Complex dielectric permittivity of rum and its mixtures with methanol, ethanol, and water at frequencies up to 15 GHz

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Complex dielectric permittivity of rum and its mixtures with methanol, ethanol, and water at frequencies up to 15 GHz

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ABSTRACT
An analysis of the complex dielectric permittivity of rum ‘Havana Club Añejo 3 Años’ and its mixtures with methanol, ethanol, and deionized water is presented in this work. Measurements are carried out using the open-ended coaxial probe technique within a frequency range from 500 MHz to 15 GHz. Curves of the real and imaginary parts as well as the permittivity spectra (Cole–Cole diagram) and relaxation parameters of the liquids-under-test are presented. Distribution of the relaxation parameters for the different inclusion levels is analyzed and modelled. Comparison between the measured data and permittivity mixture theory is carried out; comments on the accuracy of each are discussed. A validation of the measurement process is realized by means of an evaluation of the experimental and literature dielectric permittivity values.

KEYWORDS
Complex dielectric permittivity; microwave measurements; coaxial probe technique; Cole–Cole relaxation; binary mixture

1. Introduction
The complex dielectric permittivity $\varepsilon^* = \varepsilon' - j\varepsilon''$, being $\varepsilon'$ and $\varepsilon''$ real and imaginary parts, respectively, represents a constitutive parameter of all dielectric material, and practically, it rules the interaction of the matter under an incident electromagnetic (EM) field (Chen et al. 2004). As each dielectric material has a unique permittivity coefficient $\varepsilon^*$ (Agilent Technologies 2006), its electromagnetic characterization provides useful information about its properties.

Particularly, at RF and microwave frequency ranges, several methods and techniques have been developed and proposed for estimation of the dielectric constant of solid and liquid materials (Chen et al. 2004); among them, the coaxial probe is one of the most used due to its simplicity and wideband capability (Boughriet et al. 1999; Zajiček et al. 2006; Bobowski and Johnson 2012). It consists of an open-ended coaxial line which is in contact with the sample under test, allowing the EM fields interact with the material; thus, the dielectric properties can be inferred from measurements of the reflection coefficient of the coaxial line (Ghannouchi and Bosisio 1989; Chen et al. 2004).

In practical applications, the measurement of the dielectric constant of materials at RF/microwave frequencies has been employed as a relevant technique for properties...
For material characterization, dielectric permittivity models have been proposed as a useful tool for representing the $\varepsilon^*$ spectra at continuous frequency ranges; among them, the Debye equation and Cole–Cole plots are the most used due to their simplicity and illustration capability of the material dielectric characteristics, such as relaxation-time $\tau$, static-permittivity $\varepsilon_s$, and infinite-frequency permittivity $\varepsilon_{\infty}$ (Watanabe et al. 2009; Kataria et al. 2017). However, for some alcohols and water mixtures, more complex models are used in order to obtain accuracy, particularly at the higher frequencies; in Sato and Buchner (2004), an analysis on the dielectric relaxation processes of ethanol and water mixtures is carried out from 100 MHz to 90 GHz, comparing relaxation models (Debye, Cole–Cole, Davidson–Cole) and including high-frequency Debye terms; it is important to mention that, for such a wide bandwidth analysis, combined techniques are used, including time-domain-reflectometry for the low-frequency band, and Ku-, A-, and E-band waveguide interferometers for the high-frequency band (Sato and Buchner 2004).

