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Dielectric response of wet muscovite measured along the cleavage planes

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Dielectric response of muscovite mica is measured as a function of frequency and humidity. The frequencies are varied from 10 mHz–10 kHz and the relative humidity (RH) of the atmosphere surrounding the samples changes from 0%–97%. The results of measurements show the presence of a low-frequency dispersion and a loss peak which could be related to the charge carriers either moving across limited distances or hopping between two sites.

1. Introduction

Mica is a useful mineral which finds many applications in the electrical and electronic industry. The technical importance of mica, therefore, demands for its thorough study under various conditions to meet desired specifications. Recently, the high-temperature dielectric behaviour of ruby mica has been investigated perpendicular to the cleavage planes [1]. Low-temperature dielectric measurements on ruby mica were also performed [2] parallel to the cleavage planes down to 173 K, in addition to an analysis of its contact and surface impedances with humidity as a parameter [3]. The transient dielectric response of ruby mica along the cleavage planes has also been reported by Jonscher and Ramdeen [4]. This communication is in line with our previous work and is related to the study of ruby mica under various humidity levels. These measurements were carried out over a wide frequency interval lying between 0.01 and 10 000 Hz. A literature survey conducted as far back as the beginning of this century found no data similar to those reported here.

2. Experimental procedure

It is quite easy to prepare a mica sample for investigations normal to the planes compared to along the planes, which poses a serious problem because of its layered structure. For parallel to the planes measurements, samples of ruby mica were obtained with the help of a simple technique described earlier [2], in which a large number of mica strips, each measuring 40×4 mm^2, with varying thicknesses, were cut from a single sheet and stacked together in a steel clamp. Two opposite faces were polished before applying copper foil electrodes or evaporating on them aluminium electrodes which were 30×10 mm^2 in size.

The dielectric measurements were performed using a frequency response analyser (FRA) system [5, 6] over a frequency range of 10 mHz–10 kHz. The method of obtaining various humidity levels has been described in a previous paper [1].

3. Results and discussion

The results of measurements corresponding to three types of electrode, namely copper foil, evaporated aluminium with copper foil and evaporated aluminium with silver paint, are presented in Figs 1a, b–3a, b as logarithm of real and imaginary components of complex capacitance against logarithm of frequency with relative humidity as a parameter. The same data are then normalized [7] into master curves shown in Figs 1c–3c. The presentation of dielectric data in this form relies on the fact that the complex capacitance can be expressed in terms of its real and imaginary components as

$$\begin{aligned} C(\omega) &= C'(\omega) - \mathrm{i}C''(\omega) \\ &= C'(\omega) - \mathrm{I}G(\omega)/\omega \end{aligned} \tag{1}$$

where $G(\omega)$ is the frequency-dependent a.c. conductance. The two components can be related to each other through the following relationship

$$\begin{aligned} C''(\omega)/[C'(\omega) - C_\infty] &= \cot n\pi/2 \\ &= A\,\omega^{n-1} \end{aligned} \tag{2}$$

where A is a constant. The high-frequency capacitance, C_∞, corresponds to the processes which are faster than those effective in a given frequency range available in a particular experiment. The exponent, n, can have any value between 0 and 1. The relationship is known [8] as the "universal power law" and is found to be valid for both the volume and the surface response of dielectrics [3].

Two solid lines with -1 slope are drawn at corresponding positions in Figs 1a and b–3a and b to compare how the two components of the complex capacitance behave under varied humidity levels. Both

