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STATISTICAL RADIOPHYSICS

Noise of Electrode Sensor in Pulsating Liquid Flow

V. G. Maksimenko

Kotel'nikov Institute of Radio Engineering and Electronics (Fryazino Branch), Russian Academy of Sciences, Fryazino, Moscow oblast, 141190 Russia

e-mail: max54@ms.ire.rssi.ru Received May 23, 2016

Abstract—A periodic component of the ac electrode voltage (electrode noise) is revealed in experiments. The frequency of the periodic component is equal to the pulsation frequency of the flow velocity of liquid in which the electrode sensor is immersed. For the electrode sensor that moves in liquid such a component of the electrode noise may prevail and determine the sensitivity. The sensitivity of a sensor with stainless-steel electrodes

with respect to the velocity pulsations is determined to be 1.2×10^{-6} V s/m. Pulsations of the electrode potential can be interpreted using variations in the rate of electrochemical reaction on the electrode surface.

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INTRODUCTION

Electrode sensors are used for detection and measurement of low-frequency electromagnetic fields in seas. The intrinsic noise of sensor that is generated at the electrode-liquid interface determines the threshold sensitivity of the receiver. This circumstance accounts for the practical interest in the study of the electrode noise. The noise of electrode sensors in the absence of motion relative to liquid is well studied in [1-3]. Note also the analysis of noise of electrode sensors that move in liquid [4-7], which yields a relationship of the fluctuations of the electrode potential difference and liquid velocity fluctuations. However, the existing data must be specified to determine not only qualitative but also quantitative relationship of the pulsations of electrode potential and pulsations of the liquid flow velocity.

The purpose of this work is the further study of the electrode noise in the presence of the pulsations of liquid velocity, in particular, the analysis of the effect of storage time in electrolyte, electrolyte concentration, liquid velocity, pulsations of the velocity, and electrode area and material properties.

1. EXPERIMENTAL SETUP

Figure 1 shows the scheme of the experimental setup. Steel box *1* that serves as magneto- and electrostatic shield accommodates the second steel box (shield) 2, electric motor 3, accumulator (power supply of the motor) 4, and unit 5 for control of the motor rotation rate (bank of switched voltage dropping resistors). Centrifugal pump 6 connected with motor 3 by dielectric shaft 7 is placed in box 2. The pump provides

the motion of liquid (aqueous solution of NaCl that imitates sea water). Box 2 also contains cell 8 with two electrodes (model of sensor), preamplifier 9 with power supply, reservoir 10, liquid velocity sensor 11, and preamplifier 12 with power supply. Liquid passes thorough flexible dielectric pipeline 13 with a diameter of 3.5 mm from the pump to the cell with electrodes and, then, reservoir 10. The pump is modified to increase the liquid velocity pulsations.

To suppress electric interference generated by dc collector motor, we place the motor outside box 2. The motor and pump form a single unit due to a fragment of metal tube 14 that serves as electrostatic shield. Normally, the velocity of liquid is 0.5 m/s but variations are possible due to discharging of the accumulator.

To determine the noise level related to the electric motor, we use pipeline fragment 15 that shunts the cell with electrodes. In the measurements of the electrode voltage (except for the measurements of Section 2.4), the fragment is closed using clamp 16. Preamplifiers 9 and 12 are interfaced via switching unit 17 with measuring amplifier 18 the output signal of which is fed to ADC 19 and computer 20.

In the measurements using an E20-10 ADC, computer records electrode voltages and signals of the velocity sensor (series consisting of 20 samples with a duration of one second). Then, the data are processed using the LGraph2 software. The spectra of the electrode noise and spectra of signals of the liquid velocity sensor are ensemble-averaged.

To monitor the pulsations of liquid velocity, we employ photoelectric sensor 11 [8]. An opaque flag made of stainless steel is mounted on a leaf spring in the transparent fragment of the pipeline. A LED and a



Fig. 1. Block diagram of the setup for the measurement of electrode noise in the presence of liquid flow.

photodetector (photodiode) are mounted on different sides of the transparent pipe. The opaque flag partly blocks light that passes from the LED to the photodetector. Liquid flow leads to bending of the spring, so that the transmitted light is modulated in accordance with variations in the liquid velocity. The sensor is calibrated at different velocities of liquid. The corresponding signal can be used to determine quantitative relationship of the velocity pulsations and electrode voltage. However, we were unable to measure the frequency response function of the sensor due to technical reasons.

