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STATISTICAL RADIOPHYSICS

Voltage Ripple of Electrode Sensor in Electrolyte Flow

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Abstract—Voltage ripple of electrode sensor that moves relative to electrolyte (flow noise) is interpreted in agreement with experimental data using variations in the adsorption rate of oxygen on the electrode surface upon variations in the thickness of the diffusion layer. A relationship of electrode potential and flow velocity jumps is derived. It is shown that the electrode voltage ripple is proportional to the pulsation of the liquid flow velocity and is inertial with respect to such a pulsation with a time constant of several milliseconds. It is also shown that the sensor sensitivity to velocity pulsations exponentially decreases with a time constant of several hours. The dependences of the ripple amplitude on the pulsation frequency of the velocity, electrolyte concentration, and storage time of electrodes in electrolyte that are obtained using the above relationship are proven in experiments.

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INTRODUCTION

A pair of electrodes in electrolyte forms an electrode sensor of electric field that can be employed in geophysics for the measurement of electric fields in ocean, electromagnetic flowmeters, and systems for communications at ultralow frequencies and significant depths [1–3]. The potential difference of two electrodes in electrolyte exhibits variations in time (electrode noise) [4, 5]. Such noise results from electrochemical processes (in particular, oxidation of electrode metal) on the electrode surface. The electrode noise limits sensitivity of electrode sensors of electric field. When a sensor moves relative to electrolyte, the electrode noise increases, and the experiments of [6–9] show that the voltage ripple is related to the velocity pulsations.

The reasons for the flow noise of electrodes is poorly studied. Electrode voltage ripple caused by deformation of double electric layer (DEL) on the electrode surface in the presence of the liquid flow has been considered in [10]. However, the resulting voltage ripple is less than the experimental levels by an order of magnitude.

The purpose of this work is the interpretation of the flow noise of electrodes of an electrode sensor.

1. THEORETICAL SUBSTANTIATION

In air, oxygen is adsorbed on the polished surface of a metal electrode and forms monoatomic layers on surface fragments. When the electrode is immersed in electrolyte (in particular, sea water) oxygen adsorption on the surface is continued. Adsorbed oxygen is involved in reactions with metal, and the resulting oxide molecules are oriented in such a way that negatively charged oxygen ions are located on the surface. In the absence of oxidation (e.g., for noble metals), an electron from metal is attached to oxygen and a negatively charged oxygen ion also emerges on the surface. In this case, the potential of metal is positively biased relative to electrolyte [11]. Such processes lead to the formation of an excess negative charge on the electrode surface. An excess positive charge of the ions of dissolved salt is formed in electrolyte in the vicinity of the surface. Thus, a DEL that is formed on the electrode surface represents a lossy capacitor one plate of which is a liquid plate. The potential of the electrode surface relative to the electrolyte is determined by the ratio of surface charge Q to DEL capacitance C_D:

$$\varphi = Q/C_D. \tag{1}$$

Such a potential changes, since the adsorption of oxygen and oxidation involving adsorbed oxygen are continued.

In the electrolyte layer in the vicinity of the electrode surface, the concentration of atomic oxygen is less than that in the bulk of electrolyte, since oxygen atoms are either involved in reactions with metal or adsorbed on the surface. Such a layer is called diffusion layer, since oxygen arrives at the surface of metal predominantly due to diffusion. In accordance with the first Fick law, the oxygen deposition rate per unit area of the electrode surface (diffusion flux density) is

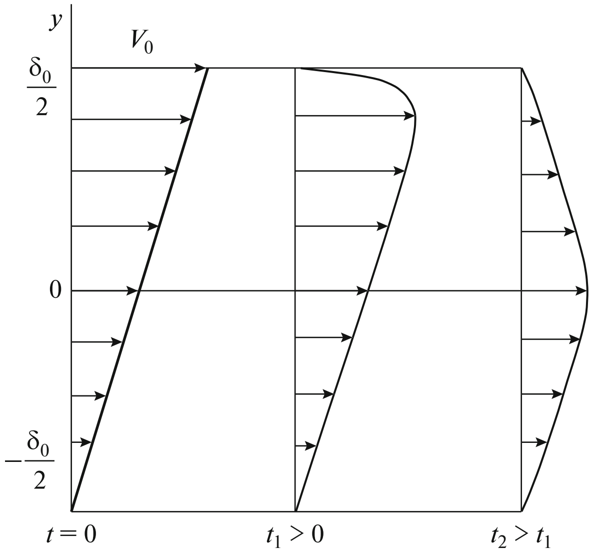

Fig. 1. Diagrams of the liquid-velocity vector in the boundary layer upon a stepwise decrease in the flow velocity to zero at different time moments.

