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Temperature Dependent Dielectric, Impedance Spectroscopy and Hopping Mechanism in NiFe₂O₄ Modified Pb_{0.75}Nd_{0.25}TiO₃ Based Multiferroic Composites

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ABSTRACT

Multiferroic composite ceramics of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFe}_2\text{O}_4)$ where $x = 0.50$ and 0.40 have been synthesized using mechanical mixing method. For multiferroic composites synthesis, both ferroelectric and magnetic component have been prepared individually. The mechanical mixing of NiFe₂O₄ with Nd³⁺ modified lead titanate (Pb_{0.75}Nd_{0.25}TiO₃) in above mentioned stoichiometric proportions. The temperature dependent electrical properties such as dielectric permittivity, electrical impedance and conductivity have been explored. The effect of NiFe₂O₄ on electrical conductivity and conduction behavior of prepared ceramic composites has also been studied. The value of dielectric permittivity (both ϵ' and ϵ'') first increases upto certain value of temperature and then decreases. The decrease in value of real dielectric permittivity with first increases upto certain value of Temperature and then starts decreases. The conductivity increases with increasing temperature. The increase of oxygen vacancies due to temperature may results for uninterrupted increase in electrical conductivity.

1. Introduction

Multifunctional materials which exhibit ferroelectric, magnetic or elastic ordering simultaneously become interesting candidates for various industrial field like storage devices, sensing applications, used as energy storage and energy harvesters etc. Those materials in which such types of ordering co exists are known as multiferroic and change of one of such ordering with effect of other ordering is termed as magneto-electric coupling and materials known as magneto-electric multiferroics. The multiferroic materials are either single phase or composites ceramics. The coexistence of such ordering contradicts each order because empty d-orbital results in Ferro electricity whereas partial filled or unpaired electrons responsible for magnetic ordering. So, it is impossible for materials show such type of behavior that is why multiferroic materials are fewer. Single phase materials can be synthesized using mixed perovskite approach in which partial substitution of transition metals in purely ferroelectric materials established the magnetic character in ferroelectric materials along with ferroelectric ordering. On the other side, composite multiferroics have been synthesized using mechanical mixing of ferroelectric and magnetic component (prepared individually). In composite multiferroic, ferroelectric ordering appears due to ferroelectric component whereas magnetic component gives magnetic ordering [1-8].

For synthesis of multiferroic composite, Nd³⁺ modified PbTiO₃ (Pb_{0.75}Nd_{0.25}TiO₃) has been selected as ferroelectric component whereas NiFe₂O₄ as magnetic component. The purpose of Nd³⁺ substitution in PbTiO₃ results in reduction of ferroelectric transition temperature (T_c^{FE}) and near T_c^{FE} , materials exhibit maximum value of dielectric constant. The modification of PbTiO₃ has been carried out using La³⁺ substitution at Pb²⁺ because rare earth reduces ferroelectric transition temperature reported by Ram et al. [9] and dielectric constant near transition temperature is supposed to be maximum. The variation of 'n' (Obtained from Universal Power Law fitting of conductivity data) vs. T(K) shows that samples initially follow small polaron tunnelling (NSPT) model up to 540 K before switching to the correlated barrier model above 540 K in Nd³⁺ modified PbTiO₃ reported by Akshey et al. [10].

So, in this research work, mechanical mixing method has been used for preparation of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFe}_2\text{O}_4)$ where $x = 0.50$ and 0.40

ceramic composites; and effect of NiFe₂O₄ on dielectric properties and conduction mechanism has been studied in details.

