass diffusion were also discovered to influence the interfacial reorientation. Beyond simple interfacial tilting, laminar flow of density-stratified fluids together with Dean vortices have been shown to generate different types of curved interfaces (Xuan et al. 2011). The Dean force in side-by-side two fluids system engenders a distorted but symmetric interface (Yamaguchi et al. 2004). This is enhanced if the two fluids have a density mismatch and the distorted interface becomes nonsymmetrical given appropriate angle between gravity and initial fluid interface. While these studies have provided significant insights into density-stratified multistream flow, they were carried out using relatively high flow rates and low Re/Fr^2 , and the resulting reorientation were generally below 0.5π .

In this study, we explored the interface reorientation associated with creeping flow of two miscible, density-stratified fluids at much greater Re/Fr^2 than reported previously. The two fluids are introduced side by side into simple straight microchannels and nano-particles are dispersed in the stream of the lighter fluid. The density stratification is found to generate a gravity-driven flow transition, which induces a swirling motion of the nanofluid. The dependence of the nanofluid swirl on density stratification, hydrodynamic drag, and channel geometry are systematically studied using confocal microscopy imaging and computational fluid dynamics modeling.

Materials

Rhodamine B ($M_W = 479$ Da), glass coverslips and spherical-shaped polystyrene nanoparticles were purchased from Fisher Scientific (Fair Lawn, NJ). The nanoparticles (100 nm in diameter) are internally dyed with Firefly™ Fluorescent Red (Ex 542/Em 612 nm). The nanoparticles have a narrow size distribution and are well dispersed in the suspension (Fig. 1). For microfluidic chip fabrication, SU-8 photoresist and developer were purchased from MicroChem (Newton, MA). Silicone elastomer and a curing agent were from Dow Corning (Midland, MI). Phosphate-buffered saline (PBS) was obtained from Mediatech (Herndon, VA). Lyophilized bovine serum albumin (BSA), sodium dodecyl sulfate (SDS), and sucrose were from Aldrich Chemical Co. (Milwaukee, WI).

Experimental procedure

Microfluidic devices were fabricated (Fig. 2) using the standard soft lithography technique. First, SU-8 photoresist was patterned on a silicon wafer by photolithography to form a negative mold. Next, a 10:1 mixture of poly(dimethylsiloxane) (PDMS) prepolymer and curing agent was poured onto the mold and baked at 70 °C until cured. After cutting and removing the PDMS channels from the SU-8 mold, the channels were bonded permanently to glass slides through oxygen plasma at a forward power of 50 W and pressure 100 mT above the base pressure for 30 s. The channels were then blocked with 1 % BSA in PBS to reduce nonspecific binding on the channel walls before further experiments.

Devices used in this study contain a Y-junction merging into a main channel, as shown in Fig. 2. Two main channel geometries were tested with dimensions of 40 mm × 2 mm × 200 μm, and 40 mm × 200 μm × 160 μm (length × width × height), respectively. The corresponding cross-sectional aspect ratios (h/w) are thus 0.1 and 0.8. Polystyrene nanoparticles were diluted in PBS/0.3 % SDS to a final particle concentration of 0.2 % (v/v). This concentration of SDS does not affect the Newtonian property of the fluid (Kang et al. 2010), but helps to maintain a dispersed state of the nanoparticles as confirmed by light scattering measurements (data not shown). The solution of 25 % sucrose was made by dissolving sucrose in PBS/0.3 % SDS.

To study nanoparticle transport in the devices, the nanofluid containing polystyrene beads was injected from one inlet, and the aqueous sucrose solution was injected from the other by a syringe pump (Chemyx Inc. Fusion 400, Stafford, TX). BD Luer-Lok™ 1 mL syringes (Franklin Lakes, NJ) were used for sample injection when flow rates were higher than 0.1 μL min⁻¹. Lower flow rates were introduced by VICI C-160 10 μL syringes (Houston, TX). Confocal images were taken on a fluorescent confocal microscope (Olympus IX81, Center Valley, PA). The images from the same y coordinates were automatically processed by FLUO-VIEW Viewer (Center Valley, PA) to form the cross-sectional images.

