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Thermal-electrical impedance spectroscopy for fluid characterization

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Abstract:

Electrical impedance spectroscopy (dielectric or electrochemical impedance spectroscopy) is a widespread characterization method for solids or fluids in industrial applications. Here we present a novel combination of the electrical impedance spectroscopy with its thermal equivalent, that we thus call “thermal impedance spectroscopy”. With this combination, a deeper insight of the fluid properties, especially at different fluid mixtures, is possible.

Key words: thermal-electrical impedance spectroscopy, thermal conductivity, fluid quality measurement, oil sensor, process fluid control

Motivation

The electrical impedance spectroscopy measures the frequency dependent system response to an AC voltage signal and is determined by the electrical conductivity, capacity and inductivity of the material in combination with the sensor arrangement. We measure in addition the frequency dependent thermal system response of a thermal excitation that is determined by the thermal conductivity and capacity, using an adaption of the well-known 3-omega method [1]. With this method, an alternating current of frequency ω is applied to the heater, causing temperature oscillations and thus a modulation of the temperature dependent heater resistance with the double frequency 2ω This results in a 3ω part of the measured heater voltage spectrum that can easily be separated and from that the amplitude of the temperature oscillation can be determined [1-3]. The amplitude gets higher with lower thermal capacity, conductivity and density, material properties that are determining its thermal impedance.

The combination of both measurement methods on a MEMS based chip and the detailed analysis of their signals due to phase and amplitude arises additional information compared to the individual measurement. This further information allows e.g. the determination of mixing ratios between to fluid or an additional change in the ionic concentration.

Measurements

Since the19th century it is known that the current and the voltage in an AC circuit do not act similar to a DC circuit if capacitive or inductive are within this circuit and will even change depending on the frequency [4]. In this case the electrical impedance, the mathematical complex written resistivity,

$$Z = R + jX \qquad (1)$$

has to be determined, with R its real or ohmic part and X its imaginary part. To measure this impedance one has to determine the AC current amplitude and the voltage amplitude as well as its relative phase compared to the current signal. Depending on the frequency of the alternating current the impedance can vary over several orders of magnitude. The measurement can be done using a known, purely ohmic, shunt resistor in series to the device under test (DUT) and measuring the time depending voltage amplitude over both devices. Using a soft- or hardware lock-in technique [5] the amplitude as well as the phase can be easily determined.

The electrical impedance spectroscopy is a well-known and established technology hence the method to determine the thermal properties is less common. The 3-omega technology developed by Cahill [1] is commonly used to determine the thermal properties of very thin films [6, 7] as there are few methods to measure those samples and their change over

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temperature. A detailed description about the 3-omega method and how to extract the thermal properties from the measured data can be found here [2, 3]. The 3-omega method is also used to determine the thermal properties of gases and liquids [8-11] but so far no commercial sensor has been published.

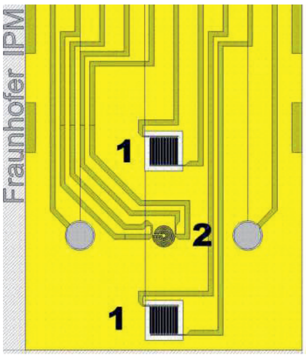

Fig. 1.: Layout-example for the sensitive microstructures that are used for the combined thermal-electrical impedance spectroscopy: interdigital capacitors (1) and a heater structure (2) in form of a disc. The electrode structure is grey, the insulator structure yellow.

The presented results are obtained using a recently developed small electronic tongue system and a MEMS-based measurement structure shown in Figure 1. Using one output of a Redpitaya measurement tool [13], an AC current is applied on the heater structure and the current and the voltage over the measurement structure is determined using the analog inputs of the Redpitaya system in combination with differential instrument amplifiers. To avoid electrical impedance mismatches and to measure several sensor structures with one Redpitaya device, a preamplifier and multiplexing card was developed which is located between the sensor and the Redpitaya. The obtained transient datasets are analyzed by means of a software based lock-in technique, which determines the amplitude and phase for different frequencies in the current and voltage signals. The output of this analysis is the electric or the thermal impedance (the temperature amplitude of the 3-omega measurement), respectively. The complete electronic measurement and analyzing system fits within a 10x10x20 cm³ high frequency tight box.

