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Д.В. Сокольский атындағы «Жанармай,
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ИЗВЕСТИЯ

НАЦИОНАЛЬНОЙ АКАДЕМИИ НАУК
РЕСПУБЛИКИ КАЗАХСТАН
АО «Институт топлива, катализа и
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NAS RK is pleased to announce that News of NAS RK. Series of chemistry and technologies scientific journal has been accepted for indexing in the Emerging Sources Citation Index, a new edition of Web of Science. Content in this index is under consideration by Clarivate Analytics to be accepted in the Science Citation Index Expanded, the Social Sciences Citation Index, and the Arts & Humanities Citation Index. The quality and depth of content Web of Science offers to researchers, authors, publishers, and institutions sets it apart from other research databases. The inclusion of News of NAS RK. Series of chemistry and technologies in the Emerging Sources Citation Index demonstrates our dedication to providing the most relevant and influential content of chemical sciences to our community.

Қазақстан Республикасы Ұлттық ғылым академиясы "ҚР ҰҒА Хабарлары. Химия және технология сериясы" ғылыми журналының Web of Science-тің жаңаланған нұсқасы Emerging Sources Citation Index-те индекстелуге қабылданғанын хабарлайды. Бұл индекстелу барысында Clarivate Analytics компаниясы журналды одан әрі the Science Citation Index Expanded, the Social Sciences Citation Index және the Arts & Humanities Citation Index-ке қабылдау мәселесін қарастыруда. Web of Science зерттеушілер, авторлар, баспашылар мен мекемелерге контент тереңдігі мен сапасын ұсынады. ҚР ҰҒА Хабарлары. Химия және технология сериясы Emerging Sources Citation Index-ке енуі біздің қоғамдастық үшін ең өзекті және беделді химиялық ғылымдар бойынша контентке адалдығымызды білдіреді.

НАН РК сообщает, что научный журнал «Известия НАН РК. Серия химии и технологий» был принят для индексирования в Emerging Sources Citation Index, обновленной версии Web of Science. Содержание в этом индексировании находится в стадии рассмотрения компанией Clarivate Analytics для дальнейшего принятия журнала в the Science Citation Index Expanded, the Social Sciences Citation Index и the Arts & Humanities Citation Index. Web of Science предлагает качество и глубину контента для исследователей, авторов, издателей и учреждений. Включение Известия НАН РК в Emerging Sources Citation Index демонстрирует нашу приверженность к наиболее актуальному и влиятельному контенту по химическим наукам для нашего сообщества.

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**ELECTROCHEMICAL BEHAVIOR OF SILVER
IN A POTASSIUM FERROCYANIDE SOLUTION**

Abstract. One of the main priorities of the innovative industrialization of the country is the development of metallurgy and the production of finished products. In this regard, the application of new methods for producing metals and their compounds or extracting them from solutions is an important task of our time. One of the promising methods applied currently is the polarization by alternating current of industrial frequency, which arouses the interest of researchers both from the point of view of oxidation and destruction of the passivating film. The peculiarity of the processes occurring under the action of industrial alternating current is that, by changing the direction of the current, it is possible to remove the oxide film and create conditions for the metal to further dissolve.

The work shows the distinctive features of electrochemical processes occurring on a silver electrode during electrolysis by industrial alternating current in a solution of ferrosyne-sintered potassium by the method of rational mathematical planning. The optimal conditions for the dissolution of silver are determined by studying the effect of current density at the electrodes, the concentration and temperature of the electrolyte, the duration of the electrolysis and the frequency of the alternating current. It is shown that when polarized with an alternating current of silver in a pair with a titanium electrode, the passivation process of the silver electrode is eliminated and the dissolution rate of the metal increases.

Keywords: alternating current, silver, electrode, electrolysis, closeness of current, current output.

Recently in electrochemical researches special attention is paid to the processes proceeding with an electrode involvement by superimposed a. c. technique [1-7].

Use of the nonstationary mode of electrolyzing expands the possibilities of a research of the mechanism of the cathode and anode processes, opens essentially new opportunities of using them for the solution of different technological questions.

Electrochemical processes are used to solve various topical issues in the field of production. Rather than direct current, alternating current allows to create simple, yet rational technological processes [8].

