International Journal of Advanced Research in Engineering and Applied Sciences

ISSN: 2278-6252

POWER LAW BEHAVIOR OF $MAL_{2-2X}Y_{2X}O_4$ (M = Mg, Ni and Zn) ALUMINATE NANOPARTICLES

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Abstract: *In the present work, investigations were carried out in order to address the issue of power law dependence* $\sigma_{ac} = B\omega^n$*, for a series of* $MgAl_{2-2x}Y_{2x}O_4$*,* $NiAl_{2-2x}Y_{2x}O_4$ *and* $ZnAl_{2-2x}Y_{2x}O_4$ *(x = 0.00, 0.01, 0.02, 0.03, 0.04, 0.05, 0.07 and 0.10) cubic spinel nanoparticles. The power law behavior of all the samples in the frequency range from 1 kHz to 1MHz suggested that the frequency exponent n increases with an increase in frequency for all the series. The increase in the value of n at higher frequencies suggests that the degree of interaction between the ions and the lattices which surround them increases.*

1. INTRODUCTION

The properties of materials in the nanoregime are totally different when compared with their bulk counterparts in the micrometer regime. This modification in the properties upon particle size reduction is attributed to a variety of reasons, namely particle size, shape and grain boundaries. Because of these modified properties nanostructured materials can be effectively utilized in various applications [1].

In the recent years nanocrystalline spinel aluminates, MAl_2O_4 (M =, Mg, Ni, Zn) have received considerable interest due to their unique electrical and optical properties [2-4]. These materials exhibit high electrical resistivity, low dielectric constant, low dielectric loss and optical bandgap in the ultraviolet region, which allow them to be used as a dielectric and optical material for technological applications [5-7]. Studies on the dielectric properties of pure and doped nanosized aluminates were carried out by many researchers [8-9], but the reports on correlation between ac conductivity and frequency for the nanosized spinel aluminates has not been reported in the literature.

In our recent study, it has been reported that the addition of rare earth yttrium in nanosized spinel aluminates strongly influenced their structural and dielectric properties [10]. In this work, we present in detail the correlation between ac conductivity vs frequency of Y

subsituted MAl_2O_4 (M =, Mg, Ni, Zn) nanoaluminates.

2. EXPERIMENTAL

All the samples were prepared from Merck Germany GR grade chemicals *viz. Mg* $(NO_3)_2.6H_2O$, *Ni* $(NO_3)_2.6H_2O$, *Zn* $(NO_3)_2.6H_2O$, *Al* $(NO_3)_3.9H_2O$, *Y* $(NO_3)_3.9H_2O$ and aqueous NH_3 (Merck India, 30%). The samples were prepared by using chemical coprecipitation technique at pH 10.

The synthesis of pure $MgAl_2O_4$ was done by using stoichiometric quantities of $Mg(NO_3)_2{\cdot}6H_2O$ (5.128 g) and $Al(NO_3)_3{\cdot}9H_2O$ (15.005 g) then dissolving them separately in 100mL deionized water and adding simultaneously into a flask containing 200mL deionized water. Ammonia solution was added drop wise till the pH value 10 was attained. The solution was continuously stirred by a magnetic stirrer for 1 h and aged at room temperature overnight. The precipitates were filtered and washed with deionized water and then dried at 120°C for 16 h in a hot air oven. The dried samples were calcined at 950°C in air in a tube furnace programmed at a fixed heating rate of 5°C/min for 8 h.

For the synthesis of pure $NiAl_2O_4$ and $ZnAl_2O_4$ the stoichiometric quantities of Ni $(NO_3)_2{\cdot}6H_2O$ (5.816 g) and Zn $(NO_3)_2{\cdot}6H_2O$ (5.949 g) were used respectively. In addition to this the nanoparticles of $ZnAl_{2-2x}Y_{2x}O_4$ series were calcined at 800°C instead of 950°C. All the other experimental conditions were maintained similar as mentioned above. The yttrium doped derivatives of all the series were prepared by adding the appropriate stoichiometric quantities of dopant salt Y $(NO_3)_3.9H_2O$ and following the same procedure.

