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Dielectric Properties and Impedance Spectroscopy of Ferroelectric Ceramic PbBaTiO₃

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ABSTRACT: Polycrystalline, ferroelectric ceramic 'PbBaTiO₃' belonging to the Barium Titanate family is synthesized by solid state reaction technique, in an attempt to analyse its crystal structure and to investigate its dielectric, impedance spectroscopic and conductivity behaviour. The formation of perovskite structure in tetragonal crystal system is detected by X - ray diffraction studies. Dielectric, impedance and conductivity properties of the pelletized ceramic sample are measured at different frequencies and temperatures and the dependence on temperature and frequency are studied. Frequencies within a range of 42Hz- 5MHz is applied to the samples at five temperatures 30⁰C, 500⁰C, 855⁰C, 900⁰C and 950⁰C in a computer controlled LCR impedance analyser in conjunction with a non-inductive furnace and a specially designed sample holder. Space charge polarization and low frequency dielectric dispersion (LFDD) are confirmed to be present in the ceramic. Non Debye relaxation behaviour and conductivity behaviour according to Jonscher's power law is observed.

KEYWORDS: Polycrystalline ferroelectric ceramic PbBaTiO₃, Dielectric properties, Impedance spectroscopy, Conductivity, Non Debye behavior, Space charge polarization, Low frequency dielectric dispersion, Jonscher's power law.

I. INTRODUCTION

Barium Titanate is one of the perovskite ferroelectric ceramic which has been the subject of intense research due to its structural stability, compositional flexibility and unique ferroelectric, piezoelectric, and pyroelectric properties. BaTiO₃ is the first perovskite discovered with ferroelectric behavior [1]. BaTiO₃ has high relative permittivity but low values for piezoelectricity and Curie temperature [2]. The literature is rich with the technological uses and applications of BaTiO₃ as high capacitance capacitors, piezoelectric sensors, positive temperature coefficient devices, ferroelectric random access memories [3], printed circuit boards [4] and electro optical devices [5]. Out of the many applications, potential applications of ferroelectric materials are in microwave tunable devices such as tunable mixers, delay lines, filters, and phase shifters for steerable antennas [6-8]. Barium titanate, since in ceramic form, can be prepared at low cost, and hence becomes a potential candidate for the above devices. The newly synthesized ferroelectric ceramic, 'PbBaTiO₃' belongs to the Barium Titanate family. The authors analyzed its crystal structure and in this paper investigated its dielectric, impedance spectroscopic and conductivity behaviour. Barium titanate tetragonal crystal structure is shown in fig.1.

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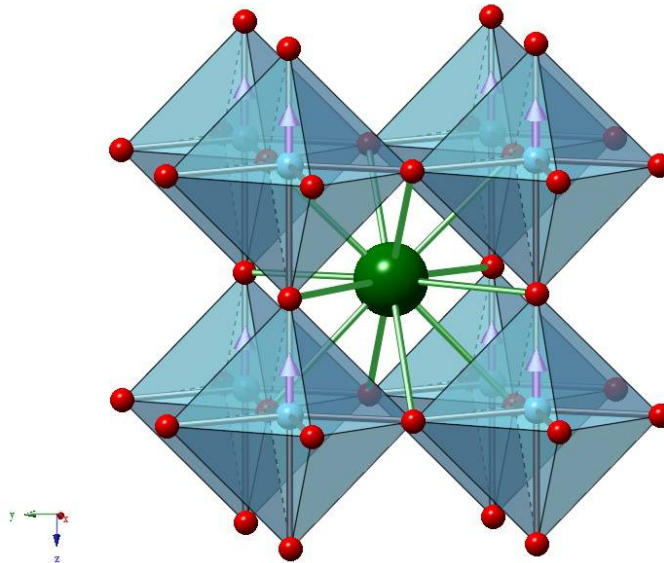


Fig. 1. Tetragonal structure of barium titanate

The reason for dielectric polarization in polycrystalline ceramics is the presence of grain boundaries, interfaces and micro cracks which act as dielectric media. Here shifting of electric charges from equilibrium position, resulting in dielectric polarization of charge centres, takes place at grain or particle interfaces when an electric field is applied. Here the effects of temperature and frequency on dielectric parameters such as dielectric constant, dielectric loss, impedance and conductivity of PbBaTiO_3 are studied. Both ac and dc conductivity are analysed. Dielectric spectroscopy or impedance spectroscopy measures the properties of a dielectric medium as a function of frequency [9]. In order to (i) distinguish between grain and grain boundary effects, (ii) make an interpretation of the microscopic processes that allow discerning between dielectric relaxation and long range conductivity processes (i.e., localized and non-localized conduction etc.) [10] and (iii) allow distinguishing between the different contributions of the intra grain conductivity, impedance spectroscopy has been widely used in last couple of years, being a powerful technique for the study of the dielectric behavior of crystalline and polycrystalline ceramic materials, especially ionic conductors, ferroelectrics and ferromagnetic [10-13].

II. THEORY OF DIELECTRIC PROPERTIES

2.1. DIELECTRIC CONSTANT

Dielectric constant or relative permittivity is a number denoting, the ratio of the permittivity of the material to the permittivity of free space or the ratio of the capacitance of a capacitor with dielectric material between the conducting plates to the capacitance of a capacitor with free space or vacuum between the plates. Dielectric constant is a measure of the degree of polarization in the presence of an external field. Dielectric constant (ϵ) in a parallel plate capacitor (Fig.2) is given by

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

$$C_0 = A\epsilon_0/d \text{----- [2] where}$$

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 (8.85×10^{-12} F/m)

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A = Area of the prepared sample
d = Thickness of the sample

