Methods for Constructing High-precision Potentiometric Measuring Instruments of Ion Activity

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Abstract – **Corresponding con-version equations are obtained, their static characteristics are constructed, measurements errors, emerging as a result of using one or another construction princi-ples are investigated. As a result of the research, it was found that in order to im-prove the measurement accuracy, it is advisable to introduce an additional measuring temperature channel. Of the proposed three methods for measuring the activity of ions, the third option with a relative error of $6.5 \cdot 10^{-7}$% in the measurement range from 0.3 to 7 pX, which is based on the principle of converting voltage into frequency, has the highest measurement accuracy.**

Keywords – analytical equation, mathematical model of the measuring channel, characteristics of measurement errors, static characteristics, measuring channel of ion activity

I. INTRODUCTION

Study of the characteristics of the measuring quality of the substance ions activity, based on different princeples of digital sensors construction is an actual task, aimed at improvement of the measurements reliability, their calibration, certipalion and determining the interval betwer the inspections. One of the ways of improving the reliability of the measurements is the decrease of the mathedical errors of measurements by means of improving the method of the measurement of the ions of the substance activity. The most widely used physical-chemical methods of the ions activity (concentration) are: amperometrical method, method of high frequency titration, spectrophotometric method, chronu conductometric method and ionometry.

Amperometrical method is based on the measurement of the current strength, passing across electrochemical element at known conditions. The current strength depends on the concentration of the electric charge carriers. Under such conditions on the base of the current strength measuring it is possible, after the graduation to determine the concentration of the available types of substances.

The given method of measuring is the most efficient at optimal concentration of ions 10^{-3} mol/dm^3. At higher concentrations "erroneous sections" are observed, at lower concentrations – rectangular sections are observed [1, 2]. Amperometrical method has the following disadvantages: strong dependence of the diffusion current on the temperature and time of the electrolysis that characterizes differential thickness of the diffusion layer.

Method of high frequency titration involves the the determination of the capacitive resistance on high circular frequency of the capacitance of the investigated emulsion. Great values of the frequency and capacitance result in considerable reduction of the capacitive resistance, that makes it possible to conduct analytical definitions. The given method enables to perform measurements of the concentration in the range from several moles per 1 dm^3 to 10^{-5} mol/dm^3 [3 - 5]. The drawbacks of high frequency titration method include the impossibility to use aqueous solutions, low sensitivity in the area of small steepness of the characteristic curve slope, necessity to prepare special titrants, that takes much time and complicates the realization of measuring procedure. Besides, there exists undesired possibility of the titration system indicator double calculation, that occurs when the curve passes across the maximum.

Spectrophotometric method is based on the registration of the solution absorption change, being titrated as the titrant is added. Graphic representation of such measurements is the titration curve, constructed on the base of the experimental data, this curve has the culminating point (singular point). Taking the culminating point on the curve as the final point of titration, the titrant consumption is found and applying the formulas of titrometric analysis calculations are carried out. The absorbtion parameter of the investigated solution can be measured, using the flow of high degree of monochromaticity, i. e., while determining the wavelength λ in case of using the spectrophotometer or in a certain interval of wavelength, working with photometer or photoelectric colorimeter when monochromaticity is performed by means of the light filters. In analytical practice the first variant of titration is commonly referred to as spectrophotometric and the second variant - photometric one. The given method enables to measure very low concentrations up to 10-12 mol/dm^3 with the fast acting of 15–20 min. But the important

drawback of the photometric method is rather high, as compared with the methods, considered above, relative error, which is 2,8 – 3% as well as the impact of the dissipated radiation, possibility of using not sufficiently pure indicators, impact of the errors as a result of the deviation from Beer's law and necessity to use the additional equipment [5-7].

