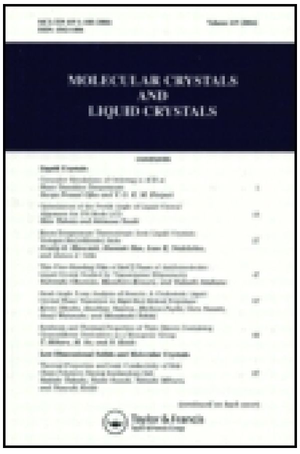

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DIELECTRIC PROPERTIES OF GLASS-DISPERSED LIQUID CRYSTALS

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Abstract Liquid crystals can be dispersed in gel-glass silica matrices by sol-gel processes. The resulting gel-glass dispersed liquid crystals (GDLCs) are similar to polymer-dispersed liquid crystals; however, the use of an inorganic silica matrix yields a material with enhanced optical and mechanical properties. Dielectric properties of these materials are highly dependent on the manufacturing conditions. Impedance spectroscopy of the isolated silica matrix and the GDLC composite has been employed to characterize the system between 200 mHz and 8 MHz. The results have been compared to theoretical results based on several equivalent circuits.

INTRODUCTION

Glass-Dispersed Liquid Crystal (GDLC) devices are made of liquid crystal microdroplets embedded in a silica matrix using sol-gel processes[1,2]. A thin film of silica matrix-liquid crystal composite is sandwiched between transparent electrodes to produce a scattering medium. Behavior of GDLCs is similar to PDLCs. The main difference comes from the inorganic nature of the matrix. This produces a non-flexible material with enhaced optical properties in the NIR, visible and near UV spectral ranges.

Once formed the composite, when an electric field is applied across the heterogeneous medium (matrix and liquid crystal), the electro-optical switching of the device depends on the dielectric properties of each separate phase as well as on their mutual interaction.

Impedance spectroscopy was used to study the dielectric behavior of both, isolated matrices and composites. This technique is a powerful tool to determine dielectric properties of materials[3]. These properties are closely related to electro-optical switching, thus being useful to optimize optical and dynamic properties of GDLC devices.

EXPERIMENTAL

A simple electrical circuit (Figure 1) was built to measure the impedance spectra. A frequency range from 0.2 Hz to 8 MHz (i.e. almost 8 decades) was employed. To ensure a linear response of the dielectric medium, a small AC signal (600 mVpp) was used; therefore, no switching is produced in the GDLC samples. Two kinds of test samples were prepared to perform impedance measurements: the first kind contains the isolated silica matrix and the second is the matrix-liquid crystal composite. The impedance data for the test sample are obtained by measuring simultaneously signals V_1 and V_2 (modulus and phase). A 4-channel oscilloscope (Tektronix TDS-520) was used to perform these measurements. This oscilloscope displays automatically the delay (i.e., the phase) between signals introduced into two channels.

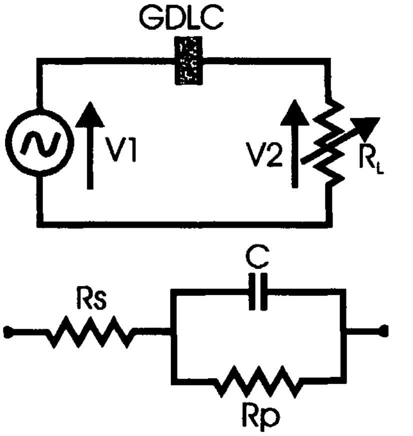

Figure 1. Experimental setup and equivalent circuit.

RESULTS AND DISCUSSION

The following expression for the impedance of the test sample can be written:

$$Z_c = R_L \left(\frac{V_1 - V_2}{V_2} \right) \quad (1)$$

As shown above, the sample (either the isolated silica matrix or the matrix-liquid crystal composite) is assumed to be a parallel planar capacitor with a non-ideal dielectric. In this case, the following relationships are met:

$$Y_c = \frac{1}{Z_c} = j\omega\varepsilon^* C_0 = j\omega C_0(\varepsilon' - j\varepsilon'') = \omega C_0\varepsilon'' + j\omega C_0\varepsilon' \quad (2)$$

where Y_c is the admittance of the capacitor, ε^* is the complex dielectric permittivity, and ε' and ε'' are respectively the real and imaginary parts of the complex dielectric permittivity. C_0 is the capacitance when the capacitor is filled with air as dielectric medium. If the cell has been previously calibrated, the value of C_0 will be given by

$$C_0 = \varepsilon_0 A/d \quad (3)$$

where ε_0 is the dielectric permittivity of the vacuum ($\varepsilon_0 = 8.85\times10^{-12}$ F/m), A is the effective area of the electrodes and d is the sample thickness.

