# Electrochemical Purification of Oil-Containing Shipping Waters

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**ABSTRACT**

The article describes the modern problems of formation and purification of marine oil-containing waters. The efficiency of using electrocoagulation to remove oil from water-oil emulsions of different mineralization using aluminum and iron anodes was studied. Treatment of water-oil solutions with an oil content of 100 mg/dm$^3$ by electrocoagulation in a single-chamber electrolyzer provides 98–99% oil removal using these electrodes at an anode current density of 0.57–2.11 A/dm$^2$ for highly mineralized waters and 0.34 A/dm$^2$ for freshwater treatment during the first 15 minutes provides a reduction in oil concentration from 100 mg/dm$^3$ to values at the level of 1.55–2.93 mg/dm$^3$. When the water treatment time is extended to 45 minutes, greater efficiency in highly mineralized waters is provided by the aluminum anode.

**Keywords:** oil-containing waters, bilge water, electrocoagulation, ship wastewater, electrolysis, aluminum anode, iron anode.

## INTRODUCTION

For many years, world shipping has been almost the only high-tonnage carrier. The rapid development of shipping requires the creation of a modern fleet with powerful equipment, large carrying capacity, high speed, accurate automated control systems for shipping equipment. On the other hand, modernity requires the reduction of energy and material consumption, as well as the minimization of harmful discharges during the operation of ships [Nugroho et al., 2019]. According to statistics, shipping is the cause of approximately 45% of oil pollution in the world's oceans. For comparison, it can be said that the pollution of the world ocean from oil production on the shelf is only 1% in accident-free mode [Dembovych et al., 2013]. The analysis of monitoring observations indicates that the largest part of the pollution of the hydrosphere by oil and oil products is associated with the discharge of washing, ballast and bilge waters.

Considering the risks involved in the pollution of the hydrosphere by oil and oil products, certain restrictions are constantly introduced by the International Maritime Organization. Thus, the International Convention for the Prevention of Pollution from Ships of 1973, as amended by the Protocol of 1978 (MARPOL Convention 73/78), allows the discharge in international waters of purified drinking water with a concentration of petroleum products of no more than 15 ppm (15 mg/dm$^3$) [International convention for the prevention of pollution from ships, 1973]. In the future, it is predicted that the legislation will be more stringent and the permissible content of oil products in the water discharged into the sea will be reduced to 5 ppm.

The bilge waters are understood as the entire set of waters of the ship's sewage wells, which are in the lower part of the ship. The composition of liquid water is diverse: fuel, lubricants, hydraulic fluids, antifreeze, solvents, and detergents. The form of finding oil and oil products in the wastewater from ships is also characterized

**Table 1.** The form of finding oil and oil products in the wastewater from ships

| Oil form | Floating | Dispersed | Emulsified | Dissolved |
|---|---|---|---|---|
| Particle size, μm | > 100 | 10–100 | 0.1–10 | <0.1 |

by diversity (Table 1). The average daily accumulation of oil-containing waters depends mainly on the power of the main engine [Nugroho et al., 2019] and ranges from 5 to 30 $m^3$. The content of oil products in drinking water is on average 2000–3000 ppm (2–3 $g/dm^3$).

Due to the wide range and fluidity of oil concentrations in ship wastewater, as well as due to the many morphological forms of oil in wastewater, many methods are proposed for their treatment. The main methods of cleaning oil-containing waters are physical, chemical, biological methods in various ways of their implementation [Mysore et al., 2006]. It is often suggested to use combined methods of cleaning such waters. Each of the proposed methods has its advantages and disadvantages.

In particular, the separation method is characterized by the low cost of the process, ease of maintenance; however, the limitation of the separation of fractions is noted. A special problem is caused by emulsified oil [Yong Li et al., 2017], which is the dominant form of oil in oil-bearing waters. The methods of coagulation water purification are characterized by low cost and ease of implementation, but the efficiency of purification from surface-active substances is low [Yong Li et al., 2017]. Filtering methods are also characterized by the low cost of equipment, but their application poses the problem of the formation of washing water and its disposal [Rana et al., 2016].

The advantage of adsorption methods for cleaning oil-containing waters is high cleaning efficiency and a small required area for equipment, although the equipment itself is expensive, and, in addition, these methods are characterized by the difficulty of regeneration of spent adsorbents [Kratochvil et al., 2017].

