

# Synthesis of Spinel $\text{MgFe}_2\text{O}_4$ Ferrite Material and Studying its Structural and Morphological Properties Using Solid State Method

S. YONATAN MULUSHOA<sup>1\*</sup>, M. TULU WEGAYEHU<sup>1</sup>,  
G. TEWODROS AREGAI<sup>1</sup>, N. MURALI<sup>2</sup>, M. SUSHMA REDDI<sup>1</sup>,  
B. VIKRAM BABU<sup>1</sup>, T. ARUNAMANI<sup>1</sup> and K. SAMATHA<sup>1</sup>

<sup>1</sup>Department of Physics, Andhra University, Visakhapatnam, Andhra Pradesh, India

<sup>2</sup>Advanced Analytical Laboratory, Andhra University, Visakhapatnam, Andhra Pradesh, India  
*tafe.mule06@gmail.com*

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**Abstract:**  $\text{MgFe}_2\text{O}_4$  ferrite has excellent magnetic behavior and high resistive material. The properties of Mg ferrite were investigated by solid state reaction method. In this work, our aim is to synthesize  $\text{MgFe}_2\text{O}_4$  using oxide precursors (*i.e.*  $\text{MgO}$  and  $\text{Fe}_2\text{O}_3$ ). The samples were pre-calcined at 800 °C for 5 hours and calcined at 1000 °C for 6 hours and have the single phase cubic spinel structure. The phase composition, micro-morphology, elemental analysis and Wyckoff sites of the products of Mg-Fe-O system were characterized by x-ray diffraction (XRD), scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS) and Fourier transform infrared (FTIR), respectively. Spinel Mg ferrite belongs to Fd-3m space group. From the XRD result the crystallite size, lattice constant and cell volume of  $\text{MgFe}_2\text{O}_4$  was found to be 44.94 nm, 8.426 Å and 598, respectively. From SEM result the magnesium ferrite particle size formed was ranged from 251 nm to 414 nm in diameter for the given calcinations temperature at 1000 °C. The EDS spectra confirm the presence of Fe, Mg and O in the compound. In FT-IR analysis vibrations of ions in the crystal lattice of Mg ferrite observed in the range of 1200-400  $\text{cm}^{-1}$  and the high-frequency band ( $\nu_1$ ) lies in the 650-530  $\text{cm}^{-1}$  range and the low-frequency band ( $\nu_2$ ) in the 446-400  $\text{cm}^{-1}$  range.

**Keywords:** Mg ferrite, Solid state reaction, Structural properties, Crystallite size

## Introduction

Magnetic materials which have combined electrical and magnetic properties are known as ferrites. Iron oxide and metal oxides are the main constituents of the ferrites. Ferrite materials are insulating magnetic oxides and possess high electrical resistivity, low eddy current and dielectric losses, high saturation magnetization, high permeability and moderate permittivity. Ferrites are magnetic oxide materials with semiconducting nature which are of great technological importance by virtue of their interesting electrical and magnetic properties. Ferrites are useful magnetic materials because of their versatility, low cost and high electromagnetic performance over a wide frequency range<sup>1</sup>.

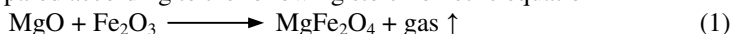
Mg ferrites belong to the normal, or the inverse, spinel structure ferrites group. Magnesium ferrite ( $\text{MgFe}_2\text{O}_4$ ) has a cubic spinel-type structure<sup>2,3</sup>. It is well known as soft magnetic *n*-type semi conductive material, with high resistivity and low magnetic and dielectric losses. Among various ferrites, magnesium ferrite ( $\text{MgFe}_2\text{O}_4$ ) enjoys a special attention because of its vast applications such as one of the candidates for high density magnetic recording<sup>4-8</sup>, microwave absorbents, sensors and electronic device, high frequency devices, color imaging<sup>9-11</sup> and fabrication of radio frequency coils, transformer cores and chock coils, noise filters, recording heading and rod antennas<sup>12,13</sup>. Magnesium ferrites are the potential materials for various applications due to their high electrical resistivity, low magnetic and dielectric losses<sup>14,15</sup>.

