Understanding Ferrofluid Spin-Up Flows in Rotating Uniform Magnetic Fields

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Abstract: Ferrofluid spin-up flow has been studied in finite height cylinders and spheres subjected to a uniform rotating magnetic field. Ultrasound measurements in finite height cylinders with a free surface show the presence of surface driven flows as well as bulk flows. In finite height cylinders with a cover, only bulk flows exist. Ultrasound measurements in fully filled spheres of ferrofluid give negligible measureable flow.

The bulk spin-up flow has been attributed to the demagnetizing non-uniform magnetic field associated with the shape of the fluid, the non-uniform distribution of fluid magnetization, and to spin-diffusion. COMSOL Multiphysics simulations help confirm that the real mechanism of bulk spin-up flow is due to non-uniform magnetic properties of the ferrofluid, either imposed externally or associated with the shape of the ferrofluid.

Keywords: ferrofluid, spin-up, demagnetizing fields, spin viscosity, spin-diffusion

1. Introduction

Ferrofluids are stable colloidal suspensions of single domain magnetic nanoparticles in a carrier fluid such as oil or water. The nanoparticles are usually ferro- or ferrimagnetic particles with typical diameters of order 10 nm coated with a surfactant layer of 1 to 2 nm. Their small size allows them to be easily dispersed by Brownian motion and prevents them from agglomerating under gravity, while their surfactant layer prevents them from sticking to each other from van der Waals and magnetic attraction forces. Ferrofluids exhibit super-paramagnetic behavior and the nanoparticles typically make up to 10% of the total fluid volume.

Spin-up flow is the term given to describe the process in which a fluid in a container reaches a state of rotation due to a rotating uniform or non-uniform magnetic field. In the case of spin-up flow of ferrofluid in a stationary cylindrical container with and without a cover, ferrofluid flow is set into rigid-body-like motion driven by a uniform rotating magnetic field. The mechanism governing the spin-up flow of ferrofluids in rotating uniform fields has been a topic of interest since its first experiment [1].

2. Spin-Up Mechanisms in Ferrofluid Literature

2.1 Surface Driven Flows

Historically, such rigid-body observations were made only on the rotating free surface since velocity distributions in the bulk of the ferrofluid could not be easily measured due to the opacity of the ferrofluids. Rosensweig [2] showed that for ferrofluids in uniform rotating magnetic fields, magnetic surface shear stresses caused by the shape of the meniscus at the free surface drives surface flow. A perfectly flat surface would not rotate, while a concave or convex shaped meniscus creates flow that counter-rotates or co-rotates respectively to the magnetic field rotational direction. He observed that the angular rotational rate of the free surface increased with a decrease in the internal diameter of the cylindrical container, contrary to expected results in viscous flows. He concluded that "surface stress rather than volumetric stress is responsible for the spin-up phenomenon" and that surface flows entrain the bulk fluid layer below.

For this theory to hold, a ferrofluid filled cylinder with no free surface would not have any spin-up flow. Ultrasound velocimetry measurements [3-6] (Figure 2), were able to show that in a ferrofluid container with or without a free surface there is a bulk flow that co-rotates with the rotating uniform magnetic field direction. In their experiments with a free surface, the flow near the surface would co-rotate or counter-rotate with a concave or convex shaped meniscus respectively, supporting Rosensweig's theory [2], but the bulk flow would always co-rotate with the uniform magnetic field direction. The mechanisms attributed to explaining bulk flow are described in the next two sections.

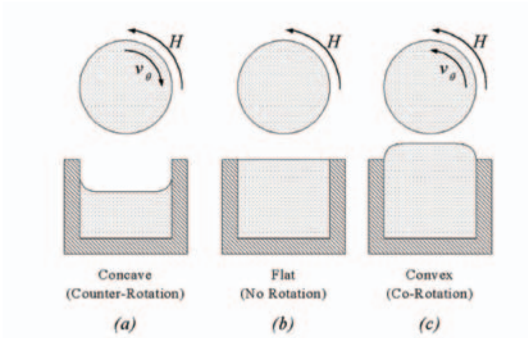

Figure 1. Experimental observations of driven ferrofluid free-surface flow. a) Concave shaped free-surface results in fluid counter-rotating with respect to magnetic field. b) Flat surface results in no discernible motion, c) Convex shaped free-surface results in fluid surface co-rotating with magnetic field [2, 7, 8].

