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Structural, electromagnetic, and dielectric properties of lithium-zinc ferrite ceramics sintered by pulsed electron beam heating

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ABSTRACT

Structural, electromagnetic, and dielectric properties of $Li_{0.4}Fe_{2.4}Zn_{0.2}O_4$ lithium-zinc ferrite sintered by 2.4 MeV pulsed electron beam heating at 1050 °C for 2 h were investigated. The formation of ferrite with a single-phase cubic spinel structure was confirmed by X-ray diffraction analysis. The average grain size of ferrite ceramic was determined by SEM analysis and its value was 1.7 μm. The radiation-thermal sintered samples are characterized by a saturation magnetization of 67.8 emu/g, the Curie temperature of 508 °C, AC electrical resistivity of 2.4×10^4 Ω cm (at 25 °C). The frequency dependences of permittivity and the loss tangent were obtained in $(20 - 2\times10^6)$ Hz frequency range. The behavior of ε′ is characterized by high dispersion caused by relaxation polarization in the investigated frequency range. The results were compared to the LiZn ferrite characteristics sintered by traditional thermal heating.

1. Introduction

It is known that the lithium ferrites on base of $Li_{0.5}Fe_{2.5}O_4$ are the most important functional materials of modern microwave electronics [1–3]. In addition, they are used as cathode material in lithium-ion batteries as well as sensor elements of gas sensors [4–6]. Basic requirements applied to microwave lithium ferrites, are: high electrical resistance providing a small dielectric loss of materials, temperature stability determined by the Curie temperature of materials, and nominal value of saturation magnetization.

The magnetic and electrical properties of $Li_{0.5}Fe_{2.5}O_4$ can be modified by substitution of Fe^{3+} ions with magnetic and/or diamagnetic cations in either sublattice. The substitution of Fe^{3+} ions in lithium ferrite to Zn^{2+} ions leads to increasing saturation magnetization. [7–9]. Thus, substituted lithium-zinc (LiZn) ferrites with the chemical composition of $Li_{0.4}Fe_{2.4}Zn_{0.2}O_4$ are characterized by high values of saturation magnetization and Curie temperature [10,11] and, thereby, provide active interaction with electromagnetic waves in the low frequency band of the microwave range.

As a soft magnetic material LiZn ferrites are very sensitive to structural and phase imperfections. Obviously, the microstructure (porosity and grain size) affects the magnetic and dielectric characteristics of ferrites [12–14]. Sintering processes, which determine the quantitative and qualitative change in porosity, and recrystallization processes forming a grain structure occur simultaneously and are interconnected. Microstructure leads to deterioration of magnetic and dielectric properties of ferrite material when the grain sizes are sharply different from each other in it. Thus, the creation of an optimum microstructure during the sintering process is critical for ferrites production with the required set of properties.

The main method to manufacture high-density LiZn ferrite ceramics is the ceramic method implying of usage both the high temperatures and long time. However, the high temperatures lead to lithium and oxygen volatility at temperatures above 1050 °C by deteriorating the magnetic and electric properties [15].

The improvements of ferrite materials basic characteristics can be achieved by controlling the structure due to changes of technological methods in ferrites production. As it has been shown previously [16–20], the radiation-thermal heating with high-energy electron beams is the effective way of ferrites production. A large number of researches is related to the ferrite powders synthesis process to obtain ferrites with a homogeneous phase composition at low temperatures.

In [21], it was demonstrated that the ferrite ceramic samples sintered by using 2.4 MeV electrons beam heating at 1100 °C exhibit a higher density and less porosity. These samples were characterized by lower electrical resistivity and high saturation magnetization in comparison with the same samples produced by conventional thermal sintering.

This paper presents the results of structural and electrical properties of LiZn ferrite sintered by high-energy electron beam heating at

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temperature of 1050 °C. For the comparative analysis of the obtained results, ferrite sintering was carried out by two methods: conventional thermal (T) heating and radiation-thermal (RT) heating by high-energy electron beam.

2. Experimental

Fe_2O_3, Li_2CO_3, and ZnO industrial powders were used for $Li_{0.4}Fe_{2.4}Zn_{0.2}O_4$ lithium-zinc ferrite preparation. The precursors were pre-mixed and mechanical activated by the AGO-2S planetary mill (Novic, Russia) using steel grinding jars and balls at room temperature for 60 min. The weight ratio of the material to the balls was 1:10, and the ball mill rotation speed was 2220 rpm. After mechanical activation the samples were pressed into compact pallets with a diameter of 15 mm and a thickness of 2 mm using the method of single-action cold compaction. The compaction pressure was 200 MPa. Thermal synthesis of the samples was conducted at the temperature of 800 °C during 120 min in a laboratory resistance furnace to obtain ferrite powders with a homogeneous phase composition.

In order to obtain ferrite ceramic samples the ferrite powders were divided by two parts and sintered via T and RT heating at 1050 °C for 140 min in both cases. T sintering of the samples was performed using laboratory resistance furnace.

The ILU-6 pulsed electron accelerator developed in Institute of Nuclear Physics of the SB RAS (Novosibirsk, Russia) was used for RT sintering [22]. The electron energy of ILU-6 was 2.4 MeV; the pulse beam current, the pulse duration, and the pulse repetition rate were 400 mA, 500 μs, (12.5–25) Hz, respectively. The samples were heated by the electron beam in air using the special cell described in [21]. The temperature was controlled by thermocouple (type S) located in control sample that placed in the immediate vicinity of the investigated samples. The thermal processing regime was regulated by the pulse repetition frequency of the electron beam. The experimental results for temperature variation in the control sample at the different beam current densities are shown in [16].

