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Alternative Properties in Liquid Fuels and Blends

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Abstract

This work summarises the results of the research program at the Renewable Energy Group (GER) of the University of Buenos Aires on alternative properties for the characterization of liquid fuels. The study included fossil fuels: diesel fuel, gasoline, and methanol, and biofuels: biodiesel from different feedstocks and bioethanol. Blends of diesel fuel/biodiesel, gasoline/bioethanol, gasoline/methanol, biodiesel/butanol, and diesel fuel/biodiesel/butanol were also studied. The electrical, acoustical, and optical properties of fuels and blends were determined as a function of temperature and composition. From these results, the composition of blends was accurately estimated from measurements of permittivity and temperature. The research program included the study of correlations of the alternative properties with those indicated in the international quality standards for liquid fuels (kinematic viscosity, methanol content, flash point). These correlations make possible to verify the quality of liquid fuels with simpler and more convenient measurements in industrial settings, and also in the laboratory.

Keywords: Electrical properties; Refractive index; Speed of sound; Liquid fuels; Correlations

Introduction

The quality of liquid fuels must be strictly controlled to ensure a satisfactory performance to the user. To this end, it is necessary to verify that the measured values of all the properties included in the relevant international standards are within the allowable ranges [1].

Electrical, optical, and acoustic properties may be correlated with those included in international standards, and used to characterise liquid fuels and blends, in the different stages of production, distribution and utilization [2-5]. The techniques for the measurement of these alternative properties have several advantages over standard methods: they are fast, non-destructive, accurate, and do not require highly trained personnel [6-7]. In addition, they may be readily adapted to industrial and field applications.

Liquid biofuels have an increasing importance, particularly in blends with fossil fuels, for automotive applications [8]. Biodiesel and bioethanol are produced from biomass feedstocks, with different conversion processes. Biodiesel is obtained from the transesterification of triglycerides from vegetable oils or animal fats [9,10]. In contrast, bioethanol is obtained by the fermentation of biomass rich in carbohydrates such as saccharose, starch or cellulose [11]. To achieve a satisfactory quality of the biofuels, pre- and post-treatment processes are necessary [10].

The application of alternative properties is particularly relevant for the characterization of liquid biofuels and their blends with fossil fuels, including the detection of contaminants and adulterants [12-14], and for the application of online measurements in vehicles and automated production systems [15,16].

Alternative Properties

Electrical properties of liquids are described by microscopic parameters related to their molecular structure [17-19]:

Relative permittivity (ε_r): is a non-dimensional parameter that describes the response of the bound electrical charges in the molecules to the application of an electric field (dielectric polarizability). At low frequencies, it is originated by the polarization of the electronic charge density (electronic polarizability), the polarization due to the orientation of the molecular dipole moments (orientation polarization), and by interfacial polarization (when conductive liquids are in contact with metallic electrodes).

Refractive index (n): is a non-dimensional parameter defined as the ratio between the speed of light in vacuum (c = 299.792.458 m/s) and in the liquid. It is important to remark that, although the refractive index is used in the context of electromagnetic waves propagation, it is related to the relative permittivity at infrared (IR) and visible (VIS) frequencies, $n^2 = \varepsilon_r$. It depends mainly on the density and electronic polarization processes at the molecular scale.

Conductivity (σ): is defined as the proportionality factor between the conduction current density and the electric field. This parameter describes the macroscopic effect of the concentration and mobility of free charges, such as ions, in the liquid. It is measured in S/m (Siemens/meter).

Speed of sound (v_s): is defined as the speed in which small amplitude oscillating perturbations propagate in an elastic medium. In liquids, it depends on the density and adiabatic compressibility. Sound waves above 20 kHz are called ultrasound.

Experimental Techniques

Nowadays, accurate electrical, optical, and ultrasonic measurements can be carried out using low-cost equipment and do not require specialized personnel. It must be remarked that the properties discussed above depend on temperature, and therefore it must be accurately known when they are measured.

There are several commercial manufacturers of instruments to measure electrical (permittivity and conductivity) properties [20-21] and the techniques are explained in detail in reference texts [22,23].