Electrode cell δ has a cubic sealed housing made of Plexiglas. The input and output pipes are mounted on the opposite faces of the housing. The measuring electrode is placed on the cover of the housing in such a way that the working surface is located at a distance of 5-10 mm from the nozzle of the input tube through which liquid is delivered to the cell. Thus, we assume that the flow velocity in the vicinity of the electrode is equal to the flow velocity in the tube. A reference electrode with the working side surface with a diameter of 9 mm is located at a certain distance from the flow. The working surfaces of electrodes are polished and passive side surfaces are isolated from liquid (painted). The input and output tubes of the cell are made of stainless steel. The electrodes are connected to an original transformerless low-noise preamplifier 9. In the experiments, we employ two different preamplifiers for different areas of the measuring electrode, since the resistance of the electrode sensor depends on the area and frequency [2]. For the major part of the working frequency interval, the input resistance of the preamplifier is greater than the magnitude of the total resistance of the sensor by at least an order of magnitude, and the noise coefficient of the preamplifier in the experiments is no greater than 2 dB.

2. MEASUREMENTS OF ELECTRODE NOISE

The experiments with pumping of liquid show that the ac component of the electrode voltage (electrode noise) contains a periodic component, the frequency



Fig. 2. Spectra of (a) electrode voltage and (b) voltage of the velocity sensor for a liquid flow 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.

of which is determined by the rotation rate of the electric motor. An increase in the rotation rate leads to an increase in the frequency of the periodic component. Figures 2a and 2b present the averaged spectra of the electrode noise and signal of the velocity sensor for a liquid flow velocity of 0.5 m/s and storage time in electrolyte (NaCl solution with a concentration of 4.5 g/L) of 17 days. The measuring electrode with a diameter of 2.5 mm and the reference electrode with a diameter of 9 mm are made of the 12Kh18N10T stainless steel. The gain of the measuring system (including the preamplifier) is 8.75×10^5 .

The spectra of Figs. 2–6 are plotted on the logarithmic scale with respect to $1 \text{ V}/\sqrt{\text{Hz}}$. The spectra of the electrode noise are plotted with allowance for variation in the coefficient of the voltage transfer from electrodes to preamplifier related to the effect of resistance and capacitance of electrode sensor.



Fig. 3. Spectra of electrode noise for storage times in electrolyte (4.5 g/L) of (1) 1, (2) 2, and (3) 17 days at a flow velocity of 0.5 m/s and (4) the spectrum obtained in the absence of liquid flow at a storage time of 17 days.

The spectra of electrode noise have ridged structure with peaks at harmonics of the fundamental frequency (the first harmonic is 46 Hz). The peak amplitudes are higher than the background noise by 10 dB. The spectral characteristics of the signal of the velocity sensor exhibit peaks at the same frequencies.

The velocity sensor is used to estimate pulsations of the liquid flow velocity. The pulsations are 0.4% at the first and second harmonics. The calibration procedure shows that the sensitivity of the velocity sensor is 7.4 V s/m. The absolute pulsation of the velocity at first harmonic is 0.02 m/s. For such a low level of the pulsations, the sensor is assumed to be linear, so that the pulsation of the electrode voltage is proportional to the velocity pulsation.

Below, we experimentally study the influence of several factors on the electrode noise. Let us consider them separately.

