

Fabrication of Microsized Magnetic Materials by Ink-Jet Printing

L. V. Cuong¹, N. K. Thuan² and P. D. Thang^{1,2,*}

¹Key Laboratory for Micro and Nanotechnology, VNU University of Engineering and Technology,
144 Xuan Thuy, Cau Giay, Hanoi 100000, Vietnam

²Faculty of Engineering Physics and Nanotechnology, VNU University of Engineering and Technology, Hanoi 100000, Vietnam

Micro-magnetic structures including Nd-Fe-B microsized particles were produced by the ink-jet printing technique. The Nd-Fe-B particles were commercial MQP-B particles with a Nd₂Fe₁₄B phase and an average diameter of 6 μm. The particles were milled to a mean particle size of about 300 nm before being added to the standard ink, MFL-003. The resultant magnetic suspension contained nanosized Nd-Fe-B particles with a suitable weight percentage (43%) and displayed a negligible difference in parameters, such as viscosity, pH, etc., compared to those of the standard ink. The produced magnetic structure, that can be used to trap magnetic particles and can be developed into a microsized magnetic source, consists of squares with thickness of 40 μm and surface area of 500 × 500 μm². Also, the properties of the magnetic structure were discussed. The obtained results show that the ink-jet printing technique is a simple and fast method for fabricating microsized magnetic structures. [doi:10.2320/matertrans.MD201702]

(Received June 19, 2017; Accepted November 8, 2017; Published June 25, 2018)

Keywords: microsized magnetic materials, magnetic properties, ink-jet printing

1. Introduction

Microsized permanent magnets are required mostly in micro systems and micro-electro-mechanical systems¹⁻⁹. Such magnets are usually based on rare-earth compounds with large remanent induction (B_R) and energy product ($(BH)_{max}$). These magnets are able to provide magnetic forces large enough to serve as actuators or force sources in microsystems. For further enhancement of magnetic forces, magnets have usually been fabricated into special shapes and arranged in an optimal formation to create a space with large magnetic gradients. Although the microsized permanent magnets are very interesting and important subjects, the conventional construction technique has limited the advance in fabricating process. Hence, there has not been much recent discussion on this topic. Conventional techniques, like electro-deposition and sputtering, have been successfully employed for the preparation of magnetic films with thicknesses in the order of 1 to 20 microns and for the fabrication of different magnetic film structures. However, these techniques are rather complex, time-consuming and costly^{2,9}.

Currently, the ink-jet (or jet) printing technique has become a rapid, versatile, and cost-effective production technology. Several groups have shown that this technology is a great way for designing and producing magnetic materials and structures¹⁰⁻¹⁶. Magnetic ink, which is a suspension composed of magnetic nanoparticles, can be ejected by an ink-jet printer onto many kinds of substrate.

Indeed, it is claimed that jet printers and magnetic inks, with a content of Nd-Fe-B powder up to 90% in weight, could be used to fabricate single hard magnets, that are either of the bulk type or film type, with large surface area dimensions in the order of mm²^{13,15,16}. These magnets have a good, hard, magnetic property with a coercive force (H_C) of about 750 kA/m and remanent magnetization (M_R) of about 35 mT. So far, there has been no work done on arrays of Nd-Fe-B micromagnets, with good, hard, magnetic properties,

produced using jet printers.

In this work, we used the ink-jet printing technique for rapidly fabricating films and micro structures comprising of magnetic Nd-Fe-B particles. The films and structures displayed a hard magnetic property with an order of magnetization (M_S) of 0.16 emu/g ~ 1 mT, M_R of 0.076 emu/g ~ 0.5 mT and H_C of 900 G ~ 71.6 kA/m, can be used to trap magnetic particles. The surface topography and magnetic properties of the films and the structures have been studied.

