Research Article

Sintering of Soft Magnetic Material under Microwave Magnetic Field

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We have developed a simple process for sintering of soft magnetization materials using microwave sintering. The saturated magnetization (M_s) of sintered magnetite was 85.6 emu/g, which was as high as 95% of magnetite before heating (90.4 emu/g). On the other hand, the averaged remanence (M_r) and coercivity (H_c) of the magnetite after heating were 0.17 emu/g and 1.12 Oe under measuring limit of SQUID, respectively. For the sintering process of soft magnetic materials, magnetic fields of microwave have been performed in nitrogen atmosphere. Therefore, a microwave single-mode system operating at a frequency of 2.45 GHz and with a maximum power level of 1.5 kW was used. We can sinter the good soft magnetic material in microwave magnetic field. The sample shrank to 82% theoretical density (TD) from 45%TD of green body. The sintered sample was observed the microstructure by TEM and the crystal size was estimated the approximate average size is 10 nm.

1. Introduction

Sintering of magnetite (Fe_3O_4) with nanocrystals has long been of great interest because of their immense technological applications especially in the magnet, motor, and electric parts. Nanoparticles with superparamagnetic properties have great potential to achieve such desirable properties.

Recently, various methods have been developed to synthesize Fe_3O_4 particles in nanometer size range. These methods were the electrochemical synthesis [1], the reactive magnetron sputtering [2], chemical reaction [3], and so on. However, the magnetic properties of magnetite-based nanoparticles or films highly depend upon the synthesis procedure.

Microwave irradiation to materials is a new comer for our civilization with a history of only half century. The temperatures of the surroundings are colder than that of targets, that can easily be imagined by a home microwave oven. It clearly suggests that the energy transfer mechanism

in microwave heating is quite different from the traditional heating process. Roy et al. reported sintering of metal powders by microwave in 1999 [4] and decrystallization of ferrite magnetic materials by microwave magnetic field heating in 2002 [5]. Therefore, we have studied the sintering of soft magnetization under microwave heating.

2. Experimental Setup

The samples had been heated by magnetic field of microwave. Figure 1 shows a schematic drawing of experimental setup of magnetic field heating of 2.45 GHz microwave. The magnetic or electric fields of microwave can be separated on positions in the single-mode cavity. High-frequency alternated magnetic field was applied to a sample placed on the magnetic field node in the TE₁₀₃ single-mode cavity with the cross-section of 27.2 mm × 85 mm. The generator, PRJ-1000L, Ewig Co., Ltd., supplied microwave to the cavity at the frequency 2.45 GHz. The microwave power

FIGURE 1: The schematic drawing of experimental setup of microwave heating.

varied from 50 to 1500 watts controlling by the DC power supply which consisted of AC-DC inverter. The infrared pyrometer, IGAR12-LO, IMPAC Infrared Co., Ltd., measured the temperature of the sample through the 6 mm hole drilled through the end plunger of cavity. The waveguide was evacuated to 10^{-4} Pa by turbo molecular pump with 100 L/s pumping speed and changed to nitrogen gas in the cavity. During the heating, nitrogen gas was flowing in cavity.

The samples were prepared by uniaxial pressing of magnetite powder (FEO07PB) with the purity of 99 weight% from Kojundo Chemical Laboratory Co., Ltd., Japan. The sample density was up to 45% of the theoretical density (TD) by geometrical method. The grain size of the magnetite was less than 1 micrometer. The size of the sample was 8 mm diameter and 4 mm thick pellet of 0.5 (g) that was small enough not to disturb the criterion for fundamental resonance in the cavity. The four samples were put in the magnetic field maxima in the cavity supported by thermal insulator made of a lightweight alumina silica fiber board.

3. Experimental

The graphs in Figure 2 show a typical progress of process temperatures measured during microwave heating of magnetite samples. The microwave power was controlled manually. The microwave power shows remainder of input power and reflex power. For sintering experiments, the magnetite powder compacts were heated to the sintering temperature of 1000°C for 10 min followed by 70 min soak time. In initial step heating during the first 30 min, the sample temperature was dramatically changing. Thereafter, the sample temperature was settling and microwave power was lower as 270 watt.