As for biological methods of water purification from oil and oil products, these methods (microbial metabolism [Ma et al., 2015], aerobic water purification using activated sludge [Uma et al., 2018], biofilters [Vyrides et al., 2018]) are characterized by the absence of the use of chemical auxiliary substances and low cost of the process, but there are disadvantages, namely low efficiency of water purification and the difficulty of purifying its large volumes.

Disadvantages of electrochemical methods include the high cost of materials and high energy consumption. However, according to the results of a comparative assessment of the energy efficiency of oil-bearing water treatment methods [Gorbov et al., 2013], the energy costs for cleaning these waters do not exceed 10% of the power of the auxiliary diesel generator, or 3.3% of the total installed power of the ship's power plant. When modernizing ships in accordance with legal requirements, cleaning equipment can be installed without increasing the electrical power of diesel generators. The advantages of electrochemical methods of cleaning oil-containing waters are precisely the decisive parameters – a high degree of water purification, a short processing time, and small equipment dimensions [Ulucan et al., 2015].

Electrocoagulation [Aswathy et al., 2016; Karhu et al., 2012] and electroflotation [Cerqueira et al., 2014; Nonato et al., 2017] are distinguished among the electrochemical methods of water purification from petroleum products. During electrocoagulation under the action of a constant electric current, anodes are dissolved with the transition of metal ions into the solution and their subsequent hydrolysis. Oil particles are attached to insoluble flakes, which ultimately leads to the purification of water from oil products. Aluminum [Gobbi et al., 2018; El-Ashtoukhy et al., 2014], iron [Aswathy et al., 2016; Cerqueira et al., 2014] and other metal plates, cylinders, etc. are used as soluble anodes.

Electrochemical methods of water purification from oil and oil products are effective and easy to implement, so they are promising. Accordingly, in order to ensure high efficiency of water purification and prevent excessive consumption of electrode metals as well as their ingress into the environment in ionic form, it is advisable to continue research on the influence of operational parameters on the process. It is also required to pay attention to the characteristics of the source polluted water, in particular, its mineralization, as this parameter significantly affects the degree of water purification.

The purpose of this work was to evaluate the effectiveness of electrocoagulation water purification from oil using aluminum and iron electrodes.

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

1. To determine the efficiency of electrocoagulation extraction of oil from water using aluminum electrodes, depending on the initial water mineralization and anodic current density.
2. To establish the dependence of the efficiency of water purification from oil by the electrochemical method using iron anodes on the amount of dissolved substances in water and the anode current density.
3. To assess the possibilities of using iron and aluminum anodes for cleaning oil-containing waters.

## MATERIALS AND METHODS

In this work, a single-chamber electrolyzer was used, the volume of which was 300 cm$^3$. The area of the cathode was equal to the area of the anode $S_A = S_K = 0.26$ dm$^3$. The distance between the plates was 0.07 m. A metal plate made of 12X18N10T alloy steel was used as the cathode. Aluminum or iron plates were used as anodes. A direct current source was used for electrolysis. The work used water-oil model solutions that simulated fresh and sea water: 1 – 100 mg/dm$^3$ of oil, 200 mg/dm$^3$ of sodium chloride; 2 – 100 mg/dm$^3$ of oil, 30 g/dm$^3$ of sodium chloride.

Electrolysis was carried out at an anodic current density from 0.34 A/dm$^2$ (0.09 A) to 2.11 A/dm$^2$ (0.55 A), determining with a constant time interval (15 minutes) in filtered water:

- residual oil concentration by spectrophotometric method;
- chloride concentration by argentometric titration;
- by the potentiometric method of pH of the medium.

The degree of purification of water from oil (Z, %) was calculated according to the formula:

$$Z = \left(1 - \frac{C}{C_0}\right) \cdot 100, \% \qquad (1)$$

where: $C$ – the final concentration of the component after processing, mg/dm$^3$;
$C_0$ – the initial concentration of the component, mg/dm$^3$.

## RESULTS AND DISCUSSION

Electrochemical methods of water purification are characterized by high efficiency and simplicity of process design.

