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Effect of CoFe2O4 on Dielectric and Conduction Behavior Pb0.75La0.25TiO3 Based Composites

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***Abstract:** $_{(1-x)}$ $(Pb_{0.75}La_{0.25}TiO_3)$-$_x$ (Co_2FeO_4) where x = 0.50 & 0.60 Ceramic Composites have been successfully synthesized via mechanical mixing method. The substitution of $CoFe_2O_4$ with La^{3+} modified Lead Titanate $(Pb_{0.75}La_{0.25}TiO_3)$ in above mentioned stoichiometric proportions on has been explored for temperature dependent electrical properties. The both real and imaginary parts of dielectric permittivity decreases with increasing both temperature as well as ferrite content. The real part of impedance decreases with increasing temperature as well as Co_2FeO_4 content whereas maxima in imaginary part reveals the presence of temperature dependent dielectric relaxation. Both conductivity as well as dielectric loss increases with increase in Co_2FeO_4 content in stoichiometric proportion.*

***Keywords:** Multiferroics, Composites Material, Magneto-Dielectrics, Dielectrics, Impedance.*

1. INTRODUCTION

Magneto-electric multiferroics are the current research materials due to their vast usage in various field. The reason behind their importance in wide industrial field like storage devices, sensing applications, used as energy storage and energy harvesters due to co-occurance of both ferroelectric and magnetic orders. The simultaneous occurrence of these orders is important because these orders contradict to each orders. Such materials in which both ferroelectric and magnetic order occurs known as multiferroics and coupling in these ordering is known as magneto-electric coupling. Therefore multiferroic materials are defined as those materials in which more than one ferroic ordering like ferroelectric, ferroelastic, or magnetic ordering appears simultaneously. Tailoring the magnetic order with effect of electrical order or vice versa leads to an important effect termed as coupling effect in multiferroic materials due to which these materials become very important for future technology. [1-5].

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Multiferroics can be single phased mixed perovskites or composite ceramics and coupling can be defined in two types such as direct and indirect coupling. Single phase mutliferroic are $BiFeO_3$, $TbMnO_3$ whereas other type in which such properties appears in multiple phase materials in which ferroelectric ordering occurs from one phase and magnetic properties from other phase termed as multiferroic composites. Magneto dielectric effect in multiferroic composites can be realized by choosing suitable ferroelectric and ferromagnetic materials and designing their heterostructures appropriately [5-8].For this, $PbTiO_3$ has been selected for ferroelectric component whereas Cobalt ferrite as a magnetic component. The modification of $PbTiO_3$ has been carried out using La^{3+} substitution at Pb^{2+} 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 effect of magnetic field on dielectric constant termed as magneto-dielectric effect has also be reported by Tiku et al.[10].

So In this case, mechanical mixing method has been used for preparation of $_{(1-x)}(Pb_{0.75}La_{0.25}TiO_3)$- $_x(Co_2FeO_4)$ where x = 0.50 & 0.60 Ceramic Composites. The effect partial replacement of La^{3+} modified $PbTiO_3$ ($Pb_{0.75}La_{0.25}TiO_3$) with on dielectric, impedance and conductive properties have been thoroughly studied.

Experimental

$_{(1-x)}(Pb_{0.75}La_{0.25}TiO_3)$- $_x(Co_2FeO_4)$ where x = 0.50 & 0.60 Ceramic Composites have been prepared by using mechanical mixing of both ferroelectric as well as magnetic components prepared individually using solid state reaction route and Auto Combustion Method. For Solid State Reaction Route, oxides of required metals have been used whereas in Auto Combustion methods, nitrates of required metal weighed in above mentioned stoichiometric proportions. The collected mixed powder from ball milling machine and from Auto Combustion process dried and calcined at 1000 ˚C for phase formation individually. The calcined powders (Both) mixed in above mentioned stoichiometric proportion to prepare composites of $_{(x)}CoFe_2O_4$-$_{(1-x)}Pb_{0.75}La_{0.25}TiO_3$ (where x = 0.60, 0.50) by mixing both ferroelectric as well as magnetic components in mentioned stoichiometric proportion with 2 wt% PVA as binder and then pressed in pallets. The pallets were sintered at 1200˚C. The Impedance properties have been carried out over sintered pallets impedance analyszer from Keysight and furnace interfaced with each other from KEYSIGHT TECHNOLOGIES, India. The real and imaginary parts of Dielectric Permittivity, Impedance, Dielectric Loss and Electric Conductivity have been calculated using reported mathematical conversion formulae.

