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Full Length Article

Permittivity of diesel fossil fuel and blends with biodiesel in the full range from 0% to 100%: Application to biodiesel content estimation

J. Corach [a], P.A. Sorichetti [b], S.D. Romano [a,*]

[a] Universidad de Buenos Aires, CONICET, Instituto de Tecnología del Hidrógeno y Energías Sostenibles (ITHES), Facultad de Ingeniería, Departamento de Ingeniería Mecánica, Grupo de Energías Renovables (GER), Buenos Aires, Argentina
[b] Universidad de Buenos Aires, Facultad de Ingeniería, Departamento de Física, Laboratorio de Sistemas Líquidos (LSL), Buenos Aires, Argentina

HIGHLIGHTS

- The permittivity of diesel fuel and blends with biodiesel was determined at 100 kHz.
- The measurement uncertainty was below 1%.
- Linear dependences with temperature and composition were found.
- Composition is estimated from permittivity and temperature measurements.
- The RMS uncertainty of biodiesel content estimation is below 2.5%.

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ABSTRACT

The relative permittivity of diesel fossil fuel and blends with biodiesel from soybean, in the full range from pure diesel to 100% biodiesel, was determined at temperatures between 298.0 K and 333.0 K (controlled within ±0.1 K), using an airtight cell. Measurements were made in the frequency range from 1 kHz to 100 kHz; this frequency range is suitable for the use of low-cost, portable equipment and also for the development of automotive sensors. The relative uncertainty of the measurements was below 1%.

Experimental values of permittivity were satisfactorily fitted to a simple model as a function of temperature and composition. The RMS uncertainty of the fitting was 1.2%. The model parameters were determined from experimental results and verified by multiple regression analysis, with very good agreement.

In addition, a model was proposed to estimate the composition of diesel/biodiesel blends from permittivity and temperature measurements. The parameters of the model were obtained by a multiple regression analysis; the RMS uncertainty of the composition estimation was below 2.5%.

The results presented in this work describe accurately the dependence of the permittivity of diesel fuel with temperature and also validate and extend previously reported models for biodiesel-rich blends with diesel fossil fuel, allowing the estimation in the full composition range with good accuracy.

1. Introduction

Diesel fossil fuel (DF) has been widely used for many years in automotive and stationary applications. Since the last decades, the use of biodiesel (BD) as a renewable alternative [1–3], pure or in blends with DF, has increased rapidly all around the world due to environmental concerns, including the reduction of carbon dioxide emissions. In consequence, there is renewed interest in the development of low-cost, fast and accurate liquid fuel characterization techniques that can be adapted to small-scale production plants, distribution points and field measurements, particularly in emerging markets. Dielectric [2,4] and ultrasonic [5,6] techniques are promising alternatives in that direction. In particular, dielectric spectroscopy has been successfully used for the production and characterization of BD [2] and also to characterize feedstocks from different origins [7,8]. They have also been used to detect alcohol in the light phase after transesterification [9], during the purification process and in the final product

* Corresponding author at: ITHES (UBA – CONICET) – GER (Grupo de Energías Renovables), Facultad de Ingeniería, Universidad de Buenos Aires (UBA), Av. Paseo Colón 850, Ciudad Autónoma de Buenos Aires 1063, Argentina.
E-mail addresses: sromano@fi.uba.ar, silviadromano@gmail.com (S.D. Romano).

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[10,11], for the characterization of fatty acid methyl esters (FAME) [12,13] and to distinguish between vegetable oil and the biodiesel produced from it [6].

The determination of the composition of DF/BD blends is very important for legal, commercial and technical reasons. Legal and commercial issues include the verification of the actual BD content in DF/BD blends; for instance, legislation in many countries establishes that diesel fossil fuel (DF) must be blended with a certain amount of BD (usually up to 20%). Moreover, the blend composition impacts engine performance and emissions [14]. The composition of a DF/BD blend is usually indicated as "*Bx*", where "x" represents the percentage of BD (V/V). For example, B0 means pure DF and B100 means pure BD. The determination of *Bx* by standard methods [15] is usually expensive, time-consuming, requires trained personnel and it is not easily adaptable to real-time measurement systems. Also, they cannot be used for in situ measurements (for instance during fuel storage and transportation). On the other side, dielectric measurements are a low-cost and fast alternative, which does not require highly trained personnel.

It must be remarked that the development of dielectric characterization techniques for DF/BD blends requires the accurate measurement and modeling of the temperature dependence of the permittivity in the full composition range. Interestingly, although some works report DF permittivity values [4,11], the authors have not found in the literature explicit models of the dependence of DF permittivity on temperature.