In this work, the complex dielectric permittivity $\varepsilon^*$ of rum ‘Havana Club Añejo 3 Años’ (Cuban production), and mixtures with methanol, ethanol, and deionized water in several concentrations, is obtained at a frequency range from 500 MHz to 15 GHz, by using an open-ended coaxial probe. Relations between the several concentrations, mixtures, and $\varepsilon$ values are found and discussed. Results from mixture theory are obtained and compared with the experimental data. Cole–Cole relaxation plots are used to characterize the samples, showing a relation between the relaxation parameters $\tau$, $\varepsilon_s$, $\varepsilon_{\infty}$ and the different mixture concentrations. As the proposal of this work is for a fast and accurate sample-purity analysis based on instantaneous and single-method measurements up to 15 GHz, limits on the higher frequency components, concentration levels, as well as differences between experimental, modelled and predicted results are established and commented. The obtained results and procedure have the potential of being used for liquid-purity analysis of other alcoholic beverages. The substances ethanol, methanol and water are proposed in this study as they are usually used in the illegal manipulation of alcoholic beverages.
The paper is organized as follows: Section 2 describes the used setup and method of measurement; Section 3 shows the experimental results, modelled and theoretical values as well as comparisons between them; discussion of the results is stated in Section 4.

2. Measurement method

2.1. Equipment and data analysis

The used measurement setup is shown in Figure 1. A Keysight PNA-X series Vector Network Analyzer (VNA) and the 85070-E model coaxial probe are used for the measurements; the coaxial probe is directly connected to the VNA’s port 1 through a coaxial cable. A standard one-port SOL (short-open-load) calibration at plane B–B’ is realized (CASCADE Microtech); a second SOL calibration is carried out at plane A–A’ of the coaxial probe, using deionized water as the load standard (Agilent 2013).

For the processed data, the setup is configured to operate from 500 MHz to 15 GHz; curves of \( \varepsilon' \) and \( \varepsilon'' \) are obtained by using the 85070E-Keysight software included in the coaxial-probe kit (Agilent 2013) directly from the measurements; each liquid-under-test (LUT) is measured five times at a controlled sample temperature of 22 °C, and the results are averaged. The total volume of each LUT is of 20 mL, and immersion of the probe end is of 1.5 cm below the LUT level, as shown in Figure 1(b), ensuring enough EM-fields/material interaction for the accuracy of the measurements (Agilent 2013).

The Cole–Cole spectra (Chen et al. 2004) of each sample are generated by smoothing and combining the \( \varepsilon' \) and \( \varepsilon'' \) curves; relaxation-time \( \tau \) is obtained directly from the smoothed \( \varepsilon'' \) curves. The parameters static- and infinite-frequency permittivity, \( \varepsilon_s \) and \( \varepsilon_\infty \), respectively, are defined from the Cole–Cole relaxation equation

\[
\varepsilon' = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + (j\omega\tau)^{1-\alpha}},
\]

where \( \omega \) is the angular frequency and the exponential \([1 - \alpha]\) displays the Cole–Cole-like relaxation effect (Tuncer et al. 2002). When \( \alpha = 0 \) (sometimes occurring for simple

![Figure 1](image-url). The used experimental setup: (a) equipment and (b) details of the sample measurement.
substances or highly diluted mixtures with non-absorbent components (Tuncer et al. (2002)), (1) becomes into the single Debye relaxation
\[
\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + j\omega\tau}.
\]

2.2. Preparation of the samples

The analyzed samples consist of rum ‘Havana Club Añejo 3 Años’ as the base (matrix) of all the mixtures with methanol, ethanol, and deionized water as inclusions; purity of methanol and ethanol samples is >99%. Table 1 tabulates the composition of each LUT. Concentration levels are proposed to be low, up to X = 15 (15%) for all the inclusion substances.

3. Results and discussion

3.1. Analysis of the pure substances

The obtained dielectric permittivity curves of the pure-substances rum, methanol, ethanol and water (LUT’s R, A, B, and C, respectively) are presented in Figure 2; the resultant Cole–Cole spectra and their correspondent relaxation model curves are shown in Figure 3.