Properties of ferrite materials are strongly depend on the distribution of metallic ions among crystallographic crystal lattice sites, composition and micro structures and the method used to prepare those materials<sup>16</sup>.

The main objective of this study was to synthesize and to asses structural and morphological properties of spinel magnesium ferrite using solid state reaction. The employed techniques are thermal analysis using TGA/DTG, x-ray diffraction (XRD), scanning electron micrographs (SEM), energy dispersive x-ray (EDX) and Fourier transform infrared spectroscopy (FT-IR) techniques.

## Experimental

$\text{MgFe}_2\text{O}_4$  (magnesium ferrite) powder was prepared using solid state route at high temperature. Precursors used were of high purity magnesium oxide MgO (98%) and Ferric oxide  $\text{Fe}_2\text{O}_3$  (98%) and both chemicals were supplied from Hi-media Company. The appropriate amount of MgO and  $\text{Fe}_2\text{O}_3$  were prepared according to the following stoichiometric equation



The sample was well ground by adding acetone in order to bring homogeneity between the precursors. The oxides were ground for eight hours according to their proportion to promote better mixing of raw materials. First, the sample was pre-calcined at 800 °C for 5 h and ground for 3 h. Secondly, the sample was calcined at 1000 °C for 6 h and then finally ground for 3 h. For the formation of the Mg ferrite phase, the calcined precursors/powders were cooled at the rate of 5 °C/min in static air atmosphere up to the required calcined temperature and maintained at the temperature for the calcined time in the furnace.

In this work we studied synthesis, structural and morphology of  $\text{MgFe}_2\text{O}_4$  by using solid state method. The employed characterizations were TGA/DTG (thermal events and phase transformation), XRD (phase confirmation), SEM (morphology analysis), EDX (elemental analysis) and FT-IR (bond formation).

Thermal analysis was determined based on the changes in sample weight in relation to changes in temperature and this technique is primarily used for the determination of phase transformation occurring in the materials. In general, the TGA curves were plotted with the percent weight change against temperature.

X-Ray diffraction analysis was performed to identify the phase formation, Lattice constant (*a*), cell volume ( $V_{\text{cell}}$ ) and crystallite size (*D*) of the prepared ferrite materials. The crystalline phase present in the different samples were identified by the powder x-ray diffraction data of the sample which was collected on a PAN analytical x-pert pro diffractometer with diffraction angles of 20° and 90° in increments of 0.008°. The unit cell lattice parameter was obtained by the unit cell software from the 2θ and (hkl) values. The crystallite size of magnesium ferrite present was investigated based on x-ray diffraction line broadening and calculated using Scherrer equation<sup>17</sup>.

$$D = \frac{K\lambda}{\beta \cos \theta}$$

Where  $D$  is the average crystalline size,  $\beta$  is the full width at half maximum of the line broadening of the maximum reflection in radians,  $\theta$  is the angle corresponding to the peak position,  $\lambda$  is the wavelength of x-ray radiation is equal to  $1.542\text{\AA}$ ,  $K$  is the shape factor of average crystallite  $\sim 0.9$ .

The average lattice constant value ( $a$ ) was obtained using  $2\theta$  values of the most intense peaks using Bragg's diffraction<sup>18</sup> condition, given by