The ac conductivity σ_{ac} was calculated using the formula $\sigma_{ac} = 2\pi f \varepsilon' \varepsilon_0 \tan\delta$ from the dielectric data carried out using Wayne Kerr 6500B Impedance Analyzer.

3. RESULTS AND DISCUSSION

3.1 AC conductivity studies

In order to understand the conduction mechanism and the type of polarons responsible for conduction, the variation of ac conductivity ($\sigma_{a.c.}$) as a function of frequency is represented in Figs 1-3. The conductivity shows an increasing trend with an increase in frequency for all the samples. This type of frequency dependence of conductivity can be explained with the help of heterogeneous nature of the samples. According to which two layers namely grain and grain boundaries are formed in the dielectric structure. When an electric field is applied the poorly conducting grain boundaries are more active at lower frequencies and hence

long range inter-well hopping of electrons between Al^{2+} and Al^{3+} ions are less at lower frequencies. As frequency of the applied field increases, the conductive grains become more active thereby promoting the intra-well hopping of electron between Al^{3+} and Al^{2+} ions in the octahedral sites. Therefore we observe a gradual increase in ac conductivity with frequency [10]. It is well known that frequency dependent conduction in disordered solids is directly proportional to the frequency. In the present case the plots of ac conductivity are linear, indicating that conduction is due to small polarons. At higher frequencies, where conductivity increases greatly with frequency, the transport is dominated by contributions from hopping of infinite clusters [10].

3.2 Power law dependence of $MAl_{2-2x}Y_{2x}O_4$ (M =, Mg, Ni, Zn) nanoaluminates

The observed behavior of ac conductivity with frequency was found to follow the power law given by

$$\sigma_{ac} = B\omega^n \qquad (1)$$

where B is constant having the units of conductivity and n is the frequency exponent ($n \leq 1$) [11]. From the plots between logω versus log σ_{ac} (ω) as shown in Figs. 1-3, the slope directly provides the value of dimensionless frequency exponent *n*. The estimated values of *n* at room temperature for all the compositions at room temperature are tabulated in Tables 1-3, from where it can be clearly seen that the frequency exponent *n* varies with Y content and frequency.

Frequency exponent *n* is a measure of correlation between σ_{ac} and frequency. Value of *n* is 0 for random hopping of charge carriers (frequency independent σ_{ac}) and tends to 1, as the correlation between σ_{ac} and *ω* increases. In the present study, value of n increases continuously with an increase in frequency and is less than 1 for all the samples. This is attributed to the fact that the hopping of charge carriers between localized states increases with an increase in increase in frequency thereby increasing the frequency exponent *n*. The values of exponent *n* in the different frequency range are also found to composition dependent. The higher value of *n* for samples having higher x indicates that the dispersion of ac conductivity with frequency has been enhanced with the incorporation of Y at Al site, and may be correlated with the change in the distance and barrier heights of the sites available for charge carriers for conduction [12-13].

CONCLUSION

The variation of ac conductivity for all the nanostructured samples has been discussed in light of the hopping model. The dispersion of ac conductivity has been estimated in terms of frequency exponent *n*, which varies dopant content and is explained due to some type of structural transformation and formation of electrical dipoles and their ordering.