2. Experiment

Commercial magnetic Nd-Fe-B powder was used as an initial material. The powder was milled manually for 4 hours in porcelain and an environment of inert gas to obtain smaller particles. For preparation of the magnetic ink, the milled Nd-Fe-B powder was dispersed in a standard ink, called MFL-003, using an ultrasonic vibrator. This ink was selected based on its parameters that satisfy the requirements for use in of the Dimatix DMP-2831 (Fujifilm) printer. An aqueous suspension of Nd-Fe-B nanoparticles was obtained and then ejected onto commercial inkjet paper using the above mentioned printer. To avoid clogging the inkjet print-heads by agglomeration, the cartridge was vibrated for 10 minutes by an ultrasonic vibration machine before starting the printing process. The distance between the jet nozzle and the substrate, the substrate temperature, the ink temperature and the ink spouting velocity were set at 0.25 mm, 40°C, 40°C and 5×10^3 m/s, respectively. After that, a magnetic film with an area of 5×5 mm² and a magnetic structure of 10×10 squares (surface area of each square being 500×500 μm²) were printed. Both of the film and the microsized squares have thickness of 40 μm.

The particle size distribution of the Nd-Fe-B particles was determined by a Horiba particle analyzer LB-550 system. The surface morphology and the thickness of the film, as well as the microsized squares, were surveyed by NT-MDT atomic-force microscopy and Carl-Zeiss optical microscopy. The magnetic hysteresis curves of the Nd-Fe-B powder,

*Corresponding author, E-mail: thangducpham@yahoo.com

magnetic ink and printed samples were measured by a Lakeshore 7400 vibrating sample magnetometer. All measurements were carried out at room temperature.

3. Results and Discussion

Figure 1(a) shows the initial Nd-Fe-B particles with an average diameter of 6 μm , an unspecific shape, a good hard magnetic property with a coercive force of 1920 G and remanent magnetization of 45 emu/g. The mean particle size after 4 hours of milling was 300 nm. In fact, we milled the initial powder at different time frames, and found that the powder milled for 4 hours had the most uniform and smallest size. Besides that, the magnetic property and crystallographic structure of the particles remained almost unchanged after the milling (not shown here).

Before mixing with Nd-Fe-B powder, the MFL-003 ink had a particle size distribution between 2 nm to 10 nm, a density of 1.4 g/ml, viscosity of 25.6 mPa.s, pH of 9.2 and diamagnetic property with a magnetic susceptibility of -24.9×10^{-6} . After mixing the Nd-Fe-B powder into this ink using a Nd-Fe-B to MFL-003 mass ratio of 3:4, we obtained the printing ink (magnetic ink) with a viscosity of 26.6 mPa.s, pH of 9.8, particle size distribution between

2 nm to 1000 nm and a soft ferromagnetic property (Fig. 1(b)). The content of NdFeB powder in the ink is about 43% by weight (12% by volume).

Shown in Figs. 2 and 3, respectively, are the surface images, the cross section images, the magnetic hysteresis curves and the surface roughness of printed samples. Both film and microsized squares show that the shape and geometry in good agreement with the design specification. The printed samples have a homogeneous thickness of 40 μm (insets of Figs. 2(a) and 3(a)), the surface roughness of about 500 nm (Figs. 2(b) and 3(b)), and an almost isotropic magnetic response. Magnetization of the samples was measured with a field applied parallel (hereafter referred as “ip”) and perpendicular (hereafter referred as “oop”) to the film plane. The field applied perpendicular to the plane was corrected by a demagnetization factor. The hysteresis curves for the ip and oop directions almost overlap with each other. The fat hysteresis loops show that the samples possess the hard magnetic property with a high coercive force, H_C of 895 G and 820 G for the film and the squares, respectively.

The remanent magnetization of the squares, $M_R = 7.6 \times 10^{-2}$ emu/g, is about half of the saturation magnetization M_S in both of directions (Fig. 3(a)). Meanwhile for the film, an $M_R = 4.0 \times 10^{-2}$ emu/g, is only about one-third of its M_S

