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Contribution of magnetization mechanisms in nickel–zinc ferrites with different grain sizes and its temperature relationship

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HIGHLIGHTS

- Nickel–zinc ferrites with varied grain sizes fabricated by the solid-state reaction.
- Complex permeability spectra were resolved into two magnetization mechanism parts.
- Domain wall movement shifts to be dominant with grain size increasing.
- High operating temperature is favorable for domain wall movement.

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ABSTRACT

By adjusting the duration time of sintering, nickel–zinc (NiZn) ferrites with gradually varied grain sizes have been fabricated by the solid-state reaction method and their magnetization mechanisms and temperature dependence were investigated. In order to manifest the variation of permeability, the dominant magnetization mechanism was investigated by resolving the complex permeability spectra into two components including domain wall movement and spin rotation magnetization. The results show that, with grain size increasing, the dominant contribution to magnetization mechanism changes from spin rotation magnetization to domain wall movement due to the increase of amount of domain wall, and the amount of domain wall is observed by Lorentz microscopy to measure the critical diameter of magnetic domain which is around 3 μm in NiZn ferrite. The temperature dependence of the magnetization mechanism has been discussed for the sake of thermal stability. As the operating temperature increases, the ratio between two magnetization mechanism components appears to be stable around 4 after a shift from 1.76, and the domain wall movement totally dominates the variation of permeability as the main magnetization mechanism.

1. Introduction

Nickel–zinc (NiZn) ferrites have been extensively applied in various electromagnetic devices such as inductors, transformers, filters and DC–DC convertors due to the frequency-dependent physical property like magnetic permeability. Since the properties of ferrite materials are highly sensitive to the preparation methods, extensive efforts are directed at optimizing the microstructure and magnetic property of NiZn ferrite by adjusting the main composition, additives and sintering process [1]. Grain growth and magnetic property of NiZn ferrites are strongly influenced by specific additives such as MoO_3 and Bi_2O_3 [2–5]. As a key process parameter, the effect of duration time of sintering on grain size, density, saturation magnetization and other relevant magnetic properties of NiZn ferrites has been widely investigated [6]. Variation of duration time of sintering for $Cu_{0.6}Zn_{0.4}Fe_2O_4$ ferrites causes appreciable changes in its electrical and magnetic properties, along with the temperature dependent drift mobility [7]. By varying the sintering

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temperature and duration time, Bera et al. [8] have studied that initial permeability increased linearly with the grain size up to about 5 μm and after that the rate of increase was reduced due to the formation of more closed porosity.

Generally, spectra of complex permeability are the superposition of two kinds of magnetization mechanisms: domain wall motion and spin rotation [9,10]. To investigate the respective contribution from each mechanism, many works have been done recently. Su et al. [11,12] recently reported that the NiCuZn ferrite with less Zn amount could obtain high Curie temperature and the larger grain size is favorable for domain wall motion. Tsutaoka [13] have studied that the domain wall contribution to permeability declined with the decrease of the amount of domain wall when NiZn particles were coated with polyphenylene sulfide. Hu et al. [14] have taken CuO and V_2O_5 additives as variables to reduce the sintering temperature of NiZn ferrites, and it was deduced from the fitting results that domain wall motion was predominant in the magnetizing process. Similar simulation procedures were also adopted by Han et al. [15] in NiZn ferrites doped with SiO_2, and the results indicated that the components of spin rotation make larger contributions to the permeability dispersion spectra because the SiO_2 aggregated at the grain boundaries of NiZn ferrites. In short, among those works focused on the magnetic property, composition, additives and sintering process as well as the spectra of complex permeability, few report concerns on the contribution of magnetization mechanisms in NiZn ferrites as a certain function of grain size and its temperature relationship. This work demonstrates the contribution of magnetization mechanisms and temperature characteristics of NiZn ferrite with different grain sizes, and the magnetocrystalline anisotropy and magnetic domain are taken into account accompanying with the fitting results of frequency dispersion of permeability.

2. Experimental procedures

NiZn ferrites with the chemical composition of $Ni_{0.266}Zn_{0.66}Cu_{0.09}Fe_{1.968}O_{4-\delta}$ were fabricated by the solid-state reaction method. The powders which consist of different raw materials were mixed in planetary mills for 1 h. The mixed powders were calcined at 930 °C for 2 h in air. The calcined powders were mixed with 0.125 wt% MoO_3, and then second-milled for 2 h. After drying at 80 °C, the dried powders were granulated with polyvinyl alcohol (PVA) and pressed into toroids at 60 MPa. Then the green flans were sintered at 1050 °C in air and left to cool to room temperature inside a furnace. The duration time of sintering was varied from 1 to 7 h in 2 h intervals to control the microstructure evolution (grain size), and corresponding samples under different duration times were labeled from 1 to 4 sequentially. Partial sintered toroids were crushed and shaped to small spheres with the diameter of 3 mm for the measurement of magnetization curve.

