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Temperature and Frequency Dependence of Dielectric Properties of Superconducting Ceramic $GdBa_2Ca_3Cu_4O_{10.5}$

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Abstract: ***A polycrystalline, superconducting ceramic $GdBa_2Ca_3Cu_4O_{10.5}$ having nano sized particles is designed, and prepared by solid state reaction route involving selection of raw materials, mixing ,manual milling, ball milling, calcining at treating temperatures and sintering. Prepared ceramic is found to be in perovskite structure and in orthorhombic crystal system by X ray diffraction and related studies. Dielectric behaviour of the ceramic is studied observing the effects of temperature and frequency on some important dielectric parameters such as dielectric constant, dielectric loss, impedance and conductivity. High impedance and high dielectric constant in low frequency and high temperature regions are expected in the case of superconducting ceramics. Dielectric parameters should be reduced with increasing frequencies and decreasing temperatures and saturation values should be reached. Dielectric studies are conducted in a selected frequency range with in 42Hz-5MHz ,at three different temperatures 30^0C, 500^0C and 900^0C in a frequency response analyzer which is connected with a computer and a heating arrangement. Impedance spectroscopic studies and conductivity studies are also done. Debye relaxation behaviour, space charge polarization and Maxwell Weigner model of low frequency dielectric dispersion (LFDD) are observed in the sample. Conductivity is found to obey, Jonscher's power law of conductivity.***

Keywords: Polycrystalline superconducting ceramic $GdBa_2Ca_3Cu_4O_{10.5}$, Dielectric constant, Dielectric loss, Impedance, Conductivity, Debye behavior, Space charge polarization, Maxwell Weigner model, Low frequency dielectric dispersion, Jonscher's power law.

1. Introduction

Crystalline ceramics exhibiting good dielectric properties are super desirable, nowadays because of its comparatively lenient and easy manufacturing techniques, economic feasibility, high resistivity, presence of high dielectric constant, low dielectric loss and high tensile strength. Dielectric ceramics have numerous technological applications. Dielectrics with low dielectric constant as well as high dielectric constant are important in electronic industry. Electronic insulation devices make use of low dielectric constant material whereas capacitive elements in electronic circuits utilize high dielectric constant materials. Since $Ba_2Ca_{(n-1)}Cu_nO_{(2n+4-\delta)}$ where n=2,3,4 etc. is an eminent superconducting phase in the high temperature super conducting family [1], a characteristic set of dielectric properties are expected for $GdBa_2Ca_3Cu_4O_{10.5}$ (n=4 in the above mentioned superconducting phase) such as dielectric loss and dielectric dispersion in the low frequency region and at low temperatures.

Generally a polycrystalline ceramic sample shows dielectric polarization due to the presence of inter grain boundaries and micro cracks which can act as dielectric media. In this work, the extent of dielectric polarization, dielectric loss, impedance and conductivity of the prepared ceramic is considered. Dielectric polarization is the displacement of different atomic cloud centers or alignment and space orientation of the electric dipoles or separation of electric charges at the grain or particle interfaces in the presence of an applied electric field. Here the temperature and frequency dependence of some of the significant dielectric properties such as dielectric constant, dielectric loss, phase angle, impedance and conductivity of $GdBa_2Ca_3Cu_4O_{10.5}$ is studied. Impedance spectroscopic and conductivity studies for both ac and dc conductivity are done. Dielectric spectroscopy or impedance spectroscopy measures the properties of a dielectric medium as a function of frequency [2]. It analyses the behaviour of a sample as a function of frequency and temperature. The relevant parameters required are the relative permittivity, permeability, and conductivity and they are measured using modern equipments.

We know that in a dielectric material, the application of an external electrical field produces electric dipole polarisation. Electric dipole polarisation is characterised by a vector quantity called electric dipole moment which constitutes a positive point charge +q and a negative point charge –q. The dipole moment is represented by a vector pointing from the negative charge to the direction of the positive charge [3-5]. Moreover, the study of dielectric constant, impedance, conductivity and dielectric loss factor, as a function of temperature and frequencies is one of the most convenient and sensitive methods of studying the ceramic structure [6]. The dielectric properties (dielectric constant, impedance, conductivity and dielectric loss) of a number of ferroelectric ceramics have been investigated in the last two decades [7-12].

