

## Dielectric Properties of $Y_3Fe_5O_{12}$ (YIG) Prepared at Different Molarities of NaOH

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Doi : <https://doi.org/10.47011/13.3.5>

*Received on: 08/08/2019;*

*Accepted on: 23/02/2020*

**Abstract:** In this study, Yttrium Iron Garnet ( $Y_3Fe_5O_{12}$ ) (YIG) powders were synthesized *via* co-precipitation method, followed by calcining the precipitates at 1100°C. The garnets produced were obtained from aqueous iron and yttrium chloride mixtures using different molarities of NaOH (M=2, 3, 4 and 5) at pH=12. The phase formation and crystallography were investigated using X-ray diffraction (XRD), the morphology was investigated using transmission electron microscopy (TEM) and the dielectric properties were measured using an impedance analyzer in the frequency range 0.5 - 5MHz, in a temperature range 22 - 350°C. X-ray diffraction peaks showed the formation of cubic YIG with lattice parameter varying between 12.334 and 12.339 Å. The grain size, measured from TEM images, decreased with the increase of the molarity of NaOH. Plots of the real part of the dielectric constant  $\epsilon'$ , the imaginary part of the dielectric constant  $\epsilon''$ , loss tangent  $\tan \delta$  and ac conductivity  $\sigma_{ac}$  as functions of frequency and temperature, respectively, were obtained. It was observed that the highest values of the dielectric constant were obtained in the 2M sample.

**Keywords:** Dielectric properties, Yttrium Iron Garnet (YIG), Co-precipitation method, NaOH molarity.

### Introduction

Yttrium Iron Garnet ( $Y_3Fe_5O_{12}$ ) (YIG) belongs to a class of magnetic oxides which is the garnets. It has 160 unit cells in 3 different sites that are octahedral, dodecahedral and tetrahedral. The importance of YIG resides in the fact that it is of technological significance due to its various properties. It possesses the highest quality factor in the microwave regime, the smallest linewidth in magnetic resonance among the magnetic materials, controllable high saturation magnetization, large Faraday rotation and low propagation loss [1-2]. These, in addition to other properties, have made it a basis for many high-technology devices, such as telecommunication devices, lasers, phase shifters, sensors and oscillators [3]. Specifically, the dielectric properties of YIG, such as its low dielectric loss tangent in the microwave region,

have grabbed a lot of attention and made it possible for the YIG to be used in electronic devices as tunable microwave devices. For this reason, the manipulation of dielectric properties of YIG under various conditions in different ranges has been a major research topic recently. For instance, Ramesh et al. [4] studied the effect of the sintering temperature on the magnetic and dielectric properties of YIG, while Ahmad et al. [5] mentioned that doping YIG by samarium (Sm) has an effect on its dielectric permittivity. Several techniques have been used to prepare YIG powders, including sol-gel [6], Pechini method [7], metallo-organic decomposition [8] and chemical co-precipitation [9]. These top-down techniques are all capable of producing YIG samples; however, the Pechini method results in the production of large-sized particles

and the metallo-organic YIG has a high percentage of impurities. In our work, coprecipitation is chosen to prepare YIG based on a variety of parameters that can be studied, as well as on cheap cost and simplicity. The aim of this work is to investigate the dielectric properties of YIG prepared using different molarities of NaOH during the titration process to reach pH = 12, as there are no reports yet of a similar study.

## Experimental

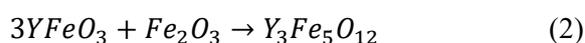
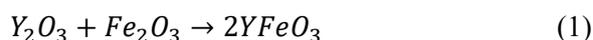
Polycrystalline Yttrium Iron Garnet (YIG) samples were synthesized using the coprecipitation method.  $YCl_3 \cdot 6H_2O$  and  $FeCl_3 \cdot 6H_2O$  were weighed in a stoichiometric ratio Y:Fe = 3:5 and dissolved in de-ionized water to form an aqueous solution of chlorides. This solution was titrated drop-wise with stirred NaOH solutions of different molarities (2, 3, 4 and 5M) to reach pH = 12. Then it was heated at 90°C for 4 hours with constant-speed stirring. The obtained mixtures were then washed with distilled water followed by drying for 18 hours at 100°C. Subsequently, the dried precipitates were ground and moved to a tube furnace to be calcinated at 1100°C for 2 hours.

