Journal of Magnetism and Magnetic Materials 322 (2010) 2819–2823

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Effect of sintering temperature on structural and magnetic properties of NiCuZn and MgCuZn ferrites

M. Penchal Reddy [a,*], W. Madhuri [b], M. Venkata Ramana [a], N. Ramamanohar Reddy [c], K.V. Siva Kumar [a], V.R.K. Murthy [d], K. Siva Kumar [e], R. Ramakrishna Reddy [a]

[a] Ceramic Composite Materials Laboratory, Department of Physics, Sri Krishnadevaraya University, Anantapur 515055, India
[b] School of Sciences, VIT University, Vellore 632014, India
[c] Department of Materials Science and Nanotechnology, Yogi Vemana University, Kadapa 516003, India
[d] Microwave Laboratory, Indian Institute of Technology, Chennai 600036, India
[e] Department of Physics, Sri Venkateswara University, Tirupati 517502, India

ARTICLE INFO

Article history:
Received 4 January 2010
Received in revised form
8 April 2010
Available online 24 April 2010

Keywords:
Ferrites
Microwave heating
Initial permeability
Saturation magnetization

ABSTRACT

The low temperature microwave sintered NiCuZn and MgCuZn ferrites with compositions $Ni_{0.35}Cu_{0.05}Zn_{0.60}Fe_2O_4$ and $Mg_{0.35}Cu_{0.05}Zn_{0.60}Fe_2O_4$ were synthesized by conventional mixed oxide method. NiCuZn and MgCuZn ferrite samples obtained showed better sintered densities at 950 and 900 °C, respectively. The scanning electron micrographs of both the ferrite samples appear to be very much similar. The magnitude of initial permeability of MgCuZn ferrite samples is found to be obviously higher than those of NiCuZn ferrite samples at all sintering temperatures. This is mainly due to the fact that MgCuZn ferrite has smaller magnetocrystalline anisotropy constant and magnetostrictive constant. NiCuZn ferrites have higher saturation magnetization than MgCuZn ferrites, which is due to the higher magnetic moment of NiCuZn ferrites. Our results indicate that the microwave sintering method seems to be a potential technique in the MLCI technology.

1. Introduction

Spinel ferrites are used in the fabrication of multilayer chip inductors (MLCIs) as surface mount devices for miniaturized electronic products such as cellular phones, notebook computers, video camera recorders, floppy drives, etc. [1]. The chip inductor is fabricated by arranging alternate layers of ferrite and silver electrode. This multilayer ceramic metal composite should be co-fired below 950 °C to suppress the interfacial diffusion of Ag metal into ceramics as the melting point of Ag is 961 °C. Till now, Ni–Cu–Zn ferrites have been the dominant core materials for multilayer chip inductor (MLCI) applications due to their high density, high initial permeability at high frequency and low sintering temperature (<960 °C) [2–6]. MgCuZn ferrites have similar magnetic properties to those of NiCuZn ferrites with the advantage that they are economical [7] and easy to synthesize. Therefore, MgCuZn ferrite will be a promising material for MLCIs with better performance and low cost.

In the present work, the NiCuZn and MgCuZn ferrite samples were synthesized by the microwave sintering method, which is an exciting new field in material science with enormous potential for synthesizing new materials and novel microstructures [8,9]. The growing interest during the past decade is essentially due to the possibility of a reduction in manufacturing cost on account of energy savings, high energy efficiency, shorter processing times and improved product uniformity and yields [10,11].

Few studies on the comparison of electromagnetic properties of NiCuZn and MgCuZn ferrites are available in the literature [12]. We aimed to produce low-cost ferrite materials according to the desired application by easy and low-cost synthesis technique. To the best of our knowledge, the detailed comparison of low-temperature microwave sintered NiCuZn and MgCuZn ferrites has not yet been reported. In this paper, the physical, microstructure and magnetic properties of the polycrystalline NiCuZn ferrite were investigated and compared with the same composition of MgCuZn ferrite sample.

