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Dielectric relaxation spectroscopy of phlogopite mica

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ABSTRACT

An in-depth investigation of the dielectric characteristics of annealed phlogopite mica has been conducted in the frequency range 0.1 Hz–10 MHz and over the temperature range 653–873 K through the framework of dielectric permittivity, electric modulus and conductivity formalisms. These formalisms show qualitative similarities in relaxation processes. The frequency dependence of the M'' and *dc* conductivity is found to obey an Arrhenius law and the activation energy of the phlogopite mica calculated both from *dc* conductivity and the modulus spectrum is similar, indicating that same type of charge carriers are involved in the relaxation phenomena. The electric modulus and conductivity data have been fitted with the Havriliak–Negami function. Scaling of M', M'', *ac* conductivity has also been performed in order to obtain insight into the relaxation mechanisms. The scaling behaviour indicates that the relaxation describes the same mechanism at different temperatures. The relaxation mechanism was also examined using the Cole–Cole approach. The study elaborates that the investigation regarding the temperature and frequency dependence of dielectric relaxation in the phlogopite mica will be helpful for various cutting edge applications of this material in electrical engineering.

1. Introduction

Dielectric materials have focused the interest of many research groups because of their technologic applications that mainly concern their dielectric characteristics. There are several kinds of dielectric materials, with distinct properties, but mica is in general very stable and has tremendous applications in science and technology. It has a distinct layered structure, and possible to cleave into very thin, optically flat sheets [1,2]. Unique properties (low dielectric loss, best for mica capacitors, perfect cleavage, high flexibility, good physico-chemical stability at high voltages and temperature etc.) of mica, makes it an appropriate material for various cutting edge applications in nanotechnology, material science, electrical engineering, high temperature and advanced power electronics, spacecraft, supersonic aircraft, dosimetry and radiation research [1–12]. There are many types of mica, but muscovite and phlogopite are cheapest, easily available and have better thermal and dielectric properties which make them useful for various technological applications.

Mica (monoclinic structure) is phyllosilicate, with unit structure consists of one octahedral sheet between two opposing tetrahedral sheets [1,2,13]. It crystallizes in a layered structure and cleaves easily into thin translucent sheets. These sheets form a layer that is separated from adjacent layers (Tetrahedral and Octahedral) by planes of non-hydrated interlayer cations (e.g., Cs, K, Na, NH_4, Rb, Ba and Ca).

The simplified formula of mica can be written as

$$IX_{2-3}\Delta_{1-10}T_4O_{10}A_2$$

where, I are the interlayer cations, X are the octahedrally coordinated cations (Li, Fe, Mg, Mn, Zn, Al, Cr, V and Ti), Δ represents the vacancy, T is commonly Be, Al, B, Fe and Si, A is commonly Cl, F, OH, O and S.

There are three major divisions within the mica: (a) the true micas, (b) the brittle micas and (c) the interlayer-deficient micas. These three divisions are further divided into dioctahedral (muscovite, boromuscovite, etc.) and trioctahedral groups (e.g. biotite and phlogopite). Phlogopite [$K(Mg)_3AlSi_3O_{10}(F, OH)_2$] is a yellow, greenish or reddish brown category of the mica.

Although the dielectric characteristics of various materials have been studied by different authors [14–29], it is important to obtain dielectric information about mica. Apart from few works on electrical and dielectric properties of mica [30–32], we could not find more detailed data in particular phlogopite mica. Today world of electrical engineering has continuously encouraged the development of new and interesting materials in fascinating and real applications. The investigation of the dielectric properties of mica is interesting both from the fundamental and technological point of view. This appeared to us to be important because

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frequency and temperature dependent dielectric phenomena in phlogopite, remains poorly understood and more information is required to form a clear picture. In the present investigation, the dielectric characteristics of annealed phlogopite mica have been conducted in the frequency range 0.1 Hz–10 MHz and the temperature range of 653–873 K using the dielectric relaxation spectroscopy. This type of investigation will provide information for various industrial applications and as well as the better understanding of the temperature response of phlogopite mica.

2. Experimental details

In the present investigation, Phlogopite mica sheets ($\approx$200 μm thickness) were procured from Shree GR Exports Private Limited, Kolkata, India. The Phlogopite mica sheets were annealed at different temperatures (473–873 K) for 2 h in a muffle furnace. Dielectric constant (ε') and dielectric loss (ε'') have been measured for heated phlogopite mica sheets in the frequency range (0.1 Hz–10 MHz). It has been found that at low frequency region, the dielectric constant and dielectric loss increases with increasing the annealing temperature and show maxima near 673 K. This annealed (at 673 K) phlogopite mica was preferred for further detailed dielectric characterization. All the dielectric characterization was performed using NOVO-CONTROL (Alpha-A) High Performance Frequency Analyzer in the frequency range of 0.1 Hz–10 MHz (installed at UGC-DAE consortium for scientific research, Indore, India). The theoretical dielectric analyses of the measured data were carried out using the Winfit software from NovoControl. Employing the dielectric modulus formalism ($M^*=1/\varepsilon^*$), the data was analyzed and fitted with Havriliak–Negami function [33] with conductivity term given as below

$$\varepsilon^*(\omega)=\varepsilon'-i\varepsilon''=-i\left(\frac{\sigma_{dc}}{\varepsilon_0\omega}\right)^n+\left\{\frac{\Delta\varepsilon}{\left[1+(i\omega\tau)^{\alpha}\right]^{\beta}}+\varepsilon_\infty\right\} \quad (1)$$

