

# Comparative Study of Structural and Magnetic Properties of $Mg_{1-x}Zn_xFe_2O_4$ Prepared by Ceramic and Sol-Gel Auto Combustion

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**Abstract:** The samples of  $Mg_{1-x}Zn_xFe_2O_4$  spinel ferrites with ( $x = 0.0, 0.50$  and  $1.00$ ) were prepared by ceramic and sol-gel auto combustion method. X-ray diffraction (XRD) technique was used to confirm the single phase formation of samples. The crystallite size ( $t$ ) was determined using Scherrer's formula and found to be  $4 \mu m$  and  $38 nm$  for ceramic and sol-gel prepared samples respectively. The lattice constant, unit cell volume, X-ray density of all these structural parameters are obtained from XRD data, shows the strong effect of particle size. The magnetic properties were investigated using pulse field hysteresis loop technique at room temperature. The saturation magnetization ( $M_s$ ) and coercivity ( $H_c$ ) increases with increase in particle size. Thus, the particle size of the Mg-Zn sample strongly influences the structural and magnetic properties of  $Mg_{1-x}Zn_xFe_2O_4$ . With the zinc substitution the lattice constant, X-ray density increases whereas magnetic properties are decreased.

**Keywords:** Nanocrystalline, Magnesium-Zinc, Magnetization, Spinel Ferrite.

## I. INTRODUCTION

Among the several magnetic oxide materials, ferrites have recognized as a best candidate for application in many fields due to the combined electrical and magnetic properties and have been extensively studied for their structural, electrical, dielectric and magnetic Properties by many workers [1-3]. The high electrical resistivity, low eddy current and dielectric loss, high saturation magnetization, high Curie temperature, high permeability, chemically stable, low cost etc. are the prime characteristics of ferrite material. These important properties of ferrite can be modified so as to suit the desired applications. One of the best ways of modifying the properties of ferrite is the method of preparation. The method of preparation plays an important role in governing the properties of ferrite. The difference in magnetic properties is due to particles size effect [4, 5].

The saturation magnetization ( $M_s$ ), Coercivity ( $H_c$ ) and remanence magnetization values are different for ceramically and Sol-gel prepared samples but are in reported range. These important properties of ferrite can be modified so as to suit the desired applications. The method of preparation plays an important role in governing the properties of ferrites. It is reported that the properties of ferrite material prepared by two technique show different properties due to particle size effect [4, 5]. Usually ferrites are prepared by standard ceramic method which produces particles of large size usually of the order of more than  $100 \text{ \AA}$ . In the recent years ferrites are commonly synthesized by wet chemical method in order to prepare nano size particles [6, 7]. The nanosize spinel ferrite is gaining importance due to their interesting and superior proprieties that are different from their bulk counterpart [8,9].

In the literature it is reported that Magnesium ferrite with zinc substitution has been prepared by ceramic technique [10, 11]. However, Mg-Zn spinel ferrite in nanosize form has not been prepared by sol-gel method to our best knowledge. To understand the effect of particle size on the various properties of spinel ferrite, it is necessary to synthesize the same spinel ferrite by two different methods which can produce particles of different sizes. According to literature magnesium ferrite is a partially inverse spinel ferrite whose degree of inversion depends on heat treatment. [12, 13]. Zinc ( $Zn^{2+}$ ) ion when substituted in spinel ferrite may gives rise to various magnetic structures [14, 15].

To understand the effect of particle size on the structural and magnetic properties Mg-Zn spinel ferrite, attempt have been made to synthesize the Mg-Zn spinel ferrite with general formula  $Mg_{1-x}Zn_xFe_2O_4$  with  $x = 0.00, 0.50$  and  $1.0$  by ceramic, sol-gel auto combustion method and obtained properties were compared to understand the effect of particle size.