Measurements of refractive index in liquids are usually made in the visible range [23,24], at the sodium D line (589 nm), with refractometers commercially available from different manufacturers [25,26].

There are several techniques for the measurement of speed of sound in liquids [27]. Commercial instruments are available for laboratory and industrial settings [28,29].

Main Results

In the last 17 years, several systematic studies of the properties of liquid fossil fuels, biofuels, alcohols, and their blends, were undertaking at the Renewable Energy Group (GER) of the Universidad de Buenos Aires. In addition to the properties indicated in the international standards such as ASTM D975 [30], EN 590 [31], ASTM D6751 [32], EN 14214 [33], ASTM D7467 [34], EN 16709 [35], EN 16734 [36], ASTM D4814 [37], ASTM D 5798 [38], ASTM D 4806 [39], alternative properties and their correlations were exhaustively explored. These results were applied to characterize liquid biofuels and feedstocks, to detect contaminants, adulterants, and aging. They were also used to estimate the composition of biofuel/fossil fuel blends, and for the development of sensors based on alternative properties [16].

This work summarises the main results of the research program at GER, UBA, on the characterization of liquid fuels and blends by alternative properties. It must be remarked that GER carried out pioneering and systematic studies of the application of alternative properties for the characterization of fuels and blends. The studied systems include gasoline, bioethanol, and their blends [7], gasoline, methanol, and their blends [40], biodiesel [2,41-43], diesel fuel and its blends with biodiesel [4,44-46], biodiesel, butanol, their blends, and blends of the binary system (biodiesel/butanol) with diesel fuel [47]. Moreover, these studies also encompass the modelling of correlations between permittivity, methanol content, and flash point of biodiesel [14,48], permittivity, conductivity, and speed of sound of biodiesel [43], permittivity, conductivity, and viscosity of biodiesel [49], permittivity and viscosity of biodiesel – diesel fuel blends [50], refractive index, density, and polarizability of biodiesel - diesel fuel blends [46], and refractive index, viscosity, and flash point of biodiesel [44]. The correlations studied by GER broaden the scope of the application of alternative properties for characterization of liquid fuels in industrial settings and field applications, particularly for small – scale producers and end users.

Permittivity and Conductivity

In Corach J, et al. [42], measurements of the electrical conductivity of biodiesel samples of different feedstocks (sunflower, corn, grape, chia, canola, jatropha, coconut and cottonseed), between 300K to 343K, were fitted to an Arrhenius dependence. The pre-exponential factor strongly depends on the moisture content and the presence of conductive contaminants in the sample. On the contrary, the activation energy of the conductivity is an intrinsic property of the biodiesel, depending on the feedstock. The same Arrhenius dependence was found for the conductivity for vegetable oils, with higher values of the activation energy. The Arrhenius dependence makes possible to compare conductivity measurements of a sample at different temperatures to detect conductive contaminants, by comparison to a reference sample at the same temperature. Moreover, experimental data show that, at the same temperature, the relative permittivity of the vegetable oil feedstocks was always lower than the corresponding biodiesels. In consequence, measurements at two different temperatures of the relative permittivity of biodiesel and its feedstock can be applied to detect a partial transesterification process (i.e., the presence of feedstock remnants in the biofuel).

In Corach J, et al. [4], accurate measurements of permittivity of diesel, soybean biodiesel and their blends were carried out at a frequency of 100kHz, between 298K and 333K, in the full composition range. For all samples, permittivity decreases with temperature and increases with biodiesel content, in both cases linearly.

The permittivity of the blends, as a function of temperature and composition, is estimated by a simple model with very good agreement with the experimental data (relative RMS uncertainty lower than 1.2%). Conversely, in the full range of composition, the composition of blends may be accurately estimated from permittivity and temperature measurements, with an RMS uncertainty below 2.5%.

In Mandalunis S, et al. [7], the permittivity of gasoline, bioethanol, and their blends, at 100kHz, was measured between 298K and 323K, in the full composition range. Experimental results show that permittivity decreases linearly with temperature, as in diesel, biodiesel, and their blends. In contrast, the dependence of permittivity on composition is different in blends with lower concentrations (below 25%) of ethanol than in ethanol-rich blends. A second order polynomial gives a good fit in the first case, and at higher ethanol concentrations a linear fit is adequate.