The phase formation and crystallography were studied using X-ray powder diffraction (XRD) and the morphology was studied using Transmission Electron Microscopy (TEM). The dielectric studies were carried out by an LCR meter in the frequency range 0.5 - 5MHz, in the temperature range 22 - 350°C.

## Results and Discussion

### Structural Analysis of YIG

Fig. 1 shows XRD patterns of YIG powders prepared at different molarities of NaOH (2, 3, 4 and 5M). The diffraction peaks of the  $Y_3Fe_5O_{12}$  (JCPDS: 21-1450) [10] phase indicate that YIG crystallized into a cubic structure with space group  $Ia3d^5$ . The XRD patterns showed the existence of a second phase; the Yttrium Iron Perovskite ( $YFeO_3$ ) (JCPDS: 39-1489) [11]. From the XRD results, the garnet formation can be described as follows [12]:



The reason behind the existence of  $YFeO_3$  could be shown in the equations of formation above. The formation of  $YFeO_3$  takes place before the formation of the YIG phase and its existence after calcination is attributed to the fact that the samples were calcinated at a temperature below 1300°C [13]. The lowest intensity of the peak (121) corresponding to  $YFeO_3$  was observed in the 3M sample. The values of the lattice parameter ( $a$ ) were calculated by indexing the XRD patterns and using the equation  $a = d_{hkl} \sqrt{h^2 + k^2 + l^2}$ , where  $d_{hkl}$  is the lattice spacing and  $h$ ,  $k$  and  $l$  are the Miller indices. It is shown in Table 1 that the increase of the molarity showed a slight effect on the lattice parameter, where its value varied between 12.334 and 12.339 Å.

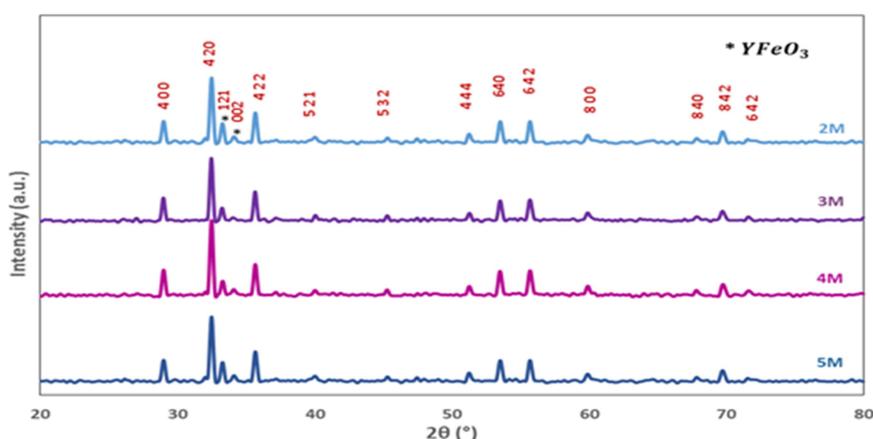


FIG. 1. XRD patterns of YIG powders prepared at different molarities of NaOH.

Fig. 2 shows the TEM photographs of the YIG samples prepared with 2, 3 and 5M of NaOH. The particles show an irregular shape

which could be resolved by increasing the temperature of calcination, whereas the agglomeration shown could be due to the piling

up of a few nanocrystals on the large-sized grains [14]. The average grain size of the particles obtained from TEM photographs

decreases from 163.42 nm to 91.09 nm as the molarity increases from 2M to 5M, as shown in Table 1.