2. Theory of Dielectric Properties

2.1 Dielectric Constant

Dielectric constant which is otherwise known as relative permittivity is a dimensionless number that can be defined as the ratio of the permittivity of the material to the permittivity of free space. Dielectric constant measures the degree of polarization in the dielectric material on the application of an external electric field. As the polarization increases the value of dielectric constant also increases. Another definition of dielectric constant is the ratio of the capacitance of a capacitor embedded with the dielectric to

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the capacitance of a capacitor embedded with free space or vacuum. Equation for calculating dielectric constant (ε) in a parallel plate capacitor is given by

$$\varepsilon = C / C_0 \quad \text{---------- [1]}$$

$$C_0 = A\varepsilon_0/d \quad \text{---------- [2]}$$

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

2.2. Dielectric Loss

Dielectric loss quantifies a dielectric material's inherent dissipation of electromagnetic energy (E.g. heat) [13]. It represents the absorption of electric energy and the dissipation of energy as heat, when electric current passes through the material. Two terms are usually used to indicate dielectric loss. Both depicts the phasor in the complex plane. One is loss angle θ and the other is loss tangent tan θ. The loss tangent is the ratio of the real part (electric field E in loss reaction) to the imaginary part (electric field E in the lossless reaction) in the phasor equation. Frequency, temperature, humidity, voltage and nature of the material influence dielectric loss.

$$\tan\theta = G\mathfrak{O}\,\varepsilon'' + \sigma / G\mathfrak{O}\,\varepsilon' \quad \text{----------[3]}$$

where
GƱ = angular frequency
σ = conductivity.
For dielectrics with small loss, this angle is ≪ 1 and
tan θ ≈ θ

2.3 Impedance Spectroscopy

Dielectric spectroscopy (sometimes called impedance spectroscopy), and also known as electrochemical impedance spectroscopy (EIS), measures the dielectric properties of a medium as a function of frequency [14-17]. It utilizes the effect of an applied electric field to the electric permittivity of the crystalline sample. Impedance is a complex quantity Z and hence impedance is often called complex impedance. Impedance is the combination of resistance and reactance. It is the effective resistance a dielectric offers to an ac current. Hence its unit is ohm. In Cartesian form, the equation of impedance is

$$Z = R + jX \quad \text{----------[4]}$$

where

$$X = \omega L \quad \text{----------[5]}$$

so

$$Z = R + j\omega L \quad \text{----------[6]}$$

R is the resistance and X is the reactance [18]. In polar form

$$Z = |Z|e^{j\theta} \quad \text{----------[7]}$$

$$|Z| = (R^2 + \omega^2 L^2)^{1/2} \quad \text{----------[8]}$$

$$\tan\theta = \omega L/R \quad \text{----------[9]}$$

The polar form of impedance contains characteristics of both magnitude and phase. Impedance spectroscopy is a very powerful technique used to analyze the dielectric responses and dielectric behavior technique of the prepared ceramic sample.This is a non-destructive technique which analyses dielectric behavior of grains present in the crystalline structure at the grain boundaries as a function of frequency. This method is also applied to understand the structure, defects and conduction routes in the crystal structure.

Two important techniques in impedance spectroscopy are

- *COLE- COLE PLOT* - plotting real and imaginary parts of complex impedance in x axis and y axis respectively (Fig.1).

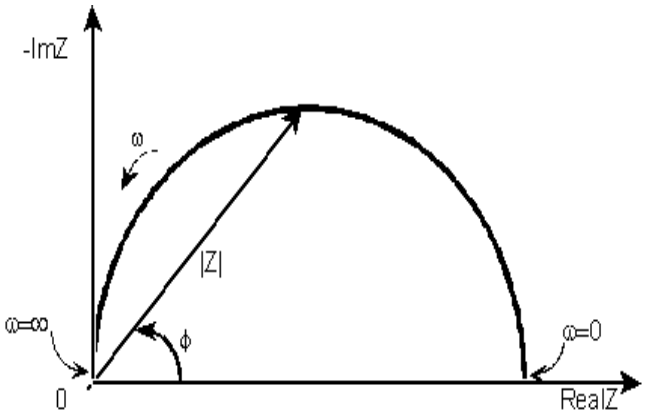


Figure 1: Schematic representation of Cole-Cole plot

- *BODE PLOT* – plotting frequency in x axis and modulus of impedance and phase angle in y axis (Fig.2).

