Ultrasound velocimetry of ferrofluid spin-up flow measurements using a spherical coil assembly to impose a uniform rotating magnetic field

Shahriar Khushrushahi, Markus Zahn*
Massachusetts Institute of Technology, Cambridge 02139, MA, USA

ARTICLE INFO

Keywords:
Ferrofluid sphere
Ferrofluid cylinder
Demagnetizing field
Spin-up flow
Spin viscosity
Spin diffusion
Ultrasound velocimetry
COMSOL simulations
Surface driven flows
Spherical coils/Fluxballs
Uniform/non-uniform magnetic fields

ABSTRACT

Ferrofluid spin-up flow is studied within a sphere subjected to a uniform rotating magnetic field from two surrounding spherical coils carrying sinusoidally varying currents at right angles and 90° phase difference. Ultrasound velocimetry measurements in a full sphere of ferrofluid shows no measurable flow. There is significant bulk flow in a partially filled sphere (1–14 mm/s) of ferrofluid or a finite height cylinder of ferrofluid with no cover (1–4 mm/s) placed in the spherical coil apparatus. The flow is due to free surface effects and the non-uniform magnetic field associated with the shape demagnetizing effects. Flow is also observed in the fully filled ferrofluid sphere (1–20 mm/s) when the field is made non-uniform by adding a permanent magnet or a DC or AC excited small solenoidal coil. This confirms that a non-uniform magnetic field or a non-uniform distribution of magnetization due to a non-uniform magnetic field are causes of spin-up flow in ferrofluids with no free surface, while tangential magnetic surface stress contributes to flow in the presence of a free surface.

Recent work has fitted velocity flow measurements of ferrofluid filled finite height cylinders with no free surface, subjected to uniform rotating magnetic fields, neglecting the container shape effects which cause non-uniform demagnetizing fields, and resulting in much larger non-physical effective values of spin viscosity $\eta^f \sim 10^{-8} - 10^{-12}$ Ns than those obtained from theoretical spin diffusion analysis where $\eta^f \leq 10^{-18}$ Ns. COMSOL Multiphysics finite element computer simulations of spherical geometry in a uniform rotating magnetic field using non-physically large experimental fit values of spin viscosity $\eta^f \sim 10^{-8} - 10^{-12}$ Ns with a zero spin-velocity boundary condition at the outer wall predicts measurable flow, while simulations setting spin viscosity to zero ($\eta^f = 0$) results in negligible flow, in agreement with the ultrasound velocimetry measurements. COMSOL simulations also confirm that a non-uniform rotating magnetic field or a uniform rotating magnetic field with a non-uniform distribution of magnetization due to an external magnet or a current carrying coil can drive a measurable flow in an infinitely long ferrofluid cylinder with zero spin viscosity ($\eta^f = 0$).

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Ferrofluid spin-up flow has often been experimentally studied using a finite height cylindrical container with and without a cover excited by an externally applied uniform rotating magnetic field. In our investigation we carefully examine various theories of ferrofluid spin-up flow in finite and infinite height cylinders and in spheres with experiments and computer simulations using COMSOL Multiphysics. This work shows that free surface magnetic stresses and the non-uniform demagnetization magnetic field that results from a finite height cylinder without a cover, even in an externally applied uniform rotating magnetic field, is the primary cause of this flow [1]. An infinitely long cylinder or a fully filled sphere of ferrofluid in a uniform rotating field, with zero spin viscosity ($\eta^f = 0$), has zero flow. We were not able to solve the ferrofluid flow equations for a ferrofluid filled sphere in a non-uniform rotating magnetic field using COMSOL; instead we solved the analogous problem assuming an infinitely long cylinder of ferrofluid in a non-uniform rotating magnetic field generated by an infinitely long permanent magnet of finite thickness, adjacent to the ferrofluid cylinder, in a uniform externally imposed rotating magnetic field [1].

