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OPTIMIZATION OF BALANCING IN A BRIDGE MEASURING CIRCUIT WITH A DIFFERENTIAL CONDUCTOMETRIC SENSOR

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The article is devoted to reducing the influence of sources of an additive error in determining local changes in the electrical conductivity of electrolyte solutions under conditions of changes in the background electrical conductivity of the measurement medium, which often occurs in biosensor and other systems with a differential pair of conductometric transducers when their electrical parameters are not identical. The goal is to provide a deep suppression of the influence of background changes with significant differences in both reactance and active resistance in the transducers of a pair of sensor. The essence of the issue, the causes and mechanism of this type of error, as well as the methods and means of its reduction, developed earlier, are briefly considered. A diagram and description of the structure of a differential conductometric channel of a biosensor system based on an AC bridge, an algorithm for its balancing operations by controlling the module and phase of the test voltage, as well as a vector diagram of currents and voltages in the bridge circuit during this process. The balancing of the bridge has been was modeled analytically, bringing it to a quasi-equilibrium state, in which changes in the background electrical conductivity do not change its output signal. Additional operations for balancing the bridge are determined to achieve such a state with significant differences in both capacitances and active resistances in the impedances of a pair of conductometric transducers of a differential sensor. The results of experimental studies of the suppression of the influence of changes in the background electrical conductivity of a solution in a differential conductometric channel with using its computer model and experimental sample of a conductometric instrument with an electrical equivalent of a differential sensor are presented. A comparison of the results obtained and the corresponding data for balancing bridge circuits by previously developed methods is given. References 16, figures 3, tables 3.

Key words: differential conductometric biosensors, impedance, measurement, common mode influences, equivalent electrical model.

Introduction. Determination of changes in the parameters of the complex electrical conductivity of electrolyte solutions can provide important information about the state of the environment, the quality of technological materials and food products, and can be used in biotechnology, medical and technical diagnostics [1–3]. In many cases, the measurement of values of active conductivity (conductometry) [4] can be used. In particular, the use of biosensor methods implemented using a two-electrode conductometric transducer (sensor) with a planar interdigitated comb topology holds great promise [5–7]. Such a transducer cover with a selective biochemical membrane. An informative physical quantity during measurements is a local change in the electrical conductivity of the buffer solution in the measuring cell due to an increase in the concentration of electric charge carriers between the sensor's electrodes in the result of a biochemical reaction in the membrane after contact with the analyte.

A serious obstacle to improving the accuracy, sensitivity and reliability of conductometric measurements is the dependence of the background electrical conductivity of the buffer solution on its temperature, changes in concentration during measurements, the presence of various impurities and other non-informative factors. To eliminate it, the method of differential measurements is used using a differential sensor, consisting of an active transducer associated with the object under study (working one), and a passive (reference one) that determines the background conductivity of medium in a measurement sell. In biosensor

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ORCID ID: [*] https://orcid.org/0000-0002-4470-4339; [**] https://orcid.org/0000-0003-1363-9252; [***] https://orcid.org/0000-0002-1043-9426; [****] https://orcid.org/0000-0002-9528-5808 systems, they constitute a differential pair of identical primary transducers with selective (active) and neutral (passive) membranes, respectively. These transducers, placed in the buffer solution of the conductometric cell, are included in opposite branches of the bridge circuit, which is powered by an alternating current voltage source with a frequency of several tens of kHz [8–10]. When an analyte is added to buffer solution, a local change in the electrical conductivity of the solution occurs in a selective membrane, which is proportional to the concentration of the analyte in the analyte. It causes the response on the biochemical reaction, the magnitude of which is determined by the change in current in the working converter and at the output of the bridge. Such technology can be used not only for biosensors, but also for many other measurements in liquid media.

The change in background conductivity in this case is common mode influence. Under ideal conditions, with identical parameters of the equivalent electrical circuit of the conductometric transducers and in the absence of an analyte, the bridge is balanced. Therefore, changes in currents in the working and reference transducers due to changes in the background electrical conductivity of the solution are mutually subtracted at the output of the bridge and do not affect the informative signal at its output. However, in practice it is very difficult to ensure the identity of these parameters [11, 12].

A simplified equivalent circuit of a two-electrode conductometric transducer at optimal frequencies is a series RC circuit in which the active resistance of its impedance R is determined mainly by the solution resistance and depends on the geometric dimensions of the electrode array. This parameter can have a sufficiently high accuracy due to the use of modern technologies for the manufacture of transducers. The parameter C is mainly determined by the reactive (capacitive) resistance $1/\omega C$ of the near-electrode double layer, which depends from the state of the electrode surfaces and cannot be stable. This leads to a change in the phase angle of the converter and to a change in both the phase and the amplitude of the current in it under constant test voltage on the transducer. Therefore, even if changes of the impedance of a transducer with series equivalent circuit are due to a change in the capacitance alone, there is also a change in voltage across the active component of the impedance – that is, across the solution under test. In this case, the increment of current in the transducer changes with the same changes in the conductivity of the solution.

In a differential conductometric system, the indicated changes in the capacities of the working and reference transducers can differ significantly. This leads to the appearance of a false response at the output of the bridge due to in-phase non-informative impact on the converters caused by changes in the background electrical conductivity during the measurement process, i.e. to the additive measurement error. That is especially noticeable when working with electrically conductive analytes [11–14].