2. Experimental procedure

The NiCuZn and MgCuZn ferrite samples having the chemical formula $Ni_{0.35}Cu_{0.05}Zn_{0.60}Fe_2O_4$ and $Mg_{0.35}Cu_{0.05}Zn_{0.60}Fe_2O_4$ were prepared by microwave sintering method at Ceramic Composite Materials Laboratory, S.K. University, Anantapur, India. The analytical grades of NiO, MgO, CuO, ZnO and Fe_2O_3 were weighed according to the corresponding composition. These oxides were

* Corresponding author. Tel.: +91 8554 255704; fax: +91 8554 255710.
E-mail address: penchu321@gmail.com (M. Penchal Reddy).

0304-8853/$ - see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2010.04.036

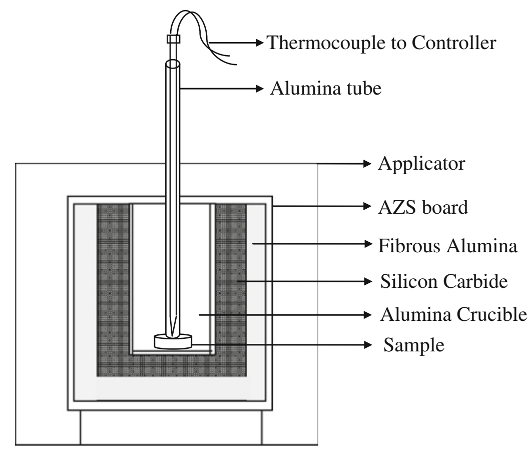

Fig. 1. Schematic view of sample environment in the microwave cavity.

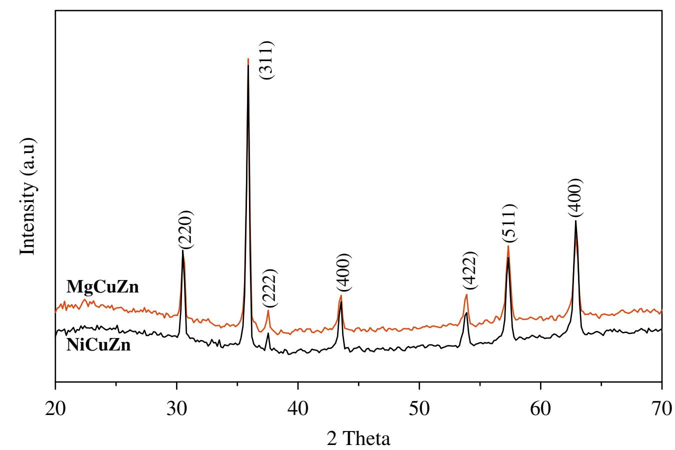

Fig. 2. XRD patterns of NiCuZn and MgCuZn ferrites sintered at 950 °C.

intimately mixed and the resulting powders were ball milled using a planetary ball mill (Restch PM 200, Germany) in agate bowls with agate balls in acetone medium for 20 h. The slurry was dried and loosely pressed into cakes using a hydraulic press. These cakes were pre-sintered at a temperature of 800 °C for 4 h in closed alumina crucibles. The pre-sintered cakes removed from the furnace were crushed and ball milled in an acetone medium in agate bowls with agate balls for another 30 h to obtain fine particle size. These slurries after drying were sieved to obtain a uniform particle size. The green powders were then pressed into disk- and toroid-shaped samples.

In this study, a modified microwave oven (Sharp; 1.1 kW, 2.45 GHz) was used to sinter NiCuZn and MgCuZn ferrite samples. Fig. 1 shows the experimental settings in the microwave oven. The general configuration of the microwave furnace has been described elsewhere [13]. The green pressed samples were finally sintered at temperatures from 850 to 1000 °C in 25 °C interval and the sintering time was 30 min.