2. Experiments

Our experiments involve using a ferrofluid in a spherical container surrounded by two concentric current carrying spherical coils with spatially orthogonal windings that generate perpendicular uniform magnetic fields. The two windings are driven by sinusoidal currents that are also out of phase by 90° in time as shown in Fig. 1, creating a uniform rotating magnetic field. A single
3. Theory

3.1. Ferrohydrodynamics

The fluid mechanics equations governing ferrohydrodynamics are conservation of linear and angular momentum equations [7]. The conservation of linear momentum equation assuming the flow is viscous dominated and incompressible \((\nabla \cdot \mathbf{v} = 0)\) is

\[
0 = -\nabla p + 2\zeta \nabla \times \mathbf{e} + \left(\zeta + \eta\right) \nabla^2 \mathbf{v} + \mu_0 (\mathbf{M} \cdot \nabla) \mathbf{H} \tag{2}
\]

The conservation of angular momentum neglecting inertial terms is

\[
0 = \mu_0 \mathbf{M} \times \mathbf{H} + 2\zeta \left(\nabla \times \mathbf{v} - 2\omega_0\right) + \left(\zeta + \lambda\right) \nabla^2 \mathbf{e} + \eta \nabla^2 \mathbf{e} \tag{3}
\]

where the variables are dynamic pressure \(p\) [N/m\(^2\)], fluid magnetization \(\mathbf{M}\) [A/m], magnetic field \(\mathbf{H}\) [A/m], spin velocity \(\mathbf{e}\) [1/s], ferrofluid dynamic viscosity \(\eta\) [N s/m\(^2\)], vortex viscosity \(\zeta = (3/2) \eta\phi_{vol}\) [N s/m\(^2\)] for small volume fraction \(\phi_{vol}\) of magnetic nanoparticles [7,8], and \(\lambda\) [N s] and \(\eta\) [N s] are the bulk and shear coefficients of spin viscosity. Many planar and infinitely long cylindrical geometry problems have \(\nabla \mathbf{e} = 0\). For all COMSOL analyses in this paper we neglect the next to last term in Eq. (3) since we assume that \(\eta\) [N s] and \(\lambda\) [N s] are of the same order and their effect is small according to the spin diffusion theory.

3.2. Spin diffusion

Rosensweig [7] predicted a dimensional value of spin viscosity using a diffusion length model

\[
\eta' \sim \eta l^2 \tag{4}
\]

where \(l\) is the average distance between the solid particles given as

\[
l = \left(\frac{4\pi}{3\phi_{vol}}\right)^{1/3} R
\]

where \(R\) is the radius of the particle and \(\phi_{vol}\) the volume fraction of the particles. The value of \(l\) and spin viscosity \(\eta'\) of Rosensweig's ferrofluid [7,9] with properties of \(\eta = 0.0012\) N s/m\(^2\), \(\phi_{vol} = 0.012\), and \(R = 5 \times 10^{-9}\) m are then \(l = 35.2 \times 10^{-9}\) m and \(\eta' = 1.487 \times 10^{-18}\) Ns. Rosensweig, using this value of spin viscosity, predicted an angular rotation rate that is a factor of 10\(^3\)-10\(^5\) smaller than what was experimentally obtained [7]. Several authors have also mentioned this discrepancy [10–13]. Schumacher et al. [14] also separately derived a theoretical expression for spin viscosity, using a modified kinetic molecular theory of an ideal gas model, with ferrofluid properties \(\eta = 3.85 \times 10^{-3}\) Pa s, \(\rho = 1187.4\) kg/m\(^3\), \(\zeta = 1.93 \times 10^{-3}\) Pa s, \(\eta' = 12.5 \times 10^{-9}\) m (magnetic nanoparticle and surfactant), and \(\phi_{vol} = 0.334\), and determines a value of spin viscosity to be \(\eta' = 6.4 \times 10^{-20}\) Ns while Eqs. (4) and (5) give \(l \sim 29.9 \times 10^{-9}\) m and \(\eta' \sim 3.25 \times 10^{-18}\) Ns.