As observed from Figure 2, the lowest and highest permittivity values, in both real and imaginary parts, correspond to ethanol and deionized water, respectively; rum sample presents higher permittivity values compared with pure methanol, but lower than that of water. In a related manner, from Figure 3, it is appreciated that the lowest \(\varepsilon_s\) can be predicted for the ethanol sample; while the highest \(\varepsilon_s\) is observed for water. Relaxation frequency \(f_r\) (and thus relaxation time \(\tau = 1/2\pi f_r\)) of rum, methanol and ethanol can be obtained directly from the summit of the \(\varepsilon''\) curves within the proposed frequency range; that of water is expected to occur at frequencies slightly higher than 15 GHz (out of the limit of the proposed experimental measurements); nevertheless, it can be predicted from its relaxation model. It can be appreciated that the pure-rum sample (LUT R) exhibits a non-Debye relaxation that can be accurately modelled using the Cole–Cole equation (1);

<table>
<thead>
<tr>
<th>LUT</th>
<th>Subs. 1 (mL)</th>
<th>Subs. 2 (mL)</th>
<th>X</th>
</tr>
</thead>
<tbody>
<tr>
<td>R</td>
<td>20 rum</td>
<td>–</td>
<td>0</td>
</tr>
<tr>
<td>A</td>
<td>20 methanol</td>
<td>–</td>
<td>0</td>
</tr>
<tr>
<td>B</td>
<td>20 ethanol</td>
<td>–</td>
<td>0</td>
</tr>
<tr>
<td>C</td>
<td>20 water</td>
<td>–</td>
<td>0</td>
</tr>
<tr>
<td>A05</td>
<td>19 rum(^a)</td>
<td>1 methanol(^b)</td>
<td>0.05</td>
</tr>
<tr>
<td>A10</td>
<td>18 rum(^a)</td>
<td>2 methanol(^b)</td>
<td>0.10</td>
</tr>
<tr>
<td>A15</td>
<td>17 rum(^a)</td>
<td>3 methanol(^b)</td>
<td>0.15</td>
</tr>
<tr>
<td>B05</td>
<td>19 rum(^a)</td>
<td>1 ethanol(^b)</td>
<td>0.05</td>
</tr>
<tr>
<td>B10</td>
<td>18 rum(^a)</td>
<td>2 ethanol(^b)</td>
<td>0.10</td>
</tr>
<tr>
<td>B15</td>
<td>17 rum(^a)</td>
<td>3 ethanol(^b)</td>
<td>0.15</td>
</tr>
<tr>
<td>C05</td>
<td>19 rum(^a)</td>
<td>1 water(^b)</td>
<td>0.05</td>
</tr>
<tr>
<td>C10</td>
<td>18 rum(^a)</td>
<td>2 water(^b)</td>
<td>0.10</td>
</tr>
<tr>
<td>C15</td>
<td>17 rum(^a)</td>
<td>3 water(^b)</td>
<td>0.15</td>
</tr>
</tbody>
</table>

\(^a\)Matrix.
\(^b\)Inclusion of the mixture.
this is thought to be due to its complex composition nature. The simple Debye relaxation (2) fits well for the pure substances methanol, ethanol and water (Megriche et al. 2012). Relaxation frequency $f_r$, relaxation time $\tau$, static- and infinite-frequency permittivities of these pure substances are reported in Table 2.

![Figure 2. Measured dielectric permittivity of LUT's R, A, B and C: (a) real part and (b) imaginary part.](image)

![Figure 3. Measured $\varepsilon''$ spectra and modelled relaxation curves of LUT's R, A, B, and C.](image)