2.1. Storage Time in Electrolyte

Solution of NaCl with a concentration of 4.5 g/L serves as electrolyte. The measuring electrode with a diameter of 2.5 mm is made of the 12Kh18N10T stainless steel. The liquid flow velocity is 0.5 m/s. The gain of the measuring system is 8.75×10^5 . Figure 3 presents the spectra of electrode noise corresponding to storage times of 1, 2, and 17 days and the result obtained in the absence of liquid flow for a storage time of 17 days. The peak of the spectral characteristic of electrode noise at the first harmonic of the velocity pulsation decreases with time. For storage times of 1, 2, and 17 days, the peak amplitudes are -26.5, -29.8, and -33.2 dB, respectively. During the measurements, the peak of the spectral characteristic decreases by 6.7 dB. Note that the background electrode noise (the signal in the intervals between the peaks) also



Fig. 4. Spectra of electrode noise for a stainless-steel measuring electrode with a diameter of 2.5 mm at a flow velocity of 0.5 m/s, a storage time of 17 days, and NaCl concentrations of (1) 4.5 and (2) 27 g/L.

decreases. The mean background noise decreases from 22 dB at a frequency of 10 Hz to 5-6 dB at frequencies of 60-100 Hz. At frequencies of greater than 120 Hz, the motion of liquid does not affect the electrode noise. The peak amplitude is higher than the background level by 10-12 dB.

2.2. Electrolyte Concentration

Figure 4 shows the spectra of electrode noise at NaCl concentrations of 4.5 and 27 g/L. The measuring electrode with a diameter of 2.5 mm is made of the 12Kh18N10T stainless steel. The liquid flow velocity is 0.5 m/s. The storage time in electrolyte is 17 days. The gain of the measuring system is 8.75×10^5 . In the first experiments, the spectra are measured at a concentration of 4.5 g/L. Then, electrolyte is partly removed from reservoir 10, electrolyte with a higher concentration is added, and the pump is switched on. After mixing over several minutes, the second measurement is performed. An increase in the salt concentration by a factor of 6 leads to a decrease in the peak of the spectral characteristic at the first harmonic of the velocity pulsation by 6.2 dB, which is in agreement with the result of [4]. The background electrode noise slightly decreases only at frequencies of higher than 50 Hz. Apparently, such a decrease (about 2 dB at a frequency of 60 Hz) is due to a decrease in the active part of the

2.3. Area of Measuring Electrode

impedance of the electrode cell and a corresponding

decrease in the thermal noise.

To determine the dependence of fluctuations of the electrode voltage on the area of the measuring electrode, we use a single unit consisting of four insulated identical electrodes made of titanium and a stainless-



Fig. 5. Spectra of electrode noise for titanium electrodes with areas of (1) 0.78 and (2) 3.12 mm^2 at a storage time of 9 days and electrolyte concentration of 4.5 g/L.

steel electrode. The electrodes represent polished ends of wire with a diameter of 1 mm, so that the area of a single electrode is 0.78 mm^2 . The electrodes are stored in electrolyte (NaCl solution with a concentration of 4.5 g/L) over 9 days. The area of the titanium measuring electrode increases due to parallel connection of sections.

Figure 5 presents the spectra of electrode noise for two areas of the titanium measuring electrode. The liquid flow velocity is 0.5 m/s. The gain of the measuring system is 7.5×10^5 . When the area increases by a factor of 4, the spectral density of the background electrode noise decreases by about 6 dB. The peak of the spectral characteristic of the electrode voltage at the first harmonic of the velocity pulsation decreases by less than 2 dB. The remaining measurements also show insignificant effect of the electrode area on the peak amplitude. The difference between the peak amplitude and the background level increases to 15 dB.

2.4. Material of Measuring Electrode

Figure 6 shows the spectra of electrode noise for titanium and stainless-steel electrodes. Electrodes with a diameter of 1 mm (see Section 2.3) are stored over 8 days in electrolyte with a concentration of 4.5 g/L. The liquid flow velocity is 0.55 m/s. The gain of the measuring system is 7.5×10^5 . For the steel electrode, the peak at the first harmonic (-31.5 dB) is higher than the background level by 8.5 dB. For the titanium electrode, the peak at the first harmonic (-35 dB) is higher than the background level by 8.5 dB. Thus, the peak for the steel electrode is higher than the peak for the titanium electrode by 3.5 dB. The same difference is obtained for background electrode noise in a frequency interval of 30–100 Hz. Note that high background noise prevents observation of the peak of the spectral characteristic of the electrode



Fig. 6. Spectra of electrode noise for (1) titanium and (2) stainless-steel electrode at electrode diameters of 1 mm, a flow velocity of 0.55 m/s, a storage time of 8 days, and an electrolyte concentration of 4.5 g/L.

noise at the frequency of velocity pulsation at a relatively short storage time in electrolyte.

