Materials Letters 122 (2014) 129–132

Contents lists available at ScienceDirect

Materials Letters

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

The effect of sintering temperature on the electromagnetic properties of nanocrystalline MgCuZn ferrite prepared by sol–gel auto combustion method

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ARTICLE INFO

Article history:
Received 3 November 2013
Accepted 8 February 2014
Available online 17 February 2014

Keywords:
Magnetic materials
Sintering
Electroceramics
Nanocrystalline materials
Initial permeability
Electromagnetic properties

ABSTRACT

In this work, the nanocrystalline MgCuZn ferrite powder was prepared through the nitrate–citrate auto-combustion route. The as-burnt powder after calcination at 600 °C, sintered at different temperatures below silver melting point in the range of 850–950 °C for 4 h. The structural and magnetic properties were investigated as a function of sintering temperatures. The X-ray diffraction patterns exhibited the formation of a single phase cubic spinel structure. The microstructural evaluations showed homogeneous grains and also revealed that the variation of sintering temperature significantly affected the densification and grain growth of the samples. The density, grain size, initial permeability (μ_i) and saturation magnetization (M_s) of all the samples increased with sintering temperature. The relative quality factor (RQF) also showed highest value for the sample sintered at 950 °C.

1. Introduction

The polycrystalline spinel ferrites are important ceramic materials due to their high electrical resistivity, high Curie temperature and thermal stability. These materials are widely used in the fabrication of multi-layer chip inductors (MLCIs) to miniaturize electronic products such as notebooks, cell phones and video cameras. MLCIs are fabricated by arranging alternate layer of ferrite and silver electrodes and then co-fired below silver melting point (961 °C) to prevent the interfacial diffusion of silver [1]. NiCuZn ferrites are used widely for the fabrication of chip inductors. However, the magnetic properties of MgCuZn and NiCuZn ferrites are same but the magnetostriction constant of MgCuZn is lower. Therefore, MgCuZn ferrite will be a potential candidate material for MLCIs with high performance and low cost. Generally, the magnetic properties of ferrites are strongly dependent on the chemical composition and method of preparation [2]. The sol–gel auto combustion method is a promising technique for the preparation of nanosized ferrite powders with high surface energy that exhibit high-sintering activity to dense at less than 950 °C with an optimized microstructure [3]. The aim of this work is to study the structure, microstructure and electromagnetic properties of nanocrystalline MgCuZn ferrites as a function of sintering temperatures (T_s).

2. Experimental method

The $Mg_{0.3}Cu_{0.2}Zn_{0.52}Fe_{1.98}O_{3.99}$ powder was prepared through the nitrate–citrate auto combustion technique using the analytical grade $Ni(NO_3)_2 \cdot 6H_2O$, $Zn(NO_3)_2 \cdot 6H_2O$, $Cu(NO_3)_2 \cdot 3H_2O$, $Fe(NO_3)_3 \cdot 9H_2O$ and citric acid. First, the solution of metal nitrates were dissolved in water. Then, an aqueous citric acid solution was added to the mixture in 1:1 M ratio of nitrates to citric acid. After adjusting the pH value with ammonia to 7, the resultant solution was heated at 80 °C under constant stirring to transform into a xerogel. During the heating process, the dried gel burnt out in a self-propagating combustion manner to form a fluffy powder. The as-burnt precursor powder was calcined at 600 °C in air for 2 h, granulated using 2 wt% PVA as a binder and uniaxially to form toroid. Finally, the pressed samples were sintered separately at 850, 875, 900, 925 and 950 °C for 4 h.

The structural, microstructural and electromagnetic properties of nanocrystalline MgCuZn ferrites were investigated by X-ray diffraction, scanning electron microscopy, vibrating sample magnetometer

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http://dx.doi.org/10.1016/j.matlet.2014.02.027
0167-577X

and RF-Impedance analyzer (Agilent 4991A) with test fixture (Agilent 16453A).

3. Results and discussion

Phase analysis: Fig. 1 shows the XRD patterns of the as-burnt, calcined and sintered ferrites. The calcined powder presents the main peaks corresponding to typical Mg–Cu–Zn ferrite spinel phase with good crystallization. Clearly, no any other impurity phases are detected in the pattern. The XRD patterns of the samples sintered at different temperatures show intense sharp peaks that can be indexed as a single phase cubic spinel structure using the standard JCPDS Card no. 08-0234. The figure also shows that the broadening of the diffraction peak decreases from as-burnt to sintered ferrites, which indicates an increase in the nanocrystalline size of samples. The lattice parameter 'a' was calculated using the relation $1/d^2=(h^2+k^2+l^2)/a^2$ where (hkl) are the Miller indices and d is the interplanar distance. The values of 'a' show no significant variation by T_s, that is in good agreement with [4]. The values of the lattice parameter and the X-ray density are 8.401 Å and 5.118 g/cm^3, respectively.