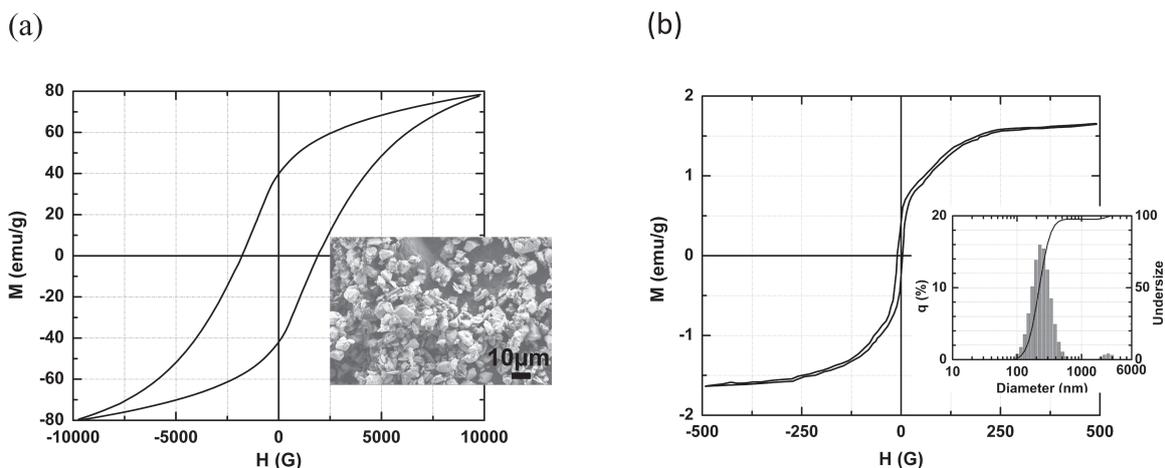


Fig. 1 (a) Magnetic hysteresis curve of Nd-Fe-B initial particles (inset: SEM image of initial particles). (b) Magnetic hysteresis curve of magnetic ink (inset: the particles size distribution of magnetic ink).

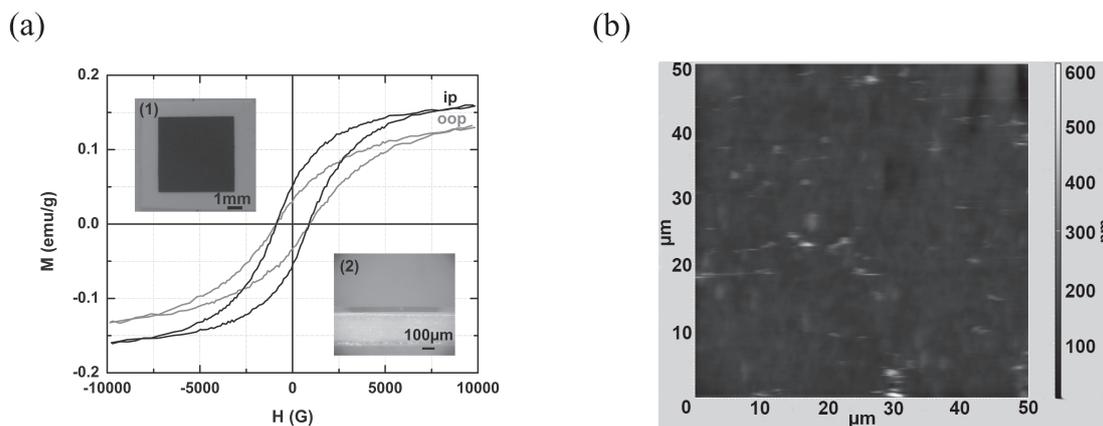


Fig. 2 (a) Magnetic hysteresis curves of $5 \times 5 \text{ mm}^2$ film. Inset: (1) surface and (2) cross-section image of film. (b) Surface AFM image of film.

(Fig. 2(a)). The hysteresis curves in the squares are of a squarer shape and enclose a larger area compared to the curves of the film, implying lower hysteresis losses. The area enclosed by the hysteresis curves of the film is 37% less than that enclosed by the hysteresis curves of the squares. This can be explained as the demagnetizing field in the squares being larger than that in the film. Indeed, the higher the concentration of Nd-Fe-B powder in the magnetic ink is, the harder the magnetic properties of the film, and the squares have been also enhanced. However, a magnetic ink with too high a concentration of Nd-Fe-B powder (more 43%) usually obstructs and damages printer nozzles. So far, in this work, we only focused on a concentration of Nd-Fe-B powder in the magnetic ink that was most suitable for the printer.

In order to investigate the magnetic strength at surface of the squares as well as the potential applications, we have simulated the z axis component of magnetic induction (B_z) at the surface of the squares and observed movement of magnetic particles on the surface of the square (Fig. 4). The simulation has shown that the squares create large magnetic induction near their surface and thus they are able to trap magnetic particles. Figure 4(a) shows the magnetic induc-

tion B_z of the squares at some heights d from their surface, along a line centered on the squares and parallel to edge of the squares (the x axis in inset (1) of Fig. 3(a)). The maximum value of 0.45 mT was obtained for B_z . Generally, at different values of d , B_z was almost unchanged and thus its gradient along the z axis is insignificant, just about 18 T/m. Obtained results show that the squares can create magnetic forces to trap nano magnetic particles at different heights.

