

**Dielectric Behaviour Of Nano- Crystalline Al Doped M-Type Barium Hexaferrite****S. P. Pareek\*<sup>1</sup> Dr.Rajesh Jain\*<sup>2</sup>****Associate Professor Lecturer****Department Of Physics<sup>1&2</sup>****S. S. Jain Subodh PG College\*<sup>1</sup>****Jaipur****L.B.S. Govt.College\*<sup>2</sup>****Kotputli****Corresponding Email Id- [shyamsubodh2006@yahoo.co.in](mailto:shyamsubodh2006@yahoo.co.in)****{Received:15 January 2019/Revised: 20 January 2019/Accepted: 28 January 2019/Published: 2 February 2019}****Abstract**

A series of hexaferrite with composition,  $BaAl_xFe_{12-x}O_{19}$  ( $x= 0.2, 0.4...1.0$ ) have been synthesized by sol-gel auto combustion route. The gel is prepared from metal nitrates and citric acid. The dried nitrate – citrate gel exhibits auto- catalytic combustion behavior, which can be used to synthesize the nano crystalline ferrite powders. To get pure phase, samples were calcinated at 950 °C. The dielectric measurements were carried out at room temperature in a frequency range of 20 Hz to 2MHz using inductance capacitance resistance Meter Bridge (An Agilent E4980A precision LCR meter). All the samples show the frequency dependent phenomena, i.e. the dielectric constant decreases with increasing frequency and then reaches a constant value.

**Keywords: Auto-Combustion, Al Doped, Dielectric Constant, Hexaferrite.****Introduction**

There has been an increasing demand of magnetic materials for high-frequency applications such as telecommunications and radar systems, as microwave technology requires higher frequencies and bandwidths up to 100 GHz. Ferrimagnetic iron oxides (ferrites) are especially useful because they combine the properties of a magnetic material with that of an electrical insulator. Ferrites have much higher electrical resistivity than metallic ferromagnetic materials, resulting in minimization of the eddy current losses, and total penetration of the electromagnetic (EM) field. Furthermore, the widespread use of ferrites rests upon a remarkable flexibility in tailoring the magnetic properties, ease of preparation, and a favourable cost price. Hence ferrites are frequently applied as circuit elements, magnetic storage media like read/write heads, non-reciprocal and non-linear devices, such as phase shifters and Faraday rotators[1].

M-type strontium (SrM) and barium (BaM) hexagonal ferrites with dimensions in nanometers have attracted much attention because of their different magnetic and electrical properties

than those of their bulk counterpart and potential applications in various fields. Their large magneto crystalline anisotropy, high saturation magnetization, high value of coercivity, high electrical resistivity and corrosion resistivity make these materials useful as components in high frequency devices, plastic and permanent magnets, high density recording media and microwave devices[2-6]. A large amount of researches has been realized to modify the magnetic properties of barium hexaferrite by substitution of  $\text{Fe}^{3+}$  ions with other trivalent cations or divalent and tetravalent cation combinations and/or  $\text{Ba}^{2+}$  ion by other cations [7-10]. The magnetic properties of substituted ferrites depend directly of both electronic configuration and preference to occupy the non-equivalent  $\text{Fe}^{3+}$  sublattices of the substituted cations on hexagonal structure. Replacing  $\text{Fe}^{3+}$  ions by other less magnetic moment, paramagnetic or diamagnetic cations leads to change in the exchange interactions between the magnetic sublattices and to the appearance of new positions of  $\text{Fe}^{3+}$  ions. It causes reduction of the high uniaxial anisotropy field of the Ba ferrite. In addition, substituted Ba hexaferrites are very promising materials as electronic wave absorbers, especially at frequencies up to 50 GHz [11].

In the present work, our attention was focused on the study of dielectric properties of  $\text{BaAl}_x\text{Fe}_{12-x}\text{O}_{19}$ , where  $x = 0.0, 0.2, \dots, 1.0$ . The sol-gel auto-ignition method is used to speed up the synthesis of complex materials. It is a simple process, a significant saving in time and energy consumption over the traditional methods[12]. Small crystalline size of the resultants may have an important influence on the particles of the materials prepared. This method is employed to obtain improved powder characteristics, more homogeneity and have a narrow particle size, thereby influencing structural, electrical, and magnetic properties of hexaferrites.