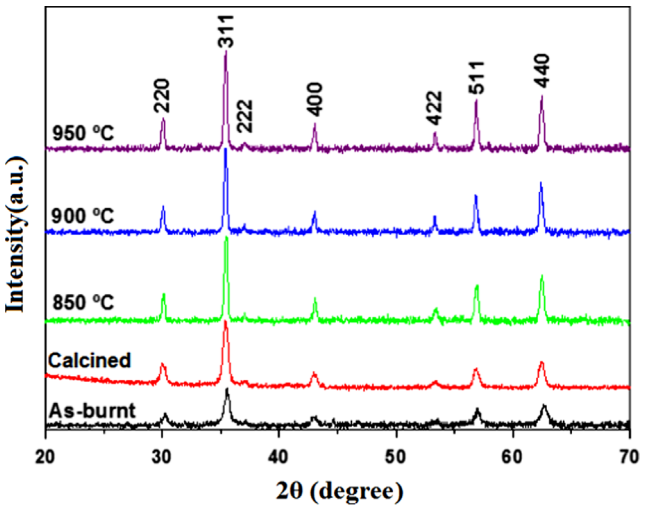

Fig. 1. X-ray diffraction patterns of the MgCuZn ferrite for as-burnt, calcined and sintered samples at different temperatures.

Table 1
Bulk density (ρ_b), grain size (D), initial permeability (μ_i) and resonance frequency (f_r) of MgCuZn ferrite sintered at different temperatures.

T_s (°C)	ρ_b [g/cm^3]	D(μm)	μ_i (1 MHz)	f_r (MHz)
850	4.51	0.30	57.68	24.23
875	4.55	0.41	67.34	20.79
900	4.60	0.62	133.41	11.07
925	4.77	0.99	195.78	6.87
950	4.82	1.21	229.6	4.68

Microstructure: Density plays a key role in controlling the properties of polycrystalline ferrites. The variation of bulk density (ρ_b) is shown in Table 1. It can be seen that the density increases with the sintering temperature. During the sintering process, a force that is generated by the thermal energy drives the grain boundaries to grow over pores and as a result reduces the pores volume and their grain boundaries. The strength of the driving force depends upon the diffusivity of individual grains, sintering temperature and porosity [5]. Obviously, the reduction of pore volume makes the material dense.

Fig. 2 shows the typical TEM and SEM photographs of the calcined nanopowder and sintered MgCuZn ferrites, respectively. The calcined powder indicates the spherical particles with an average particle size of 96 nm (Fig. 2(a)). As shown in Fig. 2(b–d), the microstructures reveal that the grain size is influenced by the sintering temperature. With the increase of sintering temperature from 850 to 950 °C (Fig. 2(b–d)), the grain size increases, from ~0.3 to ~1.2 μm. One can see that the grain size increases, while the porosity decreases with sintering temperature [6]. The micrographs of the samples that were sintered below 900 °C show the existence of many pores distributed at the grain boundaries. Such porous structure can explain why the bulk density is lower than the X-ray density. The results of the bulk density are listed in Table 1. The sample that is sintered at 950 °C shows a dense microstructure and larger grain size.

Magnetic properties: It is well-known that permeability and saturation magnetization are influenced by not only the intrinsic factors such as preferential site occupancy, but also the extrinsic factors like density and microstructure (grain size and porosity) [7].

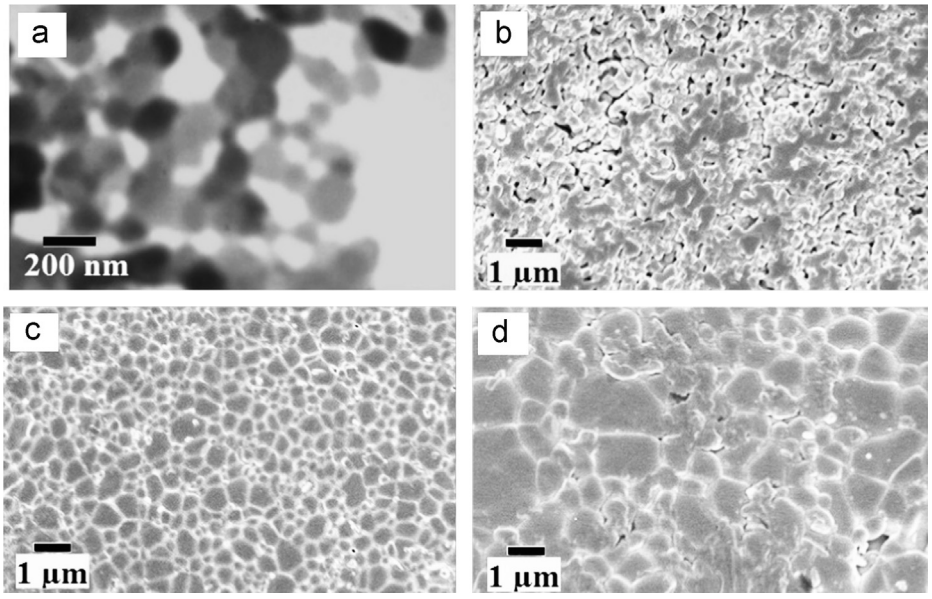

Fig. 2. The micrographs of calcined powder (a) and sintered samples at 850 (b), 900 (c) and 950 °C (d).