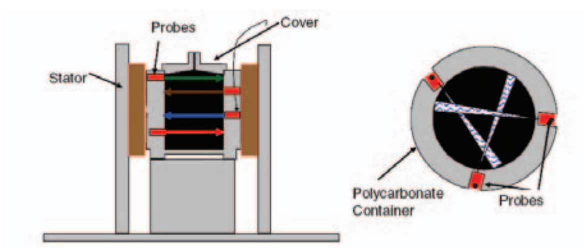

Figure 2. Experimental setup illustrating measurement of bulk ferrofluid flow profiles in cylindrical geometry. The cover ensures no free surface and zero flow velocity at the surface. Left: Ultrasound transducers placed at various heights in the container surrounded by the stator. Right: Top view of ultrasound probes oriented at different directions to measure flow velocities along different angles [6].

2.2 Spin Diffusion Theory

When a ferrofluid is subjected to a magnetic field the nanoparticles try to align their internal dipole moments in the direction of the local magnetic field. However, this alignment is impeded by two processes: rotational Brownian motion and Néel redistribution of particle magnetic domains. As a result, these delays lead to a lag between the magnetization **M** and the applied rotating field **H** such that they are not collinear. This creates a body-torque density, given by $\mu_0\mathbf{M}\times\mathbf{H}$, which results in spinning nanoparticles dragging the fluid around it and converting some of its internal angular momentum to the angular momentum of the fluid. In effect, this short-range transport of internal angular momentum would only result in macroscopic motion if there were a non-uniform distribution of spin cancellation that depends on particle/wall interactions. This process is known as "spin-diffusion".

Bulk flow measurements [3-6] were fit to spin-diffusion theory [9]. The spin-diffusion theory explained early measurements [1] of fluid rotation in near rigid-body motion right up to a thin boundary layer near the solid wall surface [7]. This theory assumes that the magnetic field throughout the ferrofluid region is uniform with uniform magnetization of the ferrofluid. This results in a magnetic body force of exactly zero and a uniform magnetic body couple.

The theoretical determination of spin viscosity, using spin-diffusion theory, is many orders of magnitude smaller than reported experimentally fit spin viscosity values [3-5]. Rosensweig [10] predicts a dimensional value of spin viscosity using a diffusion length model

$$\eta' \approx \eta l^2 \qquad \textbf{(1)}$$

where η is the dynamic viscosity of the ferrofluid and l is the average distance between the solid particles given as

$$l = [(4\pi)/(3\varphi_{vol}))]^{1/3} R \qquad \textbf{(2)}$$

with R the radius of the particle and φ_{vol} the volume fraction of particles. The value of l and spin viscosity η' of Rosensweig's ferrofluid [1, 10] with properties of η=0.0012 [Pa-s], φ_{vol}=1.2%, and R=5 [nm], are then $l \approx 35.2$ [nm] and $\eta' \approx 1.487\text{x}10^{-18}$ [N-s]. Rosensweig, using this value of spin viscosity, predicts an angular rotation rate that is a factor of 10^3-10^4 smaller than he experimentally obtained [1, 10]. Several authors have also mentioned this discrepancy [11-14]. A theoretical expression for spin viscosity [15] was also derived that is about the same order predicted by Rosensweig's Eq. (1).

For instance, the ferrofluid EMG900_2 [3] with properties, η=4.5x10^{-3} [Pa-s], ρ=1030 [kg/m^3], $\zeta\approx1.5\eta\varphi_{vol}$=2.9x$10^{-4}$ [Pa-s], R=7 [nm], φ_{vol}=4.3%, the spin viscosity was estimated from fits to experiment to be $\eta' \approx 10^{-8}$-10^{-12} [N-s] while Eq. (1) and Eq. (2) give $l \approx 32.2$ [nm] and $\eta' \approx 4.67\text{x}10^{-18}$ [N-s].

2.3 Non-Uniform Magnetic Properties Driving the Flow

Other works [11, 14] show that non-uniform magnetic properties drive the ferrofluid flow. In addition, the assumption of a uniform magnetic field throughout the ferrofluid is incorrect [14] and the spin-up flow is attributed to the inherent non-uniform field generated within the ferrofluid due to the demagnetizing effects of the finite height cylinder.

