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PERFORMANCE OF ELECTROCOAGULATION PROCESS USING IRON AND ALUMINUM ELECTRODES WITH AND WITHOUT PERFORATIONS

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Abstract. Electrocoagulation process is widely used for the removal of pollutants from the industrial wastewater. In the present study, an attempt was made to investigate the performance of electrocoagulation process using aluminum and iron electrodes to treat the metal ions present in the synthetic galvanic wastewater. The electrodes used are with and without perforations and it was observed that the efficiency of electrodes with perforation (80 %) was higher than without perforations (50 %). The removal efficiency of heavy metal ions increased with retention time and direct current. The optimized values of residence time, voltage, pH, current, electrode spacing were 160 min, 6 V, 5, 0.2 A, and 3 cm, respectively. The maximum removal percentage of nickel and copper ions using perforated iron electrodes was 90.7 % and 86.0 %, respectively, and for chromium using a combination of perforated iron and aluminum electrodes it was 93.1 %. The removal of metal ions followed pseudo second order kinetic model with current dependent parameters.

Keywords: synthetic galvanic wastewater, electrocoagulation process, heavy metals, electrodes, water treatment.

1. Introduction

In the developing nations, particularly in India, the monitoring of the water pollution is based on the concentration limits of various pollutants present in discharge effluents. Industries prefer dilution of discharge effluents using fresh water to meet the concentration limits. Treatment and reuse of wastewater with low cost, environmental friendly technology is the most acceptable method rather than dilution and disposal. The latter method increases the contamination of natural water resources. Out of all the pollutants discharged in natural water bodies, heavy metals are considered to be lethal to the living organisms. Several treatment methods are used for the removal of heavy metals from wastewater like adsorption, precipitation, coagulation, ion exchange, electro dialysis, electro winning, electrocoagulation, cementation, and reverse osmosis.

The electrocoagulation/flotation process involves applying an electric current to sacrificial electrodes inside a reactor tank. The current generates coagulating agents (metal ions) and gas bubbles. Metal ions generated from sacrificial electrode coagulate with pollutants in the water. The process is similar to the addition of coagulating chemicals such as alum and ferric chloride. It allows for easier removal of the pollutants by sedimentation and flotation. Electrocoagulation or electroflotation are the technologies that are developed based on the concepts of electrochemical cells, specifically known as "electrolytic cells".[1] Though electrocoagulation is an energy intensive process, it is easy to operate and produce clean, odorless water. The electrocoagulation treatment process has the advantage of removing the smallest colloidal particle, because the applied electric field sets them in fast motion, thereby facilitating the coagulation. This process avoids the uses of chemicals, and so there is no problem of neutralizing excess chemicals. There is no possibility of secondary pollution caused by chemical substances added at high concentrations. The gas bubbles produced during electrolysis can carry the pollutant to the top surface of the solution, where it can be easily concentrated, collected, and removed. The electrolytic processes in the electrocoagulation cell are controlled electrically with no moving parts, thus requiring less maintenance. Several studies are reported in literature for the removal of heavy metals using various electrocoagulation processes.[2,3] Forat Y. Al Jaberi *et al.*[4] investigated the performance of innovative electrocoagulation reactor for the removal of oil content and turbidity from real oily wastewater. The parameters were optimized using response surface methodology and minitab-17 statistical program. Concentric aluminum tubes were used as electrodes and obtained positive results. Marco-Morales *et al.*[5] evaluated the removal efficiency of suspended solids from the chocolate manufac-

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turing industry wastewater by integrating the coagulation process with sand filtration. Substantial reduction in turbidity, color, and chemical oxygen demand (COD) was observed using this combination treatment technique. Electrocoagulation process was employed to the effluents of Gaza wastewater treatment plant (GWWTP). The performance was studied in the optimized conditions for the removal of nitrate, total hardness, calcium and magnesium. Good quality of treated water was obtained for reuse purpose.[6] Jagadal *et al.*[1] applied electrocoagulation process to treat wastewater from dairy industry**.** They concluded that electrocoagulation process with series electrode configuration could be an alternative for cost effective treatment of dairy effluents. Sadiq Muhsun *et al.*[7] studied the removal of heavy metal ions, cadmium, nickel and lead from a synthetic wastewater using a lab scale electrocoagulation system. Results showed that the optimal operating conditions for the maximum removal efficiencies occur at pH 7, current density, 12.5 mA/cm^2, inner electrode distance, 1cm for both aluminium and iron electrodes. The detention time was 150 min for aluminium, and 120 min for iron electrodes and the performance of aluminium electrodes was found to be better than iron electrodes. The present work was carried out to study the performance of iron, aluminium, and combination of both electrodes with and without perforations for the treatment of metal ions present in synthetic galvanic wastewater.

