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ESTABLISHMENT OF TECHNOLOGICALLY FEASIBLE MODES OF ELECTROCOAGULATION PURIFICATION OF WASTEWATER FROM NICKEL IONS

The object of research is the technology of electrochemical purification of wastewater from heavy metal ions. The work, in particular, is devoted to the purification of effluents with a low concentration of the Nickel ion. The main task of the experimental research was to select the material of the electrodes and the mode of electrochemical dissolution of the anodes, under which the efficiency of wastewater purification will be maximum, as well as to confirm the practical possibility of deep purification of the specified wastewater to the standards that correspond to the maximum permissible concentrations ($MPC_{Ni}=0.5 \text{ mg/dm}^3$).

It was established that in the process of electrocoagulation wastewater purification there is an induction period (10 min), during which the coagulant accumulates in the electrolyzer and the degree of purification increases sharply. As a result of an increase in the current density from 10 A/m^2 to 20 A/m^2 , the degree of purification on iron electrodes increased from 60 to 84 % for a process duration of 20 minutes. This is explained by the intensification of the anodic dissolution of the metal and the increase in the concentration of $\text{Fe}(\text{OH})_3$. Increasing the current density to 30 A/m^2 practically does not affect the degree of purification, which is explained by the phenomenon of polarization of the anodes and is confirmed by the increase in the process voltage from 2.40 V (10 A/m^2) to 12.59 V (30 A/m^2). Therefore, it is impractical to increase the current density in the future. For iron anodes, it was not possible to achieve the required degree of purification ($\geq 98.3 \%$), the maximum degree of purification did not exceed 85 %, and the content of Ni^{2+} ions in purified water exceeds the MPC by an order of magnitude.

It was experimentally established that it is advisable to use aluminum electrodes for the process of electrocoagulation purification of wastewater from Nickel ions. At a current density of 20 A/m^2 and process duration of 40 minutes, the concentration of Ni^{2+} ions did not exceed the MPC of Ni. When using aluminum electrodes, an increase in the current density from 15 A/m^2 to 20 A/m^2 does not lead to polarization of the electrodes, and the process in both cases takes place at a steady state at a voltage of $\sim 6.7 \text{ V}$. The technologically appropriate operating mode of the electrolyzer is chosen: aluminum electrodes at an anodic current density 20 A/m^2 and the duration of the purification process – $\geq 40 \text{ min}$. The obtained results can find practical use in the design of waste water purification systems of galvanic industries.

Keywords: wastewater purification, electrochemical purification, heavy metals, Nickel compounds, electrocoagulation, aluminum electrodes.

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1. Introduction

The lack of quality drinking water is a global problem of humanity. Scientists predict that by 2025, about 60 % of the planet's inhabitants will suffer from a lack of fresh water. This can negatively affect many aspects of modern life [1]. Therefore, at the UN Summit on Sustainable Development, one of the global directions of human development was declared «Goal 6. Clean water and adequate sanitation», the purpose of which is: «to ensure the availability and sustainable management of water resources and sanitation for all» [2].

Humanity in the process of its historical development has always been closely connected with nature, being its integral part. With the growth of the Earth's population and the industrialization of society, the anthropogenic influence of man has increased significantly, in particular, the problem of irreversible pollution of water bodies with sewage has arisen, which makes it unsuitable for consumption by people, animals and plants. The problem of water pollution with heavy metal compounds, which can have a negative impact on human health and the environment, is particularly acute [3]. One of the main pollutants is Nickel

compounds, which are named by the World Health Organization as one of the most dangerous environmental pollutants [4]. Therefore, the problem of wastewater purification is definitely relevant.

The use of electrochemical methods of water purification is technologically and economically feasible for the purification of galvanic effluents from heavy metal ions. According to modern views, the process of electrochemical purification has tangible advantages over traditional technologies, namely: complete removal of persistent pollutants, environmental friendliness, and ease of integration with existing technologies, smaller volumes of formed sediments, a high level of separation and a small duration of purification [5, 6]. In addition, electrochemical installations are simple, compact, and easily amenable to automation, practically do not require reagents, have no moving parts, can use renewable sources of electrical energy, in particular, solar and wind energy.

