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Hard magnetic properties of FePd nanoparticles*

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Abstract. Magnetic nanoparticles Fe_xPd_{100-x} (x = 42, 50, 55, 60, 63) with small size of around 5–10 nm were prepared by sonochemistry from palladium acetate and iron acetate. The compositions x can be controlled by changing the ratio of the above precursor chemicals. Under the effect of annealing at various temperatures from 450 °C to 650 °C, structure change was observed and samples show hard magnetic properties with high coercivity up to 2.1 kOe. Magnetic properties of samples were then systematically discussed in dependence of x and annealing temperatures.

1 Introduction

Alloy nanoparticles with the structure type $L1_0$ are one of the candidate materials suitable for the ultra-high density magnetic storage applications due to their large uniaxial magnetocrystalline anisotropy and good chemical anisotropy [1]. Among them, FePt and FePd with large uniaxial magnetocrystalline anisotropy $K_u \sim 7 \times 10^7$ erg cm^{-3} and $K_u \sim 1.8 \times 10^7$ erg cm^{-3}, respectively, have been paid much attention [2–14]. However, only several approaches to preparation of FePd nanoparticles have been reported including epitaxial growth by electron beam deposition [8–10], chemical synthesis modified from FePt synthesis process [11,12,15], modified polyol process [13], microwave irradiation [3]... They do not show exclusively the ordered $L1_0$ phase transition similarly as in FePt preparations. Especially, FePd prepared by Chen and Nikles [15] did not transform to $L1_0$ phase after annealing at sufficient high temperature of 700 °C. Furthermore, their magnetic properties were not investigated systematically. Recently, magnetic properties of FePt nanoparticles prepared by sonoelectrodeposition have been reported by Nam et al. [16]. In this study, we report the hard magnetic properties of FePd nanoparticles synthesized by sonochemistry, which was developed to make nanoparticles [17]. Their magnetic properties were investigated in dependence of chemical compositions and annealing temperatures.

2 Experimental

The synthesis of Fe_xPd_{100-x} nanoparticles was conducted by sonochemical reaction. The mixture of palladium(II) acetate [$Pd(C_2H_3O_2)_2$] and iron(II) acetate [$Fe(C_2H_3O_2)_2$] with distilled water were prepared in a 150 mL flask. The solution in flask was ultrasonicated with power of 375 W, frequency of 20 kHz emitted by a Sonic VCX 750 ultrasound emitter within 90 min. The FePd nanoparticles were washed and separated from the solution by using a centrifuge with alcohol at 9000 rpm for 30 min and then dried at 70 °C–75 °C. As-prepared samples then were annealed at various temperatures from 450 °C to 650 °C under continuous flow of (N_2 + Ar) gas at heating rate of 5 °C/min for 1 h.

The structure of the as-prepared and the annealed FePd samples at various temperatures were studied by X-ray diffractormeter (Bruker D5005). The energy dispersive spectroscopy (EDS) measurements were carried out in order to study the chemical composition of Fe:Pd. The composition ratio factor x is defined as the number calculated from amounts of the precursor chemicals. The composition ratio estimated from EDS measurements is close to the nominal composition x. Therefore, we use calculated x as the ratio factor from now on in the chemical composition of Fe_xPd_{100-x}. The morphology and size of particle were investigated by using transmission electron microscope (TEM) JEM1010, JEOL. Magnetic properties of samples were studied by using a vibrating sample magnetometer (VSM) DMS 880 and a physical property measurement system (PPMS, Quantum design) Evercool II.

