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Synthesis and Properties of $Mg_{(x)}Fe_{(1-x)2}O_4$ Series nanoparticles

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ABSTRACT

Magnesium ferrite Series nanoparticles have been prepared in a less time by sol-gel auto combustion synthesis method. The main objective in this current work is to influence by increasing the concentration of ferrite and decreasing the concentration of magnesium. Prepared $Mg_{(x)}Fe_{(1-x)2}O_4$ (x = 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8 and 0.9) is to investigate the difference among each sample in their properties. The prepared nano particles were characterized to find Structural, Optical Properties and thermal stability of particles. Using X-Ray diffraction (XRD), Scanning electron microscope (SEM), UV-Visible spectroscopy, Thermal analysis was done by TG/DTA, Average particle sizes were calculated by using (Particle Size Analyzer) PSA analysis. XRD revealed the formation of $Mg_{(x)}Fe_{(1-x)2}O_4$ is in cubic structure for the sample with Mg-Fe equal ratio (x=0.5). The crystalline size estimated using Scherer formula in the range of 12-23 nm. SEM images illustrates the porous nature in all $Mg_{(x)}Fe_{(1-x)2}O_4$ series. UV-visible spectra shown noticeable peaks of Magnesium ferrite.

Keywords: Magnesium Ferrite; Combustion method; XRD; SEM; TG/DTA; UV-visible;

1. INTRODUCTION

In present research days, spinel ferrites have been investigated for their useful electrical and magnetic properties. This can be used for different applications in information storage systems, telecommunication devices, sensors, catalysis, magnetic drug delivery, permanent magnets, recording heads, antenna rods, loading coils, magnetic refrigeration, magnetic liquids, as a microwave absorber [1]. Spinel ferrite compounds have a general formula $A[B_2]O_4$ where A represent divalent metal ions Zn^{2+}, Mg^{2+}, Ni^{2+},Co^{2+}, and B represent trivalent metal ions such as: Fe^{3+}, Cr^{3+}, Al^{3+}, Mn^{3+} [2]. In this one of important ferrite is Magnesium ferrite ($MgFe_2O_4$). It has a cubic structure of normal spinel-type and a soft magnetic n-type semiconducting material, with high resistivity and low magnetic and dielectric losses. Which finds a number of applications in heterogeneous catalysis, sensors, and also extensively used in various technological and biomedical applications in the past decades [3-5]. It covers wide range of applications including humidity sensor, switching circuits, contrast agent in magnetic resonance imaging, tissue repair, detoxification of biological fluids, targeted drug delivery and magnetic hyperthermia [6-7]. The physical and chemical properties of ferrites are dependent upon factors such as sintering temperature, sintering time, rate of heating, rate of cooling, etc [7].There different types of methods to synthesize magnesium ferrite nano particles[8-14]. In this work magnesium ferrite series $Mg_{(x)}Fe_{(1-x)2}O_4$ is synthesized with accurate weight of precursors for each samples, and found different colour for each magnesium ferrite sample form X= 0.1 to 0.9.

2. PREPARATION MAGNESIUM FERRITE NANO PARTICLES

A soft magnetic n-type semiconducting $MgFe_2O_4$ Nano particles powder was prepared using sol-gel auto combustion technique [15]. The materials used as precursors were Magnesium nitrate $Mg(NO_3)_2$, Iron

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Figure 1: $Mg_{(x)}Fe_{2(1-x)}O_4$ Series nanoparticle samples

nitrate $Fe(NO_3)_2$ and glycine. All of them were of high purity. Glycine possesses a high heat of combustion. It is an organic fuel providing a platform for redox reactions during the course of combustion. Initially the Weights of Magnesium nitrate and Iron nitrate are taken as mentioned in the below calculation. For x = 1 sample that is $Mg_{0.1}Fe_{0.9}O_4$, the weight of Magnesium nitrate is 0.197gr and weight of Iron nitrate is 2.80gr. These are dissolved in separate 500ml Beaker with each 25ml of Distilled Water and keep stirring for 30 min, after that add both chemicals slowly with Glycine of Weight 0.932gr and keep stirring for 30min.Then keep the Beaker on Furnace and maintain 80^0C to 100^0C till to get Thick Gel form. After that increase & maintain the temperature in range of 180^0C to 200^0C. Slowly Fumes & Flames Takes Place then the nanocrystalline $Mg_{0.1}Fe_{0.9}O_4$ powder was formed within a few minutes. It was sintered at 650^0C temperature for 4hr. Then we got a Dark Brown color of Magnesium Ferrite **(**$Mg_{0.1}Fe_{0.9}O_4$) and also found different colors for different sample as shown in Figure 1.

2.1. Calculation of $Mg_{(x)}Fe_{(1-x)2}O_4$ (Magnesium Ferrite) Series

According To Rocket Fuel Chemistry, The Valence Of Carbon, Hydrogen, Nitrogen, Oxygen, Magnesium And Iron Are Given By C = 4, H = 1, N = 0, O = -2, Mg = +2, Fe = +3 respectively. Then Total Valence Of Glycine ($C_2H_5NO_2$), Magnesium Nitrate ($Mg(NO_3)_2$), Iron Nitrate ($Fe(NO_3)_3$) are 9, -10, -15 respectively.

$$\psi = \left| \frac{Fuel\ Oxidation\ State}{Oxidizer\ Oxidation\ State} \right| = 1$$

$$\psi = \left| \frac{9n}{n_1(-10) + n_2(-15)} \right| \quad (1)$$

Molecular weight of Glycine, Magnesium Nitrate, Iron Nitrate are 75.07, 256.41,404 respectively.

2.2. Calculation for $Mg_{0.1}Fe_{0.9}O_4$ Material if x = 0.1in $Mg_{(x)}Fe_{(1-x)2}O_4$ Series-

For total 3gr of magnesium nitrate and iron nitrate, we calculated the weights of magnesium nitrate (n_1) is 0.19761gr and Iron Nitrate (n_2) is 2.8023gr for x = 0.1 by using molecular weights.

