

Low Frequency Dispersion in Non-conducting Contacts in Humidity

Altaf HUSAIN¹, S. Shakeel AHMED²

¹Condensed Matter Research Laboratory, Department of Physics, University of Karachi,
Karachi - 75270, PAKISTAN

e-mail: altafhussain850@hotmail.com

²Pakistan Council of Scientific and Industrial Research (PCSIR) Laboratories Complex,
Karachi - 75280, PAKISTAN

Received 24.08.2004

Abstract

To develop an enhanced understanding of an aspect of the charge transport mechanism in systems composed of solid-solid insulating contacts, the dynamic dielectric response of sand grains and of silica-silica rod contacts was investigated at dry and at 97% relative humidity (RH), in the frequency range 10^{-2} to 10^5 Hz. In dry atmosphere no appreciable amount of conductivity is observed, while in humidity the response follows a strongly dispersive behaviour. Sharp rise in capacitance gives a clear evidence for the storage of charge in the transport process. Collectively, the conductance of the sand grains increases super-linearly with the number of contacts suggesting filamentary conduction paths. The conductance G of the silica-silica contacts varies almost inversely with the number of contacts pointing towards a uniformly distributed conduction mechanism. It is found that the conduction process, despite a reduction in the geometrical area at the point of contact, is not significantly limited at the interface between the connecting surfaces.

Key Words: Surface transport, dynamic response, silica-silica contacts, filamentary conduction.

1. Introduction

Dielectric properties of a material are mainly related with the inherent microscopic mechanisms of electric polarization, especially in the presence of an electrical stress. Dielectric spectroscopy is an analytical technique, which is used to investigate the electrical conduction/polarization processes in different types of non-conducting materials under specified conditions. The technique comprises application of a time varying voltage to a material and measuring the components of current, in phase and out of phase with the voltage, as a function of frequency of the applied signal. The dielectric parameters such as susceptibility, capacitance, permittivity etc., are therefore generally expressed as frequency dependent complex quantities. Before technological advancement in instrumentation, measurements were limited to a minimum frequency of 1 Hz, but now it is possible to measure the response below 1 Hz.

The present study is an effort to achieve an understanding of the role of the solid-solid contact in controlling the dielectric properties of systems consisting of an assembly of macroscopic discrete components, e.g. grains in sand and soil, at normal temperature and pressure. A wide range of experimental studies on a variety of materials [1–4] has shown that the charge transport on humid insulating surfaces under time

varying electric field excitation follows a strongly dispersive pattern known as the Low Frequency Dispersion (LFD). A perfect example of LFD is observed in the dielectric response of humid sand and soil [5–6], where the complex susceptibility shows a strong dispersion at low frequencies. The susceptibility is a macroscopic property of a material and is described as the volume averaged response to an external field. Mathematically, the complex susceptibility can be expressed as the relation

$$\chi^*(\omega) = \chi'(\omega) - i\chi''(\omega), \quad (1)$$

where the real part $\chi'(\omega)$ and the imaginary part $\chi''(\omega)$ correspond to storage and loss of energy, respectively, in the system.

A similar relation applies in terms of the complex capacitance is given by

$$C^*(\omega) = [C'(\omega) - C_\infty] - iC''(\omega) \propto (i\omega)^{n-1}, \quad (2)$$

where the real $C'(\omega)$ and imaginary $C''(\omega)$ components of the complex capacitance $C^*(\omega)$ correspond to the capacitance and dielectric loss, respectively, whereas C_∞ is the value of capacitance at high frequency limit. The exponent n at low frequencies is close to zero.

In contrast to the classical Debye response, which comes about due to a system of noninteracting permanent dipoles [7], the LFD response arises due to a highly dispersive and lossy system yielding a frequency independent ratio,

$$C''(\omega)/C'(\omega) = \tan \delta = \cot(n\pi/2). \quad (3)$$

Equation (3) represents the ratio of the energy dissipated per radian to the energy stored at the peak of polarization.

The LFD behaviour is shown schematically in Figure 1, and is uniquely exhibited by a considerable number of dielectrics in which charge migration takes place either through the material bulk or surface, in particular the carrier dominated non-conductors [8].