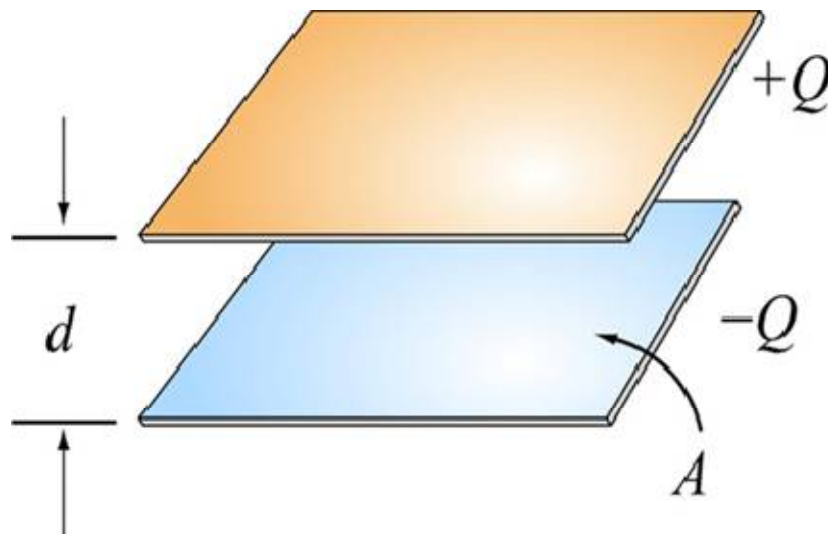


Fig. 2. Illustration of a parallel plate capacitor

2.2. DIELECTRIC LOSS

Dielectric loss quantifies a dielectric material's inherent dissipation of electromagnetic energy (E.g. heat) [14]. Dielectric loss is denoted by two terms, namely, loss angle θ and loss tangent $\tan \theta$. The loss tangent is usually given by the ratio of the imaginary and real parts of dielectric constant. Frequency, temperature, humidity, voltage etc. are some factors which affect the values of dielectric loss. Dielectric loss is given by

$$\tan(\theta) = \epsilon''/\epsilon' \text{-----}[3]$$

For dielectrics with small loss, this angle is $\ll 1$ and $\tan(\theta) \approx \theta$

2.3. IMPEDANCE SPECTROSCOPY

Dielectric spectroscopy (sometimes called impedance spectroscopy), and also known as electro chemical impedance spectroscopy (EIS), measures the dielectric properties of a medium as a function of frequency [15-18]. Impedance is a complex quantity and it is a combination of resistance and reactance or we can call it as the effective resistance offered by the dielectric material when an ac current is passed through it. In Cartesian form, impedance is given as,

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

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

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

R is the resistance and X is the reactance [19]. In polar form impedance is

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

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

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

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Magnitude and phase information of impedance is given by the polar form. Impedance spectroscopy is very useful non-destructive method used for analysing the frequency related dielectric responses of grains at grain interfaces of the ceramic and in understanding the defects and imperfections in the crystal lattice.

2.4. CONDUCTIVITY ANALYSIS

Electrical conductivity simply denotes the extent to which a material allows the passage of electric current through it. Sometimes it is called specific conductance. Conductivity is calculated as the reciprocal of resistivity with unit 'mho'. Conductivity is an inherent property of a material. It is unaffected by the shape, size or mass of the object. Changes in frequency and temperature can produce changes in the values of conductivity. From conductivity we can extract conductance, an extrinsic property which is the reciprocal of resistance. Conductivity is given by

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

G is conductance, σ is conductivity, A is the total surface area and d is the thickness of the given sample of material.

JONSCHER'S UNIVERSAL POWER LAW

In conductivity against frequency plot (frequency in x axis, conductivity in y axis), three regions, a low frequency region, an intermediate frequency region (intermediate plateau region) and a high frequency region can be observed. Conductivity is directly proportional to frequency in the low frequency region as well as in the high frequency region. In the intermediate plateau, conductivity is independent of frequency. Frequency dependent conductivity is called ac conductivity and frequency independent conductivity is called dc conductivity. Jonscher's universal power law has equations denoting ac conductivity as well as dc conductivity and they are given below.

$$\sigma_{dc} = 1/R_b A \text{-----}[11]$$

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

R_b is the bulk resistance, ω is the frequency and A is the area.

2.5. POLARIZATION, DISPERSION & RELAXATION

Dielectric polarization in a material is due to any one or a combination of different polarization mechanisms. Five major types of polarizations are

- Electronic polarization
- Ionic polarization
- Orientational polarization
- Space charge/interfacial polarization and
- Spontaneous polarization

For a polycrystalline heterogeneous ceramic material interfacial or space charge polarization plays a prominent role. This is due to the accumulation of charge at an interface between two grains or particles within the crystal lattice of

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the material because of an external field. This polarization affects free charges in the crystal. A very important point is that since charges are free charges, defects such as grain boundaries or other interfaces act as a medium for polarization. Dielectric dispersion is defined as the dependence of the dielectric constant of a material on the frequency of applied electric field. There will always be a time lag between polarization of dipoles with variation in frequency. Dielectric dispersion is a very important term while analysing the dielectric properties of a material.

When an external electric field is applied, growth of dielectric polarization of dipoles to the maximum value, takes a finite time. When the external field is withdrawn, decay of dielectric polarization of dipoles takes a finite time. There will be a time lag or time delay between polarizations of dipoles of a material with changes in electric field. This is analogous to magnetic hysteresis of magnetic materials in varying magnetic fields. Relaxation can be defined as the lagging of polarization (alignment of dipoles) behind an applied electric field. The time lag implies an irreversible degradation of Gibbs free energy.

Relaxation is a function of frequency and usually denoted in terms of permittivity. For ideal systems, with non-interacting populations of dipoles, relaxation can be explained by the Debye equation and is called Debye relaxation. If all dipoles present in the material relax with identical time of relaxation (same relaxation time), with relaxation time similar to the time period of the applied field, i.e., when relaxation time, τ , is the same as $1/\omega$, where ω is the angular frequency of the applied field, then it is called Debye relaxation. It is usually expressed in the complex permittivity ϵ of the medium as a function of the frequency

$$\hat{\epsilon}(\omega) = \epsilon_{\infty} + \frac{\Delta\epsilon}{1 + i\omega\tau},$$

where ϵ_{∞} is the dielectric constant at the highest frequency and $\Delta\epsilon = \epsilon_s - \epsilon_{\infty}$ where ϵ_s is the dielectric constant at the lowest frequency and τ is the relaxation time

Real and imaginary parts of the complex dielectric permittivity yields,

$$\epsilon' = \epsilon_{\infty} + \frac{\epsilon_s - \epsilon_{\infty}}{1 + \omega^2\tau^2}$$

$$\epsilon'' = \frac{(\epsilon_s - \epsilon_{\infty})\omega\tau}{1 + \omega^2\tau^2}$$

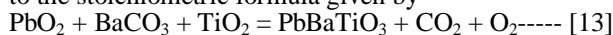
The dielectric loss is also represented by

$$\tan(\delta) = \frac{\epsilon''}{\epsilon'} = \frac{(\epsilon_s - \epsilon_{\infty})\omega\tau}{\epsilon_s + \epsilon_{\infty}\omega^2\tau^2}$$

But in many dielectric materials, dipoles have more than one relaxation time. Then they do not follow Debye equation but follows Cole- Cole equations.