For Weight of Glycine: Substitute n_1& n_2 values in above Equation 1, It was equated to 1 for 100% combustion.

So, the weight of glycine fuel (n) = 0.932gr.

Note: Similar Calculation for Total Series then,

3. RESULTS AND DISCUSSIONS

The figure 2 depicts that $Mg_{(x)}Fe_{(1-x)2}O_4$ series (x = 0.5) sample lattice parameter matches well with JCPDS (71-1232) file, remaining samples such as x = 0.1 ,0.2, 0.3, 0.4, 0.6, 0.7, 0.8 and 0.9 were not accurately match with JCPDS number due to its oxidizer percentages in the samples. The $Mg_{(0.5)}Fe_{(0.5)2}O_4$ (x = 0.5) sample having equal ratio of the oxidizers percentage. It has been matched exactly the peaks with the corresponding planes (2 2 0), (3 1 1), (4 0 0), (4 2 2), (5 1 1) and (4 4 0) observed at 30°, 35°, 43°, 54°, 57° and 62° respectively. As the width of the peak increases size of particle size decreases, which resembles that present material in nano range [16-20].

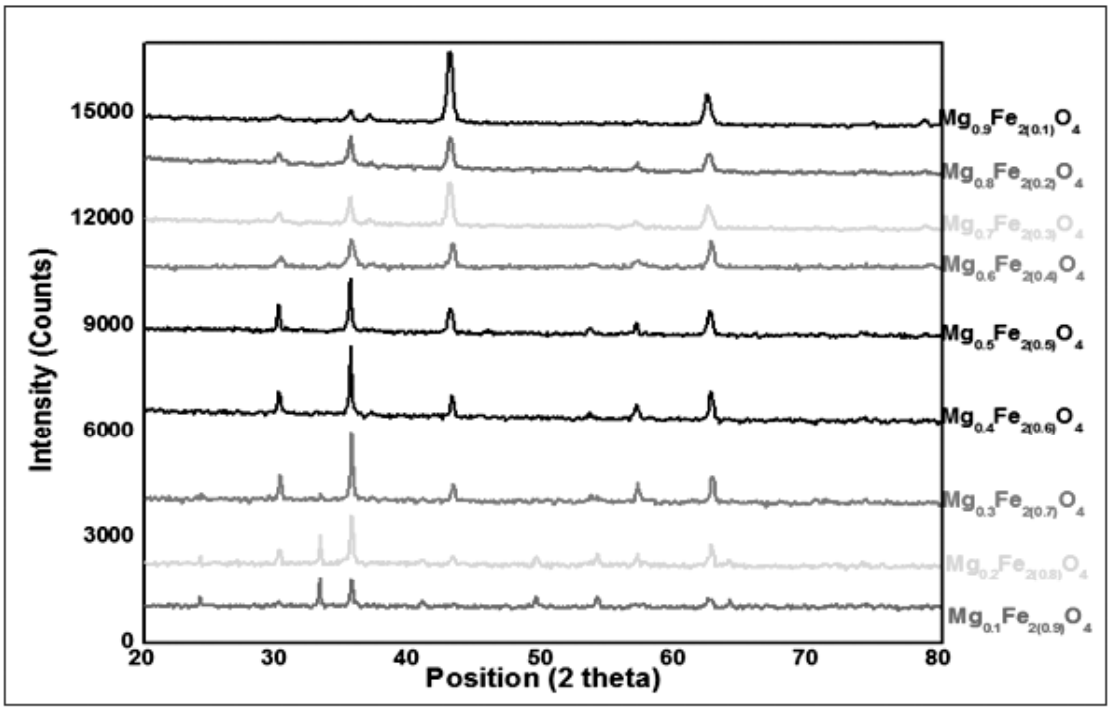


Figure 2: X-Ray Diffracted pattern of $Mg_{(x)}Fe_{(1-x)2}O_4$ series.

