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A new way to find dielectric properties of liquid sample using the quartz crystal resonator (QCR)

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ABSTRACT

The main objective of this article is to demonstrate by performing experimental measurements how the equivalent capacitance C_0 changes when a fluid sample such as water is in contact with the crystal and to relate this change with the relative permittivity of the fluid. These measurements were compared with simulations of traditional models like Butterworth–Van Dyke (BVD). To obtain the change of C_0 when the crystal is in contact with water, the relation between the series resonance frequency (f_s) and frequency at minimum impedance with zero phase (f_r) is used. To know these values, a new way of finding principal parameters in quartz crystal resonator (QCR) sensors, such as series resonant frequency (f_s), half band half width (Γ) and maximum peak of conductance (G_m), is proposed; both for an unperturbed crystal and a crystal loaded with a liquid. The method consists in measuring the current (I) that flows through the crystal and the voltage (V) between electrodes at frequency values near to resonance (sweep frequency). Additionally, the susceptance $|B|$ of the crystal is also measured, multiplying the 90 degrees shifted current of the crystal and its voltage, using a mixer. The DC component of this operation is proportional to the susceptance of the crystal. With the magnitudes of the admittance and susceptance, the real value of the conductance $|G|$ is obtained for each frequency value in the sweep. The conductance and susceptance curves were fitted with a summation of Gaussian and sine functions respectively with the minor RMSE possible.

The proposed method has been compared with simulations done in COMSOL Multiphysics in order to verify the experimental results with simulation data. MATLAB curve fitting toolbox was used to fit the experimental curves.

1. Introduction

TSM sensors and particularly the quartz crystal resonator (QCR) sensors are considered highly efficient sensing systems not only because of their low manufacturing cost but also because of their high accuracy, bio-functionalization capacity, sensitivity and reliability in measuring the deposited sample, either gaseous or liquid media. So, QCR have been widely studied for several decades by different investigators who have obtained theoretical models that explain the behavior of the sensor, especially in the frequency ranges close to the resonance frequency [1–5]. A model that stands out for its use and ease of understanding is that proposed by Butterworth–Van Dyke (BVD) [2] which is derived from the transmission network model proposed by Mason [1,2,6]. The BVD model is especially used in many publications to support the results because of the electrical representation that provides of the physical behavior of the sensor [3,7–10].

However, the results obtained from the performance of the sensor during our experimentation and from the existing simulation models, based on the equations of BVD basic electric model, do not always correspond so well when the sensor is exposed to fluids like water. This is due to the fact that small changes between series res-

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onance frequency (f_s) and frequency at minimum impedance with zero phase (f_r) are not taken into account in the basic model.

Within the BVD model, the static capacitance C_0 models losses caused by external electrical components to the sensor and the parasitic capacitance generated by the electrodes on the crystal [9,10]. This capacitance is to be significant for values far from the resonance frequency and negligible when the crystal is operating at values close to the resonance frequency [7]. Furthermore, this value is considered static and dependent only on the physical characteristics of the crystal, so in many experiments is eliminated externally [9,10]. However in practice, the results of the experiments expose that this capacitance value depends also on the properties of sample, so it is important to take it into account.

This article aims to demonstrate how the capacitance C_0 changes when a fluid sample is in contact with the crystal by experimental measures and comparison with simulations models developed using COMSOL, and how this C_0 change generates additional information about the sample. The BVD model, when the change in C_0 is taken into account, is closer to experimental results.

2. Theory and methods

The quartz crystal resonator (QCR) is traditionally considered to be a mass sensitive sensor that generates response (changes its resonant frequency) to different thin film samples or liquids in contact with its surface. For a thin film mass of the order of nanograms, the crystal response will be of the order of Hertz according to Sauerbrey Equations [4]:

$$\Delta f = -\frac{2{f_0}^2}{A\sqrt{\rho_q \mu_q}}\Delta m \tag{1}$$

$$f_0 = \frac{n}{2h_q}\left(\sqrt{\mu_q/\rho_q}\right) \tag{2}$$

where ρ_q and μ_q are the specific density and the shear modulus in quartz, respectively, f_0 is the fundamental resonant frequency in quartz, related to its thickness h_q, Δm is the thin film mass deposited A is the piezoelectrically active crystal area and n is the overtone number.