<p>| Table 2. Relaxation parameters of the measured samples. |
|-------------|-------------|-------------|-------------|-------------|--------|</p>
<table>
<thead>
<tr>
<th>LUT</th>
<th>$\varepsilon'$</th>
<th>$\varepsilon''_r$</th>
<th>$\varepsilon''_\infty$</th>
<th>$\tau$ (ps)</th>
<th>$f_r$ (GHz)</th>
<th>$1 - \alpha$</th>
</tr>
</thead>
<tbody>
<tr>
<td>R</td>
<td>61.13</td>
<td>25.18</td>
<td>7.08</td>
<td>29.12</td>
<td>5.47</td>
<td>0.9550</td>
</tr>
<tr>
<td>A</td>
<td>33.50</td>
<td>13.47</td>
<td>6.57</td>
<td>55.72</td>
<td>2.86</td>
<td>–</td>
</tr>
<tr>
<td>B</td>
<td>24.79</td>
<td>10.21</td>
<td>4.37</td>
<td>184.53</td>
<td>0.86</td>
<td>–</td>
</tr>
<tr>
<td>C</td>
<td>80.75</td>
<td>37.57</td>
<td>5.62</td>
<td>8.75</td>
<td>18.19</td>
<td>–</td>
</tr>
<tr>
<td>R05</td>
<td>60.35</td>
<td>24.86</td>
<td>7.03</td>
<td>30.11</td>
<td>5.29</td>
<td>0.9558</td>
</tr>
<tr>
<td>A010</td>
<td>58.39</td>
<td>23.98</td>
<td>7.25</td>
<td>31.18</td>
<td>5.10</td>
<td>0.9592</td>
</tr>
<tr>
<td>A15</td>
<td>57.85</td>
<td>23.71</td>
<td>7.24</td>
<td>32.09</td>
<td>4.96</td>
<td>0.9586</td>
</tr>
<tr>
<td>B05</td>
<td>59.38</td>
<td>24.34</td>
<td>6.78</td>
<td>30.53</td>
<td>5.21</td>
<td>0.9508</td>
</tr>
<tr>
<td>B10</td>
<td>57.41</td>
<td>23.38</td>
<td>6.88</td>
<td>32.33</td>
<td>4.92</td>
<td>0.9507</td>
</tr>
<tr>
<td>B15</td>
<td>55.28</td>
<td>22.36</td>
<td>7.22</td>
<td>34.63</td>
<td>4.60</td>
<td>0.9544</td>
</tr>
<tr>
<td>C05</td>
<td>62.46</td>
<td>25.85</td>
<td>6.91</td>
<td>27.82</td>
<td>5.72</td>
<td>0.9543</td>
</tr>
<tr>
<td>C10</td>
<td>63.55</td>
<td>26.34</td>
<td>6.89</td>
<td>26.64</td>
<td>5.97</td>
<td>0.9537</td>
</tr>
<tr>
<td>C15</td>
<td>64.60</td>
<td>26.89</td>
<td>6.92</td>
<td>25.26</td>
<td>6.30</td>
<td>0.9555</td>
</tr>
</tbody>
</table>

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3.2. Analysis of the mixtures

Figures 4–6 show the measured permittivity curves of samples $A_{05}$ to $A_{15}$ (rum with methanol), $B_{05}$ to $B_{15}$ (rum with ethanol) and $C_{05}$ to $C_{15}$ (rum with deionized water), respectively, which correspond to the different rum-based mixtures going from inclusion levels $X = 0.05$ to 0.15 (Table 1).

As can be observed from Figures 4 and 5, mixtures of rum with methanol and ethanol show that real and imaginary permittivities are decreasing as the concentration level is increasing; $A_{05}$ and $B_{05}$, correspondent to concentration $X = 0.05$, present higher $\varepsilon^*$ values compared to those of $X = 0.1$ and 0.15.

On the other hand, as noticed in Figure 6(a), the real permittivity tends to increase as the water content is increasing; the $C_{05}$ curve, representing the lowest concentration level $X = 0.05$ remains below the $C_{10}$ and $C_{15}$ curves with the higher presence of water. Particularly, for the imaginary permittivity, there exists a frequency region, below 3.58 GHz, where $\varepsilon''$ tends to decrease as the water content is increasing, inversely to the higher

![Figure 4](image1.png)

![Figure 5](image2.png)

**Figure 4.** Measured dielectric permittivity of LUT’s $A_{05}$–$A_{15}$: (a) real part and (b) imaginary part.

**Figure 5.** Measured dielectric permittivity of LUT’s $B_{05}$–$B_{15}$: (a) real part and (b) imaginary part.
frequencies, as seen in the inset of Figure 6(b). Such an effect is to be expected, as $\varepsilon''$ of rum tends to be higher than that of water at the lower frequencies, as seen in Figure 2(b).