It should also be taken into account that in the electrode region there are also active charge transfer resistances, which are shunted by the capacitance of the double layer. However, at frequencies optimal for measurements (tens of kHz), this shunting is not complete; therefore, the equivalent resistance R of a two-element chain has a certain component, which is determined by the charge transfer resistance and near-electrode capacitance. This component can be determined by recalculating the parameters of the parallel connection of the charge transfer resistance with the double layer capacitance into equivalent (at the operating frequency) parameters for their series connection. If the changes in the near-electrode capacitances of the differential sensor transducers are not the same, the indicated components R in their equivalent two-element circuits turn out to be different. That additionally changes the ratio of active resistances and, accordingly, worsens the suppression of the effects from the common mode influence caused by changes in the background electrical conductivity of the solution.

In order to avoid this phenomenon, in conductometric systems with a bridge circuit for comparing currents (which provides a fixed voltage mode on the measurement object), the bridge is balanced on the output current component, which is quadrature to the test voltage, by compensating of the voltage drop across the capacitances of the converters [10, 12, 13]. The result of this is the equality of the amplitudes of the supply voltage of the bridge and the antiphase test voltages on the active resistances of converters in the differential sensor and, respectively, the equality of changes in the amplitudes of the antiphase currents in them is achieved under changes the background electrical conductivity.

However, full mutual compensation of changes in sensor currents under changes in the background electrical conductivity is not obtained in this case, since the changes in currents differ in phase due to the difference in the phase angles of the conductometric transducers of the sensors [12, 13]. The way to solve this problem was first proposed in [15]. It consists in bringing the bridge to a quasi-equilibrium state by additionally adjusting the phase of the test voltage in its reference branch, due to which the collinearity of the vectors of changes in the currents of the converters is achieved. In that case the output signal of the bridge is maintained unchanged when the background electrical conductivity of the solution changes. The disadvantage of the solution described in [15] is the need to use in the bridge circuit analog signal converters: DAC, adders, inverters, phase shifters. This complicates the device circuit and its adjustment, limits the frequency range and measurement accuracy. When using bridge circuits with digital generators of test and reference voltages, analog converters may be absent. Such an AC bridge is balanced by regulating the ratios of the modules and phases of the output voltages of the generators, which are fed to the working and reference converters [16].

The collinearity of the vectors of changes in the currents of the converters is achieved by additional adjustment of the voltage phase on the reference converter. Theoretical and experimental studies of such balancing of the bridge circuit have shown possibility of a high degree of suppression of common-mode interference from changes in the background electrical conductivity with differences in the capacitances of the differential sensor transducers. However, with differences in their active resistances, its effectiveness turned out to be much lower.

The purpose of this work is to provide a deep suppression of the influence of changes in the background electrical conductivity of the solution during differential conductometric measurements under conditions of significant differences in both reactive and active resistances of the sensor's transducers. A new method is proposed for bringing the bridge to a quasi-equilibrium state, which differs from those used previously to do this.

Let us consider this problem in more detail. Balancing of the bridge on phase of the currents in the converters of the differential sensor in the device described in [16] is performed by rotating the phase of the test voltage, and not by adding a quadrature component to it, which compensates for voltage drops on the near-electrode capacitance of the converter, as in [10, 12, 13]. In this case, the normalized test voltages, which are set by the generators in the bridge branches, are distributed on the reactive and active resistances of the differential sensor converters into the corresponding quadrature components in proportion to their values. The modules and phases of these components may have different ratios in the working and reference transducers due to differences in their phase angles. If this difference is associated only with a change in the reactive component of the impedance, first, the bridge completely balanced by adjusting the amplitude and phase of the test voltage on the reference converter. Then, quasi-equilibrium state of the bridge is established by additional rotation of its phase by the phase difference of the angles of the differential pair of transducers [16]. It makes possible to achieve equality of voltages across the active resistances of the conductometric transducers. In this case, the current responses in the bridge branches to in-phase interference from changes in the background electrical conductivity of the solution are mutually compensated with high accuracy.

However, if the active resistances of the working and reference sensors differ, this compensation is not complete, since in a bridge circuit with current comparison, during balancing, the currents in the sensors are equalized, and not the voltages across them. Of course, you can use a bridge circuit with a comparison of the voltages on the sensors, but at the same time, they must operate in the mode of a given current (galvanostat). This means that voltages on conductometric transducers can vary within a wide range during measurement, which is unacceptable both for biosensors and in many other cases. This mode requires different approaches to performing measurements.

Structure and adjusting of the measuring channel.

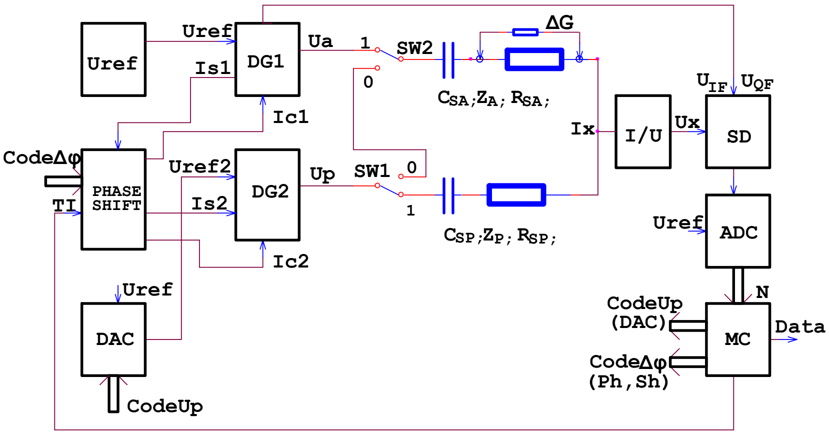

Fig. 1

A simplified structure of the differential conductometric channel of a biosensor system with a compensating-bridge circuit with current comparison, which is balanced by adjusting the ratios of test voltages on the sensors of the differential pair by modulo and by phase, is shown in Fig. 1.

