EXAMINING THE EFFICIENCY OF ELECTROCHEMICAL PURIFICATION OF STORM WASTEWATER AT MACHINE-BUILDING ENTERPRISES

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1. Introduction

Contaminated surface runoff at any industrial enterprise results from rain drops that capture particles of dust, aerosols, and products of exhaust gases in the near-earth layers of the atmosphere, as well as from the surface of water catchment areas. The concentration of pollutants in wastewater, in turn, depends on many factors, which is why it is difficult to predict and control it. Among these factors are the landscaping of a water catchment area, climatic conditions, the intensity of transport motion, features of industrial processes, other factors. Duration of a rainless period also prede-
terminates the accumulation of impurities at the territory of a water catchment area. The storm wastewater that forms at the territory of industrial enterprises contains oil products, suspended substances, nitrates and ammonium salts, chlorides, microorganisms, and other various dangerous impurities including heavy metals’ ions. These contaminants very often penetrate, without purification, groundwater, and subsequently pollute surface water sites, thereby worsening environmental conditions in industrial regions.

Storm wastewater can be used as secondary resources instead of clean tap water. Collecting rain water in tanks or water catchment reservoirs with the further planned utilization makes it possible to prevent overloading a sewerage network in the case of intense rainfall. In addition, one needs to take into consideration the fact that the rain water after purification can be used at many industrial facilities, which provides for a substantial saving in payments for water resources and significantly affects the cost of production.

Stormwaters are characterized by a high saline composition that makes it possible to include them in the category of electrolytes, and, consequently, these waters ensure high efficiency of the course of electrolysis processes at lowest electricity consumption. Therefore, it is a relevant scientific task to study the quality of surface water at industrial enterprises, to define the specificity of contamination by ions of heavy metals, and to find ways for purification that would employ modern, including electrochemical, methods.

2. Literature review and problem statement

Environmental pollution, primarily drinking water aquifers, by ions of heavy metals leads not only to deterioration in the living conditions of humans, but is also very dangerous for the health of people. As a result of anthropogenic activities, heavy metals penetrate water objects, they are accumulated by hydrobionts and bottom deposits. In addition, there are seasonal fluctuations in the content of heavy metals in water, fish, oysters [1]. They penetrate human body mainly with drinking water, which could contain significant concentrations of metals that are toxic for humans. For example, a study into drinking water undertaken by authors of [2] showed that the content of Zn, Pb, Cd, Cr and Cu in the drinking water at one of the cities in Iran were at the level of 47.01 mg/l, 3.2 mg/l, 0.42 mg/l, 5.08 mg/l, and 6.79 mg/l, respectively. Heavy metals, as reported in [3], can accumulate in the human body and may cause serious diseases of the gastrointestinal tract and can even increase the risk of cancer.

The removal of heavy metals and cleaning of industrial wastewater, as reported in [4], employs the physical, chemical, biological, and electrochemical methods. An earlier research [5] indicates the low efficiency of a settling method for the removal of heavy metals’ ions from storm wastewater. It was shown that the settling of water ensures purification only in the range of 14.7–25 % at the expense of ions sorption by suspended substances and depositing along with them. Such efficiency of purification does not meet the standards of water discharge. In addition, settling takes a long time, and, therefore, settlement structures of significant size. It is possible to intensify the process of sedimentation of suspended particles from polydisperse wastewater by the aggregation of contaminants when applying chemical coagulants and flocculants [6]. However, the use of chemical coagulants has its own special features and requires the consumption of reagents, as well as additional studies, for example, into the strength of the formed aggregates [7].

Over recent years, much research has addressed the electrocoagulation method of wastewater purification from heavy metals, which is considered one of the most promising and highly efficient [8]. Removal of dissolved substances from wastewater in the process of electrocoagulation is predetermined by the capability of Al$^{3+}$ and Fe$^{3+}$ cations to form complex compounds. The aggregates that are formed in the interaction between particles and ions of metals from soluble electrodes, or products of their hydrolysis, float with bubbles of gas formed on electrodes.