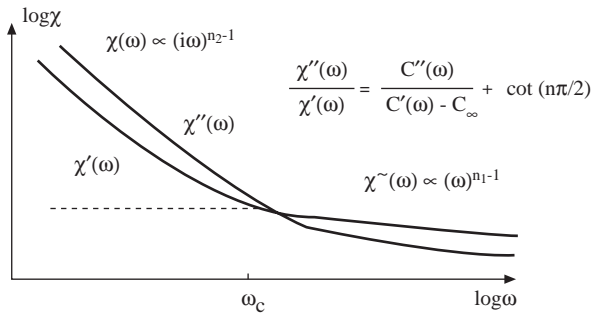


Figure 1. Schematic representation of pure LFD showing dependence of the complex susceptibility on frequency. The cross-over frequency ω_c separates the high and low frequency regions.

In general, the properties of a surface depend qualitatively on the identity, concentration and distribution of composing constituents [9–12]. In the presence of moisture, surface properties are affected in a variety of ways. Admixtures of humidity can give rise to loss due to contribution from the individual water molecules and this enhancement of loss forms the basis for the determination of structure [13–15].

In previous studies on sand [16], it was speculated that the surface makes a dominant contribution to the dielectric response and the transfer of charge at the interface is hindered by the series/parallel combinations

of hard grain-grain contacts. To elucidate these processes, the present work measures the collective dielectric behaviour of (a) a number of known sand grains placed in a trough thus forming a complex network, and (b) a series of contacts between crossed silica rods with metallic electrodes on the topmost and the bottom-most rod. Silica was chosen with a view that it is the major element in sand and is easily available in the granular form. Also, because of its highly insulating characteristics, silica has found many applications [17] in high voltage electrical installations. Composite materials containing silica as a component can be a good substitute for other materials such as ceramics [18].

Frequency domain and time-domain results for plain silica rods with circular electrodes on either side have been published [19–20], and here we report frequency dependent measurements on sand grains and on silica rod-rod contacts.

2. Experimental

2.1. Sand grains

For sand aggregates, a counted number of thoroughly cleaned grains was placed in an insulating trough of PTFE with two painted metallic electrodes at the ends, as shown in Figure 2a. Grains of almost similar shape and size, ranging from 35 to 900 in count, were selected to fill the trough starting from a single row. The assembly was kept in a controlled ambient of 97% RH. After each measurement a known number of grains was added and the measurement was repeated after allowing the sample to attain equilibrium with the surroundings. A hairspring hygrometer was used to measure the relative humidity.

2.2. Silica rod-rod contacts

Cylindrical silica rods were selected for measurements. A single silver paint electrode was applied at curved surface of each of the rod. To obtain least contact area the rods were arranged in a crosswise pattern as shown in Figure 2b and the entire assembly was fixed in an especially designed yoke made of PTFE. Rods of other diameters showed similar behaviour, except a change in magnitude of the response. We report here results of silica rods of 5 mm diameter.

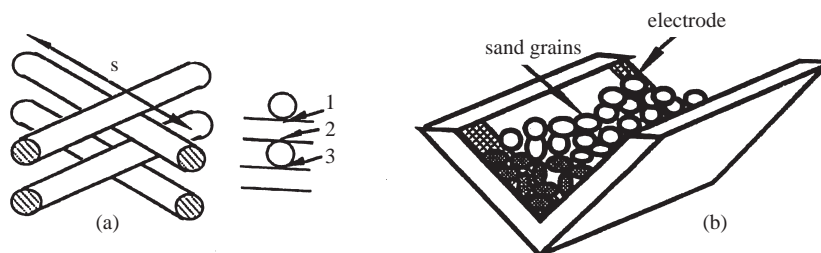


Figure 2. (a) Arrangement of silica rods to obtain varying number of contacts. (b) Sand grains in a PTFE holder.

