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Magnetoelectric Properties of Nanostructured YMnO₃ Prepared by Sol–Gel Technique

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Abstract

Nanostructured yttrium manganese oxide (YMnO₃) has been synthesized using cost effective Sol-Gel technique in order to understand effect of magnetic field on the electrical properties and its magnetoelectric coupling. XRD measurement has been carried out using powder diffractometer. To confirm the particle morphology, TEM measurement was carried out. Frequency dependent dielectric properties and impedance spectroscopy have been carried out in the range of 20Hz to 2MHz frequency at room temperature (RT). Universal dielectric relaxation (UDR) has been fitted to understand effect of magnetic field on electrical properties in detail.

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Keywords: Nanostructured; Sol-Gel; Magnetoelectric effect.

1. Introduction

Spintronic based applications have been renewed in various functional oxide materials. The giant magnetoresistance (GMR) phenomenon was the initiative of the development of applications in spintronic field, and today, spintronic materials have found with large number of applications with the advancement of the speed,

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size scaling and power requirements [1,2]. They are largely used in magnetic field sensors, high-speed data couplers or isolators, and magnetic random access memory (MRAM), etc. In recent years, various functional oxide materials like colossal magnetoresistance (CMR) manganites, diluted magnetic semiconductor, ferrites, multiferroics, etc have been investigated for the spintronic based device applications.

Yttrium manganese oxide (YMnO₃) is an important member of the perovskite family with its electrical transition temperature ~ 920K and magnetic transition temperature ~ 70K [3,4]. YMnO₃ based compounds have been studied widely for their structural, dielectric and magnetic properties [5–9]. Reports on the effect of doping on its structural and magnetic properties were available while size effect on the nanostructured YMnO₃ have been investigated by Teowee et al [10]. There exist few reports on the magnetodielectric (MD) properties of YMnO₃ [11–13] wherein magnetoelectric coupling and MD behavior have been discussed in the context of spin structure modulation, magnetic moments and ferroelectric – antiferromagnetic couplings. Few reports are available on the magnetoelectric studies on YMnO₃ based compounds [11-13] in which authors have discussed temperature dependent dielectric anomaly in the YMnO₃ sample (mostly near to its magnetic transition temperature) and variation in this anomaly with applied magnetic field has been discussed on the basis of magnetoelectric coupling. Also, Tripathy et al [11] have studied this compound in the form of nanoceramic with BiFeO₃ phase. They have studied the frequency dependent dielectric constant under different applied magnetic fields and found the suppression in dielectric constant with increase in magnetic field. Also, they have studied the field induced modifications in the impedance frequency behavior and reported the positive magnetoimpedance nature in these BiFeO₃-YMnO₃ based nanoceramics. Almost no reports are available on magnetodielectric studies on pure nanostructured YMnO₃ compound grown using sol-gel technique, since, sol-gel technique provides various aspects such as: (i) capability to synthesize wide range of materials, (ii) flexibility for selecting various starting materials, (iii) low temperature synthesis, (iv) simple set-up, (v) easy handle, (vi) vacuum free environment, (vii) identical particle size, (viii) identical particle growth, (ix) identical structure of the particles, (x) identical stoichiometry of the particles, (xi) monodispersive nature of the particles, (xii) low cost synthesis and (xiii) yields predefined stoichiometric compounds.

By keeping in mind all the above aspects of magnetoelectric / magnetodielectric studies on YMnO3 based compounds and sol-gel method, in the present communication, we have synthesized nanostructured YMnO₃ compound by Sol-Gel technique. Magnetoelectric (MD) and magnetoimpedance behaviors have been investigated by collecting magnetic field dependent electrical properties of the nanostructured YMnO₃.

2. Experimental Details

Nanostructured YMnO₃ was prepared using the conventional Sol-Gel method [14,15]. The sample was prepared by using acetate precursor route. Sigma Aldrich make yttrium acetate (99.90% purity) [Y(CH₃CO₂)₃ × XH₂O] and manganese acetate (99.99% purity) [Mn (CH₃CO₂)₂ × 4H₂O] have been used as starting materials in appropriate stoichiometric ratio. To make precursor solution, double distilled water (DDW) and acetic acid glacial (AAG) (CH₃COOH) have been used in 1:1 ratio. The mixture was then stirred at 60°C with 500 rpm of about 2 hours to get homogeneous solution. The homogeneous solution was then heated at 100°C for about 3-4 days to get the xerogel. Grinding of the xerogel for 30 minutes resulted in powder which was then calcined at 300°C for two hours. Resultant powder was subsequently palletized and sintered at 700°C for 12 hours. YMnO₃ samples was characterized by X-ray diffraction (XRD) at room temperature using CuK α X-ray radiation source for its structural behavior while frequency dependent dielectric and impedance measurements were carried out under 0 and 1.2T applied magnetic fields at room temperature using Agilent make (model no. E4980A) high precision LCR meter in the range 20Hz – 2MHz. Also, to confirm the nanophasic nature of the particles in YMnO₃ compound, transmission electron microscopy (TEM) measurement was carried out.

3. Results and Discussion

3.1. Structural Properties

Figure 1 shows XRD pattern of YMnO₃ compound synthesized by low cost and simple sol–gel technique. XRD pattern of YMnO₃ sample reveals single phasic nature with hexagonal structure having $P6_3cm$ space group and lattice parameters a = b = 6.139618 and c = 11.395924. The CS calculated using Scherer's formula [16] [CS = 0.9λ / B cos θ , where λ is the wavelength of X-rays used, B is the value of FWHM and θ is the angle of incident] is found to be ~ 26.33 nm.

