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Portrayal of Temperature Dependent Dielectric, Electric Modulus and Conduction Response in $_{1-x}$(CuO)-$_x$(Fe_2O_3) Ceramic Composites

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ABSTRACT

$_{1-x}$(CuO)-$_x$(Fe_2O_3) ceramic composites (x = 0.05, 0.10, 0.15 & 0.20)have been successfully prepared using mechanical mixing method of CuO and Fe_2O_3oxides. CuO and Fe_2O_3synthesized individually using Auto-Combustion method and composites of these oxides prepared using mechanical mixing method in above mentioned stoichiometric proportions. The effect of Fe_2O_3 on electrical properties such as dielectric permittivity, electrical modulus& conductivity with temperature have been studied.

Keywords: **Composites material; Conductivity, Dielectric Relaxation, Debye Behavior, Electrical Modulus**

INTRODUCTION

Due to scientific and technological importance of semiconducting and microelectronic industry in daily life usages, materials with semiconducting properties as well as tunable band gap are current research interest. Among such type of materials, metal oxides belongs to transition metals are prime candidates in field of semiconductors as well as microelectronic applications due to their unique, diverse, and rich physical as well as chemical properties. Transition metal oxides have been used electrode materials in various components of electronic industries in past few years. Semiconductor nanocrystal materials with narrow band gap attracts researcher from various sectors of industry including semiconducting applications, Dye Solar Cell, Catalysts etc. Because of their numerous, varied, intriguing, and rich physical and chemical properties, metal oxides are important candidates for scientific and technical investigation. Copper oxide (CuO) is a basic, most stable metal oxide belongs to family of transition metal oxides is widely attainable, environmental friendly, non-toxic, magnetic semiconductor most commonly used in various sectors of electronic industry. Because of quantum confinement effect, band gap of copper oxide, a p-type semiconductor can be tailored from ~1.2 to ~2.1 eV.

The excellent mechanical strength, thermal stability, electrical conductivity, spin dynamics, electrochemical, super-hydrophobic, photovoltaic and biocidal activities are all displayed by copper oxide nanostructures. Among all these aspects, CuO can also be used optoelectronic devices, sensors, catalysis, lithium-ion batteries, high critical temperature superconductors, super-capacitors, magnetic storage media, solar energy conversion, smart windows, optical limiters, thin film transistors, field emission emitters, and biomedicine. CuO has a space group of C2/c and crystallizes in a monoclinic crystal structure. There are four CuO molecules in a unit cell of cupric oxide. The magnetic interaction between Cu^{2+} ions is mainly mediated through the neighboring O^{2-} ions, and it depends on the Cu–O–Cu bond angle. Current research on pure and modified CuO has enormous potential in realm of microelectronics because of its extraordinarily high dielectric constant.[1-13].The particles have a regular shape, a constrained size range, and a high level of purity, according to Zhu et.'s examination into the structural characterization of CuOnano-particles made by microwave irradiating copper (II) acetate and sodium hydroxide as starting material. Kim et al. looked into the structural, optical, and electrical properties of monoclinic CuO nanoparticles [9]. The CuO nanoparticle's O is 1s and cu 2p peaks were visible in the x-ray photoelectron spectroscopy profile. At ambient temperature, a CuO nanoparticle's band gap was found to be 3.63 eV [9]. The conductivity of 0.16 S/m and dielectric constant of 14.5 has been reported by Atchaya et al. [8]

In this paper, effect of Fe_2O_3 on dielectric, Electric modulus and conduction behavior of Fe_2O_3modified ofCuO composites ($_{1-x}$(CuO)-$_x$(Fe_2O_3) ceramic composites (x = 0.05, 0.10, 0.15 & 0.20) synthesized using Mechanical mixing method have been reported.

EXPERIMENTAL METHODOLOGY & CHARACTERIZATION

Mechanical mixing technique has been employed to prepare ceramic composites of $_{1-x}CuO$-$_xFe_2O_3$ (x = 0.05, 0.10, 0.15, & 0.20). A planetary ball milling machine has been employed for milling procedure. Using a mechanical mixing technique, pure CuO and Fe_2O_3 of analytical quality that were bought from Sigma Aldrich were combined in the aforementioned stoichiometric ratio. Next, prepared CuO and Fe_2O_3 were weighed according to the weight percentage indicated above, and they were ball milled in the presence of acetone or propanol, a non-reacting solvent. For this, zirconia balls, powder, and non-reacting solvent were put into bottles and processed for a full day.