As in the case of diesel/biodiesel blends, composition in gasoline/ethanol blends can be estimated from relative permittivity and temperature measurements. For low permittivity gasoline/ethanol blends ($\varepsilon_r < 5$), composition is fitted to a quadratic formula. In the high permittivity range ($\varepsilon_r > 7$), composition is obtained from a linear function. In the intermediate range ($5 \le \varepsilon_r \le 7$), composition is estimated as a linear interpolation between the two previous functions. In the full range of temperature and composition, the mean square error in the estimation was less than 0.5%.

In Corach J, et al. [40], the permittivity of gasoline, methanol, and their blends was also determined in the full range of composition, at the same frequency and temperature range than for the gasoline/ethanol blends. Similar to diesel/biodiesel and gasoline/ethanol blends, at all compositions the permittivity decreases linearly with temperature. In contrast, at each temperature, the dependence of permittivity on methanol content is fitted to a third-degree polynomial. The estimation of permittivity of methanol/gasoline blends as a function of temperature and composition has an RMS uncertainty of 0.43.

In the same way as diesel/biodiesel and gasoline/ethanol blends, composition of gasoline/methanol blends may be estimated from permittivity and temperature. A third-degree polynomial gives the blend composition as a function of permittivity, where the polynomial coefficients depend on temperature. In the full temperature and composition ranges, the estimation of composition has an RMS uncertainty of 2%.

Refractive Index

In Colman M, et al. [46], the refractive index of low- and ultra-low sulfur diesel fuel, soybean biodiesel, and their blends, was measured, in the full range of blend composition, between 288K and 328K, for low sulfur diesel blends and 293K and 323K for ultra-low sulfur diesel blends. In all cases, it was found that the refractive index depends linearly on both temperature and composition (R^2 higher than 0.98).

In Alviso D, et al. [44], the refractive index at 313K, of low sulfur diesel fuel, biodiesel from different feedstocks (soybean, corn, olive, canola, almond, grape, and peanut oils), and their blends, was estimated from the composition of the samples. The refractive index decreases linearly with increasing biodiesel content in the blend (R^2 higher than 0.99).

In Romano SD [47], the refractive index of soybean biodiesel blended with butanol, and also biodiesel/low sulfur diesel/butanol blends was measured between 288K and 313K as a function of concentration. The full range of concentration was studied in the binary system (soybean biodiesel /butanol). In the ternary system, diesel fuel concentration was kept constant at 10% vol/vol. In all the samples, the refractive index depends linearly on temperature (R^2 higher than 0.99) and, in blends, also on butanol concentration (R^2 higher than 0.99).

Speed of Sound

In Corach J, et al. [43], the speed of sound between 303K and 323K of biodiesel from different feedstocks (soybean, corn, sunflower, olive, grape, and chia vegetable oils) is reported at the frequencies of 1.53MHz, 5.66MHz and 9.43MHz, with an uncertainty lower than 0.05%. Experimental results show that the speed of sound in biodiesel decreases linearly with temperature. Moreover, the speed of sound in the feedstocks is about 5% higher than in biodiesel, with the same behaviour with temperature. The analysis of the results shows that accurate measurements can detect the presence of vegetable oil in biodiesel (for instance, due to an incomplete transesterification).

Correlations between Properties

Permittivity, Methanol Content, and Flash Point of Biodiesel

In Romano SD, et al. [14], measurements of permittivity and methanol content in biodiesel, at different temperature, show that there is a correlation between these two properties. Based on this correlation, methanol content may be estimated from measurements of permittivity at a range of frequency between 20Hz and 2 MHz, as a function of temperature. Since the flash point is also correlated with methanol content, permittivity measurements may be used to estimate the flash point of biodiesel samples [48].