The bridge circuit contains two branches formed by digital generators DG1, DG2 of stepped quasi-sinusoidal signals and conductometric converters of the differential biosensor, which are shown as equivalent *RC* circuits R_{SA}, C_{SA} (active, working sensor) and R_{SP}, C_{SP} (passive, reference sensor) with impedances Z_A and Z_P. A stable constant reference voltage *Uref* powers the bridge. This voltage is supplied unchanged to the working branch, and to the reference branch – through a digital-to-analog converter DAC. The digital oscil-lators generate quasi-sinusoidal voltages *Ua* and *Up* with n steps per period under the influence of clock pulses I_C. The frequency f_W of these voltages is n times lower than the frequency f_C of the I_C pulses, which are formed from the clock pulses TI of the microcontroller with a frequency f_T. Their phase relationships are set by synchronization pulses: I_{S1}, which is generated by the master generator DG1, and I_{S2}, which sets the initial phase of the slave generator DG2, and is shifted in time relative to I_{S1} using the Phase Shift digital delay by an integer number of quasi-sinusoid steps. More discrete control of the phase shift is carried out by shifting I_{C2} relative to I_{C1} by an integer number of periods of frequency f_T, which is an integer number of times higher than the frequency f_C. The control codes for the phase shift Δφ and the voltage amplitude *Up* DG2 are formed by the microcontroller mkC. The bridge balancing process is controlled by the microcontroller software according to the processing result of the bridge imbalance signal I_X, which is converted into voltage U_X by the *I*/*U* converter. Then, an in-phase or quadrature component with *Ua* is extracted from it using a synchronous detector SD with mutually quadrature reference signals U_{IF} or U_{QF}, which are converted into a digital code using ADC. The configuration of the bridge circuit can be changed with the SW switches to preset the measurement channel and to measure the result of the biochemical reaction in the working sensor. The structure of this measuring channel and its functioning are considered in more detail in [16].

The microcontroller program performs the preparation of the channel for measuring of the change in the specific electrical conductivity of the solution in the working conductometric transducer in the form of a sequence of the operations for bringing the bridge circuit to its initial quasi-equilibrium state. In this state, the errors from the instability of the parameters of the equipment and the measurement environment are minimal. In [16], an algorithm and a vector model of a variant of such tuning are described in detail, which ensures its good accuracy in the case of instability of the near-electrode capacitances of transducers of a differential pair. Therefore, in this article they are shown in a simplified form, followed by a detailed description of new operations that reduce the error also from non-identities of the active resistances of the working and reference sensors. The simplified vector model of such measuring process is shown in Fig. 2.

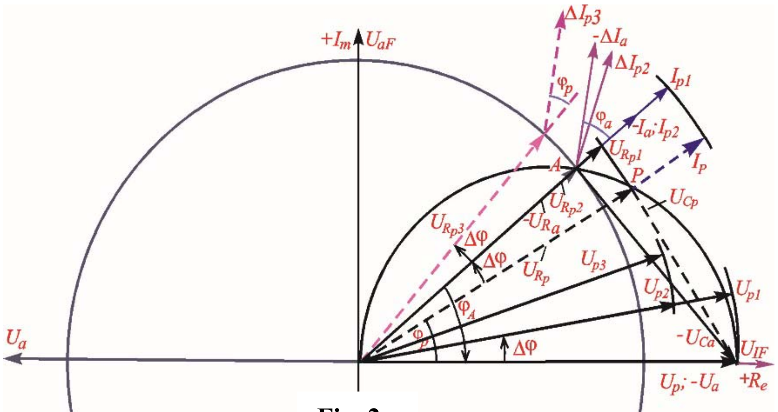

Fig. 2

The whole measurement process consists of three stages. At the first of them, a preliminary full balancing of the bridge circuit is performed, in the result of which the amplitude of the bridge output signal becomes close to zero. At this stage, the values of diagnostic parameters of the impedance of conductometric transducers R_{SA}, C_{SA} and R_{SP}, C_{SP} are determined. The values of tangents of their phase angles tgφ_A, tgφ_P are calculated. At the second stage, the bridge is set to the calculated state of quasi-equilibrium, in which the output signal of the bridge is not close to zero, but it does not change with the same, in-phase changes in the parameters R_{SA} and R_{SP}. At the third stage, a sample of the analyte is introduced into the conductometric cell. As a result of a chemical reaction, a local change in the electrical conductivity of the solution occurs on the active (working) transducer producing changing R_{SA}. That changing is converted in the bridge circuit into an informative signal, the digital code of which is proportional to the concentration of the analyzed analyte.