## II. EXPERIMENTAL

### Preparation of $Mg_{1-x}Zn_xFe_2O_4$ ( $x = 0.0, 0.50$ and $1.00$ ) spinel ferrite nanoparticles

The samples of  $Mg_{1-x}Zn_xFe_2O_4$  spinel ferrites were prepared by the standard ceramic technique for  $x = 0.00, 0.50$  and  $1.00$ . The starting materials were analytical reagent grade oxide  $Fe_2O_3$ ,  $MgO$  and  $ZnO$ . These oxides were mixed in stoichiometric proportion and wet-ground for 4 h and presintered at  $900^\circ C$  for 12 h. In the final sintering process, the material was held at  $1050^\circ C$  for 24 h and slowly cooled to room temperature ( $2^\circ/min$ ).

Analytical grade (AR) chemicals such as magnesium nitrate ( $Mg(NO_3)_2 \cdot 6H_2O$ ), zinc nitrate ( $Zn(NO_3)_2 \cdot 6H_2O$ ), ferric nitrate ( $Fe(NO_3)_3 \cdot 9H_2O$ ) and citric acid ( $C_6H_8O_7$ ) were used for the synthesis. All the chemicals used for the synthesis were analytical grade. Citric acid ( $C_6H_8O_7$ ) was used as a fuel. The metal nitrates to fuel (citric acid) ratio was taken as 1:3. Ammonia solution was added to maintain the pH 7. The temperature required for the synthesis of Mg-Zn ferrite nanoparticles was low that is around 110 °C. The as-synthesized powder is sintered at 700 °C for 5 h and then used for further investigations.

**Characterizations**

The X-ray diffraction (XRD) patterns for all the samples were recognized on ‘Philips X-ray diffractometer (PW 3710). The magnetization measurements were carried out using high pulse field hysteresis loop technique at 300 K.

**III RESULTS AND DISCUSSIONS**

Fig .1 represents typical X-ray diffraction (XRD) pattern of bulk and nano size Mg-Zn spinel ferrite with x = 1.00. The X-ray diffraction pattern shows the reflections (2 2 0), (3 1 1), (4 0 0), (4 2 2), (5 1 1), (4 4 0), (5 3 3) which belongs to cubic spinel structure.

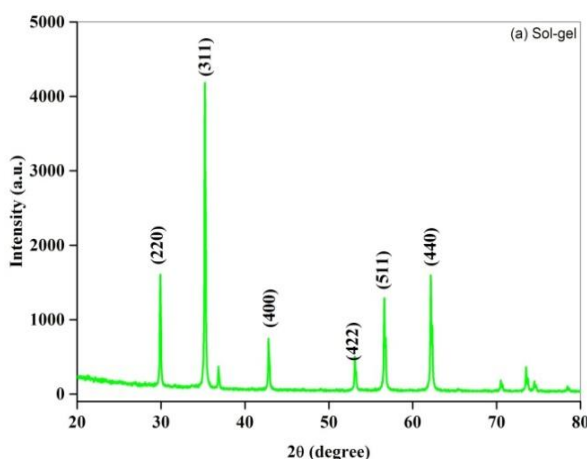
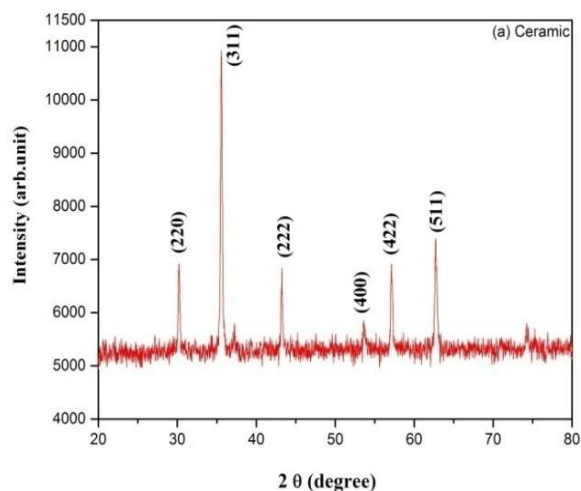


Fig.1 (a): Typical XRD patterns of  $Mg_{1-x}Zn_xFe_2O_4$  (for x = 1.00) spinel ferrite for (a) ceramic (b) sol-gel

The analysis of XRD pattern reviews the formation of single phase cubic spinel structure. A careful examination of the XRD pattern reveals that the bulk sample shows intense sharp peaks whereas nano size sample shows broad reflections may be due to particle size effect.

