

STUDIES ON EFFECT OF YTTERBIUM SUBSTITUTION IN ZINC FERRITE

Zar Zar Myint Aung¹, Soe Soe Han², Aye Aye Thant³

Abstract

Rare earth, ytterbium doped Zinc ferrites ($Zn(Yb_xFe_{1-x})_2O_4$) with $x=0.0000, 0.0125, 0.0250, 0.0375$ and 0.0500 have been prepared by the solid state method. The raw materials such as ytterbium oxide, zinc oxide and ferrite oxide have been used as the source materials. The spinel ferrite structure, cubic morphology, and the identification of functional groups of the ytterbium doped zinc ferrite have been analyzed systematically using several analytical tools. The resistivity decreases in the sample without RE dopants for both frequency regions of (10 kHz-100 kHz) and (1MHz-10MHz). Moreover, rare-earth doped samples exhibit high conductivity than undoped zinc ferrite. The electrical conductivity at room temperature is directly proportional to the reaction rate of samples.

Keywords: *X-ray diffraction, Scanning electron microscopy, electrical properties*

Introduction

Spinel ferrites are interesting materials owing to their wide range of applications in modern science and technology. They have recently attracted considerable research interest on their structural, magnetic and electrical properties. These structures are attractive for microwave applications, magnetic sensors and catalytic materials owing to their great magnetic permeability and dielectric constant, low dielectric loss, high Curie temperature as well as mechanical strength and chemical stability at low frequencies. In addition, their magnetic properties can be controlled and tailored to practical applications through the appropriate choice from a number of divalent cations in their structure.

Zinc Ferrite has many applications in high- frequency devices, and they play a useful role in technological and magnetic applications.

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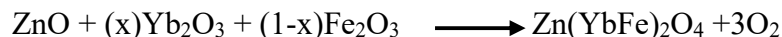
The electrical conductivity and dielectric behavior of spinel ferrites are very sensitive to the type of substituent and sintering conditions, such as temperature, time and heating rate. The aim of the present work was to study the effect of preparation conditions on ZnFe_2O_4 by solid state method in the first step. In the second step, RE ytterbium ions have been doped in ZnFe_2O_4 and investigate the effect of RE ytterbium ions on the properties of zinc ferrite.

Material and Method

Experimental procedure

Rare earth doped Zinc ferrites have been prepared ($\text{Zn}(\text{YbxFe}_{1-x})_2\text{O}_4$) with $x=0, 0.0125, 0.0250, 0.0375$ and 0.0500 by the solid state method. The raw materials such as ytterbium oxide, zinc oxide and ferrite oxide were used as the source materials. The required amount of the ZnO , Yb_2O_3 and Fe_2O_3 has been weighed with digital balance. The raw materials of ZnO , Yb_2O_3 and Fe_2O_3 have been taken in stoichiometric proportions.

These oxide materials were mixed with standard weight percentages according to their stoichiometric calculation by using the following equation.



All compositions have been mixed in an Agate mortar and ground for 4 h. After mixing and grinding, the mixture has been pre sintered at 900°C for 5 h in a furnace with heating rate of $20^\circ\text{C}/\text{min}$ and cooled to room temperature with the same rate. After that, the powder and the mixture has been ground with an Agate motor for 1h. Then, the powder has been pressed into pellets and toroids by uniaxial hydraulic press at a pressure of 5ton. The pellets have been well polished for further characterizations.

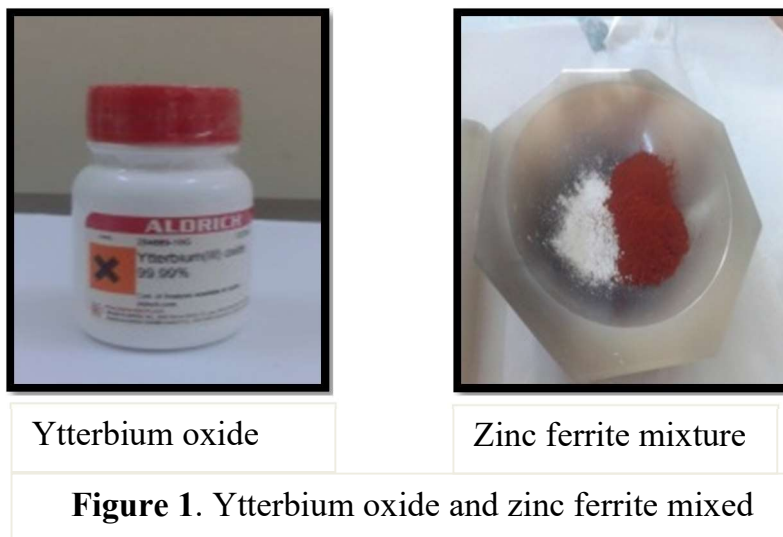
The other publication papers were $\text{MgZnFe}_2\text{O}_4$, $\text{NiZnFe}_2\text{O}_4$, $\text{CuZnFe}_2\text{O}_4$ and ZnFe_2O_4 ferrites prepared by their different temperatures. The $\text{MgZnFe}_2\text{O}_4$ was presintered at 800°C for 3 hours and final sintered at 1225°C for 6 hours in a furnace. The $\text{NiZnFe}_2\text{O}_4$ was presintered at 1100°C for 4 hours and final at 1275°C for 30mins. The $\text{CuZnFe}_2\text{O}_4$ presintered was 900°C for 3 hours and final was 1000°C for 6 hours. According to this ferrites

