



Structural and magnetic properties of manganese zinc ferrite nanoparticles prepared by solution combustion method using mixture of fuels



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ABSTRACT

The structural analysis and magnetic investigation $Mn_{1-x}Zn_xFe_2O_4$ with stoichiometry ($x=0, 0.1, 0.3, 0.5, 0.7, 0.9$ and 1.0) were synthesized by solution combustion method using mixture of fuel this is first of its kind. As synthesized Mn–Zn nanoferrites were characterized by X-ray Diffractometer (XRD), Transmission electron microscopy (TEM) at room temperature. The magnetic domain relaxation was investigated by inductance spectroscopy (IS) and the observed magnetic domain relaxation frequency (f_r) was increased with the increase in grain size. The Room temperature magnetic properties were studied using vibrating sample magnetometer (VSM). It was observed that the real and imaginary part of permeability (μ' and μ''), saturation magnetization (M_s), remanance magnetization (M_r) and magneton number (M_r/M_s) were decreases gradually with increasing Zn^{2+} concentration. The decrease in the saturation magnetization may be explained as, the Zn^{2+} concentration increases the relative number of ferric ions on the A sites diminishes and this reduces the A–B interaction. Hence synthesized materials are good for high frequency applications.

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1. Introduction

Ferrimagnetic materials mainly composed of iron oxide called ferrites. Ferrites having a spinel cubic structure and the general formula is AB_2O_4 , where A is a divalent metal ion (Mn^{2+} , Zn^{2+} , Ni^{2+} , Cu^{2+} , Mg^{2+}) and B is the trivalent metal ions (Fe^{3+} , Cr^{3+} , Sc^{3+} , Sm^{3+} , Gd^{3+}). The spinel configuration is based on a face centered cubic (fcc) lattice of oxygen ions, forming tetrahedral (A) and octahedral (B) sites that may be occupied either by A and/or B site. In normal, the transition metal ion having valency $2+$ occupy the tetrahedral sites and transition or rare earth ions having valency $3+$ occupy the octahedral sites. If divalent metal ions occupy the octahedral sites and trivalent ions are distributed among tetra and octahedral sites. Based on the occupancy of the metal ions the ferrite materials exhibit excellent properties such as structural, electrical and magnetic properties. Among all the ferrites Mn–Zn ferrites possess excellent properties because they having high electrical resistivity, high saturation magnetization, high permeability and low power loss hence they are attracted to high frequency applications [1–3]. The Mn–Zn ferrites are

magnetic oxide materials with semiconducting nature which are of great technological importance. The general important application of Mn–Zn ferrite materials used in transformer cores, antenna rods, memory chips, high density magnetic recording media, permanent magnets, transducers, activators microwave and computer technology etc. [4–6]. Furthermore, researchers using several methods to synthesis nanoferrites such as ball milling method [7], co precipitation method [8], hydrothermal method [9] and auto combustion method [10,11]. However Among these methods, the solution combustion method is a facile approach with great economic and technical advantages to obtain highly crystalline nanoparticles [7]. In the present investigation we adopt solution combustion method to synthesize Mn–Zn nanoferrite particles using urea and glucose as a mixed fuels. The first time Mn–Zn ferrites were synthesized solution combustion method using mixture of fuel. Furthermore, the substitution of Zn^{2+} at A-site shows decrees in structural as well as magnetic properties in $MnFe_2O_4$. This motivates us to synthesize the Zn^{2+} substituted $MnFe_2O_4$ nanoparticles and detailed investigation on structural and magnetic properties.

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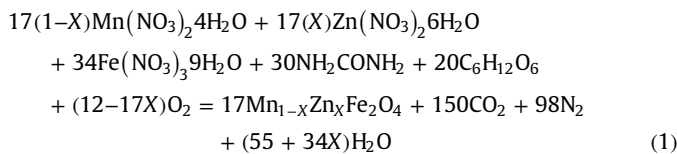
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2. Experimental

The nanocrystalline $Mn_{(1-x)}Zn_xFe_2O_4$ ($x=0, 0.1, 0.3, 0.5, 0.7, 0.9$ and 1.0) were prepared by solution combustion using urea and glucose as a fuel. The stoichiometric molar amounts of manganese nitrate [$Mn(NO_3)_2 \cdot 4H_2O$], zinc nitrate [$Zn(NO_3)_2 \cdot 6H_2O$], and iron nitrate [$Fe(NO_3)_3 \cdot 9H_2O$] as oxidizer and mixture of urea [NH_2CONH_2] and glucose [$C_6H_{12}O_6$] as fuels. The stoichiometric compositions of metal nitrates and fuels were evaluated based on the total oxidizing and reducing valences of the components and the oxidizer to fuel ratio was taken as 1: (60:40). All the metal nitrates and fuels were diluted with 30 ml double distilled water and thoroughly mixed with the help of magnetic stirrer with the speed of 800 rpm for one hour until the reactants were dissolved completely to get homogenous solution. This homogeneous solution containing redox mixture was taken in a Pyrex dish and kept in a pre-heated muffle furnace maintained at 450 ± 10 °C.

