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Citation: AIP Conference Proceedings **1700**, 050008 (2015); doi: 10.1063/1.4938446
View online: http://dx.doi.org/10.1063/1.4938446
View Table of Contents: http://scitation.aip.org/content/aip/proceeding/aipcp/1700?ver=pdfcov
Published by the AIP Publishing

Dielectric Properties Measurement Method in the Microwave Frequencies Range for Non-polar/Polar Liquid Mixtures Characterization

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Abstract. We present a method based on dielectric properties measurements over a large spectrum of frequencies, in the microwave (MW) domain, in order to characterize a liquid mixture. The liquid mixtures consist of non-polar fluids (silicone oil, diesel fuel) and polar additives, in order to increase the specific MW absorption of the mixture for further MW power processing. We have measured the MW specific absorptions for mixtures of *silicone oil* with 20% and 30% (w/w) *isopropanol*. In both cases, the mixtures are sufficiently stable over time to allow further studies of thermal convection dynamics initiated by MW heating. For a mixture of *diesel fuel* with 10% (w/w) a*lkyl polyglycoside*, the main observation was that its MW specific absorption varies over time after the mechanical mixing process.

INTRODUCTION

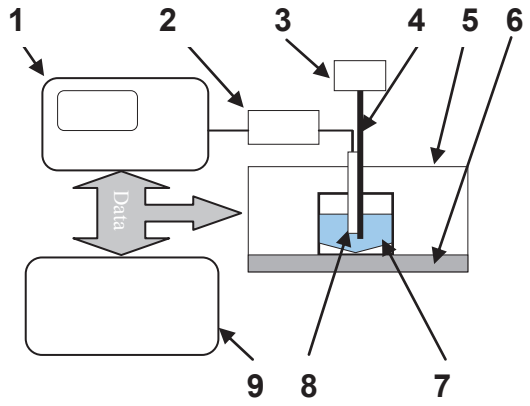
The initial goal of our research was to study thermal convection dynamics (for geological models) in liquids with specific thermal an rheological properties, internally heated by microwaves (MW) [1, 2]. For this purpose, we had to prepare liquid mixtures with specific, well-defined MW absorption at the operating frequency of our heating device (2.47GHz ± 0.020GHz) [3, 4]. The dielectric properties of such mixtures (relative permittivity ε' and dielectric loss ε") define their MW specific absorption and, in the same time, depend on their composition. The key idea is to control the specific MW absorption for a mixture of two or more liquids by measuring its dielectric properties. The resulting dielectric properties are a polynomial combination of the dielectric properties of the components, with concentration as a pondering factor [5]. We show here how this principle was applied to increase the MW absorption of some mixtures based on *silicone oil* and *diesel fuel,* both non-polar liquids. The obtained results confirm that this principle and measurement method can be used to characterize an additivation process for liquid fuels.

10th International Conference Processes in Isotopes and Molecules (PIM 2015)
AIP Conf. Proc. 1700, 050008-1–050008-5; doi: 10.1063/1.4938446

METHOD, MEASUREMENTS AND DISCUSSION

Dielectric Properties Measurement Setup

The dielectric properties measurements in the MW range are based on the *Von Hippel* method, as implemented in a PNA-L N5230A Vector Network Analyzer (VNA): a MW reflection method, which uses a coaxial performance probe, inserted in the sample or in contact with the sample's surface [6]. The dielectric properties are determined by measuring the phase- and amplitude-change in the wave reflected by the sample, as compared with the incident wave. Prior to the measurement, a specific calibration is necessary. The best results are obtained with high loss samples like water. For low loss sample measurements, a MW absorber is placed under the sample, to suppress the reflection of waves exiting the sample on the coaxial probe. Figure 1 presents the experimental setup. The dielectric measurement circuit is composed of the VNA (1), the E-Cal module (2) and the coaxial performance probe (8), together with the calibration accessories and the material measurement software (9). The sample (7) in its plastic vessel and the MW absorber (6) on the bottom are placed in a thermostat (6) at 25°C and the local temperature in the volume of the sample is measured (3, 4) close to the MW coaxial probe.



1- PNA-L N5230A Agilent Vector Network Analyzer
2- E-Cal module Agilent N4693A
3- Thermometer Omega HC4502A RTD
4- Temperature probe PT100
5- Thermostat Lauda RE207, -25ºC...+200ºC, temperature control precission 0.02ºC
6- MW absorber foam, 5mm thickness
7- Dielectric sample in cylindrical plastic vessel with conical bottom end
8- MW Coaxial Performance dielectric probe kit Agilent 85070E
9- PC-laptop, Agilent 85071E Material measurement software