Permittivity, Conductivity and Speed of Sound of Biodiesel

In Corach J, et al. [43], electrical properties (permittivity and conductivity) and speed of sound were determined at temperatures between 300K and 343K in biodiesel from different vegetable oils (sunflower, corn, soybean, grape, cotton, olive, canola, and chia). In addition, the feedstocks properties were also measured. Electrical properties were measured in the frequency range from 20Hz to 2MHz and the speed of sound at the frequencies of 1.53MHz, 5.66MHz, and 9.43MHz.

Within the temperature range studied, for all the biofuel samples, the experimental data show that an increased permittivity is linearly correlated with a higher speed of sound. The same correlation is found in the feedstocks.

Permittivity, Conductivity and Viscosity of Biodiesel

In Corach J, et al. [49], electrical properties (permittivity and electrical conductivity) and kinematic viscosity of biodiesel samples from several feedstocks (sunflower, olive, canola, corn, soybean, grapeseed and jatropha) were measured between 298K and 343K. The measuring frequency range of electrical properties was from 20Hz to 2MHz.

From the analysis of the experimental data, it is found that, in all cases, kinematic viscosity and permittivity of biodiesel are correlated. From this correlation, a model was developed to accurately estimate the kinematic viscosity of biodiesel from permittivity measurements, with an RMS uncertainty below $0.4mm^2/s$, in the full range of temperature. Moreover, kinematic viscosity is also correlated with electrical conductivity. From a model based on this correlation, it is possible to estimate the kinematic viscosity with high accuracy (RMS uncertainty below $0.07mm^2/s$).

Interestingly, the correlation between electrical conductivity and kinematic viscosity makes possible to establish a power law scaling between these two transport properties.

Permittivity and Viscosity of Biodiesel – Diesel Fuel Blends

In Corach J, et al. [50], relative permittivity and kinematic viscosity were determined in soybean biodiesel/ diesel fuel blends, between 298K and 318K (±0.1K), in the full range of composition.

Experimental data show that permittivity is correlated with kinematic viscosity. From the modelling of the results, the kinematic viscosity of soybean biodiesel/ diesel fuel blends was estimated from permittivity and temperature. The agreement between the model and the measured values is very good (the RMS uncertainty of the fitting was below $0.02mm^2/s$). It is worth mentioning that, to determine all the parameters of the model, only measurements of permittivity and kinematic viscosity of biodiesel and diesel fuel are necessary.

Refractive Index, Density and Polarizability of Biodiesel - Diesel Fuel Blends

In Colman M, et al. [46], measurements of the refractive index, between 293K and 323K, of low- and ultra-low sulfur diesel fuel, soybean biodiesel, and biodiesel-diesel blends, in the full range of composition, were reported. The results were correlated with the corresponding experimental data of density and effective polarizability. In all cases, the RMS estimation error was below 6.10^{-4}. Remarkably, the effective polarizability of the biodiesel-diesel blends depends linearly on the composition and is independent of the temperature.

Refractive Index, Viscosity, and Flash Point of Biodiesel

In Alviso D, et al. [44], experimental values of flash point of low sulfur diesel fuel, biodiesel from different feedstocks (soybean, corn, olive, canola, almond, grape, and peanut oils), and their blends, were correlated with the corresponding refractive index values measured at 313K. The flash point of the blends decreases with increasing refractive index. Flash point was also estimated by a second-degree polynomial function of kinematic viscosity and refractive index, with a more satisfactory fitting (R^2 between 0.96 and 0.99). Although for the estimation in the article, both properties were measured at the same temperature (313K), the second-degree polynomial dependence still holds even when the two properties are measured at different temperatures.

Conclusion

The advances on the accurate determination of alternative (electrical, optical, and acoustical) properties in fossil, biofuels, and blends, and their correlation with other physical and chemical properties, together with the improvements in instrumentation and experimental data processing, make possible to complement effectively the methods included in international quality standards for fuel products.

These techniques may be applied at the different stages of production, blending, distribution, and consumption of liquid fossil fuels and biofuels. They have been successfully applied to the optimization of control strategies and processes, final quality control of products, detection of alterations in the different stages of the supply chain, and assessment of fuel quality by the consumer.

In laboratory applications, such as the initial screening of samples, electrical, optical, and acoustical techniques have several advantages over conventional methods. Since they are simpler and faster, they lead to reduced testing times and workload.

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