Following the milling procedure, the powder was removed from the bottles and stay for dry. As a binder, 2% percent polyvinyl alcohol was used. Weighed polyvinyl alcohol dissolved in water and stirred until clear solution has been obtained. Solution of polyvinyl alcohol and water was completely mixed with the ball-milled powder weighed in specified stoichiometric proportion. The combined powder was compressed into a disc having diameter of 10 mm and a thickness of roughly 0.5 mm. For densification, the disc were sintered at 900 °C. The conductivity, dielectric, and impedance characteristics of the sintered pallets have been described. For these measurement, electrode (Circular Shaped circle) of conducting material (Either Silver or Gold conducting paint) has been used.

Characterization Methodology Used: Complex Impedance Spectroscopy:
Complex impedance spectroscopy has been used to study the real and imaginary components of electrical impedance as well as dielectric permittivity. The conductivity has also be studied. In material science, this is a valuable and significant characterization tool. The impedance analyzer from Keysight Technologies (E4990A) was used to determine these electrical characteristics. Using established formulas, all of these properties were computed from empirically obtained Z vs θ at different temperatures.

RESULTS & DISCUSSION

Dielectric Spectroscopy
Figures 1 illustrate temperature-dependent ε' vs. frequency in given temperature range 323K-548K of ceramic composites of $_{1-x}CuO$-$_xFe_2O_3$ (x = 0.05, 0.10, 0.15, & 0.20) in entire frequency range from 100 Hz-10^6Hz. Graphs clearlyproclaimed that prepared composites exhibits maximum value of dielectric constant (Real part of Dielectric permittivity (ε')) in lower frequency range varied from 100 Hz to 1 kHz and afterward, it starts decreases up to certain value of higher frequency region (up to ~5 KHz). Beyond this maximum value of frequency, value of dielectric constantbecome almost constant as frequency further increases in higher frequency region (Afterward from ~10 KHz)endorsed that prepared composite samples stamped for presence of normal dielectric behavior.The graphs clearlydemonstrate that maximum value of real part of dielectric permittivity in lower frequency regime (up to 1 kHz) may results due to contribution of maximum polarization named as Ionic, Dipolar, Electronic & Space Charge Polarization in value of dielectric constant and as frequency starts increases, contribution of polarization starts excluding results in decrease in value of dielectric constant.

As frequency reaches to certain maximum value of frequency, maximum polarization get eliminated and dielectric constant behaves almost constant as frequency further increases. It has been clearly visualizes from graphs that value of dielectric permittivity (ε') increases with increasing temperature up to 548 K achieved maximum value of dielectric constant. The increase in value of dielectric permittivity with increasing temperature can be due to maximum orientation of dipoles when external field has been applied. This may happen because minimum amount of energy required by dipoles to orient has been received when sample get heated to overcome thermal barrier energy. To further study in detail that either it may be Debye or non-Debye relaxation, experimental data can been fitted with theoretical model of Debye and non-Debye reported. [14-19].