2. Experimental Methods

Multiferroic composite ceramics of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFe}_2\text{O}_4)$ where $x = 0.50$ and 0.40 have been synthesized using mechanical mixing method. For composites, both ferroelectric and magnetic component have been prepared individually. The ferroelectric component (Nd³⁺ modified PbTiO₃) has been synthesized using solid state reaction route (SSRR) whereas magnetic component (NiFe₂O₄) prepared by Auto combustion method. The composites of above-mentioned stoichiometric proportion have been prepared by mechanical mixing method. For solid state reaction route, metal oxides of required metal ion have been mixed using ball mill whereas for auto combustion method, metal nitrates of required metal ions dissolved in water individually first and then mixed step by step. The after complete mixing, powder in both cases left for dry. The dried powder calcined at 1000 °C for SSRR and 800 °C for auto combustion method. The calcined powder of both (ferroelectric and magnetic component) mixed in above mentioned stoichiometric proportion along with 2%wt polyvinyl alcohol (PVA) as binder. The PVA mixed powder mixed pressed into circular disc of ~ 8 mm in diameter and ~ 0.5 mm thick. The circular disc then sintered at 1200 °C for 2 hours. A conducting electrode has been deposited on circular disc for electric properties measurements. The electrical properties have been carried out by measuring Z vs. θ at different temperatures. The Impedance properties (Z vs. θ) have been performed over sintered disc using impedance analyzer E4990A from Keysight Technologies, India and furnace from Jupiter Engineering Works New Delhi interfaced with each other. The other parameters such as Z', ϵ' , and Z'', ϵ'' and σ_{ac} etc. have been calculated from Z and θ using standard formulae.

3. Results and Discussion

Temperature dependent ϵ' and ϵ'' vs. frequency in temperature range of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFe}_2\text{O}_4)$ where $x = 0.50$ and 0.40 ceramic composites in whole range of frequency (up to 1 MHz) has been shown in Figs. 1 and 2. It has been clearly envisioned from graphs that both real and imaginary part of dielectric permittivity exhibits maximum value before 1

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kHz (means lower frequency regime) afterward decreases continuously with increasing frequency confirmed that prepared samples exhibits behavior similar to behavior of normal dielectrics. The maximum value of both real and imaginary part of dielectrics (ϵ' and ϵ'') results due to existence of all polarizations (ionic, dipolar, electronic and space charge polarization) and continuous decrease followed by linear variation may result due to elimination some polarizations (space charge polarization) at higher frequency regime. The graphs clearly emphasizes that value of dielectric permittivity (both ϵ' and ϵ'') first increases upto certain value of temperature and then decreases. Such type of trend can be explained on such hypothesis that first increase may result due to easy orientation of dipoles with respect to applied field due amount of sufficient energy received by dipoles to get easily orientates or sufficient energy to overcome thermal barrier energy comes from external heat. Apart from this range of temperature, dipole unable to responds properly results in temperature dependent dielectric relaxation.

To further study in detail that either it may be Debye or non-Debye relaxation, experimental data has been fitted with theoretical model of Debye and non-Debye reported [11,12]. The decrease in value of real dielectric permittivity with first increases upto certain value of Temperature and then starts decreases. Such type of behavior may be explained on basis of thermal energy provided to dipoles from heating. Due to this, response of dipoles towards applied filed become easy results in increased in dielectric permittivity (real and imaginary part). Further increase of temperature made dipoles so relaxed that they unable to responds to applied signal results in further decreased in value of dielectric permittivity (real and imaginary part).

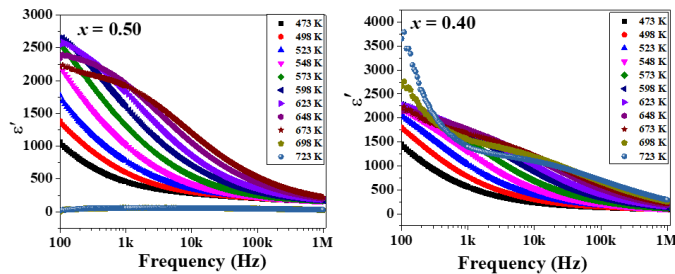


Fig. 1 ϵ' vs. frequency (Hz) at different temperature $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x=0.50$ and 0.40 ceramic composites

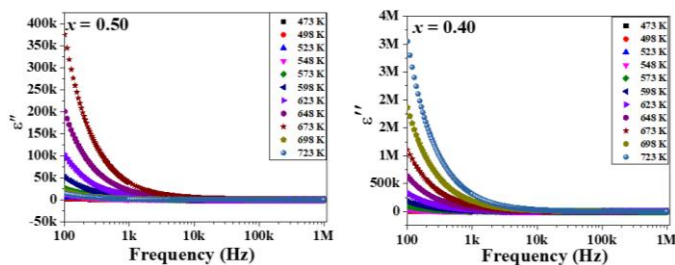


Fig. 2 ϵ'' vs. frequency (Hz) at different temperature $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x=0.50$ and 0.40 ceramic composites

