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# Investigation of Dielectric Relaxation in $Se_{78-y}Te_{20}Sn_2Cd_y$ Chalcogenide Glasses

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## Abstract

Chalcogenide glasses (ChGs) have emerged as a focal point of scientific research over the past decade due to their wide-ranging applications in electronics, optics, optoelectronics, X-ray imaging, and photonics. These multifunctional materials have attracted significant attention for their potential use in amorphous semiconductor devices, such as optical DVDs, phase change memory devices for energy conservation, RAM, waveguides, optical fibers, and low-cost solar cells. ChGs exhibit unique structural and chemical disorders resulting from their lack of long-range order and inherent defects within the mobility gap. To harness these materials for practical applications, a comprehensive understanding of their electrical transport properties is essential. Conductivity in ChGs is typically characterized as p-type, stemming from the asymmetry between the conduction and valence bands. Understanding the role of structural and chemical disorders, particularly their impact on anti-bonding and non-bonding states, is crucial. Additionally, the dielectric properties of ChGs play a significant role in predicting structural information and identifying sources of electrical loss. Temperature and frequency-dependent dielectric studies offer insights into the conduction mechanism and defects in these materials.

The investigation of a.c. conductivity in ChGs reveals insights into their conduction mechanism. Various theoretical models, such as Quantum-Mechanical Tunneling, Overlapping Large Polaron Tunneling, Non-overlapping Small Polaron Tunneling, and Correlated Barrier Hopping, have been proposed to explain a.c. conduction. While binary and ternary ChG systems have been widely studied, recent research has focused on synthesizing quaternary ChGs, presenting new opportunities for tailoring their physical properties. In this study, we investigate the dielectric relaxation and thermally activated a.c. conduction in $Se_{78-y}Te_{20}Sn_2Cd_y$ ($0 \leq y \leq 4$) glasses, exploring the influence of cadmium as a fourth element in this novel ChG system.

**Keywords:** Chalcogenide, Photonics, Waveguides, Conductivity, Tunneling

## 1. Introduction

Chalcogenide glasses (ChGs) have become the subject of intense scientific exploration over the past decade, primarily due to their versatility and myriad applications across various technological domains. These materials have piqued the interest of researchers, technologists, and scientists alike, owing to their exceptional properties and potential for advancing fields such as electronics, optics, optoelectronics, X-ray imaging, and photonics. The utility of ChGs extends to the development of amorphous semiconductor devices, encompassing optical DVDs, phase change RAM, waveguides, optical fibers, and even cost-effective solar cells. However, their intrinsic disorder, characterized by a lack of long-range order (LRO) and an array of inherent defects within the mobility gap, poses significant challenges and opportunities.

Understanding the transport mechanisms in ChGs hinges on deciphering the intricate interplay of structural and chemical disorders. These disorders are thought to have a pronounced impact on anti-bonding and non-bonding states, making it crucial to assess their effects on the materials' electrical properties. This ongoing research effort seeks a comprehensive comprehension of ChGs electrical transport properties. Chalcogenide glasses typically exhibit p-type conductivity, rooted in the inherent asymmetry between their conduction and valence bands. The disparities between these bands are driven by the assumed formation of anti-bonding orbitals in the conduction band and non-bonding orbitals or lone-pair electrons in the valence band. The extent of disorder in these glasses appears to have a more significant impact on anti-bonding states than on non-bonding states, resulting in an abundance of localized tail states near the conduction band edge.

Dielectric properties of ChGs are a key focus for unveiling structural insights and identifying the origins of electrical losses. These studies distinguish between the capacitive insulating nature and the conductive nature of the materials. They also shed light on the conduction mechanisms and defects within ChGs, with temperature and frequency-dependent analyses providing critical information about the nature and source of various losses. To understand the conduction mechanisms more profoundly, researchers have investigated the frequency and temperature dependence of a.c. conductivity in ChGs. Various theoretical models have been proposed to explain a.c. conduction, with the Correlated Barrier Hopping (CBH) model, introduced by Elliot, emerging as a particularly successful framework. The CBH model, initially designed for electrical transport in amorphous materials, has proven adaptable for comprehending the electrical conduction in ChGs, especially those with highly disordered surfaces. While binary and ternary ChG systems have been extensively studied, recent research endeavors have ventured into the synthesis and exploration of quaternary ChGs. These novel materials present exciting opportunities for tailoring their physical properties to meet specific application requirements. This study delves into the dielectric relaxation and thermally activated a.c. conduction in $Se_{78-y}Te_{20}Sn_2Cd_y$ ($0 \le y \le 4$) glasses, leveraging cadmium as the fourth element in this unique ChG system.