Many modern studies on water purification technology are devoted to advanced electrochemical processes of oxidation (electrochemical advanced oxidation process (EAOP)), which are based on the electrolysis of aqueous solutions. They include electrochemical oxidation technologies, electroprocesses Fenton and photoelectrocatalysis [7]. Electrooxidation is a promising method, which, according to modern concepts, takes place according to the mechanisms of direct and indirect oxidation. Direct oxidation of pollutants occurs directly at the anode by direct transfer of electrons. Indirect – as a result of the formation of active substances (radicals) during the electrolysis of effluents, which oxidize pollutants [8]. The paper [9] describes the results of studies of the process of electrooxidation of nickel and copper ions, to achieve the required result; the duration of the process was 60 min, at pH=4.5 and a voltage of ~28 V. In the rest, the researchers studied the purification of water from complex nickel-ammonia compounds by combination of electrooxidation and electrodeposition processes using RuO₂/Ti and stainless steel electrodes. The extraction degree of Nickel was 99 % at pH=9.0 [10].

EAOPs are effective processes for the degradation of wastewater containing hazardous pollutants. However, their economic feasibility is determined by the materials of the electrodes. Electrodes are subject to increased requirements regarding chemical resistance and catalytic activity. Usually, combined electrodes are used, which contain precious metals, which significantly increase the cost of purification. Most of the studies described in the literature were conducted on a laboratory scale, in stationary conditions. Therefore, for the perspective of industrial implementation, research is needed on pilot plants in factory conditions, using real industrial effluents [8].

Considering the current state of science and technology, electrocoagulation is the most promising industrial method of purifying wastewater from heavy metal ions. Electrocoagulation wastewater purification has the following advantages:

- compactness of installations that are easily amenable to automation and as a result – simple control of the technological process, with the possibility of creating closed circulating water supply systems;
- lack of reagent economy, insignificant influence on the course of the process of technological parameters;
- a concomitant decrease in the content of water hardness ions by 15–20 %, sulfates and chlorites, respectively, by 8–12 % and 3–15 %;
- reduction of costs for disinfection of purified water due to the bactericidal effect of electric current on microorganisms.

Electrocoagulation is a process of accelerated sticking together of colloidal particles under the action of an electric field. This method of water purification has been known since 1887, but the experience of using this method in the technology of wastewater purification from heavy metals is insignificant [11, 12].

The process of electrocoagulation is complex and its mechanisms of influence can be of different nature:

- electrochemical (anodic metal dissolution, reduction and oxidation of water, direct electrooxidation and/or electroreduction of pollutants, etc.);
- chemical (displacement of the acid-alkaline balance of water, redox processes in volumes caused by electrolysis products, etc.);
- physical (adsorption, coagulation, flotation, etc.).

All these processes can proceed both in parallel and sequentially [12]. Iron or aluminum soluble anodes are used in electrocoagulation technology due to their cheapness, good sorption properties, low solubility of electrolysis products, and non-toxicity.

Al³⁺, Fe²⁺ and Fe³⁺ ions are extremely active, therefore, they are quickly hydrolyzed with the formation of hydroxide compounds with a developed surface that absorbs pollutants well. Aluminum is usually used to purify drinking water, and iron is used to purify wastewater [11].

Electrocoagulation is recommended to be carried out in neutral or weakly alkaline environments under the following conditions:

- current density no more than 10 A/m²;
- the distance between the electrodes is no more than 20 mm;
- the speed of water movement is not less than 0.5 m/s.

Application of the method is expedient for relatively high electrical conductivity of wastewater [13].

In more modern studies [12, 14], for long-term system operation without maintenance, it is recommended to use a current density of 20–25 A/m² and to minimize the inter-electrode distance. Obviously, such differences are due to the complexity of the process, in particular, the influence of the electrode passivation effect on its implementation. Electrochemical properties of electrodes will depend on their nature, composition of electrolytes, presence of activating additives, current density, etc.