2. Materials and Methods

2.1. Chemicals and Reagents

Nickel sulfate hexahydrate ($NiSO_4(H_2O)_6$), copper sulfate pentahydrate ($CuSO_4{\cdot}5H_2O$), chromium (III) sulfate ($Cr_2(SO_4)_3{\cdot}6(H_2O)$), potassium dichromate ($K_2Cr_2O_7$), potassium permanganate($KMnO_4$), aqueous ammonia solution were purchased from Loba Chemie Pvt. Ltd. Hydrochloric acid (HCl) was procured from Fischer Scientific. Double distilled water was used to prepare all the solutions. The chemicals and reagents were analytical grade and used as pure as supplied.

2.2. Preparation of Synthetic Galvanic Solution

Metal stock solutions of 2 g/L of Ni, 2.5 g/L of Cu, and 0.7 g/L of Cr were prepared by weighing respective salts. Stock solution of 0.1 N HCl was also prepared. A synthetic galvanic wastewater solution was prepared using the individual metal ion solutions. The composition of the synthetic wastewater was prepared to represent similar conditions of real time industrial galvanic wastewater.

2.3. Concentration and pH Measurement

The concentration of metal ions in the synthetic wastewater was measured using 220 FS atomic absorption spectrophotometer. The initial pH of the synthetic wastewater was measured using Jenway 3520 pH meter.

2.4. Experimentation

All the experiments were carried out in a batch reactor. A schematic diagram representing the mechanism of electrocoagulation process is shown in Fig. 1. The electrodes were made up of iron (mild steel, ST 37) and aluminum plates ($AlMg_3$), 78 mm height, 99 mm width, 2 mm thick in dimensions. Each electrode was perforated with 28 holes of 5 mm in diameter. Fig. 2 shows the experimental setup. Two electrodes were installed vertically with a spacer to ensure fixed distance of 3 cm. A sample figure of the iron electrode before and after electrocoagulation is shown in Fig. 3.

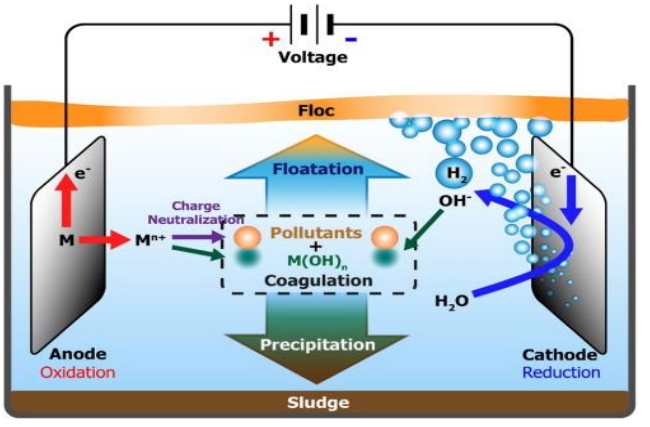

Fig. 1. Mechanism of electrocoagulation[4]

Fig. 2. Experimental set up

Fig. 3. Iron electrode before and after electrocoagulation process

2 L of synthetic galvanic wastewater was used for each experimental run. Desired current was applied and 10 mL samples were collected at each time interval of 20 min. The samples were taken from the bulk solution near the anode and cathode. The samples at anode were analyzed for the degree of metal extraction. Experiments were repeated with iron, aluminum, and a combination of both electrodes. The concentration of metal ions in the synthetic wastewater was measured by taking the samples at 20 min intervals from 0 to 180 minutes. The experiments were repeated by varying the initial pH of the wastewater at an optimum retention time, then varied current at the optimum retention time and initial pH. The set of experiments were conducted with same electrodes and a combination of electrodes with and without perforations.