Mathematical model and numerical method

ANSYS Fluent (ANSYS Inc., Canonsburg, PA) was employed to simulate steady-state 3-D laminar flows of two miscible fluids in the developing and fully developed regions. The main channel geometries used in the experiments without branching inlets were created as a computational domain by ANSYS DesignModeler. The particle tracking theory with Eulerian–Lagrangian approach was employed. The main fluid (continuum phase) was treated using Eulerian description and the dispersed particles (discrete phase) were tracked using the Lagrangian description. Two-way coupling was employed in our modeling, where the continuum phase could affect the behavior of discrete phase (nanoparticles), and vice versa. Hence, in this process a proper designed solver calculated the continuous and discrete phase equations in an alternate manner until a converged coupled solution was achieved. Particles Fluent are treated as volumeless points, but size effect is incorporated in the equations of Brownian, drag, lift, and buoyant forces. Given the low particle concentration (0.2 %) and much larger channel geometry compared to the particle size, particle–particle, and particle–wall interactions are negligible. Thus, it is reasonable to ignore the particle volume for particle tracking.

Gravitational acceleration was set as 9.8 kg m⁻³. Since the buffer solution used in the experiments has comparable density and viscosity as water, side-by-side uniform flow of 25 % sucrose ($\rho = 1,100 \text{ kg m}^{-3}$, $\mu = 2.5 \text{ cP}$), and water ($\rho = 1,000 \text{ kg m}^{-3}$, $\mu = 1.0 \text{ cP}$) was introduced as the inputs in the simulation with equal flow rates. Mass-weighted-mixing law was applied to account for the viscosity evolution, and volume-weighted-mixing law was

applied to form the density evolution. Velocity of constant magnitude normal to x - z plane was applied at the inlets. Nanoparticles 100 nm in diameter were introduced together with water at a mass flow rate of $10^{-11} \text{ kg s}^{-1}$. The volume fraction of the nanoparticles is 0.2 %. The density and viscosity of the nanofluid is 0.1 kg/m^3 and $5 \times 10^{-3} \text{ cP}$ (Masoumi et al. 2009) higher than those of the base fluid, respectively. As these differences are negligible, density, and viscosity of the base fluid (water) are applied to the nanofluid (Table 1). No-slip stationary wall with a reflecting boundary condition was chosen for nanoparticles. Zero pressure was set at the outlet.

Steady-state creeping flow of the two streams was modeled by the two-way coupling laminar flow module. The equations governing the conservation of mass and momentum of the liquid phase are (Drew 1983; Zhang and Prosperetti 1997; Kohnen and Sommerfeld 1998).

$$\nabla \cdot (\rho \vec{u}) = 0 \quad (1)$$

$$\nabla \cdot (\rho \vec{u} \vec{u}) = -\nabla p + \nabla \cdot (\bar{\tau}) + \rho \vec{g} + \sum (F_D (\vec{u}_p - \vec{u})) \dot{m}_p \Delta t \quad (2)$$

$$Re_p = \frac{\rho d_p |u_p - u|}{\mu} \quad \text{and} \quad F_D = \frac{18\mu}{\rho_p d_p^2} \frac{C_D Re_p}{24}, \quad (3)$$

where ρ is the fluid density, μ is the molecular viscosity of the fluid, $\vec{u}$ is the fluid phase velocity, p is the static pressure, $\bar{\tau}$ is the stress tensor, $\rho \vec{g}$ is the gravitational body force, $\vec{u}_p$ is the particle velocity, ρ_p is the density of the particle, d_p is the particle diameter, C_D is the drag coefficient, $\dot{m}_p$ is the mass flow rate of the particles, Δt is the time step, $F_D (\vec{u} - \vec{u}_p)$ is the drag force per unit particle mass, and Re_p is the relative Reynolds number. The last term in (Eq. 2) is the momentum source term that represents the influence of the particles on the flow field of fluids.