### **Experimental Procedure**

Al substituted M-type  $\text{BaAl}_x\text{Fe}_{12-x}\text{O}_{19}$  ( $x = 0.0, 0.2, \dots, 1.0$ ) hexaferrites were prepared using the citrate sol-gel technique. The starting materials were barium nitrate  $\text{Ba}(\text{NO}_3)_2$ , iron nitrate  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ , Aluminium nitrate  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ , Citric acid ( $\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$ ) and Ammonia  $\text{NH}_3 \cdot \text{H}_2\text{O}$  (25 %) , all of analytic purity. In a typical procedure for the preparation; Ba, Cu and Fe nitrates were taken in stoichiometric proportions then dissolved in a minimum amount of distilled water. Solution of citric acid was added to the aqueous salt solution in the ratio of 1:1 to total moles of nitrate ions and pH of the solution was adjusted to 7.0 with addition of  $\text{NH}_3 \cdot \text{H}_2\text{O}$ . The solution was heated at 80 °C with continuous stirring and maintained at constant temperature using a hot plate. Finally, as water evaporated the solution became viscous and formed a very viscous brown gel. Increasing the temperature led to the ignition

of the gel. The dried gel burnt in a self propagation combustion manner until all gels are completely burnt out to form a loose powder. To obtain different structural features of ferrites, the loose powder precursor was treated by various heating methods in a muffle furnace. The obtained powders were preheated in air at 500 °C for 4 h at the rate of 50 °C/h and then calcinated at 950 °C for 4 h to achieve ferrite particles.

### Result And Discussion

The dielectric constant of the synthesized nanomaterials is calculated using the following equation:

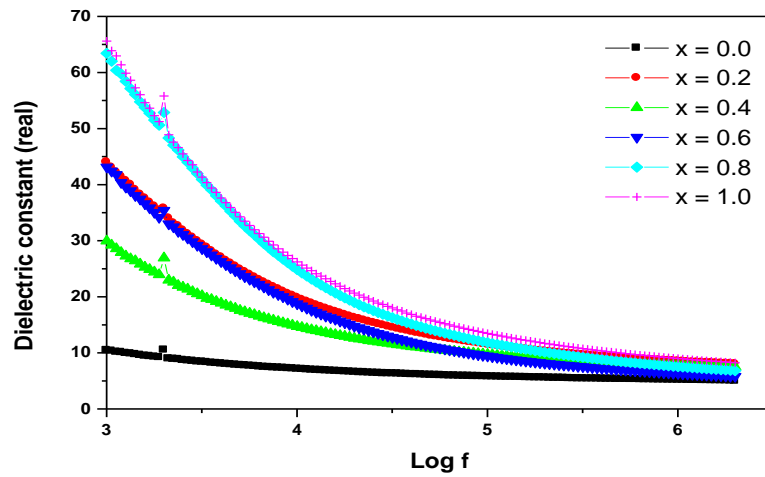
$$\epsilon' = cd / \epsilon_0 A \quad (1)$$

Where C is the capacitance of the pellet in farad, d the thickness of the pellet in meter, A the cross-sectional area of the flat surface of the pellet and  $\epsilon_0$  the constant of permittivity of free space

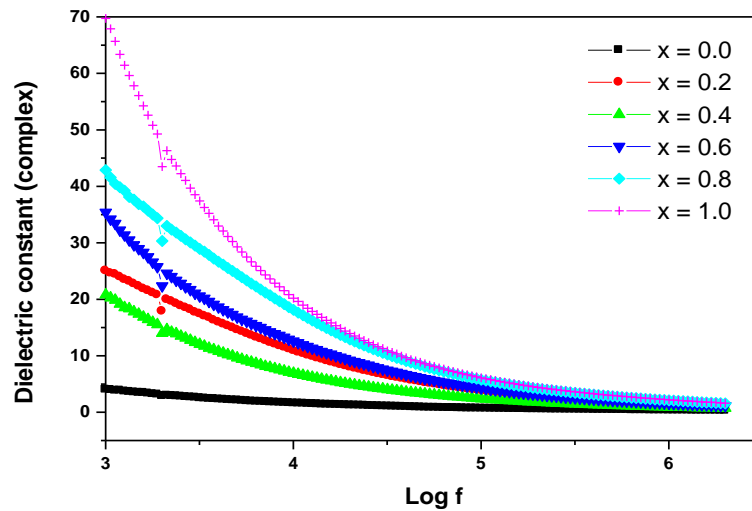
Fig.2 and Fig.3 show the variation of real and imaginary part of the dielectric constant of  $BaAl_xFe_{12-x}O_{19}$  ferrite samples with frequency 20 Hz to 2MHz at room temperature. It can be observed that all the compositions exhibit dielectric dispersion where both real and imaginary dielectric constant decreases rapidly with increasing frequency in low-frequency region while it approaches almost frequency independent behaviour in high frequency region. According to Rabinkin and Novikova [13, 14], the polarization in ferrites is through a mechanism similar to the conduction process. The polarization decreases with increase in frequency and then reaches a constant value due to the fact that beyond a certain frequency of external field, the electron exchange between  $Fe^{2+}$  and  $Fe^{3+}$  cannot follow the alternating field. The large value of 'e' at lower frequency is due to the predominance of species like  $Fe^{2+}$  ions, interfacial dislocations pile ups, oxygen vacancies, grain boundary defects, etc. [15], However the decrease in 'e' with frequency is natural because of the fact that any species contributing to polarizability lag behind the applied field at higher and higher frequencies.

