Dielectric Response and Transport Properties of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ [Lead Strontium Calcium Copper Oxide]

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High dielectric permittivity, good mechanical properties and excellent thermal stability are highly desired qualities for the dielectric materials used in embedded capacitors and energy-storage devices. This study reports the temperature dependence and very low loss factor of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ ceramics. X-ray diffraction (XRD) and scanning electron microscope (SEM) are used to analyze the sample. The dielectric properties of the material prepared has been investigated in the frequency range 42–1 MHz and in the temperature range 303–873 K. The result reveals that $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ have high dielectric constant with significant temperature and frequency dependence which makes them appropriate in the application of miniaturization of electronic devices. The dielectric constant and the loss factor decreases with increase in frequency of the applied field. Impedance studies confirm that dielectric relaxation in the material is of non-Debye type. Conductivity studies proves quality information related to the charge transport properties. The frequency analysis of modulus properties pictures a distribution of the relaxation time existing in the system. The new ceramic material $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ will be a promising candidate for uses in embedded capacitors, especially in high-temperature circumstance.

Keywords: $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$; dielectrics; impedance spectroscopy; Cole–Cole plot.

1. Introduction

Ceramic composites have always attracted research world due to their potential applications as catalysts, sensors, and biomaterials. They do not show any characteristics of micro and macro metric materials.¹ Methods like ion-sputtering,^{2–4} plasma techniques,^{5,6} cathode arc deposition,⁷ sol–gel method,^{8,9} and solid state methods open wide opportunities to study the electrical properties at direct and

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alternating currents.¹⁰ Embedded passive components like resistors, capacitors, and inductors are essentially needed for the rapid development of microelectronic research. Capacitors have grabbed attention in recent times due to their varied uses in filtering, timing, A/D conversion, termination, decoupling, and energy storage applications. The capacitance (C) of an embedded capacitor is calculated by the dielectric permittivity (ϵ), area (A) and thickness (d) of the dielectric materials as, $C = [\epsilon_0 \epsilon_r A/d]$ (where ϵ_0 is the dielectric permittivity in vacuum, 8.854 pF/m and $\epsilon = \epsilon_0 \epsilon_r$). Hence on the printing circuit boards (PCBs), the dielectric material should possess high dielectric permittivity to obtain a high capacitance. Generally, it is not possible for a single component material simultaneously to possess high permittivity and mechanical flexibility for the applications in integrated PCBs.¹¹ Through careful and controlled preparation, processing and judicious selection of material compositions it is possible to design and create new materials with unprecedented flexibility and improvements in their physical properties.¹²

Ceramic materials like BaTiO₃, PZT, and CaCuTi₄O₁₂ own remarkable dielectric permittivity and excellent thermal stability. They are widely used in devices such as memory elements, ultrasound generators, capacitors, gas igniters, etc. Lead Zirconate Titanate (Pb (Zr_xTi_{1-x}) O₃), is a most promising ceramic material which has been studied extensively. The PZT (PbZrO₃-PbTiO₃) is another reported ceramic material for a variety of microelectronic and sensing applications.¹³

Pb₂Sr₂CaCu₂O₉, new ceramic material designed and synthesized with an aim to explore the possibility of its various applications. The dielectric studies of Pb₂Sr₂CaCu₂O₉ ceramic materials are rarely reported. The structural and phase formation are confirmed by XRD analysis.¹⁴ The morphology of the sample is obtained through SEM analysis. Conductivity measurements are greatly used to investigate the nature of ionic mobility in conducting materials. Electrical behaviors of complex ionically conducting materials plotted as a function of frequency and first reported by Jonscher^{15,16} are known as universal dielectric response (UDR).¹⁷ The investigations performed into electrical properties of ceramic composites at alternating currents have confirmed that current conduction within a certain frequency range is realized by hopping exchange of electrons.¹⁸⁻²⁰

The objective of the present work is to study the dielectric and electrical impedance properties of the sample Pb₂Sr₂CaCu₂O₉ in the range of frequencies (42 Hz–1 MHz) at different temperatures with a view to understand the conductivity mechanisms involved in this new material.