Figure 4(b) shows a picture of Fe_3O_4 nanoparticles in a ferrofluidic solution attracted to locations of squares at height of $10\ \mu m$ (the structure was put under a Si sheet that has a thickness of $10\ \mu m$ to protect the surface of the structure and create a flat surface). Distribution of Fe_3O_4 particles followed the arrangement of squares. It can be seen that the shape of Fe_3O_4 particle clusters are not homogeneous and are not as square as the shape of Nd-Fe-B squares. At each square, Fe_3O_4 particle clusters are discontinuous. These are due to the uneven attraction force of Nd-Fe-B squares and the scattered distribution of Nd-Fe-B particles in each squares.

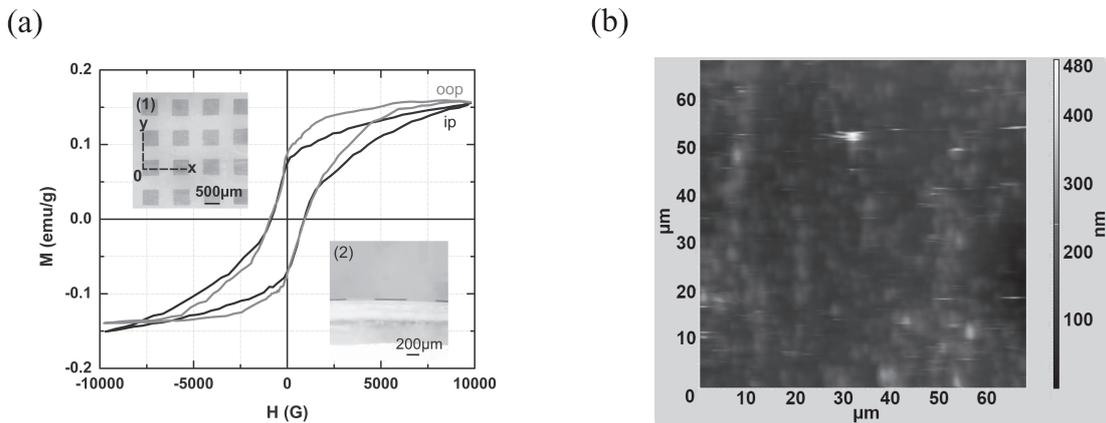


Fig. 3 (a) Magnetic hysteresis curves of squares. Inset: (1) surface and (2) cross-section image of squares. (b) Surface AFM image of one of squares.

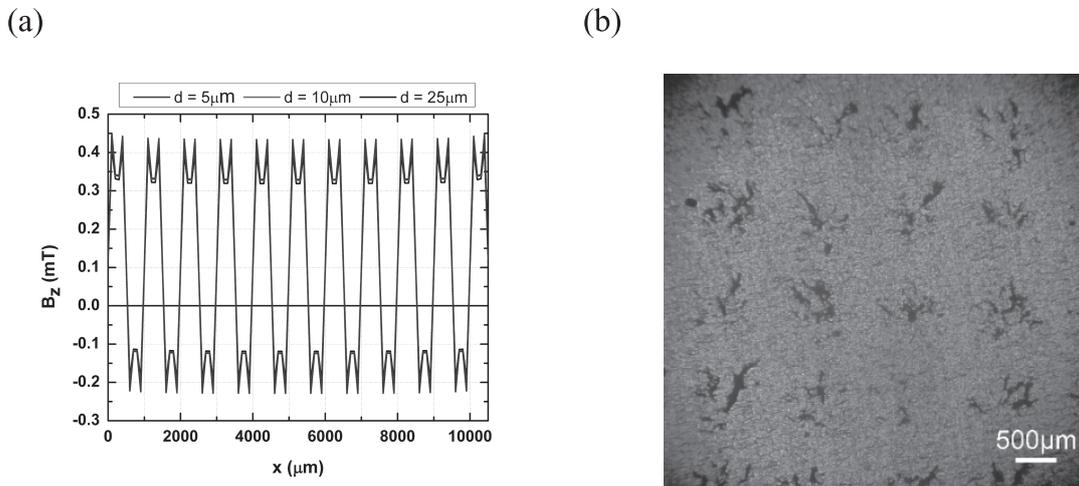


Fig. 4 (a) Magnetic induction (B_z) at some heights (d) on surface of squares along the x axis in inset (1) of Fig. 3(a). (b) Fe_3O_4 particles in ferrofluidic solution attached to locations of squares under a Si sheet.

