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RESEARCH ARTICLE

EFFECT OF TEMPERATURE AND FREQUENCY ON DIELECTRIC PROPERTIES OF TITANIUM SUBSTITUTED MANGANESE -ZINC FERRITE SYSTEM $Mn_{0.8+X}Zn_{0.2}Ti_XFe_{2-2X}O_4$ WITH X=0.10

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ABSTRACT

In this work the authors synthesized a new crystalline ceramic Titanium Substituted Manganese -Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10 by conventional solid state reaction route involving mechanical mixing, ball milling, calcination and sintering. Most important dielectric properties of the material such as dielectric constant, dielectric loss, and impedance of the material are evaluated as a function of temperature and frequency. Variation of electrical conductivity with frequency is found out and it is verified to be in accordance with Jonscher's power law.

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INTRODUCTION

Dielectric ceramics are highly desirable in this technological era because of its flexibility with its comparatively low processing temperature, high dielectric constant, low dielectric loss and high dielectric strength which enables its use for a wide range of applications such as embedded capacitors, electronic packaging, high density ceramic capacitors and dielectric resonators. Investigations in the field of dielectric ceramics are driven by the market potential of next generation memories and transducers. Thin films of dielectrics are finding very much significance in the area of MEMS applications. MEMS are finding sophisticated applications in accelerometers for air bag deployment in cars, micro-motors and pumps, micro heart valves, medical, automotive, and space applications. Highly sensitive sensors and actuators based on thin and bulk films of dielectric ceramics revolutionize day to day life with spectacular gadgets. New nano crystalline ceramic materials and their composites are important components in a large number of military and space applications.This work is aimed to find out the effect of temperature and frequency on the most significant dielectric parameters of a nanocrystalline dielectric ceramic material namely dielectric constant, impedance, conductivity and dielectric loss of Titanium Substituted Manganese - Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10. The impedance, dielectric permittivity, electrical conductivity and loss factor are very crucial quantities in the designing of a practical device. Ferrites are ferromagnetic semi conductors and the need for high resistivity ferrites led to the synthesis of various ferrites. The influence of various substituents like Ti, Zn etc considerably change in its electrical properties (Smit, 1959). Polycrystalline ferrites have very good dielectric properties (Bragg, 1915). Ferrites having very high dielectric constants are useful in designing good microwave devices such as isolators, circulators etc. Manganese-Zinc Ferrite $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ in the spinel structure is a low cost material which is generally useful for microwave - devices and memory-core applications. Due to Ti substitution electrical and magnetic properties changes and hence have much technological merits (Tarte, 1963; Srinivasan, 1980; Arumugam, 1987). The analysis and study of dielectric constant, conductivity, impedance and dielectric loss factor, as a function of temperature and frequencies is one of the most important, convenient and sensitive methods of studying the polymer structure (Scott, 1962). The dielectric properties of a number of polymers (dielectric constant, dielectric loss, impedance and conductivity) have been studied in the last two decades (Tanaka, 1970; Gulcking, 1970; Ashcraft, 1976; Srivastave, 1981; Sinha, 1985; Kulshreshtha, 1980).

Theory of dielectric properties

Dielectric constant: The dielectric constant (e) of a dielectric material can be defined as the ratio of the capacitance using that material as the dielectric in a capacitor to the capacitance using a vacuum as the dielectric. using the common equation used in parallel plate capacitor, dielectric constant at different temperatures and frequencies were calculated.

Dielectric constant (ε) is given by

$$\varepsilon = C / C_0 \quad (1)$$

$$C_0 = A\varepsilon_0/d \quad (2)$$

where

C = capacitance using the material as the dielectric in the capacitor
C_0 = capacitance using vacuum as the dielectric
ε_0= Permittivity of free space (8.85 x 10-12 F/m)
A = Area of the prepared sample
d = Thickness of the sample