The basis of the chrono conductometric method is the measurement of the solution electroconductivity during certain interval of the titration time. The obligatory condition of the chrono-conductometric method is the need to supply the titrant at a constant speed: In this case the titrant must be supplied at such speed that the reaction could come to its end. In case of chrono-conductometric titration the continuous automatic recording of the titration curves is carried out in strictly determined time intervals. Then, according to chrono=conductometric curve the length of the diagram tape is determined in milimeters from the beginning of the titration to the equivalence point. Time of titration is found by the speed of the diagram tape motion. Titration time is proportional to certain volume of the standard solution, that is why, the calculation formulas contain the concentration of the standart solution, time of flowing of certain volume of the standard solution and the titration time of the substance, being determined. Thus, automatic recording of the titration curves enables to reduce the time of the analysis, obtain the reliable results and improve the accuracy of the analysis due to rapid realization of several parallel titrations. The drawbacks of the chrono-conductometric method is conversion equation dependence on the time of titration that leads to the possibility of emerging considerable transient processes and necessity to provide constant speed of the titrant supply and diagram tape motion [8-11].

Existing methods for measuring ion activity have different ranges of ion activity measurements, their disadvantages and advantages. However, to build high-precision means for measuring the activity of ions for the study of the constituent elements of humus in soils, it is necessary to have mathematical models of transformation equations that will allow one to efficiently study the metrological characteristics for high-precision calibration of such measuring instruments. Therefore, an urgent scientific task is the development of mathematical models of measuring instruments for the activity of ions in the measurement range from 0.3 to 7 pX and the study of their accuracy.

II. ION SELECTIVE TRANSDUCER BUILT ON THE PRINCIPLE OF ADC OF SEQUENTIAL APPROXIMATION

Realization of a digital potentiometric ion selective transducer for measuring the activity of substance ions can be performed on the base of ADC of the successive approximation in its structure. Structural diagram of such transducer is presented in Fig. 1.

As it is seen from Fig. 1 digital ion selective transducer comprises: ion selective electrode pX, reference electrode pXC, scale converter (SC) that performs function of noninverted amplification, analog-to-digital converter (ADC), central processing unit (CPU), reprogrammed read only memory (EEPROM), controller for data transfer via the serial port RS232 to the computer (PC) and liquid crystal display (LCD). Signal from the output of ion selective electrode pX is amplified by the scale converter to level of reference value, set by the reference electrode, where ADC

functions and further it passes to (HI) input of ADC. ADC transforms the voltage value into binary code.
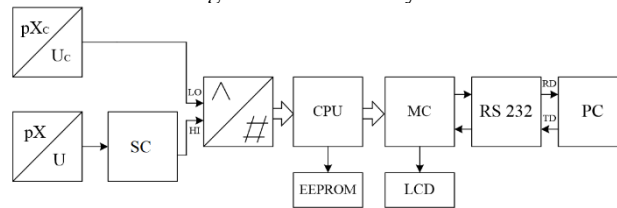

Fig. 1. Structural diagram digital potentiometric transducer of ions activity, built on the principle of ADC of serial approximation.

Model transformation equation of the suggested digital transducer of ions activity, built on the principle of successive approximation, using analog-to-digital converter, has the form:

$$N_{ADC} = \left(U_0 - \alpha \left(273,15 + t \right) n_a^{-1} pX_i \right) k \left(U_{ref} \right)^{-1} 2^m, \quad (1)$$

where U_{ref} – is the value of the reference voltage of ADC, set by the reference electrode; m – is the ADC bit rate; k – is amplification factor of the scale converter; α – is temperature coefficient of steepness S, that equals $198,4 \cdot 10^{-3}/{}^{\circ}C$; t – is the temperature of the environment being analyzed (${}^{\circ}C$); U_0 – graduation voltage, which is determined by the selection of the reference point; pX_i – concentration of ions; n_a – is the charge of ion [12-16].

Characteristic of the conversion equation of the digital ion selective transducer of ions activity, built on the principle of the successive approximation is shown in Fig. 2.