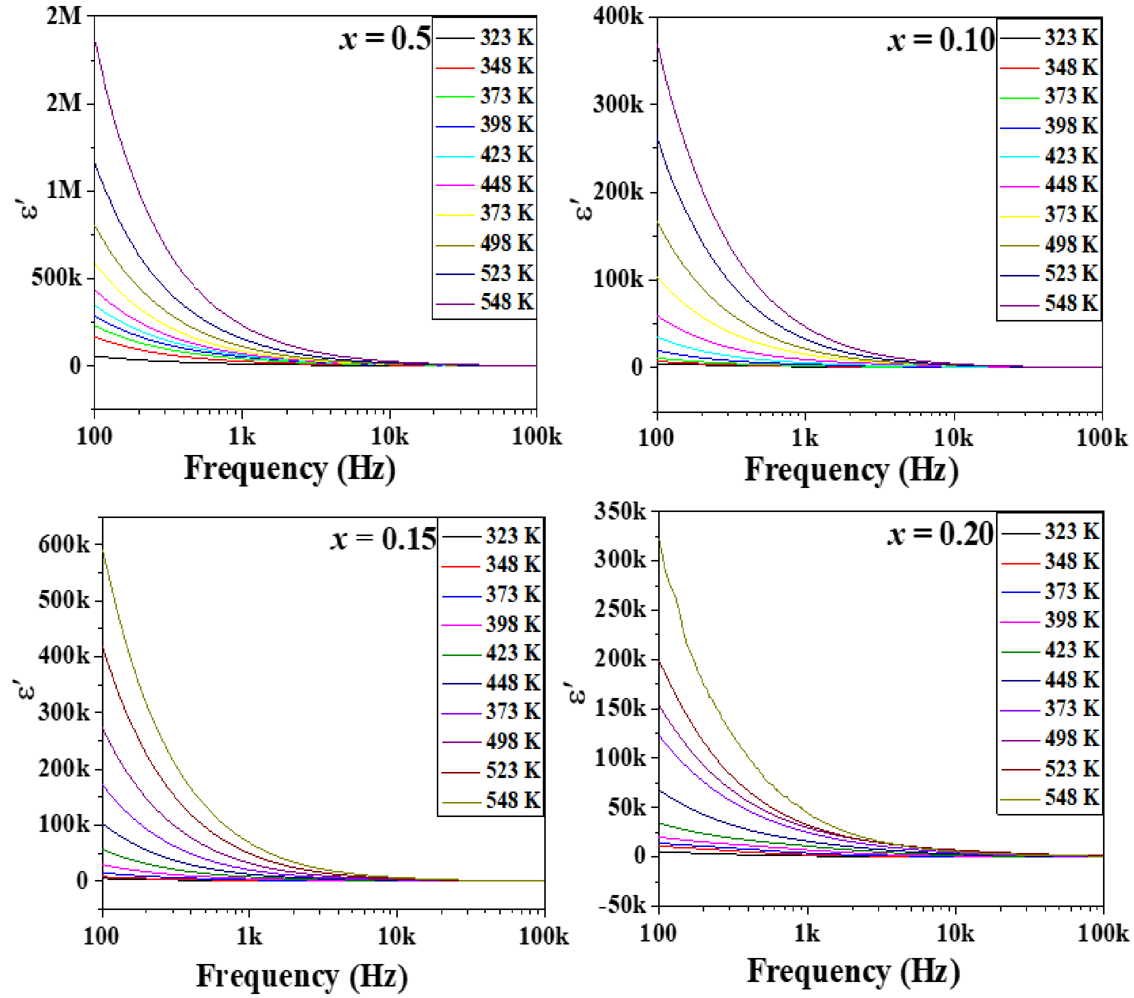

Figure 1: ε' vs. frequency temperature range 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20)

Figures 2demonstrateε" vs. frequency temperature in temperature varies from RT 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20) in entire frequency range from 100 Hz-10^6Hz. The graphs clearly shows that synthesized composites exhibits maximum value of dielectric constant (Imaginary part of dielectric permittivity (ε")) in frequency range varied from 100 Hz to 1 kHz and afterward it starts decreases up to certain value of frequency in higher frequency region (up to ~5 KHz). After this value of higher frequency region (Afterward from ~10 kHz), value of dielectric constant behaves almost linear variation according to frequency endorsed that prepared composite samples exhibits dielectric behavior similar to normal dielectric behavior. The graphs clearly represents that maximum value of real part of dielectric permittivity in lower frequency regime (up to 1 kHz) may results due to contribution of maximum polarizationstermed as Ionic, Dipolar, Electronic & Space Charge Polarization in value of dielectric constant and as frequency starts increases, contribution of such polarization starts excluding results in decrease in value of dielectric constant and in higher frequency regime, only space charge polarization can contribute in value of dielectric constant. It has been clearly visualizes from graphs that value of dielectric permittivity (ε") increases with increasing temperature up to 548 K achieved maximum value of dielectric constant.

The increase in value of dielectric permittivity with increasing temperature can be due to facile orientation of dipoles in presence of external field as well as temperature has been applied. This may happen because essential amount of energy that has been required by dipoles to orient received when sample get heated for overcome thermal barrier energy. Therefore it has been clearly depicted from graphs that value of dielectric constant increases continuously with increasing temperature. To further study in detail that either it may be Debye or non-Debye relaxation, experimental data may be fitted with theoretical model of Debye and non-Debye reported. [14-19].

