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Magnetic-field assisted mixing of liquids using magnetic nanoparticles

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ABSTRACT

Size and magnetic properties of magnetic nanoparticles (MNPs) in fluids allow special remote control of fluid flow using appropriate externally applied magnetic fields, especially when submicronic mixing is critical, inter alia, for catalytic reactions, separation and drug delivery. This work explores MNPs as nanoscale devices to control mixing at microscale by submitting the system of interest to a rotating magnetic field (RMF). Magnetic nanoparticles are harnessed by RMF and converted into nanostirrers thereby generating MNP-pinned localized agitation in the liquid phase. Using this technique, self-diffusion coefficient of water in a static diffusion cell was intensified up to 200 folds. Also, axial dispersion of capillary Poiseuille flows under RMF underwent a reduction prompted by MNP-mediated intensification of lateral mixing relative to that in absence of magnetic field. Finally a multiphase flow case concerned gas–liquid mass transfer from oxygen Taylor bubbles to the liquid in capillaries where dilute MNP solutions led to measurable enhancement of k_La under RMF.

1. Introduction

Molecular transport and diffusion in liquids play a key role in many different contexts of physics, chemistry, biology and chemical engineering [1,2]. Quite recently, an emerging trend in sciences and engineering attempts to enhance liquid phase transport operations beyond what can be accomplished in the classical diffusion paradigm by seeding fluids with an ensemble of nanoparticles [2]. For example, it has been stated that the presence of nanoparticles in liquids (so-called nanofluids) may modify heat [3–5] and mass transport [6–14] properties of the medium. Most prominently, magnetic nanoparticles (MNPs) have been applied in a few studies to direct mass transfer in absence [8,15] or in presence [7,16] of an external magnetic field. Although, the general effect of nanoparticles on liquid transport properties is still anomalous [17], the method consisting of an excitation of MNP-laden liquids with a time-varying external magnetic field appears to hold promises to achieve process intensification. In this contribution, we propose to expand the horizon of MNP possibilities by unveiling magnetically induced nanoconvection mechanism to intensify mass transport beyond the scope of molecular diffusion. A related nanomixing process is enabled when stable suspensions of single-domain MNPs are exposed to a uniform rotating magnetic field (RMF).

Briefly, external magnetic fields exert magnetic torque on the magnetic moment of MNPs suspended in liquids thus forcing the nanoparticles to align with the magnetic field direction [18]. For those MNPs whose magnetic moment is locked in the solid crystal structure (so-called rigid-dipole rdMNP), magnetic torque is felt bodily and associated momentum is transferable to the adjacent liquid phase [18]. Interestingly, the nature of mechanical interactions between magnetically excited MNPs and the bulk of liquid depends on the characteristics of applied magnetic field. For instance, a time-varying magnetic field such as RMF exerts an angular torque on MNPs forcing nanoparticles gyration relative to the contiguous liquid. In their quest to follow the ever-changing RMF, rdMNPs spin in a direction primarily imposed by the rotating magnetic field [18]. Therefore, a rigid-dipole MNP, by virtue of its Brownian relaxation pattern [18], spins bodily in a rotating magnetic field. Spinning nanoparticles in dilute colloidal suspensions – where magnetic mutual interactions among MNPs are negligible – [19,20] transfer their momentum to the surrounding liquid molecules and thus locally impart to the liquid an angular motion. Hence, nanoscale mixing effects around individual nanoparticles may be expected in so far as each MNP spins as an independent nanostirrer. A stationary magnetic field, unlike RMF, locks the magnetic moment and proscribes the nanoparticle gyration under

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shear flow. Because of the fluid viscous character, the pinned nanoparticles transfer their magnetic torque to their adjacent liquid. This torque may, on the contrary, give rise to nanostagnant zones around MNPs hindering therefore the mixing properties of the nearby liquid.

If such picture proves true, the potential of nanomixing/hindrance induced by rotating magnetic fields is expected to bear enormous implications in fields where molecular diffusion is the prime limiting factor for transport phenomena, e.g., mass/heat transfer in laminar-flow fluidics [21], diffusion-limited uptake around high-turnover-frequency catalyst sites in synthetic or physiological fluids [22,23].

The purpose of this contribution is to provide experimental data on transport phenomena measurements in the presence of magnetically excited MNPs suspended in liquids. In particular, this research is focused on mass transfer enhancement by means of magnetically excited MNPs to unveil new enhancement mechanisms with relevance to chemical engineering. Several simple hydrodynamic systems were built to examine the interplay between magnetic fields and transport phenomena. More specifically, three different hydrodynamic instances were considered: motionless liquid in a diffusion cell, Poiseuille flow and two-phase Taylor flow in capillaries. For each experiment, an apparent diffusion coefficient was measured using standard experimental techniques. It is worthy of notice that this approach stands out from other magnetic mixing processes such as those relying on an application of magnetic Kelvin force which are restricted to the boundary of magnetic and non-magnetic fluids [24] or from those systems that induce magnetic mixing in electrolytic media [25–28].