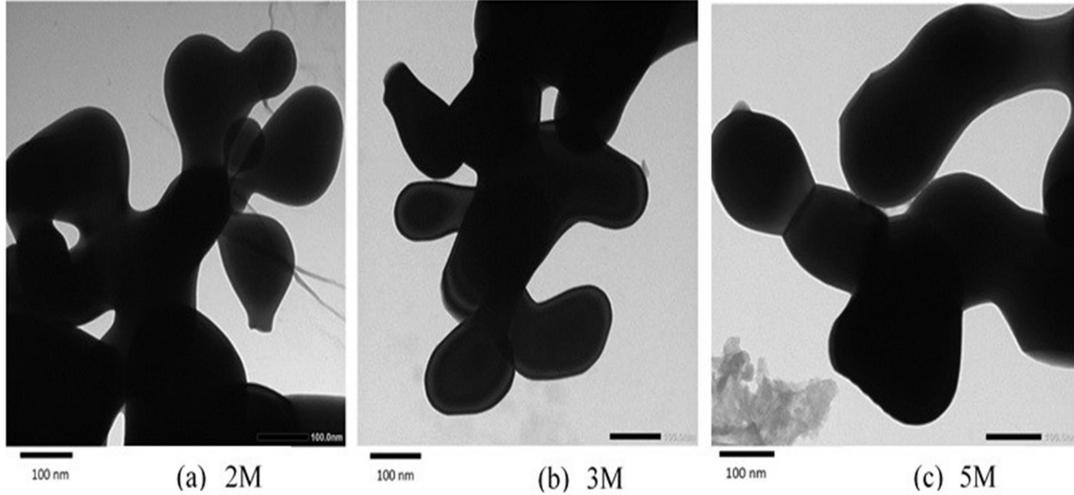


FIG. 2. TEM photographs of the YIG powders prepared with 2, 3 and 5M of NaOH.

TABLE 1. The lattice parameter ( $\text{\AA}$ ) and the grain size (nm) of the YIG powders prepared with 2, 3, 4 and 5M NaOH.

Sample	Lattice parameter ( $\text{\AA}$ )	Grain size (nm)
2M	12.339	163.42
3M	12.338	107.47
4M	12.335	-
5M	12.334	91.09

### Dielectric Studies of YIG

The real ( $\epsilon'$ ) and imaginary ( $\epsilon''$ ) parts of the dielectric constant and the ac conductivity  $\sigma_{ac}$  were calculated from the dielectric measurements using the following equations:

$$\epsilon' = Cd/A\epsilon_0 \quad (3)$$

$$\epsilon'' = \tan \delta \cdot \epsilon' \quad (4)$$

$$\sigma_{ac} = \epsilon'' \cdot \omega \cdot \epsilon_0 \quad (5)$$

where  $d$  is the sample disc thickness,  $A$  is the electrode area,  $C$  is the capacitance,  $\epsilon_0$  is the empty space permittivity,  $\tan \delta$  is the dielectric loss measured by the instrument and  $\omega$  is the angular frequency. The dimensions of the used pellets of the prepared samples are  $d = 2.2 \pm 0.1$  mm and  $A = 1.249 \times 10^{-4} \text{ m}^2$ .

Figs. 3 (a) and 4 (a) show, respectively, the influence of frequency on the real ( $\epsilon'$ ) and imaginary ( $\epsilon''$ ) parts of the dielectric constant. The highest values of both constants are obtained in the 2M sample, where the grain size is the largest. The effect of grain size on the dielectric constants can be explained by its effect on the population of domains and the movement of domain walls [15]. Both dielectric constants increase when the frequency increases. The dielectric constants arise from the polarization and movement of dipole orientation. This dipole orientation takes time, but as the frequency increases, the response time becomes comparable to the period of the field, thus the dielectric constants increase. A peak is observed in all the samples, which can be attributed to the occurrence of resonance, where the frequency of the applied electric field happens to be the same as the frequency of dipole orientation [16].

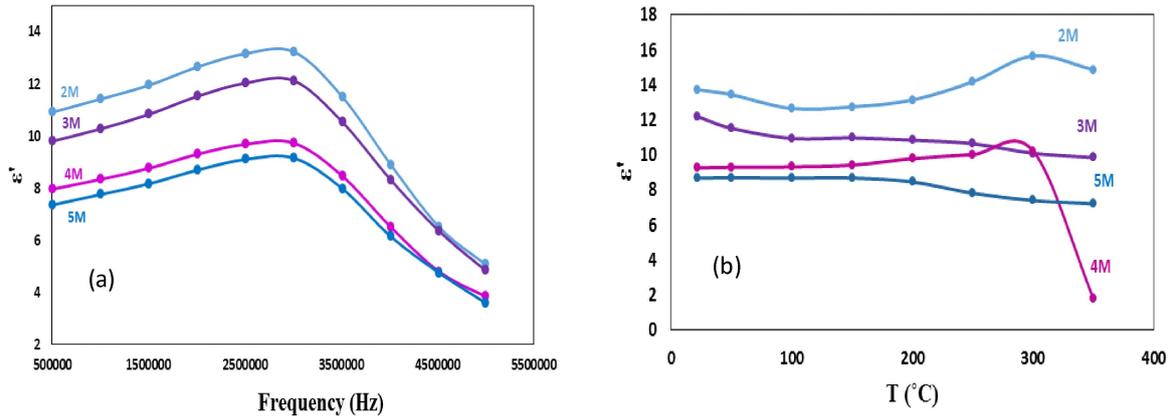


FIG. 3. Variation of the real part  $\epsilon'$  of the dielectric constant of the YIG samples (a) as a function of frequency at room temperature and (b) as a function of temperature at 2 MHz.