papers, zinc ferrite has been obtained a single phase in the temperature range of 900 °C – 1200 °C.

In addition, the solid state process requires sintered temperature more than 750 °C for phase formation and sintering temperature more than 1000 °C to achieve better densification. The influence of rare earth ions like Yb, Er, Dy, Tb, Gd, Sm substitution on structure, magnetic and electrical properties of (Zn) Yb Fe₂O₄ ferrites. Thus, ZnYbFe₂O₄ was chosen presintered at 900 °C for 5 hours and final sintered for 1000 °C and 1100 °C. The various final sintered of the sample with rare earth(pellets) dopants have been characterized by XRD and SEM. The experimental results of (Zn(Yb_xFe_{1-x})₂O₄) with x=0, 0.0125, 0.0250, 0.0375 and 0.0500 are compared to the undoped and ZnFe₂O₄ their electrical properties are investigated.

Rare Earth Elements

The rare earth elements include 15 lanthanides with the atomic numbers 57 to 71 in the periodic table. In the Figure (1) Ytterbium is a chemical element with symbol Yb and atomic number 70, which is the basis of the relative stability of its 2⁺ oxidation state. Ytterbium can be used as a dopant to help improve the grain refinement, strength, and other mechanical properties stainless steel.





(a)



(b)

Figure 2(a) Ytterbium oxide and zinc ferrite applied with agate motor and pestle

(b) The pellets and toriods prepared by uniaxial hydraulic press at a pressure of 5tons

