Low Frequency Permittivity and Conductivity of Essential Lemon Oil

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Abstract

Permittivity and conductivity values of two industrial samples of essential lemon oil have been determined from impedance and capacitance measurements in the low frequency range (500 Hz – 1000 kHz). Measurements have been performed using the Impedance Analyzer HP 4284 A and an acrylic cylindrical cell for liquids, with plane parallel electrodes of variable separation between them, designed and constructed in our laboratory. The main proposals of this work are to give some insight about the low frequency dielectric behavior and to fill partly the bibliographical data of this industrial substance.

Introduction

Tucuman province is the principal producer and exporter of lemon and its derivatives of Argentina, being one of the most important worldwide lemon producing regions. Derivatives of lemon, such as its essential oil, have a wide spectrum of industrial, medicinal, cosmetic, etc. applications, and for this reason it is of interest to characterize its physical and chemical properties.

Papers on physico-chemical properties of concentrated and clarified Argentinian lemon juice as well as Argentinian lemon peel have been published by our research group (1–4) taking into account that the more usually commercialized lemon derivatives are juice, oil and pectin.

In this work the low frequency electrical properties, permittivity and conductivity (5), of two lemon oil samples obtained from the peel of the Eureka cultivar are studied. Because information about these properties is absent from the literature, this work is intended to fill the gap.

Experimental

The lemon oil samples used in this work have been provided by two local factories. For reasons of confidentiality, we will call the samples C and T simply and no chemical composition data will be given. The dielectric measurements have been realized using the Impedance Analyzer Hewlett - Packard, HP 4284 A, and an acrylic cylindrical cell for liquids with plane parallel electrodes, designed and constructed in our laboratory. The electrodes, 54.5 mm in diameter, consist of a surgical stainless steel self-adhesive films 0.1 mm in thickness. Two O-rings have been included in order to avoid any leakage of liquid. The electrodes separation can be varied and measured to a precision of 0.01 mm. This constructive detail permits control of the electrodes' polarization in the calculation of the dielectric magnitudes. Measurements have been realized at electrode distances of 1.5; 2.0; 2.5 and 3.0 mm, at room temperature (20.5 °C), and in the frequency range between 500 Hz to 1000 kHz. The measured electrical parameters were: capacitance (Cp), loss factor (D), impedance (Z), phase angle (θ), resistance (R) and reactance (X). For all measurements, the open and short corrections recommended in the manual of procedures of the equipment (6), have been previously performed. The measurements of these electrical parameters have been realized from a series of ten measurements for each frequency, using the Analyzer in the average mode by means of which every value obtained is the average result of 10 successive measurements that the device realizes internally.

Taking into account the effect of electrodes polarization - represented by a resistance (R_e) and a capacitance (C_e) connected in series to the sample impedance, Z_s , (7–9)- the measured impedance Z is written

$$Z = Z_s + Z_e = (R_s + R_e) + j\,(X_s + X_e) \quad (1)$$

where $j = \sqrt{(-1)}$ is the imaginary unit.

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The impedance components associated with the sample are dependent on the electrode distance d, and frequency of the applied field. In turn, the impedance components associated with the electrode polarization effect depend only on frequency. The generalized complex conductivity of the sample, $\sigma^*(\omega)$, can be written

$$\sigma^*(\omega) = \sigma(\omega) + j\,\omega\,\varepsilon_0\,\varepsilon(\omega) \qquad \textbf{(2)}$$

where $\sigma(\omega)$ is the real component of the conductivity, ω is the field angular frequency, ε_0 is the permittivity of vacuum and $\varepsilon(\omega)$, the sample permittivity. Consequently, the impedance Z can be written in term of the generalized electric conductivity:

$$Z = d / (\sigma^*(\omega)\,A) + Z_e =$$
$$[d / \sigma\,(1 + j\,\omega\varepsilon_0\,\varepsilon / \sigma)A] + 2\,[\,(r_e - j / \omega\,c_e)A] \qquad \textbf{(3)}$$

being A the electrode area, r_e and c_e the electrode polarization resistance and capacitance per unit area.