Geometry of the material body plays a vital role when it is subjected to an external uniform magnetic field. If a material body of irregular shape is subjected to an external uniform field, the magnetic field inside the body is no longer uniform in direction and magnitude throughout the body. The magnetic field inside an ellipsoidal body's axis is given by

$$\mathbf{H}_{\text{internal}}=\mathbf{H}_{\text{external}}-N\mathbf{M} \qquad \textbf{(3)}$$

where the internal magnetic field $\mathbf{H}_{\text{internal}}$ [A/m] and magnetization $\mathbf{M}$ [A/m] inside the material are uniform if the external magnetic field, $\mathbf{H}_{\text{external}}$ [A/m] is also uniform where N represents the demagnetizing factor along the field direction.

Demagnetizing factors along major/minor diameters for ellipsoidal bodies in the x, y and z directions obey the following relation.

$$N_x+N_y+N_z=1 \qquad \textbf{(4)}$$

For the internal magnetic field to be uniform for an external uniform magnetic field the shape of the body must be ellipsoidal such as that of a sphere, a prolate or oblate spheroid, or an infinitely long cylinder.

A uniform externally applied field to a sphere results in a uniform internal magnetic field since a sphere has $N_x=N_y=N_z=1/3$. Only in an infinitely long cylinder with transverse magnetization with $N_x=N_y=1/2$ and $N_z=0$, is the field uniform. A finite height cylinder will have spatially non-uniform demagnetizing factors resulting in a non-uniform field within the ferrofluid volume. Such demagnetizing effects have been ignored in the works of [3-5].

3. Governing Equations

3.1 Ferrohydrodynamics

The fluid mechanics equations governing ferrohydrodynamics include conservation of linear and angular momentum equations [10]. The conservation of linear momentum equation assuming the flow is viscous dominated and incompressible ($\nabla \bullet \mathbf{v} = 0$) is

$$0 = -\nabla p' + 2\zeta \nabla \times \boldsymbol{\omega} + (\zeta + \eta)\nabla^2 \mathbf{v} + \mu_0 \left(\mathbf{M} \bullet \nabla\right)\mathbf{H} \qquad \textbf{(5)}$$

The conservation of angular momentum neglecting inertial terms is

$$0 = \mu_0 \mathbf{M} \times \mathbf{H} + 2\zeta\left(\nabla \times \mathbf{v} - 2\boldsymbol{\omega}\right) + \eta' \nabla^2 \boldsymbol{\omega} \qquad \textbf{(6)}$$

where the variables are dynamic pressure p' (including gravity) [N/m^2], fluid magnetization $\mathbf{M}$ [A/m], magnetic field $\mathbf{H}$ [A/m], spin velocity $\boldsymbol{\omega}$ [1/s], ferrofluid dynamic viscosity η [N-s/m^2], vortex viscosity $\zeta=1.5\eta\varphi_{vol}$ [N-s/m^2] for small volume fraction φ_{vol} of magnetic nanoparticles [10, 16], and η' [N-s] is the shear coefficient of spin viscosity.

A fluid mechanics module was used to represent the augmented Navier-Stokes equation in Eq. 5. A diffusion equation was used for the conservation of angular momentum equation (Eq. 6) with η' representing the diffusion constant.

3.2 Magnetic Field Equations

The ferrofluid magnetization relaxation equation derived by Shliomis [16] is

$$\frac{\partial \mathbf{M}}{\partial t} + \left(\mathbf{v} \bullet \nabla\right)\mathbf{M} = \boldsymbol{\omega} \times \mathbf{M} - \frac{1}{\tau_{eff}}\left(\mathbf{M} - \mathbf{M}_{eq}\right) \qquad \textbf{(7)}$$

where equilibrium magnetization $\mathbf{M}_{\text{eq}}$ [A/m] is given by the Langevin equation

$$\mathbf{M}_{\text{eq}}=M_s[\coth(\beta)-1/\beta](\mathbf{H}/H),\beta=(M_d V_p \mu_0 H)/kT \qquad \textbf{(8)}$$

with M_s [A/m] the saturation magnetization given as, $M_s=M_d\varphi_{vol}$, M_d = 446 [kA/m] is the domain magnetization for magnetite [10], V_p [m^3] is the magnetic core volume per particle, $\mu_0=4\pi\times10^{-7}$ [H/m] is the magnetic permeability of free space, k=1.38x10^{-23} [J/K] is Boltzmann's constant, T [K] the temperature in Kelvin, and effective relaxation time constant τ_{eff} [s] includes Brownian and Néel effects.