## 2. Literature review:

The field of chalcogenide glasses (ChGs) has witnessed significant research activities in recent years, with a focus on synthesizing and characterizing a wide range of binary and ternary systems, as well as exploring the emerging trend of quaternary glasses. These investigations have aimed to uncover the unique properties and potential applications of ChGs in various technological domains. One notable study by Kaistha et al. (2015) delved into the structural and optical characterization of chalcogenide glassy alloys. This work provided valuable insights into the composition-dependent properties of ChGs, shedding light on their structural transformations and optical behavior. Yadav and Sharma (2016) conducted a comprehensive investigation into the nonlinear optical properties of amorphous thin films within the quaternary system. Sakshi and Manish dev Sharma (2020) referred to Chalcogenide glasses as usable in Phase change memory devices for energy conservation. Furthermore, they explored the temperature and frequency dependence of a.c. conduction in ChGs belonging to the Se-Te-Bi-Pb system, contributing to our understanding of these materials' electrical transport behavior. Recent findings also suggest that the addition of a fourth element can enhance the physical properties of ternary ChGs, such as Se-Te-Sn alloys, by increasing crystallization temperature and thermal stability, although it may affect material hardness adversely (Kumar et al., 2013).

In summary, the extensive literature survey underscores the continued interest in Chalcogenides, particularly in the synthesis of novel multi-component systems. These investigations have motivated the exploration of quaternary Chalcogenides and the examination of how the addition of a fourth element can further tailor their physical properties for various applications.

## 3. Sample preparation:

Multi-component glasses within the $Se_{78-y}Te_{20}Sn_2Cd_y$ $(0 \leq y \leq 4)$ glassy system were synthesized utilizing a precise and cost-effective melt-quench methodology. In this process, meticulously measured quantities of high-purity elements were weighed using a precision electronic balance with a sensitivity of around 10 grams. The desired compositions of the materials were hermetically sealed in quartz tubes, with dimensions of approximately 6 centimeters in length and an internal diameter of roughly 10 millimeters, all under a high vacuum environment of approximately 15 Torr.

Subsequently, the sealed tubes containing the samples underwent controlled heating to reach the melting point of the constituent elements, where they were held at this elevated temperature for a duration of 8-10 hours. The gradual temperature increase in the furnace was carefully regulated at a rate of 4-5 degrees celsius per minute. Rapid cooling was then achieved by immersing the samples in ice-cooled water. The amorphous nature of the synthesized glasses was assessed using X-ray diffraction (XRD) analysis. The absence of sharp, well-defined peaks and the presence of a broad hump in the XRD patterns of the as-synthesized alloys unequivocally confirmed their amorphous or non-crystalline state. Following successful quenching, the samples were extracted from the quartz tubes through tube fracture, facilitating further characterization and investigation of their properties.