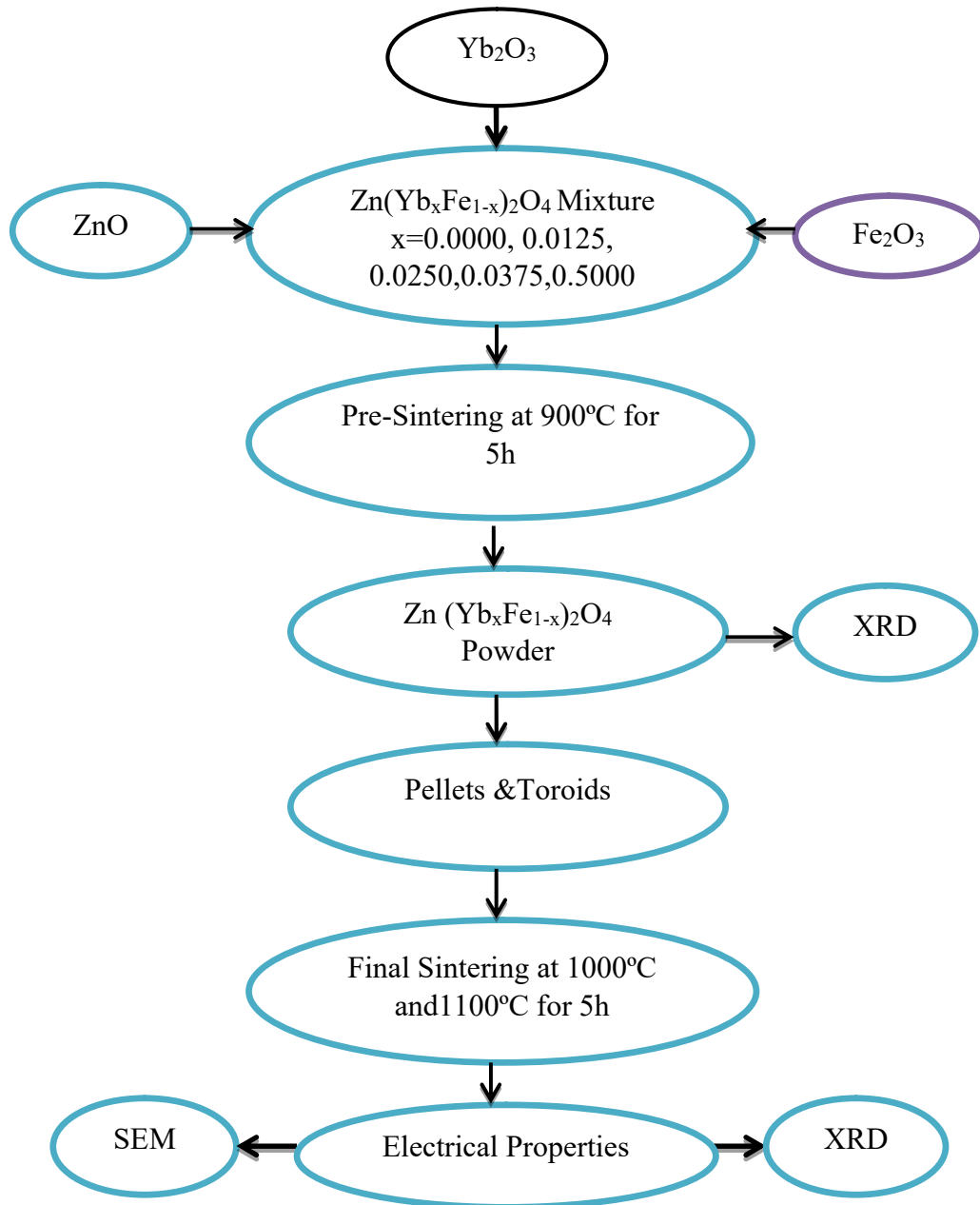


Figure 3. Flow chart of the sample preparation for Yb doped zinc ferrite

Results and Discussion

XRD Analysis

Figure 4 to 5 show XRD patterns of the as prepared and sintered samples of $\text{ZnYbFe}_2\text{O}_4$ nano particles respectively. The XRD peaks indicate the occurrence of pure spinel ferrite structure, which is represented by the reflections from the (2 2 0), (3 1 1) and (4 0 0) planes and it was compared with standard JCPDS patterns. The crystallite sizes of the $\text{ZnYbFe}_2\text{O}_4$ nanoparticles sintered at different temperatures were calculated using the Scherrer's formula, and are listed in table 1. As expected, the crystallite size increased with increasing heat treatment temperature

(1000 °C and 1100 °C) for all sample examined, ranging from 36 to 51 nm. The increasing crystallite size with increasing calcinations temperature agrees well with earlier results.

Figure 6 shows the morphology of the $\text{ZnYbFe}_2\text{O}_4$ nanoparticles at different temperatures. The micrographs clearly indicate the cubic morphology of the nanocrystals. In addition, both the crystallite size and the degree of particle agglomeration increased with increasing sintered temperature, probably because of the disappearance of the polymer at high temperatures, which is in good agreement with XRD results.