Maxwell's equations for a non-conducting fluid are

$$\nabla \bullet \mathbf{B} = 0, \nabla \times \mathbf{H} = 0, \mathbf{B} = \mu_0\left(\mathbf{H} + \mathbf{M}\right) \qquad \textbf{(9)}$$

The magnetic field can be given by

$$H = -\nabla \psi \qquad \textbf{(10)}$$

where ψ is the magnetic scalar potential and the magnetic flux equation can be given as

$$\nabla^2 \psi = \nabla \bullet \mathbf{M} \qquad \textbf{(11)}$$

Convection and diffusion modules were used to represent the magnetic relaxation equation (with zero diffusion) in the different Cartesian coordinates. Since Eq. 7 involves a time derivative, Eqs (5)-(7) were solved as a transient analysis in COMSOL with the time-derivative coefficients set to 0 for Eqs (5) & (6). The magnetic scalar potential was implemented using the AC-DC *Magnetostatics, No Currents* module.

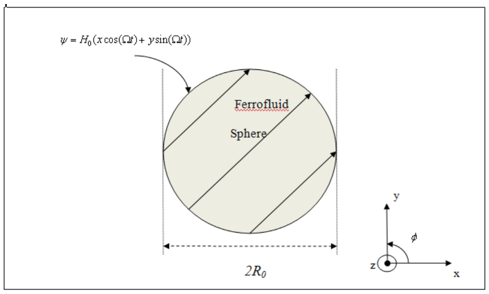

Figure 3. One region model setup with shaded circle representing ferrofluid and boundary condition on magnetic scalar potential. The scalar potential generates a magnetic field rotating in the φ direction at frequency Ω. This magnetic field represents the external magnetic field and has to be corrected for demagnetizing effects before being used in the magnetic relaxation equation. The arrows inside the stator show the uniformly distributed rotating magnetic field created inside the ferrofluid at a particular instant in time.

3.3 Boundary Conditions

Velocity boundary conditions are no slip on stationary boundaries ($\mathbf{v}$=0). If $\eta'\neq 0$ (spin-diffusion case) the spin boundary condition at the solid boundaries is taken as $\boldsymbol{\omega}$=0. If η'=0 there is no spin boundary condition. In the case of modeling the fully filled sphere in a uniform rotating magnetic field the magnetic potential boundary condition for a rotating magnetic field is given by

$$\psi(r = R_0) = H_0(x\cos(\Omega t) + y\sin(\Omega t)) \qquad \textbf{(12)}$$

where R_0 is the radius of the sphere and H_0 is the applied field strength. The pressure is also specified at one point. For the latter case involving the infinitely long cylinder with a non-uniform rotating magnetic field, the surface current boundary condition used is given as

$$\mathbf{K}(\varphi,t)=1.5K_0\cos(\Omega t-\varphi)\mathbf{i}_z \qquad \textbf{(13)}$$

4. Experimental and COMSOL Case Studies

4.1 Ferrofluid Filled Sphere in Uniform Rotating Magnetic Field

The equations are put into COMSOL Multiphysics in non-dimensional form normalized to a R_0=5 cm, Ω=190π [rad/s] (95Hz) and $\mu_0 H_0$=0.01T RMS (100 G). In this simulation, the applied field is assumed to not be strong enough to magnetically saturate the fluid. The equilibrium magnetization $\mathbf{M}_{eq}$ of the fluid is assumed to be in the linear regime of the Langevin equation and can be given by

$$\mathbf{M}_{eq}=\chi\mathbf{H}_{fluid} \qquad \textbf{(14)}$$

where χ is the fluid's magnetic susceptibility.