3. Results and Discussion

3.1 Electrodes without Perforations

2 L of synthetic galvanic wastewater was taken into the reactor system. Two iron electrodes without perforations were immersed and kept 3 cm apart. The initial pH of the solution was recorded as 2.5. 0.1 V voltage, 0.06 A current, and a current density of 0.371 mA/cm^2 was passed through the electrodes. Samples were withdrawn for every 20 minutes near anode and cathode. The samples were then analyzed for the concentration of metal ions (Ni, Cu and Cr). Fig. 4 shows the removal percentage of metal ions. The optimized value for retention time was reached after 160 min. The maximum removal percentage of Ni, Cu and Cr was estimated as 40.0 %, 30.0 % and 27.5 %, respectively. The adsorption of Ni onto $Fe(OH)_3$ flocs seems to be higher than Cu and Cr. The experiments were repeated using aluminum electrodes without perforations. The voltage, current and current density increased to 6.5 V, 1.02 A, 6.315 mA/cm^2. The initial pH and distance between electrodes maintained same. The heavy metals concentration decreased gradually and gets optimized at 160 min of retention time. The maximum removal percentage of Ni, Cu, Cr was14.4 %, 31.0 %, 12.8 %, respectively, as shown in Fig. 5. Unlike in iron electrodes, here the removal efficiency was higher for Cu. This indicates that the mass transfer resistance for the adsorption of Cu seems to be lesser than Ni and Cr. There is a possibility of parallel water electrolysis at 6.5 V, which enhances hydroxyl radicals resulting in higher flocs formation. Combination of iron and aluminum electrodes without perforations was then immersed in the synthetic wastewater and the experimental run was carried out. Iron electrode as cathode and aluminum electrode as anode was taken. The electrodes are kept 3 cm apart. The initial pH of wastewater was measured as 2.5. 3.6 V voltage, 1.02 A current and current density of 6.315 mA/cm^2 was passed through the electrodes. The removal percentage of metal ions was more or less similar to the performance of iron electrodes without perforations. The maximum removal percentage of Ni, Cu, Cr after 160 min retention time was 24.0 %, 37.5 %, 40.0 %, respectively, as shown in Fig. 6. In the combination of electrodes, the adsorption of Cr on $Al(OH)_3$ flocs was higher than Ni and Cu. At 3.6 V, there may be a possibility of parallel water electrolysis, which might enhance the formation of $Cr(OH)_3$. This will increase the chromium flocculation effect. The initial pH of the solution plays a vital role. The reaction rates during electrocoagulation process were found to be higher at neutral or alkaline pH.[8] At the low initial pH of 2.5, the removal efficiency of metals might be affected, and hence lower removal percentage efficiencies was observed. This effect was clearly observed when the experiments were conducted with increase in initial pH. The other factor that affects the removal efficiency directly is the current density. Low current density produces lesser hydroxide flocs and high current leads to agglomeration of flocs. Hence an optimum current density need to determine, which will give maximum removal efficiency of the metals.[8]

