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PREPARATION AND CHARACTERIZATION OF MAGNESIUM COPPER MIXED FERRITE AND MAGNESIUM COPPER THIN FILM

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Abstract

Magnesium-Copper mixed ferrite $Mg_{0.5}Cu_{0.5}Fe_2O_4$ was prepared by usual ceramic method at 1000°C for 8h. Structural and microstructural characteristics of the sample were characterized by using X-ray diffraction (XRD) and Scanning Electron Microscopy (SEM). Temperature dependent electrical conductivities were investigated in the temperature range of 299K-573K region. Furthermore, $Mg_{0.5}Cu_{0.5}Fe_2O_4$ thin film was deposited on p-Si substrate by using thermal diffusion method at 500°C for 1h.Microstructural characteristics and film thickness were examined by SEM. Current–Voltage (I-V) characteristics of the film were also investigated under the different illumination of visible light source.

Keywords: $Mg_{0.5}Cu_{0.5}Fe_2O_4$, XRD, SEM, Current-Voltage.

Introduction

Nanosize spinel ferrite particles have attracted considerable attention and continued efforts to investigate them for their technological importance to the microwave industries, high speed digital tap or disk recording, repulsive suspension for use in levitated railway systems, and magnetic refrigeration systems. The conventional way of preparing the ferrite is by solid-state reaction, which involves the mixing of oxides with intermittent grinding followed by high temperature sintering between 1300 and 1700°C. Though the process remains simple it has several drawbacks such as high reaction temperature, larger particle size, limited degree of homogeneity, and low sinterability.(Adam A, Ali Z, Andeltwab E & Abbas 2009)

Ferrite spinels have the formula $M(Fe_2O_4)$, where M is usually a divalent cation such as manganese (Mn^{2+}), nickel (Ni^{2+}), cobalt (Co^{2+}), zinc (Zn^{2+}), copper (Cu^{2+}), or magnesium (Mg^{2+}). M can also represent the monovalent lithium cation (Li^+) or even vacancies, as long as these absences of positive charge are compensated for by additional trivalent iron cations (Fe^{3+}). The oxygen anions (O^{2-}) adopt a close-packed cubic crystal structure, and the metal cations occupy the interstices in an unusual two-lattice arrangement. In each unit cell, containing 32 oxygen anions, 8 cations are coordinated by 4 oxygens (tetrahedral sites), and 16 cations are coordinated by 6 oxygens (octahedral sites). (Bhunia A & Bose D N 1998)

The antiparallel alignment and incomplete cancellation of magnetic spins between the two sublattices leads to a permanent magnetic moment. Because spinels are cubic in structure, with no preferred direction of magnetization, they are "soft" magnetically; *i.e.,* it is relatively easy to change the direction of magnetization through the application of an external magnetic field. (Candeia R A et al 2004)

In the present work, Magnesium-copper mixed spinel ferrite sample of $Mg_{0.5}Cu_{0.5}Fe_2O_4$ was first prepared by usual ceramic method and characterized by XRD, SEM, and temperature dependent electrical conductivity measurements to examine the phase formation and to study the

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structural, microstructural characteristics and electrical conductivity of the sample. Furthermore, $Mg_{0.5}Cu_{0.5}Fe_2O_4$ film was prepared on p-Si substrate by using thermal diffusion method and characterized by SEM and I-V characteristic measurements.

Experimental Procedure

Sample Preparation of Magnesium-Copper Mixed Spinel Ferrite

In this research, the starting materials of Magnesium Oxide (MgO), Copper Oxide (CuO), and Ferric Oxide, Fe_2O_3 were weighted with stoichiometric composition. The mixture powder sample was grounded by an agate motor for 3 h to be homogeneous and fine powders. The powders were annealed at 1000°C for 8 h in the vacuum chamber by using thermal resistive heating coil that controlled DELTA A Series Temperature Controller DTA4896. The K-type thermocouple (1300°C) was used as the temperature sensor to read-out the real temperature of the sample in the chamber. Finally, the Magnesium-copper mixed spinel ferrite, $Mg_{0.5}Cu_{0.5}Fe_2O_4$ was obtained. Experimental arrangement of the sample preparation system and the as-grown $Mg_{0.5}Cu_{0.5}Fe_2O_4$ sample are shown in Fig 1(a) and (b).