III. MATERIALS AND EXPERIMENTAL METHODS

Solid state reaction technique is used to prepare the new perovskite structured, ferroelectric ceramic sample PbBaTiO_3 from high purity ($\geq 99.9\%$) ingredients of carbonates and oxides; BaCO_3 , TiO_2 and PbO_2 . They are weighed according to the stoichiometric formula given by



The precursors are mixed thoroughly in an agate mortar for 12 hours to obtain a homogeneous fine powder. The powder sample is ball milled with zirconium beads for three months followed by attrition milling for 3 hours. Solid-state reaction was carried out in an air atmosphere. The fine powder is fired at different temperatures 300°C , 500°C , 850°C and 900°C in a special furnace. Final calcination is at optimized temperature 900°C . A part of the calcined fine powder is pressed into pellets in a hydraulic press. These pellets are sintered at 1100°C for 6 h (optimized temperature and time) and then slowly cooled to room temperature. Subsequently, the pellets are cleaned, polished and coated with

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conductive silver paste on both sides which serve as conducting parallel plates and dried at 150°C for 4 hour to remove any moisture present. Before carrying out dielectric measurements the pellets are cooled to room temperature. A pellet of 9.726 mm diameter and 2 mm thickness is inserted in a laboratory made sample holder. Pellet with silver coating acts as a parallel plate capacitor. Sample holder is inserted in the furnace which is connected to an LCR Impedance analyser and computer and different dielectric properties of the prepared sample is measured at 30°C, 500 °C, 855°C, 900°C and 950°C as a function of frequency. Frequency remains within a limit 42Hz - 5 MHz

IV. RESULTS AND DISCUSSIONS

4.1. DIELECTRIC CONSTANT

- EFFECT OF FREQUENCY**

Graphs are plotted with dielectric constant on y axis and frequency on x axis for five various temperatures, namely, 30°C, 500°C, 855°C, 900°C and 950°C (Fig.3). It is clear that dielectric constant decreases with increase in frequency up to 25KHz, after that dielectric constant remains almost constant especially for lower temperatures. Maximum value of dielectric constant (in the order of 10⁶) is obtained at 950°C at a frequency of 42 Hz. It is conclusive that dielectric constant found most higher values in the low frequency region, below 100 Hz for all taken temperatures.

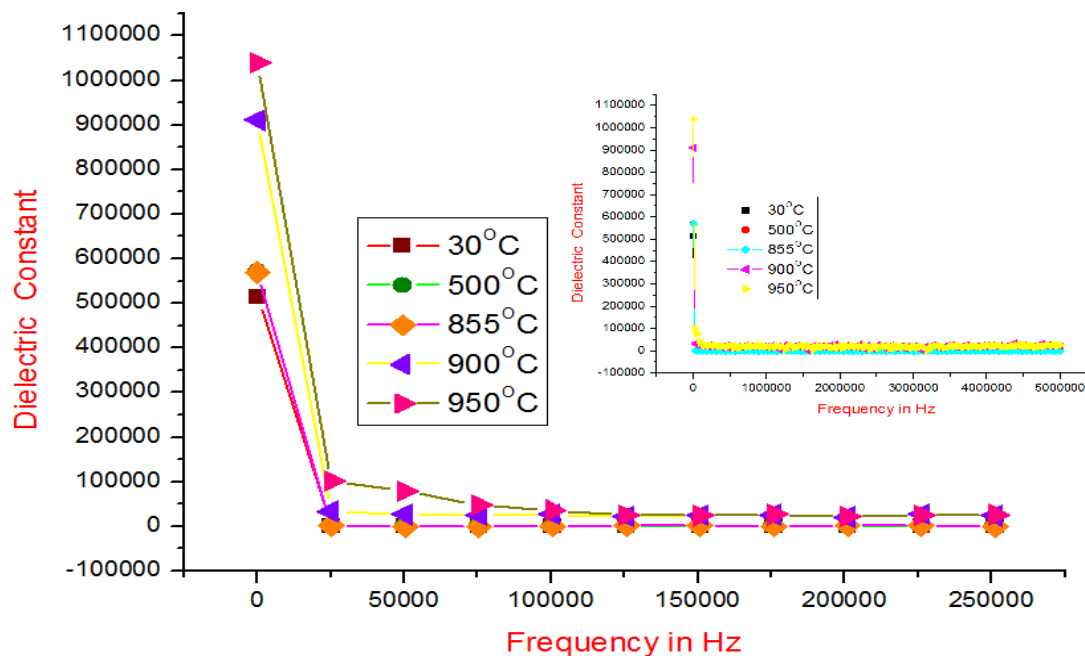


Fig.3. Variation of Dielectric constant with frequency

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• EFFECT OF TEMPERATURE

The graph given below (Fig.4) shows the relation between dielectric constant and temperature in the frequency limit 42 Hz to 5 MHz. Dielectric constant increases with temperature slowly from 30 to 500 degree Celsius and suddenly from 500 to 950 degree Celsius. Dielectric constant attains very high value in the order of 10^6 at 950 degree Celsius. Higher values of dielectric constant are obtained for low frequencies below 100 Hz. Obviously dielectric constant increases as temperature increases.