2. Experimental Procedures

The Lead Strontium Calcium Copper Oxide (Pb₂Sr₂CaCu₂O₉) belongs to cuprate family with perovskite structure. Calcium-doped ceramics are prepared by the conventional solid state reaction technique and are reported previously.¹⁴ The reagent grade chemicals of high purity (Lead Oxide, Strontium Oxide, and Calcium Oxide and Cupric Oxide powders) are used as the raw materials and are weighed

according to their molecular formula. Crystal structure of the sample is characterized using Bruker AXS D8 advance diffractometer. Morphology of the sample has been determined with the scanning electron microscopy SEM- (JEOL/EO Model JSM-6390). The obtained sample is then crushed into a fine powder and pelletized using hydraulic press so that maximum densification is obtained. The pellets with dimensions of 9.27 mm diameter $\times$ 3.07 mm thickness are prepared. All the pellets are sintered and silver paste applied on both sides to serve as conducting electrodes. The whole arrangement is kept in non-inductive furnace for heating at different temperatures.²¹ The dielectric behavior, impedance studies, and the conductivity measurements in the temperature range 30–600°C with 42 Hz to 1 MHz frequency variations have been thoroughly studied using HIOKI-3532-50 LCR Hi TESTER.

3. Results and Discussion

3.1. Structural properties

The representative XRD spectrum of sample $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at 950°C is depicted in Fig. 1.

Highly intense and sharp peaks are observed in the XRD profile. From the XRD data, the crystallite size of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ is calculated using Scherer equation (24.49 nm) and reported previously. The XRD studies conclude that this crystal belongs to the orthogonal system.¹⁴

3.2. Morphology

SEM data give the surface morphology of the sample. Figure 2 shows the surface morphology of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}$. Since the crystalline size is less, agglomeration of

Fig. 1. XRD of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at 950°C.

Fig. 2. SEM morphology of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$.

grains are visible. From the SEM data, the crystallite size is found to be in the range 25–30 nm confirming the XRD results.

3.3. Dielectric studies

The dielectric studies of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ with respect to frequency at different temperatures are conducted. The dielectric constant (ϵ) of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ ceramics exhibits strong dependence on frequency as shown in Fig. 3(a). This measures two major electrical properties of material — the capacitive nature and the conductive nature. The results in the present work can be concluded as due to total polarization. The sample exhibits strong frequency dispersion at low frequency [LFDD]. This decrease in the value of dielectric constant ϵ reaching a constant value at higher frequencies proves the presence of interfacial polarization.

Figure 3(b) shows the temperature dependence of the dielectric constant ϵ for the sample. It is clear from this graph that ϵ increases with temperature. The increase of ϵ with temperature can be related to the orientational polarization which is due to the presence of oxygen vacancies and interstitials in the sample. When the temperature is increased, the orientation of dipole is high which increases the value of orientational polarization. The increase in ϵ may also be due to the increase in the degree of crystallinity.^{22–24}

The dielectric loss ($\tan \delta$) shows a typical dispersion behavior of polarization with increase in temperature. According to Stevels,²⁵ the dielectric loss are a sum total of conduction losses, dipole losses, and vibrational losses. As the temperature is raised, the conduction losses increase. This leads to the increase of the value of ($\tan \delta$) with temperature. But the increase of the dielectric loss at low frequency is due to dipolar polarization and the migration of ions.²¹ The dependence of dielectric constant and

Fig. 3. (a) Frequency dependence of dielectric constant of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at different temperatures, (b) Temperature dependence of dielectric constant of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at different frequencies and (c) Frequency dependence of dielectric loss of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at different temperatures.

dielectric loss on varying frequencies (42 Hz–1 MHz) at selected temperatures are as depicted in Fig. 3(c).