Figure1(a) Experimental arrangement of sample preparation system

Figure 1(b) Photograph showing the top-view of as-grown polycrystalline Magnesium-copper mixed spinel ferrite, $Mg_{0.5}Cu_{0.5}Fe_2O_4$

Preparation of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si Thin Film

The obtained $Mg_{0.5}Cu_{0.5}Fe_2O_4$ powder is firstly grounded by agate motor for 1 h to be fine and homogeneous sample. 1 g of the sample and 2-methoxythanol are mixed. Obtained mixture is stirred and boiled at 100°C to get the sol state. Finally, this solution is cooled down at room temperature.

p-Si (100) wafers, dimension of (0.50 cm x 0.50 cm) are used as the substrates. These substrates are cleaned using standard wafer cleaning process. p-type silicon substrates are etched in HF:H_2O (1:5) for 10 minutes, immerse in deionized (DI) water for 10 minutes to remove native oxide. And then, the samples are immersed in acetone and methyl alcohol for 10 minutes to remove impurities. Then, the substrates are rinsed in deionized water for 10 minutes and the substrates are dried at room temperature.

After cleaning process, the precursor solution is coated onto substrates by screen-printing technique. Later, coated-layers are first dried at room temperature. The $Mg_{0.5}Cu_{0.5}Fe_2O_4$ material is deposited on p-Si at 500 °C for 1 h by using DELTA A Series Temperature Controller DTA4896 temperature controller in the vacuum condition of -150 mmHg in thin film fabrication chamber.

After the deposition, the $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si film was etched in the mixture of ethanol, DI-water and HCl (1:1:1) for 10 – 15 min to get the homogeneously surface of the film. Then, $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film was dried at room temperature. Finally, $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si film was obtained. The flow-diagram of the $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film is shown in Fig (2).