At the first stage of the work, studies were conducted to determine the effectiveness of using aluminum electrodes in electrocoagulation processes.

Thus, during the treatment of oil-containing water in a single-chamber electrolyzer with an
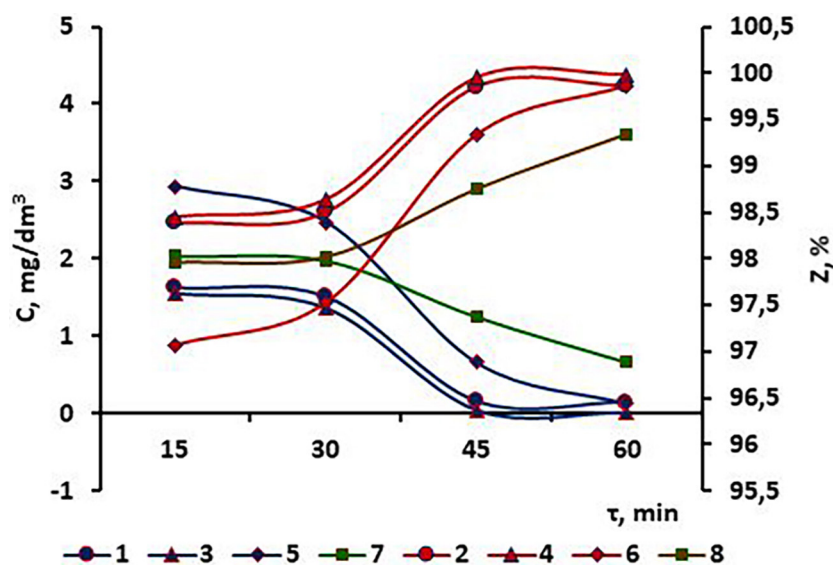


**Figure 1.** Change in the concentration of oil in water (1, 3, 5, 7) and the degree of water purification (2, 4, 6, 8) with the time of electrocoagulation of water-oil solutions (100 mg/dm$^3$ of oil, 30 g/dm$^3$ of NaCl (1– 6); 100 mg/dm$^3$ of oil, 200 mg/dm$^3$ of NaCl (7, 8)) in a single-chamber electrolyzer (V=300 cm$^3$, I=0.15 A (1, 2); 0.25 A (3, 4); 0.55 A (5, 6); 0.09 A (7, 8); j=0.57 A/dm$^2$ (1, 2); 0.96 A/dm$^2$ (3, 4); 2.11 A/dm$^2$ (5, 6); 0.34 A/dm$^2$ (7, 8)) when using an aluminum anode

aluminum anode, a high degree of oil removal was achieved (Fig. 1).

During 15 minutes of electrolysis of a model solution that simulated polluted seawater (30 g/dm³ of sodium chloride, 100 mg/dm³ of oil), at an anodic current density of 0.57 A/dm², the concentration of oil decreased from 100 mg/dm³ to 1.62 mg/dm³. The degree of water purification was 98.38%.

During further electrolysis of the solution ($\tau$=1 h), the concentration of oil in water decreased to 0.14 mg/dm³, which corresponded to a degree of oil removal of 99.86%.

With an increase in the current density to 0.96 A/dm², the efficiency of the process practically did not change compared to the current density of 0.57 A/dm², the residual concentration of oil for half an hour of electrochemical treatment was 1.36 mg/dm³ (Z=98.64%). However, when the electrocoagulation time was extended to 1 hour, the concentration of oil in water decreased to 0.02 mg/dm³ (Z=99.98%).

A further increase in the current density to 2.11 A/dm² did not lead to an improvement during the oil removal process, and during the first 45 minutes the residual oil concentrations were even higher than at low current densities, although a high degree of oil removal was ensured, at the level of 97–98%.

It should be noted that the maximum decrease in oil concentration occurred during the first 15 minutes. However, almost the entire cleaning process was completed in 45 minutes. Further electrocoagulation was impractical, as oil was not extracted.

High efficiency of the process was also observed when purifying fresh water from oil. The degree of purification was 97.96–99.34%. However, if compared with the efficiency of removing oil from seawater, it can be said that it is more efficient to remove oil from freshwater solutions. The concentration of oil in the solution decreased to 2.0–0.7 mg/dm³ during the electrolysis of the solution within an hour. It should be noted that due to the low mineralization of the solution, significant resistance was observed in the system: the current strength was 0.09 A (anodic current density 0.34 A/dm²) at a voltage of 14–16 V.