The authors of [9] investigated the processes of wastewater electrocoagulation using electrodes based on iron and aluminum, and established a possibility for almost complete (close to 100 %) removal of ions of copper, chromium and nickel over 20 minutes. According to their data, the current density was 10 mA/cm$^2$ and pH of the medium was 3.0. Consumption of energy and an electrode material was found to be 10.07 kW/m$^3$ and 1.08 kg/m$^3$, respectively. However, the authors did not examine other heavy metals, nor did they define the factors that affect the consumption of energy or the efficiency of purification.

Paper [10] investigated purification of water from copper, nickel, zinc, and manganese in an electro-coagulator of unipolar configuration. The authors established regularities in the influence of current density in the range of 2–25 mA/cm$^2$ and acidity of the medium on the efficiency of metal removal. The efficiency of cleaning from copper, nickel, zinc was 96 %, at current density 25 mA/cm$^2$ and power consumption 49 kW/m$^3$. However, the unresolved issue has been the low efficiency of purification from manganese, at the level of 72 %, which does not make it possible to consider the method suggested by the authors to be universal.

The authors of [11] examined electrocoagulation treatment from the Cu$^{2+}$, Cr$^{3+}$, Ni$^{2+}$ and Zn$^{2+}$, and managed to obtain efficiency above 97 % at current density 4 mA/cm$^2$, pH index of medium acidity 9.56, and electrocoagulation duration 45 min. However, such operational modes of equipment have a rather high power consumption, at the level of 6.25 kW/m$^3$, and consume 1.31 kg/m$^3$ of electrodes’ material, respectively.

The authors of article [12] pointed to the high efficiency of cleaning from ions of zinc and copper (at the level of 100 % at initial concentrations of 5 mg/l), as well as manganese (89 %) at current 0.1 A, pH=6, duration of water treatment 90 min. and consumption of electricity 2.55 kW/m$^3$. Paper [13] considered various approaches to the process of electrocoagulation when cleaning industrial, urban and agricultural wastewaters. However, publications [11, 12] lack the patterns of purification effectiveness predetermined by the composition of wastewater or other factors that affect the residual concentration of contaminants. The authors of [13] emphasize that the field of electrocoagulation has too many unresolved questions concerning the mechanisms and kinetics of the process, as well as there are no systemic models that would make it possible to calculate appropriate equipment.

Selection of optimum modes for the purification of wastewater using a method of electrocoagulation is defined by the efficiency of removal of impurities from water, that is, the residual concentration of contaminants in a solution upon completion of the purification process. The main indicator for wastewater treatment when applying a method of electrocoagulation is the purification effectiveness or the residual concentration of pollutants’ particles.
At present, there is no a unified approach to the optimization of parameters for electrocoagulation wastewater treatment. The scientific literature provided neither models nor guidelines that would have determined the technological parameters for operation of purification equipment, as there are significant differences between existing models and an actual pattern at a particular industrial site. In addition, we did not detect any dependences for each heavy metal, which does not make it possible to recommend conditions for treatment in each specific case. Under practical conditions, it is necessary to determine for each enterprise, depending on the composition and specificity in the formation of wastewater, the properties of each parameter of run-off, which is not always possible. Thus, the only reliable source of information about patterns in the electrocoagulation treatment of a particular, actual wastewater discharge from the territory of a specific enterprise is the experiment.

3. The aim and objectives of the study

The aim of the study that we conducted was to determine patterns in the removal of heavy metals' ions by using a method of electrocoagulation from the surface runoff from the territory of a machine-building enterprise.

To accomplish the aim, the following tasks have been set:

- to explore the composition of pollutants of wastewater from the territory of a machine-building enterprise;
- to define the factors that affect the removal of ions of metals during electrocoagulation treatment, as well as dependences of efficiency of wastewater purification on these factors;
- to investigate the dependence of purification efficiency on current density and the duration of settling following the electrocoagulation treatment of a contaminated runoff for each metal.