2. RESULTS & DISCUSSION

Figure 1 shows variation of real part of dielectric constant (ε') vs. frequency at different temperature of $_{(1-x)}(Pb_{0.75}La_{0.25}TiO_3)$- $_x(Co_2FeO_4)$ where x = 0.50 & 0.60 Ceramic Composites in temperature range vary from 373K to 598K. It has been clearly seen that in lower frequency region, value of dielectric constant is large but as frequency starts increases, dielectric constant starts decreases and after certain frequency range, dielectric constant becomes almost constant represent normal behavior of dielectric materials. The maximum value of ε′ in low-frequency region results due to responds of all polarizations such as dipolar, ionic, electronic or space charge polarization whereas lowering in value of ε′ in high-

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frequency range may be due to lack of contribution of all polarization to net dielectric constant and hence explained on basis of dipole relaxation phenomenon [11]. The graphs clearly depicts that with increasing temperature upto to 573K, ε′ increases continuously and after further increase in temperature to 598K, it decrease in $x = 0.50$ whereas in case of $x = 0.60$, ε′ continuously increases. The increase in value of ε′ with increase in temperature may because as temperature increases, sufficient energy to overcome thermal barrier energy comes from external heat at which sample get heated. Therefore ε′ increases upto to certain value of temperature and after that, it starts decreases because dipole no longer responds to electric signal at that temperature [12]. The decrease in value of real dielectric permittivity with increase of Co_2FeO_4 may results due to increasing conductive behavior or decrease of resistive behavior.

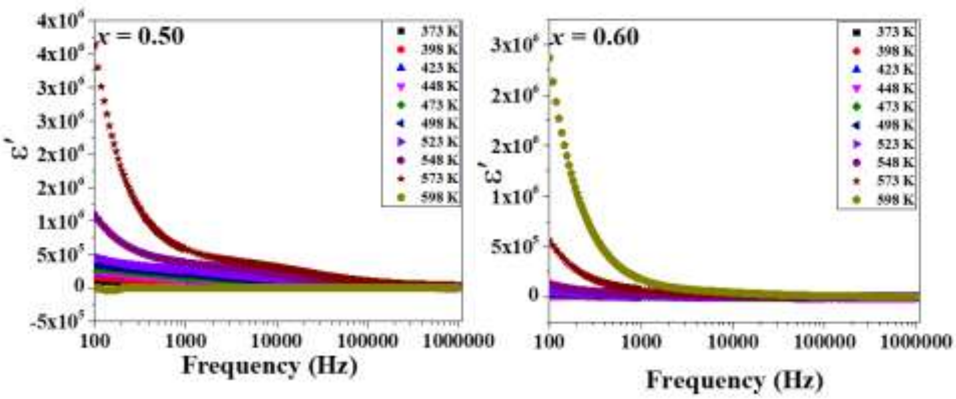

Figure 1: ε' vs. Frequency (Hz) at Different Temperature of $_{(1-x)}(Pb_{0.75}La_{0.25}TiO_3)$-$_x(Co_2FeO_4)$ where $x = 0.50$ & 0.60 Ceramic Composites

Figure 2 shows variation of imaginary part of dielectric constant (ε″) vs. frequency at different temperature of $_{(1-x)}(Pb_{0.75}La_{0.25}TiO_3)$-$_x(Co_2FeO_4)$ where $x = 0.50$ & 0.60 Ceramic Composites in temperature range vary from 373K to 598K. It has been clearly seen that in lower frequency region, value of imaginary part of dielectric constant is large but as frequency starts increases, dielectric constant starts decreases and after certain frequency range, dielectric constant becomes almost constant represent normal behavior of dielectric materials. The maximum value of (ε″) in low-frequency region and slowly regime results due to responds of all polarizations such as dipolar, ionic, electronic or space charge polarization whereas lowering in value of ε′ in high-frequency range may be due to lack of contribution of all polarization to net dielectric constant and hence explained on basis of dipole relaxation phenomenon [Kumar et al. 2002]. The graphs clearly depicts that with increasing temperature upto to 573K, (ε″) increases continuously and after further increase in temperature to 598K, it decrease in $x = 0.50$ whereas in case of $x = 0.60$, ε′ continuously increases. The increase in value of ε′ with increase in temperature may because as temperature increases, sufficient energy to overcome thermal barrier energy comes from external heat at which sample get heated. Therefore (ε″) increases upto to certain value of temperature and after that, it starts decreases because dipole no longer responds to electric signal at that temperature [12]. The

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similar behavior seen in real part of dielectric constant variation with temperature. This may results due to substitution of $CoFe_2O_4$ May results in increase in conductive behavior of La^{3+} modified Lead Titanate ($Pb_{0.75}La_{0.25}TiO_3$).