It is seen from the obtained characteristics of the dependence of ADC code change on the activity of ions that the function of the transformation of the digital potentiometric ion-selective transducer of ions activity is linear.
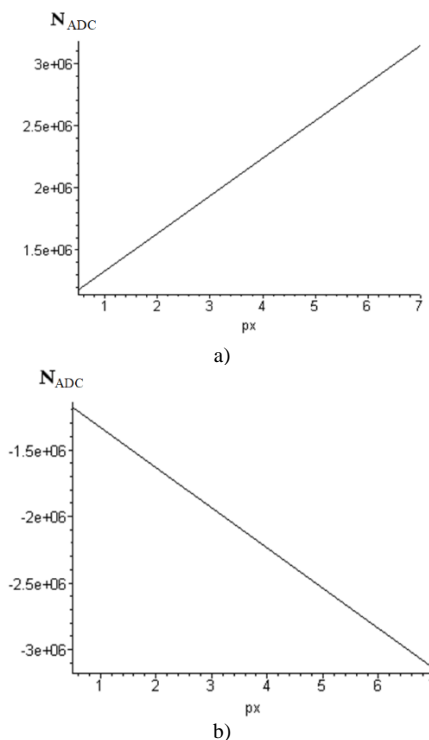

Fig. 2. Characteristics of ions activity change while realization of the digital transducer, built according to the principle of the analog-to-digital conversion of the successive approximation: a) while measuring of negatively changed ions; b) while measuring of positively changed ions.

Taking into account the conversion function (1) mathematical model of the quantization error of the digital potentiometric transducer of ions activity is described by the expression:

$$\delta_{ADC} = \frac{U_{ref}\left(U_0' - \alpha(273.15+t)n_a^{-1}pX_i\right)^{-1}}{[k2^m]}100\% \quad . \quad (2)$$

Analyzing the obtained mathematical model of the error (4) it is seen that it decreases while measuring greater values of pX ions activity and its change characteristic is nonlinear (Fig. 3).
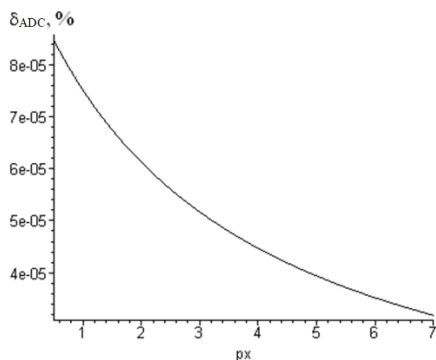

Fig. 3. Characteristic of the error change of the ions activity transducer, based on the principle of analog-to-digital conversion of the successive approximation.

As it is seen from the obtained characteristic of the digital transducer of ions activity error change (Fig. 3) the methodical component of the error does not exceed $8,6 \cdot 10^{-5}\%$ and is of nonlinear character. Ways of the methodical error decrease is the increase of the number of bits, but this will lead to the increase of digital transducer cost.

Determination of the amplitude changes of the analytical signal of ion selective electrode by means of ADC causes a number of problems, dealing with the necessity of the simultaneous provision of high capacity , fast acting, accuracy and cost.

That is why, the second variant of the circuit realization of digital ion selective transducer of ions activity, based on the principle of time-pulse conversion is proposed.

III. ION-SELECTIVE TRANSDUCER, BUILT ON THE PRINCIPLE OF TIME-PULSE CONVERSION

For measuring the instantaneous voltage values from the outputs of ion selective electrodes instead of analog-digital conversion unit the principle of time-pulse conversion, based on the sawtooth generator and comparators could be used. Also in order to improve the accuracy the additional temperature measuring channel can be added [17-20]. Structural diagram of the digital ion selective transducer of ions activity, built on the principle of time-pulse conversion is shown in Fig.4. In such transducer of ions activity, the measured voltage is converted in time interval Tx with further quantization by the pulses of the reference frequency f_0 of the quartz-crystal resonator of the microcontroller. Temperature measuring channel is realized according the similar principle (time-pulse conversion), as the primary converter thermoresistive converter will be used, and non-inverting operation amplifier will be used as the scale converter.

Basic element of the structural realization of time-pulse conversion method is comparison device (CD), realized on two comparators, sawtooth generator generator GU and RS-trigger T. Quantization of the time interval by the pulses of the reference frequency f0 is performed in the microcontroller by means of the built-in analog comparator and coincidence circuit.