Since the sphere is modeled as a one region problem, the demagnetizing effect of the sphere has to be accounted for. In the presence of an externally applied uniform magnetic field ($\mathbf{H}_{xext}$,$\mathbf{H}_{yext}$,$\mathbf{H}_{zext}$) the field inside the sphere of ferrofluid ($\mathbf{H}_{xfluid}$,$\mathbf{H}_{yfluid}$,$\mathbf{H}_{zfluid}$) can be given by the following relation.

$$\mathbf{H}_{xfluid}=\mathbf{H}_{xext}-(1/3)\mathbf{M}_x,\ \mathbf{H}_{yfluid}=\mathbf{H}_{yext}-(1/3)\mathbf{M}_y,\ \mathbf{H}_{zfluid}=\mathbf{H}_{zext}-(1/3)\mathbf{M}_z \qquad \textbf{(15)}$$

The simulation results of a water based ferrofluid MSGW11 with ρ=1200 kg/m^3, η=0.00202 [Ns-m^2], τ_{eff}=1.39x10^{-5}[s], ζ≈8.3x10^{-5} [Ns-m^2], χ=0.56, φ_{vol}=2.75%, saturation magnetization ($\mu_0 M_s$) of 154 G and η' =4.79x10^{-9} [kg-m/s] [17] (average of the values reported by [4, 5]) gives a velocity profile as seen in Figure 4 that should be measureable using ultrasound, but negligible observable flow (<1mm/s) was obtained in the experiment. Ignoring the effect of spin-diffusion by using η'=0 in COMSOL gave negligible flow supporting the conclusion that spin-diffusion is not a dominant mechanism in spin-up flow.

4.2 Ferrofluid Filled Sphere in Non-Uniform Rotating Magnetic Field

To ensure that the driving mechanism for spin-up flow is due to non-uniform magnetic properties of the ferrofluid, the ferrofluid filled sphere was subjected to a non-uniform field imposed by a coil or permanent magnet, placed on top of the sphere, in addition to a uniform rotating magnetic field. The experiment resulted in significant complicated flows that were measured using ultrasound velocimetry. Figure 5 shows velocity flows obtained using oil-based Ferrotec EFH1 fluid in the presence of a non-uniform field imposed by permanent magnets and the uniform rotating magnetic field. The velocity can be seen to switch direction indicating the presence of complex vortices.

4.3 COMSOL Simulations of Ferrofluid Flows with Zero Spin Viscosity (η'=0) in an Infinitely Long Cylinder with Non-Uniform Magnetic Fields

COMSOL 3.5a simulations of the spherical geometry in a non-uniform rotating magnetic field were too difficult to be solved in 3D. Instead a similar problem was simulated using an infinitely long cylinder, which in a uniformly applied transverse external magnetic field would have a uniform internal magnetic field due to constant transverse demagnetizing factors of 1/2. The experiments with the sphere prove that spin-diffusion is not a dominant effect and simulations of an infinitely long cylinder, with η'=0, in a uniform rotating magnetic field also give negligible flow [17, 18].

The COMSOL model is setup as shown in Figure 6. In this case, the complete Langevin equation (Eq. 7) is used since the magnet does saturate the ferrofluid near it. The *Perpendicular, Induction Currents* module was used allowing for the specification of the y-directed magnetization of the magnet ($10<\alpha<40$ times the strength of the rotating field) and a surface current boundary condition on the outside stator generates a uniform rotating magnetic field. The COMSOL model was setup and normalized to R_{cyl}=5 cm, radian frequency Ω=190π [rad/s] and surface current $3/2K_0$=100 Gauss.

By subjecting the ferrofluid filled cylinder to a non-uniform field imposed by a nearby permanent magnet and the uniform rotating magnetic field, flow with vortices are generated similar to the experiments of the ferrofluid filled sphere in a non-uniform rotating magnetic field as seen in Figure 7. The non-uniform magnetization of the ferrofluid is also shown in Figure 8.

5. Conclusions

COMSOL Multiphysics helped predict a flow velocity, that should have been measureable using ultrasound velocimetry, using spin-diffusion theory for a fully filled sphere in a uniform rotating magnetic field. This result along with negligible flow obtained through experiments confirmed that spin-diffusion is not a dominant effect for spin-up flows. It also helped to explain that the previously reported experimentally fit values of spin viscosity in a finite height cylinder in a uniform magnetic field are orders of magnitude greater than theoretically derived values because they include demagnetizing effects associated with the shape of the container. Neglecting the effect of spin-diffusion by setting the spin viscosity (η') to zero gives negligible flow, as observed in experiments, in a fully filled sphere of ferrofluid subjected to a uniform rotating magnetic field.