During electrocoagulation, the pH of the medium and the residual concentration of chlorides were also determined in the samples (Fig. 2).
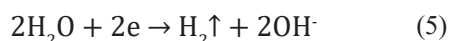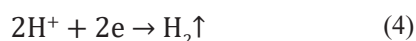
During the electrocoagulation of a highly mineralized water-oil emulsion in the first 15 minutes, the concentration of chlorides sharply decreased from 30 g/dm³ to 16.6–18.4 g/dm³ at an anodic current density in the range of 0.57–2.11 A/dm². As the process continued, there was a slow decrease in the concentration of chlorides. In weakly mineralized water, chloride concentration decreased from 200 mg/dm³ to 177–35 mg/dm³ at an anodic current density of 0.34 A/dm² within 1 hour. A decrease in the concentration of chlorides in water is explained by the formation of oxygen and/or active chlorine:
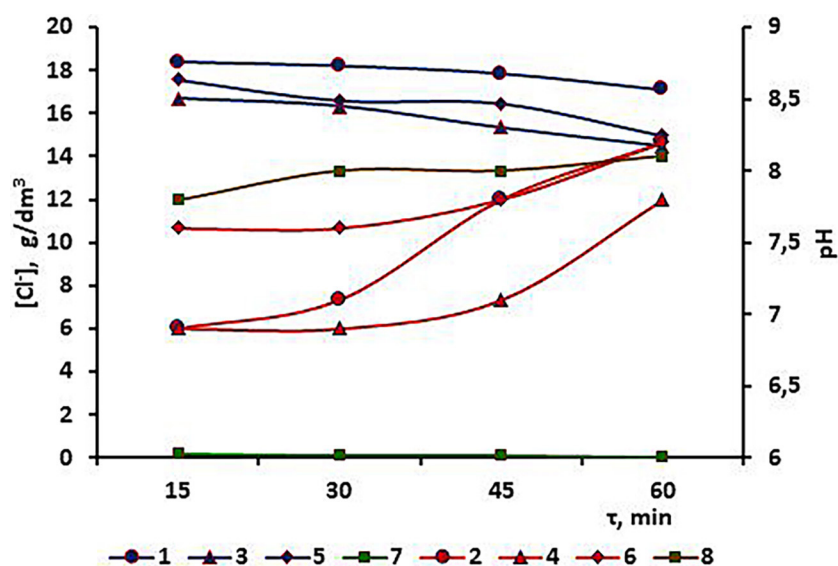


**Figure 2.** Changes in the concentration of chloride ions in water (1, 3, 5, 7) and the pH of the medium (2, 4, 6, 8) with the time of electrocoagulation of water-oil solutions (100 mg/dm³ of oil, 30 g/dm³ of NaCl (1 –6); 100 mg/dm³ of oil, 200 mg/dm³ of NaCl (7, 8)) in a single-chamber electrolyzer (V=300 cm³, I=0.15 A (1, 2); 0.25 A (3, 4 ); 0.55 A (5, 6); 0.09 A (7, 8); j=0.57 A/dm² (1, 2); 0.96 A/dm² (3, 4); 2, 11 A/dm² (5, 6); 0.34 A/dm² (7, 8)) when using an aluminum anode

$$2H_2O - 4e = O_2 + 4H^+ \qquad (2)$$

$$2Cl^- - 2e = Cl_2\uparrow \qquad (3)$$
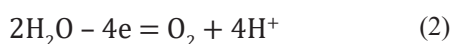
As for the pH of the medium, with the time of water treatment, it slightly increased to values of 7.8–8.2, with a less active increase in fresh water. During electrolysis at the cathode, hydrogen and hydroxide anions are formed by the following reactions:

$$2H^+ + 2e \rightarrow H_2\uparrow \qquad (4)$$

$$2H_2O + 2e \rightarrow H_2\uparrow + 2OH^- \qquad (5)$$

At the second stage of the work, research was conducted to determine the effectiveness of using iron anodes in the processes of electrocoagulation purification of water-oil emulsions.