Taking into account the above, there was a need to conduct research that will become a reliable basis for the creation of a highly efficient electrocoagulation technology for purification wastewater from Nickel ions.

Therefore, *the aim of the study* is to establish technologically appropriate modes of electrocoagulation purification of low-concentrated Ni²⁺ ion effluents (with Ni ≈ 30 mg/dm³ [15]) to the norms of the MPC of Ni (0.5 mg/dm³ [4]).

To achieve the goal, the following tasks must be solved:

- to investigate the effect of the nature of the electrode material on the process of extracting Nickel ions from wastewater;
- to study the influence of the current density and the duration of the process on the efficiency of sewage purification.

2. Materials and Methods

2.1. Description of the laboratory setup and methods of conducting experiments. *The object of the study* is technologies of electrochemical wastewater purification from heavy metal ions.

The subject of the study is technological processes of electrocoagulation purification of galvanic effluents from Nickel ions.

The research was carried out on the installation shown in Fig. 1. The main device is an electrocoagulator 1 fixed on a tripod, the body of which is made of polypropylene. 11 electrodes are placed in retainers 4: 6 anodes and 5 cathodes with a distance of 10 mm between them. The size of the electrodes is 100×60 mm, respectively, the working area of the anodes is 6 dm².

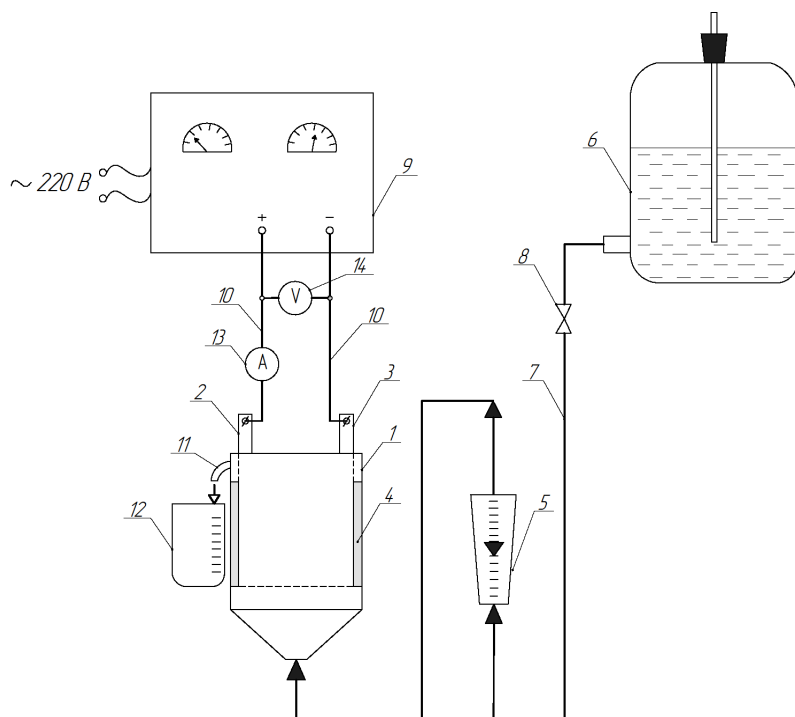


Fig. 1. Installation for experimental research on the electrochemical purification of wastewater by the electrocoagulation method: 1 – electrocoagulator; 2 – anode; 3 – cathode; 4 – dielectric insert; 5 – rotameter; 6 – pressure tank; 7 – hose; 8 – clamp; 9 – current source; 10 – current outlets; 11 – fitting for draining purified water; 12 – collective capacity; 13 – ammeter; 14 – voltmeter

Current from the rectifier 9 is supplied to the electrodes through the current taps 10. With the help of the ammeter 13 and the voltmeter 14, the current and voltage are measured and controlled. A pressure tank 6 is placed above the electrocoagulator, into which waste water is poured, which enters the main device 1 by gravity through a hose 7. The regulation and control of the water flow is carried out using a clamp 8 and a rotameter 5. The sample for analysis is taken through the fitting 11 into the collection tank 12. The selected samples were filtered through a paper filter.