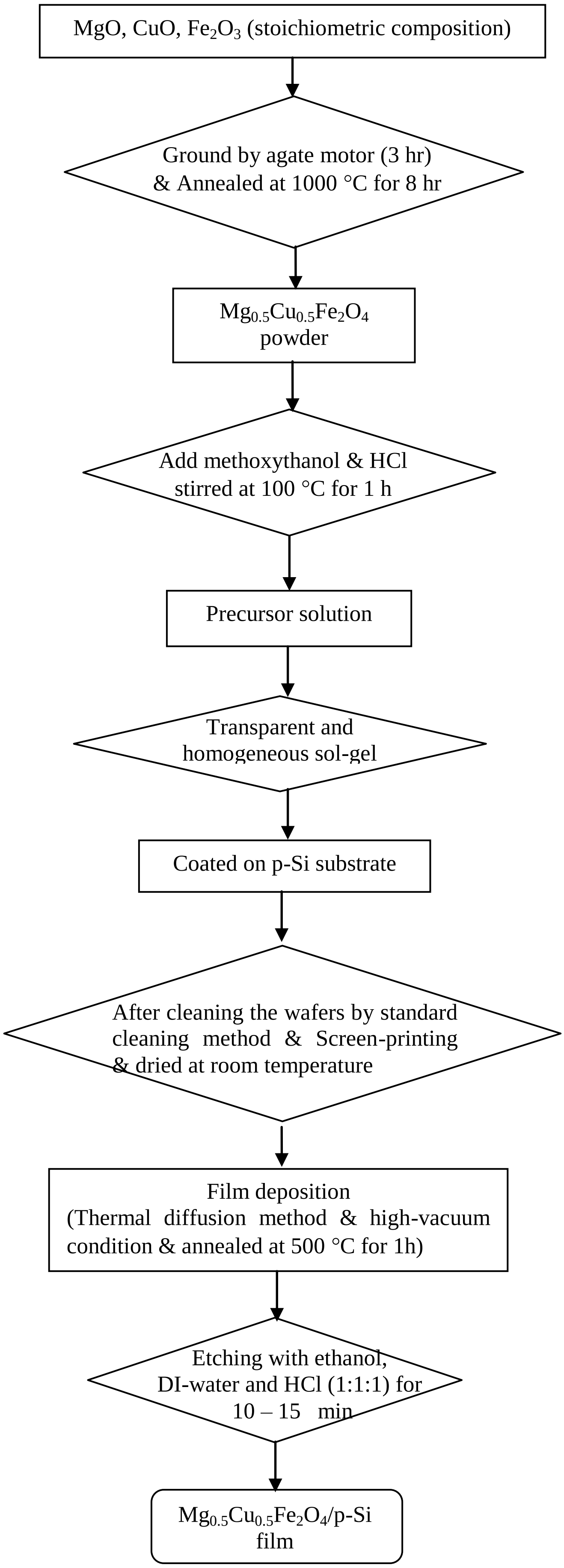

Figure 2 Flow-diagram of the $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film

XRD Measurement

Structural analysis, lattice parameters determination and crystallite size estimation of the samples were investigated by using RIGAKU MULTIFLEX X-ray diffractometer using Ni-filter with CuK_α radiation, $\lambda = 1.54056$ Å.

SEM Measurement

The morphological features of Magnesium-copper mixed spinel ferrite, $Mg_{0.5}Cu_{0.5}Fe_2O_4$ powders and $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si film are investigated by using JEOL JSM-5610LV SEM with the accelerating voltage of 15 kV, the beam current of 50 mA and 10000 times of photo magnification.

Temperature Dependent Electrical Conductivity Measurement

The samples were firstly made into pellets by SPECAC hydraulic press using 5 ton (~70 MPa). The silver paste was made over the sample to ensure good electrical contacts. The electrical resistances of the samples were observed in the temperature range of 299K-573K by the use of CAHO SR-T903 Temperature Controller. Thicknesses of the samples were measured by digital Vernier Caliper and used as 3.11 mm each. The area of the pellet was 1.14×10^{-4} m^2. The electrical resistances of the samples were measured by using FLUKE 45 Dual-display digitalmulti-meter.

I-V Characteristic Measurement

Photovoltaic effect of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film has been investigated in dark condition (0 lux) and different illumination conditions by using (60 W) electric bulb light source. The light intensity sensor of Si-photodiode (LIGHTMETER 2330LX, SEW) was placed near the sample with parallel position to record constant intensity of light source throughout the measurement. The output photovoltage of the film was observed by FUKE FK9208D digital voltmeter.

I-V (Current – voltage) characteristics in dark and in illumination condition of the film were also measured between the bias voltages of -5 V and +5 V with the step voltage of 0.2 V. In this measurement, DT-830B and FUKE FK9208X were used as the digital ammeter and voltmeter. Photograph of the experimental arrangement of I-V characteristic measurement is shown in Fig 3.