Digital transducer of ions activity, built on the principle of time-pulse conversion (Fig. 4) comprises: ion selective electrode pX, reference electrode pXC, operational amplifiers (A1-A4), multiplexers (MX1, MX2), comparison devices (CD) , microcontroller (MC) and liquid crystal display (LCD). Comparison devices consist of the sawtooth generator, two comparators and RS-trigger. Additional temperature measuring channel comprises thermoresistive converter, scale converter (SC) and CD.

Signals from the outputs of the reference electrode pXC, ions selective electrode and thermoresister reach the operational amplifiers (OA), it should be noted that the OA in the measuring channel of ion activity are provided in two variants – for the measurement of both positive and negative voltages from the outputs of ion selective electrodes. This is connected with the fact that in the process of measuring negatively charged ions the voltage at the output of the electrodes will be positive and in the process of measuring positively charged ions the voltage is negative. Process of measuring positively and negatively charged ions is controlled by microcontroller using multiplexors.

After the amplification, voltage signals enter the comparison device (CD), where they are converted into the time interval T_x.
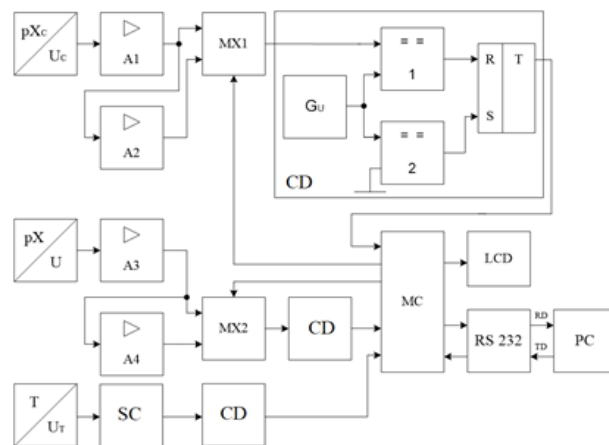

Fig. 4. Structural diagram of digital transducer of ions activity, built on the principle of time-pulse conversion.

Further by means of the built -in analog comparators, the allocated time interval is filled with pulses of the reference frequency f_0. Number of pulses, which entered in the allocated by means of the comparison device (CD) time interval is determined by the expression:

$$N_{U/T} = k\left(U_0 - \alpha\left(273,15+t\right)n_a^{-1}pX_i\right)Kf_0 \quad , \quad (3)$$

where k – is the amplification factor of the operational amplifier (OA); K – is proportionality coefficient, which depends on the steepness of linearly varying voltage of the generator G_U; f_0 – are pulses of the reference frequency of the quartz crystal resonator of the microcontroller.

Static characteristics of the digital transducer of ions activity, built on the principle of time-pulse conversion are presented in Fig. 5.

Error of the measuring channel of ions activity, taking into account model conversion equation (3) is described by the expression:

$$\delta_{U/T} = \left[k \left(U_0' - \alpha \left(273{,}15 + t \right) n_a^{-1} pX_i \right) K f_0 \right]^{-1} 100\%$$. (4)

Analysis of the obtained expression of the error of the measuring channel of ions activity, built on the principle of time-pulse conversation shows that the error decreases with the increase of pX ions activity and its cange characteristic is nonlinear (Fig. 6).
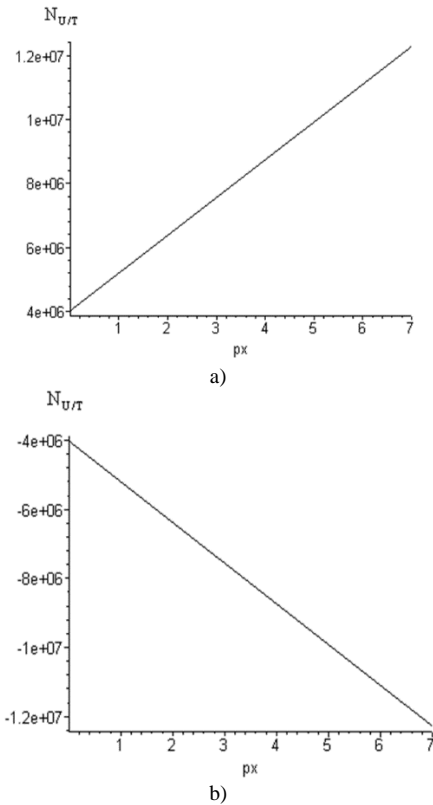

a)

b)