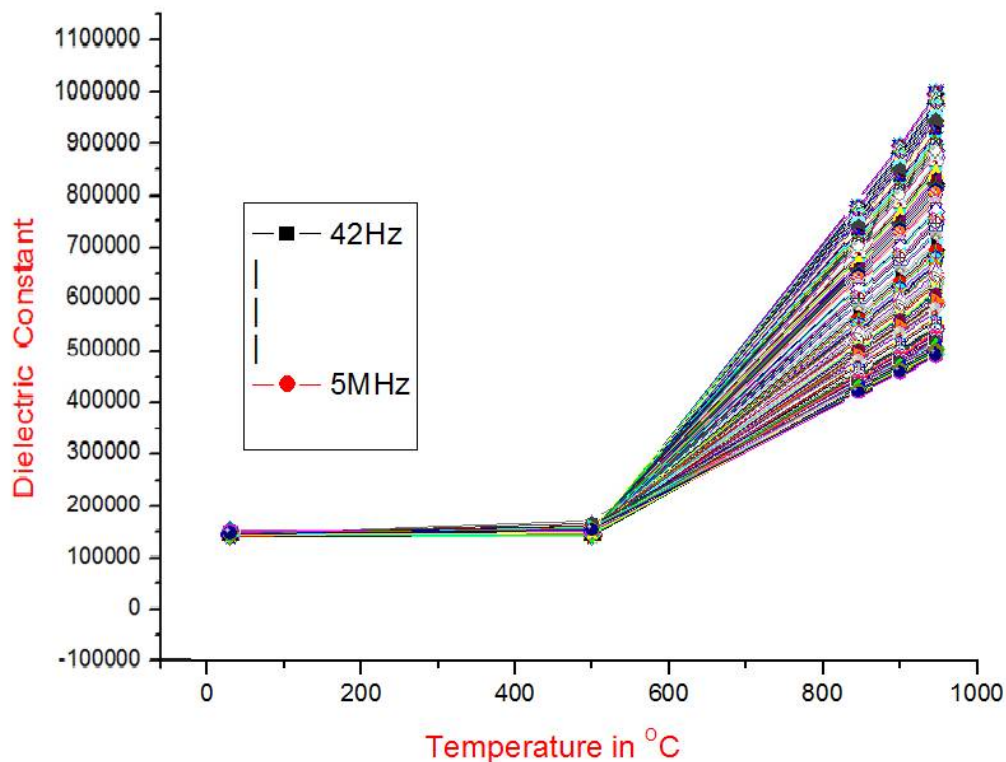


Fig.4. Variation of dielectric constant with temperature

It is evident from fig.3 and fig.4 that, dielectric constant has higher values at higher temperatures (maximum at 950°C) and at lower frequencies (below 100Hz). Dielectric constant decreases with frequency and increases with temperature.

[1] The sample exhibits a low frequency dielectric dispersion also known as LFDD characterized by the fall in dielectric polarization with advancing frequencies. Maxwell-Weigner model in dielectrics [20, 21] explains the reason for the decay of dielectric polarization with increasing frequency. We know that a polycrystalline, non-homogenous, ceramic contains grains and these grains are separated by grain boundaries. Here dielectric polarization of charge carriers, takes place at grain or particle interfaces when an electric field is applied. This type of polarization is called interfacial or space charge polarization. Space charge polarization occurs from the alignment of charges in the field direction, at the grain boundaries within the crystal lattice on the application of an external electric field. At low

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frequencies space charge polarization dominates other types of polarizations. When given frequency increases, electric field oscillates with greater frequency and the time constant of the applied electric field exceeds the dipolar polarization characteristic time (definite time for polarization) of dipoles. This causes a lagging of electric dipoles behind the applied field. This damping nature of electric dipoles results in the fall of dielectric polarization and fall in the values of dielectric constant with increasing frequencies.

[2] High temperatures are favourable for dielectric polarization. Dielectric constant increases with temperature. This is caused by the dominant space charge polarization in the ceramic sample. As temperature of a crystal increases, atoms in the crystal lattice began to vibrate with more energy. This results in expansion of unit cell. As a result, particle size or grain size increases and grain boundaries become very effective and they limit the movement of charges and larger will be the dielectric polarization. As temperature increases dielectric polarization increases and the value of dielectric constant increases.

4.2.DIELECTRIC LOSS

• EFFECT OF FREQUENCY

From fig.5 which connects dielectric loss and frequency (frequency range 42 Hz to 5000000 Hz) at various temperatures 30°C, 500°C, 855°C, 900°C and 950°C, it is observed that dielectric loss is high at lower frequencies. Loss decreases with frequency. Dielectric loss is maximum at 950°C below 100 Hz. In the lower frequency region at 42 Hz, there are continuations from a broad peaks for all temperatures. Broad peaks denoting dielectric relaxation for different temperatures are partially visible (clearly visible only if frequencies are below zero Hertz).

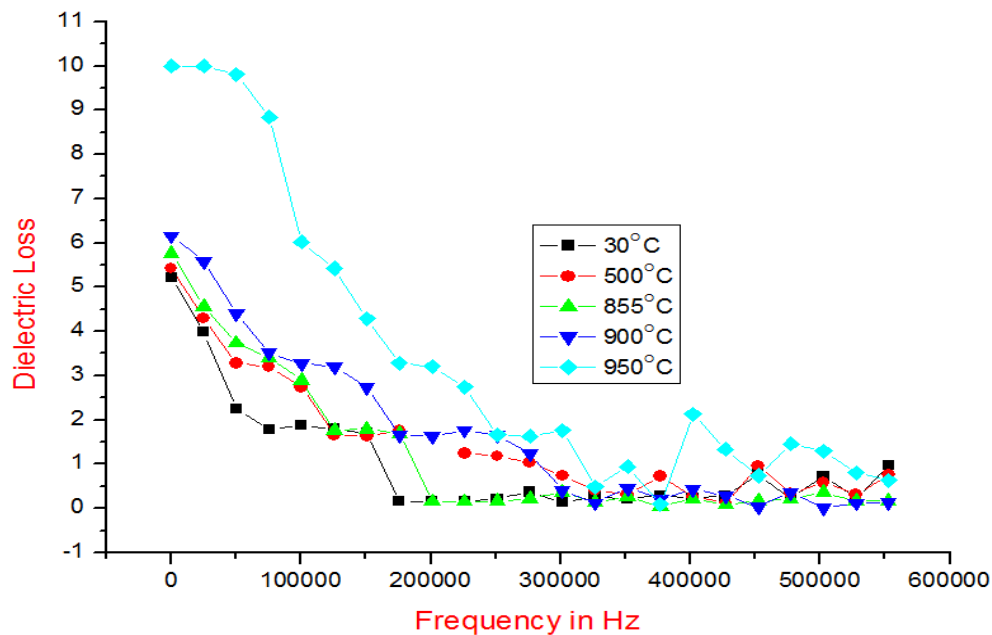


Fig.5 Variation of dielectric loss with frequency

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Fig.6 is the graph demonstrating the dependence of dielectric loss on temperature. It is observed that dielectric loss increases with temperature very slowly up to 850 degree Celsius and after that increases rapidly. In the high frequency region there are some negligible fluctuations. Maximum values of dielectric loss occur at low frequencies below 100 Hz. Peak value of loss is obtained at 950°C and at 42 Hz.