The crystallographic properties were characterized by Philips X-ray diffractometer (XRD, Cu target, K_α radiation, 40 kV, and 40 mA). The cross-section morphology of samples was observed by scanning electron microscope (SEM, JEOLJSM-6490L). The Lorentz microscopy domain wall observations of these polycrystalline ferrite samples are obtained by JEOL-2100F, and prepared samples are ion milled into the thickness of 10 nm for Lorentz transmission electron microscopy observation. From enlarged SEM micrographs of samples, average grain sizes (D) were estimated by intercept method. The complex permeability was acquired by IWATSU SY8232 B–H analyzer at 0.1–10 MHz. Magnetization curve and saturation magnetization (M_s) under various temperatures were carried out with applied static magnetic fields up to 5 kOe by a vibrating sample magnetometer (VSM, VSM-220). The density was measured by the Archimedean method.

3. Results and discussion

XRD patterns of the samples with different duration times of sintering are shown in Fig. 1. The XRD measurements show that all samples demonstrate typical spinel structure without impurity phases. The XRD densities (d_{XRD}) deduced from the XRD patterns of these samples are listed in Table 1. With the increase of duration time from 1 h to 7 h, the sintering density gradually increases while the XRD density almost stays invariable, which means the sintering densification induced by higher solid-state reaction completeness.

Cross-sectional SEM micrographs of NiZn ferrites are presented in Fig. 2. With increasing duration time of sintering, the grains grow bigger. For sample 1, intergranular pores among those small grains are mainly distributed at the grain boundaries. When the duration time of sintering rises to 3 h, the microstructure is enhanced to be more compact due to the successive grain growth and grain boundary diffusion promoted by the thermal radiation [8]. However, under the condition of 7 h sintering, the abnormal grain growth occurs.

Table 1 presents the elemental properties of the samples. M_s increases from sample 1 to sample 4 straightforward. It is well acknowledged that the joint effects of densification with molecular magnetic moment contribute to the value of M_s. For a constant molecular magnetic moment, the higher densification means more magnetic moments in the unit volume which results in the increase of M_s. An evident increase of sintering density is observed when the duration time is prolonged to 3 h which implies a drastic sintering densification and then the M_s shows a corresponding sharp increase.

The initial permeability μ_i can be described by the balance of two kinds of forces, the driving force of magnetization represented by M_s versus the resistance represented by the joint effect of anisotropy (characterized by anisotropy constant K_1) and inner stress related to porosity (P) [16]. The porosity (P), defined by $(d - d_{XRD})/d_{XRD} \times 100\%$ where d and d_{XRD} represent the sintering and XRD density, respectively, decreases initially because of the combination of reduction of pores on the grain boundaries and the growth of grain.

Besides the balance of two kinds of forces mentioned above, the transition of magnetization mechanisms is also taken into account to analyze the variation of μ_i. The average grain sizes (D) of four samples are 3.9 ± 0.2 μm, 7.5 ± 0.9 μm, 10.8 ± 1.8 μm, 12.9 ± 3.1 μm, respectively, and the grain size distribution of the homogeneous structure can be assessed by the deviation. Referring to the

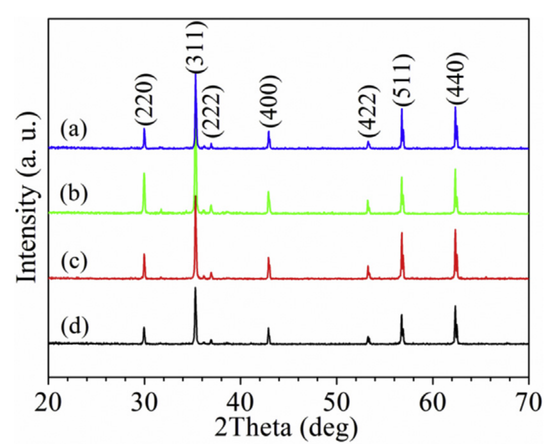

Fig. 1. XRD patterns of NiZn ferrites with different duration times (t) of sintering: (a) t = 1 h, (b) t = 3 h, (c) t = 5 h, and (d) t = 7 h.