Dielectric response of humid phlogopite*

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Abstract. The dielectric response of phlogopite mica has been measured for frequency lying between 10 mHz and 10 kHz with humidity as a parameter. The measured complex capacitance shows a low frequency dispersion (LFD) at high humidities. A weak loss peak is also present at high frequencies and low humidity levels.

Keywords. Dielectric response; phlogopite mica; amber mica; complex capacitance; humidity effects.

1. Introduction

Aluminium silicate is an integral component of all micas. However, a particular kind is distinguished from another on the basis of its metallic component. For example, muscovite or ruby mica is potassium silicate, $\text{H}_2\text{KAl}_3(\text{SiO}_4)_3$, and phlogopite or amber mica is a combination of magnesium and aluminium silicate, $\text{H}_2\text{K}(\text{Mg}, \text{F})_3\text{Mg}_3\text{Al}(\text{SiO}_4)_3$. All micas have a layered structure and hence show anisotropic behaviour (Chaudhry *et al* 1985). For instance, the thermal conductivity of phlogopite mica along the cleavage planes is higher by one order of magnitude than that perpendicular to the planes (Goldsmid and Bowley 1960; Gray and Uher 1977). Similarly other properties such as electrical conductivity and relative permittivity are also anisotropic in nature.

It has been observed that the dielectric response of mica is influenced by the presence of moisture in its surroundings (Hartshorn and Rushton 1931; Mahanti *et al* 1945). However, work carried out in the past is limited in the sense that it deals with the measurements of dielectric properties of mica as a function of relative humidity at a single frequency.

The present work on phlogopite mica is concerned with effects of humidity over 6 decades of frequency lying between 10 mHz and 10 kHz. These measurements give far more information about the dielectric sample under examination as compared to the data recorded at a single frequency (Jonscher 1983).

2. Experimental

The samples measuring $20 \times 20 \text{ mm}^2$ were directly cleaved from phlogopite sheets and were clamped between spring-loaded copper foil electrodes ($10 \times 5 \text{ mm}^2$). The procedure for making measurements and controlling the relative humidity has already been described in detail (Chaudhry and Jonscher 1985).

*Work carried out at former Chelsea College, London University, London.

3. Results and discussions

The results of measurements are presented as a function of frequency in figures 1a and b on a double log scale with relative humidity as a parameter. The same data are then normalized (Hill 1971; Jonscher 1983) to obtain a master curve (figure 2) which can be interpreted with greater assurance since all the information scattered in different curves corresponding to various relative humidities has been collected in the single master curve. This technique is based on the principle of time-temperature superposition and extends the frequency range by several decades. This also provides a direct measure of frequency shift with relative humidity of a datum point chosen arbitrarily during the normalization. It may be noted that the locus of the datum point, indicated at the bottom of figure 2, follows a straight line. To check the humidity effects at a glance, the frequency shift of the datum point is plotted against relative humidity in figure 3, and is obviously not a straight line. This means that the humidity response, in case of phlogopite mica, is not an exponential phenomena as observed previously for ruby mica paper (Chaudhry and Jonscher 1985). This difference could be related either to the nature or to the composition of the two samples. Phlogopite samples measured in the present work are crystalline in nature whereas mica paper is amorphous because it is made of finely flaked ruby mica pressed together into cohesive sheets without any binders (Ketterer 1964). Moreover, the composition of the two samples is also different as phlogopite is an aluminium silicate of magnesium while ruby mica contains potassium in addition to aluminium silicate. At this stage it is difficult to suggest which factor controls the dielectric behaviour of amber. For a conclusive result it would be necessary to study the dielectric response of crystalline and amorphous samples of each phlogopite and ruby mica. Since these were not available during the present measurements, this aspect is left for future work.

Figure 1. Effect of humidity on the dielectric response of phlogopite measured perpendicular to the cleavage planes. Each successive set in (a) and (b) is displaced vertically by one decade for clarity. The scale is drawn with respect to the lowest set in (a) and (b) which represent capacitance and loss data respectively. Two lines with -1 slope are drawn at corresponding positions for comparison of the experimental data.

Figure 2. The data of figure 1 is normalized with respect to 97% RH at 0.1 Hz. The ratio of the two lines with 0.94 slope, drawn through the real and imaginary components of the complex capacitance, is 10.58. A weak loss peak exists at higher frequency end.

Figure 3. A plot of the frequency shift of a datum point chosen arbitrarily for normalization (figure 2) against relative humidity.

It can be seen from figures 1a and b that the curves corresponding to vacuum and 23% relative humidity (RH) are similar in shape. It should be noted right in the beginning that both the capacitance and loss curves indicate a dispersion at low frequency end in addition to a small peak of loss around 1 kHz as depicted in figure 1b. As the humidity percentage grows, so does the low frequency dispersion (LFD), but the peak loss is not much affected except for a small reduction in its

Figure 4. Effect of d.c. bias at 97% RH. (a) capacitance data and (b) loss data.

amplitude. However, beyond 85% RH, the peak loss seems to disappear and LFD becomes highly predominant. It appears as if the loss peak does not move from its position yet it disappears probably owing to a strong LFD at high humidities. Phlogopite is known to have a higher water content than muscovite mica (Metsik 1963). This water is found in the interlaminal space or as the water of crystallization and thus affects the dielectric properties (Metsik 1963; Gudkov and Metsik 1973). This water is termed as bound water and is probably responsible for the high frequency loss peak observed in the present measurements. As the amount of bound water in a sample cannot be influenced by increasing the humidity in its surroundings, the loss peak should also be unaffected by any change of humidity in its neighbourhood as is the case under discussion. However, increasing the amount of relative humidity could enhance the LFD by providing extended paths to the charge carriers (Chaudhry *et al* 1984).

A sample is said to behave linearly if its dielectric response remains intact with varying fields. This was checked by applying 10 V d.c. bias in addition to the 1 V r.m.s. signal. The results of this experiment are shown in figure 4. Below 1 Hz it can be seen that both the capacitance and the loss decrease with the application of the bias. This means that the response is nonlinear in the applied bias (Jonscher *et al* 1988). At low frequencies and longer times the charges swept to one side but the d.c. bias are unable to return and thus cannot contribute to capacitance or conductance thereby decreasing both the capacitance and the loss components (Chaudhry *et al* 1984). On the other hand, at high frequencies the charges carried away by the d.c. bias can come back and enhance the conductance. However, the charge buildup cannot take place owing to the short durations corresponding to high frequencies and hence the value of capacitance remains unchanged at the high frequency side of the data reported here.

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