The values of lattice constant were determined using XRD data and are summarized in Table.1. It is seen from Table 1 that the lattice constant increases with increasing zinc substitution both the samples. The increase in lattice constant is attributed to the difference in ionic radii of  $Mg^{2+}$  and  $Zn^{2+}$ . In the present case  $Mg^{2+}$  ions with ionic radii 0.65 Å were replaced by  $Zn^{2+}$  ions having ionic radii 0.75 Å.

Therefore, the lattice constant of the present system increases with increase in Zn substitution. The comparison of lattice constant of the bulk and nano size spinel ferrite shows that the lattice constant of the nano size Mg-Zn spinel ferrite is greater than that of the bulk Mg-Zn spinel ferrite. The crystallite size (t)/particle size of all the samples were calculated using Scherrer’s formula taking into consideration the most intense peak (311) of the XRD pattern. It is found that the crystallite size for nano size ferrite is in nano meter range whereas for bulk sample it is in Å. The difference in particle size is attributed to method of preparation. The sol-gel method produces particle of nanometer dimension whereas ceramic method produces particle of coarse size, the value of particle size are summarized in Table 2.

The X-ray density ( $d_x$ ) of the samples were determined using the relation;

$$d_x = \frac{8M}{N a^3}$$

Where 'M' is molecular weight, 'a' is lattice constant and 'N' is Avogadro’s number. The values of  $d_x$  are given in Table 3.

The X-ray density of all the samples increases with Zn substitution and is attributed to the increase in lattice constant of the system. The X-ray density of nano size sample is greater than that of bulk sample. The magnetic properties were studied through pulse field hysteresis loop technique.

The magnetization measurements were carried out at room temperature with an applied magnetic field of 10 kOe. The magnetic hysteresis curve M-H plot for bulk and nano sized Mg-Zn spinel ferrite (shown in Fig. 2) which indicates that the values of saturation magnetization ( $M_s$ ), coercivity ( $H_c$ ) and remanence magnetization ( $M_r$ ).

The values of these magnetic properties were studied through pulse field hysteresis loop technique. The magnetization measurements were carried out at room temperature with an applied magnetic field of 10 kOe. Magnetic parameters for both bulk and nano size Mg-Zn Spinel ferrite are given in Table 4.