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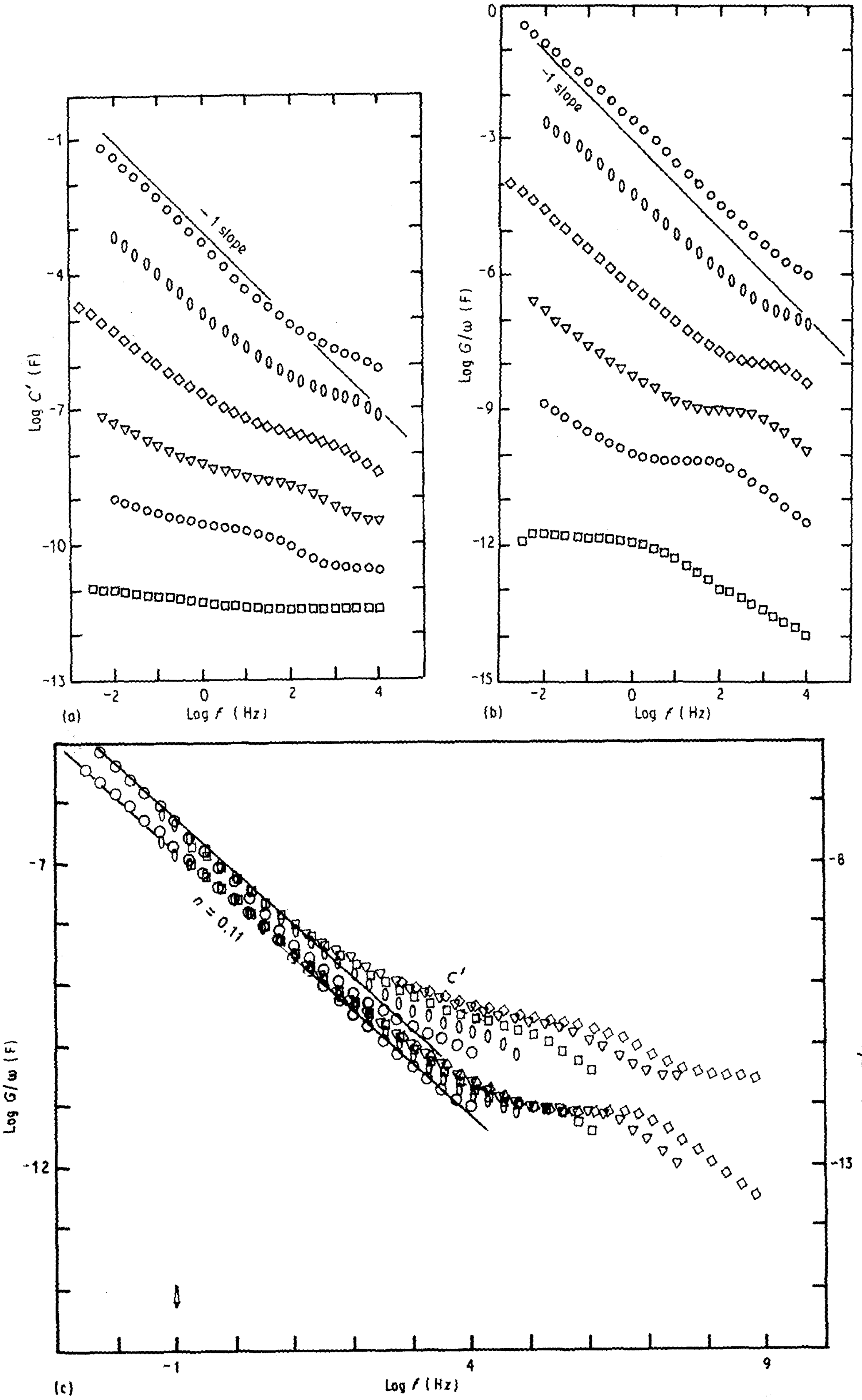

Figure 1 Effect of humidity on dielectric response of ruby mica measured parallel to the cleavage planes. A pair of copper foils pressed against two faces of the sample form the electrode system. A − 1 slope is drawn at corresponding positions in (a) capacitance, and (b) dielectric loss. The normalized plot is shown in (c). Data is scaled at 97% RH and 0.1 Hz. The two solid lines with − 0.89 slope are drawn at Kramers–Kronig compatible positions in the LFD region. A loss peak can be seen at higher frequencies. The lower part of the graph shows the shift of the datum point along the frequency axis. Fig. 1a–c (-○-) 97% RH; (0) 85% RH; (◇) 66% RH; (▽) 43% RH; (○) 23% RH; (□) vac