In the works [9-13] is provided this to polarization of a silver electrode by an industrial alternating current with a frequency of 50 Hz. It was set that in sulfate solution silver is dissolved with a high efficiency in case of low current densities, and in solution of hydrochloric acid the efficiency of dilution of silver makes only 10,4%.

It is necessary to mark that the detail researches carried out by us on a silver electrode and other researchers studying titanium, chrome, molybdenum, lead, etc. in case of polarization by an alternating current of industrial frequency, showed that the electrode processes proceeding with an electrode involvement by superimposed a. c. technique, sharply differ both on the mechanism, and according to the quantitative characteristics [14-17].

Electrochemical processes involving noble metals are interesting, especially silver, in connection with the increasing need for it in various branches of technology and industry. The most valuable properties of silver are its high electrical conductivity, reflectivity and beautiful decorative appearance of polished

sediments. Silver plating is widely used in electronics, electrical engineering and other industries to improve surface conductivity and minimize transition resistance at the points of contact, as well as to impart optical properties to the surface of a product. Technical-economic and quality indicators of the silvering process depend on the nature and composition of the electrolyte. Cyanide solutions are the most widespread ones, but they are toxic and dangerous to the environment.

The most common substitute for cyanide silvering electrolytes is a solution of ferrous sulphide potassium. This is explained by the fact that it is less toxic than cyanide electrolyte, cheaper than iodide and sulphite, and, according to [18,19], it has the best scattering ability. In this regard, it is interesting to question of the behavior of silver in a ferrosynetosuronic solution during polarization with industrial alternating current and the possibility of the formation of the corresponding complex compound of silver.

Systemic studies, which allow to establish the laws of the silver electro-oxidation with the subsequent synthesis of its compounds, can lead to both the intensification of its production and the solution of resource-saving problems.

In this regard, the study of the processes of silver ionization discharge under the action of industrial alternating current seems relevant and timely.

The purpose of this work is to study the electrochemical behavior of silver during the polarization of unsteady currents in a solution of ferrocyanide syngas potassium.

To study the electrochemical behavior of silver in a potassium ferrocyanide syrup solution, the main experiments were carried out in a 100 ml cell. A titanium wire and a silver plate were used as electrodes.

Electrodes before experiments were thoroughly cleaned, degreased and washed with distilled water.

Results and discussion

The study of the electrochemical behavior of silver in a neutral medium was carried out by the method of rational mathematical planning [20]. In accordance with the planning for the six factors, and in each factor of five levels 25 experiments were conducted. The studied factors and their levels are presented in table 1.

Table 1 – Levels of factors under study

Factor	Level				
	1	2	3	4	5
X ₁ , current density on the silver electrode, A/m ²	200	400	600	800	1000
X ₂ , current density on the titanium electrode, kA / m ²	20	40	60	80	100
X ₃ , solution concentration, M	0,5	1,0	1,5	2,0	2,5
X ₄ , duration of electrolysis, min	15	30	45	60	75
X ₅ , electrolyte temperature, ° C	20	30	40	50	60
X ₆ , frequency of alternating current, Hz	50	100	150	200	250

The experiments results after mathematical processing are shown in Table 2, in which Y₁, Y₂, etc. - particular functions, respectively, of factors X₁, X₂, etc.

Based on table 2, the graphs were plotted (figure 1). To describe point data, it is necessary to choose the right empirical formulas. The dependence Y₁=f(X₁) is described by the equation of a straight line. The equation of a line is:

$$Y_1 = aX \quad (1)$$

Table 2 – Experimental values of private functions.

Function	Level					Average value
	1	2	3	4	5	
Y ₁	70,6	73,7	61,5	55,2	49,0	62
Y ₂	52,5	65,5	70,6	63,2	58,3	62
Y ₃	73,7	64,5	60,2	54,1	46,65	62
Y ₄	73,7	68,5	63,3	55,8	49,4	62
Y ₅	73,7	67,8	60,8	56,4	51,3	62
Y ₆	73,7	68,6	59,7	55,6	52,5	62