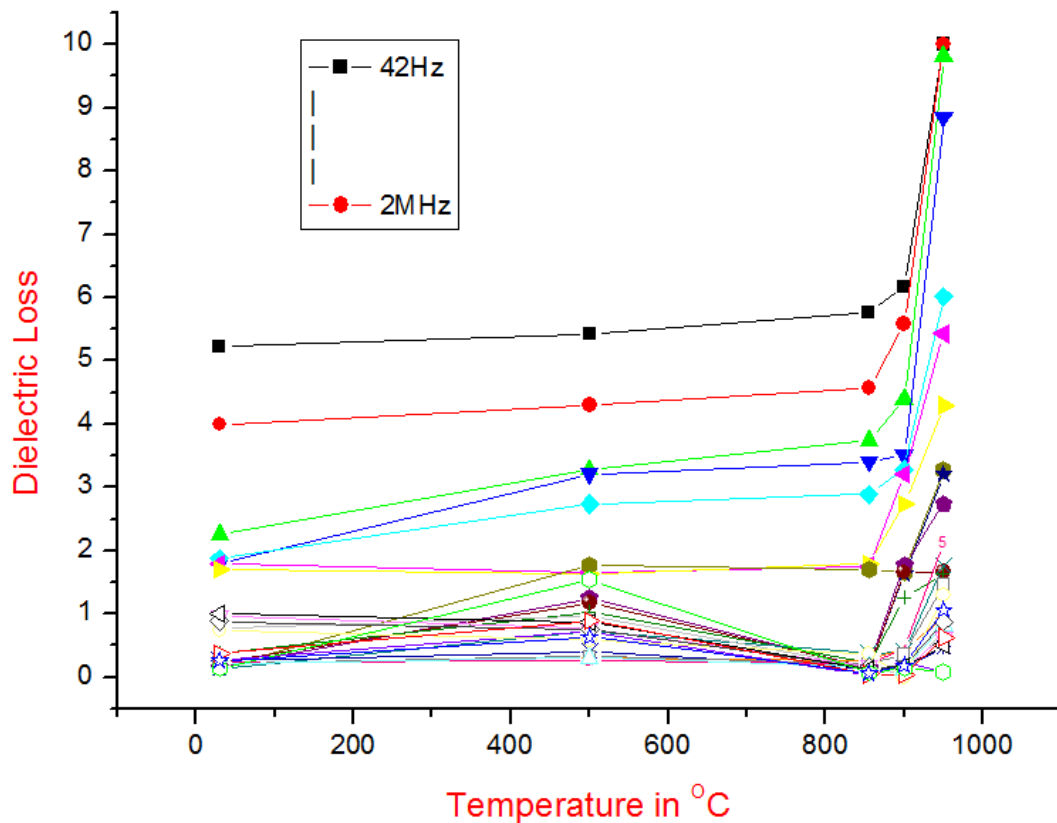


Fig.6. Variation of dielectric loss with temperature

From figures 5 & 6 it is crystal clear that dielectric loss has high values in the low frequency region (below 100 Hz) and it decreases with increase in frequency. Also maximum value of dielectric loss is obtained at high temperature (950 degree Celsius) and it decreases with decrease in temperature. So loss increases with temperature and decreases with frequency.

1. High values of dielectric loss are observed in the low frequency region. This points to the dominant space charge polarization at low frequencies. Very active grain boundaries contribute to dielectric polarization of charge carriers at grain boundaries. This results in large values of dielectric constant. As frequency increases fall in dielectric polarization causes fall in dielectric loss. Since dielectric loss depends on dielectric constant, loss also decreases with frequency just like dielectric constant.

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2. Dielectric loss is high at high temperatures because of the loss of electric energy as heat. As temperature increases vibrations of atoms within the crystal lattice increases. This causes increase in particle size. Large particles cause more hindrance to the flow of charges and this increases dissipation of electric energy as heat. Enhanced dc conduction of current (excitation of electrons from valence band to conduction band consuming thermal energy causes conduction current) in the sample at high temperatures is another reason.

3. Broad peaks obtained from dielectric loss- frequency graphs correspond to Cole – Cole model of non-Debye dielectric relaxation where different dipoles have different relaxation times. Relaxation time lags behind the time constant of the applied field.

4.3. IMPEDANCE

- **EFFECT OF FREQUENCY [BODE PLOT]**

To demonstrate the effects of frequency on impedance of the ceramic sample, Bode plots are plotted with log values of frequency on x axis and log values of dielectric constant on y axis at 30°C, 500 °C, 855°C, 900°C and 950°C (Fig.7).