High degree of purification was noted when using an iron anode for electrocoagulation extraction of oil from a water-oil emulsion in a single-chamber electrolyzer (Fig. 3).

During the first 15 minutes of electrolysis of a highly mineralized water-oil emulsion at an anodic current density of 0.57 A/dm², the oil concentration decreased from 100 mg/dm³ to 1.66 mg/dm³. At the same time, the degree of water purification was 98.34%.
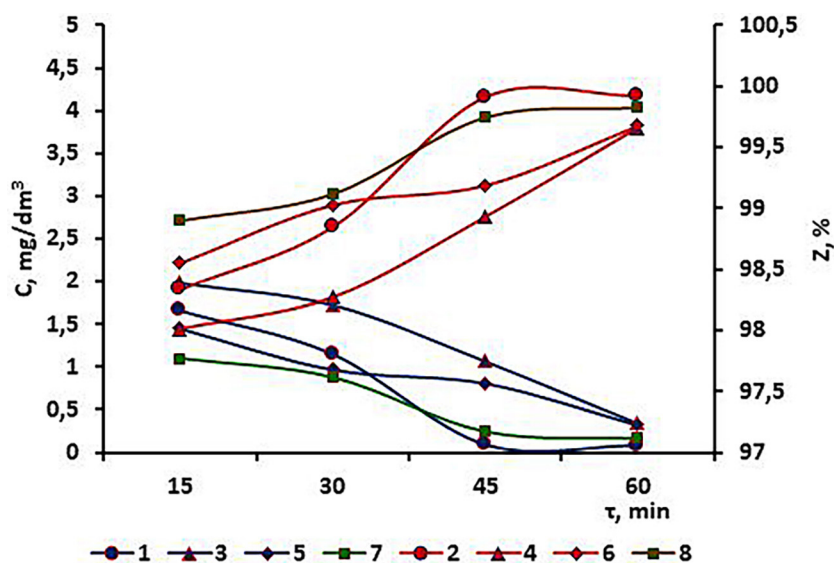
After 1 hour of electrocoagulation, the concentration of oil in water decreased to 0.08 mg/dm³, which provided a degree of oil removal of 99.92%.

A further increase in current density to 0.96 A/dm² and 2.11 A/dm² did not lead to an increase in oil removal efficiency; however, a high degree of oil removal was ensured, at the level of 98–99 %.

The maximum decrease in oil content occurred in the first 15 minutes. The water purification process was completed in 60 minutes. Only with small current densities, the removal of oil from water was completed in 45 minutes.

High efficiency of the process was also observed in the case of oil removal from fresh water. The degree of oil removal was 98.90–99.83%. Although due to the high resistance in the system, which is associated with the small content of dissolved substances in water, the anode current density was only 0.34 A/dm² (at a voltage of 15 V and a current of 0.09 A).

Regarding the change in the concentration of chlorides in water during the electrocoagulation of highly mineralized emulsions, in the first 15 minutes there was a sharp decrease in the content of chlorides from 30 g/dm³ to 15.9–15.4 g/dm³ at an anodic current density in the range of 0.57–0.96 A/dm². During the continuation of the electrolysis process, there was practically no decrease in the concentration of chlorides. Only in the case of high current density (2.11 A/dm²), in the last quarter of an hour did, the chloride concentration decrease to 8.7 g/dm³ (Fig. 4). In fresh water, the chloride content decreased from 200 mg/dm³ to 177–106 mg/dm³ at an anodic current density of 0.34 A/dm² within 1 hour.