Diffusion of sucrose into water was modeled by the species transport module:

$$\nabla \cdot (\rho \vec{u} Y_i) = -\nabla \cdot (-\rho D_{i,m} \nabla Y_i), \quad (4)$$

where Y_i is the local mass fraction of each species, and $D_{i,m}$ is the mass diffusion coefficient for species i in the mixture.

Discrete-phase module was used to track the movement of particles:

$$\frac{d\vec{u}_p}{dt} = F_D (\vec{u} - \vec{u}_p) + \frac{\vec{g}(\rho_p - \rho)}{\rho_p} + \vec{F}_b + \vec{F}_1 \quad (5)$$

$$F_{b_i} = \zeta_i \sqrt{\frac{\pi S_0}{\Delta t}} \quad \text{and} \quad S_0 = \frac{216\mu k_B T}{\pi^2 \rho^2 d_p^5 \left(\frac{\rho_p}{\rho}\right)^2 C_c} \quad (6)$$

$$\vec{F}_1 = \frac{2K(\mu/\rho)^{1/2} \rho d_{ij}}{\rho_p d_p (d_{lk} d_{kl})^{1/4}} (\vec{u} - \vec{u}_p), \quad (7)$$

where $\vec{F}_b$ is the Brownian force per unit particle mass (Li and Ahmadi 1992), $\vec{F}_1$ is the Saffman's lift force per unit particle mass due to shear (Saffman 1965), $K = 2.594$, ζ_i are zero-mean, unit-variance-independent Gaussian random numbers, T is the absolute temperature of the fluid, k_B is the Boltzmann constant, C_c is the Stokes–Cunningham slip correction, and d_{ij} is the deformation tensor. For comparison, nanoparticles were replaced by molecules ($M_W = 479$ Da) and microparticles ($d_p = 10 \mu\text{m}$) to study size dependence of the particle motion. Similar to the above procedures, laminar, species transport, and discrete phase models were applied to simulate the motion of microparticles, while laminar and species transport models were used for molecular studies. Iterative calculations to solve Navier–Stokes equations were carried out by the SIMPLE scheme for pressure–velocity coupling. Contours of discrete phase model concentration were saved at different cross-sections.

Following previous studies (Stiles and Fletcher 2003; Yoon et al. 2005; Yamaguchi et al. 2006, 2007), we hypothesize that the spatial characteristics of the nanofluid are influenced by $Re/Fr^2 (= D_h^2 g \Delta \rho / \mu v)$, the ratio of the gravitational force to the viscous force, y/D_h , the dimensionless entrance distance and h/w , the aspect ratio of microfluidic channel. Here D_h is the hydraulic diameter of the channel, $g \Delta \rho$ is the buoyancy constant, μ is the viscosity of the mixture, v is the average velocity of fluid flow along the main channel, y is the entrance length along the channel, h is the channel height, and w is the channel width. Since the two streams were introduced at the same flow rate, the ratio of average velocity between the two streams is unity and its effect on the nanoparticle dynamics and the flow structure was not investigated here.

In order to check the spectral convergence, nano-particle distribution was simulated using different mesh sizes of 32, 16, 8, and $4 \mu\text{m}$ in a microchannel with $h/w = 0.1$, $y/D_h = 0.5$, and $Re/Fr^2 = 1109$. The particle distribution was found to be comparable for mesh size of 8 and $4 \mu\text{m}$ (Table 2). Thus, $8 \mu\text{m}$ mesh size is sufficient to achieve the spectral convergence, and the numerical results presented below were generated using the mesh size of $8 \mu\text{m}$.