proportional to the concentration gradient in the diffusion layer [11]:

$$\frac{dN}{dt} = N_A D \frac{dc}{dy}, \tag{2}$$

where dN is the number of oxygen atoms deposited over time dt, N_A is the Avogadro number, D is the diffusion coefficient of oxygen, and dc/dy is the gradient of concentration c (mol/L) of oxygen along the y axis that is orthogonal to the surface. Thickness of the diffusion layer δ in the oncoming liquid flow depends on the flow velocity and is about 0.1 of thickness δ_0 of the Prandtl layer ($\delta = 0.1\delta_0$). In the interfacial layer, the liquid velocity linearly increases along the y axis that is perpendicular to the surface from zero to V_0 at the external interface δ_0 of the boundary layer. The results of [12] for a laminar flow around a semi-infinite smooth plate show that the thickness of the boundary layer in aqueous electrolyte is represented as

$$\delta_0 \approx 3\sqrt{\frac{\nu x}{V}}. \tag{3}$$

Here, ν is the kinematic viscosity of liquid, x is the distance from the front edge of the plate, and V is the liquid velocity at a large distance from the surface.

We consider the limiting diffusion flow. In this case, the oxygen atoms that arrive at the electrode surface owing to diffusion are instantaneously absorbed (involved in adsorption or reaction with metal with oxide formation). Thus, the oxygen concentration in electrolyte in the vicinity of the electrode surface is always zero and the concentration gradient can be estimated as c_0/δ (i.e., the ratio of the oxygen concentration in the bulk of electrolyte to thickness of the diffusion layer δ). Expression (2) shows that the following number of oxygen atoms are deposited on electrode surface S over time interval Δt:

$$N = N_A DS \frac{dc}{dy} \Delta t. \tag{4}$$

In this case, the surface charge increases by $\Delta Q = 2eN$ in accordance with the valence of oxygen (e is the electron charge).

We assume that electrolyte flows along the plane surface of electrode perpendicularly to the front edge along the x axis. Let V_0 be the flow velocity at a large distance from the surface. Thickness δ_0 of the boundary layer in the vicinity of the electrode surface depends on the x coordinate. At moment $t = 0$, the flow velocity (i.e., the liquid velocity at the external interface of the boundary layer) exhibits a stepwise decrease to zero. Retardation forces exerted on the boundary layer depend on the velocity gradients in the vicinity of the interfaces. At the external interface, such a force is several times less than the force at the internal interface (i.e., in the vicinity of the electrode surface). The velocity profile along the y axis becomes similar to the velocity profile of a liquid flow between two parallel plates (Fig. 1) in the presence of pressure difference Δp. In accordance with the Newton formula [12], both retardation forces are given by

$$F = \eta S \frac{dV}{dy}, \tag{5}$$

where $\eta = \nu\rho$ is the dynamic viscosity and ρ is the density of liquid. The distance between the plates is δ_0. In the steady state, the resultant force of the pressure forces is balanced by two retardation forces F.

We assume that $y = 0$ corresponds to the center of the gap. The following equality is valid for a liquid layer at the center of the gap with coordinates ranging from $-y$ to $+y$:

$$2ay\Delta p = -\eta a L \frac{dV}{dy}, \tag{6}$$

where L is the length and a is the width of the plates (electrode length and width). The integration of expression (6) under the initial condition $V = 0$ at $y = \delta_0/2$ yields the dependence of the liquid velocity on coordinate y:

$$V(y) = \frac{\Delta p}{\eta L}\left(\frac{\delta_0^2}{4} - y^2\right) = V_{\max}\left(1 - \frac{4y^2}{\delta_0^2}\right), \tag{7}$$

where the maximum velocity at $y = 0$ is

$$V_{\max} = \frac{\Delta p \delta_0^2}{4\eta L}. \tag{8}$$

We assume that $V_{\max} = 0.5V_0$, since such a velocity is observed at the initial moment in the boundary layer at