Figure 7 shows the $\varepsilon^*$ spectra of the measured mixtures and their correspondent Cole–Cole relaxation model using (1) (LUT’s $A_{05}$ to $C_{15}$). The reason of using (1) instead of (2)
for modelling the mixtures is that the rum sample (matrix substance) exhibits a Cole–Cole relaxation instead of a Debye; therefore, the proposed combinations are expected to follow a Cole–Cole behaviour.

Results in Figure 7 show the trend in dielectric constant for each of the mixtures and concentrations levels. Particularly, $\tau$ increases and $\varepsilon_s$ decreases while $X$ increases for LUT’s $A_{05}$ to $B_{15}$ (rum with methanol and with ethanol); an inverse effect in $\tau$ and $\varepsilon_s$ is observed for LUT’s $C_{05}$ to $C_{15}$, corresponding to rum with water. Values of $\varepsilon_\infty$ tend to converge around a value of 7 for all the measured LUT’s. The relaxation parameters $\varepsilon_s$, $\varepsilon''_r$ [imaginary permittivity at which relaxation occurs, $\varepsilon''(fr)$], $\varepsilon_\infty$, $\tau$, $fr$ (relaxation frequency), and $[1 - \alpha]$ are tabulated in Table 2. Although $\varepsilon''_r$ is not commonly used in this type of analysis, the authors consider that it shows valuable information for the different $X$ levels of the mixtures, as seen in the next section.

3.3. Dielectric permittivity modelling of the mixtures

As previously observed in Sections 3.1 and 3.2, a complete $\varepsilon^*$ spectra for all the analyzed samples can be modelled using the Cole–Cole relaxation equation (1) from the experimental measurements carried out from 500 MHz to 15 GHz. The relaxation parameters $\varepsilon_s$, $\varepsilon''_r$, and $\varepsilon_\infty$ of the measured samples are plotted in Figure 8; curve fitting of the parameters is included.

![Figure 8. Dielectric relaxation parameters of the samples: (a) $\varepsilon_s$, (b) $\varepsilon''_r$, and (c) $\varepsilon_\infty$.](image-url)
Although it is known that the distribution of $\varepsilon^*$ is not trivial for the different inclusion levels of binary mixtures with substances as water, methanol and ethanol (Megriche et al. 2012), it can be noticed from Figure 8(a,b) that the parameters $\varepsilon_s$ and $\varepsilon''_r$ follow a linear trend within the proposed $X$ range between 0 and 0.15, for all the analyzed samples; mixtures with methanol and ethanol exhibit negative slopes for $\varepsilon_s$ and $\varepsilon''_r$, being those of ethanol the lowest values, while mixtures with water show positive slopes for the mentioned parameters. Results of $\varepsilon_\infty$ depicted in Figure 8(c) show a nonlinear behaviour which is attributed to two main reasons: (1) the distribution of $\varepsilon^*$ at high frequencies is not linear; in fact, from data reported in Smith et al. (1997), mixtures with different concentration levels of water and methanol exhibit a parabolic-like trend for $\varepsilon_\infty$; and (2) there exist higher order relaxation components at higher frequencies involved in the mixtures (Sato and Buchner 2004); however, as their contribution is small and they are expected to occur above 15 GHz, they are not taken into account in model (1), incurring in higher differences compared with the lower frequency relaxation parameters $\varepsilon_s$ and $\varepsilon''_r$. Additionally, deviation in preparation of the samples can be seen as a source of uncertainty; however, the well-defined variation in $\varepsilon_s$ and $\varepsilon''_r$ evidence that it represents a minor contribution for the characterization.

As shown in Figure 8, $\varepsilon_s$ and $\varepsilon''_r$, are modelled following a linear trend in the form of an $ax + b$ polynomial, while $\varepsilon_\infty$ adopts the form of a second-order polynomial $ax^2 + bx + c$. The correspondent polynomial coefficients for each modelled parameter are tabulated in Table 3.