FIGURE 1. Dielectric properties measurement setup

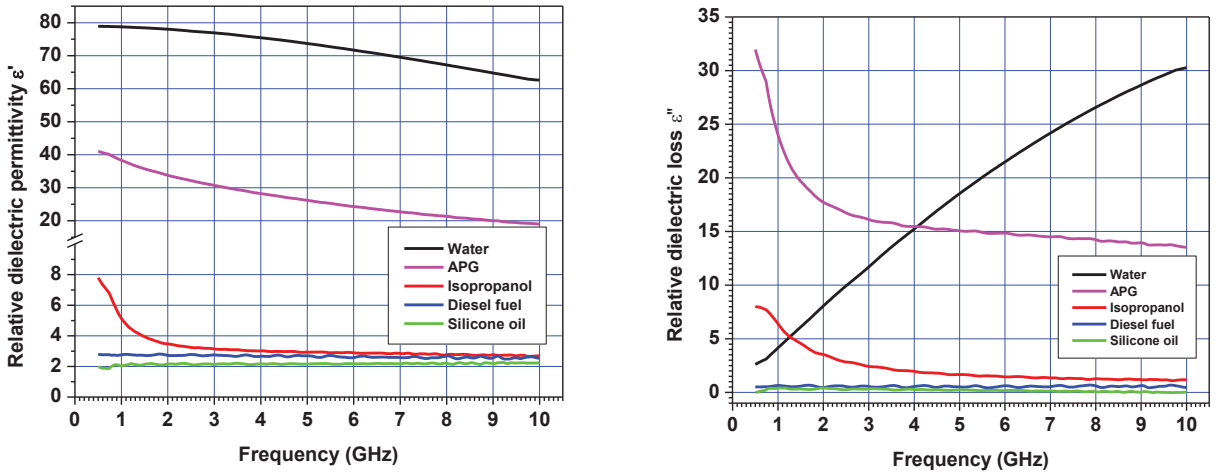
The dielectric properties of the sample are temperature-dependent, so a thermostat is needed to keep a constant sample temperature. To increase the measurement accuracy, an E-Cal module is inserted between the coaxial probe and the VNA, to calibrate the MW circuit before each measurement. Standard calibration is also performed before sample dielectric measurement. The standard calibration requires three reference calibers: MW open circuit, MW short circuit and a sample of double distilled water at 25°C, all measured consecutively, as prompted by the calibration protocol. The operating MW frequencies were selected between 0.5 – 10 GHz, in linear sweep mode, with 10^3 readings over the whole frequency band. The accuracy of the dielectric properties measurements is better than 5% for high loss samples and decreases as the sample gets further from the high loss material model or if there are other causes of receiving unwanted wave reflections from outside the sample. The repeatability and resolution are two to four times better than the accuracy [7].

Dielectric Properties Measurements

A volume of 50 ml was prepared for every liquid sample used for the dielectric measurements. Figure 2 shows the recorded data of the dielectric properties (ε', ε") for the initial materials, the polar liquids (*double distilled water, isopropanol* and 50% (w/w) solution of *Alkyl Polyglycoside (APG)* in water) and the non-polar liquids (*silicone oil* and *diesel fuel)*. Our purpose was to increase the MW absorption of the non-polar liquids by adding various concentrations of polar liquids and thus obtaining a new liquid, from the point of view of the MW absorption. We have tested three mixing homogenization techniques: mechanical, ultrasound and MW pulses treatment. The best stability over time was obtained with mechanical homogenization and we present here the results for this case.

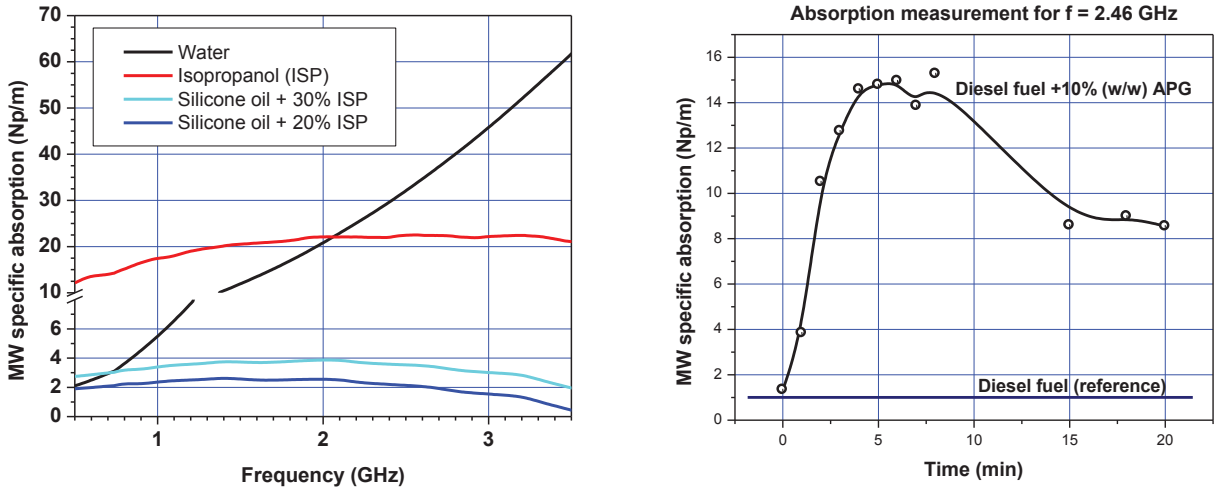
Silicone oil and *isopropanol* were selected for the experimental modeling of thermal convection dynamics in geology models. The *silicone oil* responds to the specific requirements for viscosity, density and optical transparence, but it is a low loss liquid in the desired MW frequencies range. *Isopropanol,* a liquid with relative high MW absorption, was identified as miscible with *silicone oil*. The MW absorption A can be calculated for a specific frequency f using the previously measured relative permittivity ε' and relative loss ε" (1) [3]. In Equation (1), the specific attenuation A is calculated in (Np/m). ε_0 is the free space permittivity, $\varepsilon_0 = 8.856\ 10^{-12}$ F/m. The function Re( ) represents the real part of the complex number contained between the parentheses.