It can be considered that the straight line drawn on the graph goes out from some point, taken arbitrarily on the left side of the straight line, with coordinates X_1, Y_1 . The offset of the origin to this point is fixed as follows:

$$Y - Y_1 = a (X - X_1) \tag{2}$$

Selecting on the right side of the line any point with coordinates X_2, Y_2 and substituting them into this equation, we get:

$$Y_2 - Y_1 = a (X_2 - X_1) \tag{3}$$

After the final transformation, the equation has the form:

$$Y = Y_1 + \frac{Y_2 - Y_1}{X_2 - X_1} (X - X_1) \tag{4}$$

Some graphs have the form of a parabola, we enter the extremum values into the parabola equation. In the parabola equation ($Y = aX^2$), we enter the values X_1, Y_1 with the offset of the origin:

$$Y = Y_1 - a (X - X_1)^2 \tag{5}$$

Then we select the second point with coordinates X_2, Y_2 approximately in the middle of the section of the branch passing through the experimental points, and substitute all four numbers into the final equation:

$$Y = Y_1 + \frac{Y_2 - Y_1}{(X_2 - X_1)^2} (X - X_1)^2 \tag{6}$$

The calculated values at the matrix levels of the arguments are given in table 3.

Table 3 – Calculated values of functions at matrix levels

Function	Level					Average value
	1	2	3	4	5	
$Y_1 = 93 - 0,018 (X - 300)$	71,17	74,83	67,60	64,00	60,40	67,60
$Y_2 = 91 - 0,0037 (X - 70)^2$	65,00	70,63	71,63	66,67	61,75	67,13
$Y_3 = 91 - 4,6666 (X - 0,75)$	72,16	69,83	67,5	65,17	62,83	67,49
$Y_4 = 95 - 0,29 (X - 22,5)$	77,17	72,82	68,47	64,12	59,77	68,47
$Y_5 = 92 - 0,2666 (X - 25)$	73,33	70,67	68,01	65,33	62,67	68,00
$Y_6 = 93 - 0,0029 (X - 40)^2$	72,70	72,70	70,39	65,75	58,79	68,06

The significance or insignificance of the function can be established without repeated experiments using the nonlinear multiple correlation coefficient:

$$R = \sqrt{1 - \frac{(N-1) \sum (Y_T - Y_3)^2}{(N-K-1) \sum (Y_3 - Y_{cp})^2}} \tag{7}$$

$$t_R = \frac{R \sqrt{N-K-1}}{1-R^2} > 2$$

where N is the number of points described, K is the number of active factors, Y_3 is the experimental result, Y_T is the theoretical (calculated) result, Y_{cp} is the average experimental value.

The nonlinear multiple correlation coefficient for the 5% level and the value of its significance is $t_R > 2$, which indicates the significance of the functions.

The correlation coefficient and the significance of the corresponding particular functions are listed in table 4. As can be seen, all the considered functions influence the silver dissolution.

To describe the statistical multifactor dependencies, the particular functions generalized M.M.Protodyakonov's equation [21]:

$$Y_n = \prod_{i=1}^n Y_i ;$$

$$Y_n = \frac{[93 - 0,018(X_1 - 300)] \cdot [91 - 0,0037(X_2 - 70)^2] \cdot [91 - 4,6666(X_3 - 0,75)]}{88,85^5 \cdot [95 - 0,29(X_4 - 22,5)] \cdot [92 - 0,2666(X_5 - 25)] \cdot [93 - 0,0029(X_6 - 40)^2]} \quad (8)$$

Table 4 – Correlation coefficient R and its significance t_R for particular functions

Function	R	t_R	Function significance
Y ₁	0,92	19,08	Significant
Y ₂	0,75	2,72	Significant
Y ₃	0,88	5,65	Significant
Y ₄	0,95	85,64	Significant
Y ₅	0,96	14,38	Significant
Y ₆	0,94	10,15	Significant

Based on the equation, we find the correlation coefficient for $N = 25$ and $K = 6$. It is equal to 0.4956 and the significance is $t_R = 3.02 > 2$, which indicates the adequacy of the generalized equation.

Based on the equation, 8 was determined as the optimal conditions for the electrochemical dissolution of silver: the density on the silver electrode was 200 A/m², the density on the titanium electrode was 60 kA/m², the electrolyte concentration was 0.5-1.0 mol/l, the electrolyte temperature was 20-40 °C, electrolysis duration - 15-30 minutes, frequency of alternating current - 50 Hz.

As the research results have shown, during the silver polarization by industrial alternating current, silver ions, silver dicyanoferrate are formed.