From the presented results, it is observed that a Cole–Cole estimation of the $\varepsilon^*$ spectra of rum mixed with methanol, ethanol or water, for any value of $X \leq 0.15$, can be obtained with high accuracy by following the relaxation-parameter behaviour (Figure 8), where $\varepsilon_s$ and $\varepsilon''_r$ are linearly modelled, and $\varepsilon_\infty$ is adjusted by a second-order polynomial, which is expected to fit for the proposed inclusion levels (as noticed from results of the analyzed mixtures reported in Smith et al. (1997)). Figure 9 shows the measured $\varepsilon^*$ spectra and the predicted modelling of the mixtures using the Cole–Cole relaxation (1) based on curve modelling of the parameters of Figure 8. For the sake of comparison, results obtained from the binary mixture theory proposed by Steeman and Maurer (1990) are added:

$$e_m = e_1 + 2e_2 + 2X(e_2 - e_1) 2e_1 + e_2 - X(e_2 - e_1),$$

where $e_m$ is the resultant complex dielectric permittivity of the mixture, $e_1$ and $e_2$ are matrix and inclusion complex dielectric permittivities, respectively, which are taken directly from the results of Section 3.1. The reason in the choice of (3) is because it presents the highest accuracy compared with other analytical solutions, as stated in Tuncer et al. (2001), for low concentration mixtures around $X = 0.1$. Results using (3) are

<table>
<thead>
<tr>
<th>Param.</th>
<th>Rum/methanol</th>
<th>Rum/ethanol</th>
<th>Rum/water</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon_s$</td>
<td>$a = -23.86$, $b = 61.18$</td>
<td>$a = -39.18$, $b = 61.21$</td>
<td>$a = 22.94$, $b = 61.02$</td>
</tr>
<tr>
<td>$\varepsilon''_r$</td>
<td>$a = -10.52$, $b = 25.22$</td>
<td>$a = -18.70$, $b = 25.23$</td>
<td>$a = 11.16$, $b = 25.22$</td>
</tr>
<tr>
<td>$\varepsilon_\infty$</td>
<td>$a = 6.00$, $b = 2.14$, $c = 7.13$</td>
<td>$a = -15.00$, $b = 3.23$, $c = 7.07$</td>
<td>$a = -15.00$, $b = 1.95$, $c = 7.02$</td>
</tr>
</tbody>
</table>
obtained from 500 MHz to 15 GHz, as the matrix and inclusion substances have been measured at the same frequency range.

As noticed in Figure 9, the dielectric relaxation plots from the modelled relaxation parameters are in excellent agreement with the measured data; the permittivity values calculated using (3) present higher deviation from the measured results; moreover, as the inclusion levels increase, so is the difference between the Steeman & Maurer proposal and the measured mixtures. Differences (in percentage) of the modelled relaxation parameters between the measurements and the estimated values are tabulated in Table 4; comparison with the parameters obtained using (3) are also included; however, as they are frequency-limited (500 MHz to 15 GHz), only $\varepsilon_\infty$, $f_r$, and $\tau$ are used for the calculations.

It can be appreciated from Table 4 that, differences of the modelled parameters between the measured samples and the fitted data are below 1% for $\varepsilon_s$, $\varepsilon''_r$, and $[1 - \alpha]$; maximum discrepancies of 1.05% and 1.07% are obtained for $\tau$ and $f_r$, respectively, and higher variations, up to 6.13%, are obtained for $\varepsilon_\infty$. Particularly, for $\varepsilon_s$ and $\varepsilon''_r$, the largest discrepancies are for the rum/methanol mixture (LUT’s $A_{05}$ to $A_{15}$), having average deviations of 0.57% and 0.56%, respectively, while the lowest differences for the same parameters are for the rum/water combinations ($C_{05}$ to $C_{15}$), with vales of 0.11% and 0.10%. For

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**Figure 9.** Measured, predicted, and theoretical (Steeman and Maurer 1990) relaxation spectra of the mixtures: (a) $A_{05}$–$A_{15}$, (b) $B_{05}$–$B_{15}$, and (c) $C_{05}$–$C_{15}$.
The largest differences are found for the rum/ethanol mixtures (LUT’s $B_{05}$ to $B_{15}$), in which an average deviation of 3.82% is obtained, while the lowest occurs for the composites of rum and deionized water, with average discrepancy of 1.97%.

