

BROAD BAND DIELECTRIC SPECTROSCOPY USING AN OPEN-ENDED LINE

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ABSTRACT

In this work a method of broad band dielectric spectroscopy using an open-ended line is proposed. The mathematical development of this method leads to a transcendental equation on the complex plane that possesses infinite solutions. A numerical procedure is described which is capable of obtaining the solution required avoiding the problem of infinite solutions. The study of this equation suggests that even large errors in the experimental parameters induce only small uncertainties in the complex permittivity of the sample, and the thinner the sample the lower the uncertainty. An *ad hoc* sample holder with a characteristic impedance of 50 Ω was designed and built to implement the proposed method. It allows temperature control and can be connected to three different measuring devices: the precision LCR meters HP 4284A (20 Hz – 1 MHz) and HP4285A (75 kHz – 30 MHz) and the vectorial network analyzer HP8719D (50 MHz – 13 GHz). The work frequency is limited from above by the geometric characteristics of the sample holder and the sample permittivity. The acquisition and treatment of the data have been automatized. For that purpose an interface HP-IB has been used and a code has been developed in HTBasic 8.3 for Windows designed in order that the measurements can be carried out by a laboratory technician without specific knowledge of dielectric characterization. The measurements of polar and non polar samples have established the appropriateness of all the different steps carried out for the implementation of the technique.

INTRODUCTION

Precise measurements of permittivity in the frequency domain in several frequency ranges require the use of several different experimental setups [1]. In this case, it is desirable to use the same measuring cell in all of them so as to ensure identical sample composition. For this purpose, the sample-holder is often an open-ended coaxial cell. Then circuit theory (lumped circuits) is used at low frequencies while distributed parameters are used for high frequencies [2]. In certain cases, the sample volume should not be too small to ensure reproducibility of measurements (this can prevent or minimize undesired effects such as the influence of impurities in the sample). In this case, if circuit theory is applied and the sample is polar, then its electrical length could limit the frequency range below the highest one which the experimental setup would allow. These arguments led to the decision to design and build a coaxial measuring cell and work with distributed parameters on the basis explained in the following sections.

The operating principle of the cell is based on the reflection method in a coaxial line with open-circuit terminal impedance (see Fig.1). The complex relative permittivity of the sample, $\epsilon^* = \epsilon' - j\epsilon''$, can then be determined from the measurement of the sample's input impedance Z_s or of its admittance Y_s :

$$Z_s \propto \frac{\coth x}{x} \quad (1a)$$

$$Y_s \propto x \cdot \tanh x \quad (1b)$$

where $x = \gamma_\epsilon d$, d is the sample thickness and γ_ϵ its propagation factor. This in turn satisfies

$$\gamma_\epsilon = j \frac{2\pi\sqrt{\epsilon^*}}{\lambda_0} = \alpha + j\beta \quad (2)$$

with α and β the attenuation factor and the phase factor respectively, and λ_0 the wavelength in vacuum.

For a loss-free medium, the asymptotic behavior of (1a) ($Z_s \propto \cot x/x$, $x = \beta d$) for $x \ll 1$ suggests that large errors in the impedance induce only small errors in x and hence in the permittivity of the sample. However by differentiation of

Eqns 1a and 1b (Z_s and $Y_s = Z_s^{-1}$) one finds that equal relative errors for Z_s or Y_s , $\Delta Z_s/Z_s$ and $\Delta Y_s/Y_s$ respectively, lead to the same error in x : $\Delta x = f(x, \Delta Z_s/Z_s) = f(x, \Delta Y_s/Y_s)$. For solving these transcendental equations iterative numerical methods are employed, of which the more commonly used is Newton-Raphson method [3-5]. It has been observed that convergence with this method requires a better initial approximation to the root for (1a) than for (1b). This, as well as the desire to compare results arrived at with the proposed method (distributed parameters) to those obtained with circuit theory, has lead to the use of admittance for calculations.

The two following cases were considered:

a) The propagation effects must be taken into account. In this case the admittance Y_s in (1b) can be written as

$$Y_s = Y_\varepsilon \tanh(\gamma_\varepsilon d) \quad (3)$$

where Y_ε is the characteristic admittance of the sample, related to ε^* through

$$Y_\varepsilon = \sqrt{\varepsilon^*} Y_0 \quad (4)$$

Y_0 is the free-space (vacuum) characteristic admittance.

b) The propagation effects can be neglected. Then (3) can be written

$$Y_s = Y_\varepsilon \gamma_\varepsilon d \quad (5)$$

If one measures the input admittance with sample, Y_s , and without sample, Y_v , and uses the vacuum propagation factor (2) with $\varepsilon^* = 1$ one arrives at the well-known relation used in lumped-circuits theory

$$\varepsilon^* = Y_s / Y_v \quad (6)$$

THEORY

When using distributed-parameters theory it is necessary to use (3) and not (6). On the other hand when working with an open-circuit measuring cell for liquids a dielectric bead (also called window) is needed at each side of the sample. These are required to ensure tight closure but modify the incident and reflected signals. Figure 1 shows the two windows, with widths d_1 and d_2 , between which the liquid sample of width d is located.