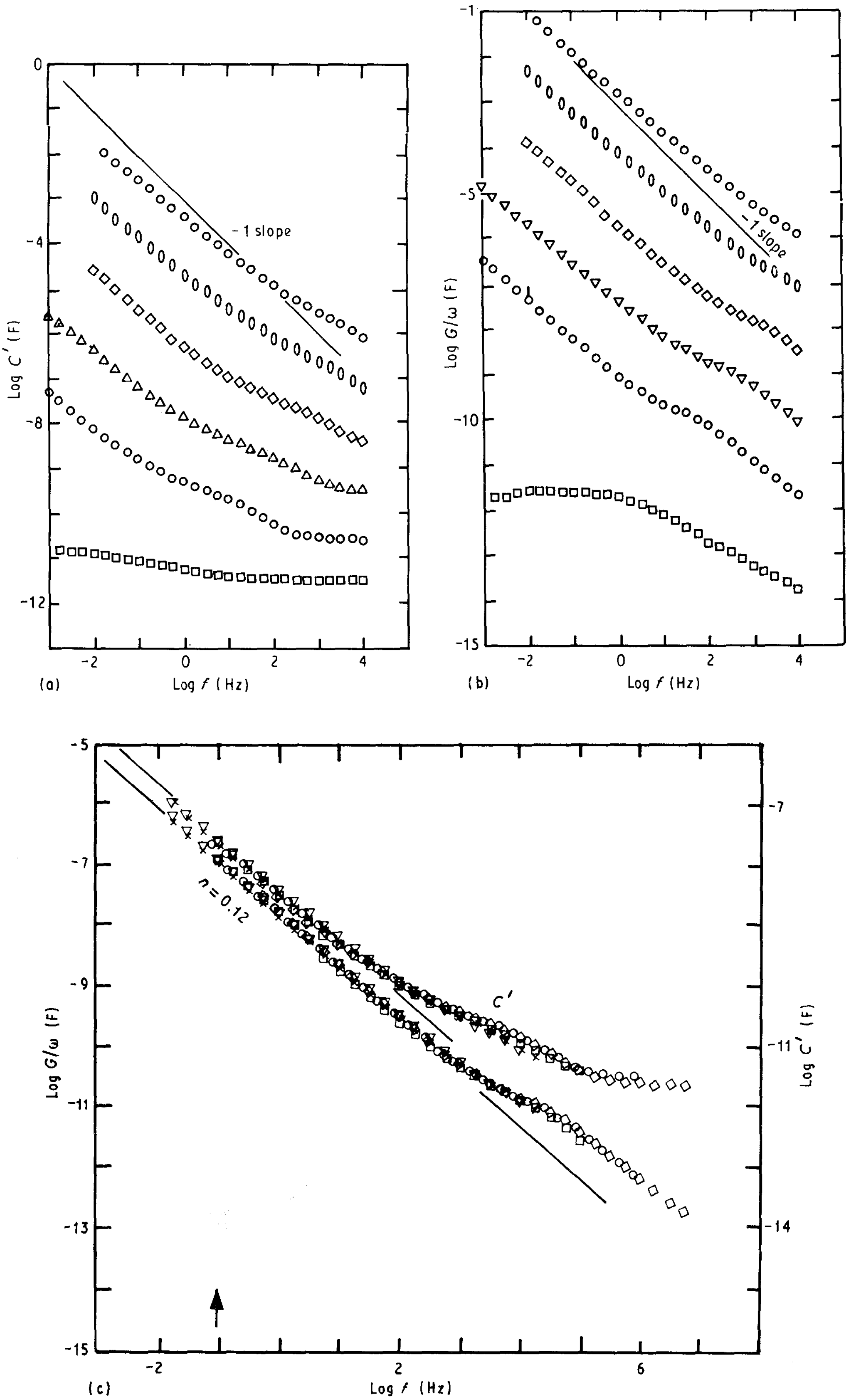

Figure 2 Dielectric response data for ruby mica measured parallel to planes with relative humidity as a parameter. Evaporated aluminium electrodes are backed by copper foil to make electrical connections. A loss peak can be seen at about 3.16 kHz (relative frequency) together with LFD in normalized plot. (a), (b): (○) 97% RH; (0) 85% RH; (◇) 66% RH; (△) 43% RH; (○) 23% RH; (□) vac. (c): (▽) 97% RH; (×) 85% RH; (□) 66% RH; (○) 43% RH; (◇) 23% RH.