where, ε_0 and ω are the vacuum permittivity and angular frequency respectively. τ is the characteristic relaxation time, ε_∞ gives the value of ε' at infinite frequency and $\Delta\varepsilon$ (relaxation strength) represents the difference between ε' at zero and infinite frequency (ε_∞) and is also proportional to the area below the relaxation peak and maximum value of ε''. In the above equation, σ_{dc} represents *dc* conductivity and n gives the exponent of the frequency dependent ε''. The exponent α (symmetric broadening parameter) gives the broadness and specifies the slope of the low frequency side of relaxation in ε''. Here β (asymmetric broadening parameter) is the asymmetry of the spectra and the value of $-\alpha\beta$ gives the slope of the high frequency side of the ε'' relaxation.

3. Results and discussion

Dielectric spectroscopy is based on the interaction of an applied electric field with the material under investigation. Dielectric permittivity ($\varepsilon^*=\varepsilon'-i\varepsilon''$), which is the material's response to the applied field, is frequency and temperature dependent function, provides information regarding the properties of the material. Dielectric permittivity (ε^*) of the material originates from the space charge polarization produced within the material. Its real part (ε') represents the polarizability, and imaginary part (ε'') represents the energy losses due to the polarization and ionic conduction. Each of the polarization mechanism prevail a certain frequency range with characteristics relaxation frequency.

Dielectric constant (ε') and dielectric loss (ε'') of annealed phlogopite samples in the frequency range (0.1 Hz–10 MHz) are presented in Figs. 1 and 2 respectively. Fig. 1 shows that in

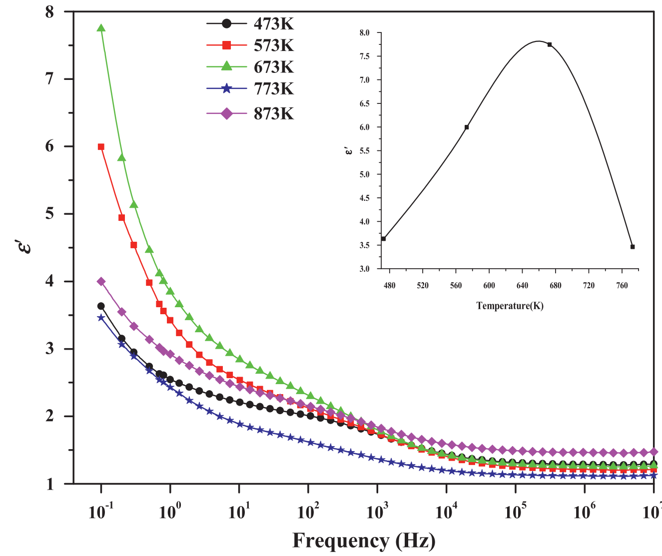

Fig. 1. Frequency dependence of dielectric constant (ε') of phlogopite mica at different annealing temperatures.

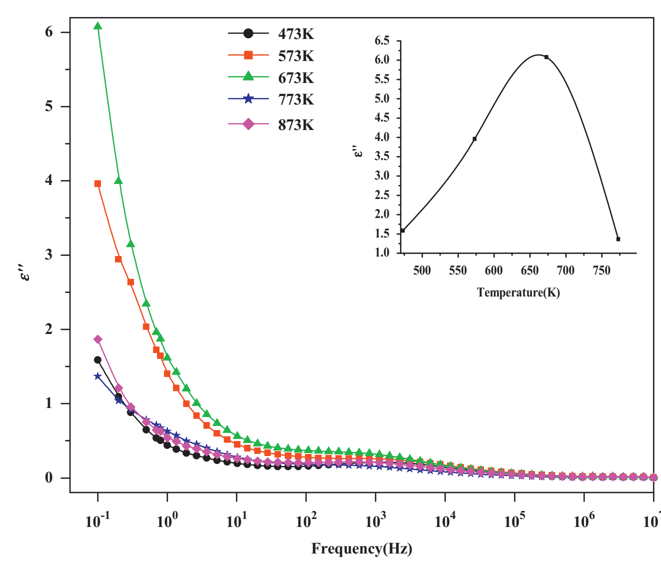

Fig. 2. Frequency dependence of dielectric loss (ε'') of phlogopite mica at different annealing temperatures.

low frequency range (0.1 Hz–1 KHz), the dielectric constant (ε') increases as the annealing temperature increases up to 673 K and then decreases at high temperatures. This increase in dielectric constant (ε') with temperature is due to the reduction of the dipole–dipole interactions. Because of the dielectric dispersion, the ε' gradually decreases as the frequency increases because the dipoles can no longer follow the field at high frequencies. Similar trend is observed for dielectric loss (ε'') at varying frequency and temperature (Fig. 2). The instability of mica near 673 K temperature [34] enhanced its dielectric properties. Therefore, the phlogopite samples annealed at 673 K for 2 h were preferred to study concisely its dielectric properties as a function of frequency and temperature [30].

The frequency dependence of real (ε') and imaginary (ε'') parts of complex dielectric permittivity (ε^*) of phlogopite mica at selected temperature range (653–873 K) are presented in Figs. 3 and 4 respectively. Significant difference for different temperatures was observed at low frequencies. Figs. 3 and 4 show that the dielectric constant (ε') and the dielectric loss