Dielectric loss: Dielectric loss quantifies a dielectric material's inherent dissipation of electromagnetic energy (e.g. heat)(13). It can be parameterized in two different ways. The first one is in terms of the loss angle δ and the second one is in terms of corresponding loss tangent tan δ. Both methods refer to the phasor in the complex plane whose real and imaginary parts are the resistive (lossy) component of an electromagnetic field and its reactive (lossless) counterpart. The loss tangent is then defined as the ratio (or angle in a complex plane) of the lossy reaction to the electric field E in the curl equation to the lossless reaction.

$$\tan\delta = \omega\varepsilon'' + \sigma / \omega\varepsilon' \quad (3)$$

Where

ω = angular frequency
σ = conductivity. For dielectrics with small loss, this angle is $\ll 1$ and $\tan\delta \approx \delta$.

Impedance spectroscopy: Impedance spectroscopy (sometimes called Dielectric spectroscopy), and also known as electrochemical impedance spectroscopy (EIS), measures the dielectric properties of a medium as a function of frequency (Sidorovich, 1984; Kremer, 2002; Prokhorov, 2003; Hippel, 1954). It is based on the interaction of an external field with the electric dipole moment of the sample, often expressed by permittivity. Impedance is represented as a complex quantity Z and the term complex impedance may be used interchangeably. The polar form conveniently captures both magnitude and phase characteristics as

$$Z = |Z| e^{j \arg(Z)} \quad (4)$$

where the magnitude |Z| represents the ratio of the voltage difference amplitude to the current amplitude, while the argument arg(Z) (commonly given the symbol θ) gives the phase difference between voltage and current. j is the imaginary unit, and is used instead of i in this context to avoid confusion with the symbol for electric current.

In Cartesian form, impedance is defined as

$$Z = R + jX \quad (5)$$

where the real part of impedance is the resistance R and the imaginary part is the reactance X (Electrical impedance, 2015). Then there is complex impedance spectroscopy technique which is used to study the electrical behaviour of the sample over a wide range of frequency and temperature. This technique provides information of the structure-property relationship of the sample. This enables us to separate the real and imaginary components of the complex impedance.

Complex impedance:

$$Z^*(\omega) = (Z' - jZ'') \quad (6)$$

where $Z' = |Z|\cos\theta$ and $Z'' = |Z|\sin\theta$

Electrical conductivity: Electrical conductivity otherwise known as specific conductance is the measure of the ability of a material to conduct electric current. It is the reciprocal of electrical resistivity. Conductivity is an intrinsic property. Conductivity is the inherent property of the material that makes up the object. No matter how the object changes in terms of shape/size/mass, as long as it is made of the same material and the temperature remains the same, its conductivity does not change. Higher conductivity also gives an object a higher conductance. The formula that relates conductivity with conductance is:

$$G = \sigma A/d \quad (7)$$

where G is the conductance, σ the conductivity, A the total surface area and d the thickness of the conductor. This formula applies for any (geometrically) Prismatic or cylindrical conductor, including cuboids.

MATERIALS AND METHODS

The dielectric ceramic having the compositional formula $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10 was prepared by solid state reaction method. The initial ingredients MnO, ZnO, TiO_2, Fe_2O_3 were of high purity. They were manually weighed and carefully mixed in correct Stoichiometric ratio. The mixture was then grounded for ten hours using agate mortar. The resulting mixture was air dried and presintered in air for 10 hours at 700°C. Temperature was controlled by a platinum-Rhodium thermocouple within the furnace. The pre sintered ferrites were then again grounded for two to three hours. This was ball milled for one month followed by attrition milling. Then again grounded and sieved. The granulated powder was then pressed into pellets and toroids at a pressure of $1N/M^2$ with the help of hydraulic press. The binder used was polyvinyl alcohol solution. The final sintering was done at for 20 hours at 1150°C followed by slow cooling to room temperature. Chopper stabilized amplifier was used as temperature control system within the furnace. Different dielectric properties of the prepared sample were measured using an LCR Impedance analyzer at 27°C, 120°C, 200°C, 300°C, 400°C, 500°C and 600°C. The measurements in a wide frequency range at different temperatures have been studied.