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FIGURES

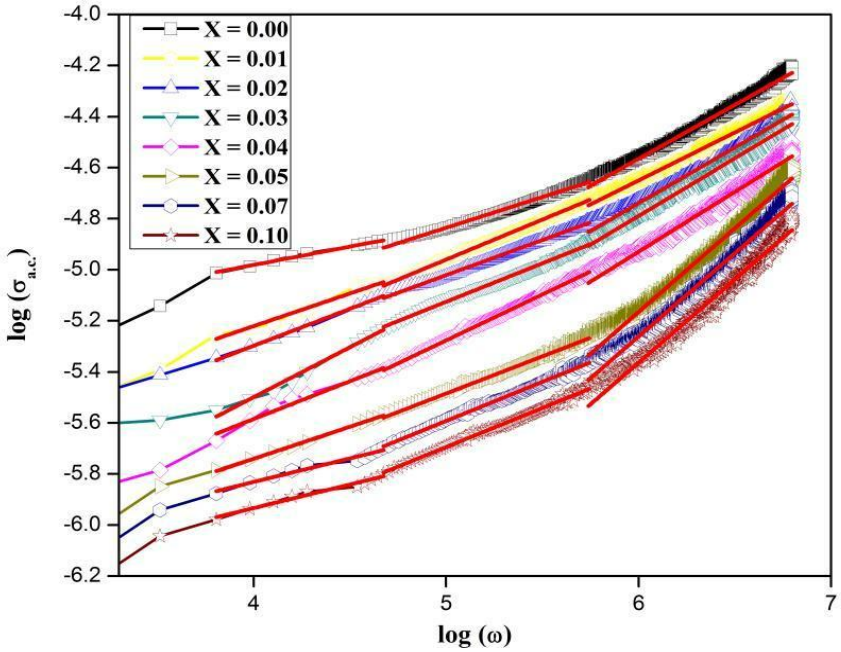

Fig.1: Variation of log (σ_{ac}) vs logω in different frequency regions for the $MgAl_{2-2x}Y_{2x}O_4$ (x=0.00-0.10) system

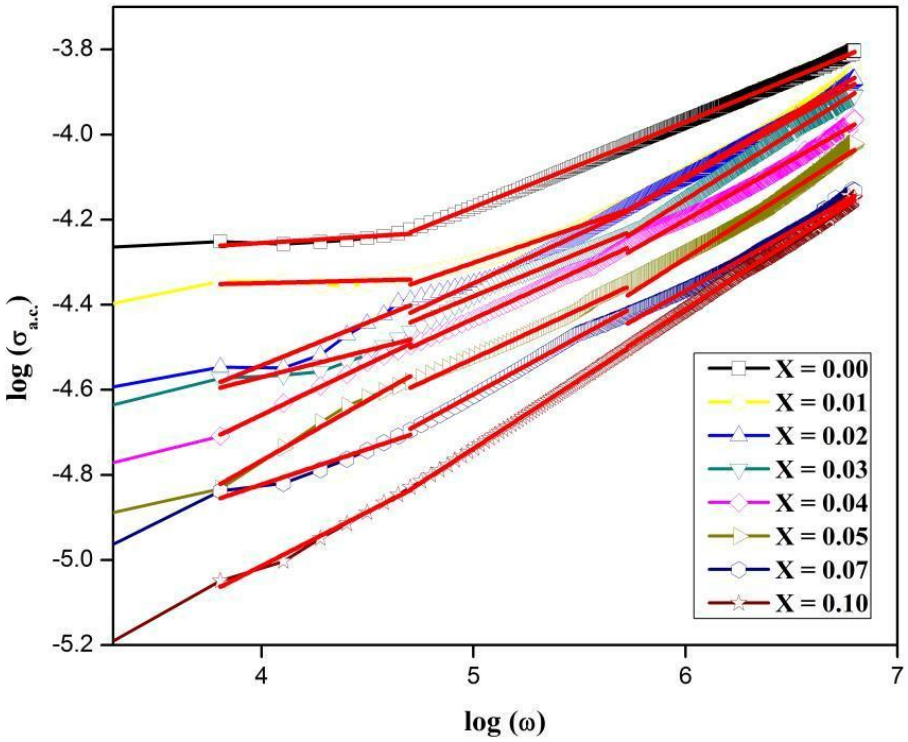

Fig.2: Variation of log (σ_{ac}) vs logω in different frequency regions for the $NiAl_{2-2x}Y_{2x}O_4$ (x=0.00-0.10) system

