Modelling the size separation of NdFeB magnetic microparticles by magnetophoresis and gravity settling

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Abstract—**Properties of sintered NdFeB magnets strongly depend on granulation and size distribution of constituent particles, which requires an efficient method for their separation into relatively narrow size fractions. We investigated two methods of magnetic particle separation from a mixture with different sizes using simulation by finite element method: magnetophoresis and gravity settling. In the case of magnetophoresis magnetic particles ranging in diameter from 1 to 10 μm were deflected from the direction of continuous laminar flow by a perpendicular magnetic field. Larger particles were deflected from the direction of laminar flow more than smaller particles. The applied flow rate and strength and gradient of the applied magnetic field were the key parameters in controlling the deflection. The gravity settling model simulated spherical particles falling in heptane. Particles of various sizes were divided according to the time they needed to reach the bottom. The model used an axially symmetric fluid-flow simulation in a moving coordinate system connected with the particle, coupled with an ordinary differential equation for the force balance of the particle (gravity and drag force). The grain accelerated from standstill and rapidly reached its terminal velocity. This velocity was approximately proportional to the square of the particle radius, which led to clear separation of 5-10 μm particles from those with a diameter of 1 μm.**

***Keywords*—NdFeB particles, separation, gravity settling, magnetophoresis.**

I. INTRODUCTION

AMONG different permanent magnets, those based on lanthanides ensure the highest magnetic fields, typically in excess of 1.4 T. Within this group, magnets utilizing nanocrystalline NdFeB [1, 2] offer the highest values of magnetic energy densities, in the range of 30-50 MGOe [3]. At the same time, they offer a relatively reduced rare-earth content. Usually such nanocrystals are bonded or sintered to produce extremely strong permanent magnets. It is of obvious interest for many applications to produce submicrometer particles of NdFeB materials for use in strong permanent magnets, since this results in a decreased weight of the magnet while at the same time the available power density is increased.

The first technological step toward NdFeB permanent magnets is to fabricate starting macroscopic particles. These can be obtained in different ways, including milling, HDDR (Hydrogenation, Disproportionation, Desorption, and Recombination), mechanical alloying, etc. [4]. As a next step it is necessary to further reduce the size of thus obtained particles to the micrometer or submicrometer range. This is usually done by fine milling, utilizing either jet milling or ball milling.

The particles processed to reduce their size will include at the end of the process a range of different particle sizes, various irregular shapes. The milling process will also cause particle agglomeration to some extent. This is the reason why a third technological step is needed, the size selection and separation [5].

Various methods are available for submicrometer particle separation. These include the use of membranes for filtering, settling from a liquid suspension, different phoretic methods (hydrophoresis, thermophoresis, magnetophoresis...), etc. [6].

Some industrial applications of strong permanent magnets require large amounts of magnetic materials. Such are for instance next generation electric motors, which are of interest for automotive applications, i.e. for modern hybrid and purely electrical vehicles. Thus it is interesting to consider methods that can be upscaled to separate NdFeB submicrometer particles at an industrial scale. Two nondestructive methods for selectively collecting or separating magnetic particles are gravity settling and magnetophoresis. In both cases magnetic particles float in a viscous liquid.

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Gravity settling or gravity sedimentation [7] is a process in which particles settle from liquid suspension due to the force of gravity and fall to the bottom. It can be applied even in the case of nanofluids, i.e. suspensions of nanoparticles with dimensions below 100 nm [8]. Magnetophoresis [9-12] is the process where magnetic particles move in a viscous liquid under the influence of magnetic field, so that combined acting of magnetic, gravitational and drag forces influence the mobility of different sizes and shapes of magnetic particles, thus causing their separation.

In this contribution we consider the applicability of the methods of gravity settling and electrophoresis for the size separation of our NdFeB particles produced by milling. To this purpose we utilize finite element method to simulate the distribution of particles when these separation methods are applied. We consider realistic conditions as encountered in our system for submicrometer particle production.

II. THEORY

In this section we briefly consider the basics of the two chosen methods for separation of different sizes of magnetic particles, the gravity settling and magnetophoresis.

1) Gravity settling

When using simple gravity settling to sediment particle from viscous liquid, two main forces act on each particle. One of them is gravity, and the second one is the drag force caused by the motion of the particle through the viscous liquid. The gravity force is not affected by the particle velocity, whereas the drag force is a function of the particle velocity.