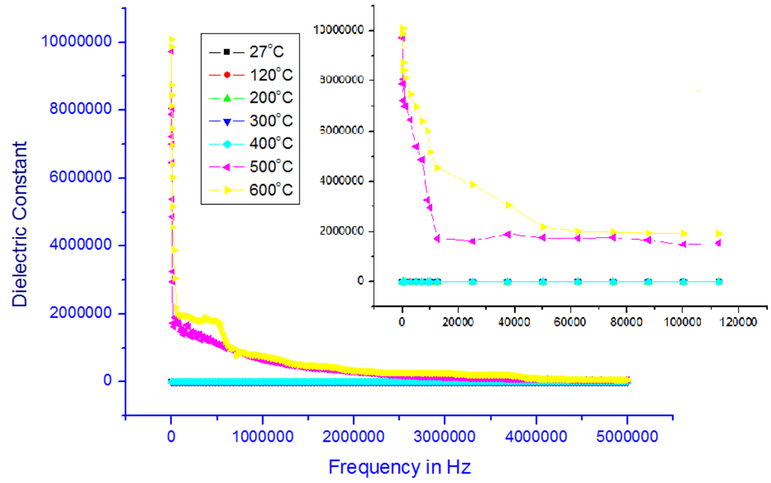

Fig.1. Variation of Dielectric constant with frequency at different temperatures of Manganese -Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10

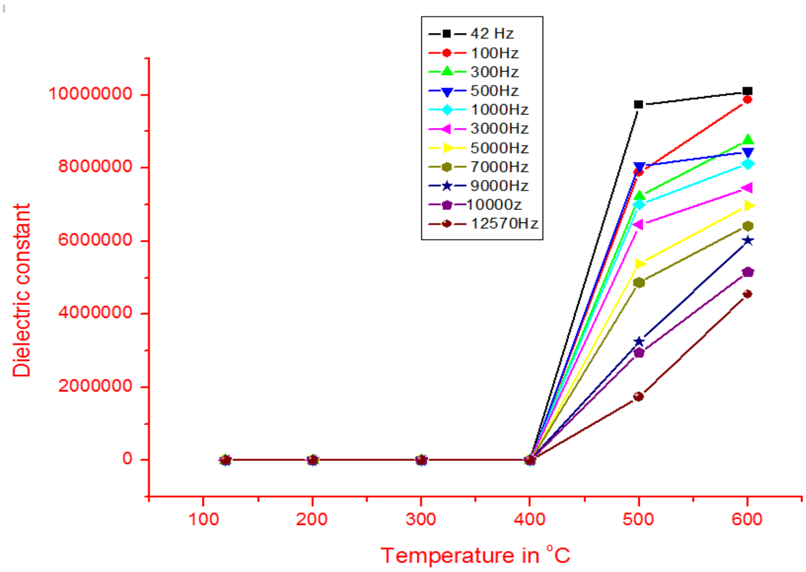

Fig.2.Variation of dielectric constant with temperature and frequency of $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10