$$a = \frac{\lambda \sqrt{h^2 + k^2 + l^2}}{2 \sin \theta}$$

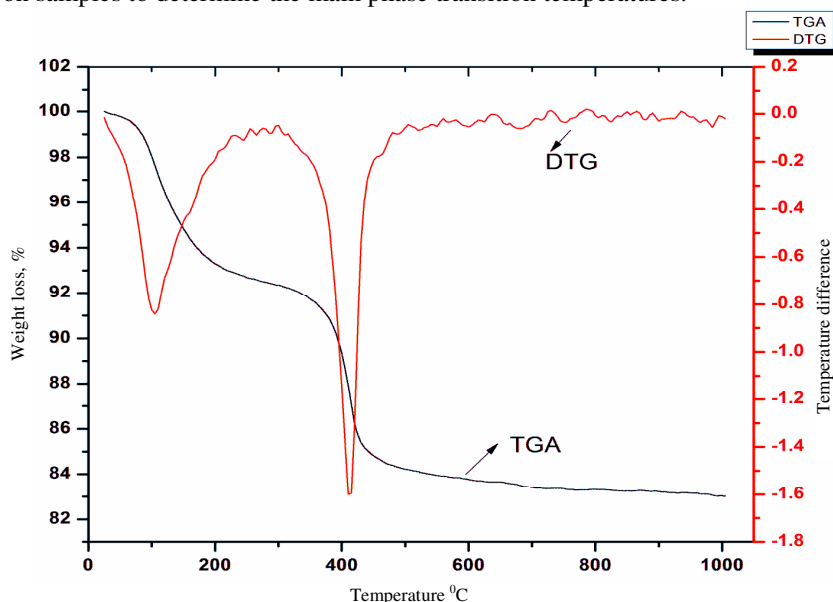
The particle morphology and elemental analysis of the powders were observed using scanning electron microscopy and energy dispersive spectra (EDS) taken from JEOL JSM-6610LV connected with Inca-Penta FETx3.JPG, Oxford Instruments.

Fourier transform infrared spectra were obtained on a Shimadzu IR-Prestige 21 spectrometer using KBr pellet technique and the wavenumber range between 400 and  $1200\text{ cm}^{-1}$ .

## Results and Discussion

### TGA/DTG analysis

The TGA/DTG measurements were conducted using Mettler Toledo TGA/DTG 851e instrument from room temperature to  $1000\text{ }^{\circ}\text{C}$  in nitrogen atmosphere at a heating and cooling rate of  $10\text{ }^{\circ}\text{C/min}$  as shown in Figure 1. TG measurements were performed on samples to determine changes in weight in relation to change in temperature. DTG is also done on samples to determine the main phase transition temperatures.