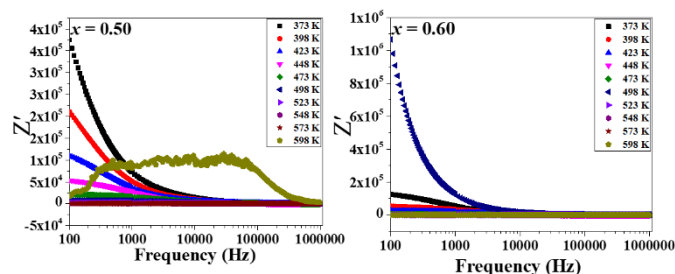


Fig. 3 Z' vs. frequency (Hz) at different temperature of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x=0.50$ and 0.40 ceramic composites

Z' vs. Frequency (Hz) in temperature range in temperature range of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x = 0.50$ and 0.40 ceramic composites in whole range of frequency (up to 1 MHz) has been shown in Fig. 3. The graphs clearly delineated that as frequency and temperature increases simultaneously, Z' decreases manifest that prepared samples exhibit negative temperature coefficient of resistance (NTCR) [13]. The decrease in value of Z' with increasing temperature directly proclaim the reduction in resistive properties followed by increasing conductive behavior due to increasing concentration of ferrite (NiFe_2O_4) in prepared ceramic composites. The decreased resistive behavior or barrier can also be justified from temperature dependent conductivity profile. The <https://doi.org/10.30799/jnst.106.26110206>

increase in conductivity with temperature may results due to increase in concentration of oxygen vacancies with increasing temperature [13,14].

Z'' vs. Frequency (Hz) in temperature range in temperature range of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x = 0.50$ and 0.40 ceramic composites in whole range of frequency (up to 1 MHz) has been shown in Fig. 4. The graphs clearly delineated that Z'' increases first continuously with increasing upto certain value frequency and then starts decreases with further increase of frequency clearly observed the presence dielectric relaxations. The continuous shift in maxima of Z'' vs. frequency directly evident for presence of temperature dependent dielectric relaxation [13-15] and merging at higher temperatures reveals for elimination of space charge polarization [16,17].

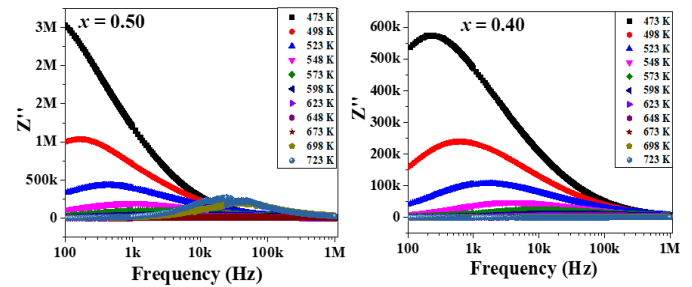


Fig. 4 Z'' vs. frequency (Hz) at different temperature of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x=0.50$ and 0.40 ceramic composites

To study the contribution of grain and grain boundaries, Nyquist plots has been studied. For this, Z'' vs. Z' graphs in temperature range of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x = 0.50$ and 0.40 ceramic composites have been plotted and studied and shown in Fig. 5. The semi-circle type plots in graphs clearly confirmed contribution of grain and grain boundaries in electrical properties. The grain boundaries have been considered as low permittivity region. Therefore, surface morphology plays an important role in electrical properties. The semi-circle types reveal variation of grain and grain boundary resistance and capacitance. Nyquist plots can fit using EC-lab software using an appropriate equivalent circuit of resistance (R) - capacitance (C) to calculate resistance and capacitance of grain and grain boundaries [18]. There may be two such combination of resistance and capacitance either it may be parallel combination of R_g and CPE_g represents grain circuit whereas R_{gb} and CPE_{gb} is for grain boundary circuit represent parallel combination. The phase element can be represented as CPE determines deviation from ideal Debye behavior. The impedance of CPE is given by $Z_{CPE} = 1/(j\omega)^\beta CPE$, where $\beta \leq 1$. The equation for the equivalent circuit can be represented by $Z^*(\omega) = Z' + jZ''$:

$$Z' = \frac{R_g}{1 + (\omega R_g C_g)^2} + \frac{R_{gb}}{1 + (\omega R_{gb} C_{gb})^2} \quad (3)$$

$$Z'' = \frac{\omega R_g^2 C_g}{1 + (\omega R_g C_g)^2} + \frac{\omega R_{gb}^2 C_{gb}}{1 + (\omega R_{gb} C_{gb})^2} \quad (4)$$

where (R_g , C_g , ω_g) and (R_{gb} , C_{gb} , ω_{gb}) corresponds to their usual meaning such as resistance, capacitance and frequency at the peaks of the semicircles for grain and grain boundaries respectively.

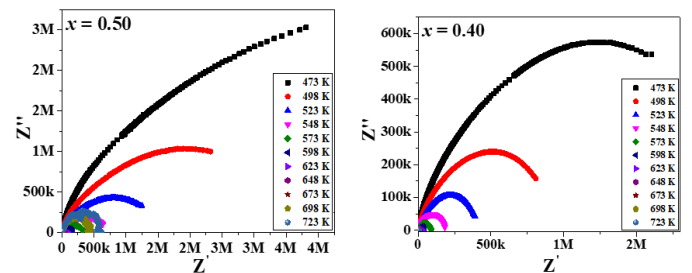


Fig. 5 Z'' vs. Z' at different temperature of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x=0.50$ and 0.40 ceramic composites

σ_{ac} vs. Frequency (Hz) in temperature range 373 K - 723 K of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x = 0.50$ and 0.40 ceramic composites in whole range of frequency (up to 1 MHz) in Fig. 6. It has been clearly seen in graphs that graphs divided into two parts (a) frequency independence represents to dc conductivity (σ_{dc}) and (b) varied with respect to frequency in higher frequency regime termed as ac conductivity (σ_{ac}). The universal Johncher's power law has been used to study

conduction mechanism from ac conductivity given as follow $\sigma_{ac} = \sigma_{dc} + A\omega^n$ where σ_{ac} = ac conductivity, σ_{dc} = dc conductivity, A = dispersion parameter representing the strength of polarizability and “n” is dimensionless parameter. It has been clearly perceived from graphs that conductivity increases with increasing temperature. The increase of oxygen vacancies due to temperature may result for uninterrupted increase in electrical conductivity [19-21]. Fig. 6 shows that electrical conductivity σ_{ac} vs. frequency in first increases upto 698 K and then decreases. The increase in value of electrical conductivity may be due to increase of oxygen vacancies created due to increase in temperature

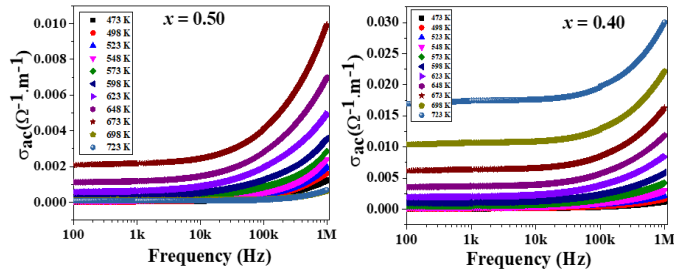


Fig. 6 σ_{ac} vs. frequency (Hz) at different temperature of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x = 0.50$ and 0.40 ceramic composites

4. Conclusion

Multiferroic composite ceramics of $x(\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3) - 1-x(\text{NiFeO}_4)$ where $x = 0.50$ and 0.40 have been synthesized using mechanical mixing method. For multiferroic composites synthesis, both ferroelectric and magnetic component have been prepared individually. The mechanical mixing of NiFe_2O_4 with Nd^{3+} modified lead titanate ($\text{Pb}_{0.75}\text{Nd}_{0.25}\text{TiO}_3$) in above mentioned stoichiometric proportions. The real part of dielectric permittivity first increases upto certain value of temperature and then starts decreases reveals that dipoles get sufficient energy from temperature for easy response to applied signal whereas further decrease shows that unable responds of dipoles towards applied signal. The real part of electrical impedance parameter decreases with increasing NiFe_2O_4 concentration and temperature shows decrease in resistive behavior which has been directly correlated with electrical conductivity behavior increasing ferrite content directly evident this.

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