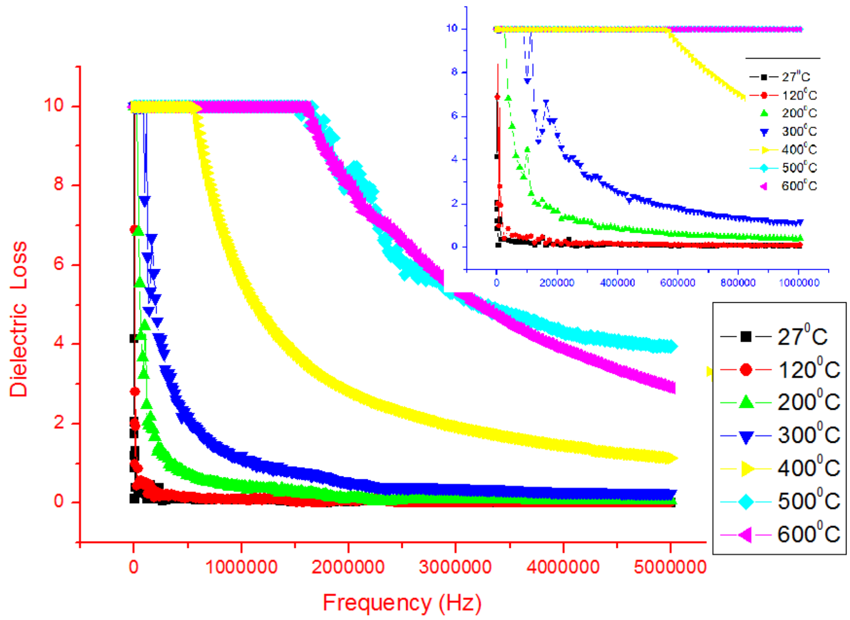

Fig.3. Variation of dielectric loss with frequency and temperature of Manganese -Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10

The calcined samples were pelletized and its dimensions were measured. Studies of dielectric and impedance spectroscopy characteristics of the ceramic sample composites are carried out in the low-frequency (100–1300 MHz) and in the high-frequency region (100 Hz–5 MHz) at different temperatures (25–600°C). The value of dielectric parameters are noted for a frequency range of 100 Hz to 5 MHZ using HIOKI 3532-50 LCR Hitester. Dielectric constant of the sample was calculated for different frequencies and temperatures using the equation used in parallel plate capacitor. Dielectric losses and impedance were directly measurable from LCR Impedance analyzer. Conductivity is calculated from the equation connecting conductivity and conductance, since conductance was measurable from Impedance analyzer. The effect of temperature and frequency on dielectric constant, impedance, and dielectric loss of the prepared sample was analyzed. Variation of electrical conductivity with frequency is found out. It is verified to be in accordance with Jonscher's power law.

RESULTS AND DISCUSSION

The values of dielectric constant, impedance and losses of Titanium Substituted Manganese - Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10 sample is observed in various temperatures 27°C,120°C,200°C,300°C,400°C, 500°C and 600°C with frequency range 42Hz to 5MHz. The observed results are analyzed using existing theories.

Dielectric Constant

***Effect of Frequency*:** In order to find out the frequency dependence of dielectric constant of the prepared sample, graphs are plotted with frequency (frequency range 42 Hz to 5MHz) against dielectric constant at various temperatures, viz, 27°C, 120°C, 200°C, 300°C, 400°C, 500°C and 600°C. The graphs are given in Fig.1. From the analysis it is observed that dielectric constant decreases with increase in frequency and reaches a constant value. At lower temperatures (27°C, 120°C, 200°C, 300°C, 400°C) dielectric constant has a very low constant value in the entire frequency range for each particular temperature. But in the case of 500 and 600 degree Celsius it is observed that dielectric constant decreases very drastically at low frequency region (say up to 1000 Hz). For the curve at 500°C, after 1000 Hz, dielectric constant attains almost a constant value but for the graph drawn at 600°C, dielectric constant then decreases gradually and attains almost a constant value only after say 6000 Hz. Constant value of dielectric constant increases with increase in each particular temperature.

Effect of Temperature: To illustrate the effect of temperature on dielectric constant, its variation as a function of temperature (27°C, 120°C, 200°C, 300°C, 400°C, 500°C and 600°C) is plotted at different frequencies (i.e 42, 100, 150, 225, 300, 1000, 10000 and 12570Hz). They are shown in the Figure 2. In the entire frequency range dielectric constant has a very low and constant value up to 400°C. From 400 to 500°C it increases rapidly up to a higher value for each particular frequency. From 500 to 600°C it again increases, smoothly for lower frequencies and rapidly for higher frequencies.