By using the relation between the admittances at two planes (in two consecutive media) such as C and D in Fig.1, Y_C and Y_D respectively:

$$Y_D = Y_t (Y_C + Y_t \tanh(\gamma_t d_2)) / (Y_t + Y_C \tanh(\gamma_t d_2)) \quad (7)$$

one can obtain the admittance at one of them if the other one is known, assuming that the propagation factor of the

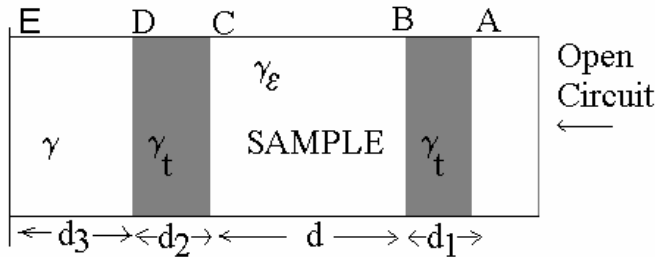


Fig.1. Diagrammatic representation of the cell containing the dielectric sample between the two windows.

dielectric bead, γ_t , its characteristic admittance Y_t and its width d_2 are also known. (7) is equally valid when written for normalized admittances. In what follows, admittances are assumed to be normalized and they will not be distinguished notationally from non-normalized quantities.

For the cell without sample and using expressions analogous to (7) one can determine (see Fig. 1 for notation): (i) Y_B from Y_A , γ_t , and d_1 ; (ii) Y_C from Y_B , Y (normalized characteristic admittance of air), γ (air propagation factor) and d ; (iii) Y_D from Y_C , γ_t , and d_2 (7).

Writing again the same expression for the planes E and D, one obtains:

$$\tanh(\gamma \cdot d_3) = Y(Y_D - Y_E) / (Y_E Y_D - Y^2) \quad (8)$$

where d_3 is the distance from the plane of the measurement to the plane D. The distance d_3 is unknown and Y_E is obtained by direct measurement.

Let us now consider the cell with sample. The symbol Y_X^M will be used to denote normalized admittance in the plane X. Y_E^M is obtained by direct measurement. Then Y_D^M can be calculated from Y_E^M and the value obtained for $\tanh(\gamma \cdot d_3)$, (8). From Y_D^M one obtains Y_C^M , which in turn is related to the already known Y_B through the equation:

$$Y_C^M = Y_\varepsilon (Y_B + Y_\varepsilon \tanh(\gamma_\varepsilon d)) / (Y_\varepsilon + Y_B \tanh(\gamma_\varepsilon d)) \quad (9)$$

The permittivity ε^* of the sample can be obtained from this equation because γ_ε and Y_ε are related to it by (2) and (4) respectively.

Two methods were used to obtain the permittivity from that transcendental equation in the complex plane. These are described below.

First method

The approximation $\tanh(\gamma_\varepsilon d) \approx \gamma_\varepsilon d$ is used in (9), so that:

$$\gamma_\varepsilon \cong \sqrt{-\left(j \frac{2\pi}{d\lambda}\right)^2 Y_C^M Y_B - j \frac{2\pi}{d\lambda} (Y_B - Y_C^M)} \quad (10)$$

where λ is the wavelength in air. We have observed that this method is valid only when the electrical length of the sample is very small. When this is not the case it has been checked that the value of Y_C^M yielded by (9) in this approximation is different from the corresponding experimental value.

Second method

The Newton-Raphson method is applied using as an initial approximation the value of $\gamma_\varepsilon d$ corresponding to calculating γ_ε from (10). The propagation factor is related to permittivity through (2).