These voltages in the bridge circuit are anti-phase, but, for clarity of the signal's comparison in the bridge, *Ua* is shown in reverse phase. The process of the preliminary balancing of the bridge is the gradual reduction of the amplitude of its output signal to the minimum possible value by successively changing the values of the phase shift codes Δφ and the amplitude *Up*. The *Up* vector rotates to the *Up*1 position and its amplitude decreases (to the *Up*2 position). The current vectors of the active (−*Ia*) and passive (*Ip*2) transducers coincide in this case (the bridge is balanced). If R_{SA} and R_{SP} are equal and C_{SA} and C_{SP} are different, the result of the first tuning stage is the alignment of the ends of the voltage vectors U_{Ra} and U_{Rp} along the arc between points P and A. The voltage vector across the active resistance of the passive transducer takes on the value U_{Rp2}. The current increment vectors $-\Delta Ia$ and $\Delta Ip2$ have the same amplitude during in-phase action on the transducers in this state of the bridge, but different phase angles relative to the current vectors (they are equal to the phase angles φ_a and φ_p, respectively). To eliminate this difference, the bridge is transferred to a state of quasi-equilibrium at the second stage of tuning by an additional rotation of the voltage vector on the passive sensor from the position U_{p2} to U_{p3} by an angle $\Delta\varphi$. After that, the increments of currents (now $-\Delta Ia$ and $\Delta Ip3$) after common-mode influence on transducers become collinear (with opposite phases) and equal in absolute value, and therefore cancel each other in the output signal of the bridge.

Achieving the necessary quasi-equilibrium of the bridge with a difference in the active resistances of conductometric transducers. There is equality of the currents of active ($-I_A$) and passive (I_{P2}) transducers in the equilibrium state of the bridge. Therefore, the voltage on them U_{RA} and U_{RP2} (that is, voltages on the solution in the transducers) turns out to be different, if the active resistances of a conductometric transducers pair are different. Consequently, the current increments with a change in the background electrical conductivity of the solution will also differ, their mutual compensation at the output of the bridge will not be complete, and the resulting additive error will not be completely suppressed.

This problem can be overcome by changing the operation of balancing the currents of the working and reference arms of the bridge in order to bring the bridge into a slightly different state of quasi-equilibrium. After reaching the equality of the modules of their currents (at the first stage of setting up the measuring channel), the reference transducer's current module is corrected, depending on the ratio of the active resistances of the differential pair of transducers. This correction transfers the bridge into a state of quasi-equilibrium (in terms of current) and equilibrium in terms of voltages across their active resistances. The correction is calculated in such a way that the voltage drops on the active components of the impedance of the working and reference transducers are as close as possible. The initial data for such calculations are the results of diagnostics of the parameters R, C, tgφ of conductometric transducers, which is performed at the first stage of channel tuning. There are some features of their definition, related to the fact that the equivalent circuit of the transducers is a serial RC-chain, and the parameters of the parallel equivalent circuit (G and ωC) are measured.

In differential conductometric devices, the measured parameter is the conductivity, and the informative signal of the bridge measuring circuit is its output current. Therefore, measurements are made using a parallel transducers equivalent circuit (active and reactive conductance, phase angle tangents and their differences), and the necessary diagnostic parameters (active and reactance according to a series equivalent circuit) are obtained by calculations. To do this, the relations known from theoretical electrical engineering are used to recalculate the parameters of a parallel circuit (indices P) into a serial circuit equivalent at a working operating frequency ω_w (indices SE) and to recalculate the parameters of a serial circuit (indices S) into equivalent parameters of a parallel circuit (indices PE):

$$R_{SE}=R_p\left(1+\left(\omega_w C_p R_p\right)^2\right)^{-1}; \qquad C_{SE}=C_p\left(1+\left(\omega_w C_p R_p\right)^{-2}\right);$$

$$R_{PE}=R_S\left(1+\left(\omega_w C_S R_S\right)^{-2}\right); \qquad C_{PE}=C_S\left(1+\left(\omega_w C_S R_S\right)^2\right)^{-1}.$$

In these expressions, the terms $\omega_w C_P R_P$ and $1/(\omega_w C_S R_S)$ are the values of the phase angle tangent of the parallel and series RC circuits at the frequency ω_w, respectively.

The above correction can be carried out by multiplying the value of the DAC control code that generates the U_{p2} voltage applied to the reference transducer after the bridge is fully balanced by some factor. It equal to the ratio of the active resistances of the reference and working transducer or the inverse ratio of their conductivities in a series equivalent circuit. However, as our studies have shown, this correction is not sufficient, since the amplitudes of current increments in transducers with a change in the specific electrical conductivity of the solution depend not only on the voltage across the solution, but also on the values of the phase angles of the transducer [12, 13]. Therefore, it is necessary to introduce an additional correction of the correction factor, which takes into account the ratio of the dependences of the increments of the of the active and passive transducer's current on their phase angles. The rationale for the values of additional corrections on the mathematical model of the process of setting the measuring channel is given below.

The complete equivalent circuit of two-electrode conductometric transducers is complex. With this in mind, it is advisable to check the compliance of their equivalent circuit with a serial two-element RC circuit at the used operating frequency of this device when performing diagnostics. It is not a problem to measure the above diagnostic parameters with (for example) a twofold change in the operating frequency, since there are no frequency-dependent blocks in bridge circuits with digital generators of test and reference voltages. The adequacy criterion is a significant excess of the change in reactive conductance compared to the change active one with a change in frequency for each of the converters. An important criterion for the quality of transducers is also the closeness of their active conductivities at both frequencies. If these criteria are not met, it is necessary either replace the sensor or, if possible, select a more optimal operating frequency.