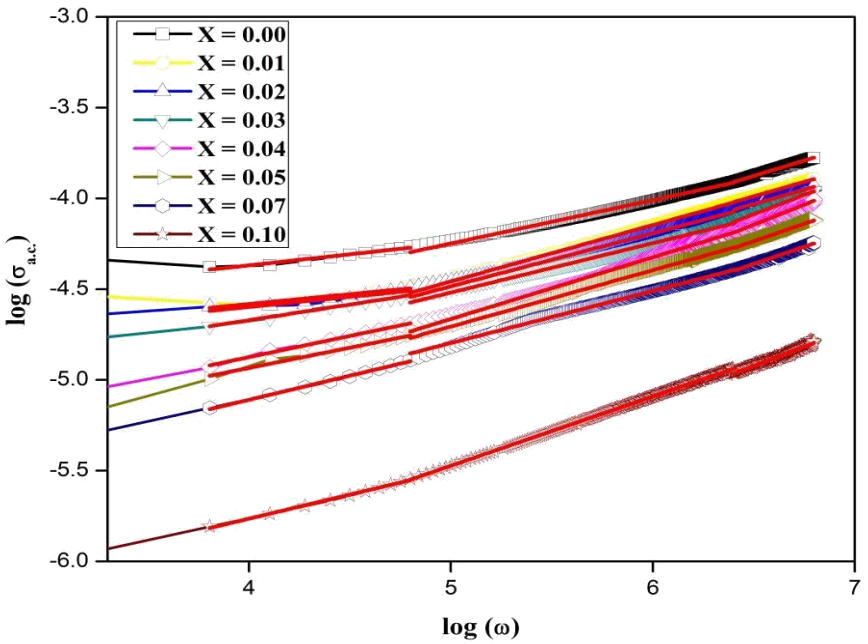

Fig.3: Variation of log (σ_{ac}) vs logω in different frequency regions for the $ZnAl_{2-2x}Y_{2x}O_4$ (x=0.00-0.10) system

TABLES

Table 1: Variation of frequency exponent n in different frequency regions for the $MgAl_{2-2x}Y_{2x}O_4$ (x=0.00-0.10) system

MgAl2-2xY2xO4	log ω (3.80 to 4.67)	log ω (4.67 to 5.74)	log ω (5.74 to 6.79)
Y concentration	Exponent factor n	Exponent factor n	Exponent factor n
0.00	0.14	0.24	0.42
0.01	0.25	0.31	0.37
0.02	0.27	0.29	0.43
0.03	0.29	0.30	0.45
0.04	0.29	0.35	0.46
0.05	0.25	0.29	0.65
0.07	0.19	0.30	0.65
0.10	0.18	0.30	0.65

Table 2: Variation of frequency exponent n in different frequency regions for the $NiAl_{2-2x}Y_{2x}O_4$ (x=0.00-0.10) system

NiAl2-2xY2xO4	log ω (3.80 to 4.70)	log ω (4.70 to 5.72)	log ω (5.72 to 6.79)
Y concentration	Exponent factor n	Exponent factor n	Exponent factor n
0.00	0.03	0.19	0.20
0.01	0.01	0.17	0.29
0.02	0.20	0.23	0.27
0.03	0.12	0.20	0.31
0.04	0.23	0.22	0.28
0.05	0.28	0.23	0.31
0.07	0.16	0.27	0.28
0.10	0.25	0.32	0.32

ISSN: 2278-6252

Table 3: Variation of frequency exponent n in different frequency regions for the $ZnAl_{2-2x}Y_{2x}O_4$ (x=0.00-0.10) system

ZnAl2-2xY2xO4	**log ω (3.80 to 4.79)**	**log ω (4.79 to 6.40)**	**log ω (6.40 to 6.79)**
Y concentration	**Exponent factor n**	**Exponent factor n**	**Exponent factor n**
0.00	0.12	0.23	0.33
0.01	0.10	0.31	0.32
0.02	0.11	0.29	0.32
0.03	0.17	0.27	0.42
0.04	0.23	0.34	0.44
0.05	0.22	0.31	0.38
0.07	0.26	0.28	0.37
0.10	0.26	0.38	0.42