$y = 0$. Then, the velocity gradient of liquid in the vicinity of the plates is written as

$$\frac{dV}{dy} = -2\frac{V_0}{\delta_0}. \tag{9}$$

The absolute value of the retardation force that is exerted by the two plates at length dx in the steady state at the velocity at the center of the boundary layer $V_{\max} = 0.5V_0$ is given by

$$dF = 4\eta a\frac{V_0}{\delta_0}dx. \tag{10}$$

In the presence of the stepwise variation in the flow velocity, the velocity at the center of the boundary layer depends on time and we use notation $V(t)$. Then, we have

$$dF = 4\eta a\frac{V(t)}{\delta_0}dx = \frac{4\eta aV(t)}{3\sqrt{\nu x/V_0}}dx. \tag{11}$$

Velocity V_0 appears in the denominator, since the boundary layer is formed over the entire surface of the plate and the layer thickness is given by expression (3). At entire length L of the plates (electrode), the retardation force is represented as

$$F = \int_0^L dF = \frac{8\eta a}{3}\sqrt{\frac{V_0L}{\nu}}V(t). \tag{12}$$

The dynamic equation for the entire boundary layer at the entire length of the electrode is written as

$$m_{\text{eq}}\frac{dV}{dt} + F = 0, \tag{13}$$

where m_{eq} is the equivalent mass of the entire boundary layer at the electrode length. Such a quantity can be calculated using the kinetic energy of the boundary layer. For a fragment of the boundary layer with length Δx, the kinetic energy is given by

$$E_{\text{k}} = 2\int_0^{\delta_0/2}\frac{\rho a\Delta x}{2}V^2(y)dy = \frac{1}{15}\rho a\delta_0\Delta xV_0^2. \tag{14}$$

The same kinetic energy can be calculated as $E_{\text{k}} = \rho a\Delta x\delta_0\bar{V}^2/2$. Thus, we find the mean-energy velocity of the boundary layer:

$$\bar{V} = \sqrt{\frac{2}{15}}V_0, \tag{15}$$

which is $0.365V_0$. The kinetic energy of the entire boundary layer is represented as

$$\begin{aligned} E &= 0.5\int_0^L \bar{V}^2\rho a\delta_0 dx \\ &= \frac{\rho aV_0^2}{15}\int_0^L \delta_0 dx = \frac{2}{15}\rho aV_0^{1.5}L^{1.5}\nu^{0.5}. \end{aligned} \tag{16}$$

The same energy can be written as $0.5m_{\text{eq}}\bar{V}^2$, and we obtain

$$m_{\text{eq}} = 2\rho aL\sqrt{\nu L/V_0}. \tag{17}$$

Then, Eq. (13) can be represented as

$$\frac{dV}{dt} + \frac{4V_0}{3L}V = 0. \tag{18}$$

Under the initial conditions $t \to \infty$, $V \to 0.5V_0$, the solution to such an equations is represented as

$$V(t) = 0.5V_0(1 - \exp(-t/\tau_1)), \tag{19}$$

where

$$\tau_1 = 3L/4V_0 \tag{20}$$

is the time constant of equilibration of the boundary-layer parameters. We assume that the same time constant characterizes the diffusion layer.

With allowance for oxygen adsorption in electrolyte, we use expressions (3) and (4) to calculate charge of electrode surface with length L at time t:

$$Q \approx 6.7FDac_0\sqrt{\frac{V_0}{\nu}}\int_0^t\int_0^L\frac{dtdx}{\sqrt{x}}, \tag{21}$$

where $F = eN_{\text{A}}$ is the Faraday number.

We assume that the flow velocity exhibits a stepwise increase by $\Delta V_0 \ll V_0$. The flow velocity at the center of the boundary layer increases in accordance with expression (19). Owing to the continuity of the flow in the boundary layer, the thickness of such a layer decreases and the gradient of the oxygen concentration increases. Formula (21) can be represented as

$$\begin{aligned} &Q \approx 6.7FDac_0\sqrt{\frac{V_0}{\nu}} \\ &\times\int_0^t\int_0^L\frac{1 + \frac{\Delta V_0}{V_0}(1 - \exp(-t/\tau_1))}{\sqrt{x}}dxdt. \end{aligned} \tag{22}$$

The integration of this expression yields

$$\begin{aligned} &Q(t) \approx 13.4FDac_0\sqrt{\frac{V_0L}{\nu}}t \\ &- 13.4FDac_0\frac{\Delta V_0}{V_0}\sqrt{\frac{V_0L}{\nu}}\tau_1(1 - \exp(-t/\tau_1)). \end{aligned} \tag{23}$$

