Synthesis, Characterization and Impedance Spectroscopy Studies of $NdFeO_3$ Perovskite Ceramics

Jada Shanker[1], M. Buchi Suresh[2], D. Suresh Babu[3]

[1]Department of Physics, Osmania University, Hyderabad, Telangana, India

[2]International Advanced Research Centre for powder metallurgy and New Materials (ARCI), Hyderabad, Telangana, India

[3]Department of Physics, Osmania University, Hyderabad, Telangana, India

Abstract: ***The ac impedance, conductivity properties of $NdFeO_3$ perovskite ceramics in the (100Hz-1MHz) frequency range with different temperatures has been studied. Compound was prepared by sol-gel auto combustion technique and pellet was sintered at 900^0C. The preliminary structural analysis of the compound by X-ray diffraction technique confirmed its single phase orthorhombic structure. AC Impedance spectroscopy was used to correlate between microstructure and electrical properties of the compound .The presence of only grain i.e bulk effects in the compound was observed. The frequency dependent electrical data was used to study the conductivity mechanism. The analysis of the electrical impedance with frequency at different temperatures as provided some information to support suggested conduction mechanism.***

Keywords: Sol-gel auto-combustion method, Conductivity, Impedance and perovskite ceramics.

1. Introduction

Rare-earth (A) transition-metal (B) oxides belong to ABO_3 perovskite structure. In the recent years perovskite shows interesting properties like high temperature (Tc) Super conductivity, colossal magneto resistance and multi ferocity **[1]**. The orthoferrites with common formula $RFeO_3$ have been the subject of intense research for variety of applications due to their unique dielectric, magnetic, magneto-optical, magneto-electric, multiferroic and perovskites. Literature survey on this material reveals that no detailed work has been reported on the temperature and frequency dependence of electric properties. Therefore we have extensively studied the frequency and temperature dependence of dielectric and impedance properties of $NdFeO_3$ perovskite ceramics. In this paper we present our extensive studies on electrical properties of $NdFeO_3$.

2. Experimental procedure

Polycrystalline powder of NdFeO3 was prepared by sol-gel auto combustion technique. For making $NdFeO_3$ we have taken 1:1 molar ratio of Stoichiometric amounts of Nd_2O_3 and $Fe(NO_3)_2.9H_2O$ and 1:3 molar ratio of citric acid were taken separately. Nd_2O_3 is converted as neodymium nitrate using HNO_3.This precursor materials were dissolved separately with double distilled water and mixed using magnetic stirrer for homogeneous solution, then add NH_3 for set PH-7.This mixed solution were stirred and simultaneously heated to 150^0C to evaporate water and until sole was obtained, then ethanediol glycol was added as a fuel. The resulting gel was combusted. Collected powders were annealed (calcinated) at 750^0C for 4hours. Resulting powder was pelletized under uniaxial pressing. The pellet was sintered separately at 900^0C temperatures for XRD, SEM, Dielectric measurements.

3. Results and Discussion

3.1. Material characterization

XRD pattern of calcineted powder of NdFeO3 is shown in **Figure 1.** This XRD pattern indicates the presence of a single phase orthorhombic (JCPDF no 251149) symmetry. The Calculated lattice parameters of $NdFeO_3$ are found to be a= 5.580(1), b=7.761(2), C=5.449(1). The Surface morphology or microstructure (i.e grain size, grain distribution and voids) of sintered $NdFeO_3$ pellet shown in **inset Figure 1**. The grain are uniformly distributed throughout the surface and the average grain size is found to be order of 100nm.The density of the ceramics was found to be around 93% of theoretical density. The presence of voids indicates that the pellet has certain amount of porosity.