The measured capacitance, C_p, can be related to the capacitance of the cell containing the sample, C_s, and the capacitance associated with the electrodes, C_e, through the relation

$$1/C_p = 1/C_e + [\omega^2\,\varepsilon_0\,\varepsilon(\omega)/\sigma(\omega)] / C_s, \qquad \textbf{(4)}$$

in the low frequency limit.

Results And Discussion

The measured values of R and X are coincident with those calculated from Z and θ measurements as:

$$R = Z\,\mathrm{sen}\theta \quad \text{and} \quad X = Z\cos\theta \qquad \textbf{(5)}$$

For both oils, the R, X and $1/C_p$ values at fixed frequency are linear functions of electrode distance as shown in Figures 1, 2 and 3 (data presented for C oil only), that is R and X data can be analytically represented by:

$$R = m_R\,d + b_R \quad \text{and} \quad X = m_X\,d + b_X \qquad \textbf{(6)}$$

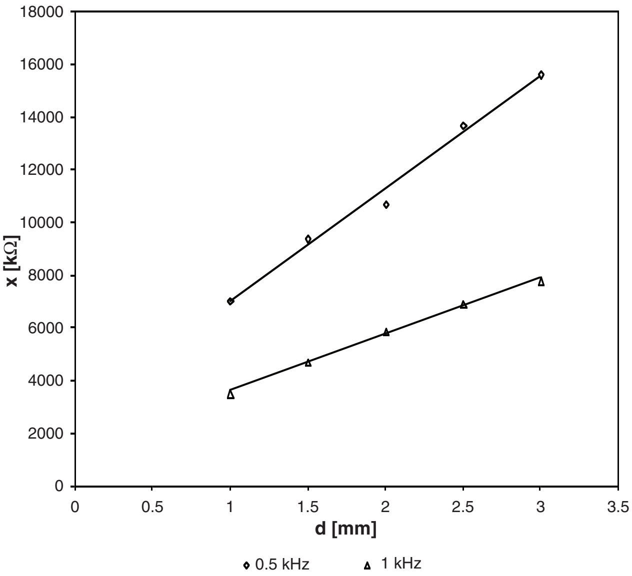

Figure 2. Reactance. X [kΩ]. vs. electrodes distance. d[mm]. of lemon oil sample C at the frequencies of 0.5 and 1.0 kHz.

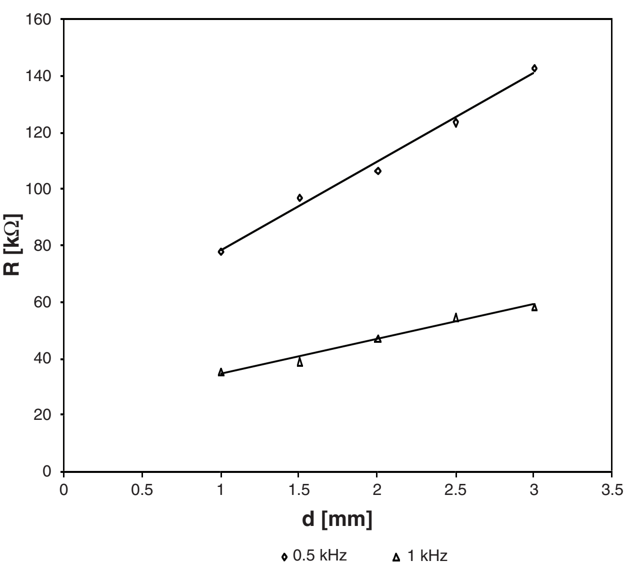

Figure 1. Resistance. R [kΩ]. vs. electrodes distance. d[mm]. of lemon oil sample C at the frequencies of 0.5 and 1.0 kHz.

