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Short Communication

Challenges associated to magnetic separation of nanomaterials at low field gradient

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ABSTRACT

Magnetic nanoparticles (MNPs) have been proposed as one of the effective tools for pollutant removal from aqueous environment. In most of the new strategies investigated, which involved the use of MNPs, the ability to reharvest back this nanomaterial by an externally applied magnetic field is always being emphasized. In this short communication, we discuss the challenges associated to the magnetic separation of MNPs from its suspending media through magnetophoresis under low magnetic field gradient. We highlight the major constraints, such as thermal energy, Stokes drag and gravitational pulling, which influence the successful separation of MNPs from aqueous environment by low gradient magnetic separation (LGMS). Dimensionless numbers are introduced to provide a more quantitative comparison between the aforementioned constraints with magnetophoresis at low field gradient. Finally, we focus our discussion on the role of (1) guided/self-assembly approaches and (2) on-site LGMS strategy as the most practical routes of using MNP for water remediation.

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1. Introduction

In last decade, we have observed a booming of research findings on the huge potential of magnetic nanoparticles (MNPs) in removing dangerous pollutants, such as arsenic [1], heavy metals [2], chlorinated compounds [3], and also organic dyes [4] from water resources. One of the great advantages of this MNP based water treatment technique is the recollection ability of MNPs, which can be easily achieved by using a hand held permanent magnet [1,4], after the hazardous compound was adsorbed onto the particle surfaces. The underlying principle behind this separation technique is remarkably straightforward. It relies on the simple fact that the magnetic materials experience magnetophoretic force in the presence of magnetic field gradients and thus these materials can be physically separated out from the surrounding fluids by a magnetic source. In addition, MNPs can also be employed to impart a magnetic dipole moment to biological cells, through immobilization on the cell surfaces, which subsequently leading to magnetophoretic separation of biological substances [5].

The rapid magnetophoretic separation of MNPs under low magnetic field gradient ($\nabla B < 100$ Tesla/m), as observed by others is very likely through field-induced reversible aggregation of

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particles [6]. Under this scenario, the particle clusters formed would migrate to the region where the magnetic field gradient is the highest. Along its migration pathway, the moving MNP clusters collide with each other and integrated into larger aggregates with higher magnetophoretic velocity [6,7]. This mechanism is the key factor for successful separation of MNPs in real time and revealed the opportunity for the implementation of low gradient magnetic separation (LGMS) for engineering applications.

In contrast to conventional industry practice in which high gradient magnetic separation (HGMS) is normally being employed, the design rules for LGMS is ill-defined and poorly understood. Moreover, the key parameters involved for implementation of LGMS in water treatment technology are also not being fully explored yet. Recently, Mandel and Hutter have briefly discussed the problems related to MNPs separation [8]. They raised an interesting point in which the use of ferrofluid as a nanoemulsion provides better alternative for water treatment purpose compared to easily agglomerated MNP suspension. However, the liquid-liquid interface between the nanoemulsion and the aqueous media can be the major barrier toward the full realization of this noble idea. Nevertheless, the key question here is how the nanosized magnetic particles can be used effectively for water treatment, and more importantly, the recollection of these particles from their suspension. It is the purpose of this paper to illustrate some general rule of thumbs related to the separation of MNPs under low field gradient.







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2. Underlying problems associated to LGMS

It is often being illustrated that by introduction of a magnetic field, the separation of MNPs from aqueous environment can be made possible in real time as shown in Fig. 1. Even though, the separation time involved is very feasible for practical usages at lab scale, the possibility for this kind of setup to be fully implemented for water treatment purposes is as good as none. By taking the example of a cylindrical NdFeB magnet, the magnetic field B_x along its symmetry axis as a function of the distance x away from the magnet pole face can be estimated quite accurately by following equation [9]:

$$B_{x} = \frac{B_{r}}{2} \left[\frac{x+L}{\sqrt{(L+x)^{2}+R^{2}}} - \frac{x}{\sqrt{x^{2}+R^{2}}} \right]$$
(1)