Let us consider mathematical expressions characterizing the process of bringing a bridge measuring circuit with a differential biosensor to a state of equilibrium and quasi-equilibrium. Each of the sensors is represented as a two-element equivalent circuit having two elements connected in series – the electrical conductivity of the solution and the electrical capacitance, which characterizes the processes in the near-electrode layer of the solution. Let us designate the impedance parameters of the sensors as follows: G_{SA} is the electrical conductivity of the active sensor, C_{SA} is the capacitance connected in series with it, G_{SP} is the electrical conductivity of the reference sensor, C_{SP} is the capacitance connected in series with it. Let us use the method of complex amplitudes for calculating electrical circuits operating under the action of alternating voltages of a sinusoidal shape with a frequency ω.

The working (active) sensor is supplied with the voltage of an independent (leading) generator with a fixed amplitude and phase, which has only an in-phase component: $\dot{U}_a = U_a$. The reference (passive) sensor is supplied with the voltage of a dependent, regulated generator, which changes when the bridge is balanced: $\dot{U}_p = \left|\dot{U}_p\right| \cdot \exp(\varphi_p)$. In this expression $\left|\dot{U}_p\right|$ – the modulus of the regulated voltage of the dependent generator, φ_p is the angle of its phase shift relative to the voltage of the independent generator. The complex currents through the sensors are defined by the following expressions:

$$\dot{I}_{a1} = \frac{U_a}{\dfrac{1}{j\omega C_{SA}} + \dfrac{1}{G_{SA}}} = U_a G_{SA} \frac{\exp[arctg(tg\varphi_A)]}{\sqrt{1+tg^2\varphi_A}};$$

$$\dot{I}_{p1} = \frac{\left|\dot{U}_p\right| \exp(\varphi_P)}{\dfrac{1}{j\omega C_{SP}} + \dfrac{1}{G_{SP}}} \left|\dot{U}_p\right| G_{SP} \frac{\exp(\varphi_P) \cdot \exp[arctg(tg\varphi_P)]}{\sqrt{1+tg^2\varphi_P}},$$

where $tg\varphi_A = G_{SA}/\omega C_{SA}$; $tg\varphi_P = G_{SP}/\omega C_{SP}$ are phase angle tangents φ_A and φ_P of sensor impedances.

Balancing the measuring circuit is to achieve equality of the modules and phase shifts of the currents I_{a1} and I_{p1}. Based on this condition, we find the relative value of the modulus of the regulated voltage, the dependent generator to the voltage of the independent generator ND1 (this is the DAC control code) and the phase shift between these voltages $\Delta\varphi_{p1}$ corresponding to the equilibrium state of the measuring circuit:

$$\Delta\varphi_{p1} = \varphi_A - \varphi_P; \qquad ND_1 = \frac{G_{SA}}{G_{SP}} \cdot \frac{\sqrt{1+tg^2\varphi_P}}{\sqrt{1+tg^2\varphi_A}}. \tag{1}$$

Let us determine the parameters of the state of quasi-equilibrium of the measuring circuit, in which the effect of changes in the background electrical conductivity on the measurement result of local changes in the electrical conductivity will be minimal. During conductometric measurements, the background electrical conductivity of the solution in the measuring cell changes and, accordingly, the active electrical conductivities in the sensors. This is equivalent to multiplying them by the same coefficient K_f:

$$G_{SA}^{\Delta} = K_f G_{SA} = G_{SA} + \Delta G_{SA} = G_{SA} + (1-K_f) \cdot G_{SA}, \tag{2}$$

$$G_{SP}^{\Delta} = K_f G_{SP} = G_{SP} + \Delta G_{SP} = G_{SP} + (1-K_f) \cdot G_{SP}. \tag{3}$$

The complex current through the active sensor I^{Δ}_a after a change in the specific electrical conductivity of the solution and its change ΔIa are described by the expressions:

$$\dot{I}_a^{\Delta} = \frac{U_a}{\dfrac{1}{j\omega C_{SA}} + \dfrac{1}{G_{SA}+\Delta G_{SA}}}; \Delta\dot{I}_a = \frac{U_a}{\dfrac{1}{j\omega C_{SA}} + \dfrac{1}{G_{SA}+\Delta G_{SA}}} - \frac{U_a}{\dfrac{1}{j\omega C_{SA}} + \dfrac{1}{G_{SA}}}. \tag{4}$$

After transformations, we have:

$$(\Delta\dot{I}_a = U_a\left(\frac{G_{SA}+\Delta G_{SA}}{1-j\cdot tg\varphi_A^{\Delta}}-\frac{G_{SA}}{1-j\cdot tg\varphi_A}\right), \tag{5}$$

where $tg\varphi_A^{\Delta} = tg\varphi_A + \Delta tg\varphi_A$, $\Delta tg\varphi_A = \dfrac{\Delta G_{SA}}{\omega C_{SA}} = (1-K_f)\cdot tg\varphi_A$.