As sensing device a tongue shaped sensor strip was developed (Figure 1) with various structures obtained by means of MEMS technology. As substrates different polymers foils as well as glass-wafers have been used. The electric impedance was measured by an interdigital capacitor (IDK, label 1, 800 squares @ 10 µm gap) with direct contact to the measured liquids. At low frequencies the impedance is basically determined by electrochemical properties and thus very sensitive to solved ions. In the following measurements we used only deionized water for the mixtures. All measurements here were made at room temperature. The “thermal impedance” was measured by an insulated micro-heater (label 2, typical length 3.7 mm, typical width 20 µm). Gold was used as an electrode material; the insulator was made by a structurable polyimide or SU-8 lacquer.

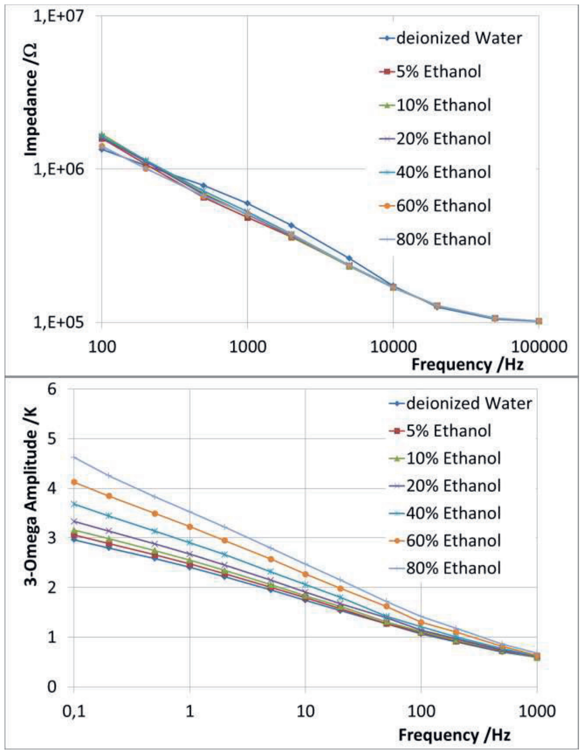

Fig. 2. The 3-omega amplitude (lower part) and the electrical impedance (upper part) over frequency for different ethanol-water (deionized) mixtures. Whereas the electrical impedance is unspecific, the concentration can be determined by the thermal signal. (Gold electrodes on polyimide substrate.)

The measurement of fluid concentrations is also possible for other water based mixtures like isopropanol or glycerin. Figure 3 shows the electrical impedance @100 Hz over thermal 3-omega amplitude @100 mHz for different concentrations of mixtures of deionized water and ethanol, isopropanol and glycerin. Again the concentrations can be measured by the thermal signal, whereas the electric impedance is unspecific. Here, even small contaminations of salts can affect the electric impedance more than the influence of large concentration changes. This is also indicated by the impedance variance of the deionized water measurement (dotted ellipse).

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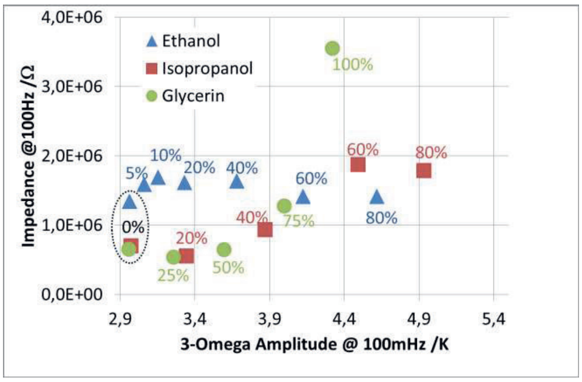

Fig. 3. Electrical impedance @100 Hz over thermal 3-omega amplitude @100 mHz for different concentrations of mixtures of deionized water and ethanol, isopropanol and glycerin. Again the concentrations can be measured by the thermal signal, whereas the electric impedance is unspecific. (Gold electrodes on polyimide substrate.)

Figure 4 shows a similar plot for a sensor chip made from gold electrodes on a glass substrate, insulated by a SU8 layer. The measurements were made thermally at 200 mHz and electrically at 200 Hz with an AC heater current of 0.4 mA_{pp}. The values for air, P3-oil, benzyl-alcohol, ethandiol, isopropanol, 50% glycerin, deionized water and 1g/l urea in deionized water are shown. The pureness of all the substances was pro analysi.

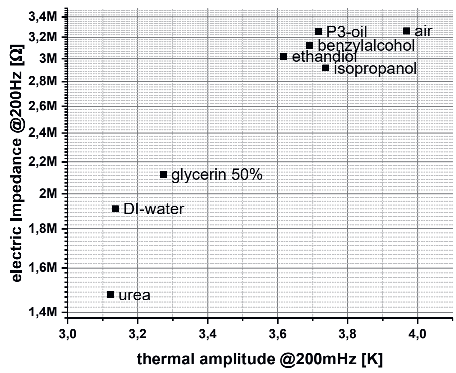

Fig. 4. Electrical impedance @200Hz over thermal 3-omega amplitude @200mHz for air, P3-oil, benzyl-alcohol, ethandiol, isopropanol, 50% glycerin, deionized water and 1g/l urea. (Gold electrodes on glass substrate.)

