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Synthesis, structural, and electromagnetic properties of $Mn_{0.5}Zn_{0.5-x}Mg_xFe_2O_4$ (x=0.0, 0.1) polycrystalline ferrites

Md.D. Rahaman [a,*], K.K. Nahar [a], M.N.I. Khan [b], A.K.M. Akther Hossain [c]

[a] Deparment of Physics, University of Dhaka, Ramna, Dhaka 1000, Bangladesh
[b] Materials Science Division, Bangladesh Atomic Energy Commision, Ramna, Dhaka 1000, Bangladesh
[c] Department of Physics, Bangladesh University of Engineering and Technology (BUET), Ramna, Dhaka 1000, Bangladesh

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ABSTRACT

Polycrystalline Mn–Zn and Mn–Zn–Mg ferrites with compositions $Mn_{0.5}Zn_{0.5}Fe_2O_4$ (MZFO) and $Mn_{0.5}Zn_{0.4}Mg_{0.1}Fe_2O_4$ (MZMFO) were synthesized by the solid sate reaction method at different sintering temperatures. Samples were characterized by X-ray powder diffraction (XRD) and scanning electron microscopy (SEM). XRD patterns showed the single phase of the spinel-type ferrite with hematite as impurity phase. Surface morphology reveals that the grain size increases with sintering temperature. The dielectric properties and ac conductivity were discussed in the light of space charge polarization and the electron hoping between the adjacent sites. The dielectric constant and loss tangent of MZMFO ferrite is lower than MZFO ferrite due to the unavailability of ferrous and ferric ions in the octahedral sites. At 1200 and 1250 °C MZFO ferrite exhibits dielectric loss peaks and MZMFO ferrite exhibits at 1200 °C. The loss peaks were appeared due to matching of the hopping frequency of electrons and the frequency of the applied field. The initial permeability of MZMFO ferrite at 1200 °C and 1250 °C is lower than that of MZFO ferrite because Mg^{2+} ion caused an increase of the magnetocrystalline anisotropy and the saturation magnetization. The increment of magnetocrystalline anisotropy is larger than that of saturation magnetization.

1. Introduction

Recently, special focus has been placed on the preparation and characterization of high performance Mn–Zn ferrite since it exhibits some unique properties such as enhanced coercivity, modified saturation magnetization, super paramagnetism, metastable cation distributions, high permeability, high resistivity, and large saturation magnetic flux density and as well as its potential applications in high frequency and low power miniaturized electronic devices such as recording heads, choke coils and electromagnetic wave absorption materials etc [1–5]. $MnFe_2O_4$ exhibits partially inverse spinels structure because manganese and ferric ions occupying both A and B-sites [6–8]. On the other hand, zinc ferrite; $ZnFe_2O_4$ has normal spinel structure in which Zn^{2+} preferably occupies the A-sites due to their affinity for sp^3 bonding with oxygen anions leaving all the ferric ions on the B-sites [9]. The normal spinel structured $ZnFe_2O_4$ is paramagnetic at room temperature due to weak superexchange interaction which could be attributed to right angle in Fe^{3+}–O–Fe^{3+} [10]. In addition, the magnetic and electrical properties of ferrites strongly depend on their chemical composition, preparation method, grain size and cation distribution between two interstitial sites [11,12]. It is reported that the structural, electrical and magnetic properties of spinel ferrite can be controlled through the substitution of nonmagnetic ions [13–15]. Recently, magnesium substituted ferrite has been emerged as new frontier of research to improve the electrical and magnetic properties of ferrite. Magnesium ferrite ($MgFe_2O_4$) is inverse spinel ferrite where Mg^{2+} cations have strong preference mainly on octahedral sites [16–18]. In the recent years, a number of investigations have been reported for studying the effect of the substitution of Mg^{2+} ions on structural, electrical and magnetic properties of spinel ferrites such as $Ni_{0.50}Zn_{0.50-x}Mg_xFe_2O_4$ [19], $Ni_{0.5}Cu_{0.5-x}Mg_xFe_2O_4$ [20], $Mn_{0.4}Zn_{0.6}Mg_xFe_{2-x}O_4$ [21], $Mn_xMg_{0.5-x}Zn_{0.5}Fe_2O_4$ [22], $Ni_{0.5-x}Zn_{0.5}Mg_xFe_2O_4$ [23], $Ni_{0.8-x}Zn_{0.2}Mg_xFe_2O_4$ [24], $Cu_{0.5}Zn_{0.5-x}Mg_xFe_2O_4$ [25], $Cu_{0.5-x}Mg_xZn_{0.5}Fe_2O_4$ [26–29], $Ni_{0.35}Mg_xCu_{0.05}Zn_{0.6-x}Fe_2O_4$ [30], $Ni_{0.25-x}Mg_xCu_{0.2}Zn_{0.55}Fe_2O_4$ [31], $Mg_{0.3}Cu_{0.2}Zn_{0.52}Fe_{1.98}O_{3.99}$ [32], $Ni_{0.5-x}Cu_{0.2}Zn_{0.3}Mg_xFe_2O_4$ [33], $Ni_{0.25-x}Mg_xCu_{0.20}Zn_{0.55}Fe_2O_4$ [34] and $Ni_{0.25-x}Mg_xCu_{0.3}Zn_{0.45}Fe_2O_4$ [35–38]. To date, there is no report available in literature on sintering temperature dependent structural, electrical and magnetic properties of $Mn_{0.5}Zn_{0.5-x}Mg_xFe_2O_4$ (x=0.0, 0.1) ferrites. Based on