The theoretical determination of spin viscosity, using the spin diffusion theory, is many orders of magnitude smaller than the reported experimentally fit spin viscosity values [3,5,6]. For the ferrofluid EMG900.2 [3] with properties \(\eta = 4.5 \times 10^{-3}\) Pa s, \(\rho = 1030\) kg/m\(^3\), \(\zeta = 2.9 \times 10^{-4}\) Pa s, \(R = 7 \times 10^{-9}\) m, and \(\phi_{vol} = 0.043\), the spin viscosity was estimated from fits to experiment to be \(\eta' \approx 10^{-9} - 10^{-12}\) Ns while Eqs. (4) and (5) give \(l \sim 32.2 \times 10^{-9}\) m and \(\eta' \sim 4.67 \times 10^{-18}\) Ns. Similarly for the water based ferrofluid MSGW11, with properties \(\eta = 2.02 \times 10^{-3}\) Pa s, \(\rho = 1200\) kg/m\(^3\), \(\zeta = 0.83 \times 10^{-4}\) Pa s, \(R = 3.95 \times 10^{-9}\) m [15], and \(\phi_{vol} = 0.0275\), the spin viscosity was experimentally estimated to be \(\eta' \approx 10^{-9} - 10^{-10}\) Ns [5,6] while Eqs. (4) and (5) give \(l \sim 21.1 \times 10^{-9}\) m and \(\eta' \sim 8.99 \times 10^{-19}\) Ns. The oil-based ferrofluid EFH1 has properties \(\eta = 7.27 \times 10^{-3}\) Pa s, \(\rho = 1221\) kg/m\(^3\), \(\zeta = 8.2 \times 10^{-4}\) Pa s, \(R = 5.3 \times 10^{-9}\) m [15], and \(\phi_{vol} = 0.0752\) with no reported experimentally fit values.
of spin viscosity; however using Eqs. (4) and (5), \(l \approx 20.2 \times 10^{-9} \text{m} \) and \(\eta \approx 2.98 \times 10^{-18} \text{Ns} \).

3.3. Magnetic field equations

The ferrofluid magnetization relaxation equation derived by Shliomis [8] is as follows:

\[
\frac{\partial M}{\partial t} + (\nabla \cdot \mathbf{v})M = \mathbf{M} \times \mathbf{M} - \frac{1}{\tau_{\text{eff}}} (M - M_{\text{eq}}) \tag{6}
\]

where equilibrium magnetization \(M_{\text{eq}} \) [A/m] is given by the Langevin equation

\[
M_{\text{eq}} = M_{s} \left( \coth(x) - \frac{1}{x} \right) \frac{H}{H_{c}}, \quad x = \frac{M_{s} V_{p} H_{0} H}{kT} \tag{7}
\]

with \(M_{s} \) [A/m] the saturation magnetization given as \(M_{s} = \phi_{\text{vol}} M_{d} \) \(M_{d} = 446 \text{[kA/m]} \) is the domain magnetization for magnetite [7], \(V_{p}\)

Fig. 2. Side profile of half of inner spherical coil with solenoidal coil or permanent magnet placed on top of fully filled sphere of ferrofluid to impose non-uniform magnetic field in addition to the uniform rotating magnetic field generated by two surrounding spherical coils [1].

Fig. 3. Measured flow velocity of Ferrotec EH1 [saturation magnetization of 421.2 G and low field magnetic susceptibility \(\chi = 1.59 \) [5]] oil-based ferrofluid under uniform magnetic field rotating counter-clockwise at 95 Hz in presence of a third coil, carrying oscillating current at 95 Hz, on top of ferrofluid filled sphere of diameter 10 cm at \(z = 5 \text{ cm} \), which causes the ferrofluid to have non-uniform variation in effective magnetic permeability. Strength of uniform rotating field precedes the suffix ‘Rot’ followed by RMS strength of magnetic field (in Gauss) generated by the third coil. Ultrasound probe is placed at bottom of ferrofluid filled sphere and measures \(z\)-velocity along the line \(x = 1.71 \text{ cm} \) with the sphere centering in \(x-z\) plane at \(x = z = 0 \). It is clear from the plot that the greatest velocity occurs near the top of the sphere where the coil is positioned, corresponding to the furthest distance from the acoustic probe. The plots clearly show that significant ferrofluid motion only occurs in presence of uniform rotating field when non-uniform magnetic field is imposed by the third coil that causes non-uniform magnetization in the ferrofluid [1].

