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Effect of Gamma Irradiation on the Structure Diffusion Coefficient of Co-Zn Doped Ferrite

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Abstract: A series of the ferrite system $Co_{0.6}Zn_{0.4}Mn_xFe_{2-x}O_4$ with (x = 0.7, 0.8, 0.9 and 1) were prepared by general ceramic method. The X-ray diffraction patterns confirmed the formation of the cubic single phase structure. The lattice parameter and the crystal size increased after irradiation which was attributed to the formation of ferrous ions of larger radius than that of ferrous ions in unirradiated samples. A small shift was observed in the diffraction peak position. This indicated the slight distortion of the cubic structure. The diffusion coefficient of electrons was found to be increased after γ irradiated samples. The concentration of electrons was estimated and was found to be increased after γ irradiation.

Key words: Ferrimagnetic, Diffusion Coefficient, X-ray Intensity

INTRODUCTION

Spinal ferrite is one of the most important classes of magnetic ceramic materials owing to their interesting applications. In the spinel structure the magnetic ions are distributed between two different lattice sites tetrahedral (A) and octahedral (B) sites. The magnetic as well as electrical properties of these ferrite depend on the relative distribution of cations at the different sites as well as the preparation conditions^[1]. In ferromagnetic state the hopping electrons are oriented in one direction (the direction of the electric field), which helps the electrons to jump from ferric to ferrous ions without scattering^[2, 3].

The diffusion coefficient of oxygen vacancies was estimated from DC conductivity measurements of $Co_{1-x}Zn_xMn_xFe_2O_3$. It was noticed that the diffusion coefficient increased after gamma irradiation for all Zn^{2+} concentrations^[4, 5]. This could be explained on the basis of the displacement of metal ion from its original sites under the effect of radiation. It was reported that the lattice parameter increased due to the formation of ferrous ions under the ionizing effect of radiation^[6].

The aim of the present work was carried out in an attempt to though light on the effect of γ -irradiation on the crystal structure, lattice parameter and the diffusion coefficient of electrons in *Co-Zn* ferrite doped with Mn. It was expected that these compositions could be used as a sensor for detecting environmental pollution.

MATERIALS AND METHODS

The pure oxides of CoO, ZnO, Fe_2O_3 and Mn_2O_3 were mixed together to take the required chemical formula^[7-9] Co_{0.6}Zn_{0.4}Mn_xFe_{2-x}O₄ (x=0. 7, 0.8, 0.9 and 1) using an agate mortar for ten hours. The powder was fired for 4 hr at 900°C. The powder was again grounded and pressed into tablets of one cm diameter and thickness 0.2 cm. The tablets were sintered at 1200°C for 2 hr. The furnace was left to be cooled slowly to the room temperature. The formation of spinel cubic phase was confirmed by X-ray diffraction patterns using X-ray diffratometer type Philips, PW 1710. The samples were irradiated by $Co^{60} \gamma$ -radiation of dose 10^6 rad hr⁻¹. The γ -irradiation was carried out at the center of the Eastern Regional radioisotope, Dokki, Cairo. After 2 days, the d.c conductivity was measured in the temperature range from room temperature to 310° C using an electrometer type Keithley 610 C.

RESULTS AND DISCUSSION

X-ray Diffraction Analysis: Figure 1a and b represent the *X*-ray diffraction patterns for samples $Co_{0.6}Zn_{0.4}Mn_xFe_{2-x}O_4$, where x=0. 7, 0.8, 0.9 and 1 before and after irradiation. The *X*-ray analysis confirmed the formation of spinel cubic phase structure. It was noticed that a slight shift of the reflected peaks was occurring in the irradiated sample. This was attributed to the distortion occurred in the cubic lattice after irradiation^[4, 5].

Lattice Parameter: Figure 2 shows the variation of the lattice parameter of the different compositions before and after irradiation. It was noticed that the lattice parameter increased with enhancing Mn concentration, this could be attributed to the substitution of Mn^{3+} of large size (r = 0.07 nm) for Fe³⁺ (r = 0.067 nm) of smaller size. The increase of lattice parameter for the irradiated samples than that of unirradiated was attributed to ionizing the ferric ions (r = 0.067 nm) of smaller size to ferrous (r = 0.08 nm) of larger size after irradiation. Our explanation was similar to that in previous work on different ferrite composition^[4-6, 9].