In the cathode half-period, gaseous hydrogen is released on the titanium electrode and silver ions are reduced. At this point, the silver electrode is in the anodic half-period and dissolves to form silver ions, dicyanoargentate and silver oxide.

With an increase in the current density at the silver electrode, the current output (CO) of silver dissolution decreases (Fig. 1a). This is due to the fact that at high current densities, the proportion of electricity affected by a side process - oxygen evolution - increases compared to the amount of electricity affected by the dissolution process of silver.

The effect of current density on the titanium electrode on the CO dissolution of silver was studied in the range of 20-100 kA/m². The maximum CO value is observed at $i_{Ti} = 60$ kA/m² (Fig. 1b).

When studying the electrolytes concentration effect on the results of electrolysis, it was found that the maximum value of the current efficiency is achieved at a concentration of 0.5 mol/l (Fig. 1c).

Studying the effect of the electrolysis duration on the dissolution process of silver during polarization with industrial alternating current showed that with an increase in the electrolysis duration, the current dissolution rate of silver naturally decreases. It should be noted that on the electrode during prolonged experiments the electrode surface is covered with a layer of dark color.

The study results of the temperature effect on the electrically dissolving the silver electrode showed that the behavior of silver in the electrolyte solution under study has a similar character with the above data (Fig. 1e). An increase in temperature adversely affects the electrochemical activity of silver. Apparently, with increasing temperature, electrode processes become more reversible, that is, compounds formed in the anodic half-period can be restored back to the cathode.

Figure 1f shows the dependence of CO dissolution of silver on the alternating current frequency. With an increase in frequency, the W of metal dissolution decreases. The decrease in the electrolytic silver dissolution with increasing frequency of the current, apparently, is due to the decrease in the duration of the anode half-period.