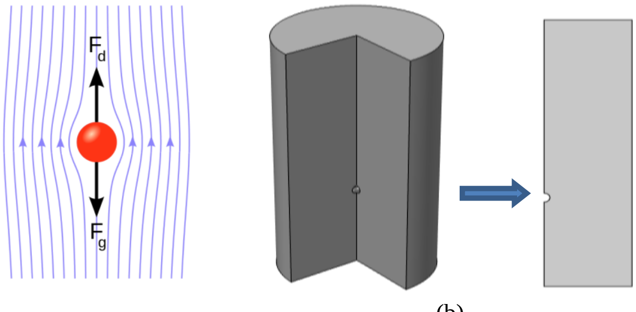
As the particle velocity increases due to gravitational acceleration, there is some point where the drag force and the gravity force become approximately equal. After that, no further changes of the particle's velocity occur. This velocity is known as the terminal velocity (settling velocity or fall velocity) of the particle. The terminal velocity is readily measurable using the rate of fall of individual particles. The terminal velocity is most notably dependent on the particle size, its shape (roundness and sphericity) and the density of the particles, as well as on the viscosity and the density of the fluid.

When a particle moves with a low velocity through a fluid described by a small Reynolds number, which is a situation corresponding to our case, the drag force caused by viscous friction is defined by the Stokes law as

$$F_d = 6\pi\eta R v. \tag{1}$$

where F_d is the frictional force known as Stokes' drag that acts on the interface between the fluid and the particle, η is the dynamic viscosity of fluid, R is the radius of the spherical object, and v is the flow velocity relative to the object. This expression is valid if the fluid flow is laminar and if the spherical particles do not interact. The fluid flow is described by the Navier-Stokes equations.

When the particle accelerates due to gravity, the drag force acts in the direction opposite to the particle's motion, preventing further acceleration (Fig. 1a). Our numerical model couples the flow simulation in cylindrical coordinates with an ordinary differential equation for the force balance of the particle. Due to axial symmetry, to facilitate calculation and decrease processor time, we modeled the flow in 2D instead of 3D. The Fig. 1b shows the modeling domain.



(a) (b)

Fig.1. (a) Creeping flow past a sphere: streamlines, drag force F_d and force by gravity F_g. (b) The modeling domain.

2) Magnetophoresis

In this case the viscous medium containing NdFeB particles may be a magnetic or non-magnetic liquid. The external magnetic field is graded. In magnetophoresis the direction and the velocity of the particles a gradient magnetic field are determined by magnetic (F_m) and drag (F_d) forces. For most magnetophoretic applications involving (sub)micron particles, the magnetic and viscous forces are dominant, while all other effects, including the gravitational force, can be neglected. By selectively controlling magnetic and drag forces it is possible to control the motion of the different size fractions and, therefore, magnetically separate mixtures into discrete size groups.

Our analysis was performed to simulate transport and separation of magnetic NdFeB microparticles in a magnetophoretic system that consists of an array of integrated magnetic elements embedded beneath a microfluidic channel serving as separation chamber over which a laminar fluid flow occurs. Perpendicular to the direction of laminar flow, a gradient external magnetic field is applied. A mixture of magnetic particles of different sizes is injected continuously into the system through the inlet channel. Magnetophoretic separation as described here is based on deflection of the particles from the direction of fluid flow. The deflection of the magnetic particles, u_{defl} can be described as the sum of two vectors: the vector for the magnetically induced flow velocity on the particle, u_{mag}, and the vector for the velocity of the hydrodynamic flow u_{hyd}:

$$u_{defl} = u_{mag} + u_{hyd}. \tag{2}$$

The magnetically induced flow, u_{mag}, is the ratio of the magnetic force, F_m , acting on the particle and the viscous drag force

$$u_{mag} = \frac{F_{mag}}{F_{drag}} = \frac{F_{mag}}{6\pi\eta R v} \qquad (3)$$

The magnetic force, F_m, is proportional to the magnetic flux density, B, and the gradient of the magnetic field of the externally applied field. The magnetic force is also proportional to the particle volume, V_p, and the difference in magnetic susceptibility between the particle and the surrounding fluid, $\Delta\chi$

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

where μ_0 is the permeability of vacuum. When introducing eq. 4 into eq. 3, it can be seen that the magnetic velocity, u_{mag}, is dependent on the size and on the magnetic characteristics of the particle for a given magnetic field and a given viscosity. It is proportional to the square of the particle radius and to the magnetic susceptibility of the particle

$$u_{mag} \sim R^2 \chi_p \qquad (5)$$

Hence, particles that are either different in size, R, or different in their magnetic susceptibility, χ_p, will be deflected from a laminar flow to a different degree, thus enabling particle separation.