2.3. Measuring technique

Frequency domain measurements were performed under the application of a sinusoidally time varying signal of amplitude 1 volt using a model 1250 Solartron Frequency Response Analyser (FRA) coupled with Chelsea Dielectric Interface (CDI) and a computer [21]. The circuit diagram of the measuring equipment is shown in Figure 3. There are two options: (i) standard technique and (ii) reference technique, either of which can be used independently. The reference technique is used in conjunction with the reference standard capacitors, while the switch S2 is kept closed and the switch S1 open. In the standard technique, the switch S2 is kept open and S1 is closed. Generally, the standard technique is satisfactory when the $\tan \delta$ of the

sample is greater than 5×10^{-3} and frequency of the applied signal is below 0.1 MHz. An advantage of $\tan \delta$ is that it is independent of geometry of the dielectric material.

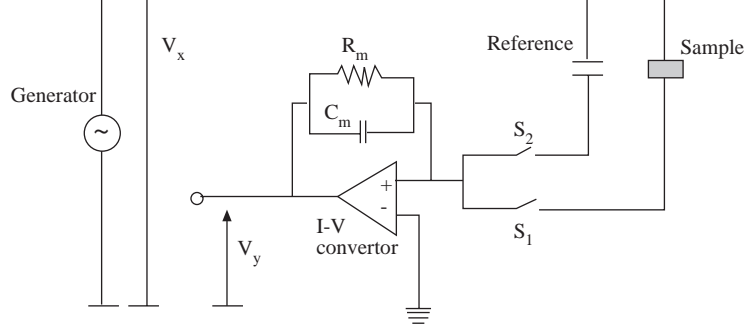


Figure 3. Circuit diagram of the frequency domain measuring technique.

For production of humid atmosphere, saturated salt solution of potassium sulphate was used and the samples were given a reasonable time to attain equilibrium with the ambient environment. All the measurements were made in the frequency range from 10^{-3} to 10^5 Hz.

3. Results

3.1. Sand grains

We report the results of measurements on sand aggregates consisting of a counted number N of grains in an insulating trough with metallic electrodes at the ends. The measured data are presented in the form of log-log plots showing the frequency dependence of the complex capacitance. The spectral response of the conductance G for a number of sand grains N is shown in Figure 4. A slowly varying trend at low frequencies with small exponent n gives a sure indication of LFD; while at high frequencies the slope approaches 0.5, a result of the consistent slow fall of $C'(\omega)$, as shown in Figure 5. For larger values of N the spectral shape of $C'(\omega)$ plots tends to become more like classical LFD. The conductance of the aggregates rises super-linearly as $N^{1.6}$, as shown in Figure 6, suggesting a rapid increase in conduction with the availability of alternate series and parallel paths.

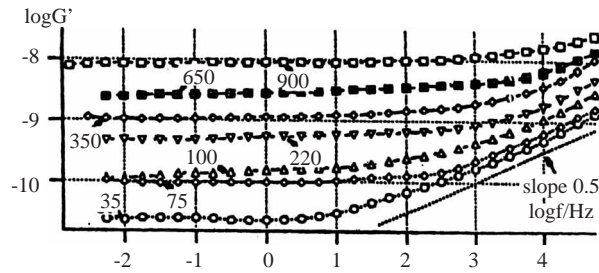


Figure 4. Dependence of the conductance G on frequency for different sets of sand grains.

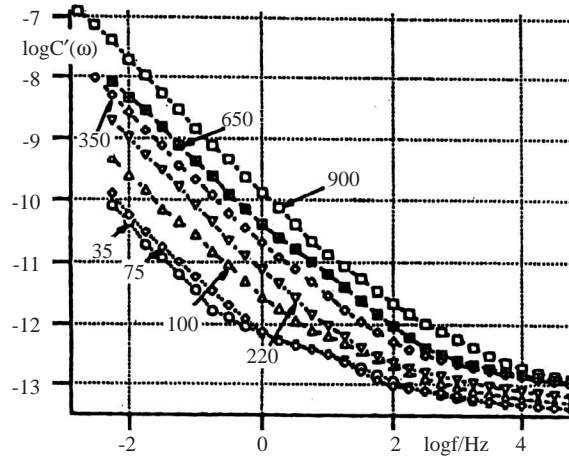


Figure 5. Variation of the frequency dependent capacitance for different sets of sand grains. The corresponding dependence of G is shown in Figure 4.