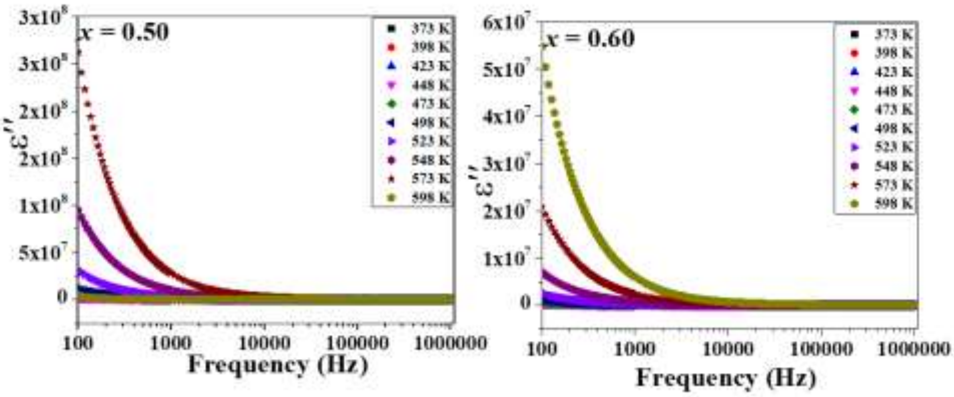

Figure 2: ε'' vs. Frequency (Hz) at Different Temperature of $_{(1-x)}$ ($Pb_{0.75}La_{0.25}TiO_3$)-$_x$(Co_2FeO_4) where x = 0.50 & 0.60 Ceramic Composites

Figure 3 shows variation of real part of impedance Z' vs. frequency at different temperature of $_{(1-x)}$ ($Pb_{0.75}La_{0.25}TiO_3$)- $_x$(Co_2FeO_4) where x = 0.50 & 0.60 Ceramic Composites in temperature range vary from 373K to 598K. The graphs clearly depicts the variation of Z'vs. frequency in temperature range 373K-598K are shown in figure 3. It has been clearly illustrated from graphs that Z'continuously decreases in magnitude with increase in temperature shows gives clear evidence for a materials to exhibit negative temperature coefficient of resistance (NTCR) [13]. The decrease in value of Z' with continuous increase in temperature with constant interval is direct evidence of increasing conductive behavior which also seen in temperature dependent conductivity profile and decrease of both real and imaginary part of dielectric permitivity. The almost constant variation or linear variation of Z' in higher frequency range results due to reduction in barrier properties. [13-14].

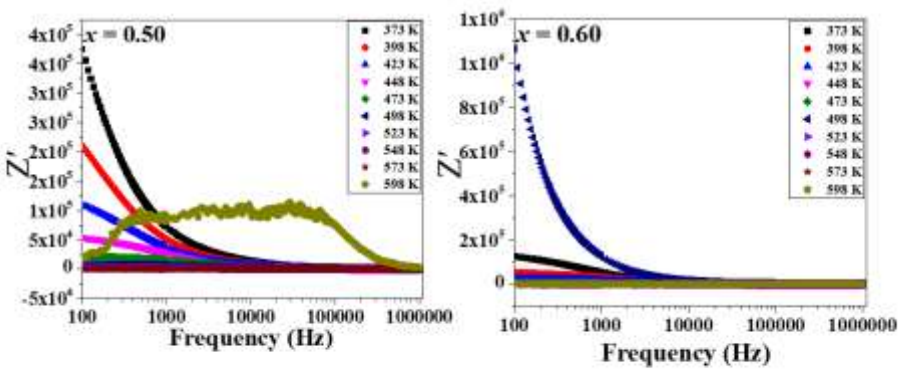

Figure 3: Z' vs. Frequency (Hz) at Different Temperature of $_{(1-x)}$ ($Pb_{0.75}La_{0.25}TiO_3$)-$_x$(Co_2FeO_4) where x = 0.50 & 0.60 Ceramic Composites

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Figure 4 shows variation of imaginary part of impedance Z'' vs. frequency at different temperature of $_{(1-x)}$ $(Pb_{0.75}La_{0.25}TiO_3)$- $_x(Co_2FeO_4)$ where x = 0.50 & 0.60 Ceramic Composites in temperature range vary from 373K to 598K. The continuous decrease in value of Z'' in higher frequency regime as well as with continuous increase in temperature interval results due to increase of conductive behavior of prepared ceramics. The maxima in Z'' vs. Frequency profile with continuous broadening with increasing both frequency as well as temperature range in regular interval shows presence of temperature dependent relaxation phenomenon due to immobility of charges at low temperature [13-15]. In high frequency regime, all peak merges at all temperature may results due to fade away of space charge polarization [16-17].