Results and discussion

First, nanoparticle migration was examined in a channel with a cross-sectional dimension of $200 \mu\text{m}$ (h) by 2 mm (w), which corresponds to an aspect ratio (h/w) of 0.1. A 25 % sucrose solution and the nanofluid were injected into the two arms of inlets by a syringe pump, and flow rates of the two fluids were kept the same and constant. After the flow reached a steady-state, confocal fluorescent images were taken in the x – z plane at different y positions. In comparison, numerical simulations were run for the same geometry and flow condition. Figure 3a shows the measured and predicted nanoparticle distribution at three x – z planes normal to the flow direction, $y = 0.5 D_h$, $28 D_h$, and $56 D_h$, respectively. The volume flow rate was $0.96 \mu\text{L min}^{-1}$ and Re/Fr^2 was 1109. A viscosity value of 1.46 cP was used to calculate Re/Fr^2 . This value corresponds to the viscosity of 12.5 % sucrose. It was picked considering fast diffusion and homogenization of small molecules with our experimental parameters, as observed in Fig. 3b. Thus, nanoparticle motion mostly happens within 12.5 % sucrose except for a short distance near the device inlets. The top image of each panel from confocal microscopy denotes the concentration distribution of nanoparticles; the middle image is the predicted nanoparticle concentration contour from simulation. The two match well in all x – z planes. The bottom graph of each panel shows the in-plane velocity vectors overlapped on the out-of-plane velocity contours.

As observed in Fig. 3a, the interface between the nanofluid and sucrose solution quickly reorients from the vertical direction. The fluorescent nanoparticles reside on the top half of the channel at $y/D_h \sim 1$, which corresponds to fully developed reorientation in previous

reports when Re/Fr^2 is on the order of 1 (Yoon et al. 2005). With Re/Fr^2 on the order of 1,000 here, the nanoparticles continue to rotate in the x - z plane along the sides of the cross-section till the device outlet. For comparison, Rhodamine B was used in the place of nanoparticles in the lower density stream and its distribution was found to equilibrate at $y/D_h \sim 1$ by confocal microscopy (Fig. 3b). As the molecular weight of Rhodamine ($M_W = 479$ Da) is comparable to that of sucrose ($M_W = 342$ Da), this suggests that the density mismatch disappears within a short distance from the fluid converging point, due to fast diffusion of sucrose.

To understand the long-lasting motion of the nanofluid, the fluid velocity field was studied and presented as the bottom image of each panel in Fig. 3a. The in-plane velocity vectors display a counterclockwise swirling pattern induced by the initial density stratification. As fluids travel along the channel, magnitude of the in-plane velocity gradually decreases, but remains undeveloped even near the device outlet. At the same time, the out-of-plane velocity develops from an asymmetrical profile to the Poiseuille flow. The same rotational direction of the fluid and nanoparticles suggests that the nanoparticles follow the fluid motion.

As the input flow rate changes, reorientation pattern of the nanofluid changes accordingly. Figure 4a–c shows the swirling pattern of nanofluid at three different flow rates, corresponding to $Re/Fr^2 = 4438, 2219$, and 1109 . The angular displacement (θ) of the nanofluid in the developing region was quantified using the leading edge of the fluorescent region (Stroock 2002) as shown in Fig. 3a. The relationship between θ and the dimensionless translational distance (y/D_h) is presented in Fig. 4d in the log–log scale using all images with clear rotational fronts. The empty symbols are from simulation and solid symbols from experiments. It is observed that as Re/Fr^2 increases or input flow rate decreases, the nanofluid experiences greater angular displacement at the same y/D_h . With all three Re/Fr^2 values tested in this work, the nanofluid swirl does not reach the fully developed state even at the device outlet. Furthermore, linear relationships between θ and y/D_h in the log–log scale are observed for all three Re/Fr^2 in the developing region. An allometric fitting of the simulation results (lines in Fig. 4d) indicates that the powers at Re/Fr^2 of 4438, 2219, and 1109 are 0.49, 0.53, and 0.60, respectively. The less-than-unity power indicates deceleration of the nanofluid rotation, which matches the in-plane velocity profile (Fig. 3a). This also supports that nanoparticle motion is driven by gravity-induced fluid flow.

The initial nanofluid reorientation is driven by the density stratification until the densifier (sucrose) homogenizes. From Fig. 4, it is observed that greater Re/Fr^2 promotes more reorientation. Greater Re/Fr^2 implies higher gravitational force relative to viscous force, and thus more in-plane-fluid momentum to drive the nanofluid swirling. Compared to the work by Yoon et al. (2005), in which the lighter fluid rotates up to $\pi/2$ in the fully developed state, we observed much greater reorientation angles. This difference results from pronounced diffusion of the densifier in our work: a uniform density is reached before the angular displacement reaches $\pi/2$, thus momentum acquired from initial density stratification continues to drive the motion of the nanofluid. On the other hand, diffusion is negligible in Yoon's work. It is difficult for the lower density fluid to overcome the gravitational force and re-orientate beyond $\pi/2$.