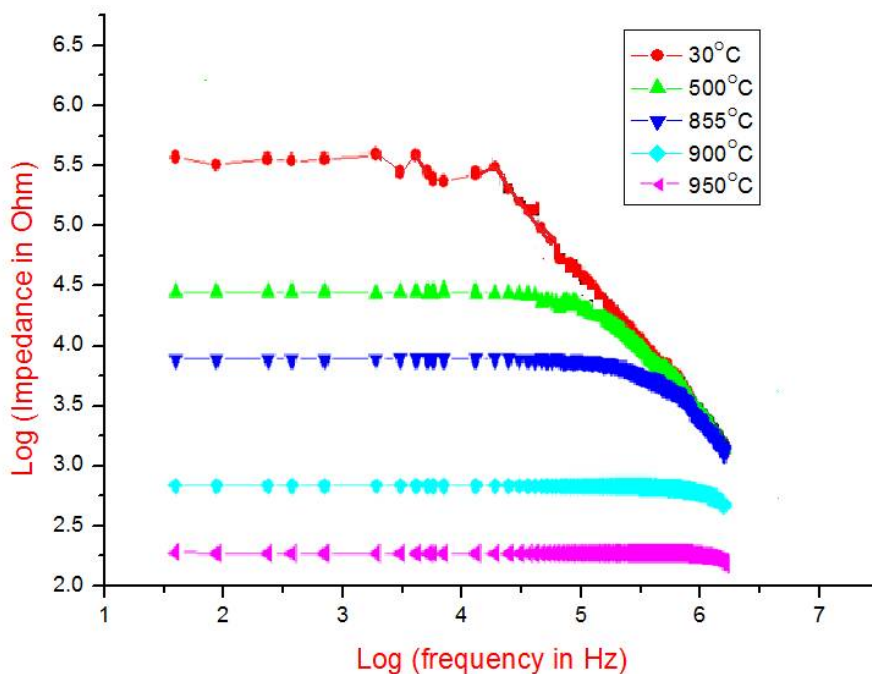


Fig.7. Variation of log of impedance with log of frequency

Impedance curves for all temperature show very small decrease with frequency in the low frequency region. Here impedance can be considered almost constant. The frequency at which impedance began to lower significantly is

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different for different temperatures. For lower temperatures decrease in impedance starts comparatively earlier and the decrease is abrupt. For higher temperatures decrease in impedance starts only at very higher frequencies and the decrease is slight. Peak value of impedance is observed at 30 degree Celsius in the low frequency region i.e. below log2.

- **EFFECT OF TEMPERATURE**

Plotting temperature in x axis and impedance in y axis we studied the effect of temperature on impedance (Fig.8). Impedance, at every taken temperature, falls with increasing temperature. At the lowest frequency taken for experiment (42 Hz) and at 30°C, maximum value of impedance is observed. For temperatures 900°C and 950°C impedance is very low. So we can conclude that high frequencies and high temperatures are not favourable for impedance. Impedance decreases with temperature and frequency.

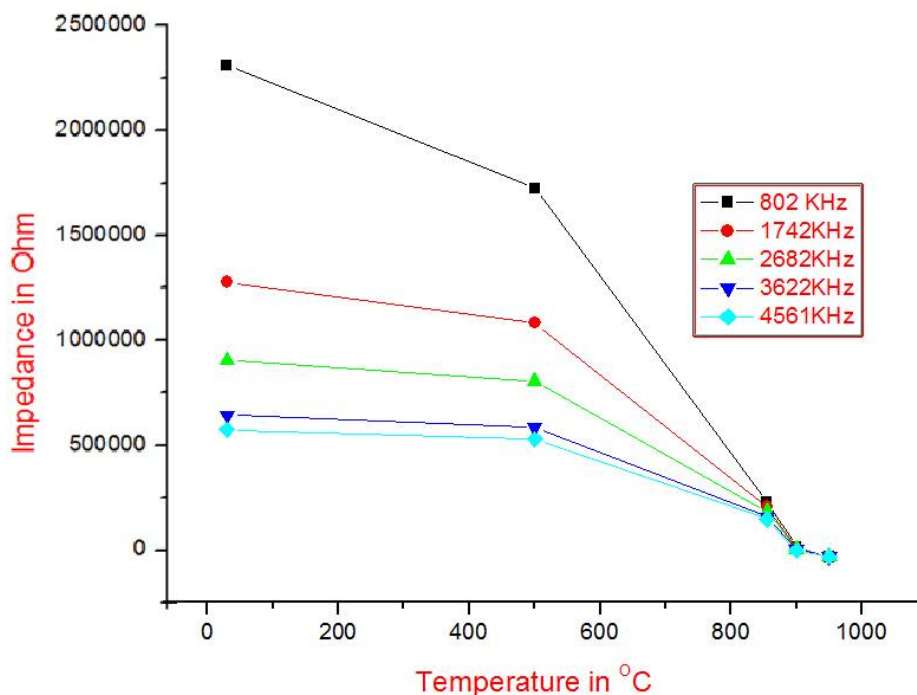


Fig.8. Variation of impedance with temperature

- **COLE-COLE PLOT**

Cole-Cole plots are constructed plotting real part of dielectric constant on x axis and imaginary part of dielectric constant on y axis (Fig.9). Semicircular curves are obtained which confirms the dielectric responses of the prepared ceramic. A variation from perfect semicircular shape is due to the loss of energy at interfaces and other defects present in the lattice. Cole-Cole model of non-Debye relaxation is also confirmed by the semicircular curves. Cole-Cole plots at various temperatures (Fig.9) are given below.

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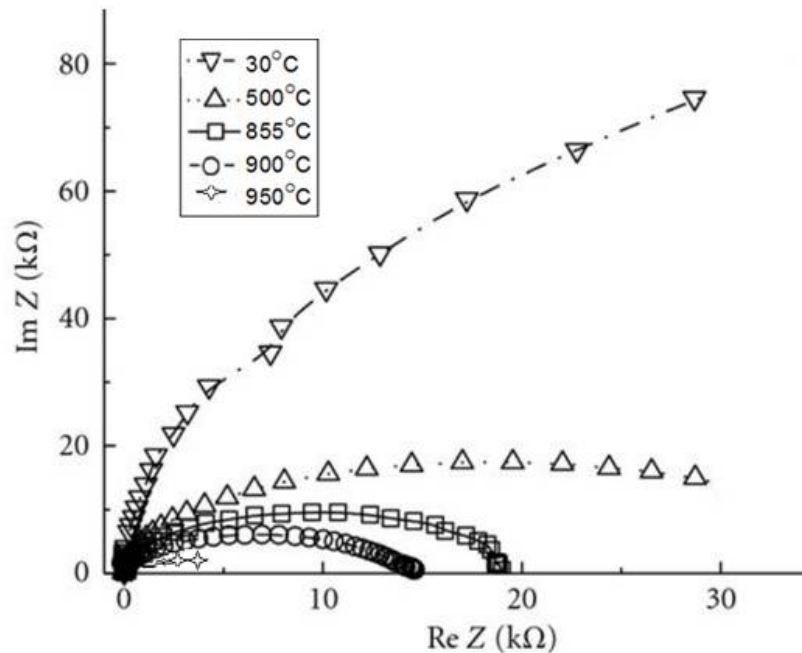


Fig. 9. Cole-Cole plot at different temperatures

Analysing the graphs shown by fig.7 & fig.8 it is obvious that the ferroelectric ceramic sample has high values of impedance at smaller frequencies. Similarly the sample has high impedance values when temperature is low. Impedance decreases with both temperature and frequency.