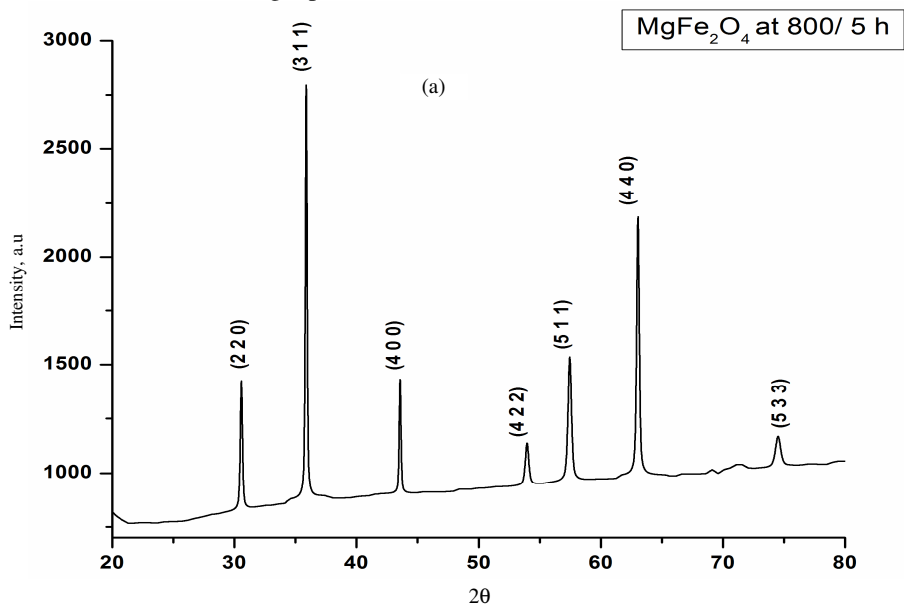


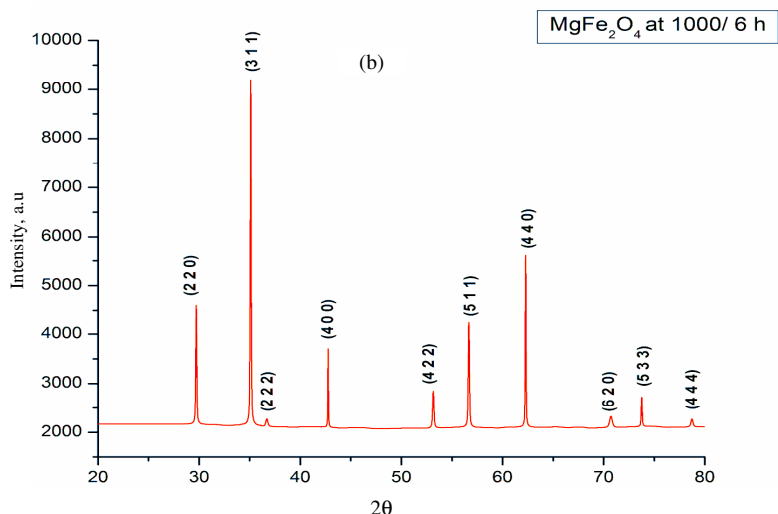
**Figure 1.** TGA/DTG pattern for the synthesized  $\text{MgFe}_2\text{O}_4$

As can be seen from the curves, there is an initial weight loss in the temperature range from room temperature to 350 °C (about ~7%). This corresponds to the evaporation of methanol used during grinding to homogenize the mixture the moisture absorbed during storage<sup>19</sup>. TGA curve of  $\text{MgFe}_2\text{O}_4$  shows significant weight loss between the temperatures 350 °C and 500 °C (about 10%). This loss was attributed to the decomposition of the precursors  $\text{MgO}$  and  $\text{Fe}_2\text{O}_3$  and the reaction between the decomposed materials in order to produce crystalline  $\text{MgFe}_2\text{O}_4$ . This is supported by sharp peak observed at 400 °C on the DTG curve. At higher temperatures the TGA curve becomes flattened, indicating the stable phase formation. Above this temperature, negligible weight loss indicated that the thermal events terminated beyond this temperature. Although the desired cubic spinel structure formation has been started earlier at 800 °C but complete single-phase is formed at higher temperature of 1000 °C which agrees with proposed synthesis temperature.

### *XRD analysis*

The XRD pattern of as synthesized  $\text{MgFe}_2\text{O}_4$  prepared powder by solid state reaction method. XRD patterns of powdered samples of  $\text{MgFe}_2\text{O}_4$  are illustrated in Figure 2a,b at different temperature. The diffraction peaks closely correspond to the standard pattern (Figure 2, JCPDS card number 17-464). The crystallite sizes of  $\text{MgFe}_2\text{O}_4$  samples were calculated from x-ray lines broadening of the reflections of (220), (311), (222), (400), (442), (511), (440), (533) and (444) using Scherrer's equation ( $D=44.94$  nm). The data were collected in a  $2\theta$  range from  $20^\circ$  to  $80^\circ$  with a  $0.008^\circ$  step size. This significant change in intensity indicates the formation of the ferrite phase which has its most intense (311) peak at  $2\theta=35.099^\circ$ . All the diffractograms showed the characteristic reflections of the spinel phase. Figure 2 also shows that the peaks are sharper and narrower with pre-calcination temperature of 800 °C (Figure 2a) followed by 1000 °C (Figure 2b) calcination temperature, the crystalline nature of the sample. No impurity phase peaks can be observed on XRD pattern at 1000 °C, indicating that the synthesized samples are single phase. But impurity is observed and there is no single phase formation at 800 °C.