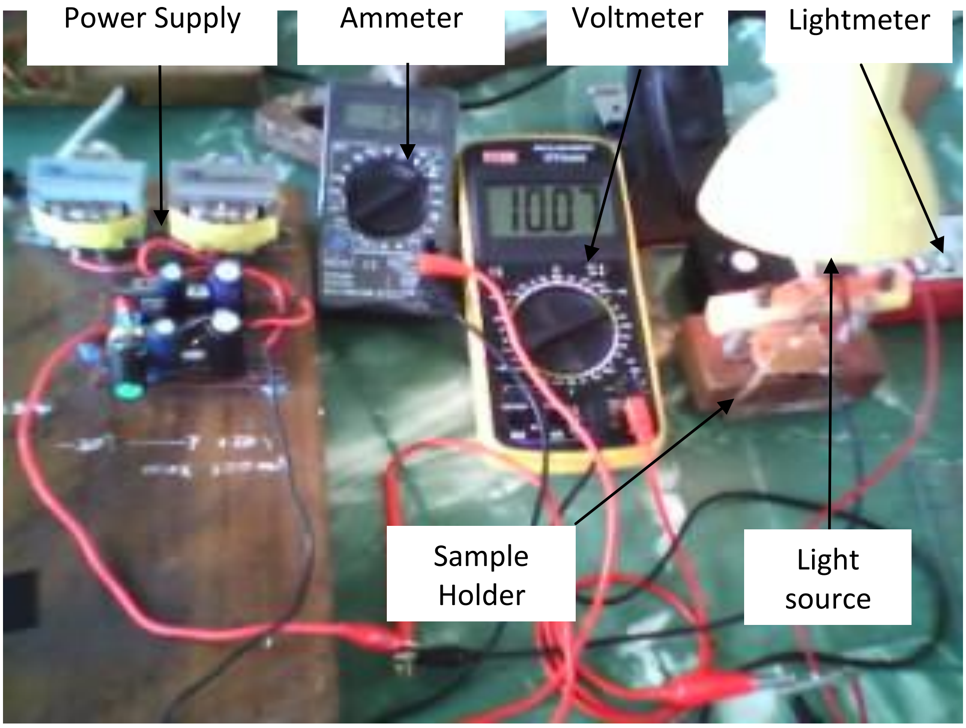

Figure 3 Photograph of the experimental arrangement of I-V characteristic measurement

Results and Discussion

XRD Analysis

Powder X-ray diffraction pattern of Magnesium-Copper mixed spinel ferrite, $Mg_{0.5}Cu_{0.5}Fe_2O_4$ is shown in Fig 4. XRD data of diffraction angle (2θ), atomic spacing (d), Miller indices (hkl), full width at half maximum (FWHM) and peak height of the sample are tabulated in Table 1. The collected XRD data are compared to those of JCPDS data library to identify the collected XRD pattern and to examine the single phase polycrystalline sample. Most of the collected diffraction lines are well assigned by JCPDS. From the XRD pattern, the material of $Mg_{0.5}Cu_{0.5}Fe_2O_4$ is found to single phase polycrystalline material belongs to cubic structure at room temperature. The lattice parameters are evaluated by using crystal utility of the equation of $\frac{\sin^2\theta}{(h^2+k^2+l^2)} = \frac{\lambda^2}{4a^2}$. Lattice parameters of the sample are $a = b = c = 8.28$ Å.

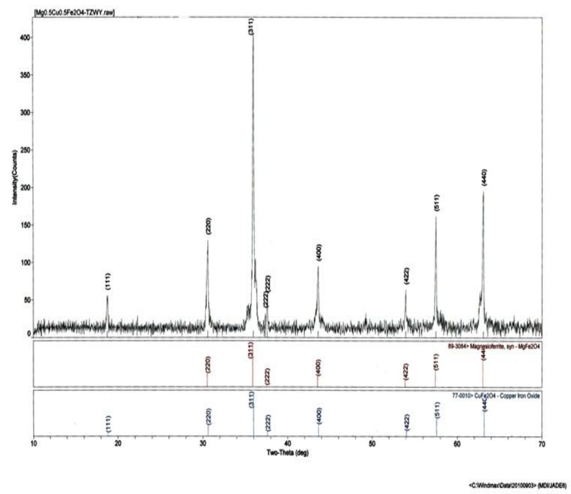

Figure 4 XRD pattern of Magnesium-Copper mixed spinel ferrite, $Mg_{0.5}Cu_{0.5}Fe_2O_4$

Table 1 XRD data of Magnesium-copper mixed spinel ferrite, $Mg_{0.5}Cu_{0.5}Fe_2O_4$

Line No	2θ (°)	(hkl)	d (Å)	FWHM (°)	I (%)
1	18.74	(111)	4.73	0.16	11.20
2	30.60	(220)	2.92	0.23	30.50
3	35.94	(311)	2.50	0.22	100.00
4	37.35	(222)	2.41	0.07	5.60
5	37.58	(222)	2.39	0.19	11.20
6	43.58	(400)	2.08	0.23	20.30
7	53.95	(422)	2.70	0.15	12.90
8	57.48	(511)	1.60	0.16	37.60
9	63.07	(440)	1.47	0.21	45.90