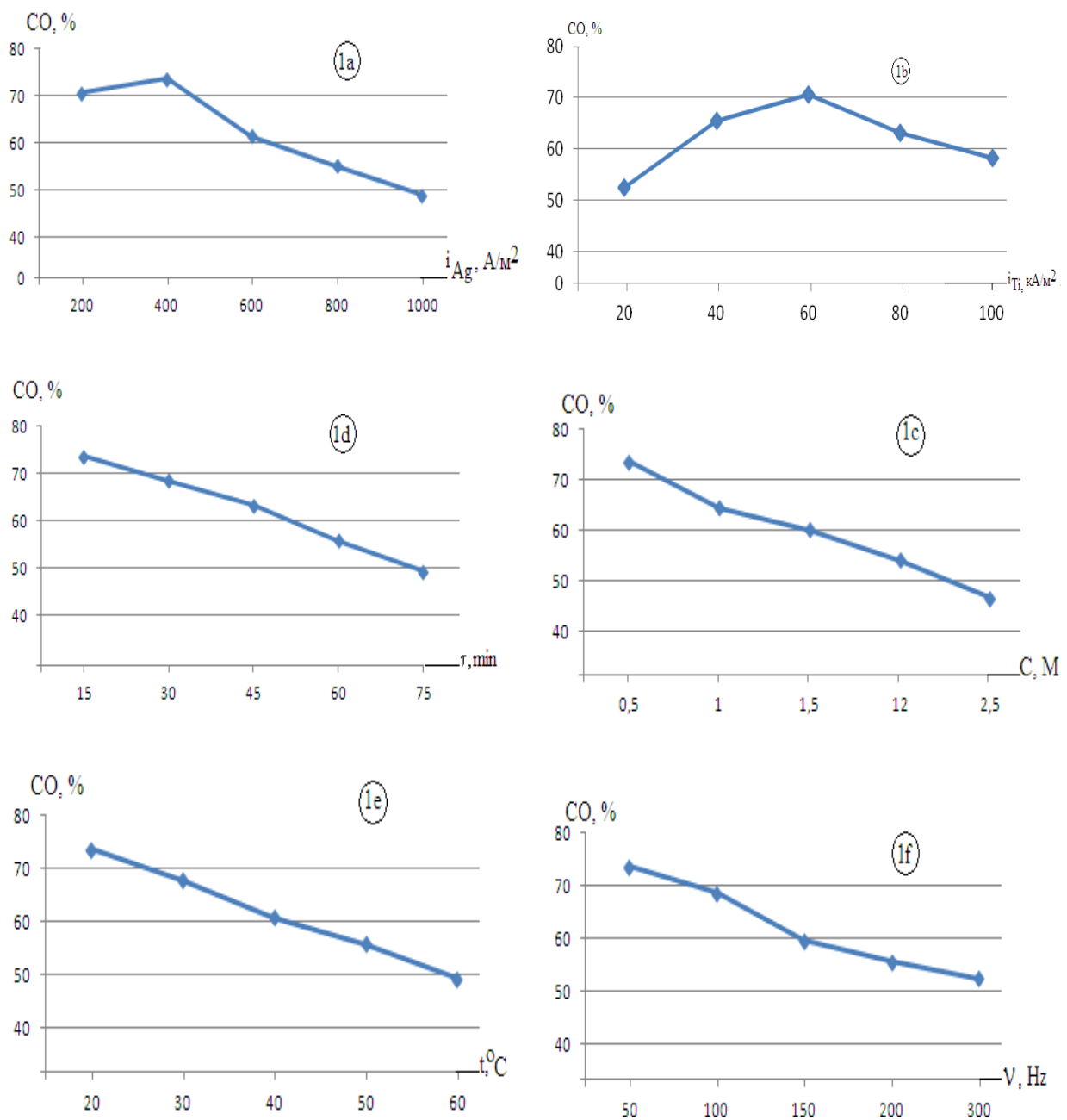


Figure 1 – Point graphs and curve approximation effect on W silver dissolution:
 a – current density on a silver electrode, b – current density on a titanium electrode, c – electrolyte concentration,
 d – electrolysis duration, e – electrolyte temperature, f – AC frequency

Thus, we studied the electrochemical behavior of silver during polarization of industrial alternating current in a potassium ferrocyanide potassium solution by rational mathematical planning, it is shown that when the system of silver-titanium electrodes is polarized by an alternating current, the silver electrode dissolves to form potassium dicyanoargentate, and it has been established that the parameters studied have a significant influence on the silver dissolution.

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КАЛИЙ ФЕРРОЦИАНИДІ ЕРІТІНДІСІНДЕГІ КҮМІСТІҢ ЭЛЕКТРОХИМИЯЛЫҚ ҚАСИЕТІ

Аннотация. Еліміздің инновациялық индустрияландыруының негізгі басымдықтарының бірі – металлургия мен дайын бұйымдарды өндіру. Осыған байланысты, металдарды және олардың қосылыстарын өндірудің жаңа әдістерін қолдану немесе оларды ерітінділерден бөліп алу өте маңызды. Қазіргі кезде қолданылатын перспективалық әдістердің бірі – өндірістік жиіліктегі айнымалы токпен поляризация. Өнеркәсіптік айнымалы токтың әсерінен өтетін процестердің ерекшеліктері ток бағытын ауыстырған кезде, оксидті пленканы алып тастауға және металды әрі қарай үздіксіз еріту үшін жағдай жасауға болады.

Бағалы металдардың, әсіресе, күмістің қатысуымен жүретін электрохимиялық процестер машина жасау мен өнеркәсіптің әртүрлі салаларында, сұраныстың өсуіне байланысты, қызықты болып отыр. Күмістің ең құнды қасиеттері – оның жоғары электр өткізгіштігі, шағылысу қабілеті және жылтыратылған тұнбаның әдемі сәндік түрі.

Калийдің ферроцианид ерітіндісі күмістендіруде цианды электролиттердің ең көп таралған алмастырғышы болып саналады. Күмістің электрототығу заңдылықтарын анықтауға мүмкіндік беретін, оның қосылыстарын синтездеуге жүргізілген жүйелі зерттеулер оның өндірісін қарқындылатуға да, сондай-ақ ресурсты үнемдеу мәселелерін шешуге де әкелуі мүмкін.

Жұмыста айнымалы токпен поляризацияланған күміс электродында калий гексацианоферраты ерітіндісінде жүретін электрохимиялық үрдістердің ерекшеліктері рационалды математикалық жоспарлау әдісі бойынша көрсетілген. Жоспарға сәйкес алты фактор үшін және әрбір фактор бес деңгейге ие 25 эксперимент жүргізілді. Статистикалық көпөлшемді тәуелділіктерді сипаттау үшін жеке функциялар – М.М. Протодьяконовтың теңдеуі қолданылды. Осы теңдеудің негізінде біз $n = 25$ және $K = 6$ үшін корреляция коэффициентін анықтадық. Оның мәні $0,4956$ және $t_r = 3,02 > 2$ және бұл жалпыланған теңдеудің сәйкестігін көрсетеді.