A solution of nickel sulfate with a concentration of Nickel ions of ~30 mg/dm³ is used to obtain an imitation of waste water. The solution was prepared from crystal hydrate (NiSO₄·7H₂O) of reactive purity and distilled water. In order to increase the electrical conductivity, 25 % of NaCl was added to the resulting solution based on the concentration of Nickel ions.

The research was carried out using iron and aluminum electrodes in flow mode. The current strength was stabilized, providing the value of the anode current density in the range of 10–30 A/m². Imitation wastewater was uniformly fed into the electrolyzer with a flow rate of 0.1 dm³/min. The research

was conducted at a room temperature of 26 °C. The volume of the electrolyzer is 1.20 dm³, so the average contact time of wastewater with the electrodes was 12 minutes. Wastewater samples were periodically taken, analyzed for the content of Ni²⁺ ions, and the degree of purification was calculated. In the course of research, changes in voltage and pH were recorded.

The experimental sample included the results of multiple measurements. At least 3 experimental values were obtained for each indicator. Therefore, it can be stated that the specified sample is sufficiently representative, and the obtained results are reliable.

2.2. Conducting method analyses and calculations in. Nickel ions in the solution before and after the experiment were determined titrimetrically.

Reagents: buffer solution (54 g of NH₄Cl and 350 cm³ of 25 % NH₄OH in 1 dm³); 0.01 N Trilon B solution; murexide indicator (murexide:NaCl=1:100).

Analysis progress. In a conical flask with a capacity of 250 ml, 50 ml of the investigated filtrate was measured with a pipette, 100 ml of distilled water and 10 ml of buffer solution were added, and 0.1–0.2 g of the murexide indicator was added. The sample was titrated with a 0.01 N Trilon B solution until the color changed from yellow to red-violet.

Nickel ions ($C_{Ni^{2+}}$, mg/dm³) was calculated according to the formula:

$$C_{Ni^{2+}} = \frac{V_{Tr} \cdot N_{Tr} \cdot E_{Ni} \cdot 1000}{V_{Ni}}$$

where V_{Tr} is the volume of Trilon B solution used for sample titration, ml; N_{Tr} is the normal concentration of trilon B, eq/dm³; V_{Ni} is the sample volume for analysis, ml; E_{Ni} is the equivalent mass of Nickel.

pH value was determined using a pH meter of the brand PH-673M (Georgia) and a pair of electrodes – glass ESL-43-07 and silver chloride EVL-1M (Republic of Belarus).

The degree of sewage purification (X , %) was calculated according to the formula:

$$X = \frac{C_{Ni^{2+}}^0 - C_{Ni^{2+}}^\tau}{C_{Ni^{2+}}^0} \cdot 100,$$

where $C_{Ni^{2+}}^0$ and $C_{Ni^{2+}}^\tau$ are the concentrations of Nickel ions in the initial sample and in the sample taken after a certain period of time, respectively, mg/dm³.

Processing, analysis and visualization of the obtained experimental results were performed using the Microsoft Excel program (Microsoft 365). To derive mathematical dependencies, the method of least squares was used with approximation of values by appropriate functions. In cases where the experimentally obtained curves were of a complex nature, the results were approximated for the corresponding intervals of the input variables. The division of the definition area into intervals was carried out on the basis of general theoretical ideas about the physico-chemical essence of electrocoagulation purification processes. Equations and values of approximation probabilities (R^2) are given in the tables below the corresponding figures.

3. Results and Discussion

In the first series of experiments, the influence of current density on the process of electrocoagulation using iron electrodes was studied. The results of research are presented in the Table 1.