SEM Analysis

Morphological features of the samples are shown in Figure 5(a). It exposes that the grain shape of the sample is snow-like circular shape and the grain sizes are about 0.25 µm – 0.80 µm. Most of samples are found to be homogeneous and well grain boundary. SEM images of $Mg_{0.5}Cu_{0.5}Fe_2O_4$ film on p-Si substrate and the diffusion layer thickness of the $Mg_{0.5}Cu_{0.5}Fe_2O_4$ sample on p-Si are shown in Fig 5 (b) and (c). As shown in recorded SEM image (see Fig 5 (b)), the grain shape of the $Mg_{0.5}Cu_{0.5}Fe_2O_4$ layer is found to snow-like circular shape and crack free layer on p-Si substrate. As shown in observed diffusion layer thickness of the sample (see Fig 5 (c)), it is found that the layer boundary of the sample and substrate are found to homogeneous and the diffusion layer of the $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film is about 3.14 µm. It is indicated that the$Mg_{0.5}Cu_{0.5}Fe_2O_4$ sample is successfully deposited on p-Si substrate by using thermal diffusion technique at 500 C for 1h in the vacuum chamber.

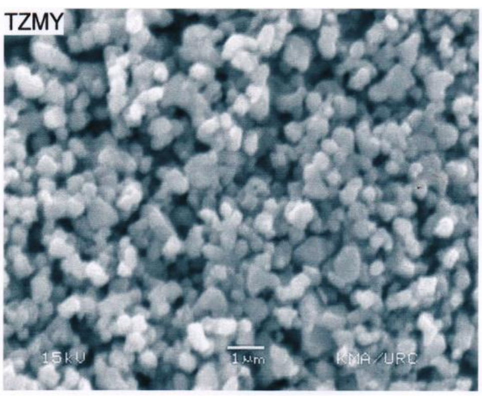

Figure 5(a). SEM image of Magnesium-copper mixed spinel ferrite, $Mg_{0.5}Cu_{0.5}Fe_2O_4$ powder

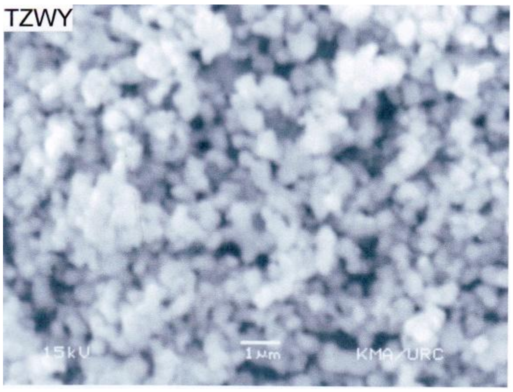

Figure 5(b) SEM image of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film

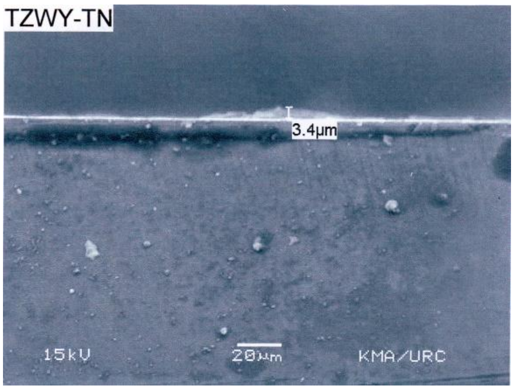

Figure 5(c) Diffusion layer thickness of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film

Temperature Dependent Electrical Conductivity

In the present work, Arrhenius plot of the variation of dc electrical conductivity of the $Mg_{0.5}Cu_{0.5}Fe_2O_4$ mixed spinel ferrite in the temperature range of 299 K - 573 K is shown in Fig 6.It shows that temperature dependent electrical conductivities of the sample are found to increase with increasing temperatures. Moreover, using the slope of the ln(σ) versus 10^3/T graph, the activation energy of the sample is calculated as 0.60 eV, according to the equation $\sigma = \sigma_0 \exp(-E_i / kT)$. From the experimental results of electrical conductivity, the $Mg_{0.5}Cu_{0.5}Fe_2O_4$ mixed spinel ferrite sample is a superionic conductor because its electrical conductivity is ($\sigma \geq 10^{-5}$ S cm^{-1}).

