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A study of ultrasonic velocity and attenuation on nanocystalline MgCuZn ferrites

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ABSTRACT

The nanocrystalline MgCuZn ferrites with particle size (~30 nm) have been synthesized by microwave–hydrothermal (M–H) method at 160 °C/45 min. The powders were densified at 750–900 °C/30 min using microwave sintering method. The sintered samples were characterized using X-ray diffraction and scanning electron microscope. The grain sizes of the sintered samples are in the range of 60–80 nm. The ultrasonic velocities have been measured on MgCuZn ferrites using the pulse transmission method at 1 MHz. The ultrasonic velocity is found to decrease with an increase of temperature. A small anomaly is observed around the Curie temperature, 520 K. The anomaly observed in the thermal variation of longitudinal velocity and attenuation is explained with the help of magneto-crystalline anisotropy constant.

1. Introduction

Recently, the surface mount devices (SMD) technology has been rapidly developed for miniaturation of electronic devices such as a multiplayer chip inductor. The multiplayer chip inductor is produced by coating ferrite and electrode layers alternatively and co-firing them [1]. To prevent the interfacial diffusion of electrode material into the ferrite layer, the multilayer structure must be sintered below the melting point of electrode material; e.g. Ag electrode melting temperature is 961 °C. It was found that these ferrites are comparatively sensitive to mechanical and magnetic properties and are easily changed or deteriorated by the stress caused at the internal electrode [2]. These problems can be reduced by preparation of MgCuZn ferrites under controlled experimental conditions and the knowledge of elastic properties.

The nanopowders of MgCuZn ferrites can be synthesized using various methods such as Co-precipitation, combustion, sol–gel, precursor, etc. [1,3,4]. Out of all the methods, the microwave–hydrothermal (M–H) method is one of the promising methods to produce ceramic nanoparticles [5–7]. Therefore, in the present investigation, MgCuZn ferrites were synthesized using microwave–hydrothermal method. The prepared powders were characterized using X-ray diffraction (XRD) and transmission electron microscope (TEM). The powders were densified using microwave sintering method [8]. A detailed study of elastic behavior of nanocrystalline MgCuZn ferrites has been undertaken and the obtained results are presented in this paper.

2. Experimental method

The nanocrystalline $Mg_{0.2}Cu_{0.3}Zn_{0.5}Fe_2O_4$ (hereafter MgCuZn ferrite) powder has been synthesized using microwave–hydrothermal method [9]. The high purity (99.9%) chemicals magnesium nitrate [$Mg(NO_3)_2 \cdot 6\,H_2O$], copper nitrate [$Cu(NO_3)_2 \cdot 3\,H_2O$], zinc nitrate [$Zn(NO_3) \cdot 6\,H_2O$] and ferric nitrate [$Fe(NO_3)_2 \cdot 9\,H_2O$] were weighed and the molar ratio of powders was adjusted to obtain composition $Mg_{0.2}Cu_{0.3}Zn_{0.5}Fe_2O_4$, and dissolved in de-ionized water. Sodium hydroxide (NaOH) was added with controlling of pH~9.2. The mixture was then treated in a Teflon lined vessel using a microwave digestion system (Model MDS-2000, CEM Corp., Mathews, NC). This system uses 2.45 GHz microwaves and can operate at 0–100% full power (1200 ± 50 W). The system is controlled by pressure and can attain maximum pressure of 200 psi, which is equivalent to 194 °C, based on steam tables. In the present investigation all the samples were synthesized at 160 °C/45 min. The reaction vessel is connected to a pressure transducer that monitors and controls the pressure during synthesis. The time, pressure and power were computer controlled. The products obtained were filtered, and then washed repeatedly with de-ionized water, followed by freeze-drying overnight. The prepared powders were weighed and the percentage yields were calculated from the expected total amount based on the solution concentration and volume and the amount that was actually crystallized. An average of 93% yield was obtained.

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All the synthesized products were characterized using powder X-ray diffraction (XRD) (PANanalytical X'pert) with CuKα radiation. Particle size and morphology of the as-synthesized powders were determined using transmission electron microscope (TEM) (Model JEM-2010, JEOL, Tokyo, Japan). The obtained ferrite powder was mixed with an appropriate amount of 2 wt% polyvinyl alcohol as a binder. Then the powder was uniaxially pressed at a pressure of 1500 kg/cm^2 to form green pellet and toroidal specimens. The compacts were microwave sintered at 750, 800, 850 and 900 °C/30 min. Sintered samples were characterized using XRD and scanning electron microscope (SEM).

A conventional ultrasonic pulse transmission technique [10] was used to measure the ultrasonic velocities (V_l and V_s) at 1 MHz at room temperature. The r.f. pulse generated by a pulse oscillator was applied to quartz transducer. The acoustic pulses were converted into electrical signals by the receiving transducers. The output signal was displaced on a digital textronic 2230 oscilloscope. The time difference Δt between two overlapping received pulse train was noted with the help of timer. The sound velocity was measured using the equation $V=L/\Delta t$, where V is sound velocity, L is length of the ferrite specimen and t is the time. The accuracy of the sound velocity measurement was $\pm 0.5\%$.

