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Effects of Ta_2O_5 addition on the microstructure and temperature dependence of magnetic properties of MnZn ferrites

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ABSTRACT

MnZn ferrites with the chemical formula $Mn_{0.68}Zn_{0.25}Fe_{2.07}O_4$ have been prepared by the conventional ceramic technique. Toroidal cores were sintered at 1350 °C for 4 h in N_2/O_2 atmosphere with 4% oxygen. Then the influence of Ta_2O_5 addition on the microstructure and temperature dependence of magnetic properties of MnZn ferrites was investigated by characterizing the fracture surface micrograph and measuring the magnetic properties over a temperature ranging from 25 to 120 °C. The results show that, when the Ta_2O_5 concentration is not more than 0.04wt%, the grain size has a slight increase with the increase of Ta_2O_5 concentration, the temperature of secondary maximum peak in the curve of initial permeability versus temperature and the lowest power loss shift to lower temperature. However, excessive Ta_2O_5 concentration (>0.04wt%) results in the exaggerated grain growth and porosity increase, which make the initial permeability and saturation magnetic flux density decrease and the power loss increase at room temperature. Furthermore, the temperature of secondary maximum peak in the curve of initial permeability versus temperature and the lowest power loss shift to about 100 °C.

1. Introduction

MnZn ferrites, capable of combining their high permeability, high resistivity, low power loss, and large saturation magnetic flux density, are widely applied as core materials for transformers in switching mode power supplies and DC/DC converters [1,2]. In application of MnZn ferrites, such as transformers, for instance, temperature of the lowest power loss is controlled to fit the operating temperature. In MnZn ferrites, the compensation temperature of magnetocrystalline anisotropy constant (T_0) coincides with the temperature of secondary maximum peak of the magnetic permeability versus temperature and also with the temperature where the power loss exhibits a minimum. So, the temperature of the lowest power loss can be obtained by changing the compensation temperature.

Properties of MnZn ferrites are determined by the main compositions, types and weights of additives, characteristics of powder and sintering condition. In order to achieve lower power loss and better magnetic properties, many different additives have to be added. There are three main categories [3]. First, the additives affect the microstructure development by introducing a liquid phase. The second category is the additives that appear at the grain boundaries as the second phase having very high resistivity. The third category is cations that are soluble in the host lattice and enter regular positions on the tetrahedral or octahedral sites. They have a fundamental influence on the intrinsic magnetic properties such as saturation magnetization, anisotropy, and the stability of the properties with time.

It is known that Ta_2O_5 can modify the grain boundary chemistry and increase the grain boundary resistivity, and can improve the power loss at the frequency range of 0.5–2 MHz in MnZn ferrites, which was prepared by the hydrothermal method [4,5]. However, there is no report about the Ta_2O_5 addition on the temperature dependence of magnetic properties in MnZn ferrites. Therefore, the influence of Ta_2O_5 addition on the microstructure and temperature dependence of magnetic properties in MnZn ferrites prepared by the conventional ceramic technique is investigated in this paper. In this study, the composition of $Mn_{0.68}Zn_{0.25}Fe_{2.07}O_4$ is chosen as the basic composition because of its better magnetic properties than other compositions in our previous investigation.

2. Experimental procedure

2.1. Sample preparation

MnZn ferrites with a composition of $Mn_{0.68}Zn_{0.25}Fe_{2.07}O_4$ were prepared by the conventional ceramic technique. The main composition was milled in an attritor with deionized water for

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1 h. After drying, the mixture of oxide powder was homogenized and calcined at 930 °C in air for 2 h. Then Ta_2O_5 was added, whose contents were 0.00, 0.02, 0.04, 0.07 and 0.10 wt%. At the same time, $CaCO_3$(0.02 wt%), TiO_2(0.05 wt%) and Co_2O_3(0.10 wt%) were added to the calcined powder. Then the resulting powder with dopants was milled in an attritor with deionized water for 4 h. After drying the milled slurry, the dried powder was granulated with about 8% poly-vinyl alcohol (PVA). The powder was pressed into samples with toroidal and disc shape, which were sintered in a computer-driven furnace at 1350 °C for 4 h with 4% oxygen and cooled at equilibrium conditions in a N_2/O_2 atmosphere. The atmosphere was controlled by using an equation for equilibrium oxygen partial pressure, which was given by Morineau [6].

2.2. Properties measurement

The XRD patterns were taken at room temperature using the Philips X'Pert PRO X-ray diffraction meter in 2θ range 20–80°. The theoretical X-ray density (d_x) of sintered samples was calculated according to the formula ($d_x = 8M/Na^3$), where M is the molecular weight, N is Avogadro's number and a is the lattice constant. The bulk density (d_m) of sintered samples was determined by the Archimedean method. The porosity percentage (P%) was calculated according to the relation $P = 100[1-(dm/dx)]$%. The fracture surface microstructures of sintered samples were observed by the JEOL JSM-6490LV scanning electron microscope. The average grain size was estimated by the intercept method [7]. The DC resistance was measured with LCR Databridge 2810 at room temperature using silver-paste contacts on both sides of sintered disc samples, and then the DC resistivity was calculated. The inductance was measured by the TH2828 precision LCR meter. Then the initial permeability was calculated. The power losses of the sintered toroids were measured at the temperature range from 25 to 120 °C by Iwatsu SY8232 B–H analyzer at 100 kHz and 200 mT.