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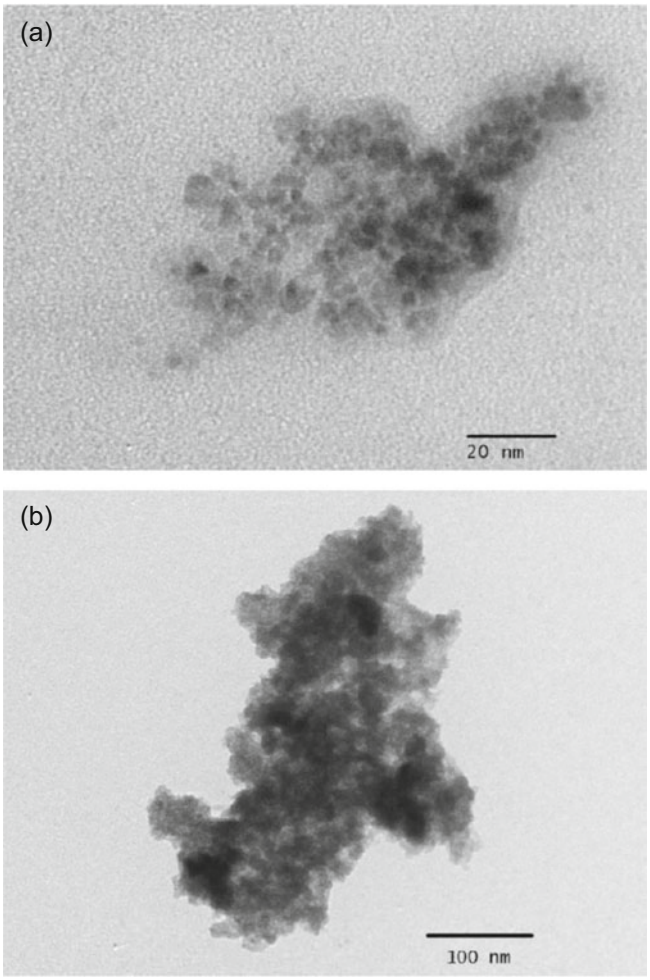

Fig. 1. TEM images of as-prepared (a) and annealed (b) $Fe_{60}Pd_{40}$ nanoparticles.

3 Results and discussion

Figure 1 exhibits the TEM images of as-prepared and annealed sample at 550 °C with $x = 60$. The as-prepared sample contains well-dispersed nanoparticles with size of around 5–10 nm. The annealed sample contains colloid with size of around 30–40 nm due to the annealing effect. The particle size of the annealed sample is larger than that of as-prepared sample due to the aggregation and particles growth.

Figure 2 shows the X-ray patterns of as-prepared and annealed samples at various temperatures with $x = 60$. The broad peaks below 30° are due to the amorphous nature scattered from the glass plate that was used as the sample holder in the experiments. The X-ray pattern of the as-prepared sample shows only the Pd diffraction peaks at 40°, 46.5° and 68° (PDF 05-0681). After annealing, samples show the tetragonal order phase of FePd alloy (PDF 02-1440) with diffraction peaks at 41°, 47°, 49°, 53°, 61° and 69°. These peaks are shifted to higher position with increasing annealing temperature. They are ascribed as (1 1 1), (2 0 0), (2 0 1) and (0 0 2) fundamental and superlattice reflections of the $L1_0$ ordered phase of

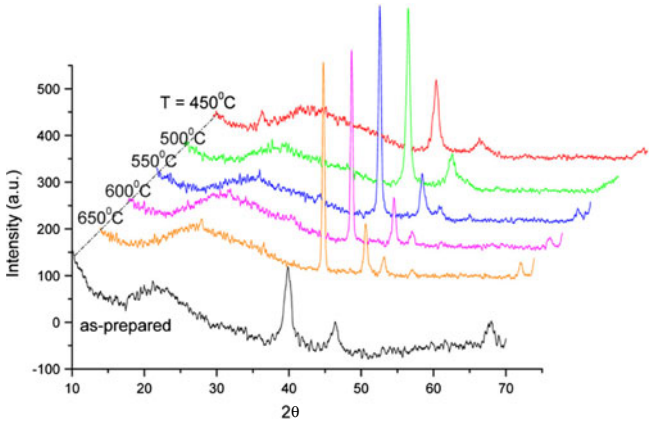

Fig. 2. X-ray patterns of as-prepared and annealed $Fe_{60}Pd_{40}$ nanoparticles.