Differences taking into account the measured samples and the estimation using (3) are considerably higher than those obtained with the relaxation-parameter polynomial model. The highest differences in $e_{\infty}$ are obtained with the rum/ethanol mixtures (LUT’s $B_{05}$ to $B_{15}$), having average deviation of 4.94%, while the lowest are with the rum/methanol ($A_{05}$ to $A_{15}$) samples with average deviation of 2.82%. For $t$ and $f_r$, the largest percentage differences are also obtained for LUT’s $B_{05}$ to $B_{15}$, with average values of 7.14% and 7.86%, respectively. In contrast, very low values of errors are found for mixtures of rum and deionized water ($C_{05}$ to $C_{15}$), with average deviations of 0.20% in both $t$ and $f_r$. Additionally, as noticed from Figure 9 and Table 4, the errors in $e^\infty$, between the measurements and the values predicted by (3), increase as the inclusion concentration increases.

### 3.4. Validation of the measurement technique

In order to validate the experimental measurement process used in this work, a comparison in $e^\infty$ is carried out using the measured permittivity of pure methanol and ethanol, and that from Gregory and Clarke (2012) at a mean temperature of 22.5 °C; as the available data in Gregory and Clarke (2012) ranges only from 100 MHz to 5 GHz, the comparison is made from 500 MHz to 5 GHz only. Figure 10 plots the reported (Gregory and Clarke 2012) and measured $e^\infty$ data.

As observed from Figure 10, both measured and reported results are in excellent agreement. The major differences for methanol are of 3.3% and 6.3% for $e'$ and $e''$, respectively, at 5 GHz both, while for ethanol, 3.4% at 1.52 GHz and 6.1% at 5 GHz for $e'$ and $e''$, respectively, are noticed as the maximum deviation values.

### 4. Comments and discussion

Based on the presented results, some important comments can be formulated. First, the Cole–Cole relaxation model (1) was useful for the rum, and consequently, the prepared mixtures, while the pure substances methanol, ethanol and deionized water are best modelled by using the Debye equation (2).

<table>
<thead>
<tr>
<th>LUT</th>
<th>$\Delta e_1$</th>
<th>$\Delta e''$</th>
<th>$\Delta e_{\infty}$</th>
<th>$\Delta \tau$</th>
<th>$\Delta f_r$</th>
<th>$\Delta [1 - \alpha]$</th>
<th>$\Delta e_1$</th>
<th>$\Delta e''$</th>
<th>$\Delta \tau$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_{05}$</td>
<td>0.59</td>
<td>0.65</td>
<td>3.19</td>
<td>0.05</td>
<td>0.09</td>
<td>0.26</td>
<td>2.17</td>
<td>1.33</td>
<td>1.42</td>
</tr>
<tr>
<td>$A_{10}$</td>
<td>0.70</td>
<td>0.80</td>
<td>2.15</td>
<td>0.19</td>
<td>0.20</td>
<td>0.20</td>
<td>2.04</td>
<td>2.08</td>
<td>2.08</td>
</tr>
<tr>
<td>$A_{15}$</td>
<td>0.42</td>
<td>0.27</td>
<td>4.81</td>
<td>0.09</td>
<td>0.18</td>
<td>0.59</td>
<td>4.26</td>
<td>2.12</td>
<td>2.24</td>
</tr>
<tr>
<td>$B_{05}$</td>
<td>0.22</td>
<td>0.21</td>
<td>6.13</td>
<td>1.02</td>
<td>1.01</td>
<td>0.56</td>
<td>2.83</td>
<td>3.34</td>
<td>3.41</td>
</tr>
<tr>
<td>$B_{10}$</td>
<td>0.21</td>
<td>0.11</td>
<td>5.30</td>
<td>1.05</td>
<td>1.07</td>
<td>0.57</td>
<td>5.05</td>
<td>6.87</td>
<td>7.46</td>
</tr>
<tr>
<td>$B_{15}$</td>
<td>0.09</td>
<td>0.27</td>
<td>0.02</td>
<td>0.38</td>
<td>0.30</td>
<td>0.08</td>
<td>6.93</td>
<td>11.20</td>
<td>12.70</td>
</tr>
<tr>
<td>$C_{05}$</td>
<td>0.18</td>
<td>0.27</td>
<td>2.50</td>
<td>0.27</td>
<td>0.17</td>
<td>0.17</td>
<td>2.13</td>
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<td>0.00</td>
</tr>
<tr>
<td>$C_{10}$</td>
<td>0.08</td>
<td>0.00</td>
<td>2.58</td>
<td>0.14</td>
<td>0.06</td>
<td>0.27</td>
<td>3.38</td>
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</tr>
<tr>
<td>$C_{15}$</td>
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<td>0.03</td>
<td>0.83</td>
<td>0.78</td>
<td>0.80</td>
<td>0.03</td>
<td>4.69</td>
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</tr>
</tbody>
</table>

