

Shape-Controlled Synthesis of Fe Nanoparticles with High- M_s Via Modest-Temperature Reduction Methods

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Recent improvements in electronic devices have led to demand for further miniaturization and higher frequency operation of magnetic devices. [1] To improve the ferromagnetic response in the high-frequency range, the ferromagnetic resonance frequency, f_r , of conventional ferromagnetic materials has to be increased by induced magnetic anisotropy, which results in a lack of isotropic magnetic properties in the high-frequency range. Superparamagnetic nanoparticles (NPs) with high saturation magnetization (M_s), such as Fe and Fe₇₅Co₂₅, can realize high susceptibility and permeability and can thus become good candidates for high-frequency magnetic materials, since superparamagnetic NPs have isotropic magnetic properties and no hysteresis loss. For the chemical synthesis of Fe NPs, the decomposition of iron carbonyl is frequently used, largely due to the ease of use and the lack of byproducts. However, a widely used synthesis procedure is the decomposition of Fe(CO)₅ at very high temperature such as 290°C for a long time (more than 1 h), in which large amounts of iron oxide easily form on the surface of Fe NPs. The magnetization of NPs with a size less than 16 nm are usually less than 130 emu/g. [2] In this paper, we report a synthesis method involving

modest-temperature reduction of an iron salt in an organic solvent by superhydride (LiBEt₃H). A combination of surfactants, alkylphosphine oxide and oily amine, were employed to control particle growth and to avoid the oxidation from surfactants. The sizes and shapes of Fe NPs were tuned by varying the reaction time and the molar ratio of the iron salt to the surfactants. The capping ligand plays a critical role in the shape-controlled process. Alkylphosphine oxide has a large steric hindrance effect and prefers to selectively absorb onto particular crystallographic facets. However, only spherical NPs can be observed using oleic acid due to its higher oxophilicity and stronger binding to NPs. Magnetic measurements reveal that the saturation magnetization of larger than 8.0 nm Fe NPs at 300 K was always over 180 emu/g, which proves that our method could open up a new way to prepare Fe NPs with high magnetization.

REFERENCES

- [1] A. Inoue *et al.*, *J. Appl. Phys.* 83, 6326 (1998)
- [2] K. S. Suslick, M. Fang, and T. Hyeon, *J. Am. Chem. Soc.* 118, 11960 (1996).

Effect of Magnetostatic Interaction on Susceptibility of Superparamagnetic Fe Nanoparticles in Nonmagnetic Matrix

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Nano-sized ferromagnetic nanoparticle (NP) shows a superparamagnetic behavior due to thermal fluctuation of its magnetic moment, and their hybrid structure with polymer can be expected as a new soft material with three dimensionally isotropic properties and no loss [1]. For the NPs/polymer hybrid structure, it should be important to clarify the correlation between the initial magnetic susceptibility, χ , and the magnetostatic interaction between NPs. In this study, we investigated the static magnetic properties of the NPs/polymer hybrid structure as a function of the size and volume fraction, v.f., of the chemically synthesized Fe NPs. The Fe nanoparticles were synthesized by a conventional thermal decomposition method or a reduction method through a superhydride technique developed by the present authors [2]. The Fe NPs covered with surfactant were dispersed into polyvinylbutyral (PVB) matrix with v.f. from 0.1 to 1.5 vol% using an appropriate solvent, and the Fe NPs/PVB hybrid structure film was packed into an epoxy resin to prevent it from oxidation. The magnetization curve and temperature dependence χ were measured by a superconducting quantum interference device (SQUID) magnetometer.

The average size from 3.2 to 9.3 nm and the saturation magnetization of 140 ~ 150 emu/g can be obtained by precisely controlling the reaction kinetics. The χ of the Fe NPs with v.f. of 0.1 vol% monotonously increases with increasing the size, which agrees well with the zero field limit calculation of the Langevin function. This suggests that the magnetostatic interaction between the Fe NPs is negligibly small for the v.f. of 0.1 vol%. The blocking temperature, T_b , and χ of the Fe NPs with 9.3 nm increase with increasing the v.f. in Fig. 1. Moreover, temperature dependent χ shows a broader peak in the higher v.f. range. These indicate that the nano-scaled locally strong magnetostatic interaction forms magnetic clusters and their size distribution is wide, in which the magnetic moments of each Fe NP are ferromagnetically aligned, resulting in increase in the effective volume of the Fe NPs with increasing the v.f.

REFERENCES

- [1] D. Hasegawa, T. Ogawa, M. Yamaguchi, and M. Takahashi, *J. Magn. Soc. Jpn.*, 30, 528 (2006).
- [2] H. Yang, F. Ito, D. Hasegawa, T. Ogawa, and M. Takahashi, *J. Appl. Phys.* (in press).

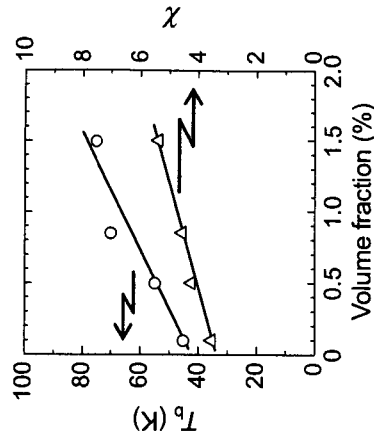


Fig. 1. Volume fraction dependence of T_b and χ of Fe nanoparticles with $D_{\text{ave}} = 9.3$ nm. Circle and triangle symbols denote T_b and χ , respectively.