After the sucrose concentration reaches equilibrium in the channel, the nanoparticles experience several in-plane forces, including the drag force, gravitational force, buoyancy force, and Brownian force, among which the drag force dominates. This in-plane drag force decelerates nanofluid rotation, thus the power between θ and y/D_h is less than one. Yoon et al. (2005) reported a constant power of 0.51 between θ and y/D_h in the developing region when diffusion is negligible with Re/Fr^2 in the range of 0.1–10. Here, we observed

comparable powers of 0.49 and 0.53 corresponding Re/Fr^2 of 4,438 and 2,219, respectively, while diffusion of the densifier is significant. The comparable power suggests that the dominant forces decelerating the reorientation may be consistent in our and Yoon's work, despite the great separation of Re/Fr^2 . Although densifier diffusion is significant in our study, net diffusion happens for a transient period compared to the residence time of fluids in the channel in the cases of high Re/Fr^2 . Thus the drag force is mostly from an invariable source, similar to that in Yoon's work in the absence of diffusion. On the other hand, when Re/Fr^2 is 1,109, the time portion with net diffusion is larger, and the drag force is from a variable source during this period. In this case, the slope between θ and y/D_h deviates slightly from the value in Yoon's work.

We further compared the angular displacement at the same cross-sectional location of $y/D_h = 28$ but with different Re/Fr^2 of 1109, 2219, 4438. The results are plotted in Fig. 5 with the solid and empty triangles denoting experimental and simulation results, respectively. A linear dependence of θ on Re/Fr^2 in the log-log scale is observed. An allometric fitting of the simulation results shows a power of 0.62. As Re/Fr^2 is inversely proportional to average velocity in the y direction and θ is measured at the same y coordinate here, the x axis of Fig. 5 scales linearly with the pattern development time. The power of θ versus Re/Fr^2 being less than 1 suggests that the average rotational velocity decrease at greater Re/Fr^2 . Interestingly, this power value matches the value of 0.61 from Yoon et al. (2005) using two miscible fluids of negligible diffusion and much lower Re/Fr^2 . This suggests again that physics behind the interfacial reorientation is comparable in both cases, despite the inclusion of solute diffusion and nanoparticle in our work. As the fluid residence time is long in our channels, the transient solute diffusion does not have a significant effect on nanofluid rotation except to enable the reorientation beyond $\pi/2$. At the same time, the inertia of nanoparticles is small hence the nanoparticles follow the secondary fluid flow.

Since the device geometry has been suggested to play an important role in the interfacial reorientation (Yoon et al. 2005), nanofluid motion in a device with an aspect ratio of 0.8 was investigated next. Similar to the above tests, the nanofluid and the denser sucrose solution were injected side-by-side into the device. At a flow rate of $0.04 \mu\text{L min}^{-1}$ for each stream and Re/Fr^2 of 530, the confocal images (images on the left of each panel) and predicted contours of nanoparticle distribution (images in the middle of each panel) are shown in Fig. 6. In comparison, the overlapped in-plane velocity vectors and out-of-plane velocity contours (images on the right of each panel) are also presented. As observed in Fig. 6, nanofluid motion reaches a fully developed state near the inlet, which coincides with the dampening of the in-plane velocity. The discrepancy between experimental and numerical results arises from a back flow as discussed below.

Figure 7 shows the quantitative relationship between the angular displacement and y/D_h for different values of Re/Fr^2 in the devices with $h/w = 0.8$. The empty symbols are from simulation and solid symbols are from experiments. The solid lines trace the simulation results and are used to guide the eye. We did not fit θ versus y/D_h in the developing region here due to fast development of the reorientation. The deviation of experimental data from numerical predictions in the developing region is caused by a backflow near inlets at extremely low flow rates: it was observed that sucrose penetrated into the nanofluid inlet branch at flow rates lower than $0.1 \mu\text{L min}^{-1}$, likely due to slight difference of flow resistance in the two branches. Thus, the reorientation pattern develops faster in the confocal images. Nonetheless, the fully developed angular displacements from experiments and modeling are well matched, and it increases with the decrease of the input flow rate or the increase of Re/Fr^2 . This is consistent with the observations in the device of $h/w = 0.1$ and supports the theory of greater momentum transfer at higher gravitational force relative to the viscous force.