**Figure 2.** The x-ray diffraction pattern of  $\text{MgFe}_2\text{O}_4$  at (a) 800 °C and (b) 1000 °C

It is found that crystal structure to be cubic with space group (SG)  $\text{Fd-3m}^{20}$  and lattice parameter was found to be 8.426 Å and cell volume is 598. The obtained lattice constant data was slightly larger than reported data of  $\text{MgFe}_2\text{O}_4$  from JCPDS file (pattern: ICDD-01-073-1720:  $\text{Fd-3m}$ ) which is crystallite size is 47 nm, lattice parameter is 8.384 and 589.3 cell volume by using Micro emulsion method<sup>21</sup>. Hankare *et al*<sup>22</sup>, has reported the value of lattice parameter as 8.37 Å. The deviation in lattice parameter can be attributed to the cation rearrangement in the nano sized<sup>22,23</sup>  $\text{MgFe}_2\text{O}_4$ . According to stoichiometric equation (Eq.1) the appropriate amount of MgO and  $\text{Fe}_2\text{O}_3$  for the preparation of  $\text{MgFe}_2\text{O}_4$  is alcinati in Table 1.

The calculated values of the lattice constant (a), unit cell volume (V) and crystallite size (D), of  $\text{MgFe}_2\text{O}_4$  particles depending upon the data of x-ray are summarized in Table 2.

**Table 1.** Preparation of Mg ferrite with total of 15 g

Compound	First precursor amount in gram/mol	Second precursor amount in gram/mol
$\text{MgFe}_2\text{O}_3$	MgO 3.0223	$\text{Fe}_2\text{O}_3$ 11.9761

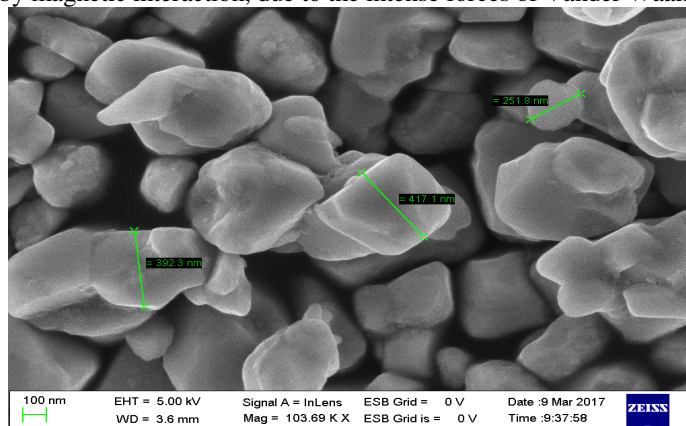
**Table 2.** Structural parameters for the prepared Mg-Fe-O system

Sample	a (Å)	V (Å <sup>3</sup> )	D, nm
$\text{MgFe}_2\text{O}_4$	8.426	598	44.94

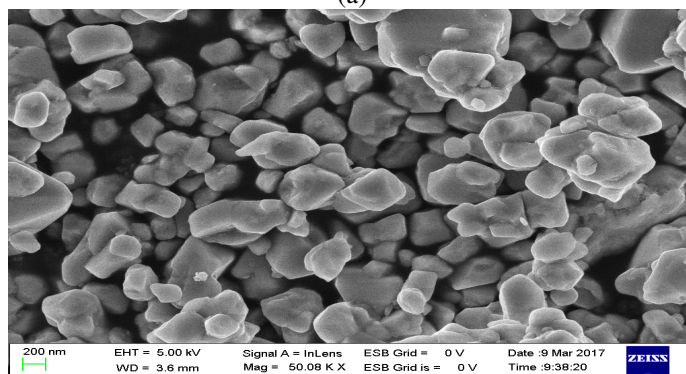
### SEM analysis

The surface and grain morphology of spinel magnesium ferrite is studied by scanning electron microscopy calcined at 1000 °C. Figure 3a to Figure 3c shows the FESEM image different magnification at 103.69KX, 50.08KX and 10.10KX. The particle size formed magnesium ferrite was ranging from diameter of 251.8 to 417. 1 nm for the samples alcinations at 1000 °C. The SEM images showed the surface morphology and grain size for the prepared samples. It is clear from the images that uniformly distributed; less agglomerated and homogenous grains have been formed. The SEM micrograph showed that spherical small particles in addition to large particles, which indicated that the Mg ferrite