The first term (for which we use notation Q_0) is a slowly varying quantity determined by a gradual increase in the electrode charge due to diffusion. The second term (ΔQ) is related to a stepwise increase in the velocity. A variation in the potential of electrode in the presence of velocity jump ΔV_0 is given by

$$\Delta\varphi = \frac{\Delta Q}{C_D} = -\frac{13.4 FDLac_0 \Delta V_0 \tau_1}{C_D\sqrt{LV_0\nu}} \times (1 - \exp(-t/\tau_1)) = -\varphi_m(1 - \exp(-t/\tau_1)). \quad (24)$$

Expression (24) corresponds to the response of a two-port with the transient characteristic $h(t) = 1 - \exp(-t/\tau_1)$ to voltage jump φ_m. In the frequency domain, the gain of such a two-port is written as

$$K(\omega) = 1/\sqrt{1 + (\omega\tau_1)^2}. \quad (25)$$

Such a gain determines the frequency spectrum of the electrode potential ripple related to the pulsations of the electrolyte flow velocity. We assume that $\Delta V_0 = V_m \sin\omega_0 t$. In this case, voltage ripple depends on the pulsation frequency of the velocity:

$$\varphi(t) = \frac{\varphi_m}{\sqrt{1 + (\omega\tau_1)^2}} \sin\omega_0 t. \quad (26)$$

The above formulas show that the electrode voltage ripple is proportional to the pulsation of the liquid velocity and exhibits inertia with respect to the velocity pulsation characterized by time constant τ_1.

When adsorption takes place, the surface of electrode is covered with a monoatomic layer of oxygen. Then, oxygen is involved in the reaction with metal that leads to the formation of an oxide film. Oxygen is deposited on the oxide surface, so that the thickness of the oxide film increases due to the diffusion of oxygen atoms to the metal surface. The deposition rate becomes less than the limiting rate due to a decrease in the concentration gradient. Regions that are free of adsorbed oxygen and regions covered with adsorbed oxygen and oxide film may simultaneously exist on the electrode surface.

Thus, the surface of a metal electrode adsorbs oxygen that diffuses from the bulk of electrolyte. The adsorbed oxygen forms a monoatomic layer and the oxide film due to reaction with metal. Evidently, adsorption rate $\Delta m/\Delta t$ on the surface covered with adsorbed oxygen or oxide is significantly less than the adsorption rate for pure metal surface, area of which S decreases with increasing time:

$$S = S_0 - m/\rho d, \quad (27)$$

where ρ is the density of the monoatomic oxygen layer, d is the layer thickness (diameter of atom), $S_0 = La$ is the total area of the electrode, and m is the mass of deposited oxygen. We use $\rho = m_0/d^3$, where $m_0 = A/N_A$ is the mass of oxygen atom (A is the atomic mass of oxygen) to obtain

$$S = S_0 - \frac{mN_A d^2}{A}. \quad (28)$$

In accordance with the Fick law [11], the mass of oxygen that is transferred over time dt through area S is represented as

$$dm = SDA\frac{dc}{dy}dt. \quad (29)$$

With allowance for formula (28), expression (29) is written as

$$\frac{dm}{dt} + \frac{Dc_0N_Ad^2}{\delta}m = \frac{Dc_0S_0A}{\delta}. \quad (30)$$

Thus, we obtain the linear inhomogeneous equation of the process with time constant

$$\tau_2 = \frac{\delta}{Dc_0N_Ad^2}. \quad (31)$$

The solution is represented as

$$m(t) = \frac{S_0A}{N_Ad^2}(1 - \exp(-t/\tau_2)). \quad (32)$$

The area of pure metal decreases as

$$S(t) = S_0\exp(-t/\tau_2). \quad (33)$$

Time constant τ_2 can be calculated. For open water, the typical concentration of oxygen atoms at a temperature of 20°C is $c_0 = 0.5\times10^{-3}$ mol/L. For oxygen atoms in aqueous electrolyte, we have $D = 2.6\times10^{-9}$ m^2/s. Thickness of diffusion layer δ for an electrode with a length of $L = 2.5$ mm at a flow velocity of $V_0 = 0.5$ m/s is about 10^{-5} m. Substituting $N_A = 6.02\times10^{23}$ mol^{-1} and $d = 1.3\times10^{-10}$ m in expression (31), we obtain $\tau_2 \approx 5\times10^3$ s. The adsorption is a relatively slow process, and the rate is determined by the concentration of dissolved oxygen.