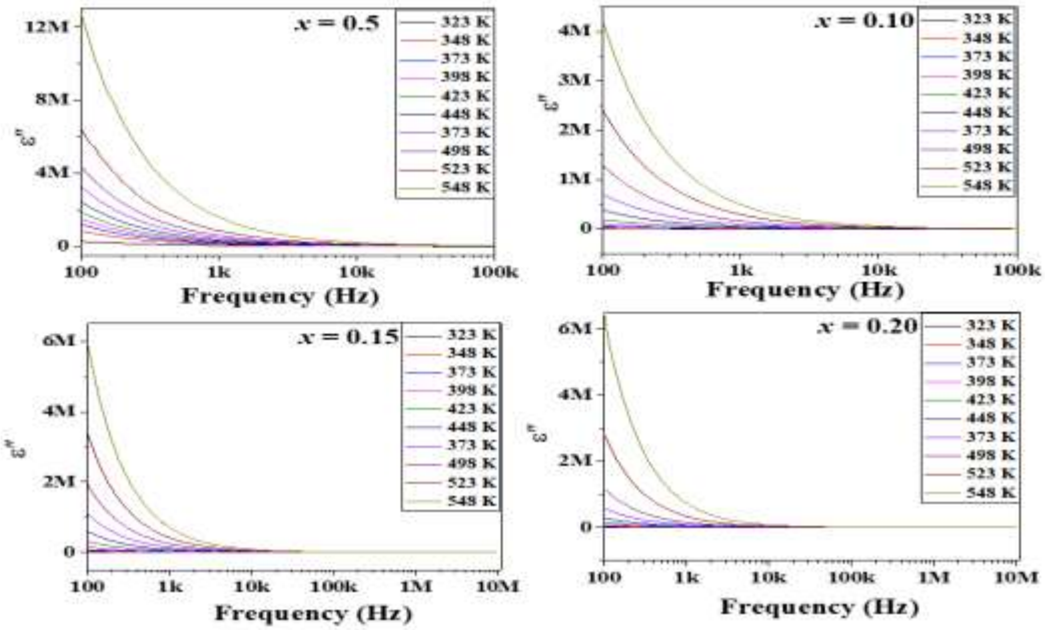

Figure 2: ε'' vs. frequency temperature range 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20)

Electric Modulus

Figures 3revealed variation of M' vs. frequency temperature in temperature varies from RT 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20) in entire frequency range from 100 Hz-10^{6}Hz. The graphs clearly delineate thatas frequency and temperature increases concomitantly, M' decreases manifest that prepared composite samplesexhibit negative temperature coefficient of resistance (NTCR) [15-16] which can also be verified from decrease in value of real part of impedance (Z') as well as continuous increased in conductivity discussed in next section. The decrease in value of M' with increasing temperature clearly proclaim that as temperature increases, resistive response in prepared ceramic samples starts reduces results in anincreased response in conductive behavior may results due to increasing concentration of iron oxide (Fe$_2$O$_3$) as well as concentration of oxygen vacancies formed when sample get heated. The decreased resistive response or barrier properties can also be explained from temperature dependent conductivity response. The increase in conductivity with temperature may results due to increase in concentration of oxygen vacancies with increasing temperature [15-16].

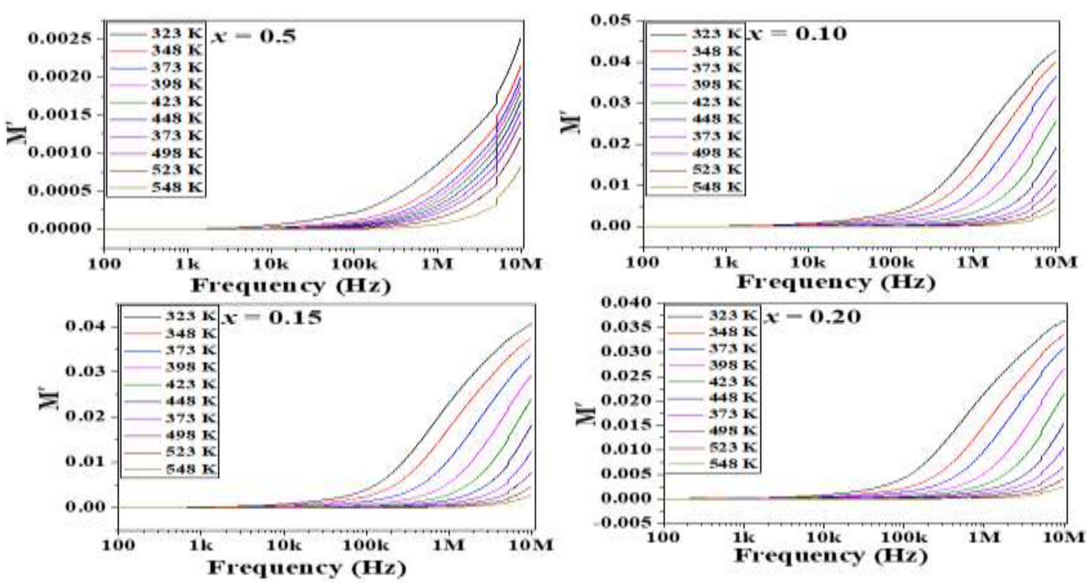

Figure 3: M' vs. frequency temperature range 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20)