**Figure 3.** Changes in the concentration of oil in water (1, 3, 5, 7) and the degree of water purification (2, 4, 6, 8) with the time of electrocoagulation of water-oil solutions (100 mg/dm³ of oil, 30 g/dm³ of NaCl (1– 6); 100 mg/dm³ of oil, 200 mg/dm³ of NaCl (7, 8)) in a single-chamber electrolyzer (V=300 cm³, I=0.15 A (1, 2); 0.25 A (3, 4) ; 0.55 A (5, 6); 0.09 A (7, 8); j=0.57 A/dm² (1, 2); 0.96 A/dm² (3, 4); 2.11 A/dm² (5, 6); 0.34 A/dm² (7, 8)) when using an iron anode
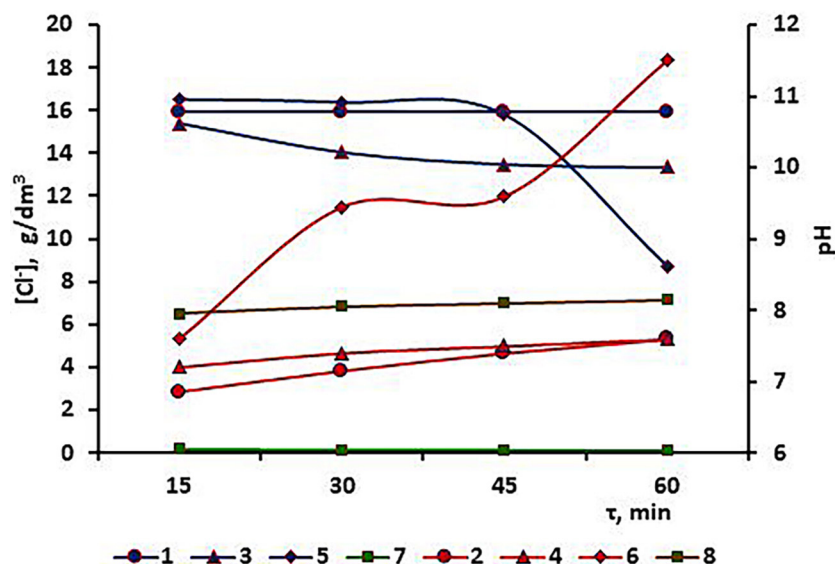
**Figure 4.** Changes in the concentration of chloride ions in water (1, 3, 5, 7) and the pH of the medium (2, 4, 6, 8) with the time of electrocoagulation of water-oil solutions (100 mg/dm³ of oil, 30 g/dm³ of NaCl (1 –6); 100 mg/dm³ of oil, 200 mg/dm³ of NaCl (7, 8)) in a single-chamber electrolyzer (V=300 cm³, I=0.15 A (1, 2); 0.25 A (3, 4 ); 0.55 A (5, 6); 0.09 A (7, 8); j=0.57 A/dm² (1, 2); 0.96 A/dm² (3, 4); 2, 11 A/dm² (5, 6); 0.34 A/dm² (7, 8)) when using an iron anode

The pH of the medium gradually increased to 7.6–8.15 with the time of electrolysis, with a slow increase in weakly mineralized water. Only at a current density of 2.11 A/dm² in the last quarter of an hour, a sharp increase in pH to 11.5 was observed.

The process of electrocoagulation takes place in several stages: the dissolution of the anode with the formation of metal cations (aluminum and iron) and the formation of their sparingly soluble compounds.

In general, the reaction of anodic oxidation of aluminum has the following form:

$$2Al - 6\bar{e} + 3H_2O \rightarrow Al_2O_3 + 6H^+ \qquad (6)$$
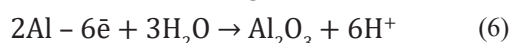
Reaction (5) is the result of a complex process, one of the stages of which is the anodic process of ionization of aluminum at the „metal-barrier oxide film" interface

$$Al - 3\bar{e} \rightarrow Al^{3+} \qquad (7)$$

The electrons released by reaction (6) enter the external circuit and form an anodizing current.

During anodization of aluminum, no noticeable release of gaseous oxygen occurs; therefore, the transition of oxygen from the electrolyte to the oxide film can be shown not as a consequence of the anodic reaction, the release of oxygen by reaction:
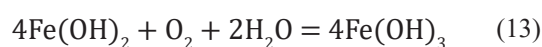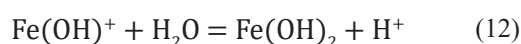
$$H_2O - 2\bar{e} \rightarrow O2 + 4H^+ \qquad (8)$$
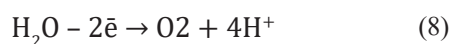
but as a direct process of oxygen ion formation

$$H_2O \rightarrow 2H^+ + O^{2-} \qquad (9)$$
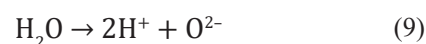
The oxygen ion, which is formed on the oxide surface by reaction (8), migrates through the barrier layer to the $Al^{3+}$ ion.