4. Materials and methods to study the purification of storm wastewater by the method of electrocoagulation

4.1. Methods of sampling and analysis of samples of storm wastewater

We collected samples during rains at the territory of one of the enterprises of machine-building complex, located in the city of Kharkiv (Ukraine). For our study, we cleaned and disconnected rain receivers from the system of storm wastewater discharge by plugging a water pipe. The sampling was conducted simultaneously at all points of water catchment areas after intensive agitation, from a filled rain receiver. The samples were taken at the roofs of facilities, by gathering and averaging the rain runoff coming from the drain pipes. That allowed us to obtain a complete picture of pollution throughout the entire water catchment area. The sampling was carried out in the mouth area by installing 10-liter containers with funnels. The samples were taken in increments of 10 minutes after intensive agitation.

Following the sampling of drain, part of the water was separated for measuring the content of suspended particles and settling. The kinetics of settling were determined under laboratory conditions in glass cylinders with a capacity of 1,000 cm³. The cylinders were filled with the averaged surface runoff. Next, by using a siphon, gradually immersed into the liquid, we took the samples every 20 minutes. Next, at a temperature of 18–20 °C, we determined by a weight method the concentration of suspended substances before and after settling.

The remaining samples were used for the further research.

4.2. Procedure for studying the electrocoagulation method for cleaning the surface runoff from heavy metals

We studied the electrocoagulation method of cleaning the surface runoff at a laboratory installation whose diagram is shown in Fig. 1. According to the diagram, the wastewater was first poured into receiving capacity 1, from which it was fed at the assigned water flow rate (volumetric velocity) to electro-coagulator 2. The water was treated in the coagulator with a coagulant, which was electrically generated as a result of the dissolution of an electrode's material, as well as by electrolysis gases. The foam that stood at the surface of the water was collected into tank 3. The magnitude of electric current at the electrodes of the electro-coagulator, assigned experimentally, was maintained by using a laboratory rectifier of electric current 5. The purified water was collected in tank 4, where it was exposed to settling, and from which, in certain time intervals, we took samples for analysis.

![Fig. 1. Diagram of experimental installation for the purification of storm wastewater using a method of electrocoagulation](image-url)

Concentration of heavy metals in the samples taken from containers 1 (prior to purification) and 4 (after settling) was determined using a method of atomic-absorption spectrophotometry.

We measured the content of heavy metals in all experiments at least 6 times, the experimental data were then processed by the method of least squares.

4.3. Procedure for treating the results of experiment

We acquired experimental data immediately after treating the water in the electro-coagulator. A chosen criterion for the estimation (response) of purification was the residual magnitude of impurities, that is, the concentration of ions of heavy metals upon purification.

For the regression analysis, we set the task to find a functional dependence of the mathematical expectation of response $M(Y)$ on values for the assigned factors $X_i$: $M(Y)=\gamma(X_1, X_2, ..., X_n)$. 

Mathematical processing of experimental results employed the software packages MS Excel and Statistica, designed for statistical processing of data from experimental research, for establishing the interdependences between factors of influence and response, for mapping them graphically and for constructing, based on this, regression models for the assigned parameters.

5. Results of research into purification of storm wastewater

5.1. Analysis of composition of surface wastewater from a machine-building enterprise

The results of sampling and analysis of the content of salts of heavy metals in the storm wastewater from an industrial enterprise have established the qualitative and quantitative composition of pollutants at a water catchment area (Table 1). The most contaminated, in terms of ions of copper (0.6–1.1 mg/dm³), zinc (0.38–2.0 mg/dm³), nickel (0.4–1.6 mg/dm³), and chromium (0.25–0.93 mg/dm³), were the storm wastewater from the territory of industrial workshops (etching, plating, machining). This is explained by the fact that the metal processing operations are the source of heavy metals’ dust, which propagates over the territory of an enterprise.