III. SIMULATION

1) Gravity settling

We performed our simulation by the finite element method using the commercial software package Comsol Multiphysics. We modeled a system of spherical magnetic NdFeB particles ($\rho = 7400$ kg/m^3) with different radii (R = 0.5, 1, 2.5, and 5 μm) in heptane ($\rho = 679.5$ kg/m^3, $\eta = 0.386 \cdot 10^{-3}$ Pa·s). We assumed that in the initial moment both particle and fluid are immobile. We connected the reference system with the particle and followed the relative changes of fluid velocity.

At the surface of the magnetic sphere the fluid velocity relative to the sphere is zero. Fig. 2a shows the velocity distribution at the final simulation time, when the grain (R = 1 μm) has reached steady state. Fig. 2b shows the falling velocity of the same particle as a function of time.

Figure 3 shows the dependence of the terminal velocity of the particle for different particle radii. This velocity is approximately proportional to the square of the particle radius. In this case this means that for a period during which all particles with a 5 μm radius reach the bottom of a vessel with a height of 50 cm (approximately 11 minutes), only those particles with a 1 μm radius will reach the bottom which were at the beginning at a height of 2.5 cm or less. The smallest particles will practically not move at all.

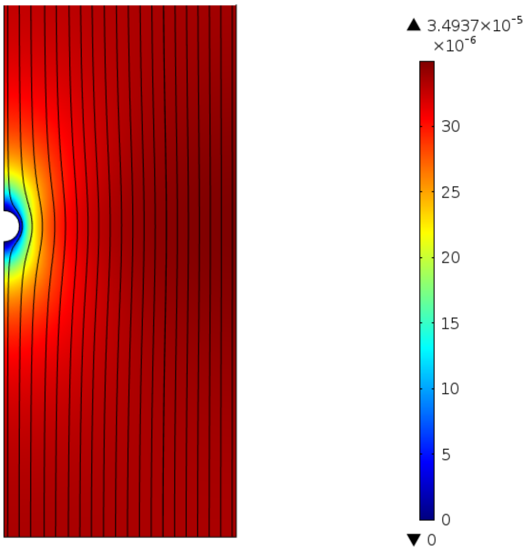


Fig. 2a. The velocity field at steady state. The velocities (in m/s) are plotted in the reference system of the sand grain.

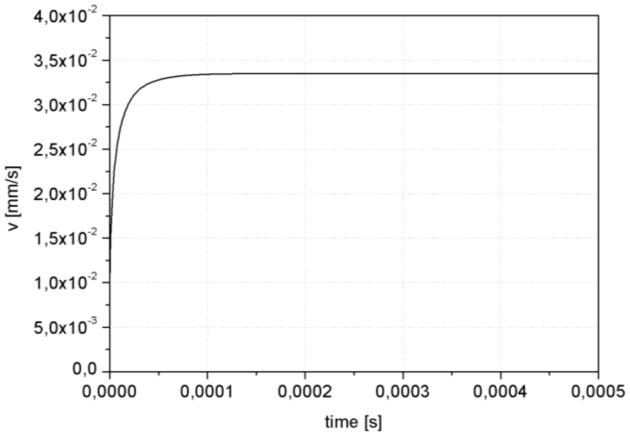


Fig. 2b. Falling velocity of the particle r = 1 μm versus time. After 0.1 ms, the velocity approaches the terminal velocity.

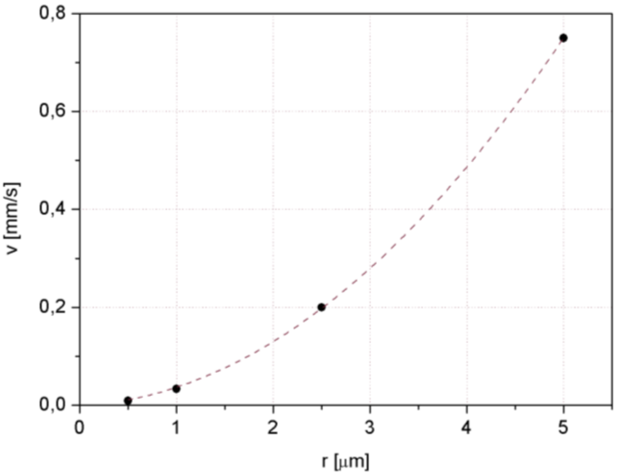


Fig.3. Velocity of gravity settling versus the radius of a spherical particle

2) *Magnetophoresis*

The modeling domain is shown in Fig. 4. Small and strong permanent magnets with $\mu_r = 1.05$, and magnetization $M = 5.510^5$ A/m) are used to generate a magnetic field within the separation flow chamber. The assumed dimensions of these magnets are 30 mm x 20 mm. Assemblies of two magnets are used to increase the magnetic flux density, and arrays of soft iron magnets direct the flux toward the separation chamber.