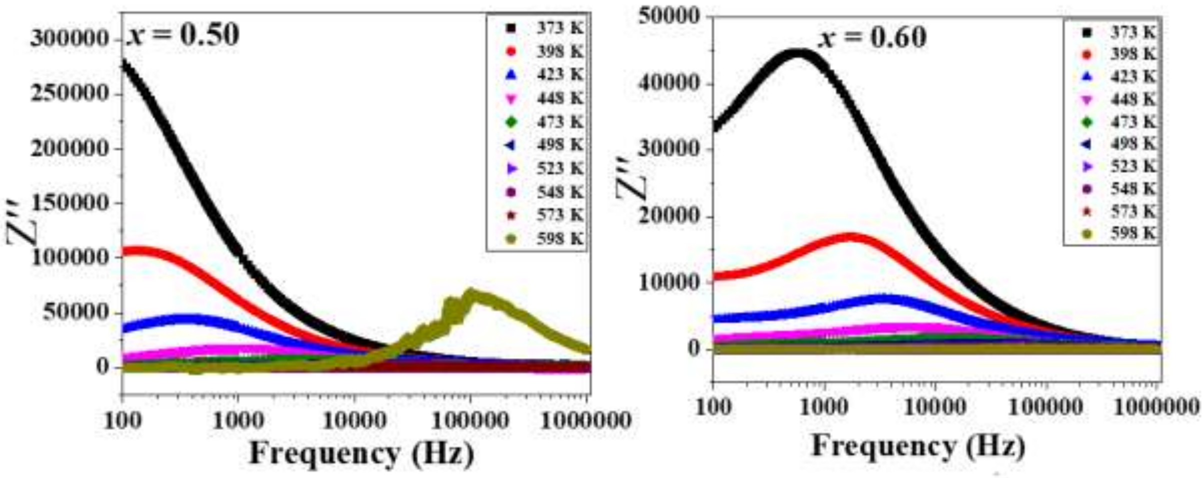

Figure 4: Z'' vs. Frequency (Hz) at Different Temperature of $_{(1-x)}$ $(Pb_{0.75}La_{0.25}TiO_3)$- $_x(Co_2FeO_4)$ where x = 0.50 & 0.60 Ceramic Composites

Figure 5 shows variation of electrical conductivity σ_{ac} vs. frequency in different temperature of $_{(1-x)}$ $(Pb_{0.75}La_{0.25}TiO_3)$- $_x(Co_2FeO_4)$ where x = 0.50 & 0.60 Ceramic Composites in temperature range vary from 373K to 598K. The σ_{ac} vs. frequency profile has been divided into two regime. Lower frequency regime in which electrical conductivity σ_{ac} varies linearly w.r.t frequency termed as dc conductivity (σ_{dc}) whereas in higher frequency range, it starts increases abruptly known as ac conductivity (σ_{ac}). The universal johncher's power has been used to study conductivity data of prepared samples as follow

$$\sigma_{ac} = \sigma_{dc} + A\omega^{n}$$

Where the symbols have their usual meaning

It has been clearly seen from conductivity profiles at different temperatures shows that conductivity continuously increases with increasing temperature with regular interval. The continuous increase of electrical conductivity may results due to increase of oxygen

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vacancies with temperature. Apart from this, it has also be clear from graphs that with increase of $CoFe_2O_4$ content, conductivity also increases means that with increasing ferrite content, conductive behavior of prepared ceramics has also be increase. The variation of Dielectric loss (Tan δ) vs. Frequency (Hz) also confirm that the ferrite content increase the conductive behavior of samples [19-21].