FePd. It has a tetragonal superlattice structure where Fe atoms can substitute Pd atoms if they have larger amount than Pd. In X-ray pattern of as-prepared sample, the reflections of Fe may very weak and can not be seen due to the fact that Fe atomic radius is much less than that of Pd similar to the formation of FePt prepared by sonochemistry [16]. The as-prepared particles were not disordered FePd but they may be formed by small domains of Fe and Pd. The broad peaks show small particles size as around 2 nm, which smaller than that observed by TEM, is also an indication of the co-existence of Fe and Pd domains in particles. The $L1_0$ ordered phase of FePd then appeared after annealing due to the diffusion process between Fe and Pd domains. The particle size is estimated to be 48 ± 5 nm, which is larger than that observed by TEM. The lattice parameters of the ordered phase is calculated as $a = 3.868 \pm 0.002$ Å and $c = 3.690 \pm 0.003$ Å for the sample annealed at 550 °C. From these values, the average ratio of Fe:Pd can be estimated as 1.47:1, which is similar to the nominal composition. The degree of the order S can be estimated as the area ratio of the peaks (2 0 0) and (0 0 2) [18]. It increases when x increases and reach maximum of 0.65 when $x = 60$, then decreases. The annealing temperature also gives an affect to the degree of the order S. With the sample $x = 60$, S increases with increasing annealing temperature and reach maximum at 550 °C then decrease when annealing temperature increases to 600 and 650 °C. The low value of maximum S indicates the chemical composition as well as the degree of the order may change from grain to grain. Figure 3 shows the EDS result of sample with $x = 60$. From EDS measurements (all data not shown), the atomic ratio of Fe:Pd was estimated to be 44:56, 49:51, 57:42, 61:39, 66:34 for sample with $x = 42$, 50, 55, 60, 63, respectively. The deviation of composition factor from EDS results is less than 5% for all samples.

Magnetic measurements of as-prepared samples (data not shown) exhibit low saturation magnetization M_S of about few emu/g and low coercivity H_C of about 10–30 Oe. The low M_S of the as-prepared sample can be assigned to the weak magnetic iron oxides and iron hydroxides. They may be formed due to the oxidation or

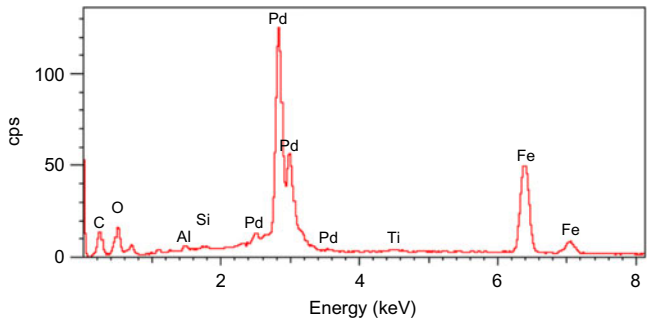

Fig. 3. EDS result of Fe_xPd_{100-x} nanoparticles with $x = 60$.

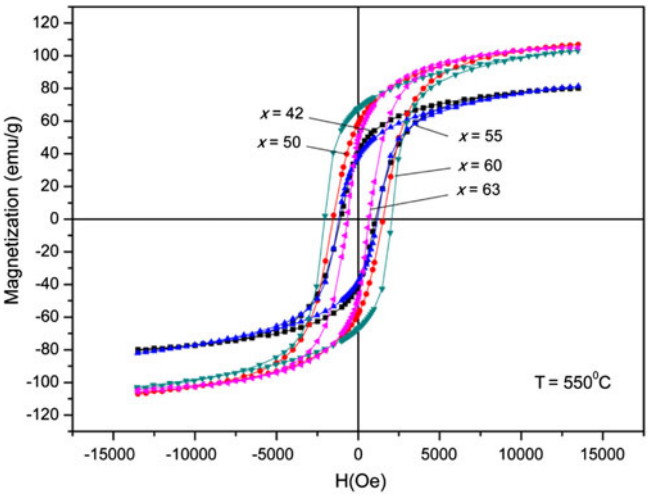

Fig. 4. Room temperature magnetization curves of Fe_xPd_{100-x} nanoparticles annealed at 550 °C with various x.

hydroxidation of Fe atoms with the suggestion of the coexistence Fe and Pd domains in as-prepared nanoparticles. Upon to the annealing at high temperature under (Ar + N_2) atmosphere, the hard magnetic FePd was formed. In order to investigate the hard magnetic properties, the as-prepared samples with $x = 42$, 50, 55, 60, 63 were annealed at temperature of 450, 500, 550, 600 and 650 °C. Figure 4 shows the room-temperature hysteresis curves of the samples with various x annealed at 550 °C. All the samples show hard magnetic properties with high coercivity H_C. When x increases from 42, H_C increases from 1 kOe and reach maximum of about 2.1 kOe at $x = 60$, then dropped to 0.7 kOe when x increases to 63. The saturation magnetization also improved significantly compared to that of the as-prepared samples. However, it does not show composition dependence similarly to that of the coercivity. The saturation magnetization is high at $x = 50$, 60 and 63, but has lower value at $x = 42$ and 55. Figure 5 shows the room temperature hysteresis curves of the samples with $x = 60$ annealed at various temperatures. Sample annealed at 450 °C shows hard magnetic properties with coercivity of 0.6 kOe. The coercivity H_C then increases with increasing annealing temperature and reach the maximum of 2.1 kOe at annealing temperature of 550 °C and has low value of 0.15 kOe at annealing temperature of 650 °C. These are