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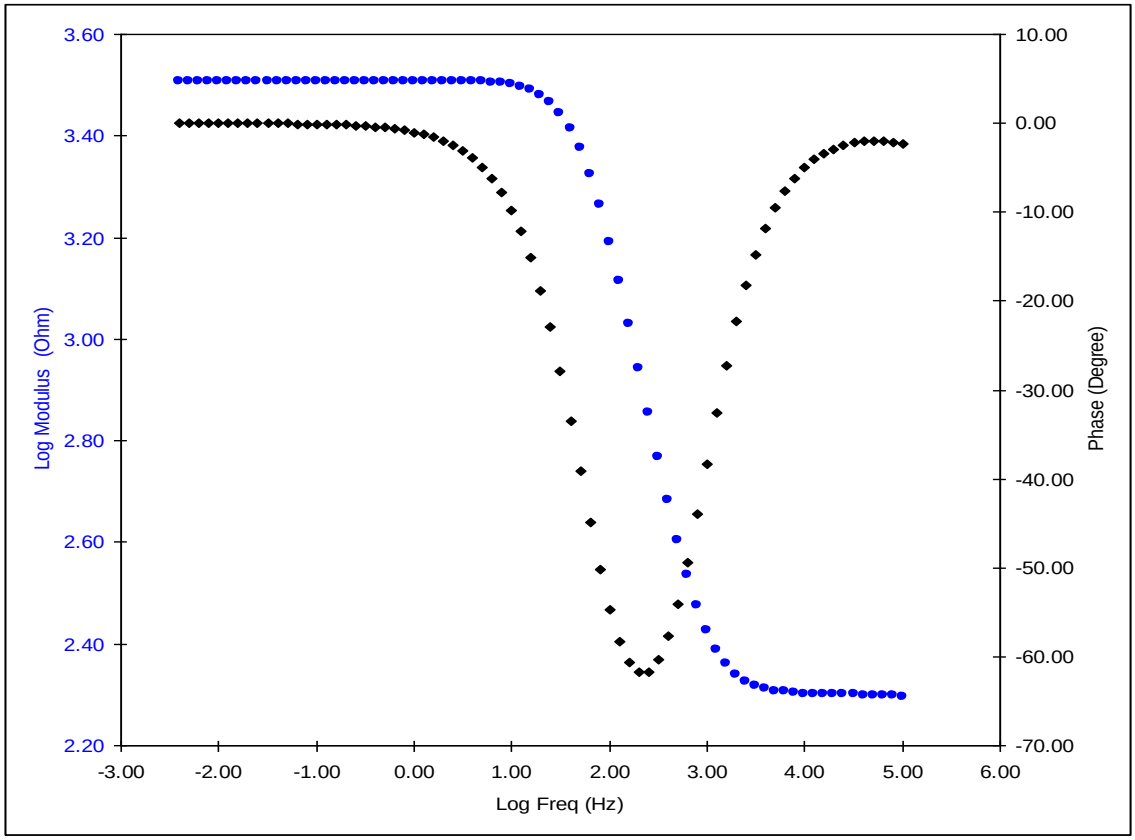


Figure 2: Schematic representation of Bode plot

2.4. Conductivity Analysis

Electrical conductivity measures the extent to which a given ceramic sample conducts electricity or electric current. It is also known as specific conductance. Reciprocal of resistivity is taken as conductivity. Hence its unit is 'mho'. Conductivity is the inherent property of a material which is independent of shape, size and mass of the object but it depends on temperature and frequency. A higher conductivity yields higher conductance. Conductance is an extrinsic property. Conductivity is given by the equation

$$G=\sigma A/d \text{----------[10]}$$

G denotes conductance, σ denotes conductivity and A denotes the total surface area and d is the thickness of the given sample of material. Almost all types of conductors having different geometry for example prismic, cuboid or cylindrical use this formula.

- **Jonscher's Universal Power Law**

When the variation of conductivity with frequency is plotted with frequency in the x axis and conductivity in the y axis, three typical regions, one with low frequency, one with intermediate frequency (intermediate plateau region) and one with high frequency can be observed. In low frequency dispersion region, conductivity decreases as frequency decreases due to space charge polarisation effects at the grain interface. So we can say that conductivity increases with frequency in the low frequency region. At intermediate frequency, conductivity has no change i.e. it is dc conductivity or bulk conductivity itself there. In the high frequency region once again conductivity becomes proportional to frequency. Jonscher's universal power law gives equations for both frequency independent dc conductivity and frequency dependent ac conductivity.

$$\sigma_{dc} = l/R_bA \text{ ----------[11]}$$

$$\sigma_{ac} = \sigma_{dc}+A\omega^n \text{ ----------[12]}$$

A is a constant, ω is the frequency and n is the number denoting the value of frequency exponent, 0<n<1

3. Materials and Experimental Methods

The perovskite polycrystalline superconducting ceramic sample with nano sized particles, $GdBa_2Ca_3Cu_4O_{10.5}$ is designed and synthesized in a specially designed furnace following steps of solid state reaction. Solid state reaction route is conventionally used for the preparation of a polycrystalline ceramic from a mixture of crystalline raw materials. The raw materials are of very high quality and greater than 99.99 % purity. Starting ingredients are Gadolinium Oxide, Barium carbonate, Calcium Oxide and Cupric Oxide. These materials are accurately weighed according to molecular formula,

$$½\ Gd_2O_3+2\ BaCo_3 + 3\ CaO + 4\ CuO=GdBa_2Ca_3Cu_4O_{10.5} + 2\ CO_2$$

These powders in correct weights and quantities are first, mixed manually in an agate mortar for a couple of hours and then mixed mechanically in a ball mill for three months with suitable zirconium beads along with mechanical grinding and sieving daily to avoid agglomeration. Then it is attrition milled for two hours.The resulting mixture after milling was treated at different temperatures 30^0C, 500^0C, 850^0C and 900^0C in a specially designed furnace. Oxygen flow is allowed into the furnace chamber throughout the calcination