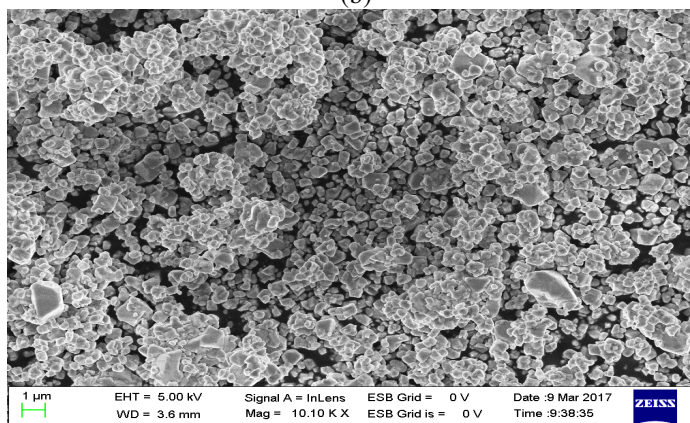
was sufficient to produce a single phase with homogeneous microstructure and uniform size at 1000 °C. Mg ferrite sample are magnetic in nature, so the particles are less agglomerated and held together by magnetic interaction, due to the intense forces of Vander Waals attraction<sup>24</sup>.



(a)



(b)

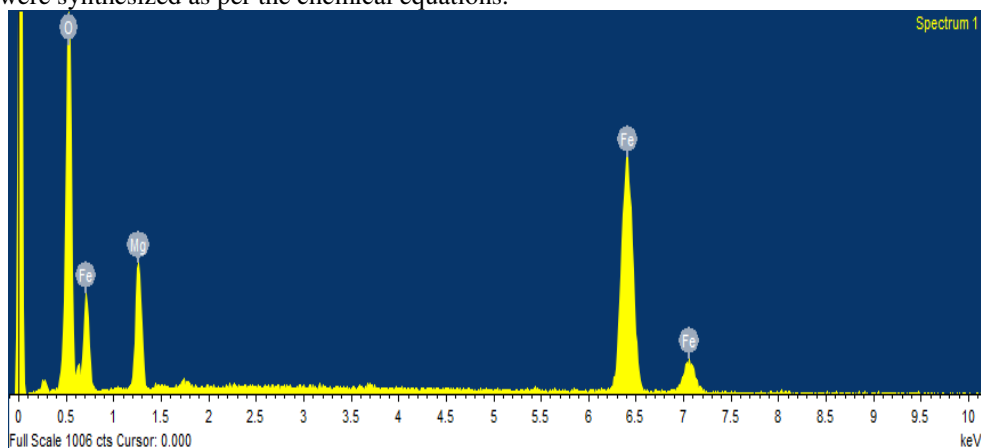


(c)

**Figure 3.** SEM images of Mg ferrites at different magnifications at (a) 103.69KX, (b) 50.08KX and (c) 10.10KX

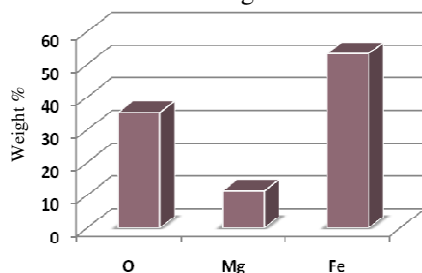
### Energy dispersive spectroscopy analysis

It is observed from the EDS spectra, as shown in Figure 4a, that the targeted compositions were synthesized as per the chemical equations.



**Figure 4(a).** EDS spectra of Mg ferrite

The EDS patterns quantitatively describe the presence of Fe, Mg and O elements in  $\text{MgFe}_2\text{O}_4$  ferrite material. The EDS analysis accurately provided estimation of the elemental concentrations as per the specifications mixed at the time of preparation. EDS patterns quantitatively describe the presence of Fe, Mg and O elements in  $\text{MgFe}_2\text{O}_4$  ferrite material is listed in the Table 3 & Figure 4b.