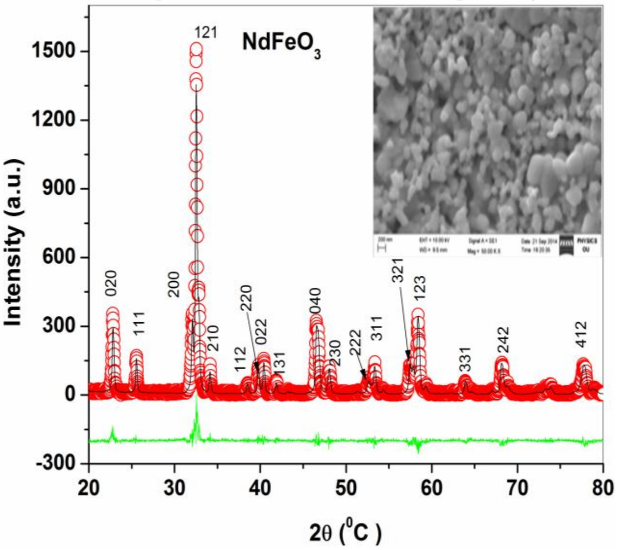

Figure 1: .XRD-rietveld refinement of Calcined powder of $NdFeO_3$. Open circles represent experimental data and solid curve shows calculated pattern. The curve below straight line

gives difference between the experimental and calculated pattern. **Inset Figure 1:** SEM Microstructure of sintered pellet of $NdFeO_3$.

3.2. Dielectric Measurements

Figure 2: Shows the frequency dependence of the dielectric constant (ε') of $NdFeO_3$ at the different temperatures. It is observed that dielectric constant (ε') decreases with increasing frequency at a given temperature. On increasing temperature, dielectric constant (ε') increases, which becomes even more significant at low frequency. The decrease in ε' is due to the space charges, which leads to the high dielectric constant and significant frequency dispersion [**2**], [**3**]. This suggests the thermally activated nature of the dielectric relaxation of the system [**3**].

Inset Figure 2: Shows the frequency dependence tanδ of $NdFeO_3$ at different temperatures. The loss tangent peak is observed at low frequency region ≥300^0C and peek shifts to words high frequency region with increase in temperatures, and also dielectric lose is increase with increase in temperature. The increase in the value of tanδ at high temperatures indicates the presence of dielectric relaxation in $NdFeO_3$ [**3**].

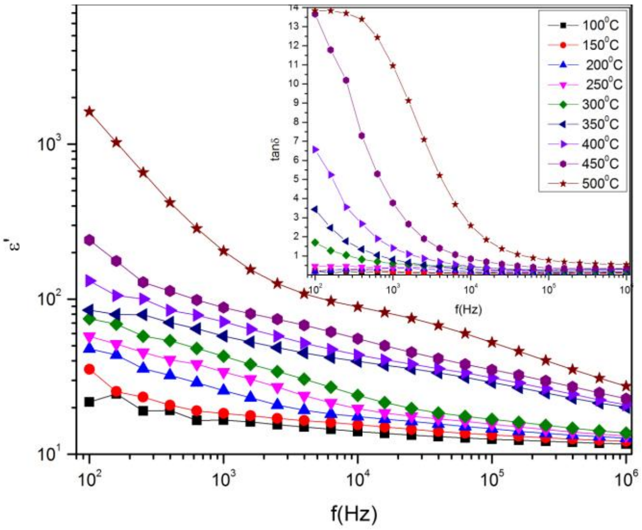

Figure 2: Frequency dependence of ε' and tanδ (inset) of $NdFeO_3$ at different temperatures.

3.3 Impedance Measurements

The frequency dependence of imaginary impedance (Z'') of $NdFeO_3$ at different temperatures is shown in **Figure 3:** A peak is observed in each curve. The position of the Z'' peak shifts towards higher frequency side on increasing temperature, and then a strong dispersion of Z'' exists. The width of the peak points toward the possibility of a distribution of relaxation times [**3**]. In such a situation, one can determine the relaxation time τ (= $1/\omega_{max}$) from the position of the peak.