3. Results and discussion

3.1. Effect of Ta_2O_5 addition on the microstructure of MnZn ferrites

In Fig. 1 the influence of Ta_2O_5 concentration on the microstructure of MnZn ferrites is illustrated. It indicates that the average grain size with 0.04 wt% Ta_2O_5 concentration (13.7 μm) is bigger and much more homogeneous than without Ta_2O_5 (12.5 μm) and 0.02 wt% Ta_2O_5 concentration (13.1 μm). However, the exaggerated grain growth phenomenon appears when the Ta_2O_5 concentration is 0.07wt%, and it becomes serious when the Ta_2O_5 concentration is 0.10 wt%.

One possible mechanism for the promotion of grain growth in MnZn ferrites doped with Ta_2O_5 addition is the reduction of the impurity and pore drag on the grain boundary motion. Ta^{5+} ions segregate to the grain boundary region and repel other segregants which are more detrimental to the grain growth, and then the grain boundary velocity will be enhanced [8]. Another possible grain growth promotion mechanism is the increased pore mobility due to the creation of excess cation vacancies by Ta_2O_5 addition. This is to maintain charge and site balance. Ta_2O_5 addition introduces Fe^{2+} ions and/or cation vacancies. For an addition of each Ta^{5+} ion, two Fe^{3+} ions are reduced to Fe^{2+}. Another alternative is to create cation vacancies. Addition of each three Ta^{5+} ions for three Fe^{3+} is accompanied by introduction of two cation vacancies. The cation vacancy flux generated by Ta_2O_5 addition increases the pore mobility and thus the mobility of pore-loaded grain boundaries. As a result the speed of the grain boundary movement increases, thereby promoting grain growth [9]. An impurity dopant, by either segregation to, precipitation on or melting at grain boundaries, can affect the grain boundary energy. According to the accepted grain growth kinetics represented by the relation [10]

$$D^n - D_0^n = K_0 t \exp(-Q/kT) \quad (1)$$

where D is the average grain size at time t, D_0 is the initial grain size, n is the kinetic grain growth exponent, K_0 and k are the constant, T is the absolute temperature and Q is the apparent activation energy. When time and temperature are constant, the final grain size is related to the activation energy Q associated with the specific grain growth. Therefore, the possible reason for the excessive Ta_2O_5 concentration that leads to exaggerated grain growth is the nonuniform segregation of Ta_2O_5 addition that decreases the activation energy of the grain boundary, which results in a coalescence of grains, and consequently the grains abruptly grow.

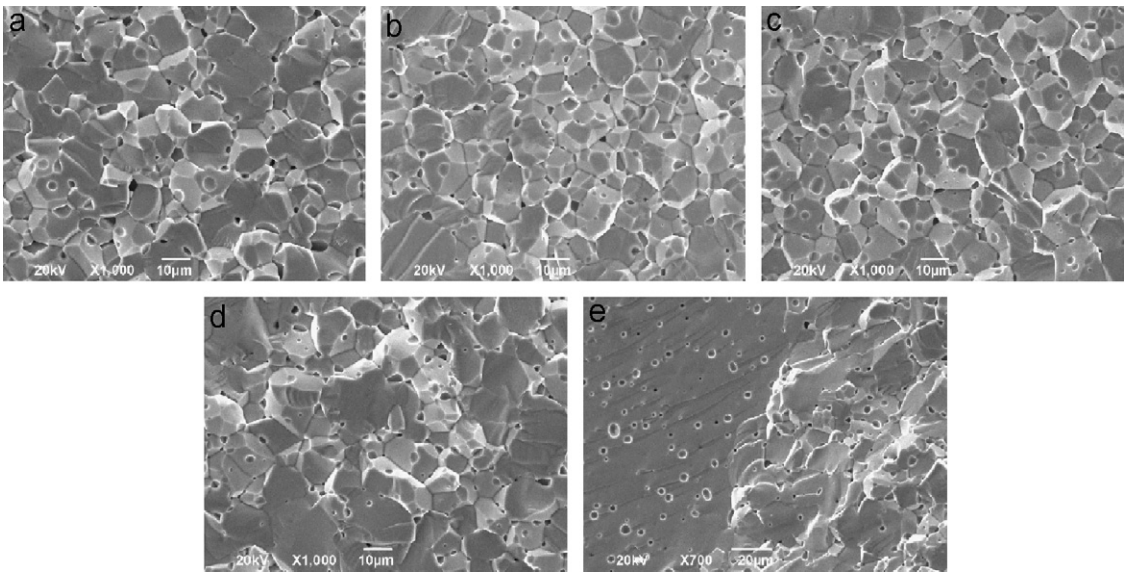

Fig. 1. SEM micrographs of sintered samples with different Ta_2O_5 concentrations. (a) 0.00 wt%; (b) 0.02 wt%; (c) 0.04 wt%; (d) 0.07 wt%; and (e) 0.10 wt%.