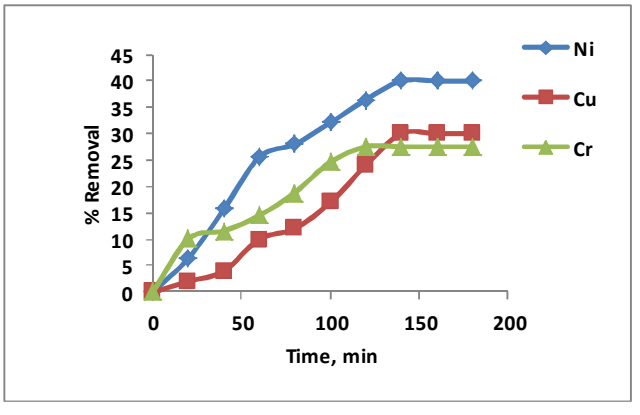

Fig. 4. Iron electrodes without perforations

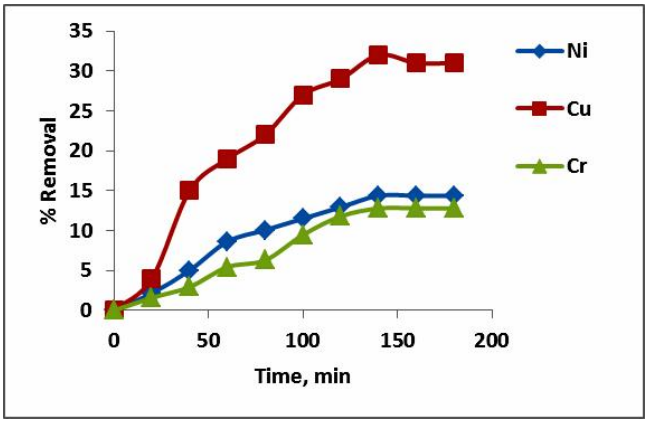

Fig. 5. Aluminum electrodes without perforations

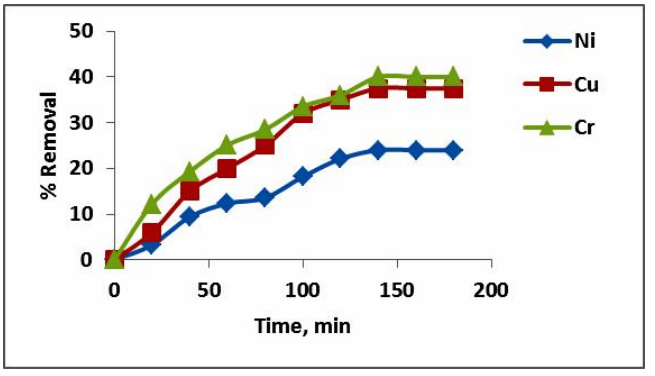

Fig. 6. Fe & Al combination of electrodes without perforations

3.2. Electrodes with Perforations

All the parameters kept same as that of iron electrodes without perforations and experiment was carried out with perforated iron electrodes. The current density was 0.385 mA/cm^2. The heavy metals (Ni, Cu, Cr) concentration was decreased gradually and become stable at 160 minutes. The maximum removal percentage of Ni, Cu, Cr was 77.0 %, 73.0 %, 44.9 %, respectively, as shown in Fig. 7. The reduction in the concentration of metal ions was almost doubled with perforations electrodes than without perforations. Perforations provide more surface for metal ions to get in contact with electrodes. The experiment was further carried out using perforated aluminum electrode by keeping same conditions as without perforations.

The current density was 6.537 mA/cm^2. Fig. 8 shows the removal percentage of metal ions. The reduction in Ni, Cu, Cr was 17.3 %, 43.0 %, 13.6 %, respectively. The removal percentage of Ni, Cu, Cr for a combination of electrodes with perforations was 40.3 %, 68.0 %, 89.7 %, respectively, as shown in Fig. 9. The performance of electrodes with perforations was better than without perforations at same experimental conditions.

