STRUCTURAL AND DIELECTRIC PROPERTIES OF $\text{MgAl}_{2-2x}\text{Y}_{2x}\text{O}_4$ NANOPARTICLES

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Abstract. A series of Nano $\text{MgAl}_{2-2x}\text{Y}_{2x}\text{O}_4$ ($x = 0.00, 0.07$ & 0.10) was prepared by chemical co-precipitation technique. X-ray diffraction (XRD) analysis reveals that a secondary phase appears at $x = 0.10$ along with the spinel phase. The broad peaks in the XRD pattern and particle size analysis by Transmission electron microscopy (TEM) confirmed nanocrystalline nature of the samples. Room temperature dielectric response as a function of frequency (1 kHz to 1MHz) shows that dielectric constant loss tangent and ac conductivity decreases with increase in yttrium content. The variation of dielectric constant as a function of frequency indicates the dielectric dispersion due to Maxwell-Wagner Polarization. The variation of AC conductivity with frequency suggests the conduction mechanism due to small polaron hopping.

Keywords: Co-precipitation, X-ray diffraction, Dielectric response.

1. Introduction

Because of their theoretical and technological relevance, ceramic spinel oxides and their related structures have been studied thoroughly [1]. Magnesium aluminate (MgAl_2O_4) is a well known ceramic compound with spinel structure. The nature of bonding in MgAl_2O_4 is ionic, which classifies this material as an insulator. Recently MgAl_2O_4 has received attention as a dielectric material. The polarization mechanism in spinel aluminates depends on the arrangement of ions in different sites. Several groups have studied the dielectric behaviour of pure and doped nano aluminates prepared by different methods [2-3]. In our present work we have systematically investigated the effect of yttrium substitution on the structural and dielectric properties of MgAl_2O_4 prepared by a simple chemical coprecipitation technique.

2. Experimental Details

All samples were prepared by co-precipitation technique at pH 10 using Merck Germany GR grade nitrate precursors described elsewhere [4]. X-ray powder diffraction was

conducted on Panalytical Xpert-PRO 3050/60 diffractometer using Cu K α radiation. Morphology of the nanoparticles was obtained using a transmission electron microscope (JEOL 2100 F) operated at 200 kV. Room temperature dielectric measurements were done using Wayne Kerr 6500B Impedance Analyzer.

3. Results and Discussion

3.1. Structural Studies

3.1.1. X-ray Diffraction

The XRD pattern of all the samples (Fig.1) shows good crystallization with well defined diffraction lines indicating the presence of cubic spinel phase. For sample $x = 0.10$ secondary phase appears at $2\theta = 29.56^\circ$. This peak is identified as $\text{Al}_2\text{Y}_4\text{O}_7$ matched with ICSD PDF # 069530. The replacement of smaller Al^{3+} with larger Y^{3+} ions increases the lattice constant (a) for all the compositions which is consistent with Vegard's law [5]. With the addition of yttrium into the spinel lattice the crystallite size (D) (Table 1) reduces which may due to the segregation of Y^{3+} ions near the grain boundaries due to which grain boundary mobility decreases and hence crystallite size decreases.

Fig.1. XRD pattern for the prepared samples.

Fig.2. TEM image of pure MgAl_2O_4

3.1.2. Transmission Electron Microscopy

Transmission electron micrograph (Fig.2) for pure MgAl_2O_4 on 20 nm scale validates the nanocrystalline nature of the sample. It was clear from TEM image that due the presence of polycrystalline aggregates particle size was larger than the crystallite size calculated from the XRD data.

3.2. Dielectric Response

3.2.1. Dependence of Dielectric Constant on Frequency

The inverse dependence of dielectric constant (ϵ') on frequency as shown in (Fig.3), can be explained on the basis of Maxwell-Wagner type interfacial polarization, and Koops

theory [6]. In the present case solid samples in the form of pellets can be imagined to consist of well conducting grains separated by poorly conducting grain boundaries. Now with the application of external field the charge carriers accumulate at the grain boundaries, thereby creating interfacial polarization. At lower frequencies the charge carriers can follow the external applied field alternations thereby contributing to the interfacial polarization whereas at higher frequencies the electron exchange between charge carriers cannot follow the external alternating field, which causes a decrease in the contribution towards interfacial polarization, thereby resulting in low dielectric constant at higher frequencies.

Fig.3. Variation of dielectric constant with log f.

Fig.4 Variation of loss tangent with log f.

3.2.2. Dependence of Dielectric Loss Tangent on Frequency

The plot of dielectric loss tangent ($\tan\delta$) with frequency (Fig.4) depicts that $\tan\delta$ decreases with increase in frequency for all samples. This variation of $\tan\delta$ was considered to be caused by domain wall resonance. At lower frequencies more energy was required for polarization in the grain boundaries due to which energy loss was high on the other hand at high frequencies polarization between grain boundaries was inhibited due to which the energy loss was found to be low at higher frequencies.

3.2.3. AC Conductivity Study

The ac conductivity ($\sigma_{a.c.}$) increases with increase in frequency for all the samples. This happens because as the frequency of the applied field increases, the conductive grains become more active, due to which the hopping conduction increases with frequency. Also the electrical conduction mechanism in terms of polaron hopping model has been discussed by Austin and Mott [7]. They have suggested that in large polaron hopping the ac conductivity decreases with frequency whereas in small polaron hopping it increases with frequency. Therefore in our present study the linear plots of ac conductivity indicates that the conduction mechanism for all the samples occurs due to hopping of small polaron type charge carriers among the localized states.

Fig.5 Variation of AC conductivity with frequency for all samples

TABLE 1. Structural and dielectric parameters for $\text{MgAl}_{2-2x}\text{Y}_{2x}\text{O}_4$ at different ‘x’ values.

(x)	a (Å)	D (nm)	ϵ' (1 MHz)	$\tan\delta$ (1 MHz)	$\sigma_{\text{a.c.}} \times 10^{-5}$ (1 MHz)
0.00	8.079	10.27	7.35	0.0143	5.88
0.07	8.115	6.23	5.41	0.066	1.92
0.10	8.118	5.87	4.81	0.058	1.52

4. Conclusion

Single phase nanocrystalline samples of $\text{MgAl}_{2-2x}\text{Y}_{2x}\text{O}_4$ have been synthesized by a chemical coprecipitation technique using ammonia as a precipitating agent. Transmission electron microscopy (TEM) reveals polycrystalline nature of the samples. A strong composition and frequency dependence was observed in dielectric response of all the samples with yttrium substitution at the octahedral site. Nanostructured samples presented in this paper possess low values of both dielectric constant and dissipation factor which favor the use of these materials in electronic industry

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