Formula (24) is obtained under the condition that oxygen is adsorbed on the entire area of the electrode surface. Therefore, area $S_0 = La$ in expression (24) must be changed by the area given by expression (33). Thus, we obtain

$$\varphi_m = \frac{13.4FDS_0c_0\Delta V_0\tau_1}{C_D\sqrt{LV_0\nu}}\exp(-t/\tau_2). \quad (34)$$

The capacitance of the DEL that is formed on the entire electrode surface is represented as

$$C_D = \frac{\varepsilon\varepsilon_0S_0}{\delta_D}, \quad (35)$$

where δ_D is the DEL thickness, $\varepsilon_0 = 8.85\times10^{-12}$ is F/m the electric constant, and ε is the permittivity between the DEL planes. For the dense part of the double layer, we have $\varepsilon \approx 3$. For the diffuse part, permittivity ε is close to 81 at a relatively low concentration of electrolyte [13]. For low-concentration solution of NaCl, the DEL thickness is [14]

$$\delta_D \approx \frac{3}{\sqrt{c_1}} \times 10^{-10} \text{ m}, \qquad (36)$$

where c_1 is the NaCl concentration in moles per liter. With allowance for expressions (33)–(36), the jump of electrode potential related to jump of flow velocity ΔV_0 is represented as

$$\begin{aligned} \Delta\varphi &= -\frac{40.2 \times 10^{-10} F D c_0 \Delta V_0 \tau_1}{\varepsilon\varepsilon_0 \sqrt{c_1} L V_0 \nu} \\ &\quad \times \exp(-t/\tau_2)(1 - \exp(-t/\tau_1)) \qquad (37) \\ &= -\Delta\varphi_m \exp(-t/\tau_2)(1 - \exp(-t/\tau_1)). \end{aligned}$$

For $c_1 = 0.077$ mol/L, $\tau_1 = 3.75 \times 10^{-3}$ s, $\Delta V_0 = 5 \times 10^{-3}$ m/s, $V_0 = 0.5$ m/s, $F = 9.64 \times 10^4$ C/mol, $L = 2.5 \times 10^{-3}$ m, $\nu = 10^{-6}$ m^2/s, and the above remaining parameter, we obtain $\Delta\varphi_m$ of several microvolts.

The analysis shows that the inertia of the electrode potential relative to the velocity pulsations caused by the inertia of oxygen atoms moving toward the electrode in the presence of osmotic pressure is characterized by a time constant of about 10^{-15} s and, hence, can be disregarded.

2. EXPERIMENTAL

The setup of [8] is used to experimentally study voltage ripple in a system that simulates two electrodes: plates made of the 12Kh18N10T stainless steel are placed in the NaCl solution with a concentration of 0.077 mol/L. The first (measurement) electrode with an area of 4.9 mm^2 (side surface of a cylinder with a diameter of 2.5 mm) is immersed in electrolyte the pulsating flow of which has a mean velocity of 0.5 m/s and a relative pulsation amplitude of 0.4% at the fundamental frequency and second harmonic of a centrifugal pump that provides the motion of liquid. The velocity vector is parallel to the plane of electrode surface. The second (reference) electrode with a significantly greater area of 63.5 mm^2 is not immersed in the liquid, so that the measured voltage ripple is related to the processes on the surface of the measurement electrode. The signal voltage of the sensor is amplified using a low-noise transformerless preamplifier with a relatively high input impedance. A coupling capacitor provides the absence of dc current. Prior to the measurements, the electrodes were stored in electrolyte over 17 days. The gain of the measurement system is 8.75×10^5. In the pipeline of the measurement setup, we use an optical sensor of the liquid flow velocity. The electrodes, pump, and pipeline are shielded to provide the absence of electric interference at the input of the preamplifier.

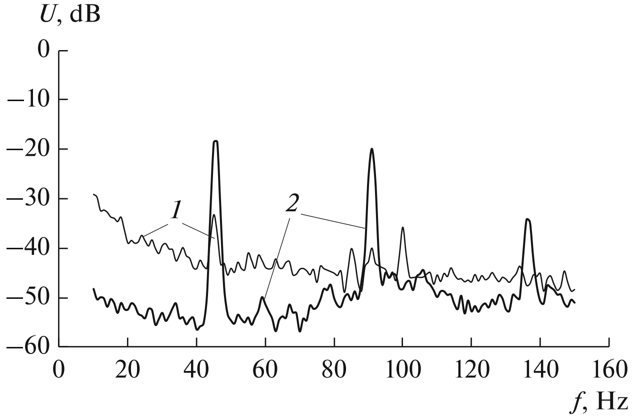

Fig. 2. Spectra of (*1*) electrode noise voltage and (*2*) voltage of velocity sensor for a liquid velocity of 0.5 m/s and a storage time in electrolyte (NaCl solution with a concentration of 4.5 g/L) of 17 days.