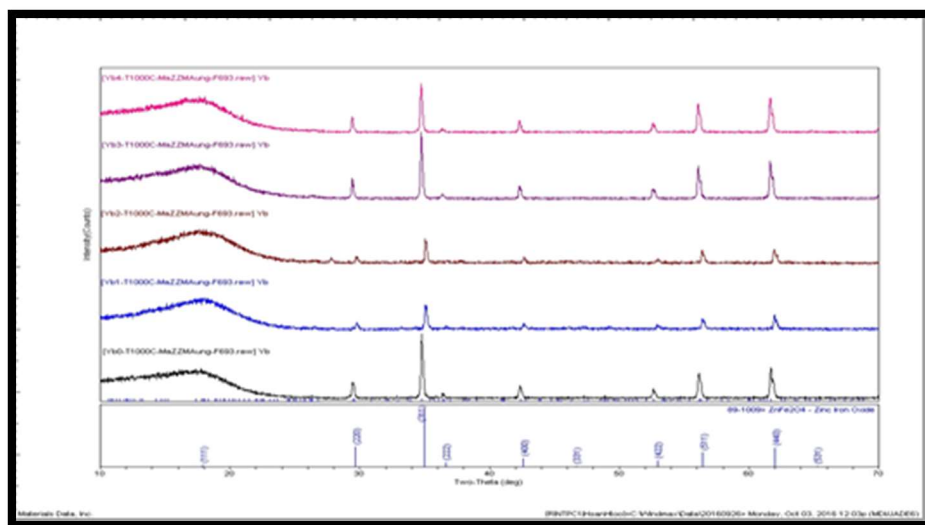


Figure 4. XRD pattern nano-particles of $\text{ZnYbFe}_2\text{O}_4$ at 1000 °C

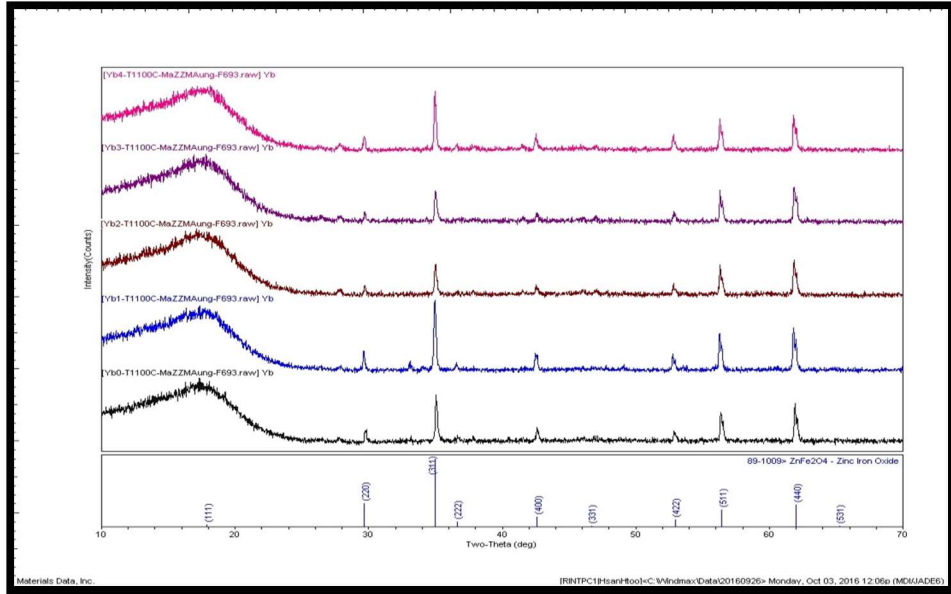


Figure 5. XRD pattern nano-particles of ZnYbFe₂O₄ at 1100 °C

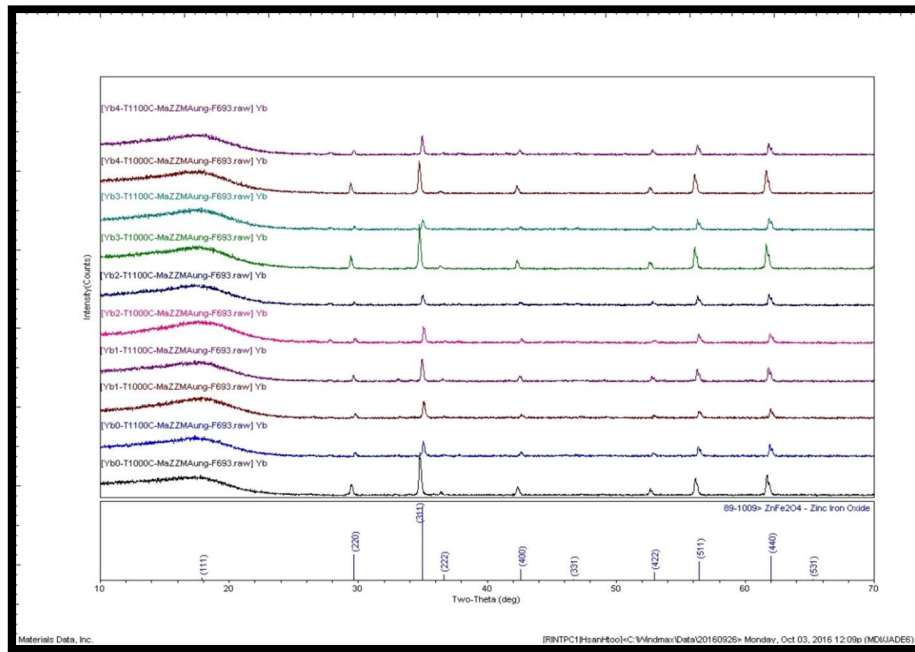


Figure 6. XRD pattern of the ZnYbFe₂O₄ samples at different temperatures

Table 1. Crystallite size of the ZnYbFe₂O₄ samples at different temperatures

| Sample | Temperature (°C) | Crystallite size (nm) |
|------------|-------------------|-----------------------|
| x = 0.0000 | 1000 | 44.02 |
| | 1100 | 42.46 |
| x = 0.0125 | 1000 | 36.78 |
| | 1100 | 43.80 |
| x= 0.0250 | 1000 | 44.29 |
| | 1100 | 44.76 |
| x = 0.0375 | 1000 | 47.30 |
| | 1100 | 41.18 |
| x = 0.0500 | 1000 | 51.77 |
| | 1100 | 49.91 |