The dimensions of the fluid flow cell were 5 mm x 20 mm, with an array of output openings 0.5 mm wide, the spacing between openings of 0.1 mm. This simulation was also done by the COMSOL Multiphysics package, using the coupled Magnetic field, Creeping flow and Particle Tracing modules.

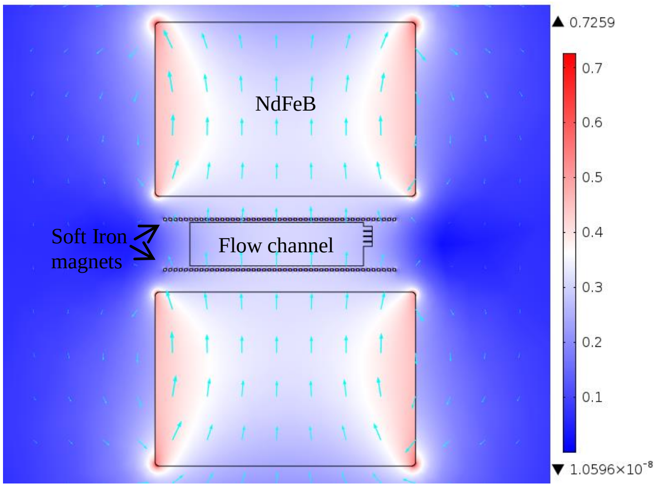
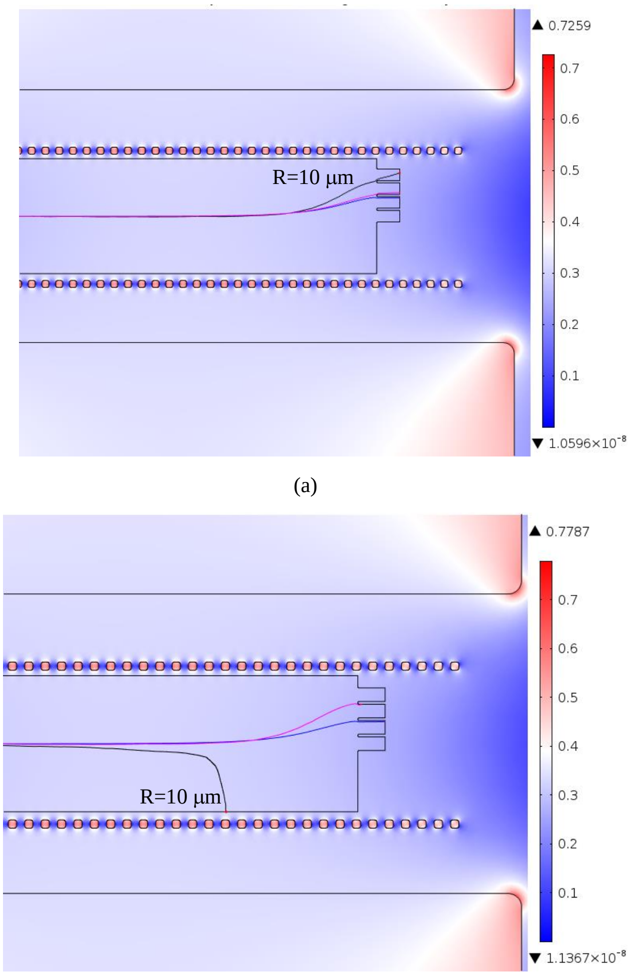


Fig. 4 Magnetic field distribution within the whole simulation domain. Surface color denotes magnetic induction intensity, arrows show the direction of magnetic induction vector.

Simulations using the mentioned three modules were performed successively; the results from the previous were used as the input for the following simulation. First we calculated the magnetic field distribution. Fig. 4 shows the distribution of the magnetic field. The magnetic induction within the microchannel is 0.3 T to 0.4 T. The input data for the simulation of fluid flow are the velocity at the channel entrance (v_f) and the pressure at the output, as well as the *no slip* boundary condition at the channel walls. We assumed that the carrier fluid was nonmagnetic (water, $\mu_r = 1$). The results of these two simulations formed the input to the Particle Tracing module, serving to calculate the drag force and the magnetophoretic force, the ratio of which determined the total deflection of the particle.