Fig. 1: The X-ray Diffraction Patterns for the Samples x = 0.6, 0.7, 0.8, 0.9 and (a) before Irradiation, (b) After Irradiation



Fig. 2: The Lattice Parameter of the Composition $Co_{0.6}Zn_{0.4}Mn_xFe_{2-x}O_4$ before and After Irradiation



Fig. 3: The X-ray Intensity Ratios before Irradiation

Intensity Ratios: Figure 3 illustrates peak intensity ratios I_{511}/I_{400} and I_{422}/I_{400} before and after irradiation which are very sensitive to the structure changes. It was noticed that the intensity ratios were sensitive to manganese content. The increase of intensity ratio I_{422}/I_{400} and I_{511}/I_{400} before irradiation was attributed to the replacement of Mn³ of larger radius for Fe³ion of smaller radius^[8, 10]. This led to increasing the intensity ratio. For irradiated samples, the observed decrease of the intensity ratio I_{422}/I_{400} could be explained by the slight distortion of the crystal lattice leading to decrease the reflected intensity. It was reported that the γ -rays irradiation caused a slight distortion the lattice structure of ferrites^[5].

In conclusion the distorted spinel cubic structure after γ -radiation decreases the intensity ratio, which is the fundamental role in sensor applications.

The Diffusion of Electrons Before and After Irradiation: The diffusion coefficient D of electrons was estimated from the relation $D = \frac{\sigma K_B T}{Ne^2}$ where, σ is the electrical conductivity, N (atom concentration) equals $4x10^{28}$ atom m^{-3[11]}, e = electronic charge, K_B is Boltzmann constant and T is the absolute room temperature.

	Х	0.7	0.8	0.9	1
After irra.	$D_{o} (m^{2} s^{-1})$	1.54x10 ⁻⁸	1.79 x10 ⁻⁸	1.67 x10 ⁻⁷	4.36 x10 ⁻⁷
Before irra.	$D_{o}(s^{-3})$	1.59 x10 ⁻⁹	2.26 x10 ⁻⁹	3.06 x10 ⁻⁹	4.28 x10 ⁻⁹
Before	E(ev)	0.566	0.592	0.635	0.633
After	E(ev)	0.498	0.476	0.475	0.456

Table 1: The Calculated Activation Energy of Diffusion Process



Fig. 4a,b: The Diffusion Coefficient of Oxygen Vacancies in the Given Ferrite before and After Irradiation

The diffusion coefficient of electrons for Co_{0.6}Zn_{0.4}Mn_xFe_{2-x}O₄ before and after irradiation as a function of 1000/T, are illustrated in Fig. 4a and b. It is observed that the diffusion coefficient increased by rising temperatures. This might be explained on the bases of the hopping conduction mechanism between ferric and ferrous ions^[2, 3]. The increase of temperature activated the hopping rate of electrons, $Fe^{3+} \rightleftharpoons Fe^{2+}$ leading to rising the diffusion coefficient through the lattice for samples before and after irradiation. Our explanation is similar to that in previous work on other compositions of ferrite^[2].

The increase of the diffusion coefficient with increasing manganese contents might be attributed to the presence of Mn^{2+} and Fe^{3+} at *A* site according to the cation distribution given in previous work^[4, 8]. They

stated that 20% of Mn^{3+} are present at *B* site and 80% of Mn^{2+} are present at *A* site.

The increase of Mn^{2+} content at *A* site gave rise to the hopping rate of electrons between Mn^{2+} and Fe³⁺, leading to increase the diffusion coefficient for an irradiated and unirradiated samples.

It is observed that the diffusion coefficient decreased after γ -irradiation this could be explained as following: when the samples were exposed to γ irradiation some of manganese ions migrate from A sites to B sites and the percentage of 80% of manganese ions decreased leading to reducing the hopping rate of electrons between Mn²⁺ and Fe³⁺. This mechanism decreased the diffusion coefficient of electrons because our samples were n type^[8, 9]. The dependence of the diffusion coefficient of temperature can be given by the equation $D = D_{e}e^{-E/K_{B}T}$ where D is the pre exponential factor and E is the activation energy for the diffusion process^[12]. The results of D₂ before and after irradiation and the activation energies of diffusion processes are given in Table 1.

The values of E after irradiation is lower than that before irradiation, which means that, the diffusion process of electrons through structural vacancies or thermal equilibrium vacancies increased. The diffusion coefficient increased after γ -radiation in ferrite as in previous work^[4] which confirmed the behavior of our results.

CONCLUSION

- * γ-radiation ionized the ferric ions of smaller size to ferrous ions of larger size which increased the lattice parameter.
- * The increase of Mn concentration increases the lattice parameter because the radius Mn³⁺ is larger than that of Fe³⁺.
- * The variation of the ratios $(I_{511}/I_{400} \text{ and } I_{422}/I_{400})$ depends on the replacement of Mn^{3+} ions for Fe³⁺ ions. These ratios changed to other values after γ irradiation.
- * Before irradiation the activation energy of the electron diffusion increased with increasing Mn concentration while after γ-irradiation it decreased with increasing Mn concentration.
- * The slight shift of the reflected peaks in X-ray diffraction patterns confirmed the effect of γ -rays which can be used in sensor technology.

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