2.5. Velocity of Liquid Flow

The effect of velocity can adequately be estimated only in the absence of variations in the pulsation frequency caused by variations in the velocity, since the response time of the electrode sensor and the frequency response function of the velocity sensor are unknown. A decrease in the velocity of liquid in the absence of variations in the pulsation frequency is reached using shunt tube 14. When the tube is opened, the flow velocity in the cell decreases by a factor of 1.2. In comparison with the system with the closed shunt, the velocity pulsation at the first harmonic also decreases by a factor of 1.2 and the peak in the spectrum of the electrode noise at the first harmonic of the velocity pulsation decreases by a factor of about 1.3. With allowance for the spread of values related to nonsimultaneous measurements, we assume that the peak amplitude of the electrode voltage is proportional to the amplitude of the velocity pulsations. Thus, we prove that the velocity sensor in our experiments is linear. In alternative experiments, an increase in the velocity of liquid due to an increase in the rotation rate of the electric motor leads to an increase in the absolute level of the pulsations of velocity and electrode voltage. However, an increase in the peak at the first harmonic of the electrode voltage is less than an increase in the velocity pulsations. This circumstance indicates significant response time of the processes responsible for the electrode potential.

We determine the electrode sensitivity to the velocity pulsations. The velocity pulsation at the first harmonic is 0.02 m/s. The first harmonic of the electrode voltage recalculated to the input of the preamplifier is 24 nV, which is in agreement with the result of [7]. Thus, the electrode sensitivity to the velocity pulsa-

tions is estimated to be 1.2×10^{-6} V s/m.

3. DISCUSSION

The pulsations of electrode potential are interpreted in [6] using deformation of double electric layer on the electrode surface in the presence of the incident liquid flow. Apparently, the effect exists but the corresponding pulsations of electrode voltage are less than the experimental pulsations by an order of magnitude. We propose an alternative effect of the generation of pulsations of electrode potential in the presence of the pulsations of liquid velocity that makes it possible to interpret the experimental data on the pulsations of electrode potential and the above dependences.

A polished electrode absorbs atmospheric oxygen as a monoatomic layer on fragments of surface. When immersed in electrolyte, the electrode absorbs dissolved oxygen. The adsorbed oxygen is involved in the reaction with metal of electrode that leads to the formation of oxide the molecules of which are oriented in such a way that negatively charged oxygen ions are concentrated on the surface. In the absence of oxide (on the surface of noble metals), electron of metal is attached to oxygen atom, so that negatively charged oxygen ion emerges on the surface. In this case, the potential of metal is shifted to positive values relative to electrolyte [9]. The shift for the oxidized electrode differs from the shift for the electrode with adsorbed oxygen. Normally, the oxidized fragments on the electrode surface are supplemented with the fragments with adsorbed oxygen. The above processes lead to an excess negative charge on the electrode surface. An excess positive charge (ions of dissolved salt) is formed in the electrolyte in the vicinity of the electrode surface. Thus, a double electric layer is formed on the electrode surface that can be considered as a lossy capacitor one plate of which is liquid. The potential of electrode surface relative to electrolyte is determined by the ratio of the surface charge to the capacitance of the double layer. Such a potential changes, since the adsorption of oxygen and oxidation in the presence of adsorbed oxygen are not terminated.