Fig. 5. Static characteristics of digital transducer of ions activity, built on the principle of time-pulse conversion: a) while measuring negatively charged ions; b) while measuring positively charged ions.
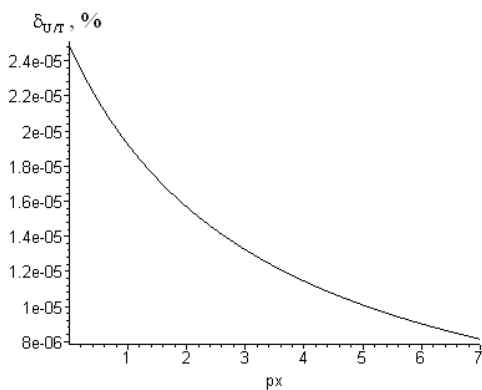

Fig. 6. Characteristic of the error variation of the ions activity measuring channel, built on the principle of time-pulse conversion.

Possible ways of the quantization error decrease (4) is the increase of the reference frequency f_0 value, proportionality coefficient K and amplification factor k.

As it is seen from the obtained characteristic of the error change (Fig. 6) its maximum value does not exceed $2{,}45 \cdot 10^{-5}\%$ and is 3,5 times less than the error of the digital

transducer, built on the base of ADC of the successive approximation [20-23]. The drawbacks of this principle of construction are low noise immunity, caused by the non-linearity of the varying voltage of the generator G_u and instability of the comparator response level. That is why, we will investigate the third variant of the digital transducer of ions activity, based on the principle of voltage to frequency conversion.

IV. ION-SELECTIVE TRANSDUCER, BUILT ON THE PRINCIPLE OF VOLTAGE INTO FREQUENCY CONVERSION

For the comparison with the previous realizations of the digital transducers of the ions activity the third variant of the digital potentiometric transducer, built on the principle of voltage to frequency conversion is suggested. Structural diagram of such digital transducer is shown in Fig. 7.

The device comprises: ion-selective converter (pX/U), reference electrode (pX$_C$/U$_C$); two operation amplifiers in each of measuring channels (MC) of ions activity (A1 – A4) to provide measuring of both positive and negative values of pX; multiplexors (MX1 and MX2); voltage into frequency converters (VFC) (U$_C$/F$_C$ and U/F) for the conversion the potentials of the reference electrode U$_C$/F$_C$ and ion selective electrode U/F into frequency; microcontroller (MC); liquid crystal display (LCD); voltage levels converter (RS232) for data transfer to the computer (PC).
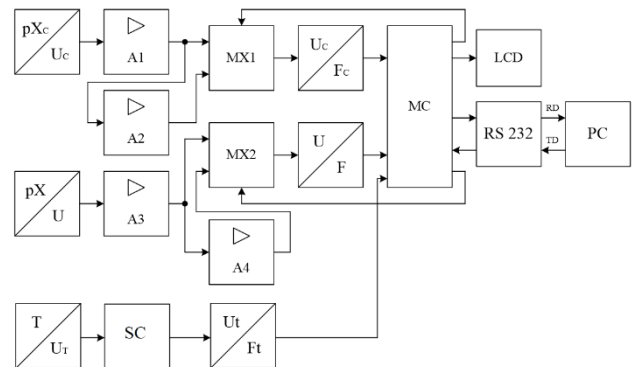

Fig. 7. Structural diagram of the digital transducer of ions activity, built on the base of voltage into frequency convertors.