1. Space charge polarization falls with advancing frequency. As a result impedance also falls with advancing frequency. Another factor responsible for fall in impedance is the presence of frequency independent dc conductivity.
2. Despite the fact that very high dielectric polarization is there at high temperatures, impedance decreases with temperature. This is because of the presence of activation energies to drive charged particles across grain boundaries. Quantum mechanical tunnelling of charge carriers provides this activation energy at high temperatures and hopping of charge carriers provides the activation energy at lower temperatures, which reduces impedance [22].
3. Semicircular curves in the Cole-Cole plots at various temperatures confirm the dielectric behaviour of the sample in an applied field. A variation from perfect semicircular shape is due to the loss of energy at interfaces and other defects present in the lattice. Cole-Cole model of non-Debye relaxation is also confirmed by the semicircular curves.

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4.4. CONDUCTIVITY

- *EFFECT OF FREQUENCY*

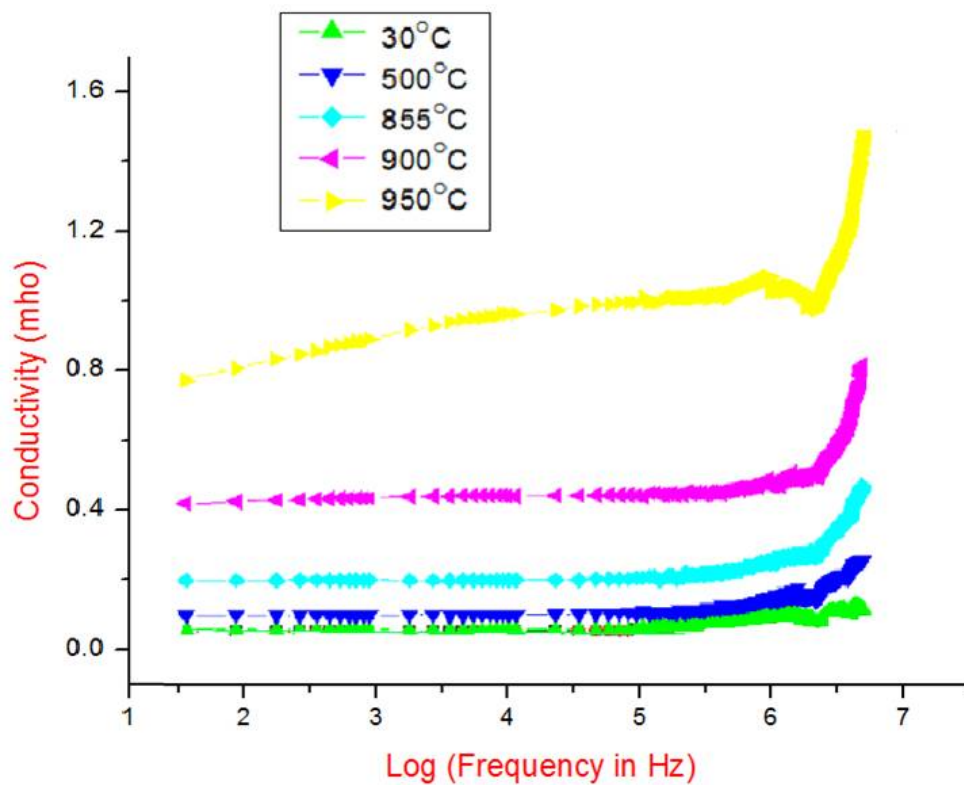


Fig.10. Variation of conductivity with log of frequency

A graph is drawn to analyse how conductivity affects the electric behaviour of the synthesised sample. Log of frequency (frequency limit 42 Hz – 5MHz) is taken at x axis and conductivity is taken at y axis. Graphs are constructed for five temperatures 30, 500, 855, 900 and 950 degree Celsius (Fig.10). In the low frequency region, values of conductivity are low for all temperatures. It shows negligible increase with frequency so that it can be considered almost constant. At the very high frequency region of the graph (Fig.10) conductivity increases very abruptly with temperature. The peak value of conductivity is obtained at 950 degree Celsius in the high frequency region.

- *EFFECT OF TEMPERATURE*

Temperature affects the dielectric properties of the sample. To study the dielectric behaviour of the sample with increasing values of thermal energy, a graph is constructed with temperature on x axis and conductivity on y axis from

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a frequency range of 42 Hz to 5 MHz (Fig.11). Graphs show that conductivity is gradually increasing with temperature for all frequencies. Maximum value of conductivity is at 950°C in the higher frequency region.