## 4. Experimental Technique :

### 4.1 Powdering and Pellet Formation:

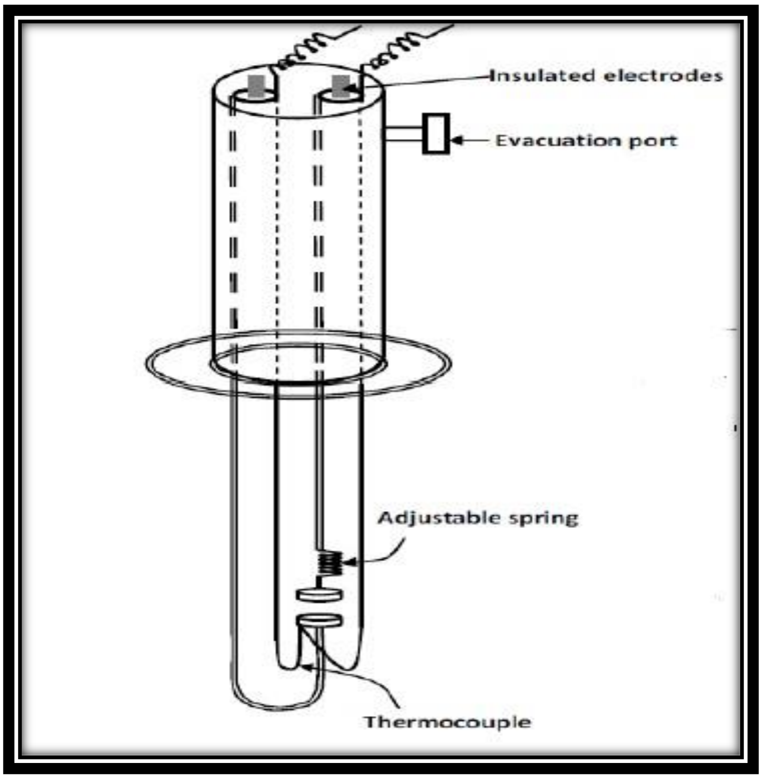
Initially, the synthesized glassy alloys were ground meticulously into a fine powder. Subsequently, pellets were created from the powdered material, with each pellet having a diameter of approximately 10 millimeters and a thickness of around 1 millimeter. The pellet formation process was facilitated using a hydraulic press.

**4.2. Consideration of Sample Form:** In the preparation of bulk samples, it is essential to be mindful of potential macroscopic effects such as the presence of gas bubbles. It has been established both theoretically and experimentally that bulk ingots and compressed pellets exhibit comparable dielectric behavior in chalcogenide glasses. This similarity arises despite suspected inhomogeneity in the case of compressed pellets within these materials. Consequently, to enhance the precision of experimental data, pellet-shaped samples with well-defined geometries were chosen for dielectric and a.c. conductivity measurements.

### 4.3 Electrode Mounting:

Each pelletized sample was carefully mounted between two steel electrodes integrated into a metallic sample holder. To ensure robust electrical contacts between the sample and the electrodes, an indium film was applied to the pellet's surface.

### 4.4. Sample Holder Configuration:



The metallic sample holder featured an evacuation port and two electrodes. These electrodes were connected to two UHF connectors for establishing electrical connections with the sample. Moreover, the outer layer of the sample holder assembly was equipped with wire serving as a heating element. Additionally, a copper-constantan thermocouple was strategically placed in close proximity to the sample to enable temperature measurements.

### 4.5 Dielectric Measurements:

The dielectric measurements were conducted under controlled vacuum conditions of approximately 13 Torr. This testing took place within a specified temperature range spanning from 300 K to 330 K, coupled with an audio frequency range from 1.2 kHz to 1.5 MHz . The dielectric measurements were executed employing a digital LCR meter, specifically the Wayne Kerr Electronics unit.

### 4.6 LCR Unit Setup:

The experimental setup for the digital LCR unit involved an electrode assembly with dual functions: first, for transmitting the applied voltage to the sample, and second, for sensing the response signals generated by the sample. The measurement of the sample's response was carried out as a function of temperature and frequency, providing valuable insights into its dielectric properties. These rigorous preparation steps and measurement techniques were essential for accurately characterizing the dielectric properties of the synthesized glassy alloys.