SEM analysis

The SEM images of undoped and rare earth (ytterbium) doped ZnFe₂O₄ pellets are shown in figures with various temperature. The pellets sintered at 1000°C showed an average particle size in the range from 0.9231µm to 7.2910µm in figure 7 whereas for the pellets sintered at 1100°C, the average particle size was in the range from 0.8056µm to 1.03125µm in figure 8. The particle size correlation of the results obtained by XRD and SEM indicates that the agglomerates are made of nanosized particles. Thus, the above discussion confirms the formation of ZnYbFe₂O₄ phase particles between temperatures of 1000°C and 1100°C. The estimate values of average grain size are presented in Table 3 and 4. It is observed that the grain size of undoped- ZnFe₂O₄ is the biggest in all samples. The grain size of Yb doped ZnFe₂O₄ ferrites are gradually decreased until x= 0.0500.

Table 2. The average grain size of the $\text{YbZnFe}_2\text{O}_4$ pellets at 1000 °C temperatures

| Composition of rare earth ions | Grain Size |
|--------------------------------|----------------------|
| x = 0.0000 | 7.2910 μm |
| x = 0.0125 | 5.4062 μm |
| x = 0.0250 | 4.4313 μm |
| x = 0.0375 | 3.7669 μm |
| x = 0.0500 | 0.9231 μm |

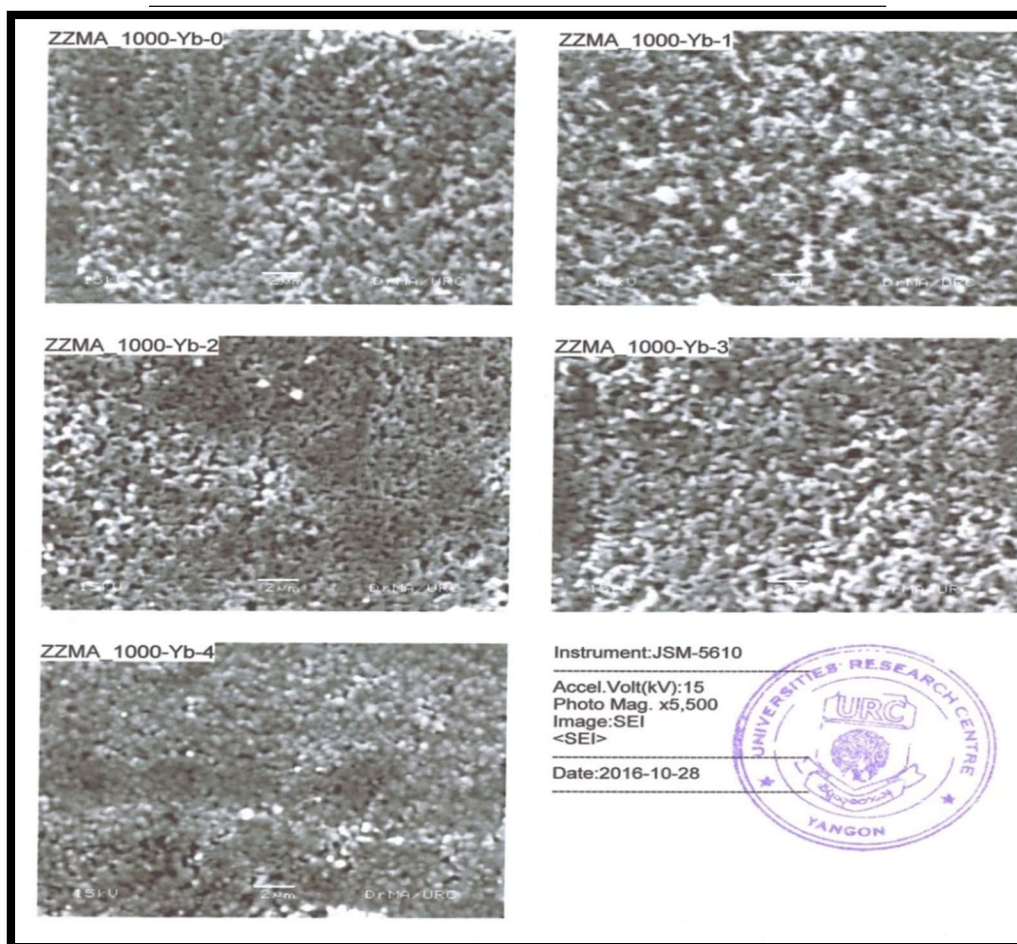


Figure 7. The SEM photograph of the $\text{YbZnFe}_2\text{O}_4$ pellets at 1000 °C temperature