$$A\left(\mathrm{Np/m}\right) = \pi f \varepsilon_0 \varepsilon'' \mathrm{Re}\left(\frac{120\pi}{\sqrt{\left(\varepsilon' - j\varepsilon''\right)}}\right) \tag{1}$$



(a) Relative dielectric permittivity ε' (b) Relative dielectric permittivity loss ε"

FIGURE 2. Dielectric permittivity measurements for *double distilled water, isopropanol*, 50% (w/w) solution of *Alkyl Polyglycosides* (*APG*) in water, *silicone oil* and *diesel fuel.*



(a) MW absorption for *silicone oil-isopropanol* mixtures. (b) MW absorption and stability over time for *diesel fuel-APG* mixture (*diesel fuel* given as reference).

FIGURE 3. MW absorption for the studied mixtures.

Figure 3a presents the MW specific absorption for two mixtures of *silicone oil* with 20% and 30% (w/w) *isopropanol* and, for comparison, *double distilled water* and *isopropanol,* all measured over the 0.5 - 3.5 GHz frequency domain. The mixture is stable for 1-2 days, after this time the *isopropanol* separates from the oil.

The *diesel fuel* with *aqueous APG* is a mixture of low stability. Because we have obtained random measurement values after the homogenization process, we have performed a sequence of permittivity measurements over a 20 min time interval, starting immediately after the mixing. Figure 3b shows the specific MW absorption of the *diesel fuel*-10% (w/w) *APG* mixture over this time and, for reference, the *diesel fuel* MW absorption.

MW Specific Absorption of Liquid Mixtures

The MW specific absorptions for the mixtures of *silicone oil* with 20% and 30% (w/w) *isopropanol* are sufficiently stable over time for the study of thermal convection dynamics initiated by MW heating (with the amendment that the mixture must be prepared in the same day as the intended experiment will be performed). Experimentally we have observed that, as the isopropanol concentration increases, the mixture is less stable over time; consequently, we have limited at 15% the *isopropanol* percentage in the mixture. In this case, the ultrasound mixing method can be used with the same good results regarding the stability. The increase of the *silicone oil* MW absorption is low, but it is enough to start the internal MW heating.

Regarding the mixture of *diesel fuel* with 10% (w/w) *APG,* the main observation is the variation over time of the MW specific absorption after the mechanical mixing process. The absorption – time dependence presented in Figure 3b is reproducible for the same preparation procedure. There are two possible explanations: during the first 5 minutes after mixing, the air bubbles formed in the liquid during the mixing process are released, and this process may be responsible for the increase in MW absorption; after that, as the excess of *APG* additive settles (a layer of sediment can be observed in the end), this separation process decreases the MW specific absorption down to a relatively constant value (which is more than 8 times higher than the *diesel fuel* MW specific absorption). After 24 hours, the *APG* additive separates completely from the rest and the sample's MW specific absorption is the same as for *diesel fuel* (in the measurement accuracy range).

CONCLUSIONS

The presented method, based on dielectric properties measurements and MW absorption analysis, is suitable for the characterization of the mixing processes of polar – nonpolar liquids. This method can be used as a feedback or in the design of the fuel additivation process. Our experiments show that, after the first 5 minutes of mechanical mixing, it is useful to apply another mixing process (MW pulses or ultrasound) for tests of improving the amount of additive mixed with the fuel.

Our main interest was to characterize this type of mixtures for further MW thermal processing; however, this method can be applied to a large variety of liquid mixtures (with polar, highly polar or ionic bi- or multiple components) used as reaction media in MW-assisted chemical processes.

In addition, dielectric measurements reveal a concentration dependence effect and complex permittivity measurements are sufficient to determine the volume fraction of one constituent in a given mixture, using a specific initial calibration. Compared with other methods which use only the relative permittivity ε' [5, 8, 9], our method gives better results since the specific MW absorption includes both aspects of MW interaction: energy *stored* and energy *lost* in material (through different internal processes).

The temperature effect on the mixing process or on the MW absorption can be significant, and further work regarding this aspect of mixtures characterization is required.

ACKNOWLEDGMENTS

The present work was funded by the Executive Agency for Higher Education, Research, Development and Innovation Funding (UEFSCDI), project no. 1 RO-FR-22-2011 and by the National Authority for Scientific Research and Innovation (ANCSI) project no. 44N/2009 PN09-44 for the Romanian team, and by the Agence Nationale de la Recherche (ANR), project no. ANR-11-IS04-0004, for the French team. The diesel fuel and related additives were provided by the JENTS CERPROD S.R.L company.

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