## 5. Results & Discussion

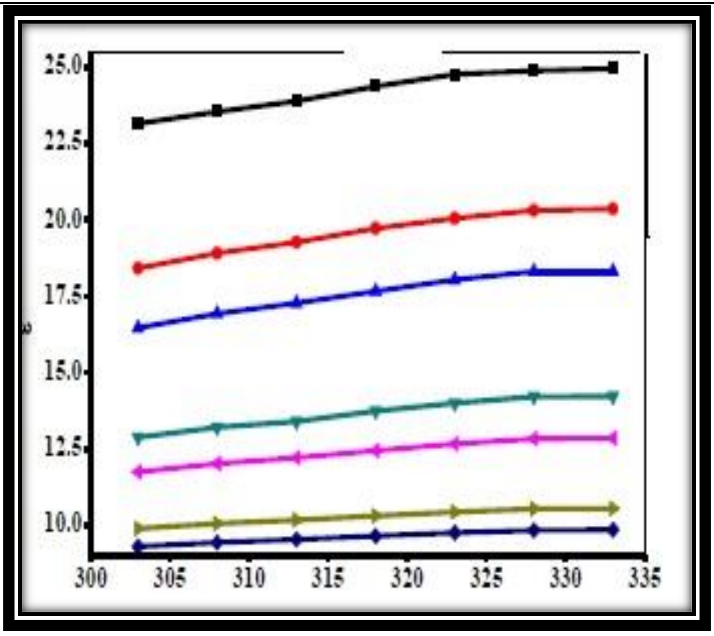
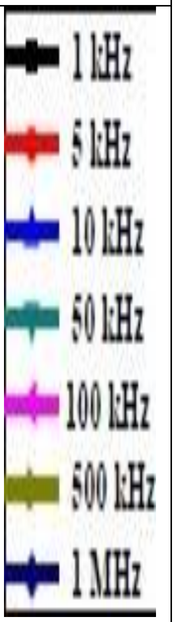
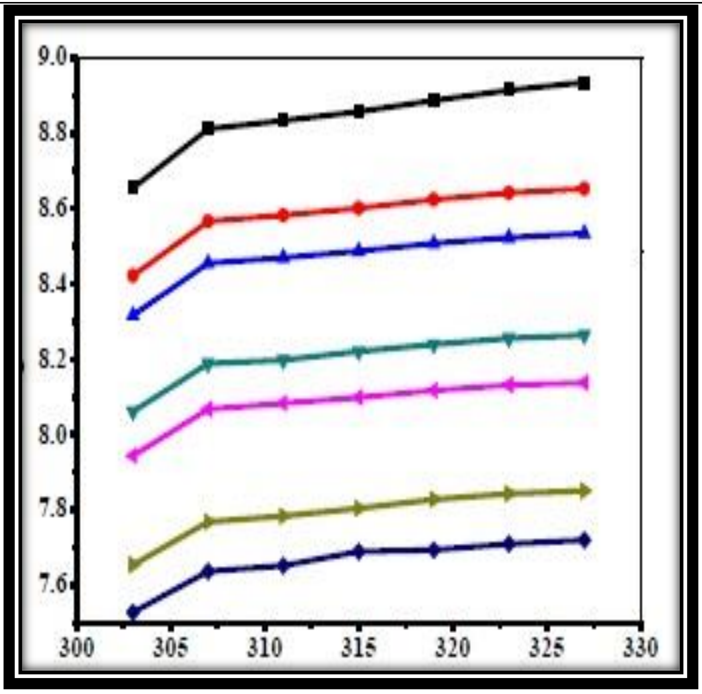
The dielectric properties of multi-component chalcogenide glassy alloys, specifically the $Se_{78-y}Te_{20}Sn_2Cd_y$ (y=0, 2, 4) glasses system, were comprehensively investigated below the glass transition temperature. Table 1 and 2 illustrate the temperature dependence of the dielectric constant ($\varepsilon'$) and dielectric loss ($\varepsilon''$) at various frequencies, respectively. The observations provide valuable insights into the dielectric behavior of these alloys.

### 5.1 Temperature Dependence of Dielectric Constant ($\varepsilon'$)

The temperature-dependent variations of dielectric constant ($\varepsilon'$) reveal intriguing patterns. Initially, for glassy $Se_{78}Te_{20}Sn_2$ and $Se_{76}Te_{20}Sn_2Cd_2$ alloys, there is an increase in dielectric constant with rising temperature. This increase is, however, frequency-dependent, exhibiting distinct trends at different audio frequencies. Consequently, the orientation polarization increases, leading to a rise in the dielectric constant ($\varepsilon'$).

### 5.2 Temperature Dependence of Dielectric Loss ($\varepsilon''$)

The dielectric relaxation phenomenon in these multi-component chalcogenide glasses involves three distinct components: i) Dipole Loss relates the relaxation of dipoles within the material. ii) Conduction losses are associated with the migration of ions over relatively long distances within the glassy network. As ions move, they transfer some energy to the lattice in the form of heat. The amount of heat dissipated per cycle is directly proportional to the product $\sigma ac\ (\omega)/\omega$. Consequently, with increasing temperature, $\sigma ac\ (\omega)/\omega$ rises, resulting in an increase in a.c. conduction loss. iii) Vibrational losses occur as a result of lattice vibrations within the material.

| | **Name of Glassy Alloy** | **Graph obtained for variation of dielectric constant with temperature** | **Frequency** |
|---|---|---|---|
| 1 | $Se_{78-y}Te_{20}Sn_2Cd_y$ (for y = 0 ) glasses |  |  |
| 2 | $Se_{78-y}Te_{20}Sn_2Cd_y$ (for y = 2 ) glasses |  |  |