Table 3. The average grain size of the $\text{YbZnFe}_2\text{O}_4$ pellets at 1100 °C temperatures

| Composition of rare earth ions | Grain Size |
|--------------------------------|-----------------------|
| x= 0.0000 | 1.03125 μm |
| x= 0.0125 | 0.8056 μm |
| x= 0.0250 | 0.8772 μm |
| x= 0.0375 | 0.8116 μm |
| x= 0.0500 | 0.6867 μm |

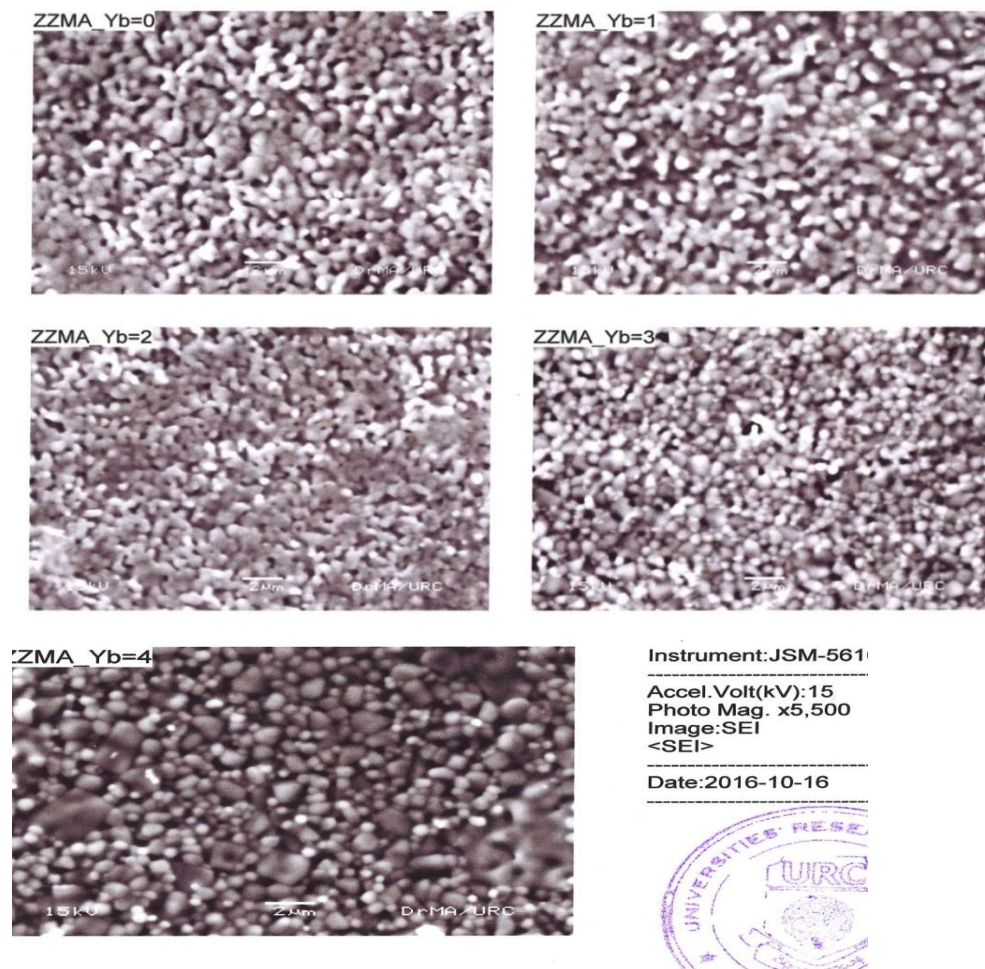


Figure 8. The SEM photograph of the $\text{YbZnFe}_2\text{O}_4$ pellets at 1100 °C temperature

Electrical Properties

Variation of Resistivity with Frequency and Sintered Temperature

The frequency dependent variation of DC resistivity (the inverse of conductivity) for all samples which are sintered at 1000 °C at lower frequency range (10kHz-100kHz) and higher frequency range (1MHz-10MHz) are shown in Fig 9 and Fig 10 respectively. It was found that the resistivity decreases in the sample without RE dopants in both frequency regions. The resistivity increases with increasing RE doping in the sample.

Again the frequency dependent variation of DC resistivity for all samples which are sintered at 1100 °C at lower frequency range (10 kHz-100 kHz) and higher frequency range (1MHz-10MHz) are shown in Fig 11 and Fig 12 respectively. The variation of resistivity shows the same trend as the result in the samples which are sintered at 1000 °C.