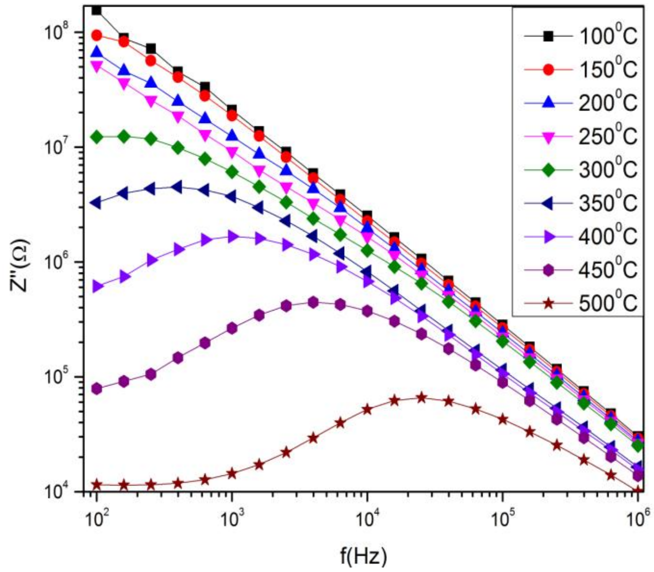

Figure 3: Frequency dependence of Z'' of $NdFeO_3$ at different temperatures

Figure 4: Shows the complex impedance spectrum Z' Vs Z" (Cole-Cole plot) of $NdFeO_3$ ceramic at different temperatures. The effect of temperature on impedance and related parameters of material become clearly visible with rise in temperature. An increasing temperature, the slope of the line decreases i.e they bend towards Z' axis and finally forms semicircles. Appearance of single arc or semicircle at a particular temperature shows that electrical properties in the materials arise mainly due to the contribution of bulk effects. The formation of full, partial or no semicircles mainly depends on the strength of relaxation and also experimentally available frequency range **[4]**.The electrical process taking place within the materials may be modelled (as an RC circuit) on the basis of the bricklayer model. The intercept of semicircular on Z'-axis gives the value of bulk resistance (R_b) of the sample, which is found to be decrease with increase in temperature. It suggests that material shows the negative temperature coefficient of resistance (NTCR) type behavior. These plots exhibit depressed semicircles having canters lying below the real axis confirming the presence of non-Debye type of relaxation phenomenon in the materials **[4]**.

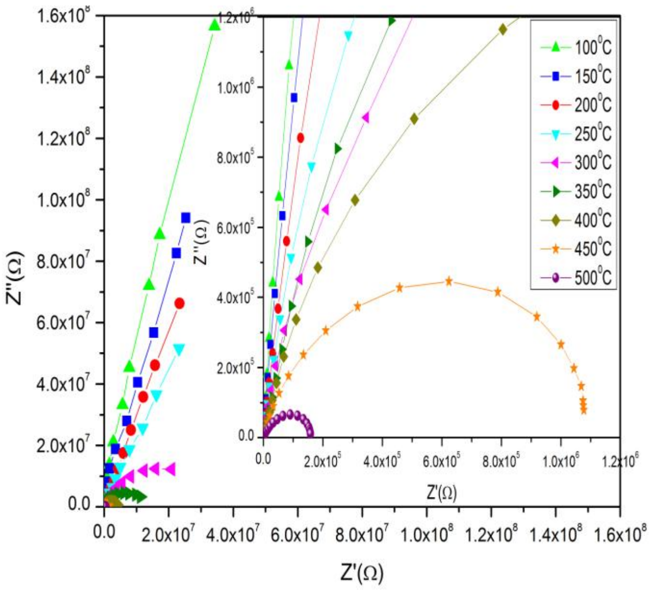

Figure 4: Z''-Z' (cole-cole) plots plots of $NdFeO_3$ at

different temperatures .**inset Figure** shows the Zoom view of same

Fig.5 shows Z''/Z''_{max} Vs f/f_{max} plots of $NdFeO_3$,the full width half maxima (FWHM) is observed that more than 1.14 decades, this behavior indicates, the observed relaxation in $NdFeO_3$ is of non-Debye type **[5].**