Also temperature measuring channel (MC), consisting of the thermoresistive converter, scale converter (SC) and voltage into frequency converter (VFC). Conversion equation of the suggested digital transducer of ions activity, built on the principle of U/F conversion has the form:

$$N_{U/F} = U_{\max} f_0 \tau \left[\left(U_0' - \alpha \left(273{,}15 + t \right) n_a^{-1} pX_i \right) k \right]^{-1}$$, (5)

where $U_{\max}$ – is the value of the reference voltage of UFC (10 V); $\tau = RS$ – constant of UFC time, used for setting the full-scale output frequency of the quartz-crystal resonator of the microcontroller (R=1 kOhм, C=47 мF); f_0 – is the frequency of the quartz-crystal resonator of the microcontroller (20 MHz); k - is amplification factor of OA [12, 15, 17].

Representations of the conversion equation (5) of the digital transducer of ions activity, built on the principle of voltage into frequency conversion, are shown in Fig.8.

From the obtained characteristics of the number of pulses change dependence on the ions activity it is seen that the conversion function of the given transducer is not linear but non linearity of VFC in the wide range of frequencies change does not exceed $2 \cdot 10^{-3}$ %.

Error of the digital transducer of ions activity, built on the base of voltage into frequency converter with the account of the conversion equation (5) is described by the expression:

$$\delta_{U/F} = k \left(U_0' - \alpha \left(273,15 + t \right) n_a^{-1} pX_i \right) \left[U_{max} \tau f_0 \right]^{-1} 100\% \qquad (6)$$

Characteristic of the transducer error change (6) is shown in Fig. 9.

Analysis of the obtained equation of the relative error of the ions activity transducer, built on the principle of voltage into frequency conversion shows that it increases with the increase of the measuring range of ions activity pX, and its change characteristic is linear (Fig. 9). Linearing of the error characteristic of the digital transducer of ions activity enables to introduce easily the corrections if necessary. As it is seen from Fig. 9, maximum error of the digital transducer does not exceed $6,5 \cdot 10^{-7}$ % which is 37 times less than the error of the digital transducer, built on the principle of time – pulse conversion and 130 times smaller than the maximal error of the digital transducer, built on the principle of analog – to – digital conversion of the successive approximation. Possible ways of decreasing the relative error of the digital transducer (6) is the increase of the reference frequency value f_0 and time constat τ.
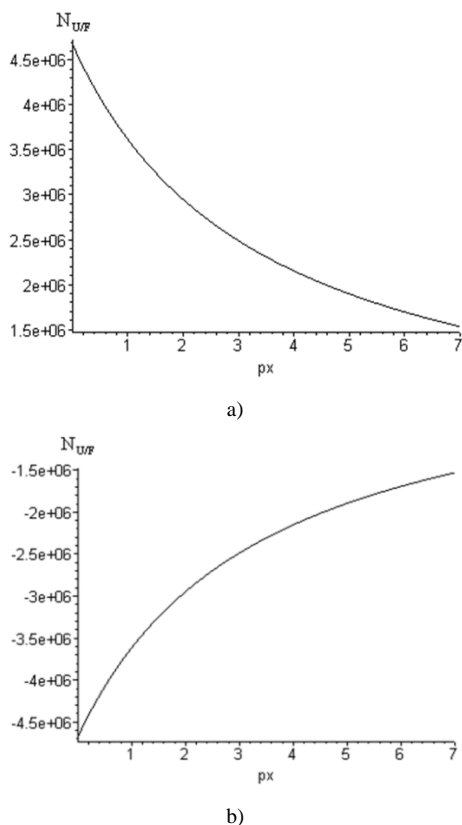
Fig. 8. Static characteristics of the measuring channel of ions activity, built on the principle of voltage into frequency conversion: a) while measuring negatively charged ions; b) while measuring positively charged ions.
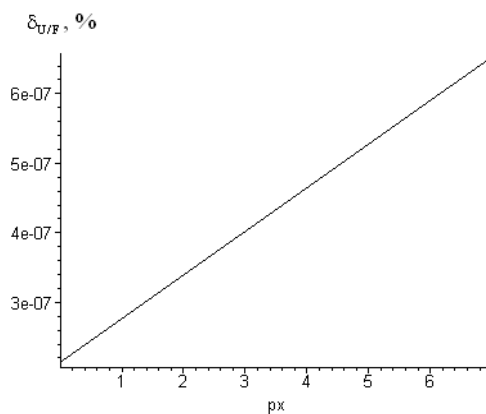
Fig. 9. Characteristic of the error variation of ions activity measuring channel, built on the principle of voltage into frequency conversion.