By comparing resistivity for different sintering temperatures, it is worth to note that the resistivity decreases with increase in sintering temperature. It has been attributed that the grains becomes bigger at higher sintering temperature and provide higher conductivity.

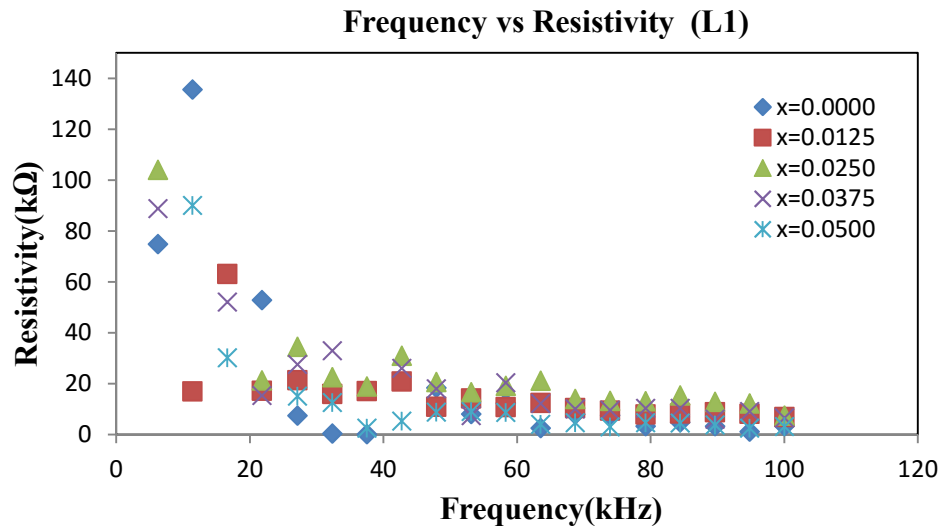


Figure 9. Frequency (10 kHz-100 kHz) dependent resistivity curves for ZnYbFe₂O₄ with different Yb content at 1000°C

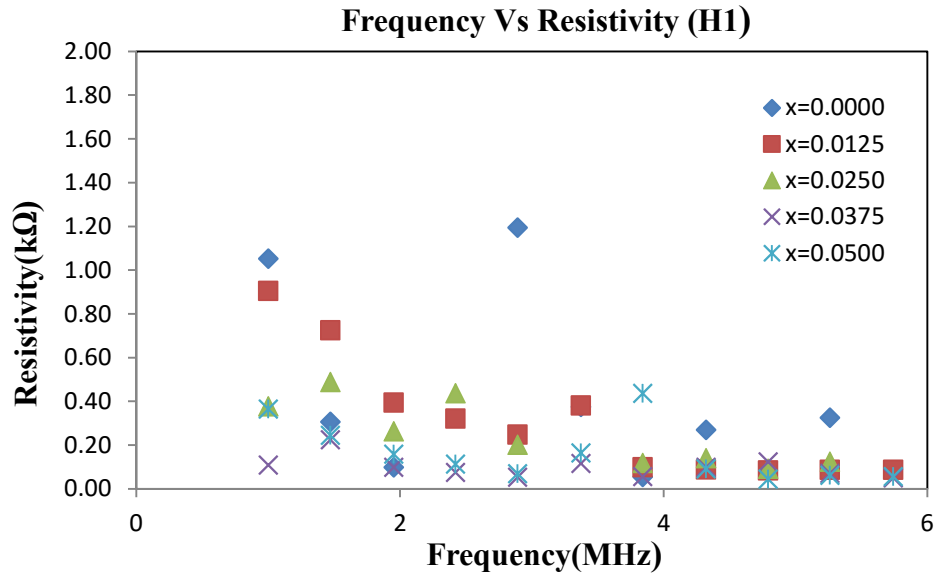


Figure 10. Frequency (1MHz-10MHz) dependent resistivity curves for ZnYbFe₂O₄ with different Yb content at 1000°C