Bringing the expression in brackets (5) to a common denominator and expanding the brackets gives:

$$\Delta\dot{I}_a = U_a\frac{\Delta G_{SA}}{1-j\cdot(2tg\varphi_A+\Delta tg\varphi_A)-tg^2\varphi_A-tg\varphi_A\cdot\Delta tg\varphi_A}. \tag{6}$$

Let's determine the module and the phase of current change through the active sensor. The phase is determined by the denominator (6):

$$\varphi_a^{\Delta} = arctg\left(\frac{2tg\varphi_A\left(1+\dfrac{1-K_f}{2}\right)}{1-tg^2\varphi_A(1+(1-K_f))}\right). \tag{7}$$

In practical measurements, the electrical conductivity changes by a few percent, therefore $(1-K_f) << 1$. Taking into account the formula for the tangent of a double angle, we can write:

$$\varphi_a^{\Delta} \approx arctg\left(\frac{2tg\varphi_A}{1-tg^2\varphi_A}\right) \quad \text{and} \quad \varphi_a^{\Delta} \approx 2\varphi_A. \tag{8}$$

Approximate value of the current change module through the active sensor:

$$\left|\Delta\dot{I}_a\right| \approx U_a\cdot\Delta G_{SA}\frac{1}{\sqrt{(1-tg^2\varphi_A)^2+4tg^2\varphi_A}} = U_a\cdot\Delta G_{SA}\cdot\frac{1}{1+tg^2\varphi_A}. \tag{9}$$

By analogy with (9), we determine the change in current through the reference sensor:

$$\Delta\dot{I}_p = \left|\dot{U}_p\right|\exp(\varphi_p)\left(\frac{G_{SP}+\Delta G_{SP}}{1-j\cdot tg\varphi_P^{\Delta}}-\frac{G_{SP}}{1-j\cdot tg\varphi_P}\right), \tag{10}$$

where $tg\varphi_P^{\Delta} = tg\varphi_P + \Delta tg\varphi_P$, $\Delta tg\varphi_P = \dfrac{\Delta G_{SP}}{\omega C_{SP}} = (1-K_f)\cdot tg\varphi_P$.

Then, by analogy with (8) and (9):

$$\varphi_p^{\Delta} \approx 2\varphi_P + \varphi_p; \qquad \left|\Delta\dot{I}_p\right| \approx \left|\dot{U}_p\right|\cdot\Delta G_{SP}\cdot\frac{1}{1+tg^2\varphi_P}. \tag{11}$$

To mutually compensate the changes of currents in the sensors when the background electrical conductivity of the solution changes, it is necessary to ensure the equality of the phase angles and absolute values of their vectors. From comparisons of the corresponding expressions in (8), (9) and (11), we find the correction values of the voltage vector on the reference sensor in phase and modulus to achieve the necessary quasi-equilibrium:

$$\text{by phase: } \Delta\varphi_{p2} = 2(\varphi_A-\varphi_P); \quad \text{modulo: } ND_2 = \frac{\Delta G_{SA}}{\Delta G_{SP}}\cdot\frac{1+tg^2\varphi_P}{1+tg^2\varphi_A} = \frac{G_{SA}}{G_{SP}}\cdot\frac{1+tg^2\varphi_P}{1+tg^2\varphi_A}. \tag{12}$$

From the performed analysis, the following conclusions can be drawn. Expressions (1) describe the voltage regulation parameters of the slave generator, corresponding to the equilibrium state of the measuring circuit before the introduction of the analytical substance into the solution. This state is achieved by applying the measurement method with balancing the bridge in quadrature and in-phase components of the output signal of the bridge towards the test signal [10, 12]. When an analyte is introduced into the conductometric cell, in addition to an informative change in the current in the working (active) sensor, the background electrical conductivity of the solution changes. In the result of it the current vectors in both sensors change. These, non-informative, changes in the general case differ from each other both in phase angle and in amplitude, which manifests itself as additive interference and the corresponding measurement error. In the method described in [16], after balancing, the operation of an additional change in the voltage phase of the regulated generator is performed by a value equal to the difference in the phase angles of the impedances of the active and reference (passive) sensors. Such a change in the phase of this voltage is determined by the first condition in expressions (11). In this case, the collinearity of the vectors of changes in currents in the sensors and the complete elimination of additive interference are achieved, if the active resistances of the sensors are the same. For complete mutual compensation of in-phase changes in currents in sensors at different values of their active resistances, it is also necessary to additionally change the voltage amplitude of the regulated generator so that the second (amplitude) condition formulated in (12) also is achieved. From the comparison of expressions (1) and (12) it follows that the coefficient of amplitude correction of the voltage of the dependent generator ND1 must be additionally multiplied by the coefficient:

$$K = \frac{\sqrt{1 + tg^2 \varphi_P}}{\sqrt{1 + tg^2 \varphi_A}}.$$

This operation allows to bring the bridge into the new quasi-equilibrium state described above.

Experimental studies of the suppression of the effect of changes in the background electrical conductivity of the solution in a differential conductometric channel.

To estimate the degree of suppression of changes in the background electrical conductivity of the solution, computer simulation of the behavior of the measuring circuit was carried out when using the prototype and the proposed method.

The simulation results are presented in Table 1. Calculations were made for the generator voltage frequency of 62.5 kHz. It is assumed that when a test substance is introduced, the background electrical conductivity of the solution changes by 1%, and the local electrical conductivity changes by the same amount due to the reaction in the active sensor. Calculations were performed for the range of electrical conductivity values from 0.2 mS to 5 mS, electrical capacitance from 1 nF to 50 nF, which corresponds to the limits of the ranges of the parameters of real conductometric transducers, which are appropriate to use in the considered biosensor systems.