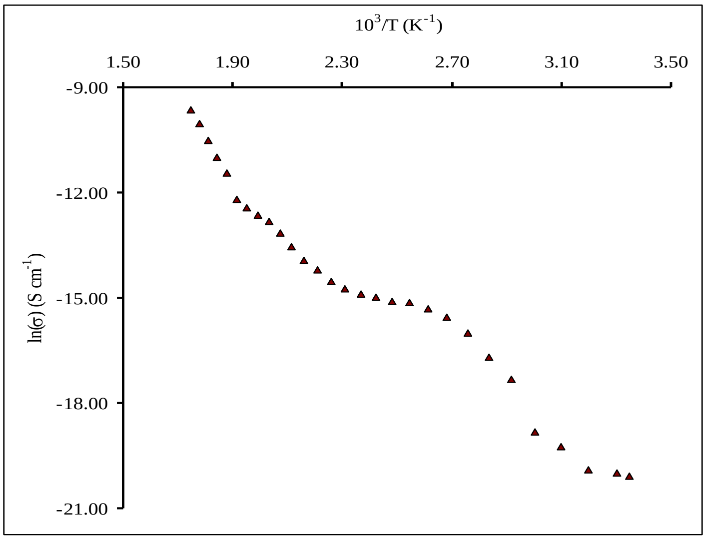

Figure 6 Arrhenius plot of the temperature dependent electrical conductivity of $Mg_{0.5}Cu_{0.5}Fe_2O_4$ mixed spinel ferrite

Current-Voltage Characteristic of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si Thin Film

Current-voltage, I-V measurements of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film devices within the bias voltages of -5 V up to +5 V (Step Up) and +5 V down to -5 V (Step Down) under the different illumination conditions to investigate the photosensitive effect and loop character. Copper are used as electrodes for the top and bottom regions. I-V characteristic curves of the $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film under dark (0 lux) and different illumination conditions of (250 lux (normal), 500 lux, 1000 lux, 1500 lux, and 2000 lux (60W, electric bulb light source) in the bias-voltage of -5 V - +5 V are shown in Fig 7(a) - (f). Among these curves, loop character of 1500 lux illumination condition is not found. Others I-V characteristic curves show the loop character of the sample in the positive bias regions.

The output current is exponentially increased with increasing bias voltage. Furthermore, comparison of the output currents as a function of (0 – 5 V) bias voltages in different illumination conditions is shown in Fig 7(g).Maximum output currents of the $Mg_{0.5}Cu_{0.5}Fe_2O_4$/ p-Si thin film at +5 V bias voltage in different illumination conditions are listed in Table 2.

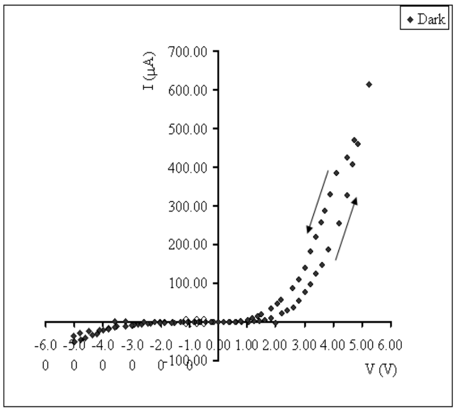

Figure 7(a) I-V characteristic curve of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film in dark (0 lux) condition