Fig. 4. Measured flow velocity of Ferrotec EH1 oil-based ferrofluid under uniform magnetic field rotating counter-clockwise at 47 Hz in presence of permanent magnets (1601–5233 G) placed on top of the ferrofluid filled sphere (north pole of magnet making contact with sphere) at \(z = 5 \text{ cm} \) that creates a large non-uniform DC magnetic field causing the ferrofluid to exhibit non-uniform variation in effective magnetic permeability. Increase in strength of magnet increases flow in a greater region of ferrofluid filled sphere. Ultrasound probe is placed at bottom of ferrofluid filled sphere and measures \(z\)-velocity along line \(x = 1.71 \text{ cm} \) with sphere centered in \(x-z\) plane at \(x = z = 0 \). There is no flow with a uniform rotating magnetic field with no permanent magnet as magnetization of ferrofluid is uniform [1].

Fig. 5. COMSOL model setup for simulating flows in ferrofluid under influence of non-uniform field of permanent magnet, as well as uniform rotating magnet field imposed by surface current boundary condition at radial distance of 10\(R_{cyl}\) away. Magnet placed at distance 0.2\(R_{cyl}\) from cylinder is magnetized in \(y\)-direction and is \(x\) \((10 \leq x \leq 40)\) times the strength of rotating field. South pole of magnet faces cylinder as shown. \(\Omega \) [rad/s] is angular frequency of rotating magnetic field [1].
\[ [m^3] \text{ is the magnetic core volume per particle, } \mu_0 = 4\pi \times 10^{-7} \text{ H/m is the magnetic permeability of free space, } \kappa = 1.38 \times 10^{-23} \text{ J/K is Boltzmann’s constant, } T [K] \text{ is the temperature in Kelvin, and effective relaxation time constant } \tau_{eff} [\text{s}] \text{ includes the Brownian and Néel effects.}
\]

Maxwell’s equations for a non-conducting fluid are

\[
\nabla \cdot \mathbf{B} = 0 \quad (8)
\]

\[
\nabla \times \mathbf{H} = 0 \quad (9)
\]

\[
\mathbf{B} = \mu_0 (\mathbf{H} + \mathbf{M}) \quad (10)
\]

Eqs. (2), (3) and (6)–(10) were put into COMSOL with the appropriate boundary conditions applied (no slip on velocity boundary condition, \( \omega = 0 \) if \( \eta \neq 0 \) and a surface current boundary condition as a source of a uniform rotating magnetic field as shown in Fig. 5) to simulate the ferrofluid flow in uniform and non-uniform rotating magnetic fields.

### 4. Measurement results and COMSOL simulations

The constant demagnetizing factors of a sphere ensured that the field inside the ferrofluid volume would be uniform and ultrasound.

![Fig. 6. COMSOL calculated distribution of velocity streamlines with \( \eta = 0 \), illustrating vortices formed, for a Ferrotec MSGW11 [saturation magnetization = 154 G, low field magnetic susceptibility \( \chi = 0.56 \)] filled infinitely long cylinder, with 2000 G permanent magnet in the 100 G RMS uniform counter-clockwise rotating field, as a function of time over one period of rotation at angular frequency of \( \omega = 190 \text{ rad/s} \). Magnetic field magnitude is represented by the colored surface plots that show the evolution in time of the total magnetic field inside and outside ferrofluid cylinder due to uniform rotating magnetic field, ferrofluid cylinder’s outside dipole field, and non-uniform magnetic field due to permanent magnet. The white region represents magnetic field strengths near the permanent magnet that are beyond the scale shown. Total magnetic field colored surface plots are normalized to strength of rotating field of 100 G and are plotted up to 200 G. Dimensional magnitude of velocity is calculated to be on the order of up to 30 mm/s according to these simulations, which corroborates with experimental results in spherical geometry [1]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)](image_url)
velocimetry measurements gave negligible flow for both MSGW11 and EFH1. Negligible flow was also verified with COMSOL Multi-physics analysis in a spherical geometry using zero bulk and shear coefficients of spin viscosity ($\lambda' = \eta' = 0$) for EFH1 and MSGW11, while simulations of the spin diffusion theory [16], with experimentally fit values of spin viscosity for MSGW11 [5,6] ($\eta' \approx 10^{-9}$ N s) and a zero spin velocity $\omega = 0$ boundary condition at $r = R$, predicted a flow (1–80 mm/s) that should have been measurable with ultrasound velocimetry [1].