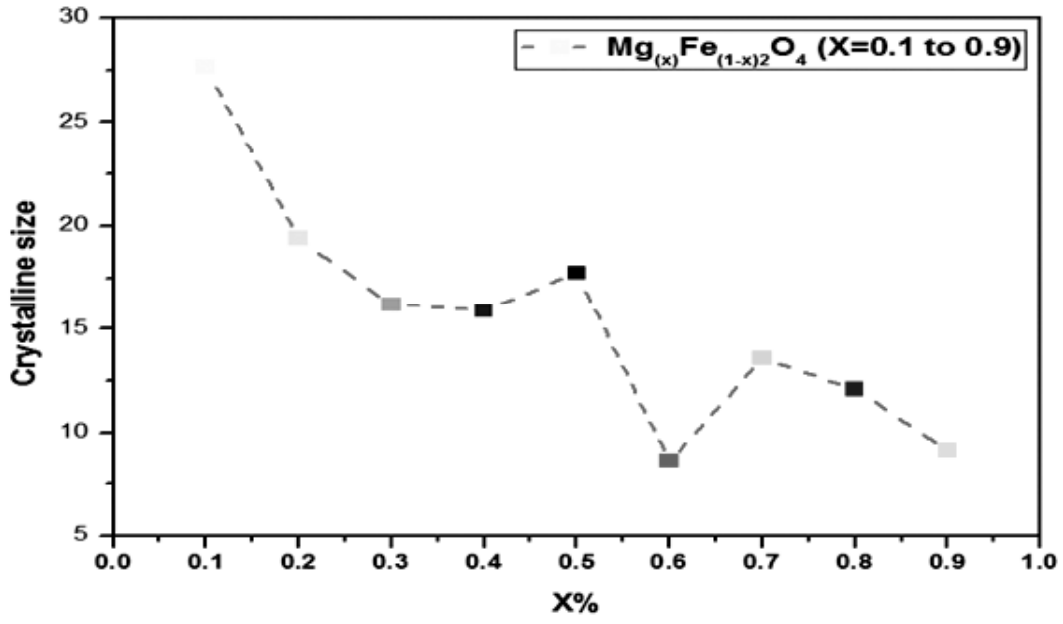
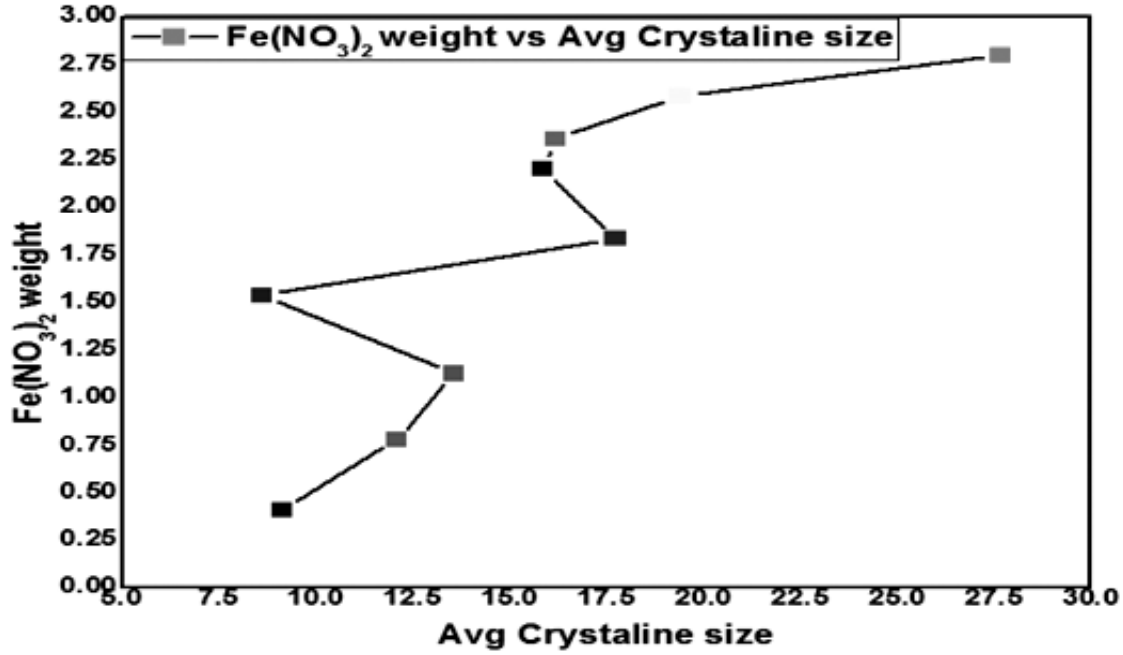


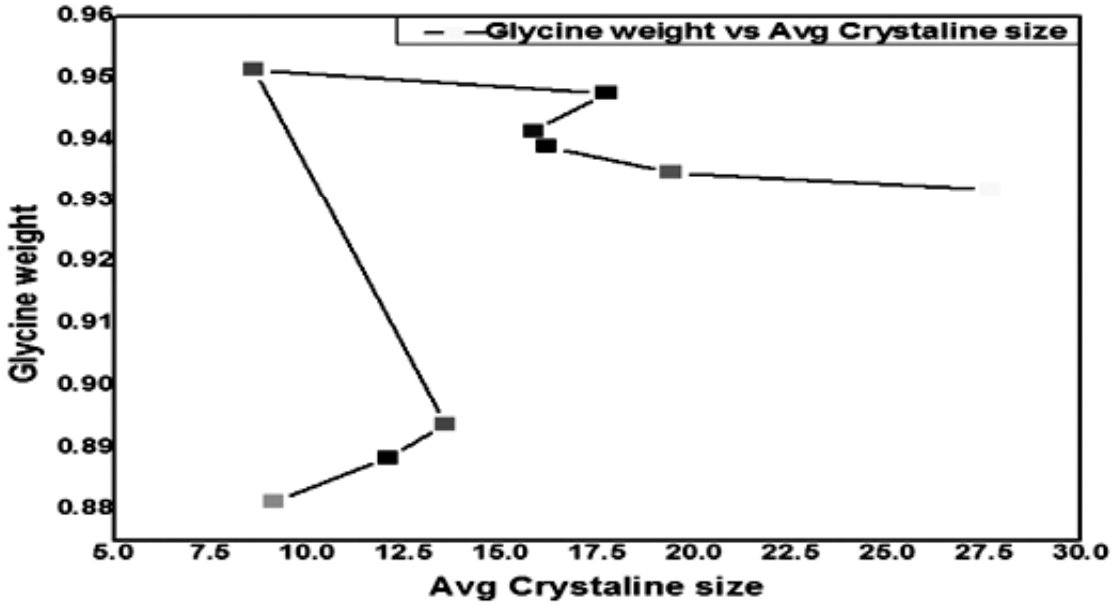
Figure 3: Plot of Average crystalline size of the $Mg_{(x)}Fe_{(1-x)2}O_4$ (x = 0.1 to 0.9) series, which calculated from Debye-Scherer formula.

The lattice parameters were obtained a = b = c = 0.83nm. This indicates that $Mg_{(0.5)}Fe_{(0.5)2}O_4$ (x = 0.5) nano particles are in the cubic structure. The Average crystallite size of prepared and annealed samples of $Mg_{(x)}Fe_{(1-x)2}O_4$ (x = 0.1, 0.2,0.3, 0.4, 0.5, 06, 0.7,0.8, and 0.9) is 27.1 nm, 19 nm, 16.2 nm, 15.4 nm, 17 nm, 8.6 nm, 13.51 nm, 12 nm and 9.1 nm respectively. Here as we observed highest crystalline size for the sample $Mg_{(0.1)}Fe_{(0.9)2}O_4$ (where x = 0.1) is 27.67 nm and lowest crystalline size for $Mg_{(0.6)}Fe_{(0.4)2}O_4$ (where x = 0.6) is 8.617 nm shown in figure 3.