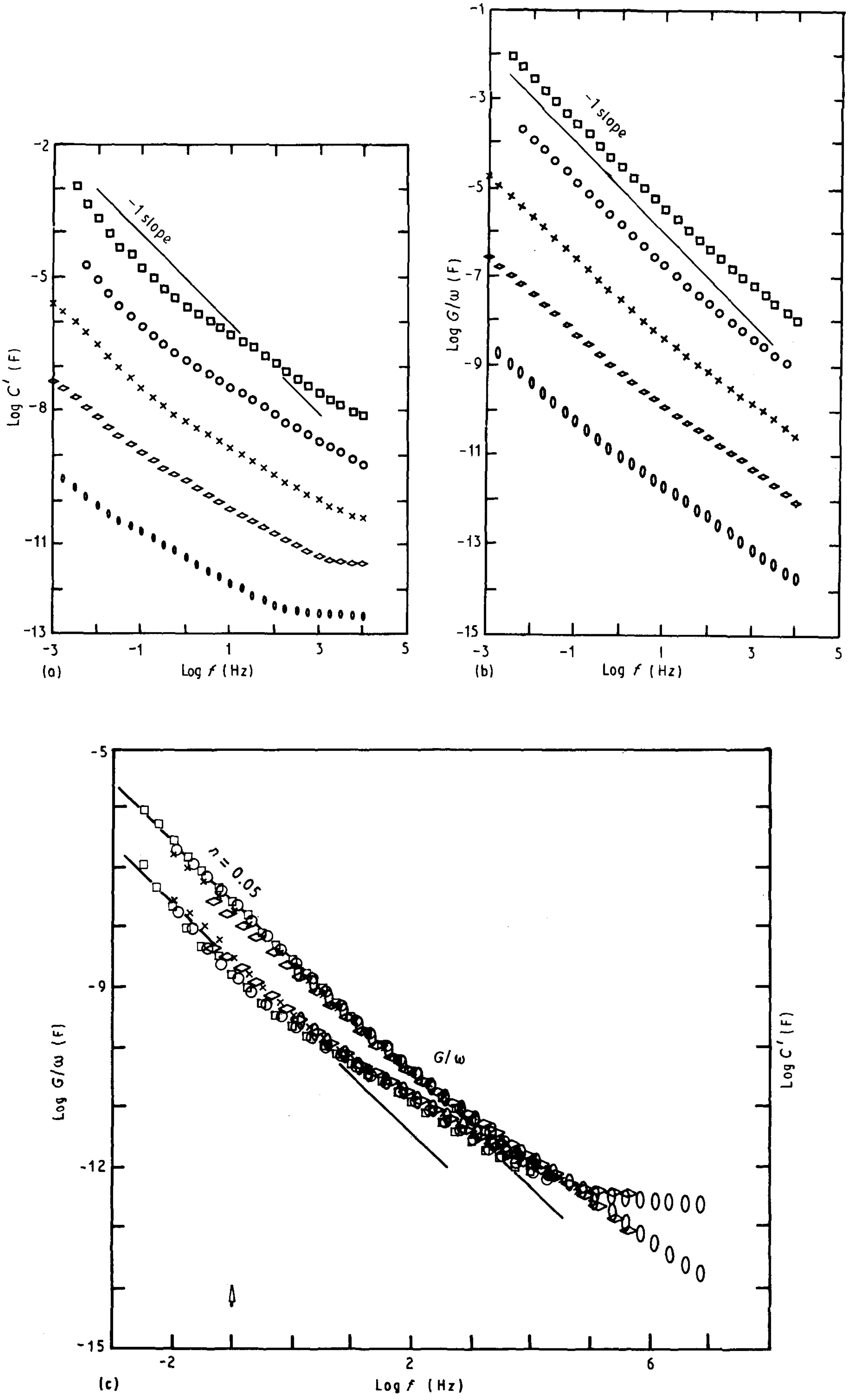

Figure 3 Dielectric response spectrum for ruby mica. Silver paint was used to stick electrical leads to evaporated aluminium electrodes. A shallow loss peak exists around 31.6 Hz on relative frequency scale in addition to LFD. (□) 97% RH; (○) 85% RH; (×) 66% RH; (◇) 43% RH; (0) 23% RH.

the dielectric loss and the capacitance increase with humidity, indicating the ease with which moisture can enter the mica parallel to the cleavage planes, compared to the direction normal to the planes.

The data corresponding to copper foil electrodes have a clearly visible loss peak in vacuum at 1 Hz in Fig. 1. This peak moved towards high frequencies with increasing relative humidity. A low-frequency dispersion (LFD) sets in at 23% RH and increases in magnitude with increasing RH. It can be seen that a smooth master curve could not be achieved on the normalization of data corresponding to copper foil electrodes; however, too much importance must not be placed on this misfit of data in the normalized curve shown in Fig. 1c because it simply means that the exponent n does not change smoothly between various levels of relative humidity.