Dielectric Loss

Effect of Frequency: The effect of frequency (frequency range 42 Hz to 5MHz) on dielectric loss at constant temperatures (27°C, 120°C, 200°C, 300°C, 400°C, 500°C and 600°C) is plotted in the graphs (Fig.3). It is observed that dielectric loss, decreases rapidly in the lower frequency region (say up to 800 KHz), for lower temperatures, with rapidness increasing with fall in temperature and remains a constant thereafter. For higher temperatures dielectric loss is a constant in the lower frequency region (say up to 1800 KHz) and then its value decreases gradually. The dielectric loss reaches the instrumental saturation value (tan δ = 9.9999) in the low frequency range, but at higher frequencies the value drops down drastically. Due to the low value of dielectric loss at higher frequencies, all the samples possess superior crystalline quality

Effect of Temperature: To illustrate the effect of temperature on dielectric loss, its variation as a function of temperature (27°C, 120°C, 200°C, 300°C, 400°C, 500°C and 600°C) is plotted at different frequencies (i.e 42Hz, 802KHz, 1742KHz, 2682KHz, 3622KHz, 4561KHz and 4875KHz).They are shown in the figure 4. It is evident from the graph that dielectric loss of all curves, except the one at 42 Hz, is constant up to say 125°C, increases with temperature up to 400°C, almost constant up to 500 °C, and then decreases. Curve at 42 Hz increases up to say 125°C and then remains constant.

Impedance

Effect of Frequency: To find out the frequency dependence of electrical impedance of the prepared sample, graphs (Fig.5) are plotted with frequency (frequency range 42 Hz to 5MHz) against dielectric constant at various temperatures, viz, 27°C, 120°C, 200°C, 300°C, 400°C, 500°C and 600°C. Impedance curves of higher temperatures are straight lines parallel to X-axis except at very high frequency region. For 27°C and 120°C, impedance decreases with frequency. For 200°C and 300°C impedance gradually decreases. Variation of real part & Imaginary part of impedance with frequency at different temperatures is as shown in the graph given (Fig.6 & Fig.7) below. Here z' is the real part, Z" is the imaginary part of impedance. From the above two graphs the relationship between real and imaginary parts of impedance it is noted that the imaginary and real parts of impedance remain almost constant as frequency increases in high temperature region. But the variation in impedance with frequency is observed in low temperature regions.

Effect of Temperature: In order to illustrate the effect of temperature on impedance its variation as a function of temperature (27°C, 120°C, 200°C, 300°C, 400°C, 500°C and 600°C) is plotted at different frequencies (i.e 802,1742, 2682, 3622, 4561, 4875KHz).They are shown in figure 8. For higher frequencies, up to 300°C impedance is constant, it decreases up to 400°C, slightly decreases upto 500°C, then remains constant. For lower frequencies impedance slightly increases up to 100°C, then decreases exponentially up to 400°C, slightly decreases up to 500°C, and then remains constant.

Conductivity

Effect of Frequency: The effect of frequency (frequency range 42 Hz to 5M Hz) on electrical conductivity at constant temperatures (27°C, 120°C, 200°C, 300°C, 400°C, 500°C and 600°C) are plotted in the graph (Fig.9) given below. Conductivity remains constant except at very high frequencies.

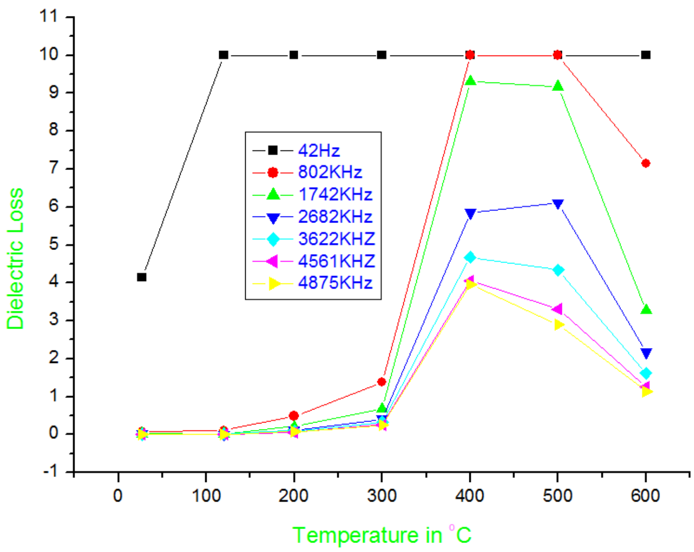

Fig. 4. Variation of dielectric loss with temperature at varied frequencies of Manganese -Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10