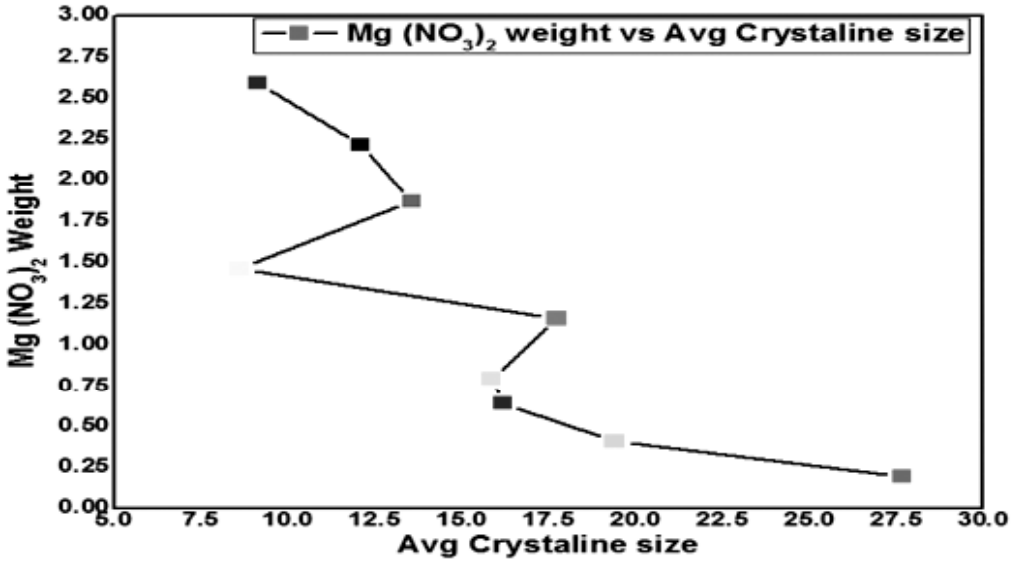
The average crystalline size verses oxidizer weights and Fuel weights were explained tentatively in figure 4. This can be show that when iron nitrate weight is increasing the crystalline size is also increasing. Where magnesium nitrate weight is increasing the crystalline size is decreasing and no effect was observed in changing the weight of Glycine.



(a)



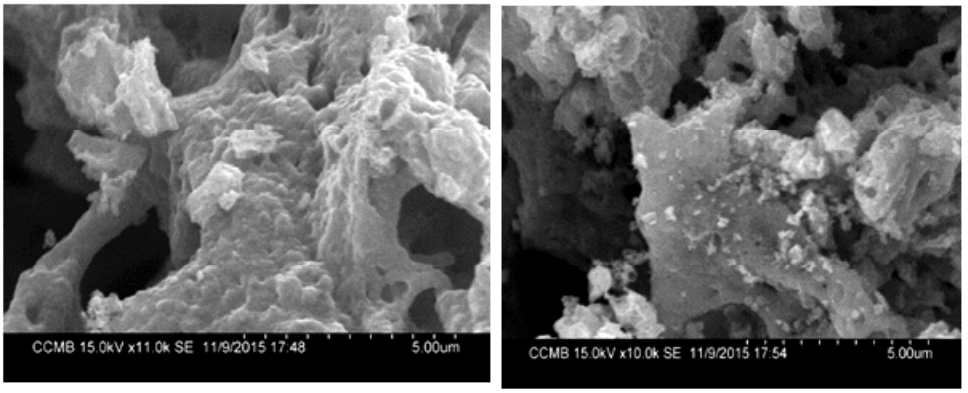
(b)



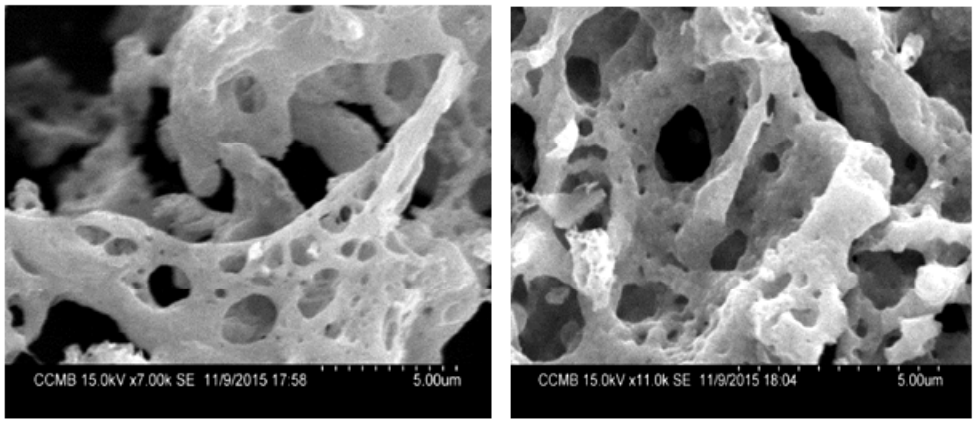
(c)

Figure 4: a) Average crystalline size verses $Fe(NO_3)_3$ weight, b) Average crystalline size verses $Mg(NO_3)_2$ weight, c) Average crystalline size verses glycine weight

The surface morphology was observed from SEM with 5um magnification. SEM micrographs show remarkable change in the structure regarding porosity, grain size of sample illustrates in the Figure 5 (a) to (i). From fig it can be concluded that these have frothy and small holes within structure, which may be due to escaping large number of gases during the combustion. It can be seen from the ûgure that sample exhibit