Using ultrasound velocimetry, the bulk flow velocity was measured in a 2/3 full ($R = 5$ cm) sphere of ferrofluid (max velocity $\leq 14$ mm/s) and in a ferrofluid filled cylinder of 6 cm height without a cover (velocity $\approx 1$–4 mm/s). These flows are due to both free surface curvature that causes magnetic tangential surface shear stress [17] as well as due to the demagnetizing effects associated with the shape of the contained ferrofluid resulting in a non-uniform magnetic field within the ferrofluid volume even though the externally applied magnetic field is uniform.

In addition to the uniform rotating magnetic field, extensive experiments were also conducted where the ferrofluid magnetic properties were disturbed using either a DC or AC current carrying coil, generating a 42 (Gauss/1) RMS maximum axial non-uniform field at the coil bottom adjacent to the ferrofluid filled sphere, or 1 in. diameter permanent magnets with surface axial field strengths 0.16–0.52 T, with a coil or magnet placed on top of the fully filled sphere of ferrofluid as shown in Fig. 2. In both cases, significant flows are obtained, with velocity plots shown in Figs. 3 and 4, as a result of the spatial non-uniformity of the ferrofluid magnetic properties due to the non-uniform magnetic field and ferrofluid magnetization as given by the Langevin equation in Eq. (7), with significant non-uniformity of the magnetic field near the top of the sphere near the coil or magnets.
Since we were not able to simulate the spherical geometry in a non-uniform rotating magnetic field using COMSOL, we instead did COMSOL simulations of an infinitely long ferrofluid filled cylinder, with zero spin viscosity (\( \eta_s = 0 \)), subjected to a uniform rotating magnetic field and a non-uniform magnetic field imposed by an infinitely long permanent magnet placed nearby. These simulations resulted in non-zero irregular and complicated two dimensional flows similar to experimental observations obtained in the spherical case even though the geometries are different.

Fig. 5 shows the COMSOL model setup with two infinitely long concentric cylinders, the inner one representing the ferrofluid cylinder of radius \( r_{cyl} \) (5 cm) while the outer one of radius \( 10r_{cyl} \) (50 cm) represents a stator winding from a surrounding 3 phase-2 pole AC motor winding. The stator winding with a surface current boundary condition generates a uniform rotating field (100 Gauss RMS), while the permanent magnet (0.5 x 0.5 in\(^2\) dimensions), magnetized in y-direction, is placed at a distance 0.2\( r_{cyl} \) (1 cm) above the ferrofluid filled cylinder making the ferrofluid magnetic field non-uniform. The strength of the magnet is a factor \( 10 \leq \alpha \leq 40 \) times greater than the strength of the RMS rotating magnetic field (permanent magnet strength 1000–4000 G). Air fills the gap between the outer stator winding and the ferrofluid cylinder.

The total magnetic field due to the uniform rotating field and the field due to the permanent magnet creates moving regions of cancellation (weak field) and addition (strong field) that rotate as seen by the colored surface plots in Fig. 6. The distribution of the ferrofluid’s magnetization clearly shows the effect of these rotating regions of strong and weak fields as a function of time in Fig. 7.