When the images are acquired at the same position in the fully developed region but with varying flow rates, the angular displacement scales with $(Re/Pr^2)^{0.61}$ (see Fig. 8). Surprisingly, the power is the same as in the device with an aspect ratio of 0.1, and is also consistent with previous results in the literature (Yoon et al. 2005), despite those studies characterized interfacial reorientation in the developing region.

A drastic difference is observed in the developing length between devices with aspect ratios of 0.1 and 0.8. For device with an aspect ratio of 0.8, secondary flow diminishes near inlets for $y/D_h \sim 1$, while for device with an aspect ratio of 0.1, secondary flow continues for a long distance even to the outlet of the channel ($y/D_h \sim 100$). The difference of development length is much greater than that of the Reynolds number, which is two times higher in the device with $h/w = 0.1$. This phenomenon can be explained by the initial momentum generated by density stratification. In the two geometries, the channel height is nearly the same but the width is eight times different. In the low aspect ratio device, movement of more fluid and thus greater momentum result in a much longer development length.

In addition to the dominant swirling motion as observed in the device of $h/w = 0.1$, simulation results from the $h/w = 0.8$ devices also reveal an inwards motion to the center of the x - z plane, forming an Archimedean spiral (inset of Fig. 8). This is likely a result of shear-induced motion (Grzelczak et al. 2010). Shear-stress distribution in a device with aspect ratio close to 1 tends to push nanoparticles inwards to the center of the cross-section. Combination of shear-induced and swirl motion results in an Archimedean spiral.

Besides channel geometry, the observed swirling effect is also sensitive to the particle size. As observed in Fig. 9a, 100 nm nanoparticles mainly swirl, 10 μm microparticles have an inward motion in addition to swirling, and ~ 1 nm molecules demonstrate fast diffusion. The different patterns can be explained by size-dependent forces. The main forces that the suspended species experience under density stratification are drag, Brownian, and lift force. Using the flow conditions and device geometry tested here, Reynolds numbers for all suspended species are much less than 1, which suggests that the inertial force is negligible relative to the drag force. Thus, all species follow the base fluid swirling through drag force. The effect of drag force can also be observed in the identical secondary flow of the base fluid in the presence of different species (Fig. 3a, b and unshown data). At the same time, diffusion length of micro- and nanoparticle is less than 10 μm within the residence time of 450 s and this diffusion length is less than 3 % of the device hydrodynamic diameter. Thus, no obvious diffusion is observed for these particles. On the other hand, for molecules of single nanometer dimension, the diffusion length is comparable to the microchannel hydrodynamic diameter within 6 s. This contributes to the observed homogenization of molecules near the device inlet. The main force that accounts for the difference between micro- and nanoparticles is the lift force. The lift force remains small for nanoparticles, but becomes significant for microparticles (Li and Ahmadi 1992). The lift force drives the microparticles away from the wall to the channel center where velocity of the base fluid swirling is low. Thus, microparticles have slower swirling velocity (Fig. 9b). For even bigger particles like millimeter-scale particles, we conjecture that the lift force may push all the particles even more toward the device center and the swirling velocity would be even slower.

Although it was not experimentally tested, it is worth noting that the diffusion coefficient of the densifier plays an important role to control the nanofluid motion. Too high a diffusion coefficient leads to an immediate homogenization, and the nanofluid does not have enough time to obtain the swirling momentum. On the other hand, too low a diffusion coefficient will lead to reorientation stopping at $\theta = \pi/2$, as further rotation requires overcoming the

gravity. Thus, densifier diffusivity needs to be carefully selected to produce desirable nanofluid swirl.