The sintered ferrites were characterized with respect to phase identification and lattice parameter determination using X-ray diffraction (PW1730 Germany) with Cu K_α radiation (1.5405 Å). Bulk density was determined from mass and bulk volume of sample pellets. The surface structure of the samples was observed by the use of a scanning electron microscope (SEM, CRL-ZESIS-EVO-MAI5). The initial permeability (μ_i) of the ferrite toroids were evaluated using the standard formula [13] from the inductance measurements carried out at 10 kHz using a computer controlled impedance analyzer (Hioki Model 3532-50 LCR HiTester, Japan). These measurements were carried out in the temperature range 30–200 °C at 10 °C temperature intervals. Saturation magnetization (M_s) measurements were carried out at room temperature using a vibrating sample magnetometer (VSM) (Lakeshore-7040) with a maximum magnetic field of 20 kOe.

3. Results and discussion

X-ray diffraction patterns of NiCuZn and MgCuZn ferrite samples sintered at 950 °C are shown in Fig. 2. The XRD patterns exhibit single phase cubic spinel structure. The typical scanning electron micrographs (SEMs) of NiCuZn and MgCuZn ferrite samples are shown in Fig. 3; apparently there are no obvious differences in NiCuZn and MgCuZn ferrite samples. The grain size seems to be increasing with increase in sintering temperature up to 950 and 900 °C for NiCuZn and MgCuZn ferrites, respectively; after that, there is a dramatic decrease in grain size in both the types of samples. At this sintering temperature, it appears that the powders have a relatively high reactivity so as to obtain appropriate speedy grain growth. When the sintering temperature is lower than the above optimum temperatures of 950 and 900 °C for NiCuZn and MgCuZn ferrites, respectively, plethoric pores might exist in sintered powders, which stunt grain growth during sintering [14].

Fig. 4 shows the X-ray density and bulk density of NiCuZn and MgCuZn samples sintered at various temperatures. The ferrite density seems to be increasing with increase in sintering temperature from 850 to 950 °C and 850 to 900 °C, respectively for NiCuZn and MgCuZn ferrites; thereafter the densities tend to decrease. According to Lange and Kellet [15], grain growth and densification are intimately related. Thus in the present investigation, the formation of pores within the grains or grain boundaries may be responsible for the observed decrease in density. A similar type of observation made by variation of the density with sintering temperature was observed by Murthy [16] in the case of NiCuZn ferrites and Haque et al. [17] in the case of MgCuZn ferrites.

In spite of a series of attempts to fire at various sintering temperatures, the MgCuZn ferrites exhibited only lower densities than those of NiCuZn ferrites. This can be attributed to the fact that magnesium has lower atomic weight (24.30 amu) than the nickel (58.69 amu) atoms. So when nickel atoms are replaced by magnesium, the density decreases [12]. From the figure it is evident that MgCuZn ferrites will get better sintering density at lower sintering temperatures than those of NiCuZn ferrites. It can also be noticed from the figure that X-ray density of each sample (NiCuZn and MgCuZn ferrites) is higher than the corresponding bulk density of sintered samples. This is attributed to the existence of the pores, which depends on sintering conditions. Also this might be due to the fact that the density of magnesium (1.74 g/cm^3) is lower than that of nickel (8.91 g/cm^3) as well as the atomic concentration of magnesium (4.3×10^{22} cm^{-3}) as compared with nickel concentration (9.14×10^{22} cm^{-3}) [18].

Fig. 5 shows the dependence of initial permeability of NiCuZn and MgCuZn ferrite samples on the sintering temperature. Initial permeability increases first and reaches its peak value with increase in sintering temperature at 950 °C for NiCuZn ferrite and 900 °C for MgCuZn ferrite, and thereafter the initial permeability begins to decrease. This behaviour of initial permeability seems to be in accordance with the densities of the samples as the maximum permeabilities are recorded for samples with higher densities. Hence increase in permeability may be attributed to the increase in sintering density.

Magnetic properties sensitively depend on the microstructure of ferrites, which in turn are highly affected by the sintering