Figures 4represents variation of M'' vs. frequency temperature in temperature varies from RT 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20) in entire frequency range from 100 Hz-10^6Hz. It has been clearly portrayed from graphs that M''continuously decreases as both frequency as well as temperature has been increased continuously clearly manifestfor presence dielectric relaxations. In sample for x = 0.5, absence of maxima shows absence of dielectric relaxation whereas in samples x = 0.10, 0.15 & 0.20, maxima represents presence of frequency dependent dielectric relaxations.

As temperature increases continuously, maxima in all samples except x = 0.5 shifts towards maximum value of frequency evidence for presence of temperature dependent dielectric relaxation. In sample x = 0.5, sample does not exhibits frequency as well as temperature dependent dielectric relaxation.The continuous shift in maxima of M''vs. Frequency with temperature accounted for presence of temperature dependent dielectric relaxation [13-15] and merging at higher temperatures reveals for elimination of space charge polarization [16-17].

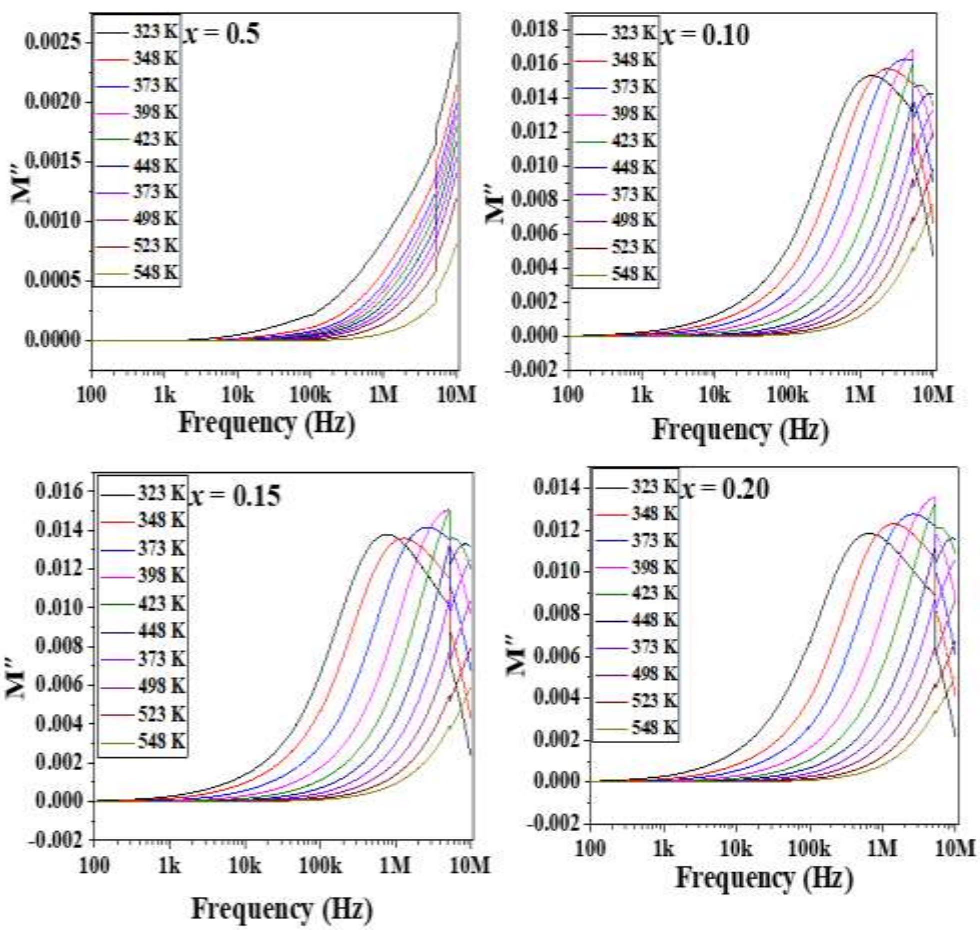

Figure 4: M'' vs. frequency temperature range 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20)