Ions of lead prevailed in wastewater from automobile roads and parking lots (0.35–5.0 mg/dm³), as well as from the territories adjacent to the warehouses of fuel and lubricant materials (0.4–2.15 mg/dm³).

The highest concentrations of suspended solids (Table 1) were found in the wastewater from the territory of automobile roads and parking lots, to 2,300 mg/dm³, from the area in front of warehouses, to 1,400 mg/dm³, and near the territory of shops, to 3,200 mg/dm³.

Data on the kinetics of settling showed that up to 80% of suspended solids settle within three hours. This indicates that the surface wastewater at a machine-building plant contains a significant number of finely-disperse impurities. The result of our study was the established fractional composition of suspended substances in wastewater (Table 2). Density of the solid mechanical impurities contained in the surface runoff at a machine-building enterprise is within 2.4–2.8 mg/dm³.

### Table 1

| Type of harmful substance       | Territories adjacent to shops | Automobile roads and parking lots | Warehouse of fuel and lubricant materials | Warehouse lot and other areas |
|--------------------------------|-------------------------------|-----------------------------------|------------------------------------------|-------------------------------|
|                                | Mechanical                    | Electroplating                    |                                          |                               |
| Za                             | 0.3–0.7                       | 0.38–2.0                         | 0.2–2.0                                  | 0.2–0.6                       | 0.01–0.015                   |
| Ni                             | 0.1–0.7                       | 0.4–1.6                          | 0.13–0.4                                 | 0.2–0.42                      | 0.01–0.1                    |
| Cr<sup>+</sup>                  | 0.1–0.5                       | 0.25–0.93                        | 0.1–0.3                                  | 0.15–0.3                      | 0.01–0.02                   |
| Pb                             | 0.01–0.4                      | 0.01–0.4                         | 0.35–5.0                                 | 0.4–2.15                      | 0.01–0.35                  |
| Cu                             | 0.1–0.5                       | 0.6–1.1                          | 0.07–0.27                                | 0.06–0.3                      | 0.0–0.05                   |
| Suspended substances           | 320–2,800                     | 400–3,200                        | 300–2,300                                | 260–1,400                     | 10–1,560                   |
| Petroleum products             | 3.1–24.9                      | 3.1–24.9                         | 28.1–168.3                               | 40.34–320.1                   | 3.1–24.9                   |

High concentrations of petroleum products are characteristic of the storm wastewater from the fuel and lubricant materials warehouse, to 40–320 mg/dm³, and the territory of automobile roads and parking lots, to 28–168 mg/dm³.

The data obtained are required when choosing and calculating facilities and technological schemes for the purification of wastewater at machine-building enterprises; they testify to the need for the preliminary settling of suspended particles before cleaning water from the ions of heavy metals.

5.2. Studying the effectiveness of electrocoagulation method for removing heavy metals

Experimental study into purification of surface wastewater at a machine-building enterprise was conducted in line with the scheme: settling – electrocoagulation – settling. The results that were obtained at a laboratory installation (Fig. 1), presented in graphs (Fig. 2), indicate the high efficiency of the developed technology for purification of stormwater runoff. The residual content of major pollutants (heavy metals’ ions) after electrocoagulation and settling over a certain time is at a level that meets the requirements for discharge to a water body.

The data shown in Fig. 2 also indicate that the residual concentration depends on the duration of settling and the current density of electrocoagulation. For example, increasing the duration of settling can increase the efficiency of purification at lower current density and vice versa, increasing the current density can reduce the time required for settling. It is also interesting that for certain metals an increase in the duration of settling cannot compensate for the effect of electrocoagulation. For example, in Fig. 2, a, the concentration of nickel does not reduce below 0.2 mg/dm³ even after 15 hours of settling at current density 2 A/m².
In the course of experimental study and when deriving regression equations for approximating curves, we constructed dependences of the residual concentration of heavy metals on settling duration at values for current density in the range of 2–50 A/m³, Table 3.