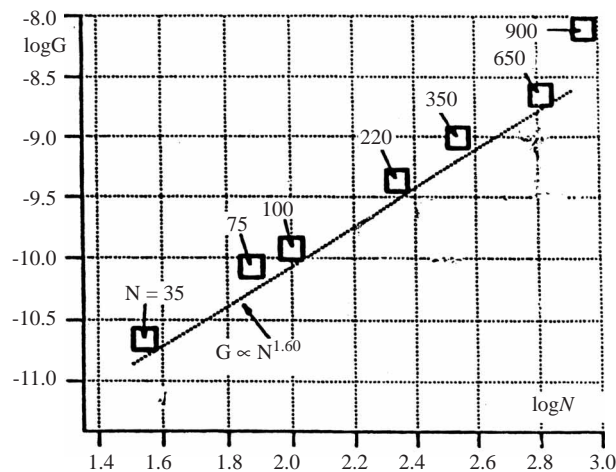


Figure 6. Logarithmic plots showing power law dependence of conductance on the number of sand grains N . All measurements were made at 97% RH.

3.2. Rod-rod contacts

The frequency dependence of the complex capacitance for a series connection from 1 to 6 contacts between identical silica rods is shown in Figure 7. All the results show them, at low frequencies, a strong dispersion in the real $C'(\omega)$ and imaginary $C''(\omega)$ parts of the complex capacitance, similar to that of sand grains but with a clearly distinguishable spectral shape. The loss component $C''(\omega)$ follows a slope of almost -1 which, in general points towards a uniformly distributed process; but in the present situation, the corresponding strong rise in $C'(\omega)$ following a slope of -1.3 , rules out the possibility of pure classical charge transport for which $C'(\omega)$ must remain frequency independent.

A falling tendency in the amplitude without any significant change in the spectral shape of all the responses implies that the rate of transport process is affected but its nature remains same.

The value of resistance $R=1/G$ at 1 Hz is found to vary almost linearly with N , as shown in Figure 8., indicating that the contacts are simply connected in series.

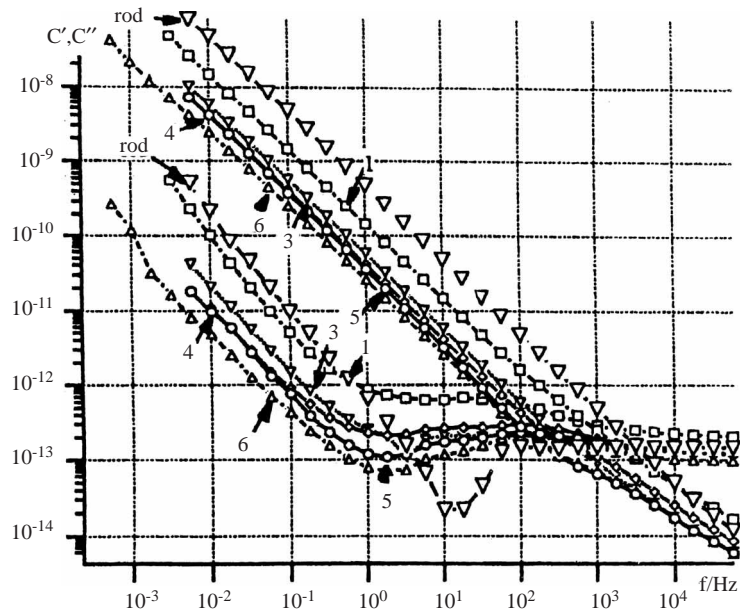


Figure 7. Frequency dependence of the complex capacitance for a number of silica rod-rod contacts.