In the vicinity of the electrode surface, the concentration of oxygen atoms is less than the bulk concentration in electrolyte, since oxygen atoms are involved in reaction with metal and adsorbed on the surface. Thus, the concentration of atomic oxygen changes in the electrolyte layer in the vicinity of the electrode. Such a layer is called diffusion layer, since oxygen arrives at the electrode surface primarily due to diffusion. In accordance with the first Fick law, the rate of oxygen deposition on a unit area of the electrode surface (diffusion flux density) is proportional to concentration gradient in the diffusion layer [9]:

$$\frac{dm}{dt} = D\frac{dc}{dy}.$$

Here, dm is the amount of deposited oxygen (in moles) per time dt; dc/dy is the gradient of concentration c (mol/L) along the y axis that is perpendicular to the surface, and D is the diffusion coefficient of oxygen ($D \approx 10^{-9} \text{ m}^2/\text{s}$). The thickness of the diffusion layer in the liquid flow that is incident on the electrode depends on the flow velocity. The concentration gradient can be calculated as a ratio of the bulk concentration of oxygen in electrolyte to the thickness of the diffusion layer. Thus, the gradient depends on the liquid flow velocity. The results of [10] for the laminar flow in the vicinity of a semi-infinite smooth plate show that the thickness of the diffusion layer can be calculated as

$$\delta \approx 3 \left(\frac{D}{v}\right)^{1/3} \sqrt{\frac{vx}{V}}.$$

Here, v is the kinematic viscosity of liquid, x is the distance from the front edge of the plate, and V is the velocity of liquid at a relatively large distance from the surface. At an infinite rate of the surface adsorption of the diffusing substance (oxygen), the diffusion flux density on the electrode surface exhibits power dependence on the flow velocity:

$$\frac{dm}{dt} = kV^n,\tag{1}$$

where k is the proportionality factor. Exponent n in formula (1) may range from 0.5 (laminar flow around smooth surface) to 0.9 (turbulent flow around rough surface) [10]. In addition, the liquid flow that is not depleted due to diffusion is incident on the front edge of the electrode, so that the tangential transportation of oxygen takes place and exponent n increases for the diffusion flow of oxygen to the electrode. For electrodes with sizes of the working surface of no greater than several millimeters (see the above experimental structure), exponent n can be close to unity.

The above effects can be used to interpret the experimental relation of the pulsations of electrode potential and velocity of liquid. In the presence of the pulsations of the velocity of liquid, the thickness of the diffusion layer is modulated and the oxygen adsorption rate and the electrode potential are varied in accordance with variations in the velocity. The electrode surface is gradually covered with oxide that exhibits dielectric properties for titanium and stainless steel. Thus, the structure of the double electric layer on the electrode surface changes, so that the diffusion rate and the sensitivity of electrode to pulsations decrease. An increase in the thickness of the electrode relative to electrolyte [2]. Such an effect also

results in a decrease in the sensitivity of the electrode to velocity pulsations. We obtain the corresponding results in the experiments.

A decrease in the electrode sensitivity to the pulsations of the velocity of liquid with an increase in the concentration of the solution can be due to an increase in the capacitance of the double electric layer. The thickness of the double electric layer in the NaCl solution with a relatively low concentration is given by [11]

$$d \approx \frac{3}{\sqrt{c}} 10^{-10} \text{ m,}$$
(2)

where c is the NaCl concentration in mol/L. An increase in concentration c causes a decrease in thickness of double layer d and an increase in the capacitance, so that the pulsation of the electrode potential decreases. In the experiments, a six-fold increase in the electrolyte concentration leads to a decrease in the pulsation of the potential of the measuring electrode by a factor of 2.04.

Chlorine ions expel oxygen from fragments of the electrode surface. The charge of chlorine ion is less than the charge of oxygen ion, so that the electrode potential is negatively shifted [9]. However, the presence of chlorine does not lead to the termination of oxidation and adsorption of oxygen continues. Note that efficient oxidation occurs at relatively small fragments of the surface and pitting corrosion may take place. In the presence of the liquid flow, electrolyte with a relatively high concentration of oxygen arrives at the electrode surface. Thus, the interchanging of the oxygen and chlorine adsorptions may lead to oscillations of the electrode potential [12]. Such random oscillations affect the background noise level.

