

Piezoelectric resonator and high-frequency admittance study of viscosity and density changes in a thin water layer adjacent to the electrode surface

Deivis Plausinaitis^a, Magne Waskaas^b, Rimantas Raudonis^a, Vytautas Daujotis^{a,*}

^a Department of General and Inorganic Chemistry, Vilnius University, Naugarduko 24, 03225 Vilnius, Lithuania

^b Faculty of Technology, Telemark University College, Kjølnes Ring 56, N3918 Porsgrunn, Norway

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Abstract

Analysis of the admittance of quartz resonator in contact with water has shown that the viscosity–density product of water at a metallic electrode surface decreases during the positive potential scan. This decrease is more pronounced when the electrode surface is coated by a thin layer of polymer. Literature data provide evidence that the density of water in the first layer attached to the surface increases when the potential is shifted into the positive direction. It is concluded that during such potential scan the decrease of viscosity is larger than the increase of density.

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1. Introduction

Many important industrial processes and community works involve transport of fluids in pipes. Examples are among others, supply of water to hydroelectric power stations, waterworks, water purification plants, transport of oil and gas in pipes, etc. A common problem connected with all forms of transport of fluids in pipes and ducts is the loss of fluid pressure due to the flow resistance. This pressure loss causes loss of energy for all processes, which includes pipe transport of fluids. It has been shown that an electric potential applied to the pipe wall can reduce flow friction [1]. This effect was tested in an industrial environment. An increase of water flow rate in the pipe of the hydroelectric power station at Vrenga (southern Norway) was observed and, subsequently, an increase in production of electrical energy up to 2% was obtained [2]. Thus, the study of the nature of this effect is of great practical importance. The profile of the flow rate in the tube suggests the increase of the flow rate near the surface when a positive potential is applied to the surface. Therefore, it is reasonable to conclude that positive potentials favour water

flow by changing the properties of the water layer adjacent to the surface.

The arrangement of water molecules at charged aqueous interfaces is an important question in physics and chemistry. Electrostatic considerations suggest that the molecules in the first layer are reoriented from oxygen-up to oxygen-down as the electrode charge changes from negative to positive [3–5]. Toney et al. [6,7] reported surface X-ray scattering measurements of the water distribution perpendicular to a Ag(111)–electrolyte interface in 0.1 M NaF at two potentials: +0.52 and –0.23 V from the potential of zero charge (PZC). They found that, first, the water is ordered in layers extending about three molecular diameters from the electrode. Second, the extent of ordering and the distance between the electrode and first water layer depends on potential, the latter being consistent with a hydrogen (oxygen) orientation towards the surface for negative (positive) electrode potential. Third, the inner water layer contains at –0.23 V $1.55 \times 10^{15} \text{ cm}^{-2}$ and at +0.52 V $2.6 \times 10^{15} \text{ cm}^{-2}$ water molecules, remarkably more than $1.15 \times 10^{15} \text{ cm}^{-2}$ water molecules expected from the bulk water density. Such a large compression shows that the molecular arrangements in the inner layer are significantly different from those in the bulk, which has not been anticipated in earlier models of charged, aqueous interfaces. Toney et al. [6,7] gave a qualitative explanation

* Corresponding author. Tel.: +370 5 2331004; fax: +370 5 2330987.
E-mail address: vytautas.daujotis@chf.vu.lt (V. Daujotis).

tion of this large density as resulting from the strong electric field at the charged Ag(1 1 1) electrode and presented a tentative model of the molecular arrangements. Later, the influence of electrode potential on the water density was confirmed for the RuO₂(1 1 0)/bulk water (0.1 M NaOH solution) interface [8].

The results obtained by Toney et al. [6,7] instigated the theoretical revision of earlier adopted views, the main of which was that the molecules become arranged in several layers adjacent to a solid interface with densities similar to that in the bulk. The particle–particle–particle-mesh method with a modification to account for the slab geometry was used to calculate the interfacial fluid structure characteristics found to be generally in good qualitative agreement with the conclusions obtained by the experimental X-ray study [9]. Similar results regarding to water compression by the local electric field were obtained from the analysis of the equation of state and the behaviour of entropy [10]. High interfacial fields ($E > 10^9 \text{ V m}^{-1}$) at electrode surfaces can cause a phase transition with an ordered layering of water at high densities similar to ice X [10].