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process and highly introduced during cooling process of the sample in the furnace. After each calcining temperature, the powder is analyzed, cooled, and again calcined till the desired crystal system and crystal structure is obtained. In order to study about dielectric properties, a part of the prepared sample is pelletized in a hydraulic press at a pressure of $1N/m^2$. Pelletization is followed by sintering at 1100^0C for 6 hours and finally cooled to room temperature. Then the pellet is electroplated with silver paste on the sides for conduction and placed in a specially designed sample holder. The diameter of the pellet is measured to be 8. 746 mm and thickness is measured to be 1. 8 mm. The sample along with the sample holder arrangement is now inserted in the furnace which is in conjunction with a computer controlled LCR Impedance analyzer and different dielectric properties of the prepared sample is measured at 30^0C, 500^0C and 900^0C. The measurements are done within a frequency range of 42Hz-5MHz. Values of dielectric constant are extracted from the equation of parallel plate capacitor using readings of capacitance obtained. Values of dielectric loss and impedance can directly be noted from LCR Impedance analyzer. Conductivity readings are calculated from conductance readings obtained from the apparatus.Frequency and temperature dependence of dielectric constant and dielectric loss are analysed. Impedance spectroscopic and conductivity studies are also done.

4. Results and Discussions

4.1. Dielectric Constant

- **Effect of Frequency**

From the graphs drawn by plotting dielectric constant on y axis and frequency on x axis for three temperatures 30^0C, 500^0C and 900^0C (Fig.3), it is observed that the value of dielectric constant decreases with increase in frequency up to 25000 Hz. After that dielectric constant remains constant i.e. attains saturation value. At 900^0C dielectric constant has very high value in the order of 10^6 in the lower frequency region. Maximum value of dielectric constant is obtained at 900^0C below 100 Hz. For 30^0C and 500^0C dielectric constant has an almost fixed constant value in the order of 10^3 in the entire frequency range from 42Hz to 5MHz.

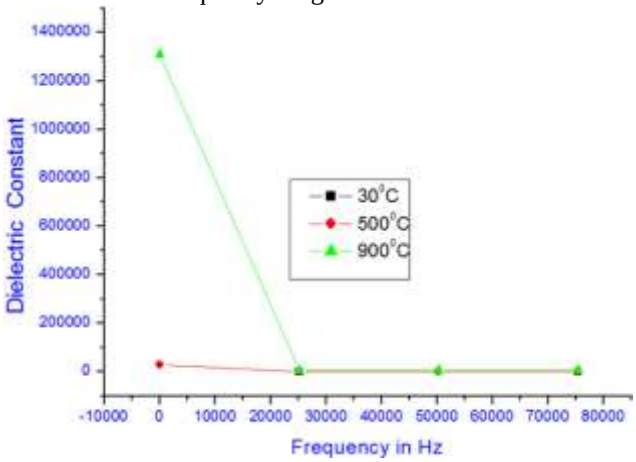


Figure 3: Variation of Dielectric constant with frequency

- **Effect of Temperature**

In the figure 4 given below the relation between dielectric constant and temperature at different frequencies from 42Hz to 5MHz of the prepared ceramic sample is plotted. Dielectric constant is found to be increasing with increase in temperature. Dielectric constant has comparatively low values in the beginning i.e. at lower temperatures in the order of 10^3 and then increases to very high values from 500 degree Celsius onwards and attains very high value at 900 degree Celsius in the order of 10^6. Higher values of dielectric constant are obtained for low frequencies below 100 Hz.

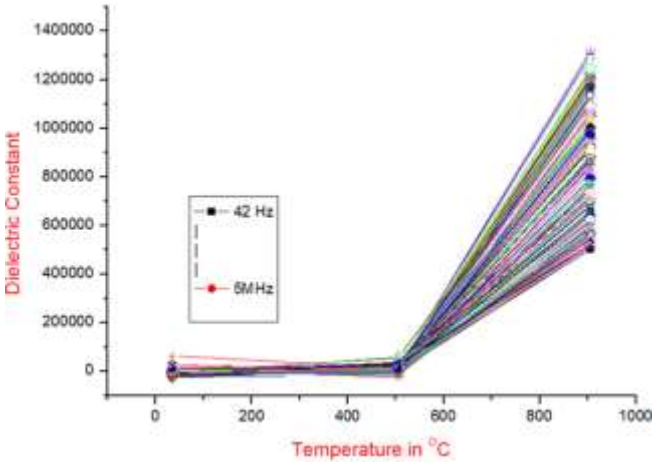


Figure 4: Variation of dielectric constant with temperature

From both fig. 3 and fig. 4 we can conclude that dielectric constant has very high values at higher temperatures (most high value at 900 ^{0} C) in the low frequency region below 100 Hz. Dielectric constant decreases as frequency increases and dielectric constant decreases as temperature decreases.