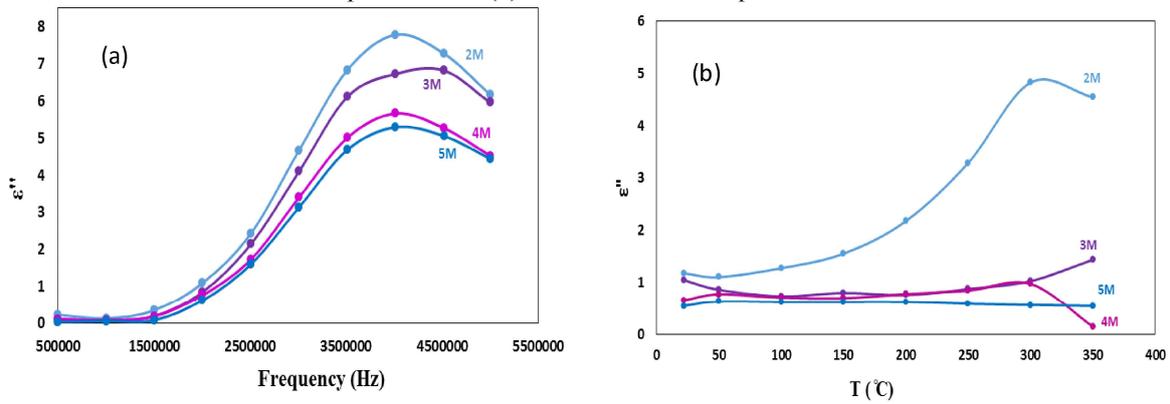


FIG. 4. Variation of the imaginary part  $\epsilon''$  of the dielectric constant of the YIG samples (a) as a function of frequency at room temperature and (b) as a function of temperature at 2 MHz.

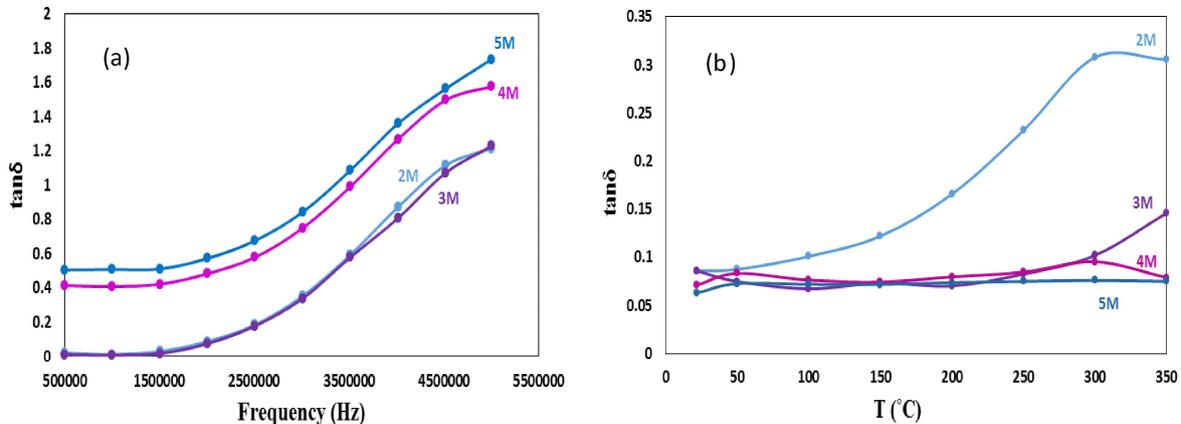


FIG. 5. Variation of the dielectric loss tangent  $\tan \delta$  (a) as a function of frequency at room temperature and (b) as a function of temperature at 2 MHz.