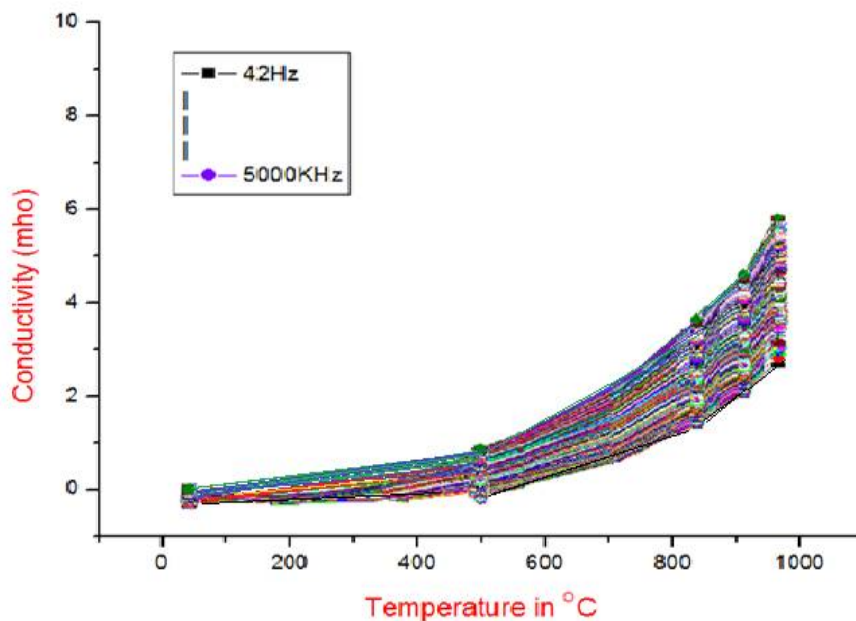


Fig.11. Variation of conductivity with temperature.

It is crystal clear from figures 10 and 11 that conductivity increases with frequency as well as temperature.

1. At lower frequencies, conductivity increases so slightly with frequency that it can be considered negligible and hence conductivity can be taken as frequency independent dc conductivity or bulk conductivity. Excitation of valence electrons from valence band to conduction band at high temperatures causes dc conductivity. At higher frequencies, conductivity increases abruptly with frequencies due to minimum polarization and due to frequency dependent ac conductivity. Both ac and dc conductivity obeys Jonscher's universal power law of conductivity.

2. Even though dielectric polarization is very strong at high temperatures conductivity increases with increasing temperature. Explanation is given below. Increase in particle size favours space charge polarization at high temperatures, so usually low conductivity is expected. But if there is an accumulation of charge carriers trapped in the grains, grain boundaries and voids of the lattice, conduction across grain boundaries is possible, provided that, there is sufficient activation energy. In our sample at low temperatures activation energy is supplied by hopping of charge carriers. At high temperatures, activation energy is provided by quantum mechanical tunnelling.

V. CONCLUSIONS

Synthesized polycrystalline ferroelectric ceramic 'PbBaTiO₃' by solid state reaction method using a special furnace and investigated its dielectric, impedance spectroscopic and conductivity behavior at different temperatures (at 30°C, 500°C, 855°C, 900°C and 950°C) and frequencies (ranging 42 Hz-5 MHz). The material shows very good dielectric properties

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with very high values of dielectric constant (up to the power of 10^6) and impedance, very low values of conductivity and dielectric loss, at frequencies between 42-100 Hz.

[1] The low frequency dielectric dispersion exhibited by the sample is characterized by the fall in dielectric polarization with advancing frequencies. Maxwell-Weigner model in dielectrics explains the reason for the decay of dielectric polarization with increasing frequency. In a polycrystalline, non-homogenous, ceramic there are grains and grain boundaries. Here dielectric polarization of charge carriers, takes place at particle interfaces when an electric field is applied. This type of polarization is called space charge polarization. At low frequencies space charge polarization dominates other types of polarizations. When given frequency increases, electric field oscillates with greater frequency and the time constant of the applied electric field exceeds the dielectric polarization characteristic time of dipoles. This causes a lagging of electric dipoles behind the applied field. This damping nature of electric dipoles results in the fall of dielectric polarization and fall in the values of dielectric constant with increasing frequencies.

[2] High temperatures are favourable for dielectric polarization. Dielectric constant increases with temperature. As temperature of a crystal increases, atoms in the crystal lattice began to vibrate with more energy. This results in expansion of unit cell and particle size. Grain boundaries become very effective and they limit the movement of charges and larger will be the dielectric polarization. As temperature increases dielectric polarization increases and the value of dielectric constant increases.

[3] High values of dielectric loss are observed in the low frequency region due to dominant space charge polarization. Very active grain boundaries contribute to dielectric polarization of charge carriers at grain boundaries. This results in large values of dielectric constant. As frequency increases fall in dielectric polarization causes fall in dielectric loss. Since dielectric loss depends on dielectric constant, loss also decreases with increasing frequency just like dielectric constant.

[4] Dielectric loss is high at high temperatures because of the loss of electric energy as heat. As temperature increases particle size also increases. Large particles cause more hindrance to the flow of charges and this increases dissipation of electric energy as heat. Enhanced dc conduction of current in the sample at high temperatures is another reason for high dielectric loss at high temperatures.

[5] Broad peaks obtained from dielectric loss - frequency graphs correspond to Cole – Cole model of non-Debye dielectric relaxation where different dipoles have different relaxation times. Relaxation time lags behind the time constant of the applied field.

[6] Space charge polarization falls with advancing frequency. As a result impedance also falls with advancing frequency. Another factor responsible for fall in impedance is the presence of frequency independent dc conductivity.

[7] Despite the fact that very high dielectric polarization is there at high temperatures, impedance decreases with temperature because of activation energies which drive charged particles across grain boundaries. Quantum mechanical tunnelling of charge carriers provides this activation energy at high temperatures and hopping of charge carriers provides the activation energy at lower temperature.

[8] Semicircular curves in the Cole-Cole plots at various temperatures confirm the dielectric behaviour of the sample in an applied field. Variations from perfect semicircular shape are due to the loss of energy at interfaces and other defects present in the lattice. Cole-Cole model of non-Debye relaxation is also confirmed by the semicircular curves.

[9] At lower frequencies, conductivity increases so slightly with frequency that it can be considered negligible and hence conductivity can be taken as frequency independent dc conductivity. At higher frequencies, conductivity

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increases abruptly with frequencies due to minimum polarization and due to frequency dependent ac conductivity. Both ac and dc conductivity obey Jonscher's universal power law of conductivity.

[10] Even though dielectric polarization is very strong at high temperatures conductivity increases with increasing temperature. Increased particle size favours space charge polarization at high temperatures, so usually low conductivity is expected. But if there is an accumulation of charge carriers trapped in the grains, grain boundaries and voids of the lattice, conduction across grain boundaries is possible, provided that, there is sufficient activation energy. In our sample at low temperatures activation energy is supplied by hopping of charge carriers. At high temperatures, activation energy is provided by quantum mechanical tunnelling.

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