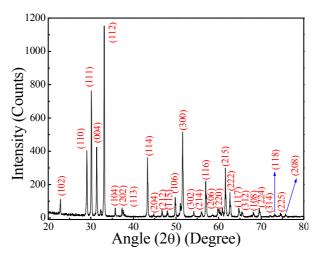


Fig. 1. XRD pattern of Sol-Gel grown nanostructured YMnO₃.

3.2. Microstructural Properties

To confirm the particle size of presently studied sol–gel grown YMnO $_3$ compound, TEM image was carried out, as shown in figure 2. It is clear from TEM image that presently studied nanostructured YMnO $_3$ compound possesses high enough crystallinity with the particle size ~ 20.37 nm which is very near to the size calculated using XRD results.

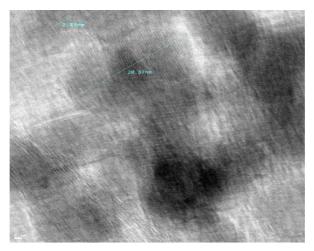


Fig. 2. TEM image of Sol-Gel grown nanostructured $YMnO_3$.

3.3. Electrical Studies

In order to understand magnetoelectric coupling of nanostructured YMnO₃, magnetic field dependent dielectric and impedance spectroscopy have been studied at 0 and 1.2T magnetic fields. Figure 3 (a) shows the variation in dielectric permittivity (ϵ ') with frequency for Sol-Gel grown nanostructured YMnO₃ with and without magnetic fields in the range of 20Hz to 2MHz performed at room temperature. From figure 3 (a) it is seen that dielectric permittivity increases when magnetic field is applied to nanostructured YMnO₃. Positive MD [MD = [(ϵ '_H - ϵ '₀) / ϵ '₀] × 100] effect has been found for the presently studied samples which can be understood as: when magnetic field is applied to the sample, magnetic phase gets modified which in turn modifies the structural distortion and hence dielectric permittivity increases with applied magnetic field due to inverse DM interaction [17]. To understand the effect of magnetic field for dielectric response of nanostructured YMnO₃, UDR (Universal dielectric response) model has been plotted. Figure 3 (b) shows log (f ϵ ') vs. log f plot for, both, the conditions, with and without magnetic fields. UDR model fits well for both the conditions suggesting that crystal cores contribute for dielectric response.

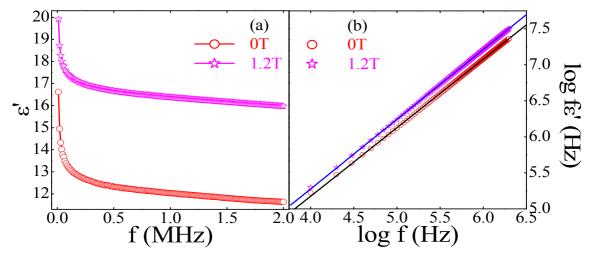


Fig. 3. (a) Frequency dependence of dielectric permittivity and (b) UDR model for Sol-Gel grown nanostructured YMnO₃.

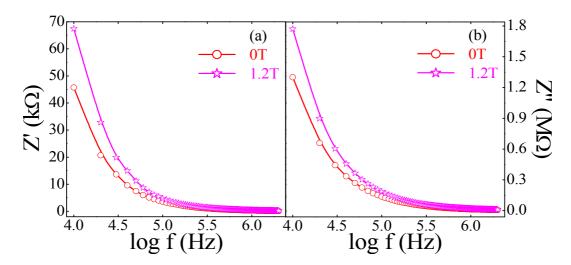


Fig. 4. Frequency dependent (a) real and (b) imaginary parts of impedance for Sol-Gel grown nanostructured YMnO₃.

Figure 4 shows the frequency dependent (a) real (Z') and (b) imaginary (Z") impedance plots for the nanostructured YMnO₃ under 0 and 1.2T magnetic fields. Reduction in the real impedance with applied magnetic field suggests the presence of negative magnetoimpedance effect in YMnO₃. This can be ascribed to the magnetic field induced weak ferromagnetism in the present sample which acts as a driving force for 'zener double exchange' mechanism and hence impedance decreases with applied magnetic field. Also, imaginary impedance (Z") decreases with increase in magnetic field because of the mobility of the charges get increased as the spin aligned with magnetic field which further induces the activation of the charges in the crystal core.

4. Conclusion

In the present study, nanostructured YMnO₃ compound was successfully synthesized by cost effective, simple, vacuum free, easy and eco-friendly acetate precursor based modified Sol-Gel technique. Final product was sintered at 700oC to obtain the nanostructured phases in the compound. XRD measurement and analysis confirm the hexagonal unit cell structure with $p6_3cm$ space group. Average crystallite size, obtained using scherer's formula, is found to be ~ 26.33 nm. Recorded TEM image of presently studied YMnO₃ compound reveals the high enough crystallinity with the particle size almost similar to that obtained from the XRD analysis. With increase in applied magnetic field, dielectric constant of nanostructured YMnO₃ compound gets enhanced throughout the frequency range studied. This effect of magnetic field has been understood on the basis of magnetic field induced modifications in the electronic processes. Similarly, the relaxation mechanisms, possible for the present case of nanostructured YMnO₃, have been discussed in the light of UDR model. Nanostructured YMnO₃ compound shows a reduction in real and imaginary parts of impedance upon an application of magnetic field which has been understood in terms of field induced weak ferromagnetism.

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