All the characteristics of the errors changes are built in the range of ions activity change from 0,3 to 7 pX at the temperature of 20°C. As it is seen from the obtained characteristics of the relative errors change (Fig. 3, Fig. 6, Fig. 9) the smallest value of the error $7,7 \cdot 10^{-7}$% has the third variant of the realization of the digital ion selective transducer of ions activity, based on the principle of voltage into frequency conversion. The only drawback of such realization is nonlinearity of the static characteristic. However, the range of binary code change is great and this nonlinearily is of minor importance for measuring ions activity.

V. CONCLUSIONS

Thus, the scientific novelty of the research lies in obtaining mathematical models of transformation equations based on the principles that are laid down in the schemes on which the measuring channels of the activity of ions of the constituent elements of humus in soils are built. The resulting mathematical models allow us to investigate the measurement error that occurs in the corresponding measurement ranges. In addition, on the basis of the obtained mathematical models of the transformation equations, it becomes possible to study static metrological characteristics by expanding them into a Taylor series.

On the base of the above - mentioned, the conclusion can be made that the best variant of realization of the unified system of the automated control of humus constituents in the soil with increased methodical component of the control reliability will be the system, built on the base of the improved method of the ionometry, using in the structure of measuring channel (MC) method and means of voltage into frequency conversion. Also in the process of construction of the system of the automated control the combination of the method of voltage into frequency conversion with the method of time-pulse conversion is possible, for instance, usage of one of the methods in temperature MC, and other method – in MC of ions activity or vice versa.

As a result of the research, carried out, it is established that the error, emerging due to the deviation of the real value of the temperature at least for 1 °C from the theoretical value of the temperature (25 °C), taken into account in the transformation equation, while using industrial ion selective electrodes, in the process of measuring negatively charged ions may reach the value of 0,36%/ °C , and while measuring positively charged ions it may reach the value of 10,4%/ °C . That is why, hile construction digital ion selective transducer

it is expedient to introduce the additional temperature measuring channel. In this case, as a result of the research it is established that the error of the temperature measuring channel must be 0,05 % for the account of 1 °C temperature change while measuring at low boundary of 0,3 pX.

REFERENCES

[1] Michael Jacobs, Carlos G. Lopez, and Andrey V. Dobrynin, Quantifying the Effect of Multivalent Ions in Polyelectrolyte Solutions, *Macromolecules,* 2021, *54* (20), 9577-9586.

[2] Voltammetric and Amperometric Methods. DePauw University, 12 Sept. 2021, https://chem.libretexts.org/@go/page/154799.

[3] Kang, S. D., & Chueh, W. C., Galvanostatic Intermittent Titration Technique Reinvented: Part I. A Critical Review. *Journal of The Electrochemical Society*, 2021, *168*(12), 120504.

[4] Ryan Sayko, Michael Jacobs, Andrey V. Dobrynin, Universality in Solution Properties of Polymers in Ionic Liquids, *ACS Applied Polymer Materials,* 2022, *4* (3), 1966-1973.

[5] Parthasarathy, S., Ganesh Kumar, V., & Subbarao, R. V., A remote conductometric titration method for simultaneous determination of uranium and nitric acid concentrations at elevated temperatures. *Journal of Radioanalytical and Nuclear Chemistry*, 2021, *327*(3), 1087-1093.

[6] Thipwimonmas, Yudtapum, et al. "A Simple and Rapid Spectrophotometric Method for Nitrite Detection in Small Sample Volumes", *Chemosensors* 9.7 (2021): 161.

[7] Mabrouk, M. M., Hamed, N. A., & Mansour, F. R., Simple Spectrophotometric Method to Measure Surfactant CMC by Employing the Optical Properties of Curcumin's Tautomers. *Journal of Chemical Education*, 2021, *98*(8), 2603-2609.