Dependence of the concentration of Nickel (+2) ions on the duration of the process and the current density on iron anodes

Time (τ), min	$i=10 \text{ A/m}^2$			$i=15 \text{ A/dm}^{2*}$			$i=20 \text{ A/dm}^2$			$i=30 \text{ A/dm}^2$		
	C_i , mg/dm ³	U , V	pH	C_i , mg/dm ³	U , V	pH	C_i , mg/dm ³	U , V	pH	C_i , mg/dm ³	U , V	pH
0	32.45	2.40	5.43	32.16	3.64	5.52	32.45	10.37	5.45	32.45	12.59	5.47
10	14.16	2.20	6.15	14.75	3.93	6.25	6.49	10.63	5.98	8.85	12.11	6.06
20	14.16	2.10	6.21	13.28	3.74	6.41	5.90	11.68	6.06	5.16	11.86	6.15
30	12.98	2.20	6.22	13.28	3.52	6.21	5.31	10.37	6.18	5.16	12.30	6.21
40	12.98	2.19	6.28	11.51	3.40	6.31	5.31	10.20	6.26	5.16	12.53	6.26
50	10.62	2.19	6.26	10.77	2.99	6.21	5.31	10.58	6.24	4.72	11.85	6.23
60	–	–	–	10.77	3.00	6.24	4.13	10.14	6.25	4.72	11.50	6.25

Note: * – NaCl content in the emitate is 10 % of the concentration of Nickel ions

In the process of electrocoagulation due to the cathodic decomposition of water, the pH value of the solution increased from 5.43–5.52 to 6.24–6.28.

The dependence of the degree of wastewater purification on the current density and the duration of the process on iron anodes are shown graphically in Fig. 2.

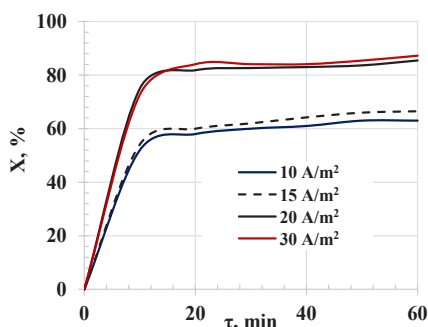


Fig. 2. Dependence of the degree of wastewater purification on the current density and duration of the process on iron anodes (Table 2)

Table 2

Approximation equation of the experimentally obtained results of the study of the dependence of the degree of wastewater purification on the current density and the duration of the process on iron anodes (Fig. 2)

Current density, A/m ²	Stages of the process and their duration	
	1 st stage, 0–10 min	2 nd stage, 10–60 min
	Equations and values of approximation probabilities (R^2)	
10	$X=5.4\cdot\tau$; $R^2=0.9986$	$X=0.0001\cdot\tau^3-0.0135\cdot\tau^2+0.6258\cdot\tau+50$; $R^2=0.9827$
15	$X=5.4108\cdot\tau$; $R^2=1$	$X=7\cdot 10^{-5}\cdot\tau^3-0.0122\cdot\tau^2+0.8077\cdot\tau+47.449$; $R^2=0.9865$
20	$X=7.28\cdot\tau$; $R^2=0.9964$	$X=0.0004\cdot\tau^3-0.0442\cdot\tau^2+1.7291\cdot\tau+61.818$; $R^2=0.9989$
30	$X=7.3382\cdot\tau$; $R^2=0.9997$	$X=0.0006\cdot\tau^3-0.069\cdot\tau^2+2.6048\cdot\tau+53.467$; $R^2=0.9579$

The nature of the change in the degree of purification of wastewater from Ni²⁺ ions (Fig. 2) is approximately the same for all current densities. At first, within 10 minutes, there is a sharp increase in the degree of purification from Ni²⁺ ions, then the degree of purification increases slightly, and after about 20 minutes, the degree of purification practically does

Table 1

not change. This type of dependence can be explained by the presence of the so-called induction period, that is, the time (10 min) during which Fe²⁺ ions accumulates in the system. As mentioned above, as a result of the cathodic decomposition of water, the pH of the solution increases, which significantly increases the rate of oxidation of Fe²⁺ ions by water-soluble oxygen to Fe³⁺ [16, 17]. Fe³⁺ ions are hydrolyzed to form an active coagulant – Fe(OH)₃. At a certain moment (in 20 minutes from the start of the experiment), the reactor reaches a state of saturation in terms of Ferrum compounds and a dynamic equilibrium is established – the amount of coagulant formed and removed from the reactor is the same, so the degree of purification does not change.