The obtained dependences make it possible to determine, for example, the duration of settling after electrocoagulation in order to achieve the required degree of purification at the assigned parameters of current density. Mathematical processing of experimental results using the method of regression analysis in the Statistica software made it possible to derive the following generalizing equations for dependence of the concentration of pollutants \( y \) on settling duration \( x \) and current density \( t \):

\[
Y_{Ni} = 0.4761 - 0.049x - 0.0193t + 0.0023x^2, \quad (1)
\]

\[
Y_{Zn} = 0.3556 - 0.0507x - 0.0027t + 0.0014x^2, \quad (2)
\]

\[
Y_{Cu} = 0.2899 - 0.0381x - 0.0027t + 0.0014x^2, \quad (3)
\]

\[
Y_{Pb} = 0.6059 - 0.0954x - 0.0089t + 0.0041x^2, \quad (4)
\]

\[
Y_{Cr^{3+}} = 0.2917 - 0.0514x + 0.003t + 0.0022x^2. \quad (5)
\]

Fig. 2. Dependence of residual concentrations of ions of heavy metals (mg/dm³) on density of electric current (A/m³) and settling duration after electrocoagulation (hours): \( a \) – nickel, \( b \) – copper, \( c \) – zinc, \( d \) – lead, \( e \) – chromium
By conducting a comparative analysis of values for the coefficients of the derived models (1) to (5), we can conclude that it is the settling duration after electrocoagulation that has the greatest impact on reducing the residual concentrations of salts of heavy metals in storm wastewaters from an industrial enterprise (approximately 1.4–3 times larger than the magnitude of current). The constructed dependencies make it possible to define specific conditions for the purification of surface runoff from heavy metals at a machine-building enterprise. Depending on strength of the current, duration of settling after electrocoagulation or wastewater flow rate, it is possible to predict efficiency of purification and to select the required technological parameters in order to achieve the desired efficiency.

The merits of this research are in that the experiments into wastewater purification using a method of electrocoagulation were carried out not on model, but actual, wastewater, collected at the territory of one of the enterprises of a machine-building complex. Therefore, the proposed mathematical models in the form of regression equations (1) to (5) take into consideration the most important factors, and the results of the calculation of the process of removal of metals’ ions based on them will be very close to actual conditions. The equations derived are of important practical significance because the examined parameters, duration of settling and amperage during electrochemical treatment, are decisive, required to control effectiveness of purification.

The main drawback of this study is the limited number of pollutants that we explored. However, this opens up avenues for further research in order to determine the effectiveness of electrocoagulation when cleaning from other metals and contaminants.

### 6. Discussion of results of studying the electrochemical removal of heavy metals from surface runoff

An analysis of experimental data from the examined samples of surface runoff has revealed (Table 1) that the range of change in concentrations, even within an enterprise, is quite wide and exceeds the permissible standards for discharge to water bodies. Among the pollutants, of especial environmental hazard is the heavy metals that are distributed non-evenly at the territory of a machine-building enterprise with clusters near production facilities. The suspended particles that are washed away by surface runoff contain above 45 % of the fraction the size less than 50 µm, which poorly settles and affects the choice of a method for water purification.

As shown by the results of laboratory experimental studies (Fig. 2), the effectiveness of cleaning surface wastewater from ions of heavy metals depends mainly on the following parameters:

- duration of settling wastewater after electrocoagulation;
- current density during electrolysis.

In addition, the impact of current density is significant only when cleaning from nickel, and increasing the current density almost does not affect purification from other heavy metals. Comparative analysis of the results of research into reduction of the concentration of salts of heavy metals, given in charts in Fig. 2, indicates that at current density 50 A/m³ the optimal settling duration to remove nickel and lead is 9 hours, for copper and zinc 12 hours, and it is possible to reduce the concentration of chromium to safe concentrations at current density 10 A/m³ and settling duration 4 hours. These conditions are considered optimal as they reduce the level of contamination of stormwater runoff to the norms for discharge to water reservoirs at reasonable time and at acceptable consumption of electricity.