Impedance data may be represented by a Bode plot, which shows the modulus, $|Z_c|$, and the phase of the impedance against the logarithm of frequency. This allows to define an equivalent electrical circuit which describes the behavior of the sample. Two different approaches are studied to describe this electrical behavior. The first one is extracted from the Bode plots and the second one is derived from fitting parameters.

Bode-plot Model

The so-called *Randles Circuit*[4] is one of the simplest and most widely used equivalent circuits for electromechanical systems. This circuit has been used for modelling PDLC devices[5] thus it seems appropriate for GDLC modelling. The circuit (Figure 1) is made of a series resistor R_s associated mainly to the resistance of the electrodes, and an additional parallel combination formed by a resistor R_p and a capacitor C. The parallel resistor simulates the non-ideality of the actual capacitor cell.

It is convenient to carry out an asymptotic study of modulus and phase of the impedance as a function of frequency for this circuit. The complex impedance for the Randles circuit will be given by

$$Z(j\omega) = \frac{(R_s+R_p) + j\omega R_s R_p C}{(1 + j\omega R_p C)} = (R_s+R_p)\left(\frac{1 + j\omega \frac{R_s R_p}{R_s+R_p} C}{1 + j\omega R_p C}\right) \quad (4)$$

$$\log|Z(j\omega)| = \log|(R_s+R_p)| + \log\left|1 + j\omega \frac{R_s R_p}{R_s+R_p} C\right| - \log\left|1 + j\omega R_p C\right| \quad (5)$$

Taking logarithms, the following expressions result:

$$\Phi_{Z(j\omega)} = \arctan\left(\omega \frac{R_s R_p}{R_s+R_p} C\right) - \arctan\left(\omega R_p C\right) \quad (6)$$

Resistances in this circuit are represented by horizontal lines (for $\omega \to 0$ y $\omega \to \infty$) in the Bode plots, while the capacitance is related to the line whose slope is -1. Figure 2a capacitance is extracted from the fitted curve . The time constant can be taken from the phase curve at 45° slope. Using this number, the time constant R_sC is 68 ns approximately. The R_s value derived from this time constant and the above capacitance agrees with the value measured from the horizontal end of the log|Z| plot. The value of R_pC constant cannot be derived from the plot. It can be only estimated as being very high ($R_pC \geq 10$ s). This constant is related to the charge decay across the bulk of the sample.

The characteristic curves for modulus and phase of the impedance versus frequency for a silica matrix containing liquid crystal in OFF state are shown in figure 2b. The time constant in this case is $R_sC \approx 108$ ns. The resistance R_p is very high; it is not possible to get it from the plot. The best estimation for this time constant is $R_pC \geq 1.60$ s.