3.4. Impedance spectroscopy studies

Impedance spectroscopic techniques are a convenient and efficient tool to study the charge transport behavior of polycrystalline materials.²⁶ This distinguishes the contributions of electrical properties due the grains, grain boundaries, and electrode/sample interface in a polycrystalline material. Experimental data can be studied through four possible complex formalisms such as electrical impedance Z^* , (2) electric modulus M^* , (3) admittance Y^* , and (4) dielectric permittivity ϵ^* . They represent the same information expressed in different ways. The relationship between each parameter is given below.

$M^* = jwC_0Z^* = jwC_0(1/Y^*) = (1/\epsilon^*)$ where $w = 2\pi f$ is the angular frequency, $C_0 = \epsilon_0 A/d$ vacuum capacitance of the cell without sample, ϵ_0 is the permittivity of free space, (8.85×10^{-12} F/m), and A and d are area and thickness of the sample, respectively.¹⁸ The total ceramic impedance in perovskites gets mainly affected by grain impedance and grain boundary impedance. Hence, impedance data can be presented as a basic equivalent circuit consisting of two parallel resistors- capacitor (RC) elements connected in series. If (Z', M', ϵ', Y') are the real and $(Z'', M'', \epsilon'', Y'')$ the imaginary components of the complex impedance, electrical modulus, permittivity, and admittance then

$$Z^* = Z' - jZ'', \quad M^* = M' + jM'', \quad \epsilon^* = \epsilon' - j\epsilon''$$

$\tan \delta = \epsilon''/\epsilon' = M'/M'' = -Z'/Z'' = Y''/Y'$.²¹ The complex electric modulus is defined by the reciprocal of the complex permittivity. Hence, the complex electric modulus M reflects the dynamic properties of the sample alone.

The variation of Z' and Z'' , M' and M'' and ϵ' and ϵ'' with frequency at different temperatures shows that each parameter varies with the increase in both frequency and temperature (Figs. 4(a)–4(e)). The distinct behavior becomes prominent after temperature 400°C. At higher frequencies Z' (or M') attains a maximum value ($Z'_{\max}/M'_{\max}$) known as relaxation frequency (f_r). The relaxation process occurs due to the presence of immobile charges at lower temperatures, and also due to defects and vacancies at higher temperatures.²¹

Cole–Cole Plot

The electrical transport response can also be explained using Cole–Cole/Davidson–Cole and empirical expressions. The low-frequency semi-circle can be explained to the grain boundary effect, and the higher-frequency semi-circle can be explained to the grain interior. Cole–Cole plots shape suggests homogeneity of the material when perfect semicircular curves are observed. Figure 4(f) depicts the heterogeneity of the sample synthesized. The surface characteristics and the methods of preparation might have affected the shape of Cole–Cole plots thus confirming that the electrical response is due to bulk property and the presence of grain boundary.²⁷

The combined plot of imaginary parts of normalized M'' and Z'' with frequency of the sample $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at different temperatures distinguishes whether relaxation process is due to short range or long range motion of charge carriers. The peaks ($Z'' \text{ max}/M'' \text{ max}$) shift toward higher frequency range upon increase of temperature [Figs. 4(g) and 4(h)]. The mismatch in the peak positions of Z'' and M'' confirm the relaxation processes (non-Debye type) in the ceramic sample at low temperatures.²⁸ At high temperatures Z'' and M'' peaks coincide proving the gradual inclination to

(a)

(b)

Fig. 4. (a) Frequency dependence of Z' of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at different temperatures, (b) Frequency dependence of Z'' of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at different temperatures, (c) Frequency dependence of M'' of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at different temperatures, (d) Frequency dependence of M'' of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at different temperatures, (e) Frequency dependence of complex permittivity ϵ'' of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ at different temperatures, (f) Cole-Cole plot of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$, (g) and (h) Normalized imaginary parts of electrical modulus and impedance of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ as a function of frequency and (i) The scaling behavior of M'' at different temperatures for $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$.