It was noted that the values of  $\epsilon'$  and  $\epsilon''$  increase with the addition of  $Al^{3+}$  ions. The conduction in ferrites is because of the electron hopping between  $Fe^{3+}$  and  $Fe^{2+}$  ions at octahedral sites. After doping of  $Al^{3+}$  ions, it is assumed that  $Ba^{2+}$  ion is substituted for  $Al^{3+}$  ion according to the equation:  $Ba^{2+} + Fe^{3+} \rightarrow Al^{3+} + Fe^{2+}$ . With the increase of  $Al^{3+}$  doping, the chance of electron hopping between  $Fe^{3+}$  and  $Fe^{2+}$  increases. Because there is a strong correlation between hopping mechanism and dielectric polarization mechanism in ferrite. The substitution of  $Al^{3+}$  ions leads to an increase in the polarization on the sample surface, and results in the increase of dielectric constant  $\epsilon'$ . Also, with the increase of  $Fe^{2+}$

ions, the chance of electron transfer increased between Fe<sup>3+</sup> and Fe<sup>2+</sup> leading to higher dielectric loss, and  $\epsilon''$  also increased.



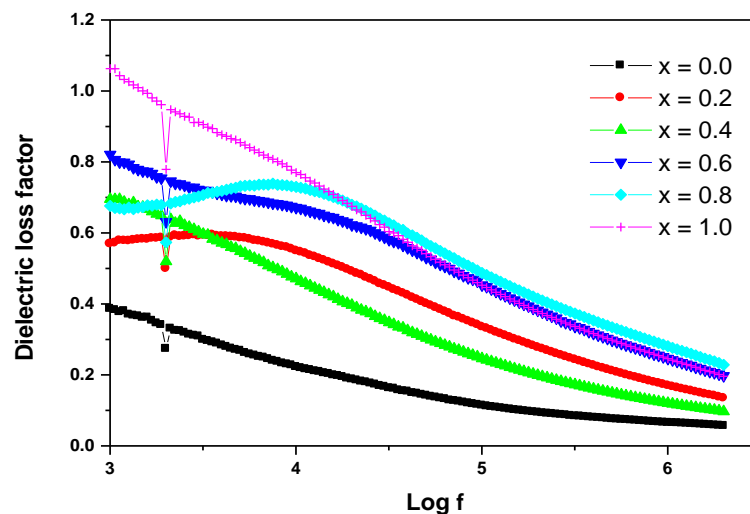
**Fig.2.** Variation Of Dielectric Constant ( $\epsilon'$ ) Versus Frequency Log f of BaAl<sub>x</sub>Fe<sub>12-x</sub>O<sub>19</sub> At Room Temperature.



**Fig.3.** Variation Of Dielectric Constant ( $\epsilon''$ ) Versus Frequency Log f of BaAl<sub>x</sub>Fe<sub>12-x</sub>O<sub>19</sub> At Room Temperature.

Dielectric tangent loss factor ( $\tan \delta$ ) is an important part of the total core loss in ferrites. Hence for low core loss, low dielectric losses are desirable. Dielectric loss factor is represented as the energy dissipation in the dielectric system. The variation of  $\tan \delta$  as a function of frequency at room temperature is shown in Fig. 4. It is seen for all the samples that it decreases continuously as the frequency of the alternating field's increases. The

decrease of  $\tan \delta$  with an increase of frequency can be explained on the basis of Koop's phenomenological theory. None of the samples exhibit the loss peak. The peaking nature occurs when the jumping frequency of electrons between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  is equal to the frequency of the applied AC field. In the low frequency region, which corresponds to high resistivity (due to grain boundaries), more energy is required for electron exchange between  $\text{Fe}^{3+}$  and  $\text{Fe}^{2+}$  ions; thus the energy loss is high. In the high frequency range, which corresponds to low resistivity (due to grain), less energy is needed for electron transfer between  $\text{Fe}^{3+}$  and  $\text{Fe}^{2+}$  in the grains and hence the energy loss is small.



**Fig.4.** Variation Of Dielectric Loss Factor Versus Frequency Log f of  $\text{BaAl}_x\text{Fe}_{12-x}\text{O}_{19}$  at Room Temperature.

### Conclusion

A series of Al doped Barium hexaferrite with composition  $\text{BaAl}_x\text{Fe}_{12-x}\text{O}_{19}$  ( $x = 0.0, 0.2, \dots, 1.0$ ) have been successfully by using Sol-Gel auto combustion route. The dielectric properties show the normal behaviour with frequency, which is explained on the basis of Koop's theory and Maxwell–Wagner model. All the samples show the frequency dependent phenomena, i.e. the dielectric constant decreases with increasing frequency and then reaches a constant value.

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