The phase composition of the samples was controlled using an ARL X'TRA x-ray diffractometer (Switzerland) with a Peltier Si (Li) semiconductor detector and Cuk_α radiation. XRD patterns were measured for $2\theta = (10 - 80)°$ with a scan rate of 0.02°/s. The phase compositions of the examined samples were determined using the PDF-4 powder database of the International Centre for Diffraction Data (ICDD). The XRD patterns were processed by the full-profile Rietveld analysis using the Powder Cell 2.5 software.

The density and apparent porosity of ceramic samples was measured by hydrostatic weighing using Shimadzu AUW 220D high-precision analytical balance. The grain morphology of the sintered samples was examined using a SEM Philips 515 type scanning electron microscope (SEM). The intercept method was used for calculation of the average grain size.

The Curie temperature (T_c) of the samples was measured by thermomagnetometry method which is the thermogravimetric TG/DTG analysis of samples in the magnetic field [23,24]. TG/DTG measurements were performed with the STA 449C Jupiter thermal analyzer (Netzsch, Germany).

The saturation magnetization (M_s) was measured at room temperature with the vibrating sample magnetometer with the maximum field of 10 kOe.

The AC electrical resistivity (ρ) and dielectric parameters were measured by two-electrode method using the LCR Meter. Silver electrodes were deposited on the sample surface (sample thickness of 0.24 mm) through thermal evaporation in vacuum. The diameter of the measuring electrode was 5 mm. The measurements were carried out in the $(20–2\times10^6)$ Hz test signal frequency range at room temperature.

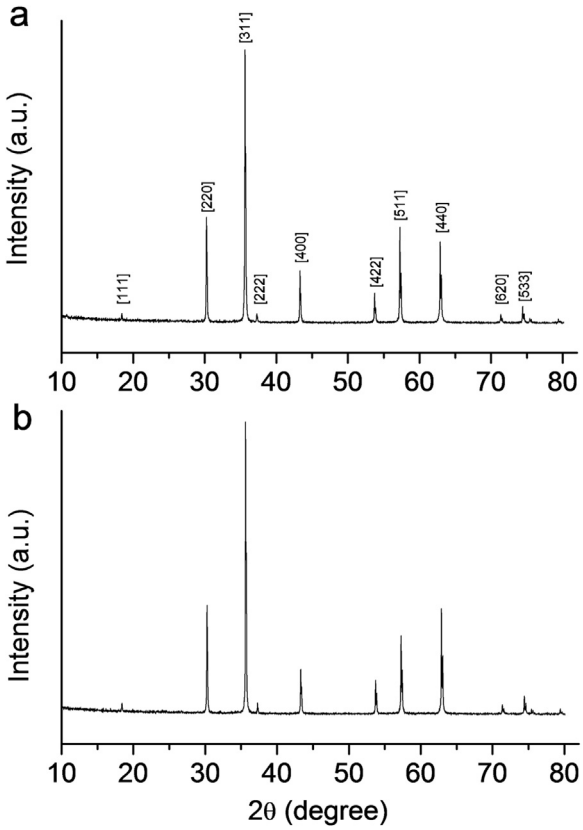

Fig. 1. X-ray diffraction patterns for samples sintered by RT (a) and T (b) methods.

3. Results and discussion

The X-ray diffraction patterns, shown in Fig. 1 for LiZn ferrite samples sintered by T and RT methods at 1050 °C, confirmed the well-defined homogeneous single phase cubic spinel structure in the samples with $Li_{0.4}Fe_{2.4}Zn_{0.2}O_4$ composition (PDF No 04–007-4738). The (*hkl*) values corresponding to the diffraction peaks are marked in Fig. 1. The crystallite size of the sintered samples was calculated by Williamson-Hall's formula, and the lattice parameter which was calculated by PowderCell 2.5 software, are summarized in Table 1. As it can be seen, there is a slight increase in the lattice parameter and a decrease in the crystallite size for RT sintered samples compared to the T sintered samples.

Fig. 2 shows SEM micrographs for LiZn ferrite samples produced by RT and T sintering at 1050 °C. For RT sintered samples, the ferrite ceramic is characterized by a polycrystalline structure with well-formed grains. The average grain size is 1.7 μm. In this case, the relative density attains 90.1% of the theoretical density determined by XRD analysis. Porosity is lower than 10%.

The T sintered samples are characterized by a smaller average grain size and larger porosity compared to the RT sintered samples. The relative density of such samples is 79.5%. SEM micrograph shows an intermediate sintering stage characterized by fritting the powder particles in the contact areas without well-formed grid of the grain boundaries.

The saturation magnetization values presented in Table 1 are similar for both types of ferrite ceramic and are equal to the nominal operating characteristics of M_s for lithium-zinc ferrite. The saturation magnetization depends mainly on the chemical composition of the ferrite, and the synthesis process determines its value. The aim of this synthesis is to obtain a ferrite with a single-phase composition. Since the synthesis, regimes were the same for both types of ferrites it is possible to expect close values of saturation magnetization.

Fig. 3 presents the thermogravimetric (TG) and derivative thermo-

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