The increase of the temperature facilitates the segmental mobility, thus easing the dielectric polarization and increases the dielectric constants [10], as shown in Figs. 3 (b) and 4 (b). The peaks obtained are explained by Bunget et al. [17], where at a certain temperature, the decrease in relaxation polarization overcomes the increase in the number of polarons, so that the dielectric constants begin to decrease.

In Fig. 5 (a), it is obvious that the loss tangent increases with the increase of frequency and the lowest values are obtained in the 2M and 3M samples. This is caused by the presence of leakage loss aside with the polarization loss [18]; whereas the increase of the loss tangent with the increase of temperature, as shown in Fig. 5 (b), is attributed to the perturbation in the phonon system caused by the applied electric field, where the energy transferred to the phonon is dissipated in the form of heat [19].

The impact of the frequency on the ac conductivity is shown in Fig. 6 (a). The ac conductivity increases with the increase of frequency and this increase is the largest in the 2M sample. This lift is an indicator of the presence of small polaron conduction. On the other hand, the increase of the ac conductivity with temperature shown in Fig. 6 (b) could be attributed to the increase of the number of polarons with temperature [18] and the peak

obtained might be a result of the flipping of conduction mechanisms at a certain temperature. The irregular behavior of the dielectric parameters above the temperature of  $300^\circ\text{C}$  for samples at molarities 2M and 4M of NaOH can be attributed to the ferroelectric behavior of these samples. This behavior can be explained by the existence of the yttrium iron perovskite ( $YFeO_3$ ), where a ferroelectric phase transition can take place at high temperatures [20].

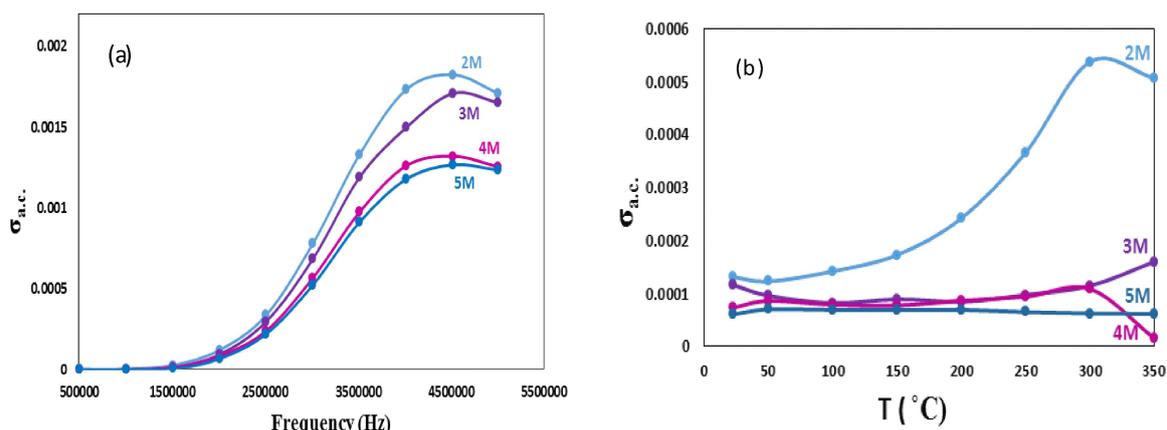


FIG. 6. Variation of the ac conductivity  $\sigma_{ac}$  (a) as a function of frequency at room temperature and (b) as a function of temperature at 2 MHz.

## Conclusion

In this work, we prepared YIG powders by co-precipitation method from chlorides of Iron and Yttrium. Dielectric properties of the prepared samples were studied over a wide range of frequencies and temperatures for different values of molarity of NaOH ( $M=2, 3, 4$  and  $5$ ). The change in the behavior of the dielectric constants as a function of frequency and temperature can be interpreted in terms of polarization and segmental mobility, respectively, while the variation of the ac conductivity can be interpreted in terms of polaron conduction. It can be seen that the highest dielectric constants and highest ac

conductivity values were obtained for the 2M sample. The dielectric properties of YIG can be manipulated by changing the molarity of NaOH, making it suitable for a wider range of applications, such as bio-antennas.

## Acknowledgement

Most of this work was done in the Materials Science Laboratory, Physics Department, Faculty of Science, Beirut Arab University, Beirut, Lebanon; in collaboration with the Physics Department, Lebanese University, Beirut, Lebanon.

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