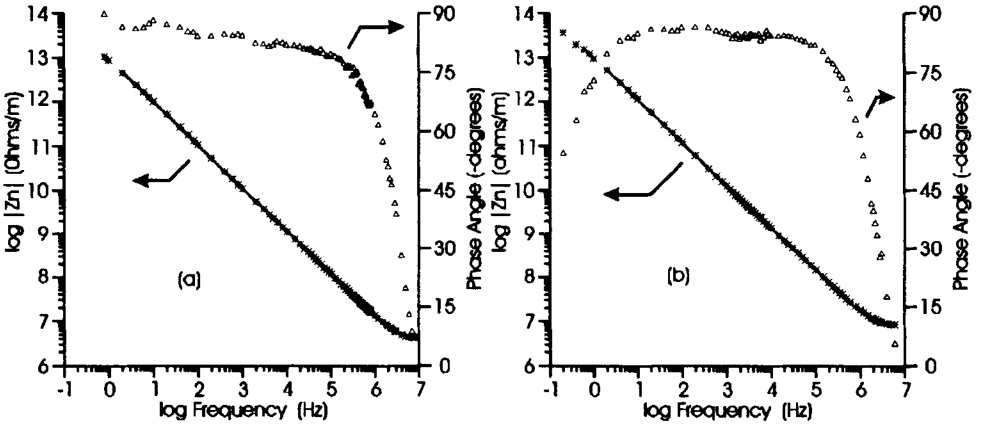

Figure 2. Bode plot of silica matrix and GDLC composite.

As seen in figure 2, impedance spectra with and without liquid crystal are fairly similar, the major differences being found in the impedance phase at low frequencies. This is related to a decrease in R_p upon addition of the liquid crystal. The values for ε' and ε'' can be extracted from the equivalent electrical circuit. The real and imaginary parts of $Y(j\omega) = 1/Z(j\omega)$ in the Randles circuit are related to ε'' and ε' respectively. Figures 3a (silica matrix) and 3b (GDLC composite) show the variation of both permitivities with frequency. Comparing calculated and experimental values, large differences are found, indicating that the model is too simple for explaining the features of this dielectric system.

<u>Fitting Model</u>

Experimental results point to the presence of relaxation mechanisms that the Randles circuit does not take into consideration. The loss factor of GDLC samples at low frequencies, increases abruptly, while a second relaxation phenomenum appears in the kHz range.

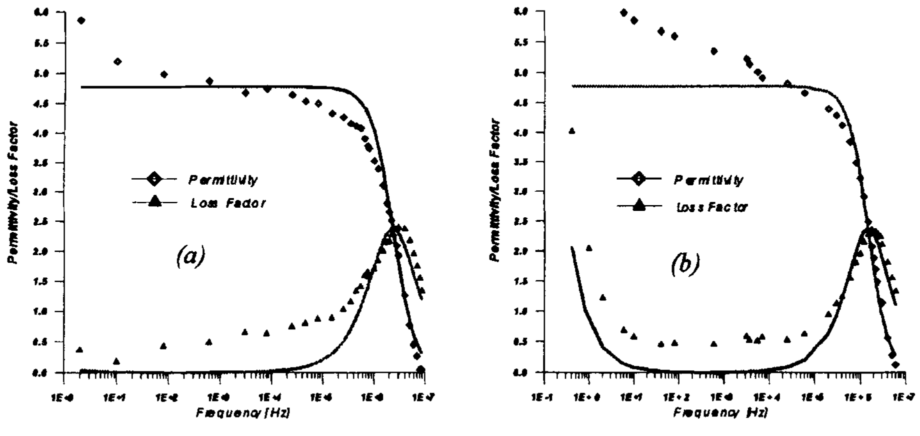

Figure 3. Permittivity and loss factor of Silica matrix and GDLC composite. Lines are calculated values from Randles equivalent circuit.

These two contributions have been atributed in similar systems to ionic contributions and liquid crystal relaxation processes[6]. Ionic charge carriers have low mobility, therefore their contribution to relaxation appears only at the lowest frequencies. Quantification of these two contributions lead to a second model having the following features:

Low frequencies: A term due to free charges (ions) is described as a contribution $1/\omega^n$, where n is an exponent close to 1.

Medium frequencies: A small contribution weighed with exponent α_1. This is in principle assigned the liquid crystal, as the peak is not detected in silica matrix samples.

High frequencies: The term due to the R_sC time constant. The same term as in the Randles model, but includes a weighing exponent α_2. The resulting exponent is close to 1.