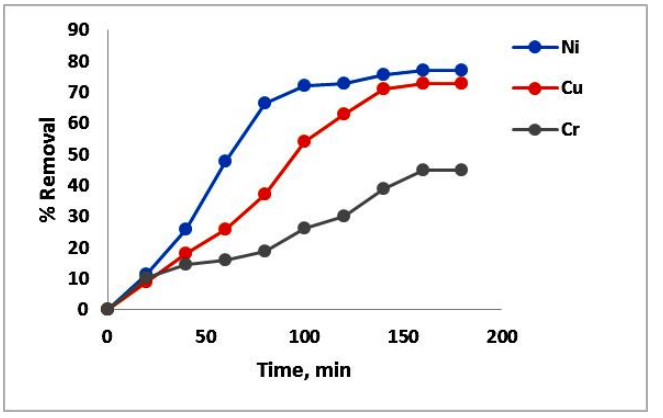

Fig. 7. Iron electrodes with perforations

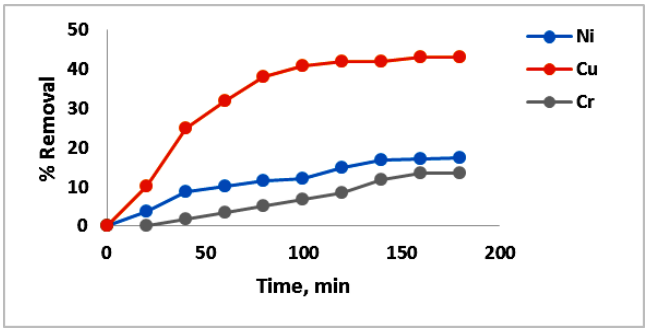

Fig. 8. Aluminum electrodes with perforations

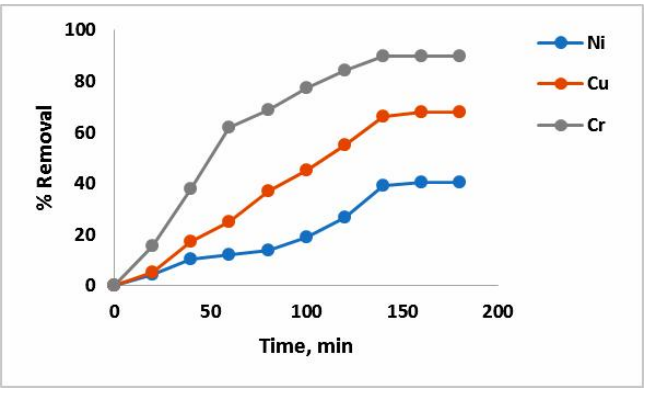

Fig. 9. Fe & Al combination of electrodes without perforations

Keeping all the parameters same, the effect of current variations on individual electrodes was also investigated. Table 1 shows the maximum removal percentage of metals with variations in current.

Table 1. Removal percentage of metals ions at different currents

Metal Ion	Aluminum Electrode		Iron Electrode		
	0.5A	1.0A	0.05A	0.1A	0.2A
Ni	17.98 %	12.14 %	76.42 %	78.57 %	90.71 %
Cu	46.46 %	55.00 %	83.00 %	84.00 %	86.00 %
Cr	82.75 %	87.93 %	46.42 %	48.21 %	55.93 %

3.3. Effect of pH

The effect of pH was studied in combination of electrodes. Experimental runs were conducted at three initial pH conditions 3.5, 4.0 and 4.5. 0.1 N HCl solution was used to maintain the required pH. All the other parameters were kept same as earlier experimental runs with the combination of perforated electrodes. At pH 3.5, the removal percentage of Ni, Cu, Cr was 67.7 %, 73.0 %, 90.0 % respectively as shown in Fig. 10. Figs. 11 and 12 show the removal percentage of metal ions at pH 4 and 4.5, respectively. At pH 4, the removal percentage of Ni, Cu, Cr was 36.7 %, 79.0 %, 93.1 %, respectively, and at pH 4.5 it was 19.4 %, 84.7 %, 91.9 %, respectively. It is obvious that the reaction rates increase as pH increases and hence the removal percentage efficiency enhanced. The adsorption of Cr on to $Al(OH)_3$ flocs is higher than Ni and Cu and also the formation of insoluble $Cr(OH)_3$ during the process is another factor that enhances the removal efficiency of Cr.

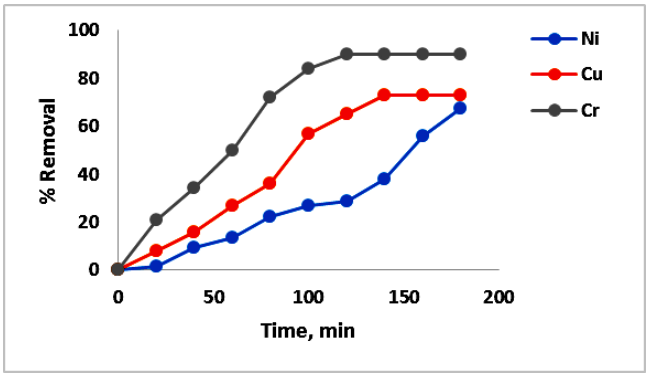

Fig. 10. pH 3.5, Fe-Al electrodes with perforations

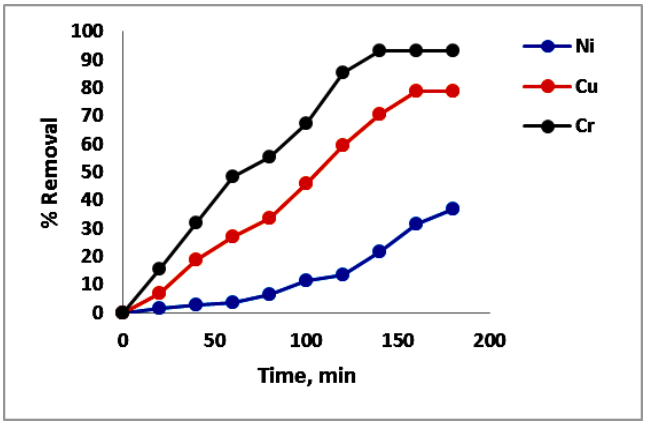

Fig. 11. pH 4, Fe-Al electrodes with perforations

3.4. Kinetic Model

The prediction of removal rate of heavy metal ions gives an important information for designing the batch electrocoagulation system. Information on the kinetics is required for selecting optimum operating conditions for the full-scale batch process. The process of removal of heavy metal ions using electrocoagulation consumes low energy. The kinetic model demonstrated that the removal of heavy metal ions follows the second order Eq. (1) with current dependent parameters.