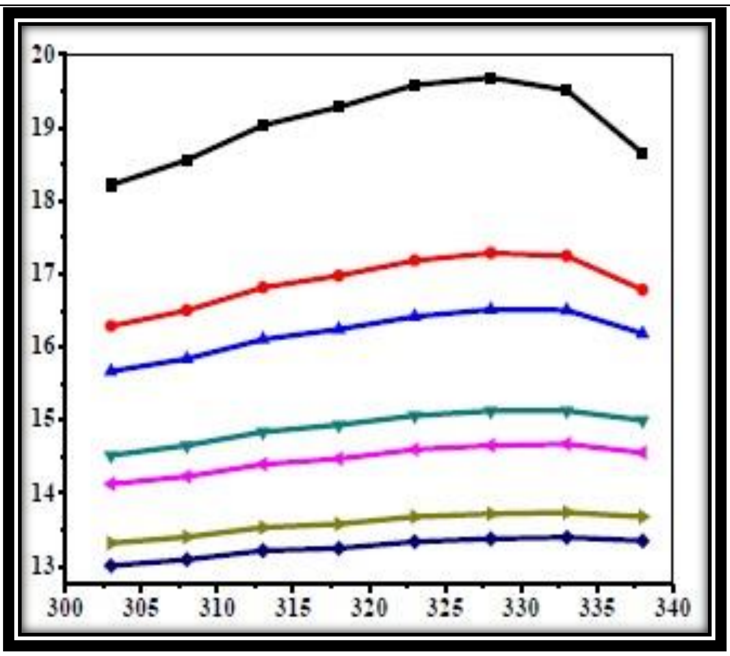
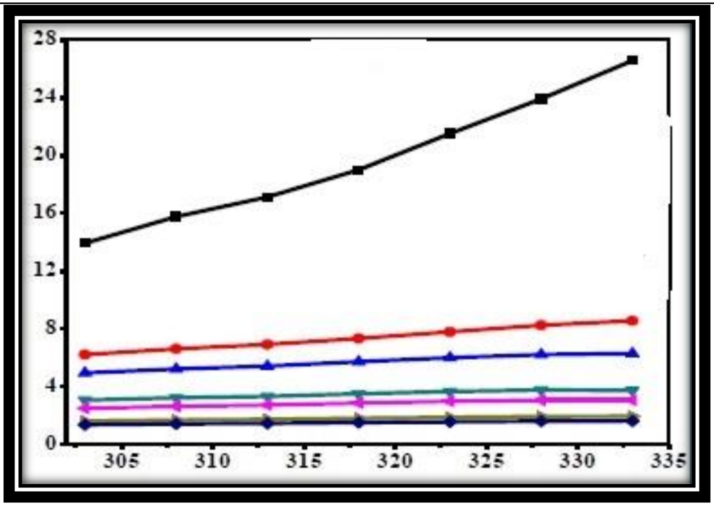
| | | | |
|---|---|---|---|
| 3 | $Se_{78-y}Te_{20}Sn_2Cd_y$ (for y = 4 ) glasses |  | 1 kHz<br>5 kHz<br>10 kHz<br>50 kHz<br>100 kHz<br>500 kHz<br>1 MHz |

**Table 1.0: Showing variation of dielectric constant with temperature**

At lower temperatures, the dielectric loss shows minimum values. This can be attributed to the relatively low values of conduction, dipole, and vibration losses under these conditions. However, at higher temperatures, all three types of losses, namely conduction, dipole, and vibration losses, contribute significantly to the dielectric loss. This collective effect leads to an increase in dielectric loss with rising temperature.

| | **Name of Glassy Alloy** | **Graph obtained for variation of dielectric loss with temperature** | **Frequency** |
|---|---|---|---|
| 1 | $Se_{78-y}Te_{20}Sn_2Cd_y$ (for y = 0 ) glasses |  | 1 kHz<br>5 kHz<br>10 kHz<br>50 kHz<br>100 kHz<br>500 kHz<br>1 MHz |