A four-fold increase in the area of the measuring electrode leads to a two-fold decrease in the background noise and weakly affects the peak amplitudes of the spectral characteristic of electrode noise at the pulsation frequency of the velocity of liquid and the corresponding harmonics. As was mentioned, the double electric layer that exists at the electrode-electrolyte interface and the oxide film that covers the electrode surface form a lossy capacitor. The pulsation of electrode potential gives rise to a pulsating electric charge at the liquid plate of the capacitor. Correlated pulsations of velocity at the sections of electrode lead to the correlated pulsations of electric charge. When four sections are connected, the electrode charge increases by a factor of 4 but the electrode capacitance also increases by a factor of 4, so that voltage pulsation at the electrode does not depend on the electrode area. Uncorrelated electrode noise is summed up with respect to power, and the results of [3] show that the amplitude spectrum of the electrode noise must decrease by a factor of 2 when the electrode area increases. We observe such a dependence of the electrode noise on the area of the measuring electrode in the experiments. A four-fold increase in the area of the measuring electrode leads to a decrease in the spectral density of the background noise by 6 dB and the corresponding difference of the peak amplitude at the first harmonic in the spectrum of electrode noise and the background level increases to 15 dB. A minor decrease in the peak amplitude in the spectrum of electrode noise at the pulsation frequency of the velocity of liquid for the system with the parallel connection of the electrode sections can be due to the absence of total correlation of the velocity pulsations in the vicinity of the sections.

The experimental results show that the electrochemical reaction (oxidation of metal electrode surface) is the main reason for the electrode noise in the presence of electrolyte flow. The growth rate of the oxide layer gradually decreases, and the electrode noise is stabilized. Figure 2 presents the corresponding spectral characteristics.

The turbulence caused by the pipeline and velocity sensor slightly affects the experimental results, since the velocity pulsations in the intervals between the peaks of the spectral characteristic are lower than the pulsations at the first and second harmonics of the pump rotation rate by 30-35 dB (Fig. 2). In the spectrum of electrode noise, the background noise level between the peaks is less than the peak amplitudes by 10-20 dB. Thus, the effect of turbulence pulsations is not observed.

To determine the level of noise caused by electric motor, we close the input pipe of the cell with electrodes while the shunting pipe remains opened. The pump provides the electrolyte flow that does not pass through the cell, and we measure the electrode voltage in such a configuration. The experiments show the absence of electric interference caused by the motor in the measurement cell. The dependence of the peak amplitudes in the spectra of electrode noise on the electrolyte concentration additionally proves the absence of the effect of external noise. However, the peaks at a frequency of 100 Hz in the spectra of Figs. 3, 5, and 6 are due to the second harmonic of the mains frequency and the peak at a frequency of 85 Hz in Fig. 2a is related to unknown external interference.

Thus, the fluctuations of electrode voltage are directly related (proportional) to the fluctuations of the velocity of liquid. The effect can be used in practice in the study of turbulence in liquid flow [13]. Metals with the minimum intrinsic electrochemical noise (e.g., titanium and stainless steel) must be used as electrode materials.

CONCLUSIONS

Electrochemical processes on the electrode surface serve as the main reason for the electrode noise. Pulsations of the velocity of liquid affect the rate of such processes and, hence, pulsations of the electrode voltage. The pulsations of the electrode voltage are proportional to the pulsations of the velocity of liquid. The pulsations decrease with an increase in the storage time in electrolyte and electrolyte concentration. As distinct from the background electrode noise, the pulsations of electrode voltage are almost independent of the electrode area. The noise of electrode sensor with larger electrodes (an area of greater than 1 cm²) that is stored in electrolyte over at least ten days is predominantly determined by the pulsations of the flow velocity. The sensitivity of the sensor with stainless-steel

electrodes is 1.2×10^{-6} V s/m. Note that velocity pulsations of less than 0.5% cause pulsations of electrode voltage that are greater than the background noise level by 10–15 dB. Such a result indicates that the electrode sensor can be used for measurements of pulsations of the velocity of liquid.

Both pulsation of electrode voltage related to the velocity pulsation and the background noise decrease with an increase in the storage time in electrolyte and NaCl concentration. An increase in the area of the measuring electrode results in a decrease in the back-ground noise and weakly affects the voltage pulsation at the frequency of the velocity pulsations, so that the corresponding peak in the spectrum of electrode noise becomes relatively higher. Titanium electrodes are superior to stainless-steel electrodes with respect to the noise (the difference is about 3.5 dB).

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