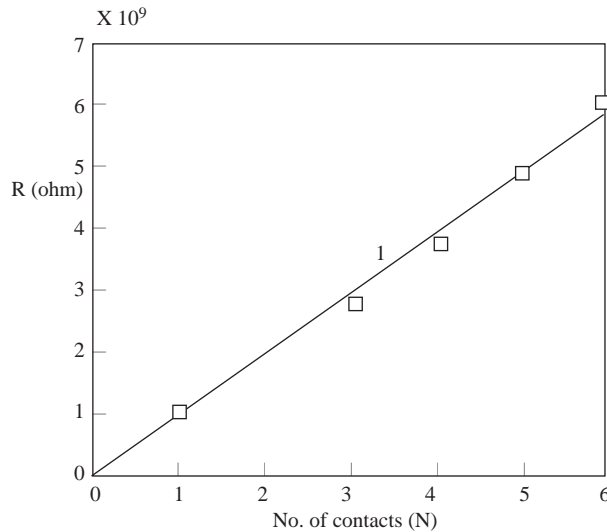


Figure 8. Variation of the effective resistance R with the number of rod-rod contacts N .

4. Discussion of Results

Our results provide clear evidence of the existence of LFD in the very different systems of silica rod-rod contacts and aggregates of sand grains. In both systems, the rapid rise in capacitance $C''(\omega)$ at low frequencies mentions storage of charge distributed along the length of the inter-electrode path. To our understanding, the strong dispersion with small values of the exponent n invokes the ratio of the energy lost per cycle E_l to the energy stored E_s in the system,

$$\cot(n\pi/2) = \text{Energy Lost/Energy Stored} = E_l/E_s. \quad (4)$$

In humidity, progressive development in conductance G suggests the formation of filamentary paths with the probability of following easy paths much larger than uniform flow. In sand, the set of smallest number

of grains, 35, behaved as if connected as a single filament of connected grains, whereas larger number sets behaved as if conducting through more series-parallel filaments. The suggested conduction paths are shown in Figure 9.

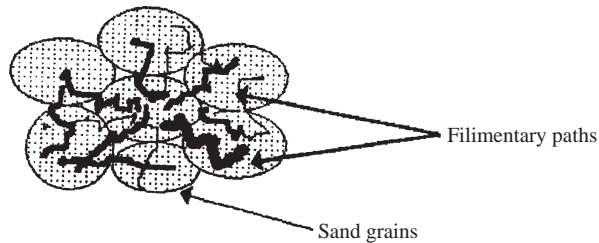


Figure 9. Formation of filaments on the surface to provide conduction paths between sand grains.

Absence of dry conductivity suggests that the filamentary charge flow is mainly due to adsorption of ambient humidity and is limited to the surface. The conduction comprises ionic charge flow resulting due to dissociation of water molecules into the components.

In sand, the amplitude of the response increases with the number of contacts, while it shows a decrease in the silica contacts. However, individually in both cases, the spectral shape remains independent of the number of contacts. The existing difference in the spectral shape associated with the silica rods and sand, points towards different transport mechanisms. Apart from the dissociated water ions, it is very likely that the ionic species present in the sand grains also makes a contribution to the response along with the adsorbed water.

Our results on rod-rod contacts show a very different behaviour compared with the granular contacts in sand. The resistance of a single rod-rod contact is found to be more than four times the resistance of a single plain rod with two silver paint electrodes for nearly the same separation. It means that the area or circumference of the contact cannot be a dominant influence in the process. As a possible physical interpretation of the results, we perceive that in the presence of humidity the limiting process is determined by condensation of water through capillary action in the narrow space at the contact, thus forming a pool of water, which enlarges the effective area, as shown in Figure 10. Due to collection of water, relatively large number of carriers can contribute to the response and this would also explain the relatively small increase of resistance of a single contact compared to a plain rod itself.

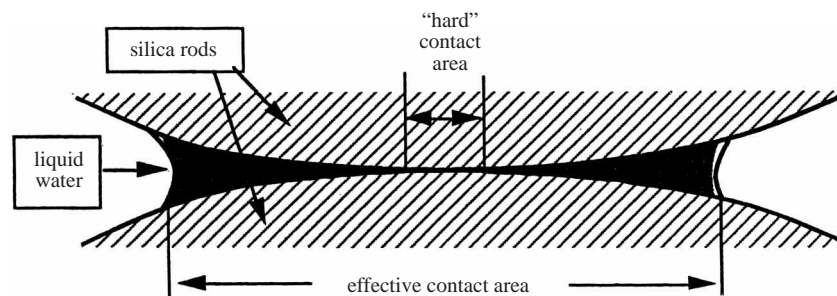


Figure 10. Hypothetical view of a single contact between two solid silica rods in humidity. Condensation of water vapour around the point of contact has considerably increased the contact area.