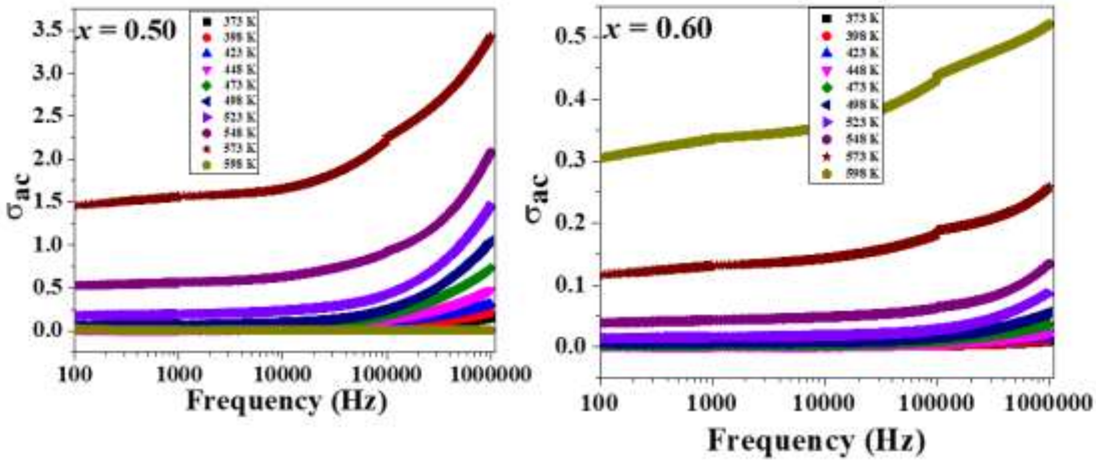

Figure 5: σ_{ac} vs. Frequency (Hz) at Different Temperature of $_{(1-x)}(Pb_{0.75}La_{0.25}TiO_3)$-$_{x}(Co_2FeO_4)$ where x = 0.50 & 0.60 Ceramic Composites

Figure 6 shows variation of Dielectric Loss (Tan δ) vs. Frequency (Hz) in different temperature of $_{(1-x)}$ $(Pb_{0.75}La_{0.25}TiO_3)$- $_{x}(Co_2FeO_4)$ where x = 0.50 & 0.60 Ceramic Composites in temperature range vary from 373K to 598K. It has been clearly visualized from graphs that the dielectric loss continuously increases with increasing $CoFe_2O_4$ content results in increasing conductive behavior. The increase in conductive behavior has been directly visualized from both increasing conductivity, decreasing real part of dielectric permittivity as well as impedance parameter.

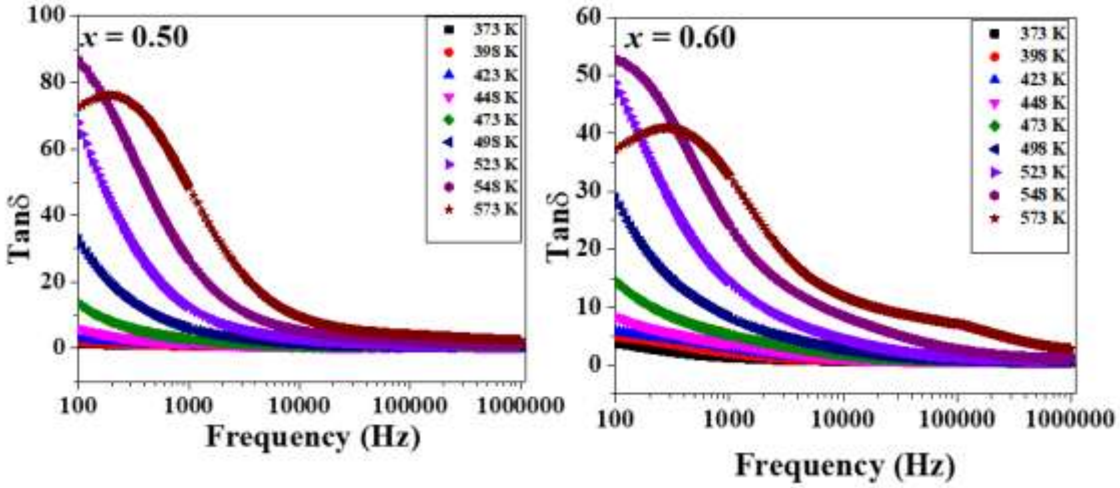

Figure 6: Dielectric Loss (Tan δ) vs. Frequency (Hz) at Different Temperature of $_{(1-x)}$ $(Pb_{0.75}La_{0.25}TiO_3)$- $_{x}(Co_2FeO_4)$ where x = 0.50 & 0.60 Ceramic Composites

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3. CONCLUSION

$_{(1-x)}$ $(Pb_{0.75}La_{0.25}TiO_3)$- $_x(Co_2FeO_4)$ where x = 0.50 & 0.60 Ceramic Composites have been successfully synthesized via mechanical mixing method. The phase purity and orthorhombic structure of these samples was confirmed using X-ray diffraction technique reported by Tiku et al. The real part of dielectric permittivity and Impedance parameter decreases with increasing $CoFe_2O_4$ concentration shows decrease in resistive behavior with increase of ferrite content. The increase in conductive behavior with simultaneous decrease in conductivity has also been confirmed from increases in conductivity as well as dielectric loss with increasing ferrite content directly evident this.

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