(a) **(b)**



(c) **(d)**

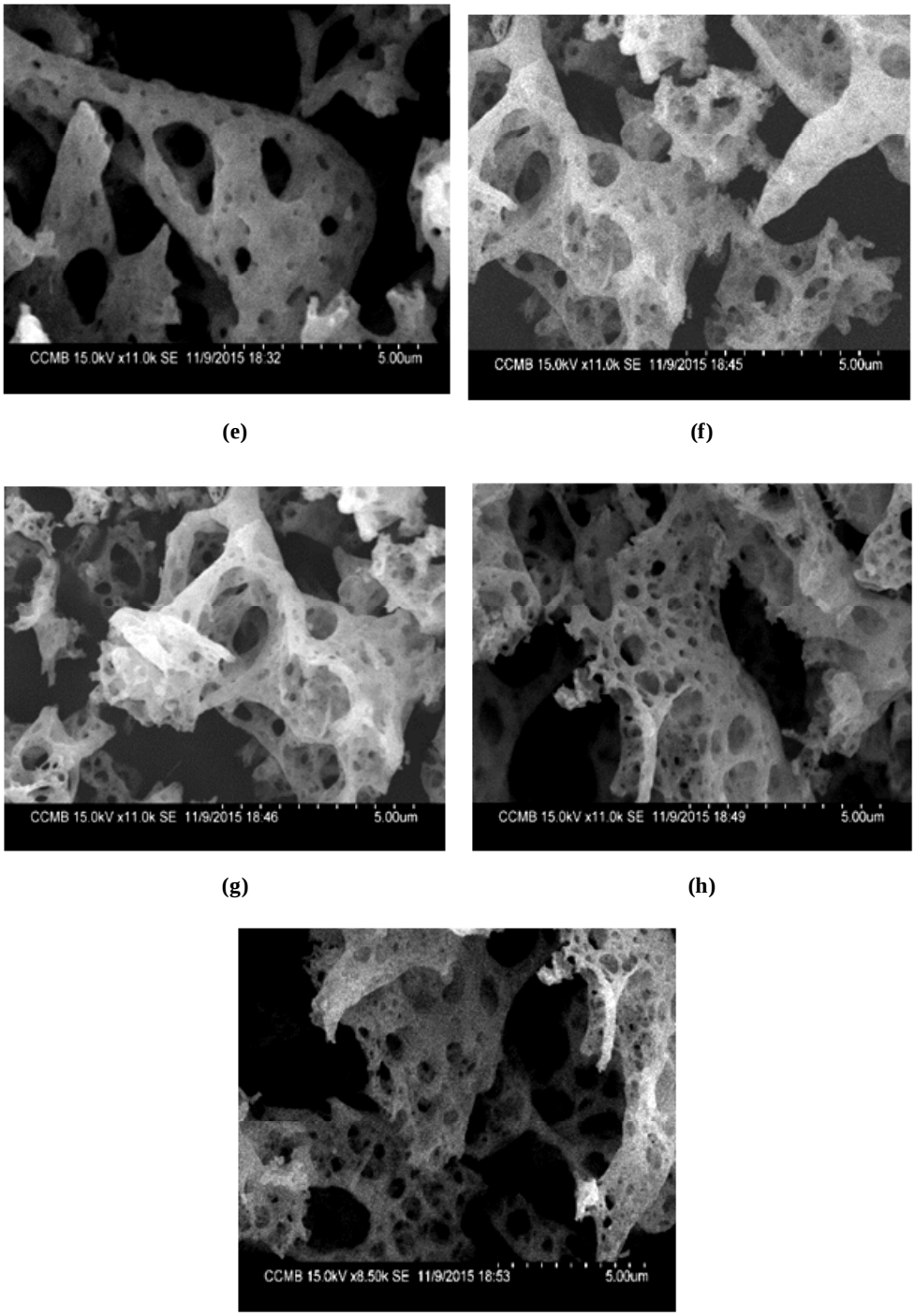


Figure 5.SEM Images of (a) $Mg_{0.1}Fe_{(0.9)2}O_4$, (b) $Mg_{0.2}Fe_{(0.8)2}O_4$, (c)$Mg_{0.3}Fe_{(0.7)2}O_4$ (d) $Mg_{0.4}Fe_{(0.6)2}O_4$ (e) $Mg_{0.5}Fe_{(0.5)2}O_4$ (f) $Mg_{0.6}Fe_{(0.4)2}O_4$ (g)$Mg_{0.7}Fe_{(0.3)2}O_4$ (h) $Mg_{0.8}Fe_{(0.2)2}O_4$ (i) $Mg_{0.9}Fe_{(0.1)2}O_4$

network with voids and pores. The porosity in all cases is found to be entirely intergranular. The appearance of spongy structure was attested a better crystallinity of spinel phase. These SEM images of $Mg_{(x)}Fe_{(1-x)2}O_4$ (x = 0.1, 0.2, 0.3, 0.4, 0.5 ,0.6, 0.7, 0.8 and 0.9) shows that the porosity is increasing as expected due to the increasing the percentage of the magnesium while decreasing the percentage of ferrite.

Figure 6 depict the reflectance spectra of pure and magnesium ferrite nanoparticles in the spectral range of 300-900 nm. The Optical properties of the samples were studied using Systronics UV–Visible spectrometer-2202.

It is a clear all the samples show optical properties in the visible region. In pure $Mg_{(x)}Fe_{(1-x)2}O_4$ (x = 0.5) sample reflectance percentage decreases with equal ration of Mg-Fe percentage. But in the case of remaining samples $Mg_{(x)}Fe_{(1-x)2}O_4$ (x = 0.1 to 0.9) reflectance percentage almost equal. All the samples in the series are representing two peaks around 336 nm and 834 nm wavelength with small blue shift and red shift variation with respect to nano size in the particles.