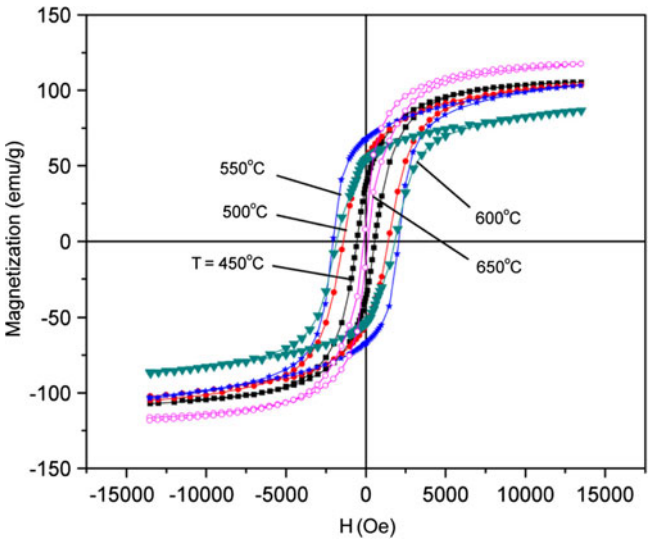

Fig. 5. Room temperature magnetization curves of $Fe_{60}Pd_{40}$ nanoparticles annealed at various temperatures.

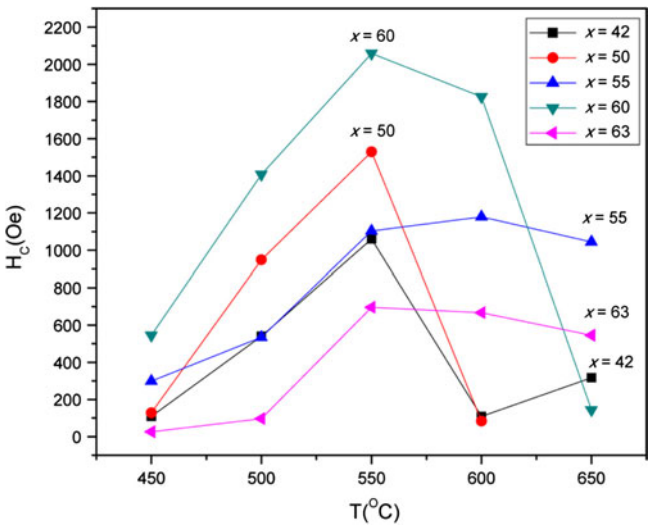

Fig. 6. The annealing-temperature dependence of coercivity of the Fe_xPd_{100-x} nanoparticles.

in agreement with the change of the degree of the order S due to the diffusion of Fe atoms upon annealing. Figure 6 shows the overview of all x and annealing temperatures dependences of the coercivity H_C. Samples with $x = 42$, 50, 60 and 63 show similar annealing temperature dependence of the coercivity H_C, which increases with increasing annealing temperature and has maximum value at annealing temperature of 550 °C then decreases. Sample with $x = 55$, however, shows highest coercivity H_C at annealing temperature of around 600 °C. This sample also has low value of saturation magnetization as shown in Figure 4. These can be understood that the diffusion processes between Fe and Pd domains are varied from sample to sample with different x. Over all, the sample with $x = 60$ shows highest H_C at almost all annealing temperatures. The degree of the order of this sample also has highest value, indicating that the hard magnetic properties strongly depend on the order phase of the $L1_0$ of FePd nanoparticles.

4 Conclusions

Hard magnetic properties of FePd nanoparticles prepared by sonochemistry were systematically studied and show strong dependence on chemical composition factor x from 42 to 63 and annealing temperatures from 450 °C to 650 °C. In general, the coercivity H_C shows high value at around annealing temperature of 550–600 °C for all x and the highest one up to 2.1 kOe of sample with $x = 60$ annealed at 550 °C. The chemical order degree shows similar tendency, indicating the hard magnetic properties strongly depend on the order of the $L1_0$ of FePd nanoparticles. The forming of this order phase depends on the chemical composition of Fe:Pd and the annealing temperature.

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