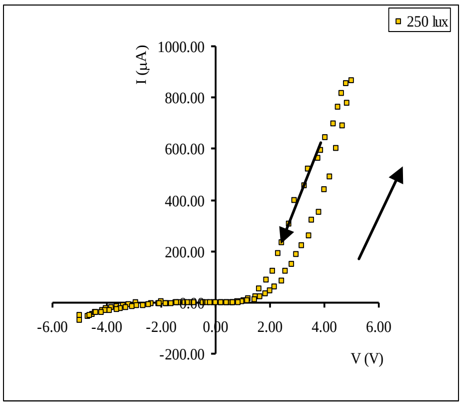

Figure 7(b) I-V characteristic curve of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film in ambient (250 lux) Illumination condition

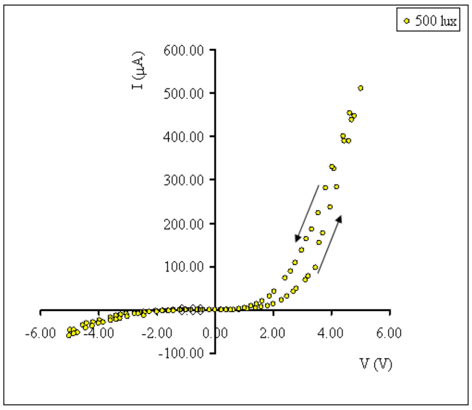

Figure 7(c) I-V characteristic curve of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film in 500 lux illumination condition

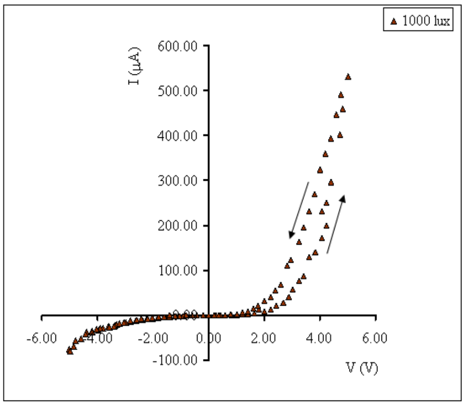

Figure 7(d) I-V characteristic curve of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film in 1000 lux illumination condition

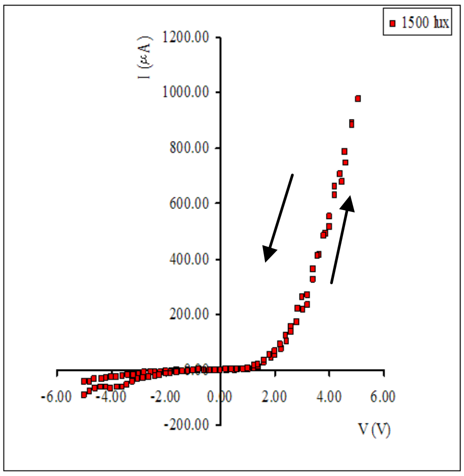

Figure 7(e) I-V characteristic curve of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film in 1500 lux illumination condition

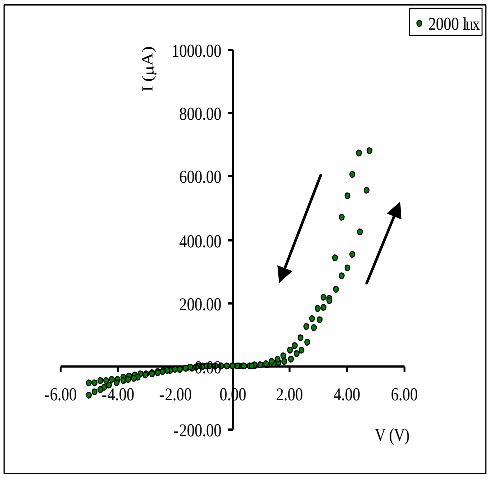

Figure 7(f) I-V characteristic curve of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film in 2000 lux illumination condition