1) Dielectric constant decreases with increase in frequency because charge carriers cannot follow applied external electric field at higher frequencies and hence the dielectric responses become ineffective. As frequency increases, oscillations of the given field increases and dipolar polarization characteristic time exceeds time constant of the external field and hence the degree of dielectric polarization decreases and hence corresponding values of dielectric constant. This means dielectric polarization is dominant in low frequency region and it decreases with increasing frequencies. This behaviour is typical in materials with very small particle sizes. This confirms the presence of space charge polarization which is dominant in low frequency region. Strong dielectric dispersion or strong dielectric polarization at low frequencies is called LFDD i.e. low frequency dielectric dispersion. The presence of this behaviour in crystalline ceramics can be explained on the basis of Maxwell-Weigner model in dielectrics as follows. The crystalline ceramic lattice is made up of well conducting grains separated by grain boundaries. When frequency is low, grain boundaries are very effective and as a result there is a hopping of charge carriers. This results in high dielectric polarization and hence high values of dielectric constant. As frequency increases, responses of dipoles decreases and they lag behind the applied field and this damping nature of dipoles reduces dielectric constant.

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2) At high temperatures dielectric polarization is very high and it decreases with decrease in temperature. Another noteworthy point is that LFDD increases with temperature. Increase in the values of dielectric constant with increase in temperature is because of space charge or interfacial polarization present in the ceramic sample. Space charge polarization is due to the accumulation of charge carriers in the interface between grains. This is very common in dielectric materials and also when electrodes are connected to the dielectric material. Space charge polarization is different from orientational and ionic polarization because it affects free charges as well. Hence it is very common in polycrystalline materials. As temperature increases, grain or particle size increases due to increased lattice vibrations of atoms. Increased grain size means increased hindrance to the flow of charge carriers and hence enhancement in dielectric constant values.

4.2. Dielectric Loss

- **Effect of Frequency**

From the graph (fig.5) connecting dielectric loss and frequency (frequency range 42 Hz to 5000000 Hz) at various temperatures 30^0C, 500^0C and 900^0C, it is observed that dielectric loss has higher values in the low frequency region and it decreases with increasing frequency.Maximum value of dielectric loss is obtained at 900^0C below 100 Hz. Dielectric loss reaches a low saturated constant value at high frequency (almost near to 5 MHz). We can observe the continuation form a sharp peak at 900^0C. For the other two temperatures 30^0C and 500^0C dielectric loss has high values at very low frequenciesand attains a constant saturation value after 500 Hz. Sharp peaks are partially visible in the low frequency region.Peaks will be clearly visible only if we lower the frequency range to negative values.

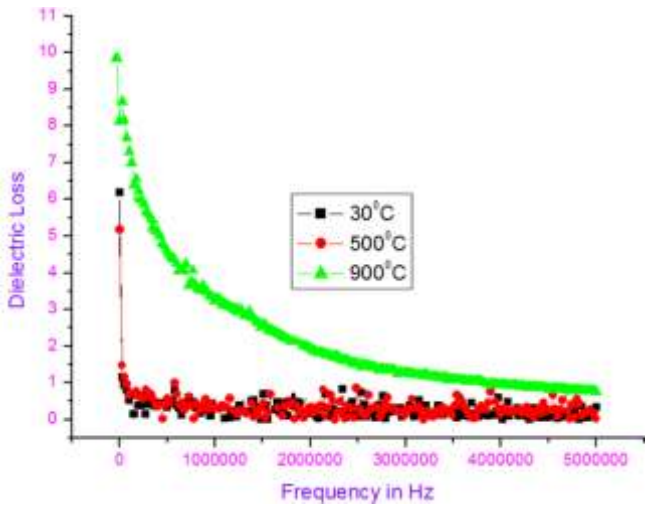


Figure 5: Variation of dielectric loss with frequency

- **Effect of Temperature**

To demonstrate the effect of temperature on dielectric loss, its variation as a function of temperature (30^0C, 500^0C and 900^0C) is plotted at different frequencies (i.e. 42HZ-5 MHz) in Fig.6. From the graph we can see that dielectric loss increases with temperature. A very high increase in the values of dielectric loss is noted at low frequencies below 100 Hz, from 500 degree Celsius onwards. Maximum value of loss is at 900^0C below 100 Hz.