Following the procedure described in [3], one concludes that:

- i) Once the solution for $\gamma_\varepsilon d$ is found the others can be calculated from it.
Using polar notation $\gamma_\varepsilon d = T_0 \angle \tau$; $45^\circ < \tau < 90^\circ$, once one solution $T_0 \angle \tau$ has been found, other solutions can be found from that one by using the Newton-Raphson method with initial approximations $T_n = T_0 + n\pi$, with $n = 1, 2 \dots N$ and N the number of solutions sought.
- ii) For usual substances, T_0 is close to zero. The next solution, obtained by using $T_0 + \pi$ as an initial approximation, leads to permittivity values of the order of 10^4 . Therefore for usual substances there is no ambiguity [5]
- iii) In the given neighborhood, there are no other roots close to the solution sought. Thus there is no need to use any discerning algorithm [3].

This second method, more general than the first, is the one that has been used in the software developed for data treatment. Also, at all stages in the calculation it has been considered that measurements had been done in air and not in a vacuum. For this purpose it has been taken into account that propagation speeds in these two media are different.

SAMPLE HOLDER FOR LIQUID SAMPLES

Implementation of this method required the construction of a coaxial sample-holder. It was designed with a characteristic impedance of 50Ω and built in steel with low thermal expansion coefficient. The sample is located between two windows of 99.9% SiO_2 , ensuring tight closure while centering and holding the internal conductor (see Fig. 1). Previously, a prototype of this sample-holder had been built in brass with Teflon windows. The sample is injected by means of two small holes in the external conductor. The liquid arrives at the holes through two funnels embedded in a thermal enclosure allowing temperature to be stable within 0.05°C . The funnels are kept filled with liquid during measurements to avoid sample evaporation. Connection between the measuring devices, an HP 4284A (20Hz-1MHz) and an HP 4285A (75kHz-30MHz) both precision LCR meters (with four coaxial measurement terminals), and the sample holder was made through the four terminal adapter HP16085A. The connection to the network analyzer ANA HP8719D (50 MHz – 13 GHz) was done using a HP85131 test post cable. As open circuit at the end of the line a precision open-circuit termination GR-900-WO was used. Fig. 2 shows a schematic diagram of the experimental setup for the LCR meters. This is similar to that for the ANA.

The acquisition and treatment of the data have been automatized. For that purpose an interface HP-IB has been used and a code has been developed in HTBasic 8.3 for Windows designed in order that the measurements can be carried out by a laboratory technician without specific knowledge of dielectric characterization.

EXPERIMENTAL RESULTS

Measurements with both polar and non-polar liquids were made following the method proposed above. The static

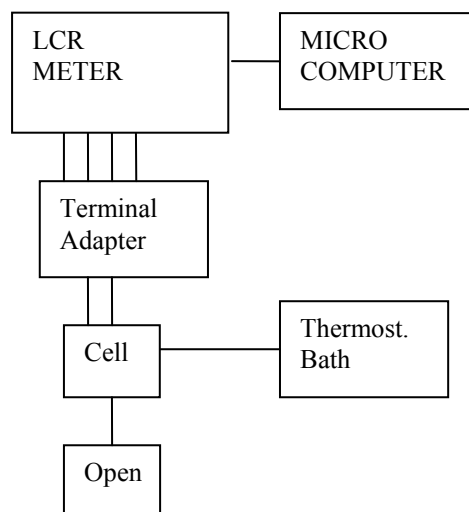


FIG. 2. Schematic representation of the experimental setup.

permittivity, ϵ_s , measurements of non polar standard samples have established the appropriateness of all the different steps carried out for the implementation of the technique since the values of the real part of the complex permittivity differ less than 0.6 % from those in the literature and the values of the loss factor, ϵ'' , lie between $4 \cdot 10^{-3}$ and $7 \cdot 10^{-4}$.

Several measurements were made on primary alcohols as polar samples. The chemicals, supplied by Aldrich, were recently acquired and kept in an Argon (N-55) atmosphere as soon as the bottles were opened. They were degassed ultrasonically and stored over freshly activated molecular sieves (Union Carbide, type 4 Å) to avoid moisture absorption because of their hygroscopic behavior.

The values we obtained for ϵ' at 100 kHz differ less than 0.1 % from those of the ϵ_s , extrapolated from the values obtained by Bezman *et al.* [6]. These values also differ slightly from those reported by other authors [7-9], which in turn differ among themselves because they were obtained by extrapolation from the behavior at microwave frequencies and therefore bear little comparison to measurements directly made at low frequencies.

Something similar happens with the permittivity at high

frequencies, ϵ_∞ . That is why we question the reliability of the comparison without further tests of the values of ϵ' obtained by interpolation at single frequencies. These values were thus checked and their behavior (particularly for alcohols with long chains, since the relaxation frequency diminishes with increasing chain length) is similar to that reported in the literature.

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