The penultimate two columns of Table 1 show the ratio of the change in the nonequilibrium current modulus due to the change in the background electrical conductivity of the solution to the informative change in the nonequilibrium current modulus due to the local change in the electrical conductivity as a result of the reaction in the active sensor. Column δ1 corresponds to the method without amplitude correction, column δ2 corresponds to the method developed in this work. The right column shows the values of the ratios δ1/δ2, characterizing the increase in the coefficient of suppression of changes in the background electrical conductivity using the proposed method compared with the prototype. The first and last two rows show simulation results for the most typical values of solution resistance in the sensor, about 1 kΩ, with differences in sensor capacitances up to ±20 percentage. As can be seen from the data in the table, the influence of non-informational changes in the background electrical conductivity when using the developed measurement method decreases at least 37 times compared to the method that does not have additional voltage correction on the active resistance of the passive sensor.

Table 1

№	G_{AS}, mS	C_{AS}, nF	$\mathrm{tg}\varphi_A$ φ_A,°	G_{RS}, mS	C_{RS}, nF	$\mathrm{tg}\varphi_R$ φ_R,°	φ_{B2},°	ND_1	ND_2	δ_1, %	δ_2, %	δ_1/δ_2
1	1	5,44	0,4681 25,084	1	4,352	0,5851 30,333	-10,498	1,0493	1.1011	4,7	0,08	58,75
2	5	40	0,3183 17,657	5	48	0,26526 14,856	5,6015	0,9858	0,9719	1,5	0,04	37,5
3	5	1	12,733 85,509	4,167	1	10,61 84,616	1,787	1,0013	0,8356	19,8	0,017	1164
4	0,2	1	0,5093 26,99	0,25	1	0,6366 32,482	-10,984	0,8451	0,8927	5,4	0,11	49
5	1	4,5	0,5659 20,505	1	5,4	0,4716 25,247	8,515	0,9622	0,9259	4	0,066	60,6
6	1	5,44	0,4681 25,084	1,2	6,8	0,4496 24,207	1,7555	0,8272	0,8214	0,7	0,01	70

Verification of the developed measurement method on a physical model. For an experimental evaluation of the effectiveness of the proposed measurement method, an operating sample of a differential conductometric meter was made with the structure and tuning algorithm considered in this paper. The studies were carried out using the electrical equivalent of a differential conductometric transducer developed earlier [13]. Also were used the results of studies of the frequency characteristics of several types of conductometric transducers with interdigitated topology used in biosensors. Scheme of the equivalent is shown in Fig. 3

resistors 1K and capacitors 5.44nF simulate the tipical electrical parameters of active and passive converters, 100K resistances simulate changes in their electrical conductivity by 1%, and 5K resistances and 1.3nF and 4.55nF capacitors allow you to simulate non-identity of the converter parameters. The 3.3K resistors can simulate near-electrode resistance to charge transfer to implement a 3-element equivalent circuit of converters (not used in these studies). In the course of these studies, the characteristics of suppression of the influence of in-phase changes in electrical conductivity in sensors of a differential pair in a new device and in devices of previous developments described in [10, 12, 13] were compared.

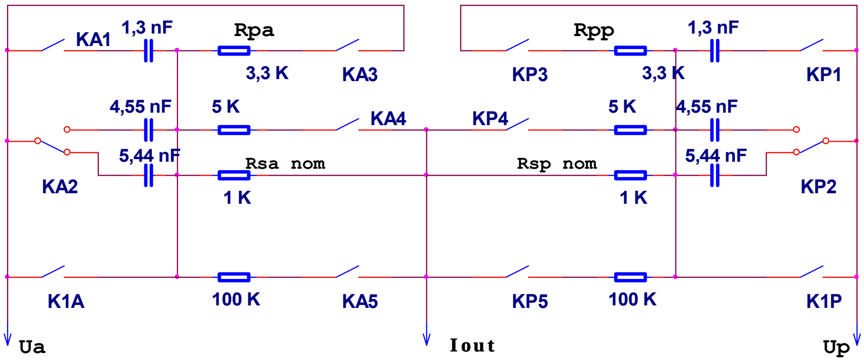

Fig. 3

Table 2 shows the values of the obtained responses of the measuring channels of previously developed devices (in conventional units, 100 c.u. ≈ 10 μS) while simulating informative and non-informative effects on the equivalents of active and passive transducers. The KA5 key simulates the informative effect, and the non-informative effect of a change in the background electrical conductivity is simulated by closing the keys KA5 and KP5 simultaneously. The ratio of the response to simultaneous non-informative influences (G_a and G_p) to the informative influence G_a is the additive error δ_{add}. The data were obtained using a bridge circuit that is fully balanced on the quadrature and in-phase components of the output signal to the bridge supply voltage [10, 12], and under using a bridge circuit that is partially balanced only on the quadrature component of this signal [13]. In the second device, the voltages on the active resistances of the sensors are equal even if when they are not identical. The results of diagnostic measurements of the ratio of equivalent values of the active resistances of the transducers R_p/R_a, the difference between the tangents tg(φ_p-φ_a) and tgφ of the active transducer are also presented. The measurements were made with the equality of the *RC*-parameters of the impedances of the converters (*Za*=*Zp*) and with changes in these parameters in the reference converter by about 20%. The response values given in the line G_a =+1%, ΔG_p =+1% are the percentage value of the additive error (δ_{add}) from the unsuppressed part of the common mode interference when it is equal to the useful signal.