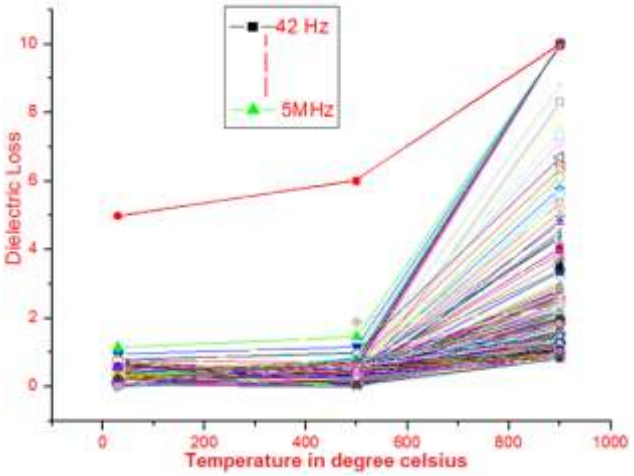


Figure 6: Variation of dielectric loss with temperature

Combining observations from graphs, fig.5 and fig.6, high values of dielectric loss are obtained in the low frequency region below 100 Hz at 900 degree Celsius. Dielectric loss increases with increase in temperature and decreases with increase in frequency.

1) The high values of dielectric loss exhibited by the ceramic sample at low frequencies, points to the presence of high dielectric polarization due to space charge polarization. Space charge polarization is dominant in lower frequencies due to effective grain boundaries. Polarization hinders charge carriers. This increases dielectric loss because dielectric loss or energy loss is calculated as tanθ where θ is the loss angle and tangent of loss angle depends on time varying electric fields.
2) The high values of dielectric loss exhibited by the ceramic sample at high temperatures, points to conduction losses. Dielectric loss is directly proportional to temperature because as temperature increases particle or grain size increases and hindrance to the flow of current increases and overall loss during conduction increases. Thus confirm the presence of dc conductivity in the dielectric material.
3) The presence of sharp peaks confirm Debye relaxation behaviour. Peaks indicate frequency dispersion positions corresponding to dielectric relaxation. If all dipoles present in the material relax with identical time of relaxation (same relaxation time τ), with relaxation time similar to the time period of the applied field 1/ω, i.e., where ωτ = 1, Debye relaxation behaviour is obtained.

4.3. Impedance

- **Effect of Frequency**

In order to study about the effects of frequency on electric impedance of the prepared ceramic sample, graph (Fig.7) is plotted with frequency (frequency range 42 Hz to 5000000 Hz) against dielectric constant at various temperatures, viz, 30, 500 and 900 degree Celsius. The graphs are given below.

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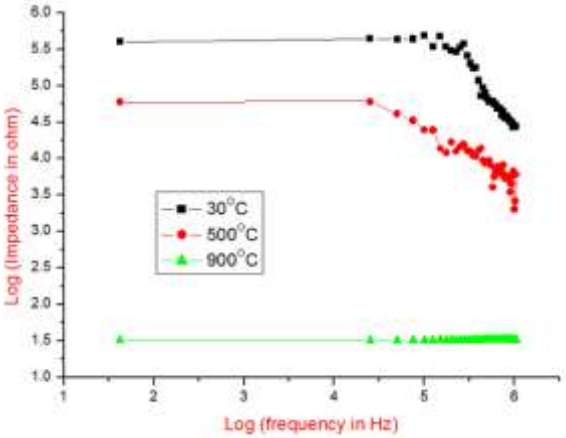


Figure 7: Variation of log of impedance with log of frequency

Three distinct regions are observed in the graphs connecting impedance and frequency. Impedance has higher values in the low frequency dispersion region and it decreases slightly as frequency increases. This is due to the low frequency dielectric dispersion which is a property of space charge polarization. This region matches with the low frequency conductivity region where conductivity increases with increase in frequency. For the next higher frequencies impedance enters a constant value and this region is called intermediate frequency or intermediate plateau region. At very high frequencies, once again impedance decreases with frequency due to quantum mechanical tunnelling of charge carriers. This corresponds to the high frequency conductivity region. High values of impedance are observed at 30 degree Celsius in the low frequency region, i.e. below 42Hz. Impedance is observed to be decreasing with temperature.

- **Effect of Temperature**

In the graph (Fig.8), temperature in x axis and impedance in y axis is plotted. From 30^0C to 500^0C impedance gently decreases with temperature and from 500^0C to 900^0C impedance decreases rapidly, for all frequencies. It is seen that at the lowest frequency 42Hz, ceramic offers very high impedance above 225000 Ohm up to 500^0C. After that temperature, impedance decreases and at final temperature 900^0C impedance attains lower values. It is observed that impedance is lower for higher frequencies and higher temperatures. Impedance decreases with increase in frequency and increase in temperature.