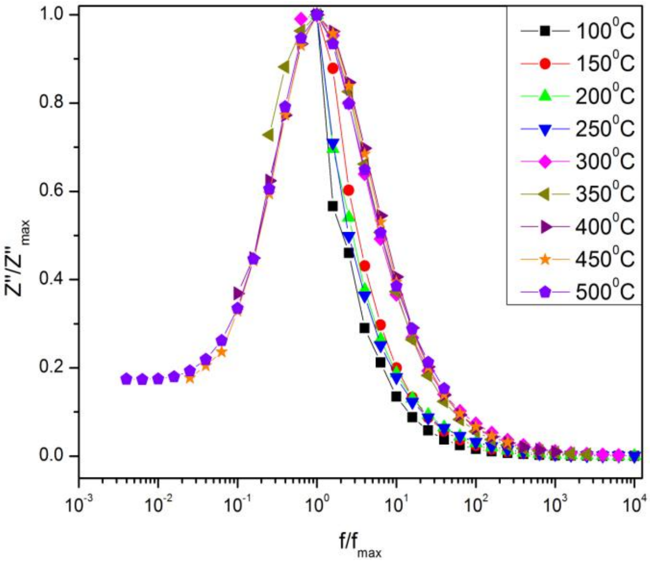

Figure 5: Z''/Z''_{max} V_s f/f_{max} plots of NdFeO3 at different temperatures

Fig. 6 shows variation of relaxation time (τ) as function of temperature. The relaxation time (τ) follows the Arrhenius law. The typical pattern suggest a temperature dependent relaxation process with spread of relaxation time in the range of 0.1-10^{-5} sec suggesting enhancement in the process dynamics in the material with rise in temperature **[3], [5]**. Fitting of these data to Arrhenius yields activation energy E_a as 1.05eV.

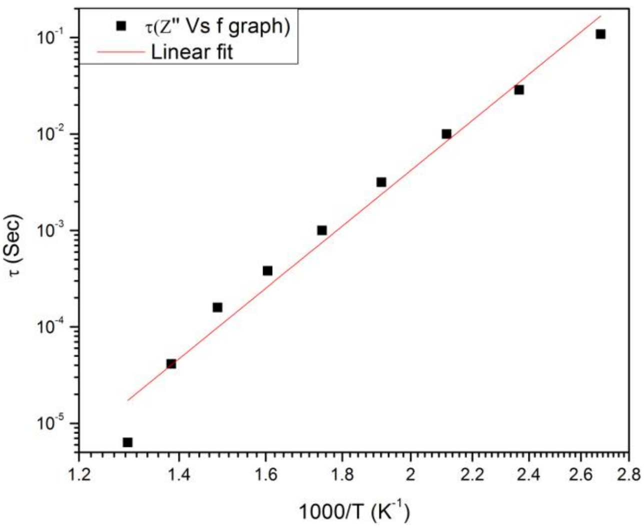

Figure: 6 Variation of relaxation time (τ) of $NdFeO_3$ as function of temperature

3.4 DC &AC Conductivity Analysis

Figure 7: Shows the dc conductivity (σ_{DC}) Vs 10^3/T plot of $NdFeO_3$.It indicates a pattern in which conductivity increases gradually with rise in temperature suggesting a thermally activated process in the materials [**3**]. The value of dc bulk conductivity has been calculated from the impedance data using the relation, $\sigma dc=t/R_bA$, where R_b is the bulk resistance, t is the thickness, and A is the area of the electrode deposited on the sample. The dc bulk (grain) conductivity graph follow the Arrhenius law. The evaluated activation energy has found to be 0.85.

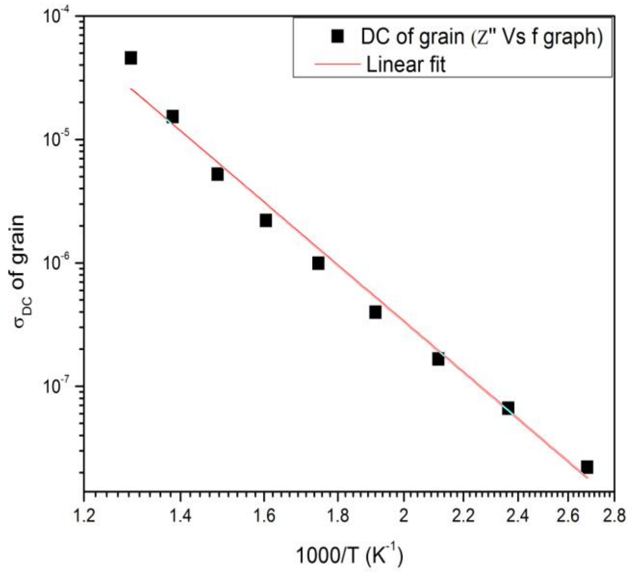

Figure 7: Variation of DC conductivity (σ_{DC}) of $NdFeO_3$ at different temperatures