Increasing the current density (Fig. 2) in general contributes to the deepening of the purification process. Thus, as a result of increasing the current density from 10 A/m² to 20 A/m², the degree of purification in 20 minutes of research increased significantly: from 60 to 84 %, which is explained by the intensification of the anodic dissolution of the metal and the increase in the yield of Fe(OH)₃. Increasing the current density to 15 A/m² does not have a noticeable intensifying effect. This is obviously due to the fact that the NaCl content in the solution was lower (10 %) compared to other experiments (25 %). Increasing the current density to 30 A/m² practically does not affect the depth of purification, and the values of the degrees of purification are practically similar to the results obtained at a current density of 20 A/m². This is well explained by the phenomenon of anode polarization, and is confirmed by the increase in process voltage to 12.59 V (Table 1). That is, it is impractical to increase the current density in the future. In the course of research, it was noticed that at constant values of the current densities, the voltage decreases slightly over time. This is due to the heating of the electrolyte in the electrolysis process and, accordingly, an increase in its conductivity.

Despite the generally positive effect on the process of increasing the current density, for iron anodes it was not possible to achieve the required degree of purification (≥ 98.3 %), in the best case, at a current density of 20 A/m², it did not exceed 85 %. This result is unacceptable, since the content of Ni²⁺ ions exceeds the MPC by an order of magnitude. Therefore, the next series of experiments was conducted using aluminum anodes.

The research conditions were similar to those for iron electrodes. The results of research are presented in the Table 3.

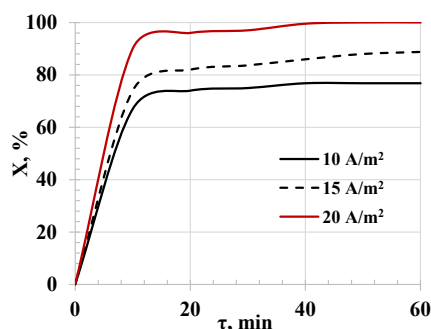
In the process of electrocoagulation using aluminum electrodes due to the cathodic decomposition of water, the pH value of the solution increased from 5.71–5.90 to 6.08–6.90.

Table 3

Dependence of the concentration of Nickel ions (+2) on the duration of the process and the current density on aluminum anodes

Time (τ), min	$i=10 \text{ A/m}^2$			$i=15 \text{ A/dm}^2$			$i=20 \text{ A/dm}^2$		
	C_i , mg/dm ³	U_i , V	pH	C_i , mg/dm ³	U_i , V	pH	C_i , mg/dm ³	U_i , V	pH
0	31.86	5.5	5.72	31.57	5.67	5.71	31.57	5.90	6.23
10	8.85	5.77	5.99	7.67	5.95	6.92	1.92	6.57	6.80
20	8.85	6.07	6.06	6.2	6.19	7.21	1.92	6.68	6.57
30	8.11	6.23	6.03	5.16	6.57	7.00	1.18	6.74	6.48
40	7.38	6.44	6.08	4.43	6.70	6.90	0.15	6.72	6.37
50	7.38	6.68	6.02	3.54	6.69	6.90	0.0	6.72	6.49
60	7.38	6.68	6.03	3.54	6.69	6.90	0.0	6.74	6.21

The dependence of the degree of wastewater purification on the current density and the duration of the process on aluminum anodes are shown graphically in Fig. 3.

**Fig. 3.** Dependence of the degree of wastewater purification on the current density and duration of the process on aluminum anodes (Table 4)**Table 4**

Approximation equation of the experimentally obtained results of the study of the dependence of the degree of wastewater purification on the current density and the duration of the process on aluminum anodes (Fig. 3)

Current density, A/m ²	Stages of the process and their duration	
	1 st stage, 0–10 min	2 nd stage, 10–60 min
	Equations and values of approximation probabilities (R^2)	
10	$X=7.2178 \cdot \tau$ $R^2=1$	$X=-6 \cdot 10^{-5} \cdot \tau^3+0.0038 \cdot \tau^2+0.0605 \cdot \tau+71.369$ $R^2=0.9614$
15	$X=7.5764 \cdot \tau$ $R^2=1$	$X=8 \cdot 10^{-5} \cdot \tau^3-0.0132 \cdot \tau^2+0.8409 \cdot \tau+68.853$ $R^2=0.9782$
20	$X=9.28 \cdot \tau$ $R^2=1$	$X=-9 \cdot 10^{-5} \cdot \tau^3+0.0068 \cdot \tau^2+0.0382 \cdot \tau+92.159$ $R^2=0.9694$