The viscosity of water adjacent to surfaces and the spatial range of influence of these surfaces on the viscosity and water density are subject to some controversy. Quantitative theoretical analysis in microscopic terms of the viscosity in the layers adjacent to the surface is still lacking. The experimental methods most used to measure surface viscosity are based on acoustic sensors [11–16]. Smooth resonators, which viscously entrain a layer of contacting liquid, exhibit a response that depends on the product of liquid density and viscosity of that thin layer, the thickness of which is lower than the decay length of the wave propagating into the bulk [11,12,16]. The measured product of viscosity and density of this liquid layer usually differs significantly from the bulk values [14]. Combining smooth- and textured-surface resonators in a monolithic sensor enables the liquid density and viscosity to be separated and extracted simultaneously [13–15].

Most of published works focus on the development of models describing the electrical responses of thickness-shear mode resonators subject to a variety of surface conditions. These resonators are mainly used as mass sensors. Their application as viscosity sensors is less widespread.

Using the surface X-ray scattering technique Toney et al. [6,7] found only a few ordered water monolayers at the surface. Corresponding calculations show that such thin layers cannot noticeably affect the response of the quartz crystal resonator. However, there are experimental observations that the orientation of water molecules in the interfacial layers results in the shift of equilibrium between water clusters in the noticeably thicker “transitional” water layer. This is discussed together with the results obtained in present work in Section 4.

The purpose of this work is to measure and examine the dependencies of the product of viscosity and density on the potential by using smooth thickness-shear mode resonators. Next, to analyse these data in relation to the earlier detected effect of the applied electrical potential on the increase of the flow velocity in the tube.

2. Experimental

2.1. Analysis of admittance behaviour of quartz resonators

For the quartz resonator used in liquid environment, where effects of added mass and viscous coupling of the resonator to the liquid can appear, the electrical equivalent of QCM is presented in Fig. 1 [11–13]. The electrical characteristics of a quartz resonator, not contacted by liquid, are modelled by region I in Fig. 1. The capacitance C_0 arises between the electrodes located on opposite sides of the insulating quartz. In parallel to this capacitance the electrical coupling to a shear mode mechanical resonance gives rise to an additional motional contribution. This motional region has inductive (L_1), capacitive (C) and resistive (R_1) elements. Under loading conditions new elements are included into the equivalent circuit. The effect of resonator surface loading by the liquid is represented by a motional inductance, L_2 , and a resistance, R_2 . Rigid mass loading increases the motional inductance described by L_3 . The interaction of the quartz crystal resonator with the contacting medium has been elucidated using network analysis according to this circuit.

In general, the admittance of quartz resonator is

$$\vec{Y} = \vec{G} + j\vec{B}, \quad (1)$$

where G is the conductance and B is the susceptance.

$$|Y| = (G^2 + B^2)^{1/2}. \quad (2)$$

The phase angle Θ is related to G and B according to the equation

$$\tan \Theta = \frac{B}{G}. \quad (3)$$

To relate explicitly the circuit elements shown in Fig. 1 to physical properties of the quartz, perturbing mass layer and liquid loading, model assumptions were used allowing to explicitly relate circuit elements to physical properties. For an ideally smooth surface in contact with a Newtonian liquid, the analysis of the general equivalent circuit (Fig. 1) produces [11,13]:

$$\rho_L \eta_L = \frac{64}{\pi} \mu_q \rho_q (K^2)^2 f_s^2 C_0^2 (R - R_1)^2, \quad (4)$$

where η_L and ρ_L are the dynamic viscosity and the density of liquid, respectively; $\mu_q = 2.957 \times 10^{10} \text{ kg m}^{-1} \text{ s}^{-2}$ the shear modulus of AT-cut quartz and $\rho_q = 2650 \text{ kg m}^{-3}$ the density of

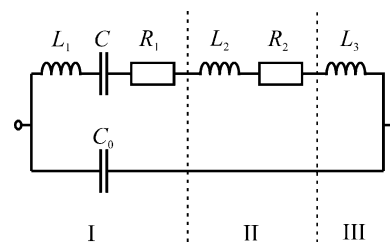


Fig. 1. The general equivalent circuit for an AT-cut quartz resonator with contributions from the mass of rigid film (region III) and the viscosity and density of a liquid (region II) in contact with one face of the quartz resonator (region I).

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