The above contributions can be expressed as:

$$\varepsilon'' = \frac{\sigma}{\varepsilon_0}\frac{1}{\omega^n} - Im\left(\left[\varepsilon_1(\infty) + \frac{\varepsilon_1(0) - \varepsilon_1(\infty)}{1 + (j\frac{\omega}{w_1})^{1-\alpha_1}}\right] + \left[\varepsilon_2(\infty) + \frac{\varepsilon_2(0) - \varepsilon_2(\infty)}{1 + (j\frac{\omega}{w_2})^{1-\alpha_2}}\right]\right) \qquad (7)$$

where σ is the sample conductivity, ω_1 and ω_2 are resonance frequencies, $\varepsilon_i(0)$ and $\varepsilon_i(\infty)$ i=1, 2 are the relative dielectric permittivities below and above the relaxation processes. The fit shown in figure 4 has been obtained with the following values:

$\sigma = 3.2188 \cdot 10^{-11}\Omega^{-1}cm^{-1}$ $\quad$ n = 0.57 $\quad$ $\omega_1 = 1.885 \cdot 10^4$ rad/s

$\omega_2 = 1.131 \cdot 10^7$ rad/s $\quad$ $\alpha_1 = 0.4867$ $\quad$ $\alpha_2 = 0.1267$

The relaxation mechanisms shown above deviate from the ideal Debye behavior, for they are modified by exponents α_1 and α_2. Therefore, frequency-independent Cole-Cole plots of ε" vs. ε' will be circles whose centers are not located on the abscissa. This is seen in figure 5. The right part of the plot corresponds to the low and medium frequency mechanisms. Fitting of this small region would require[7] a second circle.

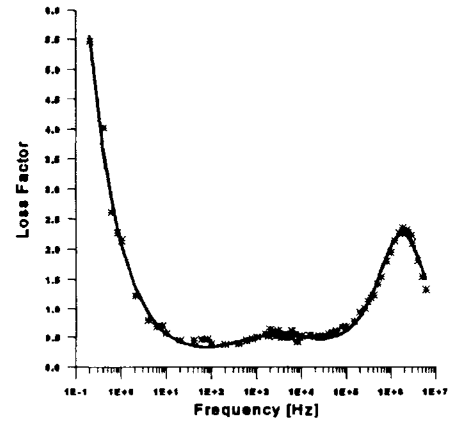

Figure 4. Fit of Loss Factor curve.

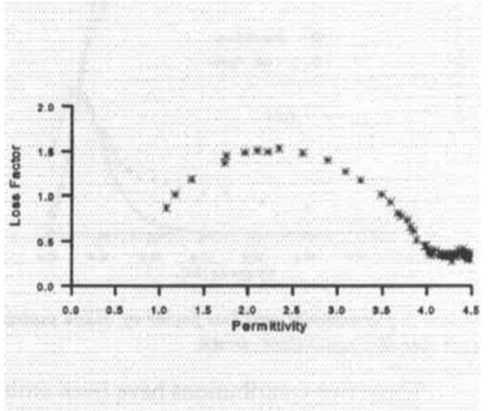

Figure 5. Cole-Cole experimental plot.

In summary, GDLC dielectric characterization demonstrates that the sample resistivity is lowered by the addition of liquid crystal. Yet the time constant R_pC is high enough, the voltage drop thus being negligible. Ions are detected at low frequencies as a significant contribution to the loss factor. The system cannot be accurately described by a simple equivalent Randles circuit, but requires separate contributions at different frequencies. This study is presently being completed with an additional set of measurements of switched GDLCs (i.e., ON state samples). The complete dielectric characterization of GDLCs would allow the estimation of relevant manufacturing parameters such as the effective dielectric permittivity and the liquid crystal filling factor.

REFERENCES

1.- D. Levy, C.J. Serna and J.M. Otón, Mater. Lett. 10, 470 (1991)
2.- J.M. Otón, J.M.S. Pena, A. Serrano and D. Levy, Appl. Phys. Lett. 66 (8) (1995)
3.- J.R. MacDonald, Impedance Spectroscopy Emphasizing Solid Materials and Systems John Wiley & Sons, New York (1987)
4.- J.E.B. Randles Discuss Farad. Soc. 1, 11 (1947)
5.- L. Welsh and L. White Proc. Soc. Informat. Display SID 90 Digest, 220 (1990)
6.- F.M. Gouda, Ph.D. Thesis, Chalmers University of Technology, Göteborg, Sweden (1992).
7.- A.K. Jonscher, Dielectric Relaxation in Solids, Chelsea Dielectrics Press, London (1983)