Күмістің электрохимиялық еруінің оңтайлы шаралары анықталды: күміс электродының тығыздығы – 200 А/м^2 , титан электродының тығыздығы – 60 кА/м^2 , электролит концентрациясы – $0,5-1,0 \text{ моль/л}$, электролит температурасы – $20-40 \text{ }^\circ\text{C}$, электролиз ұзақтығы – $15-30 \text{ мин.}$, айнымалы ток жиілігі – 50 Гц .

Титан электродымен жұптасқанда, күміс электродында пассивтену процесі жойылып, металдың еру жылдамдығы артатыны анықталды. Катодты жартылай периодта титан электродында газ тәрізді сутегі бөлінеді және күміс иондарының тотықсыздануы байқалады. Осы сәтте күміс электроды анодты жартылай периодта болады және күміс иондарының, дицианоаргентаттың және күміс оксидінің пайда болуымен ериді. Зерттелген параметрлер күмісті еріту процесіне елеулі әсер ететіні анықталды.

Түйін сөздер: айнымалы ток, күміс, электролиз, айнымалы ток жиілігі, ток тығыздығы, ток бойынша шығым.

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ЭЛЕКТРОХИМИЧЕСКОЕ ПОВЕДЕНИЕ СЕРЕБРА В РАСТВОРЕ ФЕРРОЦИАНИДА КАЛИЯ

Аннотация. Одним из основных приоритетов инновационной индустриализации страны является развитие металлургии и производство готовых продуктов. В этой связи применение новых способов получения металлов и их соединений или извлечения их из растворов является актуальной задачей современности. Одним из перспективных способов, применяющихся в настоящее время, является поляризация переменным током промышленной частоты, которая вызывает интерес исследователей как с

точки зрения оксидообразования, так и разрушения пассивирующей пленки. Особенность процессов, протекающих под действием промышленного переменного тока, заключается в том, что при смене направления тока удается снять оксидную пленку и создать условия для беспрепятственного дальнейшего растворения металла.

Интересным представляются электрохимические процессы с участием благородных металлов, в особенности серебра, в связи с возрастающей потребностью в нем различных отраслей техники и промышленности. Наиболее ценными свойствами серебра является его высокая электропроводность, отражательная способность и красивый декоративный вид полированных осадков.

Наиболее распространенным заменителем цианидных электролитов серебрения является раствор железистосинеродистого калия. Системные исследования, позволяющие установить закономерности электроокисления серебра с последующим синтезом его соединений, могут приводить как к интенсификации его производства, так и решать проблемы ресурсосбережения.

В работе показаны отличительные особенности электрохимических процессов, протекающих на серебряном электроде при электролизе промышленным переменным током в растворе железистосинеродистого калия методом рационального математического планирования. В соответствии с планированием для шести факторов, а в каждом факторе пять уровней было проведено 25 опытов. Для описания статистических многофакторных зависимостей частные функции обобщили уравнение М.М.Протоdjeяконова. На основании уравнения нашли коэффициент корреляции при $N = 25$ и $K = 6$. Он равен 0,4956 и значимость $t_R = 3,02 > 2$, что указывает на адекватность обобщенного уравнения.

Определены оптимальные условия электрохимического растворения серебра: плотность на серебряном электроде – 200 А/м², плотность на титановом электроде – 60 кА/м², концентрация электролита – 0,5-1,0 моль/л, температура электролита – 20-40 °С, продолжительность электролиза – 15-30 мин., частота переменного тока – 50 Гц.

Показано, что при поляризации переменным током серебра в паре с титановым электродом, устраняется процесс пассивации серебряного электрода, и скорость растворения металла возрастает. В катодном полупериоде на титановом электроде выделяется газообразный водород и наблюдается восстановление ионов серебра. В этот момент серебряный электрод находится в анодном полупериоде и растворяется с образованием ионов серебра, дицианоаргентата и оксида серебра. Установлено, что изученные параметры оказывают заметное влияние на процесс растворения серебра.

Ключевые слова: переменный ток, серебро, электролиз, частота тока, плотность тока, выход по току.

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