**Figure 4(b).** Bargraph of quantitative results

**Table 3.** Result of elemental analysis

Element	Weight%	Atomic%
O	35.30	60.80
Mg	11.39	12.91
Fe	53.30	26.30
<b>Total</b>	100.00	

### FT-IR analysis

Figure 5 shows FT-IR spectra of the synthesized sample prepared by solid state reaction method calcined at 1000 °C for 6 h. In order to validate the results of XRD analysis, the room temperature FT-IR spectra of the synthesized sample was performed. The Fourier transform infrared absorption spectra of the prepared sample were recorded in the wavenumber range  $1200\text{ cm}^{-1}$  to  $400\text{ cm}^{-1}$  with potassium bromide as binder. Two ranges of the absorption bands are observed from the FT-IR graph. These are the high-frequency band ( $\nu_1$ ) lies in the  $650\text{--}530\text{ cm}^{-1}$  range and the low-frequency band ( $\nu_2$ ) in the  $446\text{--}400\text{ cm}^{-1}$  range. The higher frequency band ( $\nu_1$ ) is due to the stretching intrinsic vibration of unit cell of the spinel in the tetrahedral (A) site and the lower band ( $\nu_2$ ) is caused by stretching vibration of metal(Fe)-oxygen (O) in octahedral (B) site<sup>25</sup>, which are the typical bands of the spinel structure. This difference in the band position is attributed to the vibration of tetrahedral and octahedral components in the spinel ferrites<sup>26-28</sup>.

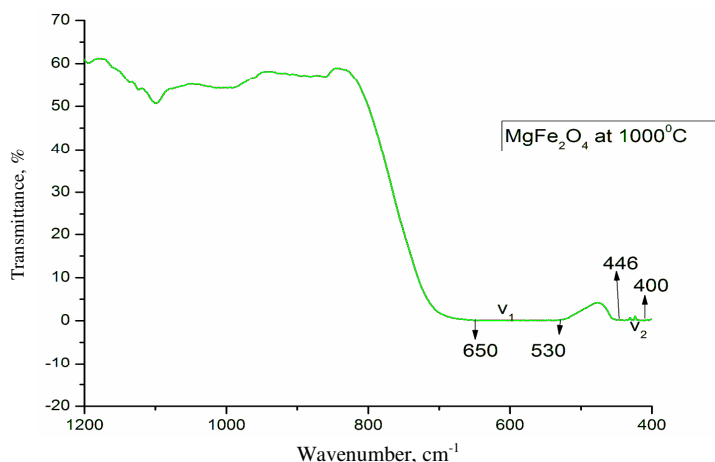


Figure 5. FT-IR spectra of  $\text{MgFe}_2\text{O}_4$  calcined at 1000 °C

## Conclusion

The powder of Spinel  $\text{MgFe}_2\text{O}_4$  ferrite was prepared by solid state method and well characterized by Thermal analysis, X-ray diffraction, Scanning electron microscopy and Fourier transform infrared spectroscopy. The result can be summarized as:

- The sample was pre-calcined at 800 °C for 5 hours at this stage impurity appear and there is no single phase observed.
- From powder calcined at 1000 °C for 6 hours we observed no impurity and single phase spinel structure.
- Single phase of crystalline  $\text{MgFe}_2\text{O}_4$  was formed by precursors  $\text{MgO}$  and  $\text{Fe}_2\text{O}_3$  at calcination temperature 1000 °C and Spinel Mg ferrite belongs to Fd-3m space group.
- The sample were found to be cubic spinel structure which is confirmed by XRD with crystalline size, lattice constant and cell volume was 44.94 nm, 8.426 Å and 598 respectively.
- From SEM result the particle size formed magnesium ferrite was ranging from diameter of 251 to 417 nm for the given calcination temperature at 1000 °C.
- The EDS patterns quantitatively describe the presence of Fe, Mg and O elements in  $\text{MgFe}_2\text{O}_4$  ferrite material.
- In FT-IR analysis vibrations of ions in the crystal lattice of Mg ferrite observed in the range of 1200-400  $\text{cm}^{-1}$  and the high-frequency band ( $\nu_1$ ) lies in the 650-530  $\text{cm}^{-1}$  range and the low-frequency band ( $\nu_2$ ) in the 446-400  $\text{cm}^{-1}$  range.

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