The inverse dependence of G on N may be attributed to the reduction in the easy filamentary paths residing on the curved surface between successive contacts. Contrary to this, the fast increase in conductance G with N in sand may arise due to availability of a large number of interconnected series-parallel paths through which ions may move with relative ease.

5. Conclusions

The silica rod contacts show a different behaviour compared with the contacts between sand grains. In rod contacts, although the inverse dependence of G on N suggests a uniformly distributed conduction mechanism, it is not a simple dc process in which capacitance remains unchanged. Absence of dry state bulk conductivity in both cases shows that the major contribution comes from the surface due to adsorption of water. The difference in the spectral shape of the response of sand and silica indicates that the nature transport mechanism is different in both cases. This also suggests that silica does not play a major role in the response of sand. Presence of an appreciable amount of capacitance and conductance suggests that the contacts do not play a major role in restricting the charge carriers at the point of contact as was speculated in previous studies. The super-linear variation of G with the number of contacts N suggests that the transport is filamentary along the contacts with the probability of finding a conducting path from one grain to another being strongly nonlinear.

References

- [1] A. K. Jonscher, *Dielectric Relaxation in Solids*, (Chelsea Dielec. Press. Lond. 1983), p. 220.
- [2] A. K. Jonscher, *Universal Relaxation Law*, (Chelsea Diel. Press. Lond. 1996), p. 150.
- [3] E. F. Owede and A. K. Jonscher, *J. of Elect. Chem. Soc.*, **135**, (1988), 1757.
- [4] D. M. Craig, R. M. Hill, and J. M. Newton: *Pharmaceutical Tech. Int.*, (1990), p. 61.
- [5] M. Shahidi, J. B. Hasted and A. K. Jonscher, *Nature*, **258**, (1975), 595.
- [6] M. Shahidi, Ph.D Thesis, University of London, 1975.
- [7] P. Debye, *Polar Molecules* (Dover Publications, New York. 1945).
- [8] A. K. Jonscher, *Philosophical Mag. B*, **38**, (1978), 487.
- [9] M. Prutton, *Surface Physics*, (Oxford Physics Series. 1987), 5–10.
- [10] Andrew Zangwill, *Physics at surfaces*, (Camb. Univ. Press, Cambridge. 1990), p. 20.
- [11] L. Holland, *The Properties of Glass Surfaces*, (Wiley, New York. 1964), p. 56.
- [12] M. Henzler, *Surface Science*, **25**, (1971), 650.
- [13] A. Saad and R. Tobazeon, Surface Conduction and Losses of An Insulator Wetted by A Liquid Dielectric, *IEEE Trans. on Elect. Insulation*, Vol. EI-19, No. 3, (1984).
- [14] Sivert H. Glarum, Dielectric Relaxation of Polar Liquids, *J. of Chem. Phys.*, **33**, (1960).
- [15] L. A. Dissado and R. M. Hill, *Philosophical Mag. B*, **41**, (1980), 625.
- [16] M. K. Anis and A. K. Jonscher, *J. Mat. Sci.*, **28**, (1993), 3626.
- [17] H. Janssen, J. M. Seifert and H.C. Karner, *IEEE Trans. Diel. Elec. Insul.*, **6**, (1999), 651.
- [18] A. P. Gonon, A. Sylvestre, J. Teyssere and C. Prior, *J. Mat. Sci., Materials in Electronics*, **12**, (2001), 81.
- [19] A. K. Jonscher and A. Husain, *Physica B*, **217**, (1-2), (1996), 29.
- [20] A. Husain and A. K. Jonscher, *Physica B*, **222**, (1996), 123.
- [21] J. Pugh, Dielectric Measurements Using Frequency Response Analyzers, DMMA, *IEE Publication No. 239*, (1984), 247.