$$\frac{1}{C(t)} = \frac{1}{C_0} + k_2 t \qquad (1)$$

where k_2 is the second order rate constant in $ppm^{-1}min^{-1}$.

Table 2 shows the rate constants and corresponding R^2 values. The experimental data was fitted to the best performed electrodes for the removal of corresponding metal ions.

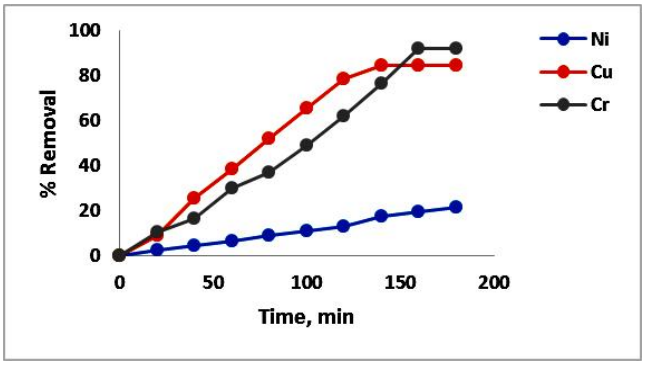

Fig. 12. pH 4.5, Fe-Al Electrodes with perforations

Table 2. Rate constants and corresponding R^2 values

Metal Ion	Rate Constant (K_2), $ppm^{-1}\ min^{-1}$ $\frac{-dc}{dt} = k_2 c^2$ $k_2 = (ppm^{-1}min^{-1})$	R^2 value
Ni^{+2}	0.021	0.924
Cu^{+2}	0.003	0.917
Cr^{+3}	0.027	0.948

4. Conclusions

The present study investigated the removal of heavy metal ions from synthetic galvanic wastewater by electrocoagulation process. The performance of Iron, Aluminum electrodes with and without perforations was evaluated. The performance efficiency of perforated electrodes was higher than without perforations. The maximum removal percentage of nickel and copper ions using perforated iron electrodes was 90.7 % and 86.0 %, respectively, and for chromium using a combination of perforated iron and aluminum electrodes it was 93.1 %. Pseudo

second order kinetic model was found to be best fit for the experimental data.

Conflict of Interest: The authors do not have any conflict of interest

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ПРОВЕДЕННЯ ПРОЦЕСУ ЕЛЕКТРОКОАГУЛЯЦІЇ З ВИКОРИСТАННЯМ ЗАЛІЗНИХ І АЛЮМІНІЄВИХ ЕЛЕКТРОДІВ З ПЕРФОРАЦІЯМИ І БЕЗ НИХ

Анотація. *Процес електрокоагуляції широко використовують для вилучення забруднюючих речовин з промислових стічних вод. У цьому дослідженні зроблено спробу дослідити проведення процесу електрокоагуляції з використанням алюмінієвих і залізних електродів для обробки іонів металів, наявних у синтетичних гальванічних стічних водах. Досліджувані електроди були з перфораціями та без них, і встановлено, що ефективність електродів з перфорацією (80 %) була вищою, ніж без перфорацій (50 %). Ефективність вилучення іонів важких металів зростала з часом утримування та постійним струмом. Оптимізовані значення часу перебування, напруги, pH, струму, відстані між електродами становили 160 хв, 6 В, 5, 0,2 А і 3 см відповідно. Максимальний відсоток вилучення іонів нікелю та міді за допомогою перфорованих залізних електродів становив 90,7 % і 86,0 % відповідно, а для хрому з використанням поєднання перфорованих залізних і алюмінієвих електродів – 93,1 %. Вилучення іонів металу відбувалося за кінетичною моделлю псевдодругого порядку з параметрами, залежними від струму.*

Ключові слова: *синтетичні гальванічні стічні води, процес електрокоагуляції, важкі метали, електроди, водопідготовка.*