All values are expressed in percentage.
Regarding the relaxation parameters used for modelling the experimental $\varepsilon''$ spectra of the rum-based mixtures, the proportional shift in $\varepsilon_s$ and $\varepsilon''_r$ tends to be linear for low inclusion levels, from $X = 0$ to 0.15, while that of $\varepsilon_{\infty}$ does not. Following adjusted curves of these parameters, first-order polynomial for $\varepsilon_s$ and $\varepsilon''_r$ and second-order for $\varepsilon_{\infty}$, an estimation of any other value of $X$ within the mentioned values can be obtained.

Theoretical results obtained by the Steeman & Maurer mixture model (3) (Steeman and Maurer 1990) are included for comparison; however, the estimated curves from the relaxation-parameter curve fitting offers higher agreement when compared with the experimental data. From the several mixture models studied in Tuncer et al. (2001), (3) is chosen because it seems to offer the highest accuracy for low-concentration mixtures.

The presented results can be useful for a fast and simple quality estimation of other alcoholic beverages without the use of complex models and methods, only by knowing the $\varepsilon''$ spectra or the relaxation parameters of each alcoholic beverage in its pure state. Specifically, from measurements of an unknown binary mixture, its concentration level $X$ (lying between 0 and 0.15) can be easily obtained graphically following the parameter models (Figure 8 and Table 3) if the inclusion substance is known; on the other hand, the qualitative identification of the inclusion (methanol, ethanol or water) can be known if the concentration level $X$ (any value between 0 and 0.15) of the measured mixture is given, following the relaxation-parameter graphics of Figure 8 or the analytical calculation using values of Table 3. The inclusion levels $X$ are proposed low as, usually, alcoholic beverages are modified by adding other substances in small amounts (Nascimento et al. 2011).

5. Conclusions

The complex dielectric permittivity of a Cuban-production rum and its mixtures with methanol, ethanol, and deionized water at low inclusion levels has been presented. A coaxial probe is used for the measurements at frequencies up to 15 GHz. The basic Cole–Cole and Debye relaxation models have been used to characterize the measured samples. Estimation of the relaxation spectra of the proposed mixtures has been obtained based on the modelled distribution of the measured relaxation parameters; the estimated results are
compared with the measured data, being in excellent agreement. The theoretical approach by Steeman & Maurer is included, showing higher deviation levels with the measured data. Finally, validation in the accuracy of the measurements has been presented up to 5 GHz by means of comparison of the complex permittivity of methanol and ethanol between the measurements and reported data.

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**Disclosure statement**

No potential conflict of interest was reported by the authors.

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