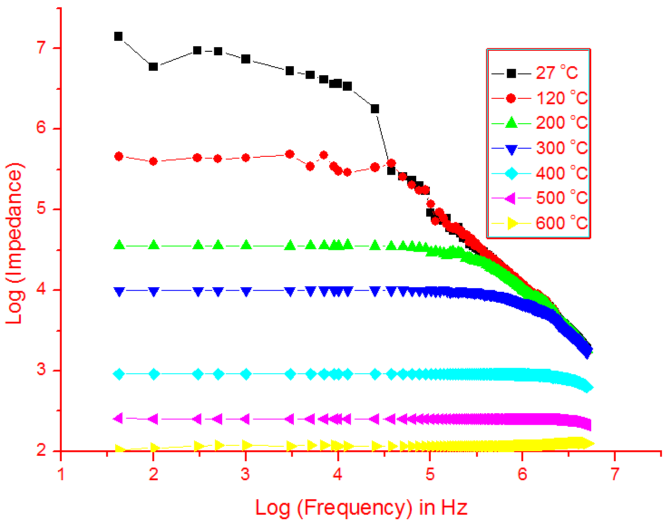

Fig.5. Variation of impedance with frequency at different temperatures of $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10

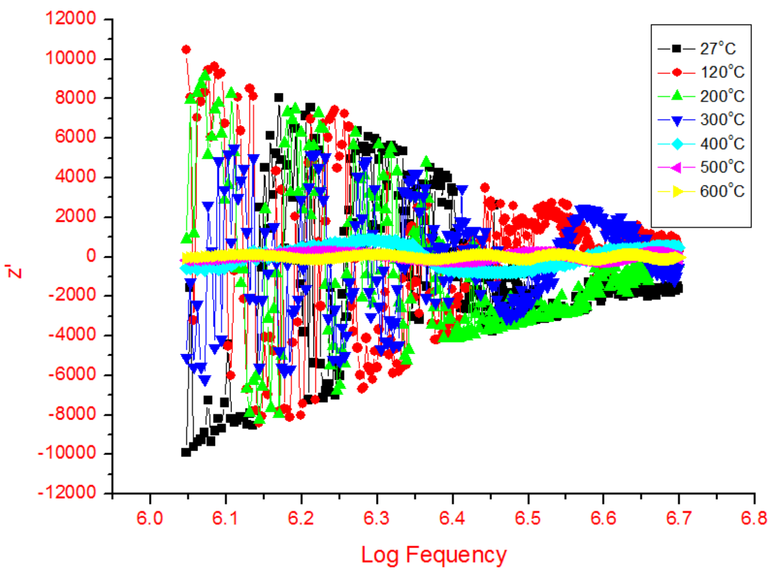

Fig.6.Variation of real part of Impedancewith frequency and temperature of $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10

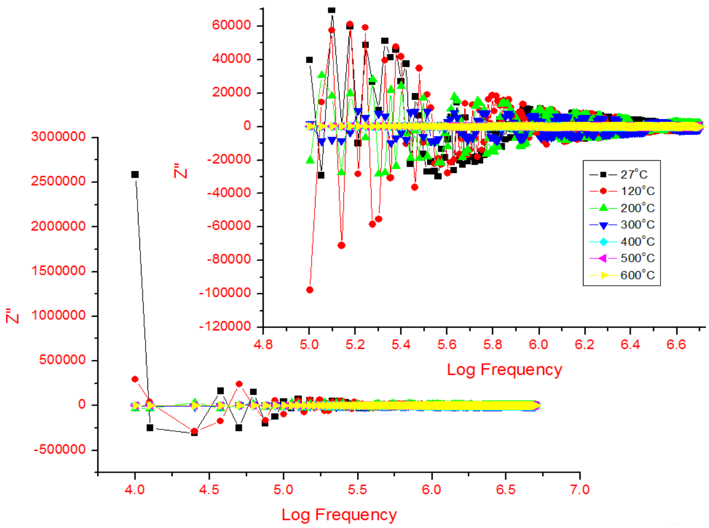

Fig.7. Variation of imaginary parts of Impedance with frequency and temperautre of Manganese -Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10