Bulk spin-up flows are a result of non-uniform magnetic properties in the ferrofluid volume as obtained by experiments subjecting a fully filled sphere to a non-uniform rotating magnetic field generated by a permanent magnet placed on top of the sphere in a uniform rotating magnetic field. The complicated significant flows that result are confirmed using COMSOL Multiphysics, albeit in a 2D infinitely long cylinder subjected to a non-uniform rotating magnetic field imposed by a permanent magnet and a uniform external rotating field.

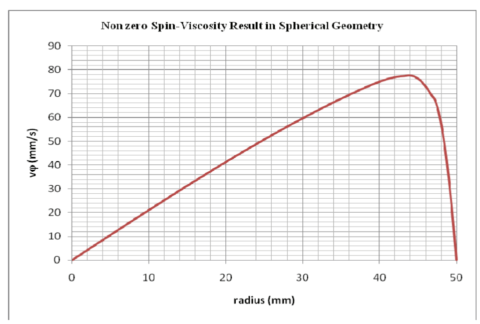

Figure 4. Dimensional rotational velocity for 10 cm diameter sphere of ferrofluid that would have been measured, with non-zero experimentally fit values of spin viscosity, in a uniform rotating magnetic field calculated using COMSOL Multiphysics 3.5a.

6. Acknowledgements

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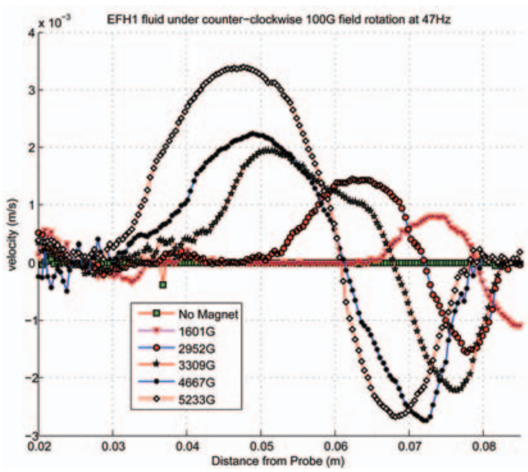

Figure 5. Measured flow velocity of Ferrotec EFH1 oil-based ferrofluid [saturation magnetization=421.2 Gauss, low field magnetic susceptibility χ=1.59] under a uniform magnetic field rotating counter-clockwise at 47 Hz in the presence of permanent magnets (1601-5233 Gauss) placed on top of the ferrofluid filled sphere (north pole of magnet making contact with the sphere) at z=5 cm that creates a large non-uniform DC magnetic field that causes the ferrofluid to have a non-uniform variation of effective magnetic permeability. Increase in the strength of the magnet increases the flow in a greater region of the ferrofluid filled sphere. The ultrasound probe is placed at the bottom of the ferrofluid filled sphere and measures the z-velocity along the line x= -1.71 cm with the sphere centered in the *x-z plane* at *x=z=0.* There is no flow with a uniform rotating magnetic field with no permanent magnet as the magnetization of the ferrofluid is uniform.

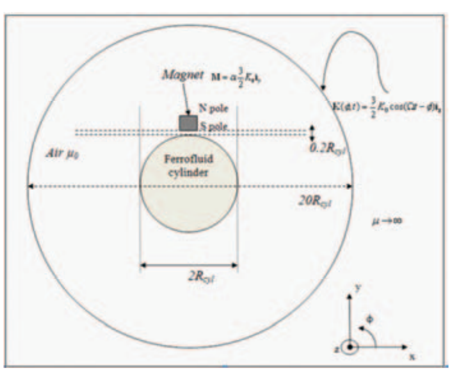

Figure 6. Model setup for simulating flows in ferrofluid under the influence of the non-uniform field of the permanent magnet and the uniform rotating magnetic field imposed by a current boundary condition far away. The magnet placed at a distance 0.2R_{cyl} above the cylinder is magnetized in the y direction and is α (10-40) times the strength of the rotating field.