Fig.8. Shows frequency dependence σ_{AC} plots of $NdFeO_3$ for various temperatures. AC conductivity of the samples were calculated from the impedance data, using the formula

$$\sigma_{AC}=2\pi f\varepsilon 0\varepsilon' \tan\delta$$

(Where f=frequency, tanδ = dielectric loss, ε_0 = dielectric constant in free space, ε'=dielectric constant)**[6]**. σ_{AC} remained constant at lower frequencies and increased rapidly at higher frequencies and increased with increasing temperature at all frequencies, which may be attributed to the increase in the number of charge carriers and their drifted mobility which are thermally activated [**6].**

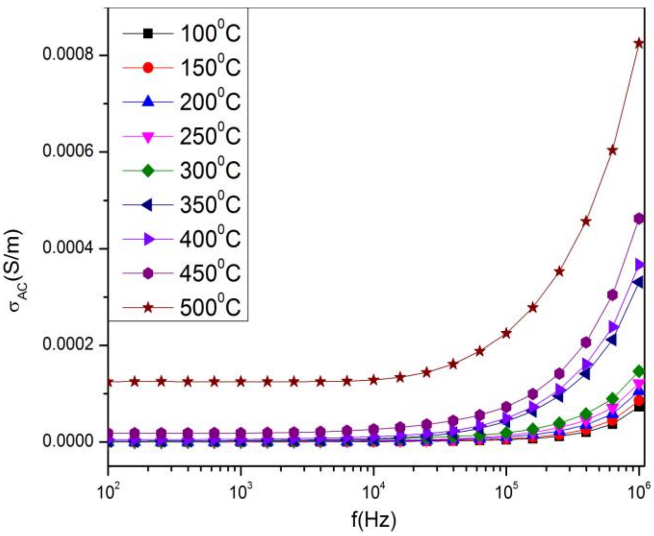

Figure 8: Frequency dependance of AC conductivity(σ_{AC}) of $NdFeO_3$ at different temperatures

4. Conclusions

Polycrystalline perovskite ceramic sample of $NdFeO_3$ were prepared by sol-gel auto combustion method. The phase formation of the $NdFeO_3$ sample is confirmed by XRD. The presence of phase charge polarization at higher temperature arises only due to the mobility of ions and imperfections in the material. The DC conductivity (σ_{DC}) and relaxation time (τ) follows the Arrhenius behavior. The real and imaginary parts of impedance properties investigated. The impedance analysis supports the typical behavior of negative temperature coefficient of resistance (NTCR) of materials. They also confirm the presence of non-Debye type relaxation phenomenon in the material.

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Author Profile

Jada Shanker did B.Sc degree from Kakatiya University, Warangal, in the year 2008. He has received M.Sc in Physics, from Osmania University, Hyderabad, in the year 2010. He is now doing Ph.D in Physics on the field of Multiferroic perovskite materials as well as he has been working in UPE-Project in the Dept. of Physics Osmania University Hyderabad. He is a Life Membership of Materials Research Society of India.

Dr. D. Suresh Babu obtained M.Sc., M.Phil., and Ph.D. in Physics from Osmania University, Hyderabad. He has been actively engaged in teaching and research for the last 35 years. He is recipient of young scientist award from Andhra Pradesh Academy of sciences, Hyderabad. He worked as postdoctoral fellow at Institut des Materiaux de Nantes, France. He visited USA, Germany, Spain, and France to carryout research work and presenting papers in International Conferences. Life Membership of Professional Bodies like Materials Research Society of INDIA, Instrument Society of INDIA, Andhra Pradesh Academy of Sciences, Association of Indian Science Congress and Magnetic Research Society of India. Presently he is working as a Professor and Chairman Board of studies in Physics at Osmania University. During his research carrier, he was involved in preparing ceramics of High Tc Superconductors, Colossal Magneto resistance materials, Colossal dielectric constant materials and Multiferroics (Perovskite oxides).

Dr. M. Buchi Suresh is working as scientist in ARCI, Hyderabad. He has done his Ph.D in physics with material science specialization. He has 10 years of experience in the field of polycrystalline oxide ceramics for various applications and published about 30 papers in international journals.