In the combustion synthesis, besides the target product (Mn–Zn ferrite), gases in the most stable form, i.e., CO_2 , H_2O , and N_2 , are produced as gas is being combusted. Carbon and hydrogen with a valence of $4+$ and $1+$, respectively, are regarded as reducing agents, oxygen with a valence of $2-$ is regarded as an oxidizing agent and the valence of nitrogen amounts to zero. The stoichiometric redox reaction for the systems in which $O/F=1$ according to the proposed equations:

Fuel: Mixture of fuel (60:40) urea [NH_2CONH_2] and glucose [$C_6H_{12}O_6$]



Initially, the solution boils then froths and ignites to yield fine powder of Mn–Zn rapidly because these are exothermic. The whole combustion process was complete in less than 20 min, whereas the reaction time of the actual ignition was less than 5 s. The formation of Mn–Zn ferrite was possible due to the gas phase reaction between decomposition products of metal nitrates (nitrogen oxides) and fuels. The obtained powder was pressed in to the cylindrical pellets by applying the pressure of 5-ton/cm² for 5 min and sintered at 1000 °C/2 h in muffle furnace.

The sintered pellet were characterized by X-ray Diffractometer with CuK_α radiation ($\lambda=0.154056$ nm). Transition electron microscopy (TEM) and magnetization was carried out by vibrating sample magnetometer (VSM) mounted on an electromagnet with a bipolar source of maximum applied field of 1.5 T at room temperature.

3. Results and discussion

3.1. Structural analysis

Fig. 1 shows the room temperature X-ray diffraction patterns of all the samples of $Mn_{1-x}Zn_xFe_2O_4$ nanoferrites and it confirms the spinel cubic structure with space group $Fd\bar{3}m$ (Oh7) for all the composition. The broad diffraction peaks indicates that the ferrite particles are of Nano-sized. It is also observed that the presences of secondary phases in the XRD pattern of some compositions (M3, M4, M5, and M6) of the 2θ is around 34° is identified as $\alpha-Fe_2O_3$ and the amount of this phase is found to be very small and it has

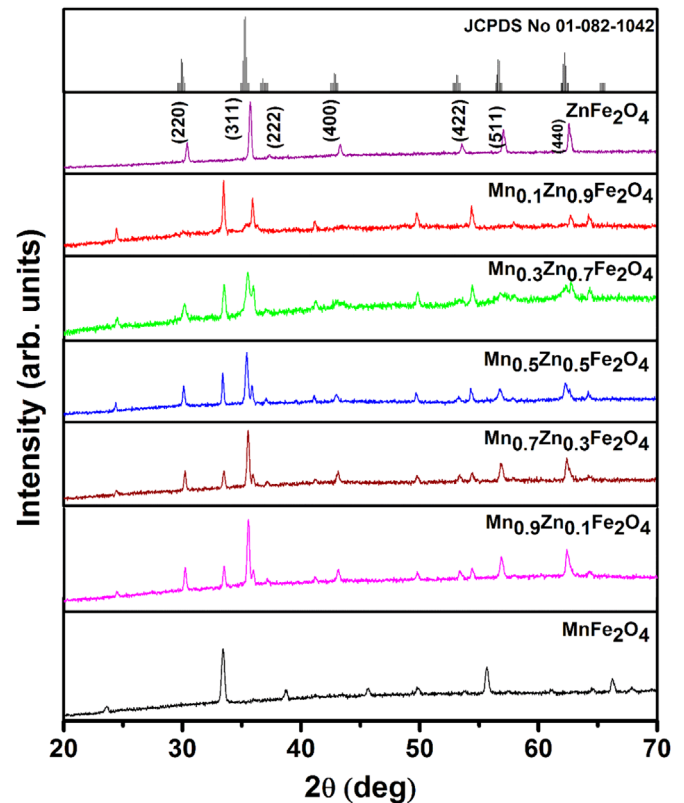


Fig. 1. X-ray diffraction patterns of $Mn_{1-x}Zn_xFe_2O_4$ ($x=0, 0.1, 0.3, 0.5, 0.7, 0.9$ and 1.0) sintered at 1273 K at 2 h.

no effect on the electrical and magnetic properties of present ferrites [12]. The average crystallite size of the each composition was calculated from the line width of the (3 1 1) peak of XRD pattern using the Scherrer formula [12].

$$t = \frac{0.9\lambda}{\beta \cos \theta} \text{ nm} \quad (2)$$

where 't' is crystallite size, ' λ ' is wavelength of X-ray radiation, ' θ ' is Bragg's angle, is full width at halfmaximum. The structural parameters such as average crystallite size, cell volume and lattice parameter are varies with Zn^{2+} concentration increases. The average particle size of all the composition varies in the range of 25–35 nm. The lattice parameter and cell volume is decrease with increasing Zn^{2+} content. The decrease of lattice parameter and cell volume is due to the larger ionic radius Mn^{2+} ions (~ 0.091 nm) replaced by smaller ionic radius of Zn^{2+} ions (~ 0.082 nm) [13]. The value of lattice parameter and cell volume is listed in Table 1.

The distances between the magnetic ions at tetrahedral (A) and octahedral (B) sites were calculated by using the following equation.

$$L_A = a \frac{\sqrt{3}}{4} \text{ and } L_B = a \frac{\sqrt{3}}{2} \quad (3)$$

The variation of hopping length of tetrahedral site (L_A) and octahedral site (L_B) is as shown in the Fig. 2(a) and (b). Both the hopping length of L_A and L_B decreases with increasing Zn concentration. This may be cause of due to the decreases in lattice parameter of all the samples of increasing Zn^{2+} concentration.

3.2. Transition electron microscopy

Fig. 3(a) and (b) illustrates the TEM images of samples of composition $x=0.0, 0.3$. The particle sizes were analyzed using

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