Only the calibrated ones have been used throughout, and all the observations were taken using either 2- or 5-s sweep speeds. One-megahertz PZT crystals are used for the measurements. Stainless steel holders were employed to house the PZT crystals and the delay imposed by the metallic ends of the stainless steel holders has been corrected. With this arrangement, measurements were possible to a high degree of accuracy as the error in the compressional velocity measurements was less than 1%.

The values of Young's (Y) modulus, shear (η) modulus and bulk (K) modulus were calculated from the given formulae. The accuracy of ultrasonic velocities and attenuation measurements was 0.01% and 0.2%, respectively.

$$Y = (V_l)^2\rho$$

$$\eta = (V_S)^2\rho$$

$$K = 2/3\eta[(1+\sigma)/(1-\sigma)]$$

where V_l and V_S are longitudinal and transverse velocities, respectively and ρ is the bulk density of the sample.

The magnetic field dependence of elastic properties at room temperature was measured keeping the sample in between the pole pieces of an electromagnet. The magnetic field was always applied parallel to the length of the sample. The magnetic field was measured using the Hall probe. This arrangement allowed us to measure the field and temperature dependence of the ultrasonic velocities and attenuation. The maximum field used in the present investigation was 0.5 T. This field is sufficient to saturate the present samples. The entire arrangement has been placed in a vacuum of 10^{-4} torr. The accuracy in the velocity and attenuation measurements is 0.05% and 0.3%, respectively.

3. Results and discussion

Fig. 1 shows the XRD patterns of as-synthesized MgCuZn ferrite powder. It can be seen from the figure that the single-phase ferrite was obtained and no other phases were detected. The average particle size (D_m) of the as-synthesized powders has been estimated with the help of XRD patterns using Scherer's equation:

$$D_m = K\lambda/\beta\, Cos\,\theta$$

where K is a constant, β is the full width half maxima and λ is the wavelength of X-rays used and θ is the diffraction angle. The average particle size calculated from the above formula for the MgCuZn ferrite is ~25 nm.

Fig. 2 shows the TEM picture of as-synthesized MgCuZn ferrite powder. It can be seen from the picture that the particles are more or less spherical in morphology and uniform in size. The particle size is found to be ~30 nm. Proper control of the pH is the key factor in synthesizing nano-phase ferrites with low particle size.

Fig. 3 shows the XRD patterns of MgCuZn ferrite sintered at different temperatures. All the diffraction peaks were indexed and matched well with the JCPDS card no: 08–0234. The values of lattice constant (a), bulk density (d_b), X-ray density (d_x) and porosity ($\%P$) were given in Table 1. The crystallinity of the sintered samples increases with an increase of sintering temperature, therefore the lattice constant increases. The bulk density of the samples was measured using Archmedic principle. As the sintering temperature increases bulk density of the sample increases due to the increase in the particle sizes. The accuracies in the density measurements will be $\pm 0.01\%$. Using the X-density and bulk density data, the value of porosity was estimated and it is found to be in the range of 1%.

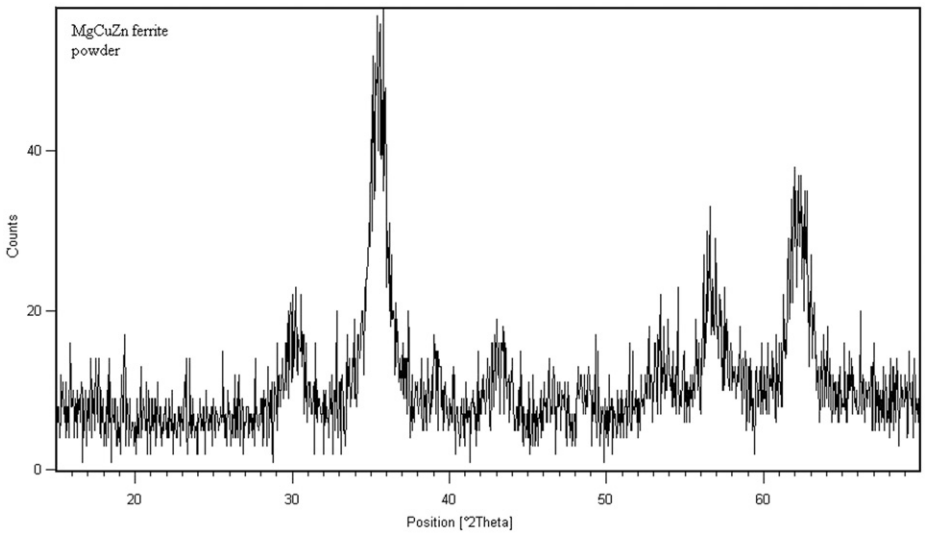

Fig. 1. XRD pattern of as-synthesized MgCuZn ferrite powders.