Figure 3 shows a magnetite sample sintered by using H-filed of microwaves at 1000°C . The sample shrank to 6.2 mm diameter from 8 mm diameter for sintering. The sintered sample was found to be oxidized on the only surface. The linear shrinkage of the sample was 17% and no remarkable change in the shape was observed. The sample shrank to 82% theoretical density (TD) from 45%TD of green body.

Figure 4 shows the X-ray diffraction profile of the magnetite after heating at 1000°C . It shows Bragg reflections of

FIGURE 2: The typical progress of process temperature measured during microwave heating.

both magnetite and hematite. The surface of sample was oxidized because the nitrogen gas was flowing in cavity during heating.

The high-resolution transmission electron microscope (TEM; a JEOL JEM-3200) observed the images of the original magnetite powder before heating and those excited in the selected magnetic field. The original crystal can see the well-ordered lattice patterns over the whole crystal; therefore the original crystal has flat and homogeneous surfaces. Figure 5 shows TEM image and selected area electron diffraction pattern. The sample heated in the magnetic field exhibits the presence of randomly oriented nano-crystal. The crystal sizes of this sample are of approximate average size 10 nm. The randomness of the lattice orientation indicates that the particle-particle magnetic interaction is negligible in

FIGURE 3: The picture of magnetite sintered by using magnetic field of microwave at 1000°C.

FIGURE 4: X-ray diffraction of samples heated by magnetic field at 1000°C.

the heating cooling process. If each magnetic domain should be highly excited by application of the oscillating magnetic field, cohesive rotation of the domains can be induced synchronously with the oscillating field. It is very interesting that the temperature increase in the sample is accelerated at a temperature close to the Curie point and never rises above 1000°C but it is well below the melting point of 1535°C.

The macroscopic magnetizations of samples were measured by SQUID without zero correction (Quantum Design MPMS-XL7). Figure 6 shows the hysteresis loop of magnetite at 300 K. As can be seen in Figure 6, the saturated magnetization (M_s) of sintered magnetite was 85.6 emu/g, which was as high as 95% of magnetite before heating (90.4 emu/g).

(a)

(b)

FIGURE 5: TEM image of magnetite sintered by using magnetic field of microwave at 1000°C.

The averaged remanence (M_r) and coercivity (H_c) of the magnetite before heating were 11.74 emu/g and 108.9 Oe, respectively. On the other hand, the averaged remanence (M_r) and coercivity (H_c) of the magnetite after heating were 0.17 emu/g and 1.12 Oe under measuring limit of SQUID, respectively. Their M_r value and H_c value of sintered sample became two digits smaller than raw sample for heating by magnetic field of microwave.

4. Conclusion

We have developed a simple process for sintering of soft magnetization materials. For the sintering process of soft magnetic materials, magnetic fields of microwave have been performed in nitrogen atmosphere. Therefore, a microwave

FIGURE 6: (a) The hysteresis loop of the magnetite sintered by using magnetic field of microwave at 1000°C. (b) The M_r and H_c of the magnetite sintered by using magnetic field of microwave at 1000°C.

single-mode system operating at a frequency of 2.45 GHz and with a maximum power level of 1.5 kW was used. We can sinter the good soft magnetic material in microwave magnetic field. The sample shrank to 82% theoretical density (TD) from 45%TD of green body. The sintered sample was observed the microstructure by TEM and the crystal size was estimated the approximate average size is 10 nm.

Using microwave sintering, the M_s of sintered magnetite was 85.6 emu/g, which was as high as 95% of raw magnetite with 90.4 emu/g. On the other hand, the M_r and H_c of the magnetite after heating were 0.17 emu/g and 1.12 Oe under measuring limit of SQUID, respectively.

From TEM and SQUID data, it is suggested that the sintered magnetite under magnetic field of microwave has super-ferrimagnetism.

We expect that it is used in transformer or electromagnetic cores. The macroscopic magnetization of sintered body using microwave can easily reverse direction without dissipating much energy, that is, hysteresis losses. Because of their comparatively low losses, they are extensively used in the cores of transformers and electromagnetic cores in applications such as car industrials.

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