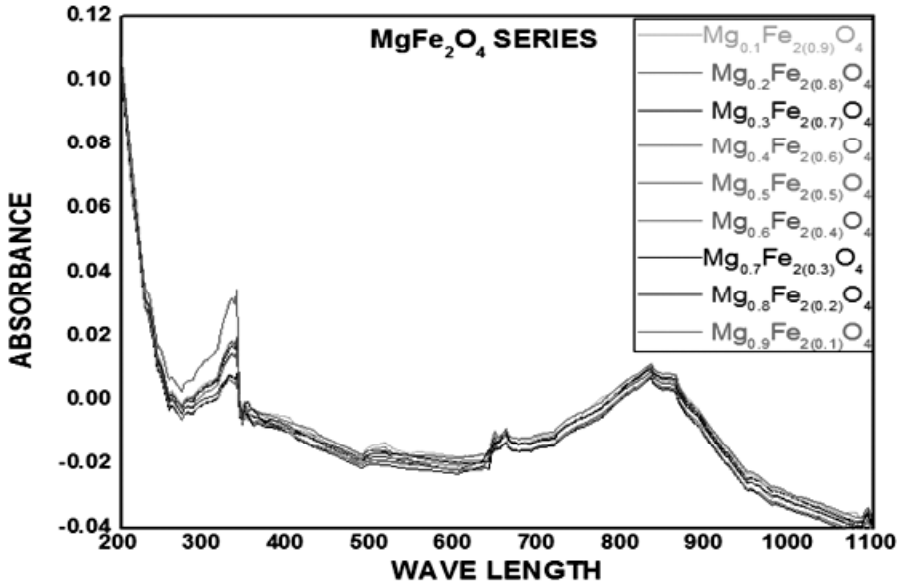
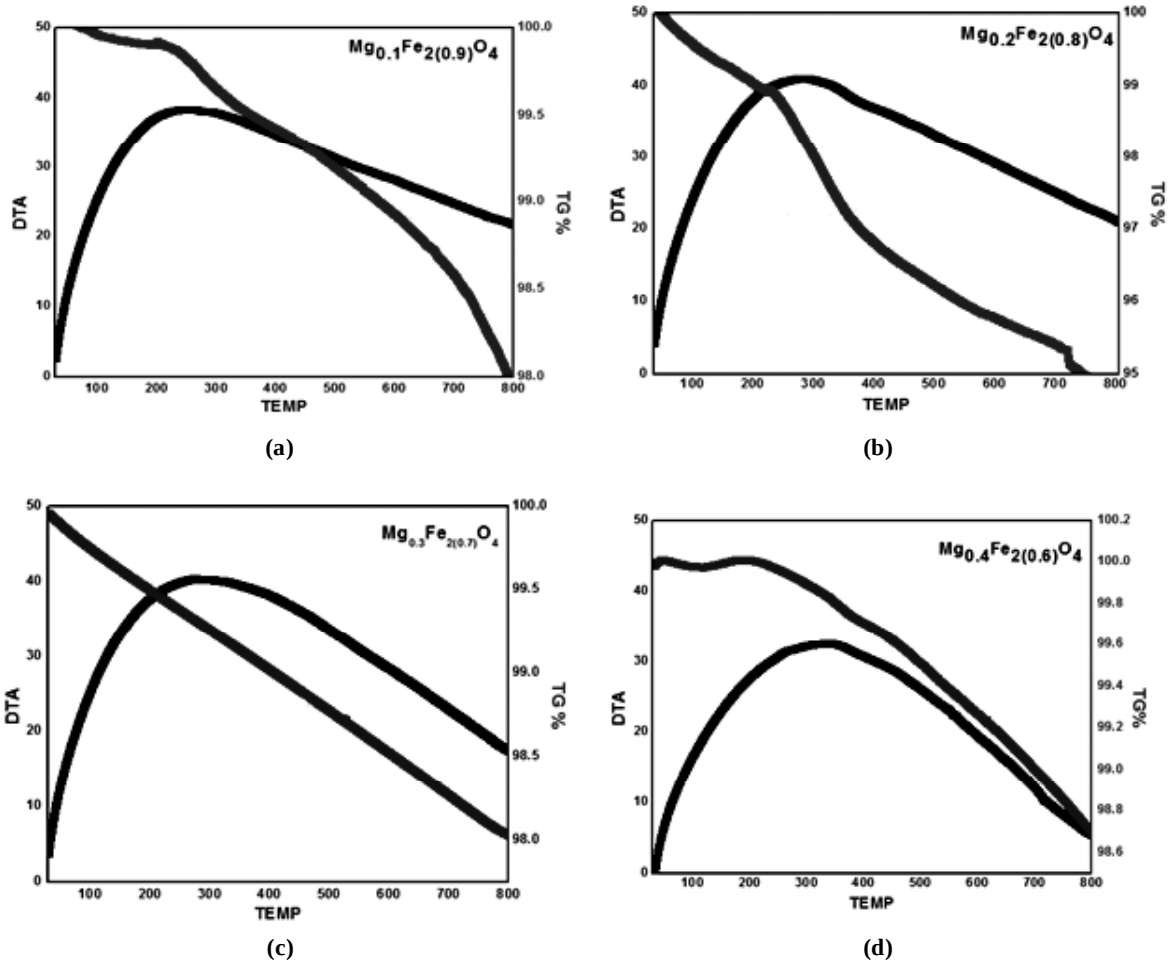


Figure 6: Reflectance spectra of $Mg_{(x)}Fe_{(1-x)2}O_4$ (x=0.1,0.2,0.3,0.4,0.5,0.6,0.7,0.8 and 0.9) nanoparticles.