where *R* is the radius of the cylinder, *L* is the length of the cylinder, B_r is the remanence or residual induction of the magnetic material. For a grade N50 NdFeB, B_r is about 1.45 Tesla. This calculated B_x has a very good match with experimentally measured results [10] and for the permanent magnet shown in Fig. 1, with a dimension of R = 0.7 cm and L = 1.5 cm, its magnetic field B_x decay rapidly from the magnet pole face (Fig. 2). Once, the MNPs has been released into environment for water treatment purposes, there is no such magnet or magnetic separation devices/strategies can be employed to cope with the length scale involved, up to kilometers, in re-harvesting them back. For an example, according to Eq. (1), in order to generate an appropriate field gradient to achieve LGMS at separation distance of 1 km (x = 1000 m) would require a cylindrical NdFeB magnet (at the same aspect ratio of the magnet shown in Fig. 1) with radius of 140 m and length of 300 m.

This analogy bring out an important message in which for any in situ water treatment technology that involved the usages of MNPs, it is misleading to emphasize on its magnetic separation. Once being released into the environment there is no way for MNPs to be re-harvested back, at least not by magnetic separation. Thus, it is more appropriate for the implementation of MNPs under the setting of a on-site treatment facility. Under this context, the application of LGMS as a downstream separation unit can be both economical and technological feasible [5]. For engineering application, the (a) inhomogeneous field gradient and (b) complex distribution of magnetic field lines in three dimensional space of a permanent magnet can be extremely challenging to be properly integrated into a LGMS system.

3. Transport behaviors of MNPs due to "nanosize effects"

By taking non-interacting particles assumption, at magnetic field *B*, the magnetophoretic force F_{mag} needed to induce separation of spherical MNPs is [11,12]:

$$F_{mag} = \frac{4}{3}\pi r_{pt}^3 (M\cdot\nabla)B \tag{2}$$

where r_{pt} and M are the radius and the magnetization (per unit volume) of the MNP, respectively. By equating the F_{mag} with viscous



Fig. 2. Magnetic field B_x extended out from a cylindrical NdFeB magnet with radius of 0.07 m and length of 0.015 m. B_x is calculated by using Eq. (1).

drag force ($F_{drag} = 6\pi\eta r_{pt} \cdot u_{mag}$) experienced by a sphere [13], the magnetophoretic velocity u_{mag} can be calculated as

$$u_{mag} = \frac{2r_{pt}^2}{9\eta} (M \cdot \nabla)B \tag{3}$$

where η is the viscosity of the suspending medium. For the case of very weak magnetic field, Eq. (2) can be further simplified to [14]:

$$F_{mag} = \frac{\Delta \chi V_p}{\mu_0} (\nabla B) B \tag{4}$$

where μ_0 is the vacuum permeability, V_p is the particle volume $(V_p = 4\pi r_{pt}^3/3)$ and $\Delta \chi$ is the different in magnetic susceptibility between the particle and the fluid. Eq. (4) raised an interesting observation, in which the F_{mag} experienced by a nanoparticle is not only dependent on the magnetic field and field gradient but the volume of the MNP involved is equally important. For any design purposes which involved transport behaviors of MNP, knowing the value of u_{mag} is vital as various transport phenomena analyses can be performed [15].

Since the particle size is in nanometer range, its motion is heavily influenced by thermal energy and viscous drag; hence, conventional dimensionless number analysis can be very helpful to characterize the flow behavior. Table 1 summarized some of the useful dimensionless numbers which are familiar to the chemical and mechanical engineers and can serve as an effective way to rationalize the transport behavior of MNPs under magnetophoresis. By using the value of *B* and ∇B as shown in Fig. 2 and hydrodynamic radius of the MNPs (Fig. 1) at 150 nm as determined by dynamic light scattering (DLS), the Reynolds (Re), Péclet (Pe) and Froude (Fr) numbers with respect to the separation distance x from the pole surface of NdFeB magnet can be calculated (see Fig. 3a). Here we used the mathematical equations presented in Table 1 for the calculation of each dimensionless number and according to Eq. (3) the MNPs would typically having a magnetophoretic velocity within the range of $0.5 \,\mu$ m/s to around 9000 μ m/s. As



Fig. 1. Magnetophoretic collection of iron oxide MNPs with ~150 nm hydrodynamic radius in real time.

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