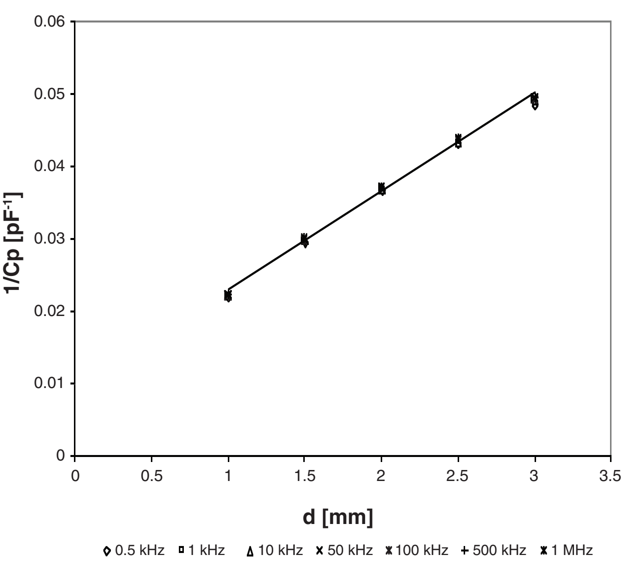

Figure 3. Inverse of capacity. 1/Cp [pF^{-1}]. vs. electrodes distance. d[mm]. of lemon oil sample C at the frequencies of 0.5 and 1.0 kHz.

After rationalizing the denominators of eq. (3) and comparing with eq. (6), the slopes m_R and m_X , can be written in term of conductivity and permittivity as:

$$m_R = 1 / \sigma [1 + (\omega\varepsilon_0 \varepsilon / \sigma)^2]A \quad (7)$$

$$m_X = (\omega\varepsilon_0 \varepsilon / \sigma) / \sigma [1 + (\omega\varepsilon_0 \varepsilon / \sigma)^2]A \quad (8)$$

with $m_R / m_X = (\omega\varepsilon_0 \varepsilon / \sigma)$.

The slopes of resistance, m_R, reactance, m_X, and inverse of capacity, $m_{1/Cp}$, have been determined using least squares fitting with r^2 equal to 0.99, 0.99, and 0.98 respectively. Finally, the permittivity and the conductivity values can be obtained from the expressions:

$$\sigma = m_R / (m_R^2 + m_X^2) A, \quad (9)$$

$$\varepsilon = m_X / (m_R^2 + m_X^2) \omega \varepsilon_0 A \quad (10)$$

which can be easily deduced by combining equations (7) and (8). The values of the slopes for both oils at different frequencies are indicated in Table I. Permittivity and conductivity vs. logarithm of the applied field frequency are represented in Figures 4 and 5.

From the analysis of the curves in Figure 4 it is deduced that the permittivity of lemon oil samples C and T were found to be very similar and practically independent of frequency. The average static permittivity values in the considered frequency range were 3.51 ± 0,03 and 3.71 ± 0,03 respectively. On the other hand, Figure 5 shows that conductivity of both oils has a strong frequency dependence changing by various orders of magnitude becoming practically equal at the greater frequencies. Taking into account that lemon oil is a complex substance composed mainly of monoterpene hydrocarbons and oxygenated monoterpenes among many other compounds, the differences in dielectric properties may well be due to a slight variation in chemical composition (in spite of the fact that they were isolated from the peel of the same lemon cultivar). In view of the permittivity of some vegetable oils - olive oil (10): 3.1 ; edible oil: 3.1(11); jojoba oil, cosmetic grade (12): 2.68; sesame oil (13): 3.0; vegetable transformer oil (14): 3.0-4.0 it can be pointed out the permittivity values of the studied lemon oil are in the same range of values.

The results also show that dielectric spectroscopy can also be a useful alternative for the characterization and differentiation of products and/or components in the citrus industry,

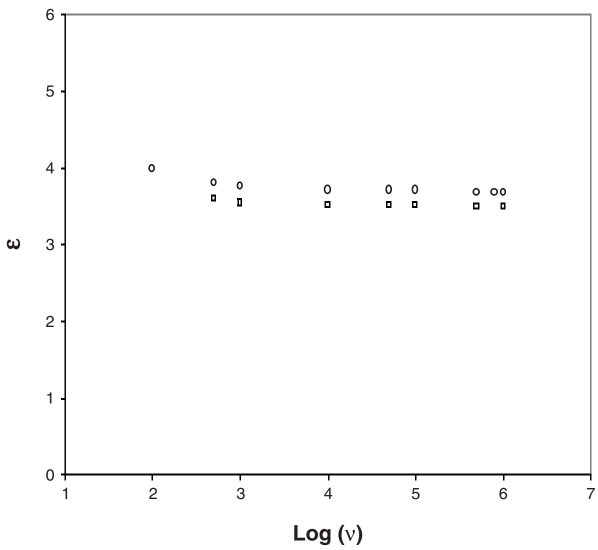

Figure 4. Permittivity. ε . vs. Log (ν) of lemon oil samples C (□) and T (○).