An early work by Tat and Van Gerpen, [16], explored the use in biodiesel blends (B0 to B100) of a commercial dielectric sensor originally designed for methanol-gasoline blends; the device operated at room temperature (296 K). The uncertainty of the composition determination was ±10%; the permittivity values of the samples were not reported. Munack at al. [4] described a sensor specifically designed for the determination of the composition of DF/BD blends (B0 to B100) in automotive applications. Graphs of permittivity values were plotted for temperatures of 278 K, 303 K and 323 K, at a frequency of 1 kHz. The sensor was successfully tested in a passenger car covering more than 50,000 km of on-the road use. The precision of blend detection was ±10%. In [17], De Souza et al. reported permittivity and conductivity measurements of DF/BD blends (B0 to B10) at frequencies between 0.1 Hz and 100 kHz at room temperature. In a previous work [18] the authors studied the dielectric properties of biodiesel-rich blends with diesel fuel (B50 to B100) at temperatures between 303.0 K and 343.0 K at frequencies between 20 Hz and 2 MHz. From these results, a model for the estimation of the composition of BD rich blends from permittivity and temperature measurements was presented. The RMS uncertainty of the estimation was below 1.5% in the full temperature and composition ranges studied.

In this work, the permittivity of pure DF, BD and their blends was measured in an airtight cell, for samples in the full composition range from 0% to 100% (V/V) of BD, at temperatures from 298.0 K to 333.0 K, between 1 kHz and 100 kHz. The range of measurement frequencies in this work makes possible the use of low-cost, portable equipment and it is also useful for the development of sensors for automotive applications, using state-of-the-art electronics. From the experimental results, a model was proposed to estimate the permittivity as a function of composition and temperature with an RMS uncertainty below 1.2%. Furthermore, a simple model is proposed for the estimation of blends composition from permittivity and temperature measurements. The RMS uncertainty of the composition estimation is below 2.5% in the full range of temperatures and compositions.

The results and models presented in this work validate and extend those presented in [18] to the full range of compositions. They are relevant for the design and implementation of accurate, economical and compact measurement systems.

2. Samples and methods

2.1. Samples

All the samples were prepared with commercial BD from soybean oil provided by a local producer, certified to met standard EN 14214 [19]. Certified pure DF samples were also provided by a local producer and also complied with international standards [20]. Since the BD and DF used for sample preparation were certified by their producers, no pre-processing procedures were carried out.

2.2. Measurement system

The relative permittivity of the samples was determined from isothermal dielectric measurements between 1 kHz and 100 kHz with a LCR meter (Tonghui TH2822C). The measurement cell is airtight to prevent systematic errors due to evaporation of the sample. To minimize polarization effects, the electrodes are made of platinized platinum. During the measurement, the cell is immersed in a thermostatic bath (Lauda), allowing a very precise temperature control (within ±0.1 K) and avoiding systematic error due to thermal gradients. Fig. 1 shows the main characteristics of the experimental setup, including the circuit configuration and the cell diagram. The system was calibrated with an uncertainty below 1% using cyclohexane as a reference liquid. All the uncertainties in this work are given as one standard deviation. The measurement uncertainty of real part of permittivity, $\Delta\varepsilon_r'$, was below 1% in all cases. In this work all the uncertainties were obtained from the statistical analysis of experimental data. Relative permittivity experimental values at each frequency were obtained as the average of 3 sets of 30 repetitions each; it must be remarked that the difference in the average values of each set was well below 1%. Besides, since no statistically significant differences in the permittivity values were observed between measurements at 1 kHz, 10 kHz and 100 kHz, the results reported in this work correspond to 100 kHz. It is important to note that measurements at this frequency minimize the influence of electrodes polarization that could occur in deficiently purified FAME samples.

In all the cases presented in Section 4, the uncertainty of the linear fittings parameters (from Eqs. (1)–(3)) was from the least-squares fitting to experimental data. The overall uncertainty of the models from Eqs. (4)–(6) was estimated as the RMS error between the experimental data and the estimated values.

3. Theory

3.1. Electrical properties

The dielectric properties of the samples are described by the relative permittivity as a function of temperature and composition [2,21,22]. As it was to be expected from results in previous works [6,18], given the high purity of the samples, it was experimentally found that dielectric losses in the frequency range studied could be neglected. Also, it was found that permittivity measurements at each composition fit very satisfactorily to a linear dependence with the absolute temperature of the sample, T:

$$\varepsilon_r'(Bx_o, T) = \varepsilon_r'(Bx_o, T_o) + \frac{\delta\varepsilon_r'(Bx_o, T_o)}{\delta T}(T - T_o) \quad (1)$$

In Eq. (1), $\varepsilon_r'(Bx_o, T)$ is the relative permittivity of a sample with composition Bx_o at temperature T. The reference temperature is T_o = 318.0 K. The temperature coefficient of the relative permittivity, $\frac{\delta\varepsilon_r'(Bx_o,T_o)}{\delta T}$, is given in K^{-1} at the reference temperature, T_o, and at the reference composition, B0.