Table 2

Measurement results	At full balance of the bridge circuit at a frequency of 68 kHz				When balancing the bridge circuit on the voltages on the capacitors at a frequency of 66 kHz			
Sensor parameters	Z_a=Z_p	R_p=R_a C_p=1,2C_a	R_p=R_a C_p=0.8C_a	R_p=0.8R_a C_p=C_a	Z_a=Z_p	R_p=R_a C_p=1,2C_a	R_p=R_a C_p=0.8C_a	R_p=0.8R_a C_p=C_a
ΔG_a = +1%	104	103	103	104	109	109	109	109
ΔG_p = +1%	-104	-109	-95	-79	-106	-110	-101	-98
ΔG_a= +1% ΔG_p= +1%	0	-6	8	25	3	-1	8	11
R_p/ R_a	1.0	0.962	1.044	0.829	0,987	1,059	1,13	0.837
tgφ_a	0,46	0,46	0,47	0,46	0,5	0,5	0,5	0,5
tg(φ_p-φ_a)	-0,008	-0,081	0,056	0,046	-0,002	-0,0005	0,0019	-0,0003

Table 3 shows the data of studies using an experimental sample of a new device, which is balancing to the quasi-equilibrium state by the developed method. The response to an informative change in conductivity in the working transducer was measured for ΔG_a = +1%. With an additional simultaneous change of 1% in the active resistances of the equivalents of the working and reference transducers, the relative value of the additive error δ_{add} of these measurements was determined. The data of the study of the new method on the physical model of the conductometric channel showed a lower value of suppression of the effect of background electrical conductivity compared to the computer model due to the limited discreteness of the phase adjustment (0.7°). That was done to simplify the device. If necessary, the value of δadd can be reduced several times more.

Table 3

№ C_s; R_s	R_{SA}, Ом	R_{SP}, Ом	R_{SP}/R_{SA}	tgφ_A	tgφ_P	ΔG_a= +1% ΔG_p= +1%	ΔG_a = +1%	δ_{add}, %
C_p=0.8C_a R_p=R_a	1009	1010	1,001	0,43976	0,52544	18	1530	1,17
C_p=1,2C_a R_p=R_a	1008	1008	1,000	0,44047	0,35149	12	1530	0,78
R_p=0.8R_a C_p=C_a	1008	846	0,8393	0,44042	0,52699	30	1519	1,89
R_a=0.8R_p C_p=C_a	846	1010	1,1939	0,52681	0,44003	25	1680	1,49

Conclusion.

The use of the differential method for measuring local changes in the electrical conductivity of solutions loses its effectiveness when the impedance parameters differ in the equivalent circuits of a pair of conductometric transducers that make up the differential sensor, especially when both reactive and active components differ. Due to these differences, a significant additive error arises from changes in the background electrical conductivity of the measurement medium. It is shown that it is possible to adjust the bridge circuit of the differential conductometric channel to such a state of quasi-equilibrium in phase and modulus of its output signal, in which the exact balance of the bridge is achieved with respect to in-phase changes in electrical conductivity in the working and reference transducers for any non-identity of their parameters. It can be performed by additional calculated correction of the phase and voltage's modulus on the reference conductometric transducer after the equilibrium state of the bridge is established. An algorithm and mathematical expressions for achieving such a quasi-equilibrium are determined. The use of the developed measurement method makes it possible to significantly improve the accuracy of determining local changes in the electrical conductivity in the working sensor by reducing the influence of changes in the background electrical conductivity of the solution on the measurement result by many times.

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ОПТИМІЗАЦІЯ БАЛАНСУВАННЯ МОСТОВОГО ВИМІРЮВАЛЬНОГО КОЛА З ДИФЕРЕНЦІЙНИМ КОНДУКТОМЕТРИЧНИМ СЕНСОРОМ

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Статтю присвячено зниженню впливу джерел адитивної похибки визначення локальних змін електропровідності розчинів електролітів в умовах зміни фонової електропровідності середовища вимірювань, що часто має місце в біосенсорних та інших системах з диференціальною парою кондуктометричних перетворювачів через неідентичність їхніх електричних параметрів. Метою є забезпечення глибокого придушення впливу фонових змін за значних відмінностей як реактивних, так і активних опорів у парі перетворювачів сенсора. Коротко розглянуто суть питання, причини та механізм виникнення цього виду похибки, а також методи та засоби її зменшення, розроблені раніше. Наведено схему та опис структури диференціального кондуктометричного каналу біосенсорної системи на основі моста змінного струму, алгоритм операцій його балансування регулюванням модуля та фази тестової напруги, а також векторну діаграму струмів та напруг у ньому при цьому процесі. Аналітично промодельоване балансування моста з приведенням його в стан квазірівноваги, за якого варіації фонової електропровідності не змінюють його вихідний сигнал. Визначено додаткові операції балансування моста, що дає змогу досягти такого стану за значних відмінностей як ємностей, так і активних опорів в імпедансах пари кондуктометричних перетворювачів диференціального сенсора. Наведено результати експериментальних досліджень придушення впливу змін фонової електропровідності розчину у диференціальному кондуктометричному каналі на його комп'ютерній моделі та на експериментальному зразку кондуктометричного приладу з електричним еквівалентом диференціального сенсора. Наведено порівняння отриманих результатів та відповідних даних у разі балансування мостових кіл раніше розробленими методами. Бібл. 16, табл. 3, рис. 3.

Ключові слова: диференціальні кондуктометричні біосенсори, імпеданс, вимірювання, синфазні впливи, еквівалентна електрична модель.

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