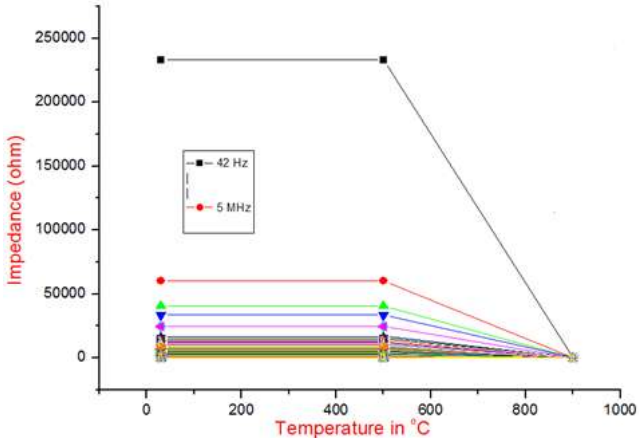


Figure 8: Variation of impedance with temperature

Analysing the two graphs (fig.7 and fig.8) it is found that the ceramic sample offers high impedance for lower frequencies and lower temperatures. Impedance decreases with frequency in the lower frequency region and higher frequency region and remains almost a constant in the intermediate frequency region which is also known as intermediate plateau region. Also it is observed that impedance decreases with temperature.

1) Impedance decreases slightly as frequency increases in the low frequency dispersion region due to the decrease in the extent of space charge polarization. In the intermediate plateau region impedance enters a constant value. In the high frequency region, it is observed that, impedance decreases with frequency, due to decrease in the values of dielectric constant.
2) Impedance decreases with temperature despite the hindrance caused by enhanced dielectric polarization at high temperatures. This is due to quantum mechanical tunnelling of charge carriers at high temperatures and hopping mechanism of charge carriers at lower temperatures [19].

4.4. Conductivity

- **Effect of Frequency**

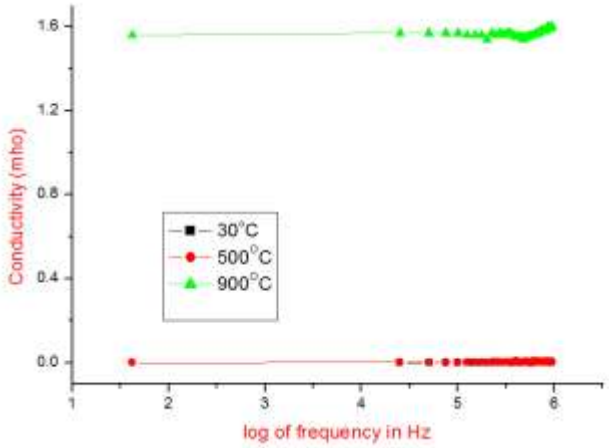


Figure 9: Variation of conductivity with log of frequency

To analyse the frequency dependence of conductivity, a graph (Fig.9) is constructed with log of frequency (frequency range 42Hz to 5MHz) against conductivity at

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three temperatures namely 30, 500 and 900 degree Celsius. Three regions are observed in the graph. Conductivity has lower values in low frequency region and it increase with increase in frequency slightly. This slightly increasing conductivity region is called law frequency conductivity region. Then conductivity enters a constant phase and this region is called intermediate frequency region. After that again conductivity increases with frequency. This region is called high frequency region. Maximum value of conductivity is observed at 900 degree Celsius at high frequency region. Conductivity decreases with temperature.

- **Effect of Temperature**

To analyse the dielectric responses of the ceramic sample to various temperatures a graph connecting conductivity and temperature is drawn (Fig.10) by plotting temperature on x axis and conductivity on y axis. Graphs are drawn in the entire frequency range (42Hz -5MHz). From 30^0C to 500^0C conductivity gently increases with temperature and from 500^0C to 900^0C conductivity increases rapidly. This behaviour is repeated for every frequency. High value of conductivity is obtained at the highest temperature taken i.e. at 900^0C.

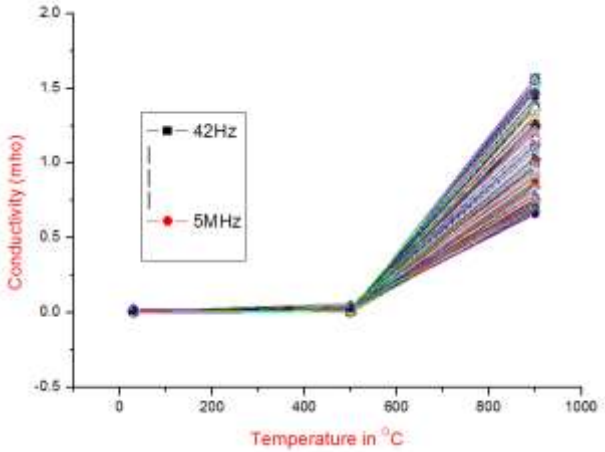


Figure 10: Variation of conductivity with temperature.

Combining results from figures 9 & 10 it can assume that conductivity increases with frequency in the lower frequency region and higher frequency region and remains almost a constant in the intermediate frequency region. Also conductivity increases with temperature.