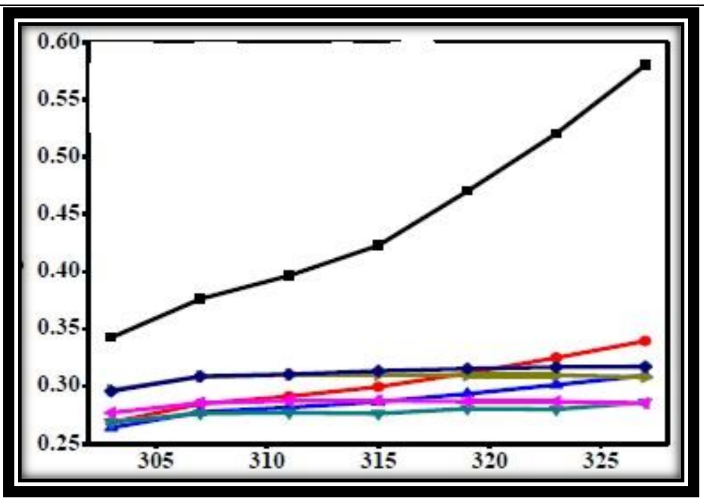
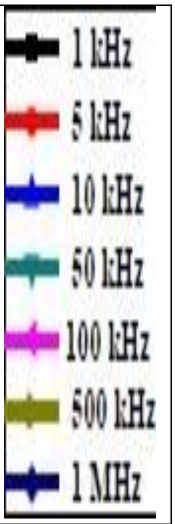
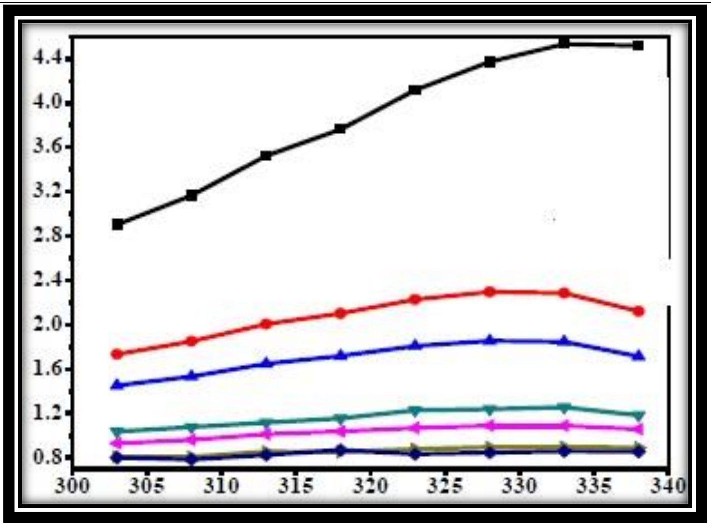
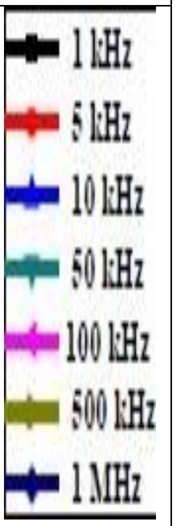
| | | | |
|---|---|---|---|
| 2 | $Se_{78-y}Te_{20}Sn_2Cd_y$ (for y = 2 ) glasses |  |  |
| 3 | $Se_{78-y}Te_{20}Sn_2Cd_y$ (for y = 4 ) glasses |  |  |

**Table 2.0 Showing variation of dielectric loss with temperature**

## 6 Conclusion:

Above shown Table 1 and 2 reveal a noteworthy phenomenon: the emergence of peaks in the temperature-dependent plots of both $\varepsilon'$ and $\varepsilon''$ for each frequency as the concentration of Cd increases. These observations align with the theory of dipolar relaxation. The presence of peaks indicates specific temperature regimes where the dielectric properties exhibit distinctive behavior. This phenomenon could be attributed to the interplay of various relaxation processes, emphasizing the complex nature of dielectric behavior in these multi-component chalcogenide glasses. In summary, the investigation of dielectric properties in the $Se_{78-y}Te_{20}Sn_2Cd_y$ glassy alloys underscores the interplay of dipole, conduction, and vibrational losses, leading to temperature-dependent variations in dielectric constant and dielectric loss. The emergence of peaks in the presence of higher Cd concentrations highlights the complex nature of dielectric relaxation and underscores the significance of multi-component alloy composition in shaping the dielectric behavior of chalcogenide glasses.

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