A similar behaviour can be observed in Fig. 2a and b for evaporated aluminium with copper foil contact electrodes. In the case of evaporated aluminium backed by silver paint contacts, the loss peak is weak but the LFD is distinctive as depicted in Fig. 3a and b. An examination of all the normalized plots shown in the present work indicates that the real and imaginary components of complex capacitance run parallel to each other with a common slope of $n - 1$. There exists a constant ratio between $C'(\omega)$ and $C''(\omega)$ as required by the Universal power law, thus indicating Kramers–Kronig compatibility of two components of the complex capacitance. The value of exponent n, together with the corresponding constant ratio, for each set of electrodes, is given in Table I.

The existence of a constant ratio between imaginary and real parts of the capacitance implies that both components have the same frequency dependence, which is completely different from the Debye response where this ratio is equal to the product of angular frequency and relaxation time, i.e. ωt. In terms of energy, it means that the ratio of energy lost per cycle to the energy stored in the system is constant. Thus the larger the value of $\cot(n\pi/2)$ the greater is the loss of energy in the system.

A close look at the normalized plots reveals that the LFD is followed by a peak whose high-frequency side has different slopes for different electrode systems. The magnitude of the loss peak is maximum for copper-foil electrodes and minimum for evaporated aluminium with silver paint electrodes. Low-frequency dispersion processes are normally related to the movement of charge carriers over limited distances in such a way that a large amount of charge accumulates in the system and depicts itself as a large dispersion in the capacitance. This is entirely different from the direct conduction processes where charge carriers move from one electrode to the other without piling up the charges in the dielectric under investigation, and thus no dispersion appears in the real part whereas the loss component varies as ω^{-1}, ω being the angular frequency.

TABLE I Value of exponent n with ratio

Electrode system	n	$\cot n\pi/2$
Copper foil	0.11	5.7
Evaporated aluminium with copper foil	0.12	5.2
Evaporated aluminium with silver paint	0.05	12.7

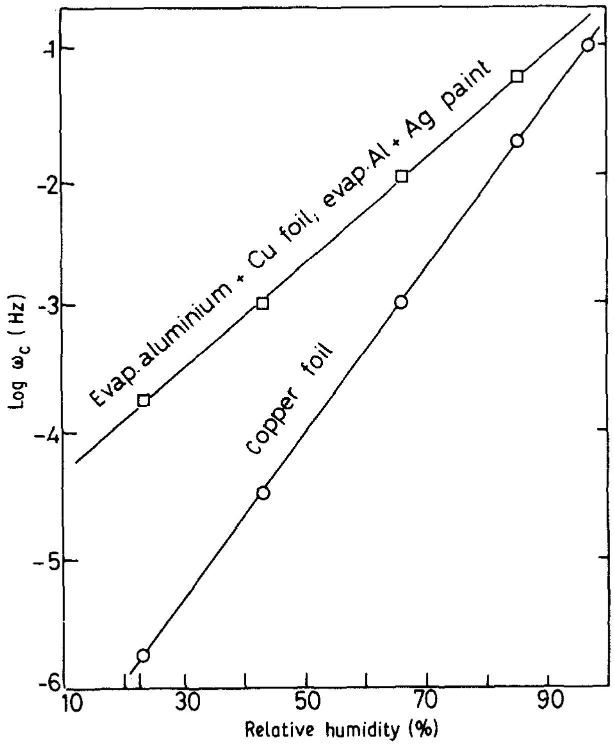

Figure 4 Frequency shift of datum point chosen arbitrarily for normalization (Figs 1–3) against relative humidity.

The loss peaks usually arise either due to dipoles or due to a charge hopping between two sites. In the present study, the samples involved are of an inorganic nature and thus have the least possibility for dipoles, except those arising from the water of crystallization which is invariably present in all varieties of mica. The magnitude and the position of the loss peak depends on the electrode material. If the observed loss peak is to be related to the water of crystallization, then its position should not be affected by the electrode material [9]. Because the experimental evidence is against this assumption, it suggests the possibility of another mechanism. In view of the inorganic nature of mica, the presence of impurities cannot be ruled out. It may be the impurity carriers which hop between two sites to develop a loss peak which changes its position and magnitude with a change in the material of the electrodes. This approach strengthens the previous view [10] about the role of electrodes with regard to the dielectric behaviour of ruby mica.

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