The non-uniform magnetic field and non-uniform distribution of magnetic properties as a result of the permanent magnet in addition to the uniform rotating magnetic field create non-zero irregular and complicated COMSOL computed two dimensional flows in Fig. 6, taking \( \eta_s = 0 \) for a Ferrotec MSGW11 water-based ferrofluid filled cylinder with a permanent magnet that is 20 times stronger (2000 G) than the uniform rotating magnetic field taken to be \( (3/2) \mu_0 K_0 = 100 \) G (RMS).

Fig. 7 is a plot of the non-uniform distribution of the magnetization of the EFH1 ferrofluid as a result of the non-uniform field imposed by a permanent magnet (1000 G), ten times stronger than the uniform rotating magnetic field of 100 G (RMS), over one rotational period taking \( \eta_s = 0 \). The blue circle representing weak magnetization, which can be seen at \( \Omega t = (3 \pi/2) \), is the magnetization due to the almost complete cancellation of the magnetic field due to the permanent magnet and the rotating field at a distance of approximately 0.115\( r_{cyl} \) from the top of the cylinder. The red region of strong magnetization at the top of the cylinder at \( \Omega t = (\pi/2) \) is due to the strong region of the magnetic field that saturates the EFH1 ferrofluid. The white region in Figs. 6 and 7 represents the magnetic field strengths or magnetizations that are greater than the colored scales (200 G maximum in Fig. 6 and 421 G maximum in Fig. 7) to better highlight the dominant effects.

In the absence of the permanent magnet, COMSOL simulations, with zero spin viscosity (\( \eta_s = 0 \)), result in negligible flow since the magnetic field internal to the ferrofluid cylindrical volume is also uniform due to equal transverse demagnetizing factors of 1/2.

5. Conclusions

COMSOL simulations, using experimentally fit spin viscosity values from previous work with finite height ferrofluid filled cylinders with a cover, for an MSGW11 filled sphere (\( \eta_s \approx 10^{-5} \) N s) with an \( a = 0 \) boundary condition at \( r = R \), predicted flow that should have been measurable by ultrasound velocimetry, but in contradiction, negligible flow was measured in a fully filled sphere, of either EFH1 or MSGW11 ferrofluid, subjected to a uniform rotating magnetic field. Measured bulk flows were obtained in a 2/3 full sphere of ferrofluid and a finite height cylinder both with a free surface, proving that the governing mechanism for the bulk flow is due to tangential magnetic stresses that drive surface driven flows and due to non-uniform magnetic properties within the ferrofluid volume either imposed by an external source (magnet/coil) or created due to the demagnetizing effects associated with the shape of the ferrofluid volume.

The main mechanism for flow in recent fitting of experimental flow velocity to the spin diffusion theory was actually due to the non-uniform fields generated inside the ferrofluid volume due to non-uniform demagnetizing field effects associated with the finite height cylindrical container (with a top cover), which the authors [3,5,6] did not consider. It is for this reason that experimentally fitted values of spin viscosity are many orders of magnitude greater than those theoretically predicted by the spin diffusion theory [7,14].

This also explains the reason why many authors [7,10,11,13] have asserted that there is a serious discrepancy between the theoretical results that predict a small flow velocity due to a small theoretical value of spin viscosity \( \eta_s \leq 10^{-18} \) Ns as compared to experimentally fitted non-physically large values of spin viscosity \( \eta_s \sim 10^{-8} - 10^{-12} \) Ns, while the actual spin-up flow velocity mechanism is from ignored non-uniform fields generated inside the ferrofluid volume due to demagnetizing field effects.

Acknowledgements

The authors would like to thank MIT visiting scientist Professor Gwan Soo Park of Pusan National University in South Korea for his experimental help and expertise, MIT Professors Jeffrey Lang and Jacopo Buongiorno for their helpful discussions and suggestions, and Dr. R.E. Rosensweig for a helpful critique of a draft version of this paper. The authors would like to acknowledge the Binational Science Foundation for partial financial support for this project (BSF Grant no. 20004081).

References