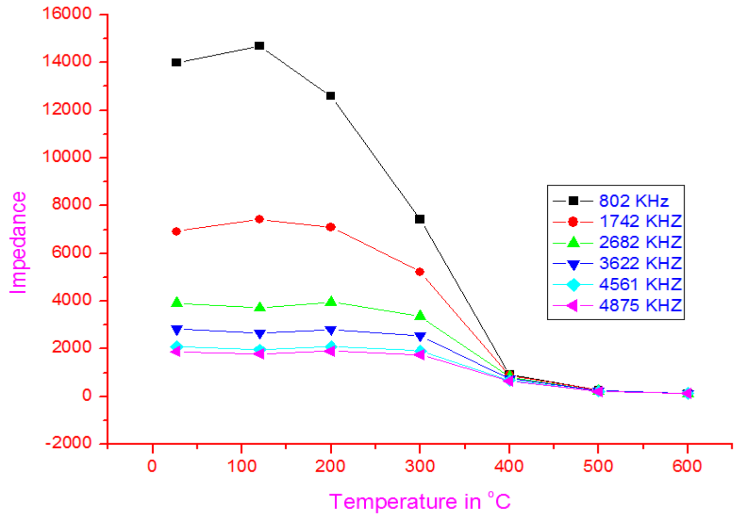

Fig.8 . Variation of impedance with temperature at different frequencies of Manganese -Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10

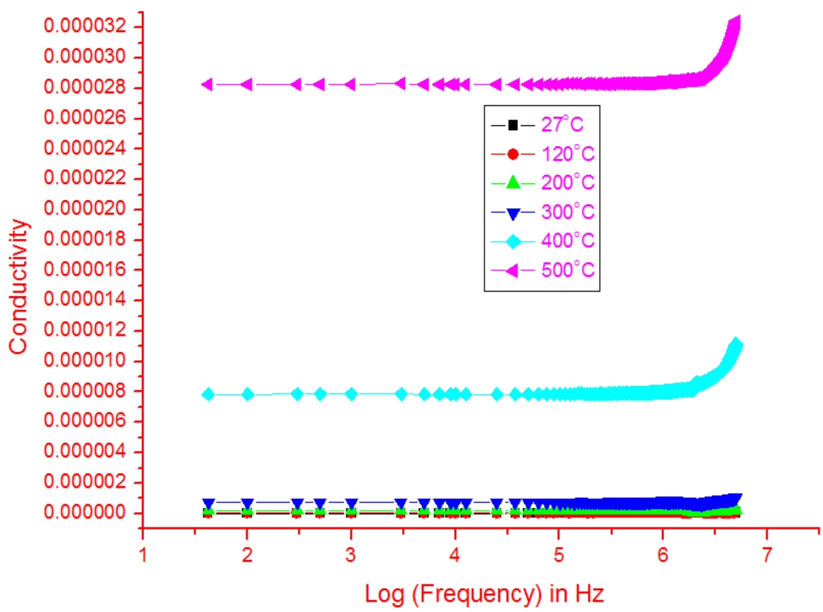

Fig.9.Variation of conductivity with frequency at different temperatures of Manganese -Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10