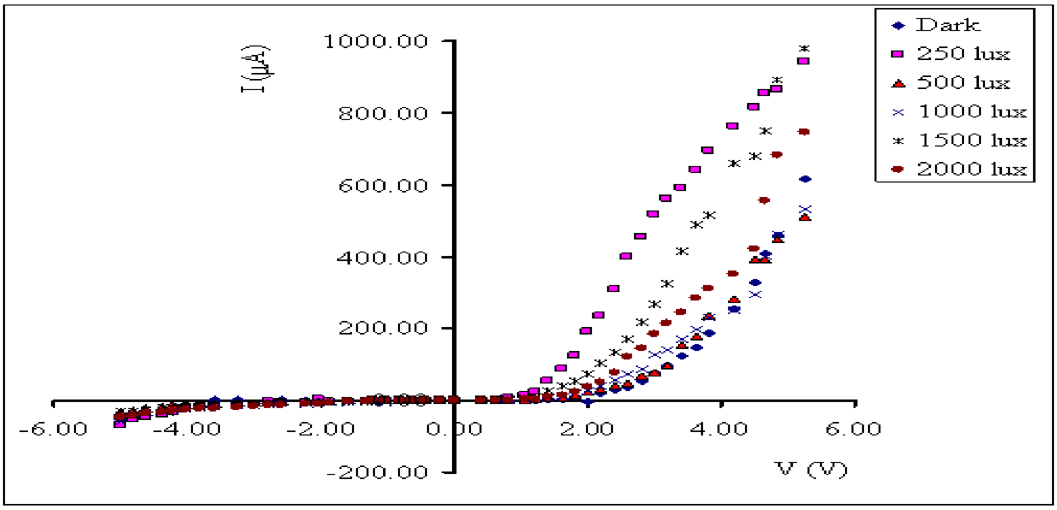

Figure 7(g) Comparision of the I-V characteristic curve of $Mg_{0.5}Cu_{0.5}Fe_2O_4$/ p-Si film in dark and in different illumination condition

Table 2 Maximum output currents of the $Mg_{0.5}Cu_{0.5}Fe_2O_4$/p-Si thin film at +5 V bias voltage in different illumination conditions

Sr No	Illumination (lux)	Output Current (μA)
1	0	616
2	250	943
3	500	512
4	1000	533
5	1500	979
6	2000	745

Conclusion

Magnesium-copper mixed spinel ferrite sample of $Mg_{0.5}Cu_{0.5}Fe_2O_4$ was prepared by usual ceramic method at 1000°C for 8 h in vacuum chamber.

Structural, microstructural and temperature dependent electrical conductivity of the as-grown $Mg_{0.5}Cu_{0.5}Fe_2O_4$ sample were reported by means of XRD, SEM and temperature dependent electrical conductivity measurements. From the XRD pattern, $Mg_{0.5}Cu_{0.5}Fe_2O_4$ mixed spinel ferrite belongs to cubic structure and the lattice parameters are $a = b = c = 8.28$ Å. It is found to single phase polycrystalline material. From the SEM micrograph, the grain shape of the sample is snow-like circular shape and the sizes are about 0.25 µm – 0.80 µm. Most of samples are found to be homogeneous and well grain boundary. From the temperature dependent electrical conductivity results, $Mg_{0.5}Cu_{0.5}Fe_2O_4$ is the superionic conductor at high temperature and the activation energy is also evaluated.

In addition, $Mg_{0.5}Cu_{0.5}Fe_2O_4$ was successfully deposited on p-Si substrate at 500°C for 1 h in the vacuum chamber by using thermal diffusion method. Morphological features and layers thickness of the film were investigated by SEM method. Electrical characterizations of the film were examined by I-V characteristic measurements in the bias-voltage of -5 V - +5 V under different illumination conditions. From the SEM micrograph, the grain shape of the $Mg_{0.5}Cu_{0.5}Fe_2O_4$ layer is found to snow-like circular shape and crack free layer on p-Si substrate. The diffusion layer thickness of the sample is about 3.14 µm.

The results of these studies are promising and suggested that Magnesium-copper mixed spinel ferrite, $Mg_{0.5}Cu_{0.5}Fe_2O_4$ based films are attractive for use as storage elements for ferromagnetic memory applications.

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