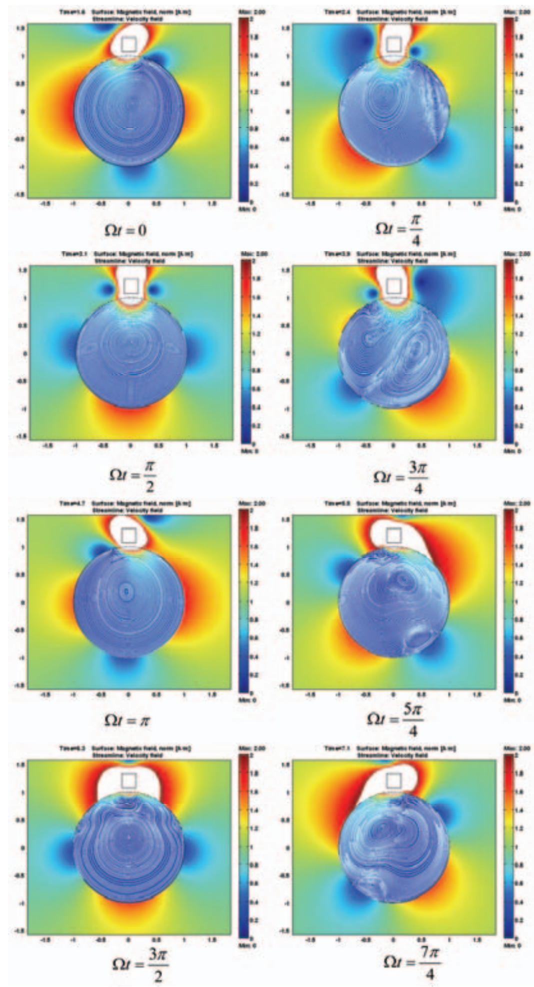

Figure 7. COMSOL 3.5a calculated distribution of velocity streamlines with $\eta'=0$, illustrating vortices formed, for a Ferrotec MSGW11 filled infinitely long cylinder, with a 2000 Gauss permanent magnet in the 100 Gauss RMS uniform counter-clockwise rotating field, as a function of time over one period of rotation at an angular frequency of Ω=190π [rad/s]. The magnetic field magnitude is represented by the colored surface plots which show the evolution in time of the total magnetic field inside and outside the ferrofluid cylinder due to the uniform rotating magnetic field, the ferrofluid cylinder's outside line dipole field, and the non-uniform magnetic field due to the permanent magnet. The white region represents magnetic field strengths near the permanent magnet that are beyond the scale shown. The total magnetic field colored surface plots are normalized to the strength of the rotating field of 100 Gauss and are plotted up to 200 Gauss. Dimensional magnitude of velocity is calculated to be on the order of 3-30 mm/s according to these simulations which corroborates with experimental results in spherical geometry.

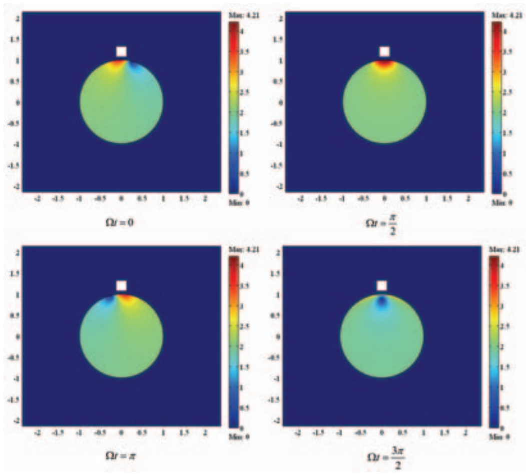

Figure 8. COMSOL 3.5a calculated distribution of magnetization magnitude in an EFH1 ferrofluid filled infinitely long cylinder with $\eta'=0$ as a function of time in a non-uniform magnetic field generated by a 1000 Gauss permanent magnet with a 100 Gauss RMS uniform rotating magnetic field at angular frequency $\Omega=190\pi$ [rad/s]. The colored surface plot of magnetization magnitude is normalized to the strength of the uniform rotating magnetic field. The blue circle which represents a weak field, due to near complete cancellation of the rotating magnetic field and the field due to the permanent magnet, can be clearly seen to rotate in an arc near the top of the sphere. The red region represents the strong field that saturates the EFH1 with a normalized saturation magnetization of 421/100=4.21.

7. References

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