Conclusions

In conclusion, nanofluid swirl patterns induced by density stratification are observed in a steady laminar flow of two miscible fluids in microchannels. The nanofluid migration patterns are investigated in both entrance and fully developed regions in two geometries for various fluid velocities. Stream rotation forms when the device aspect ratio is 0.1, and Archimedean spiral-shaped patterns are observed when the device aspect ratio is ~ 1 . The angular displacement varies with dimensionless position from the inlet in the entrance region and the ratio between gravitational and viscous force, Re/Fr^2 . Through both experiments and simulation, the swirling motion of nanofluid is shown to follow secondary flow of the base fluid. The interaction of gravitational force, buoyancy force, drag force, Brownian force and diffusion can be utilized to explain the observed swirling patterns. As the particle size varies from nanometer to micrometer the swirling motion and migration pattern of particles alters. The patterns observed and predicted in the present work may have application in nanoparticle focusing, sorting, and mixing in microenvironments.

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List of symbols

x, y, z (μm)	Spatial coordinates
h (μm)	Microfluidic channel height
w (μm)	Microfluidic channel width
l (μm)	Microfluidic channel length
D_h (μm)	Microfluidic channel hydraulic diameter
L_p (μm)	Developing length
ρ (kg m^{-3})	Density of the fluid
μ (Pa s)	Viscosity of the fluid
p (Pa)	Static pressure
$\overline{\tau}$ (Pa)	Stress tensor

$\vec{g}(\text{m s}^{-2})$	Gravitational acceleration
$\vec{u}(\text{m s}^{-1})$	Fluid phase velocity
Y_i	Local mass fraction of each species
$D_{i,m}(\text{m}^2 \text{s}^{-1})$	Diffusion coefficient of species i in the mixture
$\rho_p(\text{kg m}^{-3})$	Density of the particles
$d_p(\mu\text{m})$	Particle diameter
$\vec{u}_p(\text{m s}^{-1})$	Particle velocity
$\dot{m}_p(\text{kg s}^{-1})$	Mass flow rate of the particles
$\Delta t(\text{s})$	Time step
C_D	Drag coefficient
$F_D(\text{kg m s}^{-2})$	Drag force
$T(\text{K})$	Absolute temperature of the fluid
k_B	Boltzmann constant
$d_{i,j}(\text{Pa})$	Deformation tensor
Fr	Froude number
Re	Reynolds number
Re_p	Relative Reynolds number

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Fig. 1. Scanning electron microscope images of nanoparticles used in this study. The particle diameter is measured to be 91.7 ± 7.2 nm ($n = 2,000$)

Fig. 2.

A schematic of microfluidic devices used in this study. All devices contain a Y-junction merging into a main channel. Two main channel geometries, $40\text{ mm} \times 2\text{ mm} \times 200\text{ }\mu\text{m}$, and $40\text{ mm} \times 200\text{ }\mu\text{m} \times 160\text{ }\mu\text{m}$ ($l \times w \times h$) were used in our work with the cross-sectional aspect ratios of 0.1 and 0.8, respectively

Fig. 3.

a Left A schematic showing the *top view* of the microchannel. Right confocal images (*top image* of each group) and predicted contours (*middle image* of each group) of nanoparticle distribution at three locations corresponding to $y/D_h = 0.5, 28, 56$. In-plane velocity vectors with velocity contours in the y direction are shown as the *bottom images* of each group. The swirling motion of the nanoparticles follows the in-plane fluid motion. The range of the color scale bar is from 0 to 1 kg m^{-3} for the nanoparticle distribution maps (*middle* of each group), and $0\text{--}180 \text{ } \mu\text{s}^{-1}$ for the out of plane velocity. **b** Confocal images (*top image* of each group), predicted contours (*middle image* of each group), and secondary flow vector with y direction velocity contours (*bottom image* of each group) of a small fluorescent molecule, Rhodamine B, along the flow direction. Rhodamine quickly equilibrates in the cross-sectional image. In both **a** and **b**, $h/w = 0.1$ and $Re/Fr^2 = 1,109$

Fig. 4.

a–c Confocal images (*left*) and predicted contours (*right*) of particle distributions in developing regions for $Re/Fr^2 = 4,438$ (**a**), $2,219$ (**b**), and $1,109$ (**c**). Good match between the two are observed. **d** The angular displacement (θ) as a function of dimensionless distance from the inlet y/D_h for $Re/Fr^2 = 1,109$ (*squares*), $2,219$ (*circles*), and $4,438$ (*triangles*). The *empty symbols* are from simulation and solid symbols from experiments. Allometric fitting of the simulation data points is also shown as *solid lines*

Fig. 5.