Figure 2 presents the averaged amplitude spectra of voltage ripple U for the two electrodes (curve *1*) and pulsations of the signal of the velocity sensor (curve *2*). The spectra are plotted on the logarithmic scale. The zero level on the ordinate axis corresponds to a spectral density of the noise voltage of 1 $\mathrm{V}/\sqrt{\mathrm{Hz}}$. The spectra exhibit corrugated structure the peaks of which correspond to the harmonics of the fundamental frequency related to the rotation rate of the centrifugal pump (the frequency of the first harmonic is 46 Hz). Note correlation of the peaks of the velocity-sensor signal and electrode noise. The peak amplitude at a frequency of 46 Hz in the spectrum of electrode noise is greater than the background noise level by 10 dB. To determine the time constant that characterizes the inertia of the voltage ripple relative to the velocity pulsations, we increase the peak amplitude at the second harmonic in the spectrum of velocity pulsations (curve *2* in Fig. 1) by 2 dB to a level of the first harmonic. The corresponding peak in the spectrum of electrode voltage is also increased by 2 dB. The resulting peaks in the spectra of electrode voltage at the first and second harmonics of the velocity pulsations correspond to frequency characteristic (25) with $\tau_1 = 4.4$ ms, which is close a value of 3.75 ms calculated with the aid of expression (20) for $L = 2.5$ mm and $V_0 = 0.5$ m/s.

An increase in the electrolyte concentration by a factor of 6 (from 4.5 to 27 g/L NaCl) leads to a decrease in the peak in the spectrum of electrode voltage at the velocity pulsation frequency by a factor of 2.05.

3. DISCUSSION

The velocity of the liquid flow on the sensor electrodes has a pulsating component caused by turbulence of variations in the mean velocity relative to the sensor. Such a velocity pulsation leads to voltage ripple of the sensor electrodes (i.e., flow noise).

Formula (37) makes it possible to interpret experimental regularities including correlation of the spectral peaks of electrode voltage and voltage of the velocity sensor. The peak in the spectrum of the sensor voltage at the second harmonic of the pulsation frequency is significantly lower than the peak at the first harmonic, and the peak at the third harmonic is not observed. The above analysis shows that such results are related to the inertia of electrochemical processes on the surface of the measurement electrode. The peak at a frequency of 85 Hz is caused by unknown interference, and the peak at a frequency of 100 Hz is related to the noise at the second harmonic of the industrial frequency. A decrease in the sensor sensitivity to liquid-velocity pulsations with an increase in concentration of the solution c_1 results from an increase in the DEL capacitance. An experimental decrease by a factor of 2.05 is in good agreement with a decrease by a factor of 2.45 obtained with the aid of formula (37). In the experiments, a decrease in the sensitivity of sensor to velocity pulsations with an increase in the storage time in electrolyte is due to a decrease in the oxygen adsorption rate (i.e., the effect of factor $\exp(-t/\tau_2)$). Oxygen adsorption in static electrolyte is even slower. Thus, the electrodes are stored in electrolyte over at least 3 days prior to the measurements.

The calculated electrode voltage ripple is in qualitative agreement with the experimental result. In particular, the peak amplitude of the spectral characteristic of the electrode voltage (about 30 nV) is less than $\Delta\varphi_m$ calculated with the aid of formula (37) by two orders of magnitude. Such a difference may result from long-time storage of electrodes in electrolyte.

The surface of electrodes is gradually covered with oxide. For metals that are used in the experiments (tantalum, titanium, and stainless steel), oxides exhibit dielectric properties. An increase in the thickness of the oxide layer leads to a decrease in the capacitance of electrode relative to electrolyte, so that the electrode sensitivity to velocity pulsations additionally decreases.

The electrolyte contains chlorine ions that expel oxygen from fragments of the electrode surface. The charge of chlorine ion is less than the charge of oxygen ion, and we obtain a negative bias of the electrode potential [11]. However, electrode oxidation is not terminated in the presence of chlorine and the adsorption of oxygen is continued. Moreover, efficient oxidation that occurs on relatively small surface fragments may lead to pitting corrosion. The liquid flow toward the electrode surface provides delivery of electrolyte with relatively high content of oxygen. Thus, the local interchange of the oxygen and chlorine adsorption may lead to oscillations of the electrode potential [13]. Such random oscillations affect the background noise level [9, 15].

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