In contact with liquids, the crystal is capable of giving information about the density-viscosity product $(\rho\eta)^{1/2}$ [1,11] of the fluid by changing its resonant frequency and quality Q-factor according with Kanazawa equations [5]:

$$\Delta f = -\sqrt{n}{f_0}^{3/2}\sqrt{\frac{\rho_L \eta_L}{\pi \rho_q \mu_q}} \tag{3}$$

$$\delta = \sqrt{\frac{2\eta_L}{\omega \rho_L}} \tag{4}$$

where ρ_L and η_L are the density and viscosity in fluid respectively. Eq. (4) shows the decay characteristic length (δ) as proportional to the ratio viscosity to density of the liquid and as inversely proportional to the angular frequency (ω) [12]. For 10 MHz AT cut quartz crystal this length is: δ = 178 nm.

Additional to Sauerbrey and Kanazawa equations, the quartz crystal comportment can be seen as an electrical resonator circuit [2]. There are some electrical models like Mason or BVD that explain the crystal behavior near to the resonance frequency.

Fig. 1 shows the equivalent conductance comportment (G_{eq}) for unperturbed crystal (kept in air) and for the top face of the crystal immersed in liquid. It is important to note that the maximum value takes place at resonance frequency [3,13]. The bandwidth shift ($\Delta\Gamma$) gives essential additional information, for example, it

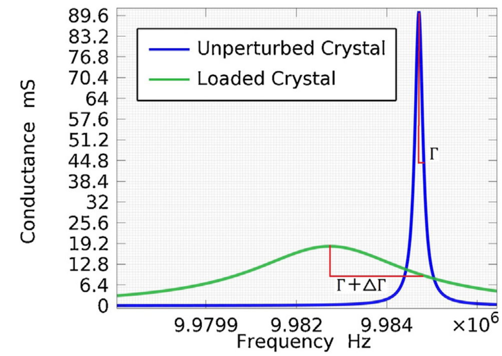

Fig. 1. Change in morphology of the conductance curve for bare crystal and in contact with a liquid.

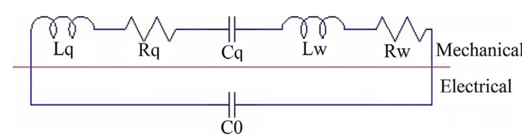

Fig. 2. Schematic of BVD (Butterworth–Van Dyke) model for quartz crystal resonator.

is useful to determine the viscoelastic properties of the fluid. Both values are related through the quality factor Q:

$$Q = \frac{F_s}{2\Gamma} \tag{5}$$

According to theoretical models, when the QCR electrode is in contact with a sample, the morphology of the conductance curve changes as it is shown in Fig. 1, shifting it to the left on the frequency axis, increasing its bandwidth and decreasing its magnitude. The electrical model BVD explains this phenomenon by adding impedance that represents the characteristics of the sample. For liquids in contact with the crystal, the equivalent impedance is the series of an inductance and a resistance [10–12] (See Fig. 2). The values of these impedances are related with the characteristics of the samples according with the Eqs. (6)–(11) [1,2,7,10,12]:

$$C_q = \frac{8{e_{26}}^2 A}{(N\pi)^2 \overline{C_{66}} h_q} \tag{6}$$

$$L_q = \frac{1}{{\omega_s}^2 C_q} \tag{7}$$

$$R_q = \frac{\eta_q}{\overline{C_{66}} C_q} \tag{8}$$

$$L_w = \frac{\omega_s L_q}{N\pi}\left(\frac{2\rho_L \eta_L}{\omega_s \overline{C_{66}} \rho_q}\right) \tag{9}$$

$$R_w = \frac{\omega_s L_q}{N\pi}\left(\frac{2\omega_s \rho_L \eta_L}{\overline{C_{66}} \rho_q}\right) \tag{10}$$

$$C_0 = \frac{\varepsilon_{22} A_e}{h_q} \tag{11}$$

where, $\overline{C}_{66}$ is the piezoelectrically stiffened elastic constant for loss-less quartz (2947 × 1010 N/m^2), e_{26} is the piezoelectric stress constant for quartz (953 × 103 A s/m^2), ε_{22} is the quartz permittivity (3.982 × 10^{-11} A^2 s^4/kg m^{-3}), A is the active electrode area (m^2), h_q is the quartz crystal thickness (m), η_q effective viscosity of quartz (3.5 × 10^{-4} kg/m s), ω_s is the series resonance frequency ($2\pi F_s$). For the liquid, ρ_L and η_L are the density of liquid sample (kg/m^3) and the viscosity of the liquid sample (kg/m s) respectively [3].

The model shown in Fig. 2 explains the behavior of QCR In contact with a sample of liquid. In this theoretical model, the maximum