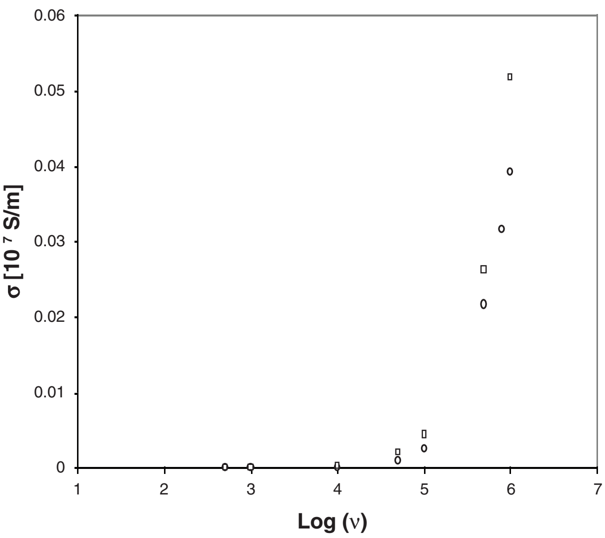

Figure 5. Conductivity. σ .10⁷ [S/m]. vs. Log (ν) of lemon oil samples C (□) and T (○).

Table I. Resistance, m_R , reactance, m_X, and inverse of capacity slopes, $m_{1/Cp}$, for lemon oil samples C and T at different frequencies.

ν (kHz)	m_R (C)	m_R (T)	m_X (C)	m_X (T)	$m_{1/Cp}$ (C)	$m_{1/Cp}$ (T)
0.5	31.34	1830.00	$1.33\ 10^{-2}$	$1.26\ 10^{-2}$	$1.30\ 10^{-2}$	$1.29\ 10^{-2}$
1.0	12.26	605.80	$1.35\ 10^{-2}$	$1.27\ 10^{-2}$	$1.30\ 10^{-2}$	$1.30\ 10^{-2}$
10.0	1.10	8.00	$1.36\ 10^{-2}$	$1.29\ 10^{-2}$	$1.30\ 10^{-2}$	$1.30\ 10^{-2}$
50.0	$2.01\ 10^{-1}$	$4.67\ 10^{-1}$	$1.36\ 10^{-2}$	$1.29\ 10^{-2}$	$1.30\ 10^{-2}$	$1.30\ 10^{-2}$
100.0	$9.60\ 10^{-2}$	$1.64\ 10^{-1}$	$1.36\ 10^{-2}$	$1.29\ 10^{-2}$	$1.30\ 10^{-2}$	$1.30\ 10^{-2}$
500.0	$1.60\ 10^{-2}$	$1.90\ 10^{-2}$	$1.37\ 10^{-2}$	$1.30\ 10^{-2}$	$1.30\ 10^{-2}$	$1.30\ 10^{-2}$
800.0	$1.10\ 10^{-2}$	$1.10\ 10^{-2}$	$1.37\ 10^{-2}$	$1.30\ 10^{-2}$	$1.30\ 10^{-2}$	$1.30\ 10^{-2}$
1000.0	$8.00\ 10^{-3}$	$1.30\ 10^{-2}$	$1.37\ 10^{-2}$	$1.30\ 10^{-2}$	$1.30\ 10^{-2}$	$1.30\ 10^{-2}$

as described by Martinez et al (15). In particular, dielectric properties play an important role in the capacitor and transformer industries since, compared with other vegetable oils, lemon oil shows a superior biodegrability, lower toxicity, higher flash and fire point and better heat conductivity. This industry is looking for vegetable-based insulating oil as substitute of mineral-based oils since they offer potential benefits of safety, environmental health compared to conventional oils with permittivity values ranging from 2.5 to 3.5 at 25ºC (16). The permittivity of lemon oil leads us to speculate that it would have a higher breakdown voltage than other vegetable oils and would be similar to some mineral oils. Further measurements are necessary for confirmation.

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