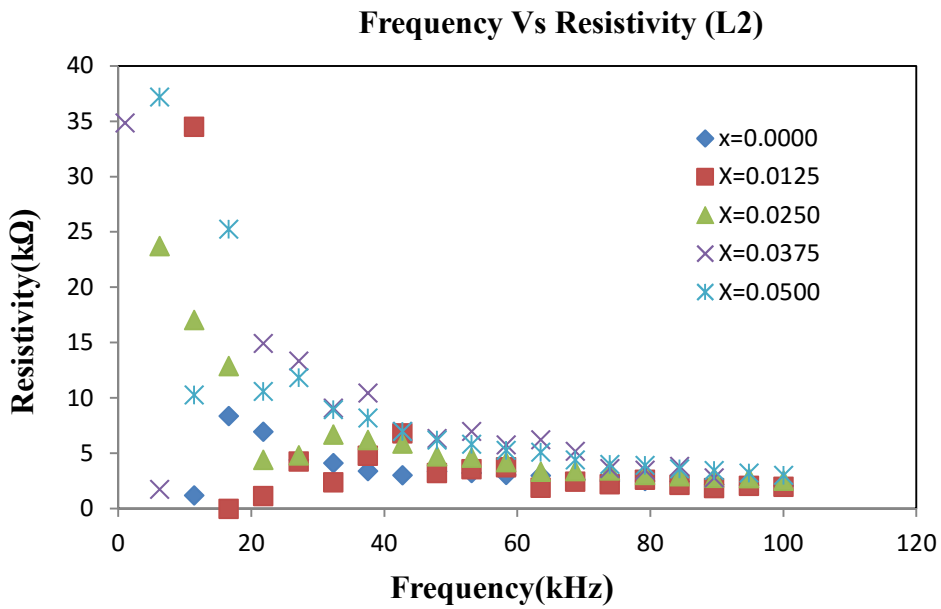


Figure 11. Frequency (10 kHz-100 kHz) dependent resistivity curves for ZnYbFe₂O₄ with different Yb content at 1100°C

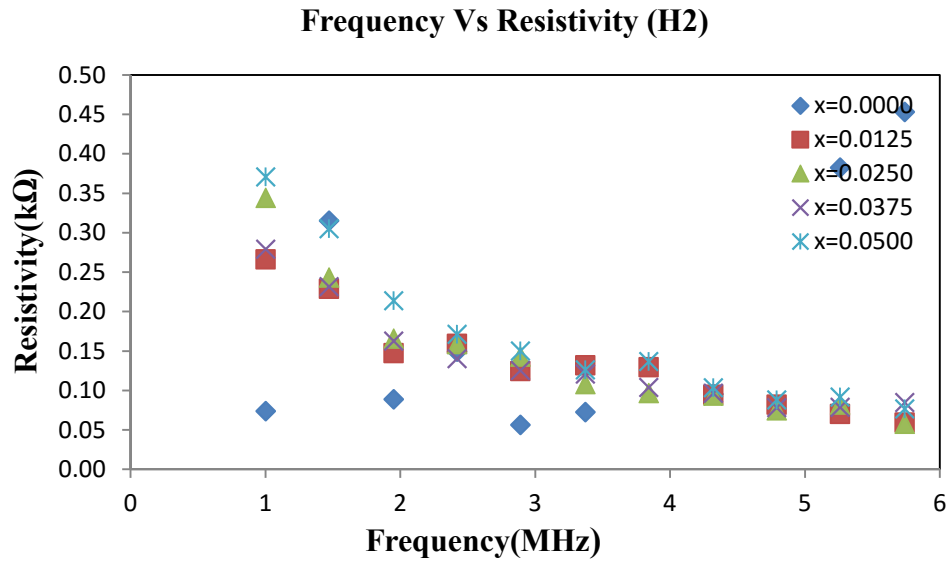


Figure 12. Frequency (1MHz-10MHz) dependent resistivity curves for ZnYbFe₂O₄ with different Yb content at 1100°C

Conclusion

From the above experimental results, it is clearly evident that the nano size of the ferrite particles in Yb doped zinc ferrite ZnFe₂O₄ has been obtained by using solid state method. The microstructure with fine grains has been observed to evaluate the applicability of the ferrite. Moreover, the grain size which is less than 1µm confirmed the formation of the smaller grains under the sintering temperature of 1000°C and 1100°C. Therefore, it is concluded that ZnFe₂O₄ with the homogeneous microstructure has been formed for further investigation with RE dopants.

In this work has been reported a study on the influenced of ytterbium doped on structure, morphology and electrical properties of the Zn (Yb_xFe_{1-x})₂O₄ (x=0.0000, 0.0125, 0.0250, 0.0375 and 0.0500). The resistance and capacitor have been measured in the frequency ranges (10 kHz-100 kHz) and (1MHz-10MHz) with different temperatures 1000°C and 1100°C respectively. The resistivity decreases in the sample without RE dopants in both frequency regions. The resistivity increases with increasing RE doping in

the sample. Moreover, the resistivity decreases with increase in sintering temperature. It has been attributed that the grains becomes bigger at higher sintering temperature and provide higher conductivity. This is proved that the substitution of small amount of RE ions in ferrite can tune the electrical properties.

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