1) Conductivity-frequency graph (Fig.9) yields three typical regions, a low frequency region, an intermediate frequency region and one high frequency region. In low frequency region, conductivity slightly increases with increase in frequency due to the decrease in polarization effects with frequency. Since conductivity increases with frequency, conductivity is frequency dependent ac conductivity and obeys Jonscher's universal power law of ac conductivity. In the intermediate frequency region, conductivity is independent of frequency and hence is called dc conductivity or bulk conductivity and obeys Jonscher's universal power law of dc conductivity. In the high frequency region, again conductivity increases with increasing frequencies due to low values of dielectric constant, Obeying Jonscher's universal power law of frequency dependent ac conductivity.
2) Conductivity increases with temperature despite the hindrance caused by enhanced dielectric polarization at high temperatures, as a result of quantum mechanical tunnelling at high temperatures. This is expected in high temperature superconductors. At lower temperatures conductivity has high values due to hopping mechanism of charge carriers.

5. Conclusions

The polycrystalline, superconducting ceramic $GdBa_2Ca_3Cu_4O_{10.5}$ is prepared by solid state reaction route and dielectric behaviour of the ceramic is studied observing the effects of temperature and frequency on some important dielectric parameters such as dielectric constant, dielectric loss, impedance and conductivity. Studies are done in a selected frequency range within 42Hz-5MHz, at three different temperatures 30^0C, 500^0C and 900^0C. Debye relaxation, space charge polarization and low frequency dielectric dispersion (LFDD) are observed in the sample. At low frequencies and high temperatures the ceramic acts as a good dielectric material. Superconducting properties will be revealed at temperatures below zero degree Celsius (nearer to transition temperature) according to literature.

[1] Dielectric constant decreases with increase in frequency because charge carriers cannot follow applied external electric field at higher frequencies. So dielectric polarization due to space charge polarization is dominant in low frequency region and it decreases with increasing frequencies. Strong dielectric dispersion or strong dielectric polarization at low frequencies is called LFDD i.e. low frequency dielectric dispersion. The presence of this behaviour in crystalline ceramics can be explained on the basis of Maxwell-Weigner model. When frequency is low, grain boundaries are very effective and as a result there is a hopping of charge carriers. This results in high dielectric polarization and hence high values of dielectric constant. As frequency increases, responses of dipoles decreases and they lag behind the applied field and this damping nature of dipoles reduces dielectric constant.

[2] At high temperatures dielectric polarization is very high and it decreases with decrease in temperature. Increase in the values of dielectric constant with increase in temperature is because of space charge or interfacial polarization present in the ceramic sample. Space charge polarization is due to the accumulation of charge carriers in the interface between grains. As temperature increases, grain or particle size increases due to increased lattice vibrations of atoms. Increased grain size means increased hindrance to the flow of charge carriers and hence enhancement in dielectric constant values.

[3] The high values of dielectric loss, exhibited by the ceramic sample at low frequencies points to the presence of high dielectric polarization at low frequencies due to effective grain boundaries. Polarization hinders charge carriers. This increases dielectric loss, dielectric loss or energy loss is calculated as $\tan\theta$ where θ is the loss angle and tangent of loss angle depends on time varying electric fields.

[4] The high values of dielectric loss, exhibited by the ceramic sample at high temperatures points to conduction

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losses. Dielectric loss is directly proportional to temperature because as temperature increases particle or grain size increases and hindrance to the flow of current increases and overall loss during conduction increases. So it can confirm the presence of dc conductivity in the dielectric material. The presence of sharp peaks confirms Debye relaxation behaviour. Frequency dispersion positions are corresponding to dielectric relaxation. If all dipoles present in the material relax with identical time of relaxation (same relaxation time τ), with relaxation time similar to the time period of the applied field the relaxation is Debye relaxation.

[5] Impedance decreases slightly as frequency increases in the low frequency dispersion region due to the decrease in the extent of space charge polarization. In the intermediate plateau region impedance enters a constant value. In the high frequency region, again we can observe that, impedance decreases with frequency, due to decrease in the values of dielectric constant.

[6] Impedance decreases with temperature due to quantum mechanical tunnelling of charge carriers at high temperatures. At lower temperatures impedance has high values due to hopping mechanism of charge carriers.

[7] Conductivity-frequency graph yields three typical regions, a low frequency region, an intermediate frequency region and one high frequency region. In low frequency region, conductivity slightly increases with increase in frequency due to the decrease in polarization effects with frequency. Here conductivity is a frequency dependent ac conductivity and obeys Jonscher's universal power law of ac conductivity. In the intermediate frequency region, conductivity is independent of frequency and hence is called dc conductivity and obeys Jonscher's universal power law of dc conductivity. In the high frequency region, again conductivity increases with increasing frequencies due to low values of dielectric constant obeying Jonscher's universal power law of frequency dependent ac conductivity.

[8] Conductivity increases with temperature as a result of quantum mechanical tunnelling at high temperatures. At lower temperatures conductivity has high values due to hopping mechanism of charge carriers.

6. Acknowledgement

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