* Corresponding author. Fax: +880286136.
E-mail address: dalilurrahaman1976@gmail.com (Md.D. Rahaman).

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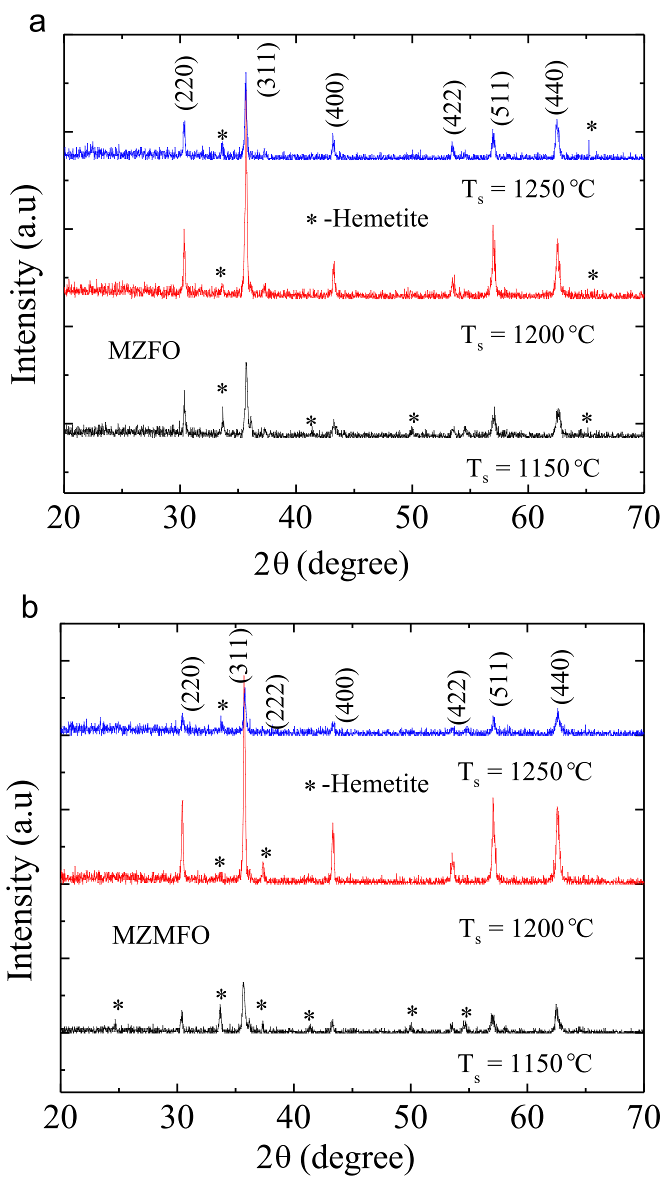

Fig. 1. Typical X-ray diffraction (XRD) patterns of (a) $Mn_{0.5}Zn_{0.5}Fe_2O_4$ (MZFO) and (b) $Mn_{0.5}Zn_{0.4}Mg_{0.1}Fe_2O_4$ (MZMFO) ferrites sintered at 1150–1250 °C for 5 h.

the abovementioned issues, our main aim is to synthesize $Mn_{0.5}Zn_{0.5}Fe_2O_4$ (MZFO) and $Mn_{0.5}Zn_{0.4}Mg_{0.1}Fe_2O_4$ (MZMFO) ferrites at different sintering temperatures. In the present work, effect of sintering temperature on structural, microstructural, elastic and electromagnetic properties of MZFO and MZMFO ferrites has been reported.