In general, the nature of the obtained dependencies of the degree of purification over time is similar to the results of research with iron electrodes. At the same time, the purification performance is much better. Thus, at the 20th minute of the experiment, the degree of purification for the current densities, A/m²: 10, 15 and 20 were, %: 74.0; 82.0 and 95.5, and after 40 minutes from the beginning of the experiment, it was possible to achieve the following values of the degree of purification of water from Nickel ions, %: 76.8; 86.0 and 99.5, respectively. That is, at a current density of 20 A/m² and a process duration of 40 min, the concentration of Ni²⁺ ions did not exceed the MPC, and later the degree of purification approached

100 % (the concentration of Ni²⁺ ions was beyond the sensitivity of the analysis).

The dependence of the electrolysis voltage on the current density and the duration of the process on aluminum electrodes (Table 3) are significantly different from the similar one for iron electrodes (Table 1). At current densities of 15 and 20 A/m² in the electrolysis process, a slight increase in voltage was observed from 5.7 and 5.9 V to approximately 6.7 V, respectively. From the 40th and 20th minutes (for densities of 15 and 20 A/m²), the process in both cases took place stationary at voltages of ~6.7 V. That is, it can be stated that the process proceeds without polarization of the electrode, and a slight increase in voltage at the beginning of the process is associated with the activation of the surface of aluminum electrodes.

The conducted studies showed high efficiency of electrocoagulation wastewater purification from Nickel ions, have practical value for designing wastewater purification systems of galvanic industries. However, for the effective practical implementation of the process in industry, a deep understanding of a number of features and limitations is necessary, in particular:

- electrocoagulation purification is accompanied by the dissolution of anodes, therefore, in order to maximize the use of electrode material, electrolysis must be carried out in reverse mode;
- polarization phenomena on the electrodes are determined by the limiting stage of electrolysis, which can be of different nature (diffusion, chemical, electrochemical, concentration, phase, etc.). For the industrial scaling of electrocoagulation installations, it is necessary to experimentally establish the type of overvoltages under various investigated conditions.

The conditions of martial law in Ukraine affected the conduct of research; in particular, the forced transition to a distance form of education significantly complicated the conduct of the experiments described above. Therefore, the subject of further research will be the study of the influence of electrolysis modes, Ni²⁺ concentration in wastewater, electrolyte conductivity, pH value, temperature, and the average contact time of effluents with electrodes on the efficiency of the electrocoagulation plant.

4. Conclusions

1. The presence of an induction period, a time (10 min) during which the coagulant accumulates in the electrolyzer and a sharp increase in the degree of purification is characteristic of the process of electrocoagulation purification of wastewater from Nickel ions on iron and aluminum electrodes.

2. An increase in the current density from 10 A/m² to 20 A/m² intensifies the process of anodic dissolution of iron, which leads to an increase in the concentration of coagulant and the achievement of a maximum degree of purification of 85 %. A further increase in the current density is impractical, since at a current density of 30 A/m² the anodes are polarized and the degree of sewage purification practically does not change.

3. In the case of using iron anodes, the degree of wastewater purification does not meet environmental protection standards ($X \geq 98.3$ %), the maximum degree of purification did not exceed 85 %, and the concentration of Nickel ions exceeds the MPC of Ni by 8–9 times.

4. It is technologically expedient to use aluminum anodes for the process of electrocoagulation purification of wastewater from Ni²⁺ ions. With an anodic current density of 20 A/m² and the duration of the purification process ≥40 min, the wastewater is practically completely cleaned of Nickel ions.

Conflict of interest

The authors declare that they have no conflict of interest in relationship that this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

Financing

The study was performed without financial support.

Data availability

Data will be made available on reasonable request.

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