A log-log plot of the angular displacement (θ) as a function of Re/Fr^2 at $h/w = 0.1$, $y/D_h = 28$. The *empty symbols* denote the predicted results and *solid symbols* denote experimental measurements. Allometric fitting of the simulation data points is also shown as the *solid line*

Fig. 6.

Left a schematic showing the *top view* of the microfluidic device. *Right* the images on the left are from the confocal microscopy; the middle ones are from numerical simulation with the *color* corresponding to particle concentration from 0 to 1 kg m⁻³ and the images on the *right* is the in-plane velocity vectors overlapped with the out-of-plane velocity contours for $y/D_h = 0.1, 0.6, 1.2$ and 1.8 . Here $h/w = 0.8$ and $Re/Fr^2 = 530$

Fig. 7.

Log-log plots of the angular displacement (θ) as a function of y/D_h for different Re/Fr^2 with fully developed confocal images and predicted contours of nanoparticle distribution for different Re/Fr^2 in the insets. The squares represents results from $Re/Fr^2 = 530$, circles from $Re/Fr^2 = 1,060$, and triangles from $Re/Fr^2 = 2,120$. The empty symbols are from simulations, and the solid symbols are from experiments. In the fully developed region, experimental results match with simulation results. The angular displacement quickly reaches a fully developed state. The solid lines trace the simulation results and are used to guide the eye

Fig. 8. A log–log plot of the angular displacement (θ) as a function of Re/Fr^2 for fixed $y/D_h = 2.4$. The *empty symbols* are from simulation and *solid dots* are from experiments. Allometric fitting of the simulation data points is shown as the *solid line*

Fig. 9.

a Predicted contours of distributions for $10\ \mu\text{m}$, $100\ \text{nm}$ particles and $\sim 1\ \text{nm}$ molecules in otherwise identical stratified flow. In all cases, $Re/Fr^2 = 1,109$. Slower swirling velocity is observed for microparticles. The microparticles also exhibit an inward motion, which is not observed in the nanofluid swirling with the same channel geometry. **b** The predicted angular displacement (θ) as a function of dimensionless distance from the inlet y/D_h for particles with different diameter at $Re/Fr^2 = 1,109$. The *round symbols* are from $100\ \text{nm}$ particles and *square symbols* from $10\ \mu\text{m}$ particles. Allometric fitting of the simulation data points is also shown as *solid lines*

Table 1

Properties of fluid and nanoparticles in simulation

Properties	Value
Density of water	1,000 kg m ⁻³
Viscosity of water	1.0 cP
Density of 25 % sucrose	1,100 kg m ⁻³
Viscosity of 25 % sucrose	2.5 cP
Density of nanoparticles	1,050 kg m ⁻³
Diameter of nanoparticles	100 nm
Drag coefficient	0.47
Mass diffusion coefficient of sucrose ^a	2.14–5.22 × 10 ⁻¹⁰ m ² s ⁻¹

^aThe value range corresponds to mass diffusivity of sucrose in water up to 25 %. In Fluent simulation, mass-weighted-mixing law was applied to determine the viscosity at each point in the channel during mixing. The mass diffusion coefficients were then calculated by the program using the viscosity and Einstein equation

Table 2

Predicted nanoparticle distribution with different mesh sizes

Number of elements	Mesh size (μm)	Predicted nanoparticle distribution
37,800	32
327,600	16
2,500,000	8
20,000,000	4

8 μm mesh size is sufficient to achieve the spectral convergence, and the numerical results presented in the paper were generated using the mesh size of 8 μm