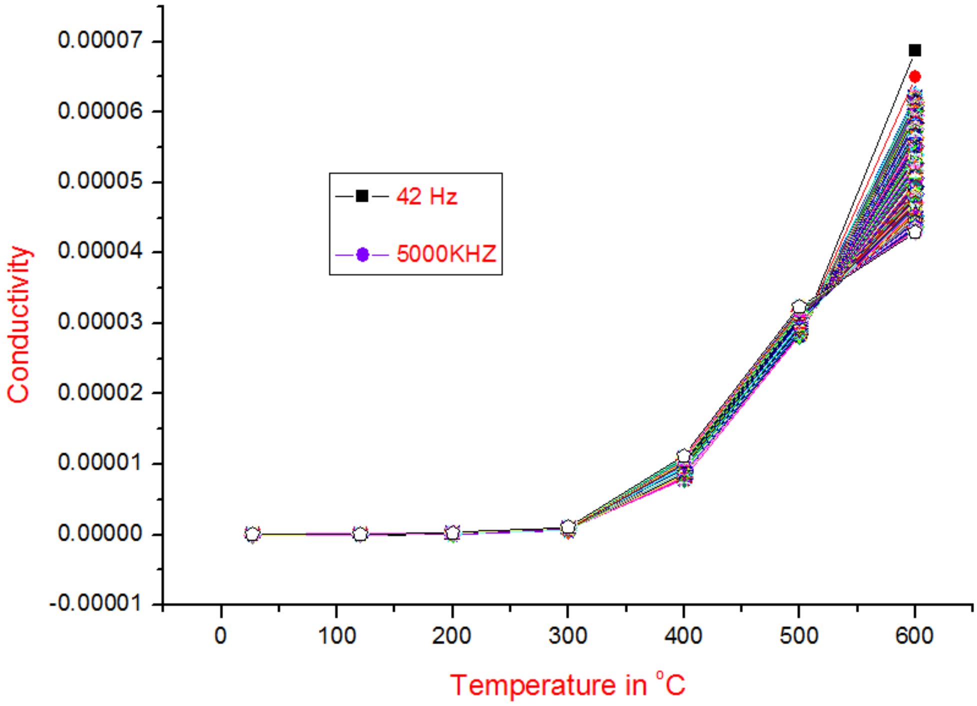

Fig.10.Variation of conductivity with temperature and frequency of Manganese -Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10

At higher frequencies conductivity increases exponentially. This can be explained according Joncher's power law. i.e. σ ac = σ dc + $A\omega^s$

Effect of Temperature: To illustrate the effect of temperature on conductivity its variation as a function of temperature (27°C, 120°C, 200°C, 300°C, 400°C, 500°C and 600°C) is plotted at frequencies ranging from 42Hz to 5MHz. It is plotted in figure 10. Conductivity remains almost a constant for all frequencies up to 300^0C. Afterwards conductivity increases with increasing temperatures.

DC conductivity or bulk conductivity obeys Jonscher's universal power law of dc conductivity. Conductivity very slightly increases with frequency due to decrease in polarization effects along with dc conductivity which excites electrons in valence band to conduction band in the crystal lattice. The sudden increase in conductivity for 600 degree Celsius near maximum frequency is due to ac conductivity which obeys Jonscher's universal power law of frequency dependent ac conductivity along with low polarization at high frequencies. Even though dielectric polarization is very high at high temperatures, conductivity increases with increasing temperatures due to the conduction of charges caused by quantum mechanical tunneling at high temperatures. Hopping of charge carriers causes conductivity at low temperatures.

Conclusion

Most important dielectric properties of Titanium Substituted Manganese - Zinc Ferrite System $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.10, viz, dielectric constant, dielectric loss, electrical conductivity and impedance varies with frequency and temperature. The ceramics with high permittivity, low dielectric loss, and near zero temperature of dielectric constant are used as components for wireless communications (Wersing, 1991; Wersing, 1996; Negas, 1991). Dielectric constant remains at a constant value for lower temperatures and decreases with frequency in the lower frequency region and then remains constant for higher temperatures. Dielectric loss decreases rapidly with frequency in the lower frequency region and then remains constant for lower temperatures and remains constant in the lower frequency region and then decreases gradually for higher temperatures. Impedance is a constant except at very high frequencies. For lower temperatures it decreases gradually with frequency. Conductivity remains constant except at very high frequencies. At higher frequencies conductivity increases exponentially. Conductivity remains almost a constant for all frequencies at lower temperatures. Afterwards it increases with increasing temperatures. The variation of dielectric constant and dielectric loss with temperature and frequency implies the presence of some internal field within the dielectric composite material along with the external A C field.

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