![Approximating curves](image-url)

| Type of pollutant | Current density, A/m³ | Regression equations for approximating curves | R²  |
|-------------------|-----------------------|---------------------------------------------|------|
| Ni                | 2                     | \[y = 0.0049x^2 - 0.0723x + 0.4671\]          | 0.9975 |
|                   | 10                    | \[y = 0.0177x^2 - 0.2035x + 0.58\]            | 0.989 |
|                   | 50                    | \[y = 0.0133x^2 - 0.1688x + 0.5543\]          | 0.9975 |
| Zn                | 2                     | \[y = 0.0123x^2 - 0.1527x + 0.5\]             | 0.9924 |
|                   | 10                    | \[y = 0.0088x^2 - 0.1219x + 0.4786\]          | 0.9955 |
|                   | 50                    | \[y = 0.0145x^2 - 0.1712x + 0.51\]            | 0.9851 |
| Cu                | 2                     | \[y = 0.0031x^2 - 0.069x + 0.35\]             | 0.9919 |
|                   | 10                    | \[y = 0.0069x^2 - 0.1047x + 0.3994\]          | 0.9837 |
|                   | 50                    | \[y = 0.0105x^2 - 0.1285x + 0.3947\]          | 0.9796 |
| Pb                | 2                     | \[y = 0.024x^2 - 0.2914x + 0.8797\]           | 0.9973 |
|                   | 10                    | \[y = 0.0143x^2 - 0.2142x + 0.8207\]          | 0.9999 |
|                   | 50                    | \[y = 0.0267x^2 - 0.3088x + 0.8763\]          | 0.9879 |
| Cr³⁺              | 2                     | \[y = 0.0069x^2 - 0.1047x + 0.3994\]          | 0.9837 |
|                   | 10                    | \[y = 0.0031x^2 - 0.069x + 0.35\]             | 0.9919 |
|                   | 50                    | \[y = 0.0105x^2 - 0.1285x + 0.3947\]          | 0.9796 |

### 7. Conclusions

1. It was established that the territory of a machine-building enterprise is polluted by different pollutants unevenly; the contaminants of surface wastewaters from the area adjacent to production shops are dominated by ions of copper, to 1.1 mg/dm³, zinc, to 2.0 mg/dm³, nickel, to 1.6 mg dm³, chromium, 0.93 mg/dm³, and lead, to 5.0 mg/dm³. In addition, high concentrations of petroleum products are characteristic of storm runoff from warehouses with fuel and lubricants, to 320 mg/dm³, and of the territory of automobile roads and parking lots, to 168 mg/dm³. Wastewater from any plot is also contaminated with finely-disperse (about 45 % of the fraction the size less than 50 µm) suspended substances at concentrations up to 3,200 mg/dm³.

2. It was established that the removal of ions of metals during electrocoagulation purification is significantly affected by the duration of wastewater settling after electrocoagulation and current density during electrolysis. It was found that the conditions for deposition of each metal differ both in terms of current density and settling duration. For the removal of nickel and lead, the optimum current density is 50 A/m³ and settling duration of 9 hours. To remove copper and zinc, the duration of settling must be 12 hours. It is possible to reduce the concentration of chromium to safe concentrations at current density 10 A/m³ over settling duration of only 4 hours.
3. We have constructed mathematical dependences of the process, which include the dependence of response (residual concentration) on settling duration and current density during electrocoagulation treatment. It was established that each ion has its own removal dependence and it is possible to define the optimal parameters for the course of the process – settling duration and current power. An increase in current during purification and settling duration significantly increases the efficiency of purification from the ions of metals, with almost 1.4–3 times higher settling efficiency than that at an increase in current density.

Such data make it possible to control efficiency of purification depending on fluctuations in the concentration of pollution: by increasing current density or by prolonging the duration of settling it is possible to improve purification effectiveness to the normative value.

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