Figure 5 shows the effect of a salt contamination of 1 g/l NaCl in deionized water, 50 vol% and 80 vol% ethanol, respectively. The electrical impedance @100 Hz over the thermal 3-omega amplitude @1 Hz with an AC heater current amplitude of 0.6 mApp. Again, SU8-insulated gold electrodes on glass substrates were used. As expected, only the electric measurements are affected by the salt. A determination of the alcohol content is still possible thermally.

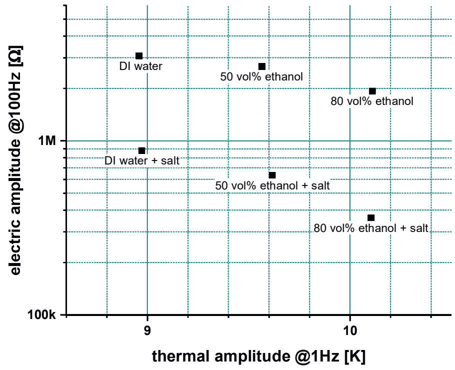

Fig. 5. Influence of salt on the electrical impedance @100 Hz over thermal 3-omega amplitude @1 Hz for different water ethanol mixtures without and with 1 g/l NaCl. (Gold electrodes on glass substrate.)

The measurements results of the figures 4 and 5 are excerpts from frequency depending sweeps as shown in figure 3. As these measurements require between 10 to 30 min for each sweep pair they are unpractical to supervise a process. As can be seen in the figures 4 and 5 for a known process only measurements at one or few frequency points might be needed to control a process. These analyses indicate that it is possible to reduce the required measurements and analysis electronics for a future integrated sensor system to be implemented within a plug or small box directly located at the MEMS sensor.

Applications beside liquids

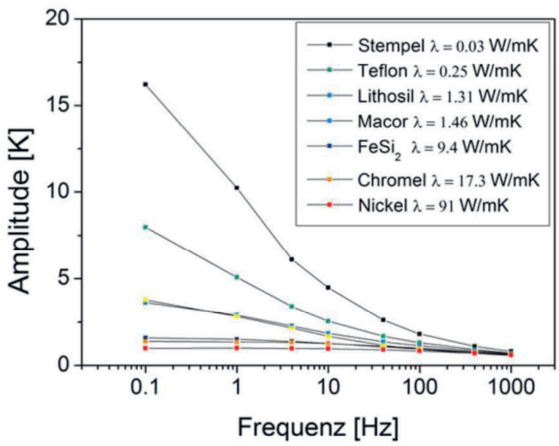

Fig. 6. Thermal characterization of different bulk specimen measured with a sensor pressed on a polished surface. (Gold electrodes on polyimide substrate.)

As mentioned before the 3-Omega was originally used to measure the thermal conductivity of bulk and thin film samples. In this case sensor structures had to be lithographically added on the surface of the measurement specimen. Using a flexible polymer sensor we were able to measure various bulk samples just by pressing the sensor structure on a polished test specimen. Results on different specimen are shown in Figure 6.

Conclusion and next steps

The measurement of the thermal impedance is a reasonable extension to electrical impedance measurements. The thermal measurements are affected by changes in density, thermal conductivity, heat capacity or currents. In addition, there is also a depth information in the frequency dependent signal (not shown here, see e.g. [7]), since thermal waves at lower frequencies have deeper penetration depths than at higher frequencies. Thus, fluid exchange processes at wetting surfaces, turbulences and scaling effects [11] can be monitored. Currently, such thermal-electric impedance spectroscopy measurements are tested for process and quality control in fluids. But also solids can be characterized by this method. Here, one possibility is to fabricate measurement chips with insulated test structures and cover them by a sample layer for measurement [12]. Another possibility is using foils with the sensor structures pressed onto the polished surface of a sample by a stamp [6].

MEMS based sensors enable the combination of multiple sensing structures on a small surface. By combination of the individually seen often unspecific sensor signals, even complex processes like mixing or material synthesis can be monitored. Due to recent improvements in the microcontroller development complex integrated data acquisition and analyzing systems can be realized at low cost with reasonable power consumption to operate those new multi parameter sensor devices. Compensating temperature effects are a crucial point for the usage of those sensors in real life.

Acknowledgement

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