The particles in simulation were again spherical NdFeB particles ($\rho = 7400$ kg/m^3, $\mu_r = 1.05$), their radii being 1 μm, 5 μm and 10 μm. Fig. 5a shows the trajectories for these three particle sizes if the fluid input velocity is $v_f = 0.85$ mm/s and $M = 5.5$e5 A/m. It can be seen that the largest deflection is obtained for 10 μm particles, its value being ~1 mm larger than the deflection of the 5 μm particles.

Changes of the fluid velocity and the magnetization of permanent magnets significantly influence the shape of the trajectory of magnetic particles. Fig. 5b shows trajectories of the same particle when $v_f = 0.25$ mm/s, and $M = 5.9$e5 A/m. In this case the motion of the largest particle is such that it remains confined within the flow channel.



(a)

(b)

Fig. 5 Trajectories of the NdFeB particle with different radius under magnetophoretic/drag forces, for two values of the flow velocities: v_f=0.85 mm/s (a), and v_f=0.25 mm/s (b). Surface color denotes magnetic induction intensity.

IV. CONCLUSION

We applied finite element method utilizing the Comsol Multiphysics package to numerically simulate two methods for magnetic particle separation from a mixture with different sizes, the magnetophoresis and gravity settling. We assumed spherical particle shapes with diameters 1 to 10 μm. Realistic process vessel geometries and sizes were used. The calculations showed that the deflection from laminar flow in the case of magnetophoresis is proportional to the particle size. The fluid flow, the particle magnetization and the external magnetic field gradient and intensity can be used as the controlling parameters in the process. In the case of gravity settling, the sedimentation velocity was proportional to the particle cross section. In both situations clear separation occurred between smaller and larger particles. The modeling could be further improved to take into account different particle geometries. To this purpose we utilize realistic shapes as determined by SEM characterization of microparticles we produced utilizing milling.

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REFERENCES

[1] D. Brown, B.-M. Ma, Z. Chen, "Developments in the processing and properties of NdFeb-type permanent magnets," *Journal of Magnetism and Magnetic Materials*, vol. 248, Iss. 3, pp. 432-440, 2002.

[2] M. Sagawa, S. Fujimura, N. Togawa, H. Yamamoto, Y. Matsuura, "New material for permanent magnets on a base of Nd and Fe," *Journal of Applied Physics*, vol. 55, Iss. 6 pt 2A, pp. 2083-2087, 1983.

[3] Y. Kaneko, "Rare-Earth Magnets with high energy products," *Proceedings of the 16th International Workshop on Rare-Earth Magnets and Their Applications*, Sendai, Japan, pp. 83, 2000.

[4] A.-H.Lu, E.L. Salabas, F. Schüth, "Magnetic nanoparticles: Synthesis, protection, functionalization, and application," *Angewandte Chemie Internat. Ed.*, vol. 46, Iss. 8, pp. 1222-1244, 2007.

[5] K. Simeonidis, C. Sarafidis, E. Papastergiadis, M. Angelakeris, I. Tsiaoussis, O. Kalogirou, Evolution of $Nd_2Fe_{14}B$ nanoparticles magnetism during surfactant-assisted ball milling, *Intermetallics*, vol. 19, pp. 589-595, 2011.

[6] X. Xuan, J. Zhu, C. Church, *Microfluidics and nanofluidics*, Springer Science+Business Media, New York, NY, 2010.

[7] J.-F. Berret, O.Sandre, A. Mauger, "Size distribution of superparamagnetic particles determined by magnetic sedimentation," *Langmuir*, vol. 23, Iss. 6, pp. 2993-2999, 2007.

[8] S. Ganguly, S. Chakraborty, "Sedimentation of nanoparticles in nanoscale colloidal suspensions," *Physics Letters A*, vol. 375, Iss. 24, pp. 2394–2399, 2011.

[9] V. Prigiobbe, S. Ko, C. Huh, S.L. Bryant, "Measuring and modeling the magnetic settling of superparamagnetic nanoparticle dispersions," *Journal of Colloid and Interface Science*, vol. 447, pp. 58-67, 2015.

[10] N. Pamme, A. Manz, "On-Chip Free-Flow Magnetophoresis: Continuous Flow Separation of Magnetic Particles and Agglomerates," Anal. Chem., vol. 76, pp. 7250-7256, 2004.

[11] Hui Yan, Hongkai Wu, "Magnetophoresis," in *Encyclopedia of Microfluidics and Nanofluidics*, Ed: Dongqing Li, pp.1-8, Springer Science+Business Media, New York, NY, 2008.

[12] S.-E. K. Fateen, M. Magdy, "Simulation of a magnetophoretic Device for the Separation of Colloidal Particles in Magnetic Fluids," Proc. COMSOL Conference Boston, 2010.