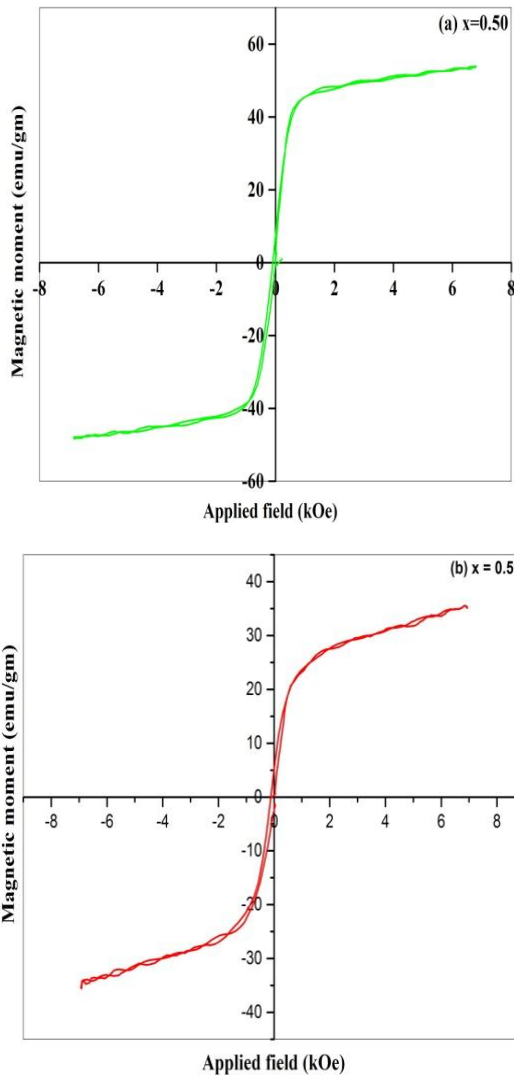


Fig.2 Hysteresis loops of the system  $Mg_{1-x}Zn_xFe_2O_4$  for  $x = 0.50$  (a) sol-gel method (b) ceramic method

It is evident from Table 4 that as Zn increases the magnetization and other magnetic properties decreases.

Table 4: Magnetic parameters for bulk spinel ferrite for the system  $Mg_{1-x}Zn_x Fe_2O_4$

[	Ceramic			Sol-gel		
	Ms (emu/gm)	Mr (emu/gm)	Hc (Oe)	Ms (emu/gm)	Mr (emu/gm)	Hc (Oe)
0.0	58.71	2.192	37.33	41.66	1.58	132.96
0.5	50.43	1.729	58.68	28.43	0.35	154.25
1.0	25.20	0.942	69.70	7.17	0.01	222.64

#### IV CONCLUSIONS

Zinc substituted magnesium ferrite sample with chemical formula,  $Mg_{1-x}Zn_xFe_2O_4$  ( $x = 0.00, 0.50$  and  $1.00$ ) were synthesized successively by ceramic method and sol-gel auto combustion method to obtain the particles of different size. The sol gel method produces nano size particle, X-ray diffraction technique confirms the formation of single phase spinel phase samples. The lattice constant found to increase with increasing Zn substitution  $x$ .

This is due to the fact that the Zn is a non-magnetic ion and occupies tetrahedral A-site. The comparison of magnetic data suggests that the values of  $M_s$ ,  $M_r$  and  $H_c$  are greater than the reported values for Mg-Zn bulk ferrite. The discrepancy in the magnetic parameters of nano size and bulk Mg-Zn spinel ferrite is due to the particle size effect. Mg-Zn spinel ferrite prepared by sol-gel auto combustion method shows better properties as compared to their bulk Mg-Zn spinel ferrite. Similar results of magnetic properties are reported in the literature.

Table 1: lattice constant (a), Particle size for sol-gel and Ceramic method for the system  $Mg_{1-x}Zn_xFe_2O_4$

x	a (Å) (sol-gel)	a (Å) (ceramic)	Sol-gel method	Ceramic
			t (nm)	t (µm)
0.00	8.355	8.366	38	4.1
0.50	8.371	8.414	33	4.3
1.00	8.385	8.441	37	3.9

Table 2: The values of  $d_x$ ,  $d_B$  and percentage Porosity  $p$  the system  $Mg_{1-x}Zn_xFe_2O_4$  for sol-gel method

x	$d_B$ (gm/cm <sup>3</sup> )	$d_x$ (gm/cm <sup>3</sup> )	P%
0.00	2.832	4.553	37.80
0.50	2.882	4.505	36.02
1.00	2.936	4.446	33.96

Table 3: The values of Bulk density ( $d_B$ ), X-ray density ( $d_x$ ) and porosity (P) for  $Mg_{1-x}Zn_xFe_2O_4$  ( $0 \leq x \leq 1.00$ ) for ceramic method

x	$d_B$ (gm/cm <sup>3</sup> )	$d_x$ (gm/cm <sup>3</sup> )	P (%)
0.00	3.275	4.539	27.77
0.50	3.260	4.919	34.56
1.00	3.245	5.332	38.57

The porosity increases with increasing Zn substitution for ceramic sample whereas it decreases for sol-gel prepared samples. Saturation magnetization and coercivity are found to be greater for sol-gel prepared samples as compared to ceramically prepared samples. The structural and magnetic properties were strongly influenced by method of preparation thereby particle size.

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