4. Conclusions

In this work, a simple and low-cost inkjet printing technique has been developed for the fabrication of Nd-Fe-B films and microsized structures. Hard magnetic properties have been obtained using these printed objects. Preliminary results showed that the microsized structures are suitable for positioning magnetic particles. Further work on improvement of the magnetic properties of printed materials is underway.

Acknowledgments

This study was supported by Vietnam National Foundation for Science and Technology Development (NAFOSTED), grant number 103.02-2015.80.

REFERENCES

- 1) B. Azzerboni, G. Asti, L. Pareti, M. Ghidini: *Magnetic Nanostructures in Modern Technology*, (Springer, Berlin, 2008) pp. 105–125.
- 2) J.P. Liu, E. Fullerton, O. Gutfleisch, D.J. Sellmyer: *Nanoscale Magnetic Materials and Applications*, (Springer, Berlin, 2009) pp. 661–680.
- 3) D. Niarchos: *Sens. Actuators A Phys.* **106** (2003) 255–262.
- 4) V. Zablotskii, A. Dejneka, S. Kubinova, D. Le-Roy, F.D. Bouchiat, D. Givord, N.M. Dempsey and E. Sykova: *PLOS ONE* **8** (2013) e70416.
- 5) O. Osman, S. Toru, F.D. Bouchiat, N.M. Dempsey, N. Haddour, L.F. Zanini, F. Buret, G. Reyne and M.F. Robbin: *Biomicrofluidics* **7** (2013) 054115.
- 6) P. Wang, K. Tao, Z. Yang and G. Ding: *J. Zhejiang Univ-Sci C (Comput&Electron)* **14** (2013) 283–287.
- 7) J. Pivetal, D. Royet, G. Ciuta, M.F. Robin, N. Haddour, N.M. Dempsey, F.D. Bouchiat and P. Simonet: *J. Magn. Magn. Mater.* **380** (2015) 72–77.
- 8) D.L. Roy, G. Shaw, R. Haettel, K. Hasselbach, F.D. Bouchiat, D. Givord and N.M. Dempsey: *Materials Today Communications* **6** (2016) 50–55.
- 9) Z. Hu, H. Qu, D. Ma, C. Luo and H. Wang: *J. Rare Earths* **34** (2016) 689–694.
- 10) P. Tiberto, G. Barreca, F. Celegato, M. Coisson, A. Chiolerio, P. Martino, P. Pandolfi and P. Allia: *Eur. Phys. J. B* **86** (2013) 173–178.
- 11) S. Schwarzer, B. Pawlowski, A. Rahmig and J. Topfer: *Materials in electronics* **15** (2004) 165–168.
- 12) T. Speliotis, D. Niarchos, P. Falaras, D. Tsoukleris and J. Pepin: *IEEE Trans. Magn.* **41** (2005) 3901–3903.
- 13) B. Pawlowski and J. Topfer: *J. Mater. Sci.* **39** (2004) 1321–1324.
- 14) H. Song, J. Spencer, A. Jander, J. Nielsen, J. Stasiak, V. Kasperchik and P. Dhagat: *Journal of Applied Physics* **115** (2014) 17E308_1–17E308_3.
- 15) C. Huber, C. Abert, F. Bruckner, M. Groenefeld, O. Muthsam, S. Schuschnigg, K. Sirak, R. Thanhoffer, I. Teliban, C. Vogler, R. Windl and D. Suess: *Appl. Phys. Lett.* **109** (2016) 162401–162405.
- 16) M.P. Paranthaman, C.S. Shafer, A.M. Elliott, D.H. Siddel, M.A. Mcguire, R.M. Springfield, J. Martin, R. Fredette and J. Ormerod: *J. Magn.* **68** (2016) 1978–1982.