The first set of experiments aims at measuring liquid self-diffusion coefficients in dilute MNP suspensions retained in a diffusion cell centered horizontally inside the magnet bore and kept under pure hydrostatic conditions to assess nanoconvective mixing upon triggering a magnetic field.

The second set of experiments consists of axial dispersion coefficient (K) measurements in capillary Poiseuille flows to identify the effect of magnetically excited MNPs on Taylor dispersion and laminar velocity profile under various types of magnetic fields. Taylor dispersion is a phenomenon originating from combination between axial convective displacement of a tracer slab under laminar Poiseuille flow and its diffusive radial migration [29]. Hence in a given experimental condition, any phenomenon that alters molecular diffusion could be detected indirectly through its effect on K [29]. Residence time distribution (RTD) measurements are thus performed using a Taylor dispersion capillary cell from which axial dispersion coefficients are obtained. MNPs in our Poiseuille experiments were excited by rotating magnetic fields to probe their influence on the lateral molecular transport mechanism in a capillary flow. Also, we put forward an approach to correlate (and infer from impulse RTD) the shape of laminar velocity profile under magnetic fields from the breakthrough time of the capillary tube.

The last part is devoted to an investigation of gas-liquid mass transfer coefficient (k_La) in a microchannel in the so-called Taylor flow regime. This regime corresponds to an alternating succession of equally long bubbles intertwined between liquid slugs [30]. We attempt to broaden the scope of MNP-mediated mixing toward the Taylor flow regime as the most commonly observed gas–liquid regime in microchannels [31]. Although in such contactors radial mixing is improved in the vortex region due to the recirculation patterns within liquid slug, molecular diffusion seems to play a prominent role in the micron-size thin lubricating film which encloses the bubbles [32]. Thus, our goal is to unveil how magnetic nanoparticles seeding the liquid, whether or not stimulated by magnetic fields, will affect gas–liquid mass transfer.

Table 1
Magnetic properties of EMG 705 from magnetometry measurement.

Saturation magnetization, M_s (kA/m)	18.7
Initial susceptibility, χ_0	2.9
MNP volume fraction, ϕ (v/v)	0.042
Estimated median magnetic core diameter, d_p (nm)	16.0

2. Experimental

2.1. Colloidal suspension

Dilute ferrite-in-water nanoparticle suspensions (ϕ = 0.001–0.01, v/v magnetic content) were prepared from a commercial ferrofluid, EMG705 (FerroTec). The magnetic properties of EMG 705 were measured by an alternating gradient magnetometer, MicroMag model 2900 (Princeton Instrument Co.) at 298 K in low-field (for initial susceptibility, χ_0) and high-field (for saturation magnetization, M_s) asymptote of magnetization curve. Using these values, particle magnetic core diameter was estimated following a method proposed by Chantrell [33]. Table 1 summarizes the magnetic properties of EMG-705 ferrofluid. Likewise, the particle size distribution of the dilute ferrofluid suspensions was measured via magnetometry for different MNP concentrations. The hydrodynamic diameter of MNP suspension was also measured based on dynamic light scattering technique on a Zetasizer Nano 6 (Malvern Instruments Ltd.). The particle size distribution with number average diameter and standard deviation, respectively, of 24.8 nm and 7.25, were registered. The particle hydrodynamic diameter thus measured was found to be larger than the core MNP diameter and is consistent with literature findings. It was verified that cluster or chain formation during the course of experiments was highly unlikely at these dilute concentrations [19,20]. Similarly, particle mutual interactions may be neglected [20] considering the average MNP particle–particle distance is several times longer than the MNP diameter, e.g., $d_{p\text{-}p} \sim 10^2$ nm for ϕ = 0.0025 [18]. Thanks to the small particle sizes (ca. 16-nm magnetic core), MNPs can be easily dispersed by thermal agitation while the surfactants decorating the EMG705 ferrofluid particles prevent them from sticking to each other under short-range Van der Waals attraction forces. For these reasons, any enhancement or retardation in transport phenomena can be interpreted as stemming from MNP individual manifestations under magnetic field.

2.2. Magnet

A tubular two-pole three-phase magnet was designed and built in collaboration with MotionTech LLC (Arizona, USA) and Windings Inc. (Minnesota, USA) with bore dimensions as 55 mm height and 45 mm inner diameter. The three-phase stator assembly (Fig. 1a) consists of a magnet stator with three identical coil pairs, spatially shifted from each other by 120° in azimuthal direction (Fig. 1b). Each coil can be energized separately or coupled to other coils in various configurations (Fig. 1c). Therefore, the magnet can be used to generate different magnetic field types including rotating (RMF), static and oscillating magnetic fields with moderate intensity at the center axis (up to 50 mT). In this work, only rotating magnetic fields will be illustrated where in absence of any magnetic object, each ampere-rms generates a RMF of nearly 186 Gauß at the center of the bore. The resistance and inductance of the stator windings were measured as 14.6 Ω per coil (single winding) and 150 mH per coil at 200 Hz, respectively.

To generate a RMF coils are fed by three balanced AC currents, 120° out of phase from a variable frequency drive (ABB, ACS150,