[8] Muthuri, L. K., Nagy, L., & Nagy, G., Chronopotentiometric method for assessing antioxidant activity: A reagentless measuring technique. *Electrochemistry Communications*, 2021, *122*, 106907.

[9] Barros, K. S., Marti-Calatayud, M. C., Ortega, E. M., Perez-Herranz, V., & Espinosa, D. C. R., Chronopotentiometric study on the simultaneous transport of EDTA ionic species and hydroxyl ions through an anion-exchange membrane for electrodialysis applications. *Journal of Electroanalytical Chemistry*, 2020, *879*, 114782.

[10] O. Vasilevskyi, V. Didych, A. Kravchenko, M. Yakovlev, I. Andrikevych, D. Kompanets, Y. Danylyuk, W. Wojcik, A. Nurmakhambetov, Method of evaluating the level of confidence based on metrological risks for determining the coverage factor in the concept of uncertainty, Proceedings Volume 10808, Photonics Applications in Astronomy, Communications, Industry, and High-Energy Physics Experiments 2018, 108082C.

[11] Zagatto, E. A., Oliveira, C. C., Townshend, A., & Worsfold, P., *Flow analysis with spectrophotometric and luminometric detection.* Elsevier, 2012; doi:10.1016/c2010-0-66895-7.

[12] O. Vasilevskyi, Assessing the level of confidence for expressing extended uncertainty through control errors on the example of a model of a means of measuring ion activity, Acta IMEKO 10(2) (2021) pp. 199-203.

[13] Toshio Fuchigami, Mahito Atobe, Shinsuke Inagi, Fundamentals and Applications of Organic Electrochemistry: Synthesis, Materials, Devices, (2014), 240 P. ISBN: 978-1-118-65317-3.

[14] Fuller, T. F., & Harb, J. N., *Electrochemical engineering*. John Wiley & Sons, 2018.

[15] O. M. Vasilevskyi, P. I. Kulakov, I. A. Dudatiev, V. M. Didych, A. Kotyra, B. Suleimenov, A. Assembay, A. Kozbekova, Vibration diagnostic system for evaluation of state interconnected electrical motors mechanical parameters, Proc. SPIE 10445, Photonics Applications in Astronomy, Communications, Industry, and High Energy Physics Experiments, 104456C, 2017.

[16] M. Nagasawa, "Physical Chemistry of Polyelectrolyte Solutions", Advances in Chemical Physics, 2015.

[17] O. Vasilevskyi, P. Kulakov, D. Kompanets, O. Lysenko, V. Prysyazhnyuk, W. Wójcik, D. Baitussupov, A new approach to assessing the dynamic uncertainty of measuring devices, Proc. SPIE 10808, Photonics Applications in Astronomy, Communications, Industry, and High-Energy Physics Experiments, 108082E, 2018.

[18] V. S. Bagotsky, "Fundamentals of Electrochemistry", 2005.

[19] E. W. Abel, "Organic Spectroscopic Analysis", Tutorial Chemistry Texts, 2007.

[20] Febiana, H., et al. "Automotive engineering chemistry module: Exploring acid base and electrochemistry topic in vehicle context", *Journal of Physics: Conference Series*., Vol. 1156, No. 1, IOP Publishing, 2019.

[21] O. M. Vasilevskyi, Calibration method to assess the accuracy of measurement devices using the theory of uncertainty, Int. J. Metrol. Qual. Eng. 5(4) (2014), art. no. 403.

[22] Shishkanova, T. V., Sapurina, I., Stejskal, J., Král, V., & Volf, R., Ion-selective electrodes: polyaniline modification and anion recognition, *Anal. Chim. Acta*, 2005, *553*(1-2), 160-168.

[23] M. Nagirnyi, R. D. Apostolova, A. S. Baskevich, and E. M. Shembel', "Electrolytic Deposition of Molybdenum Oxide from Aqueous Solutions at Room Temperature," Russian Journal of Applied Chemistry, vol. 77, no. 1, pp. 71–73, Jan. 2004.