Conductivity & Conduction Mechanism

Figures 5shows variation of σ_{ac} vs. Frequency (Hz) in temperature varies from 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20) in entire frequency range from 100 Hz-10^6Hz.It has been clearly visualized from graphs that graphs composed of two parts (a) Linearly variable or frequency independent behavior termed as σ_{dc}(dc conductivity) & (b) Frequency dependent or frequency varied termed as σ_{ac} (ac conductivity) shown in figure 5.

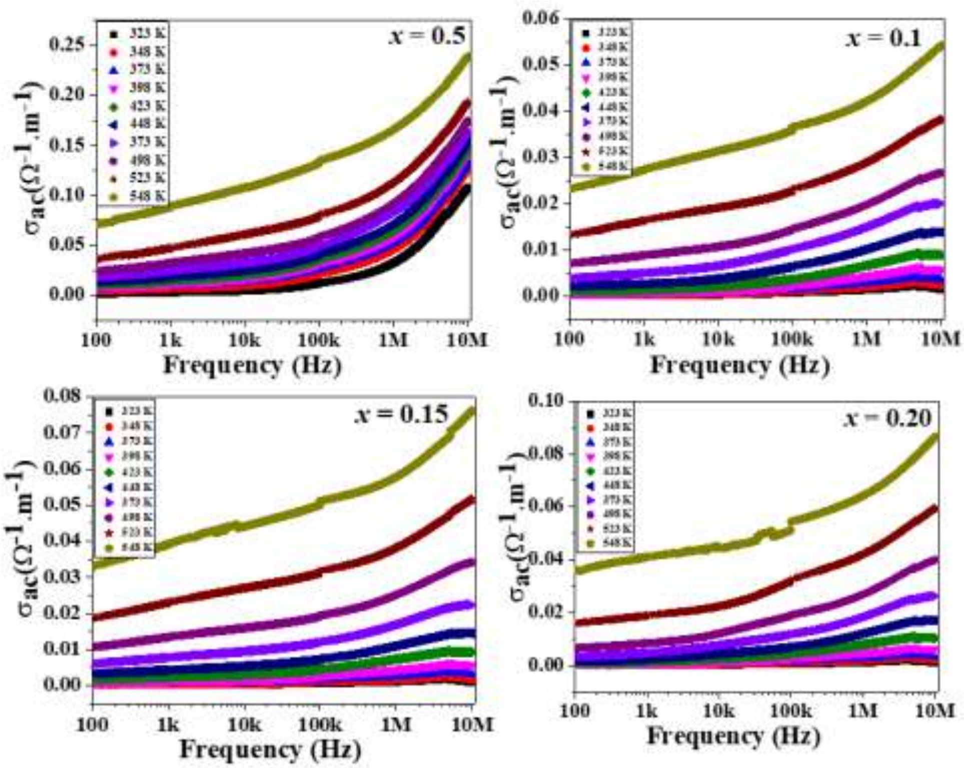

Figure 5: σ_{ac}vs. frequency temperature range 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20)

The conductivity continuously increases with increasing both temperature as well as content of Fe_2O_3 shows increase in conductive behavior of prepared ceramic composites which also verified from temperature dependent electric modulus (M'). The increase in conductivity with temperature may results due to increase in concentration of oxygen vacancies with increasing temperature. To further study conduction mechanism, experimentally collected has been fitted using universal johncher's power law 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&"n" is dimensionless parameter [20-22].

CONCLUSION

$_{1-x}$(CuO)-$_x$(Fe$_2$O$_3$) ceramic composites (x = 0.05, 0.10, 0.15 & 0.20) have been successfully prepared using mechanical mixing method of CuO and Fe_2O_3oxides. The both real and imaginary part dielectric (ε' & ε") increases with increasing temperature can be due to maximum orientation of dipoles when external field has been applied may happen because minimum amount of energy required by dipoles to orient has been received when sample get heated to overcome thermal barrier energy. The decrease in value of $M'$with temperature results for reduction in resistive behavior with simultaneous increased conductive response of prepared ceramic composites. The increased in conductivity with both temperature as well as content of Fe_2O_3may results due to increase in concentration of oxygen vacancies.

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