(a) **(b)**

(c) **(d)**

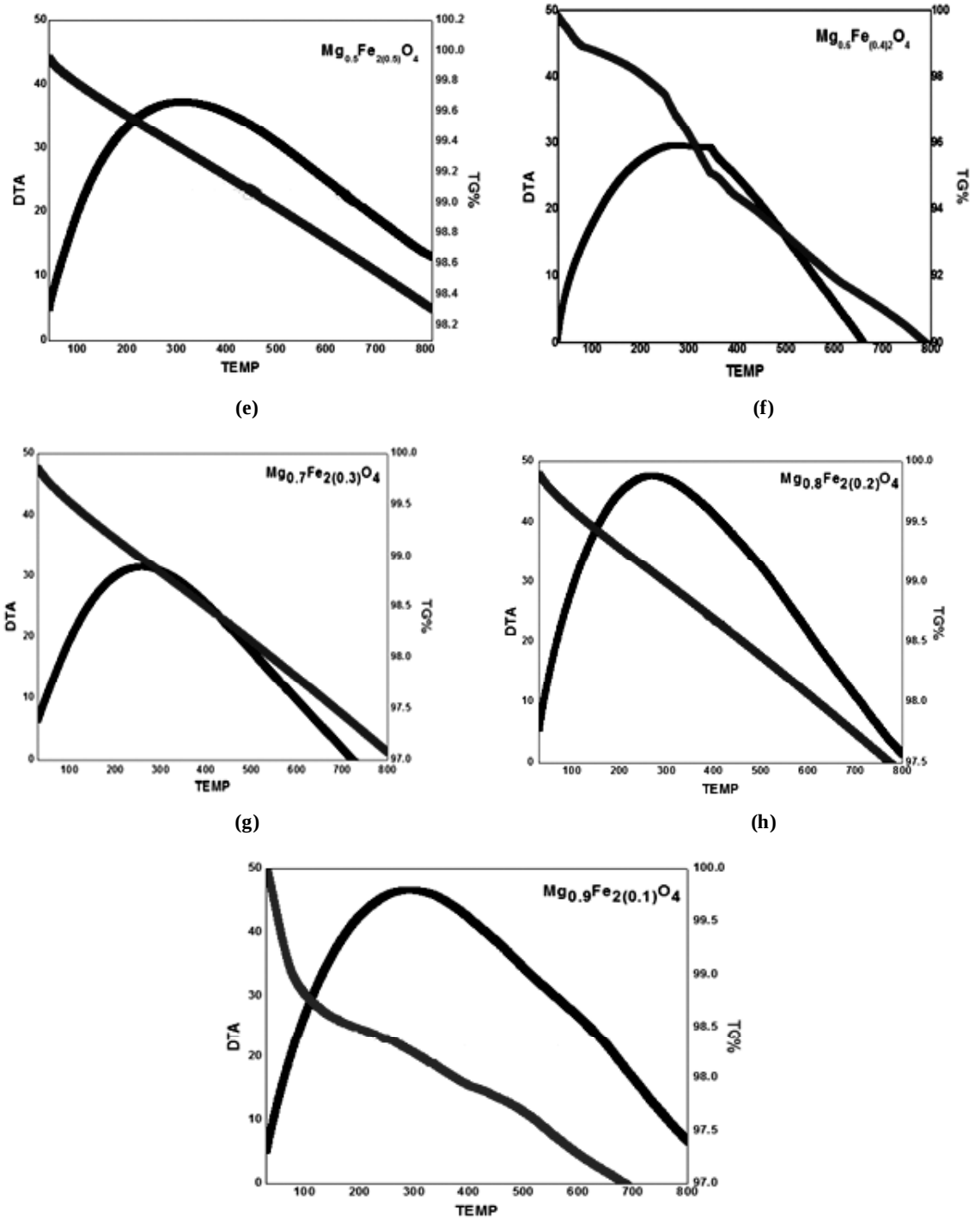
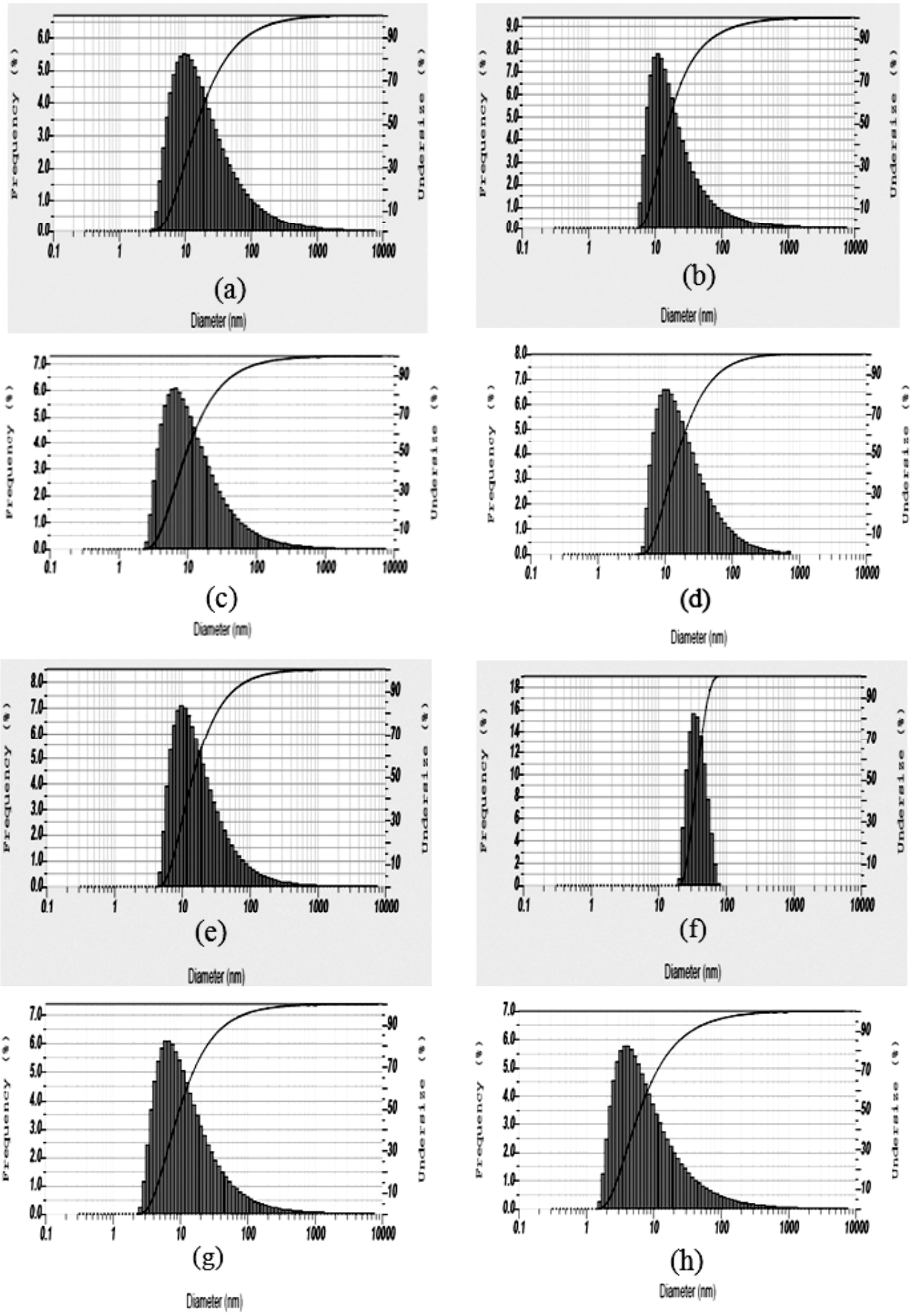