(c)

(d)

(e)

Fig. 4. (Continued)

(f)

(g)

(h)

Fig. 4. (Continued)

Fig. 4. (Continued)

Debye-like nature.²⁷ These curves show two apparent relaxations — the left region indicating a conduction process, while the right region indicating the localized motion of ions. The peak shifts toward higher frequency and exhibits the correlation between the motions of mobile charge carriers. The nature of modulus spectrums obtained proves the existence of hopping mechanism in the electrical conduction of the materials.²⁹

For confirming the dielectric relaxation dynamics scaling behavior of M'' is studied. Figure 4(i) shows the scaling at different temperatures. Since there is lack of overlap among the peaks, the temperature dependency and deviation from the ideal Debye behavior is evident. Thus, relaxation process is proved to be thermally activated.

3.5. ac conductivity

In order to understand the conductivity mechanism of $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$, frequency-dependent conductivity at different temperatures are studied. Figure 5 depicts the ac conductivity of the sample as a function of frequency at different temperatures.

The phenomenon of conductivity dispersion is analyzed using Jonscher's power law, $\sigma_{\text{ac}}(\omega) = \sigma_{\text{dc}} + A\omega^n$, where σ_{dc} , frequency-independent conductivity (frequency-independent plateau in the low-frequency region), “ A ” is the temperature-dependent pre-exponential factor and “ n ” the frequency exponent (has values between 0 and 1). The factor “ n ” is temperature and material dependent and but not on frequency. Three regions can be observed in the conductivity graph — (i) dispersion in the low frequency part, (ii) an intermediate plateau, and (iii) conductivity dispersion at high frequency part. The low frequency region which is generally known as hopping frequency shifts toward higher frequency side with increase of temperature.³⁰ The increase in value of σ_{ac} with frequency rise is due to the disordering of cations between

Fig. 5. Frequency dependence of the electrical conductivity for $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$.

Table 1. Temperature dependence of the frequency exponent “ n ”.

T (K)	373	473	573	673	773	873
n	0.22	0.12	0.028	0.016	0.021	0.023

neighboring sites, and due to space charge. The dispersion disappears as frequency is increased. The values of frequency exponent, “ n ” obtained are listed in Table 1.

The temperature dependence of “ n ” gives information to specify the suitable mechanism involved for ac conductivity. In most cases, “ n ” is found to be between 0.6 and 1 for ionic conducting materials.⁸ Here it is clear from the results that “ n ” decreases with rise in temperature up to 400°C and then shows a marginal increase. This can be attributed to hopping conduction of mobile charge carriers (ions) over barrier between two sites. The variation of “ n ” with the temperature corresponds to a thermally activated process.¹⁷ The conduction mechanism can be explained by the correlated barrier hopping (CBH) model till 400°C. At higher temperatures “ n ” has temperature-independent behavior which can be explained by the quantum tunneling model. Hence it can be concluded that similar charge barriers are responsible for both conduction and relaxation process in the sample $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$.

4. Conclusion

A detailed study of the transport properties of the new crystalline sample $\text{Pb}_2\text{Sr}_2\text{CaCu}_2\text{O}_9$ is done using impedance and modulus spectroscopy. Structural and morphological studies are conducted through XRD and SEM. Both the dielectric

constant and dielectric loss increases with temperature and decreases with frequency. The dielectric constant variation is due to orientation and space charge polarization. The temperature dependence of the dielectric loss contributes for the conduction loss. The variation of the real part of the dielectric constant as a function of frequency at different temperatures exhibits LFDD. Scaling of M'' plots results in mis-match in overlap of M'' curves suggesting temperature independence of the relaxation dynamics for low temperatures but gets overlapped for higher temperatures. The electrical transport properties have also been found to obey Jonscher's universal law. The behavior of exponent " n " confirmed that the conduction mechanism in the material follows CBH model for low temperatures. The results of conduction and the relaxation process in the sample are found to be in close agreement with each other establishing the involvement of similar charge barriers in both the processes.

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