2. Experimental details

2.1. Preparation method

The MnZn and MnZnMg ferrite samples having the chemical formula $Mn_{0.5}Zn_{0.5}Fe_2O_4$ (MZFO) and $Mn_{0.5}Zn_{0.4}Mg_{0.1}Fe_2O_4$ (MZMFO) were prepared by solid state reaction method. The raw materials $MnCO_3$, ZnO, MgO and Fe_2O_3 (with purity 99.9%) were weighed in stiochiometric proportions; the powders were grounded with agate mortar and pestle for 4 h, after the powder samples were calcined at 1100 °C for 5 h in air atmosphere using programmable muffle furnace with a heating rate of 10 °C min^{-1} and cooling rate of 5 °C min^{-1}. Then, the powders were again grounded with small amount of polyvinyl alcohol (PVA) using as a binder. The powder was palletized into small disks and toroids using with a uniaxial pressure of 34.5 MPa and sintered at 1150, 1200, and 1250 °C for 5 h. The heating and cooling rate were similar as the calcination process.

2.2. Measurement and characterizations

Structural study of sintered MZFO and MZMFO ferrites prepared by the conventional solid state reaction method were performed by Philips PanAnalytic Xpert Pro X-ray diffractometer with a Cu anode (Cu-Kα radiation source with λ=1.541 Å) operated at 40 kV and 30 mA. The average grain size and morphology of the samples were observed by a SEM (JEOL-instrument JEOL JSM-6460, Japan). The grain size of the samples was determined by the linear intercept technique [39]. The structural parameters (Crystallite sizes (D_{311}), lattice constants, X-rat density (ρ_x), bulk density (ρ_B) and porosity (P) were calculated according to Hossain et al. [19]. The electromagnetic properties such as initial permeability, dielectric constant and dissipation factor were measured with a Wayne Kerr Impedance Analyzer (B 6500 series). Initial complex permeability ($\mu_i^* = \mu_i' - j\mu_i''$) was calculated using the following relations: $\mu'_i = (2\pi L_s/\mu_0 N^2 h)\ln(r_{outer}/r_{inner})$ and $\mu''_i = \mu'_i \tan\delta$, where L_s is inductance, N is the number of turns of copper wire on torroid, h is height of the torroid in meters, r_{outer} is outer diameter of torroid in meters, r_{inner} is the inner diameter of torroid in meters, and tan δ is the magnetic loss [40].

3. Results and discussion

3.1. Structural analysis

Typical XRD patterns of MZFO and MZMFO ferrites sintered at 1150, 1200, and 1250 °C are depicted in Fig. 1(a) and (b) respectively. The peaks could be indexed as (111), (220), (311), (222), (400), (411), (511) and (422), which are characteristics of cubic spinel structure with impurity phase of hematite [41]. The lattice parameter was calculated using the relation,

Table 1
Values of lattice constant (a), bulk density (ρ_B), X-ray density (ρ_x), porosity (P), Grain size, crystallite size (D_{311}) of the $Mn_{0.5}Zn_{0.5}Fe_2O_4$ and $Mn_{0.5}Zn_{0.4}Mg_{0.1}Fe_2O_4$ ferrites sintered at different sintering temperatures (T_s).

Compositions	Sintering temperature (°C)	a (Å)	ρ_B (g/cm^3)	ρ_x (g/cm^3)	P (%)	Grain size (μm)	D_{311} (nm)
$Mn_{0.5}Zn_{0.5}Fe_2O_4$ (MZFO)	1150	8.3383	4.87	5.40	10	2.10	40
	1200	8.3474	4.76	5.39	12	2.84	41
	1250	8.3496	4.45	5.38	17	3.13	47
$Mn_{0.5}Zn_{0.4}Mg_{0.1}Fe_2O_4$ (MZMFO)	1150	8.3542	4.61	5.28	13	2.00	37
	1200	8.3361	4.38	5.31	18	3.00	45
	1250	8.3225	4.57	5.34	14	3.10	38