Figure 7: TG-DTA Curves of $MgFe_2O_4$ series (a) $Mg_{0.1}Fe_{(0.9)2}O_4$, (b)$Mg_{0.2}Fe_{(0.8)2}O_4$, (c) $Mg_{0.3}Fe_{(0.7)2}O_4$ (d) $Mg_{0.4}Fe_{(0.6)2}O_4$ (e) $Mg_{0.5}Fe_{(0.5)2}O_4$ (f)$Mg_{0.6}Fe_{(0.4)2}O_4$ (g) $Mg_{0.7}Fe_{(0.3)2}O_4$ (h) $Mg_{0.8}Fe_{(0.2)2}O_4$ (i) $Mg_{0.9}Fe_{(0.1)2}O_4$

Figure 7 shows the TG-DTA curves for magnesium ferrite samples. The TG analysis was observed from room temperature to 800ºC from which the stability of the sample can be known from weight loss calculations. The weight loss of the samples observed at room temperature to 100ºC due to the evaporation of water molecules, whereas 100ºC to 400ºC the weight loss caused by evaporation of inorganic materials. After 400ºC the weight loss occurs due to the evaporation of un-reacted materials which is involved in the samples. The total weight loss of the $MgFe_2O_4$ series nano particles was measured as 2.1%, 5%, 2.2%,


Frequency (%)
Undersize (%)
Diameter (nm)
(a)
(b)
(c)
(d)
(e)
(f)
(g)
(h)

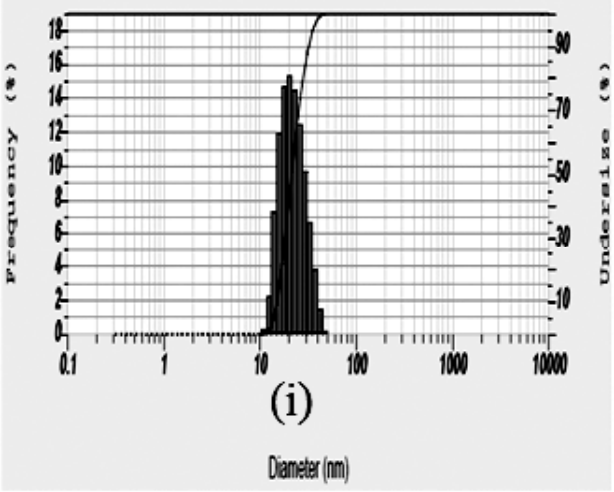


Figure 8: Histograms from Average particle size Analyzer of (a)$Mg_{0.1}Fe_{(0.9)2}O_4$, (b) $Mg_{0.2}Fe_{(0.8)2}O_4$, (c) $Mg_{0.3}Fe_{(0.7)2}O_4$ (d) $Mg_{0.4}Fe_{(0.6)2}O_4$ (e)$Mg_{0.5}Fe_{(0.5)2}O_4$ (f)$Mg_{0.6}Fe_{(0.4)2}O_4$ (g) $Mg_{0.7}Fe_{(0.3)2}O_4$ (h) $Mg_{0.8}Fe_{(0.2)2}O_4$ (i)$Mg_{0.9}Fe_{(0.1)2}O_4$

1.3%, 1.71%, 10%, 2.9%, 2.5% and 3%. From these results 1.3% shows the $Mg_{0.4}Fe_{2(0.6)}O_4$ has least weight loss in the series.

Figure 8 shows histograms of Magnesium ferrite series. The mean value of the histogram taken as the average particle size. According to the graphs the particle size distributions of nanoparticles range from 10 nm 100 nm. From the particle size analyzer the average particle size was obtained as 34.7 nm, 52.31 nm, 47.4 nm, 31.8 nm, 38.1 nm, 30.2 nm, 32.41 nm, 27.12 nm and 22.5 nm, which was shown in the following figure. From this results $Mg_{0.9}Fe_{2(0.1)}O_4$ shows the least average particle size. Hence, these results were supported to XRD average crystallite size.

4. CONCLUSION

In the present work, nanoparticles of $Mg_{(x)}Fe_{(1-x)2}O_4$ (x = 0.1, 0.2, 0.3, 0.4, 0.5, 06, 0.7, 0.8, and 0.9) are synthesized using sol–gel auto combustion route in less time, with a view to understand the changes of Magnesium ferrite properties in the nano Scale range. In actual sense the distribution of the cations becomes the cause of the structure and properties of ferrites. Structural, morphological and optical properties are studied successfully by influencing changing the concentration of the Mg-Fe percentage. The synthesized $Mg_{(x)}Fe_{(1-x)2}O_4$ (x = 0.1, 0.2, 0.3, 0.4, 0.5, 06, 0.7, 0.8, and 0.9) nanoparticles affecting on several factors including the method of preparation, chemical composition and grain structure or size of particles while changing the concentrations.

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