In the case of using an iron anode, the process takes place in three stages. At the first stage, the formation of $Fe^{2+}$ ions occurs, followed by their further oxidation to $Fe^{3+}$ and the formation of insoluble hydroxide.

$$Fe - 2e = Fe^{2+} \qquad (10)$$

$$Fe^{2+} + H_2O = Fe(OH)^+ + H^+ \qquad (11)$$

$$Fe(OH)^+ + H_2O = Fe(OH)_2 + H^+ \qquad (12)$$

$$4Fe(OH)_2 + O_2 + 2H_2O = 4Fe(OH)_3 \qquad (13)$$

If the efficiency of oil extraction from aqueous solutions with the use of aluminum and iron anodes is compared (Fig. 5), the following conclusions can be drawn.

In general, it should be noted that both aluminum and iron anodes ensure high efficiency of oil removal from water. At the first stages of electrochemical water treatment, a more effective application of an iron anode is observed. However, over time, in the end, the maximum reduction in oil concentration is provided by the aluminum anode. At 45 minutes of electrocoagulation, the aluminum electrode reduces the oil concentration to 0.05–0.15 mg/dm³ at anodic current densities of
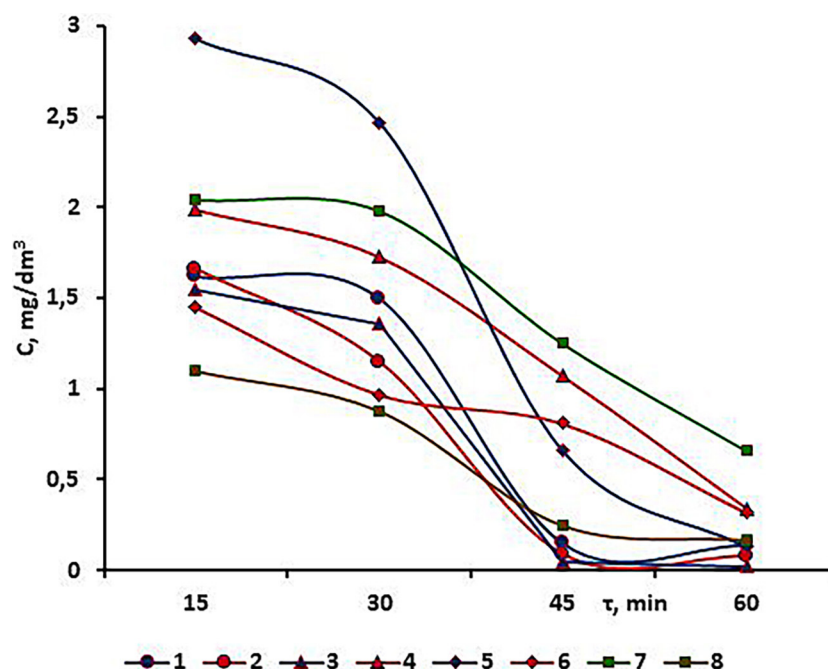
**Figure 5.** Reduction of oil concentration in water with time of electrocoagulation of water-oil solutions (100 mg/dm$^3$ oil, 30 g/dm$^3$ NaCl (1–6); 100 mg/dm$^3$ oil, 200 mg/dm$^3$ NaCl (7, 8)) in a single-chamber electrolyzers (V=300 cm$^3$, I=0.15 A (1, 2); 0.25 A (3, 4); 0.55 A (5, 6); 0.09 A (7, 8); j =0.57 A/dm$^2$ (1, 2); 0.96 A/dm$^2$ (3, 4); 2.11 A/dm$^2$ (5, 6); 0.34 A/dm$^2$ (7, 8)) at using aluminum (1, 3, 5, 7) and iron (2, 4, 6, 8) anodes

0.57 A/dm$^2$ and 0.96 A/dm$^2$. A further increase in the anodic current density to 2.11 A/dm$^2$ does not provide such an effect, which may be explained by partial passivation of the metal.

At the same time, when using an iron anode, the oil concentration decreased in 45 minutes to 1.07–0.81 at an anode current density of 0.96 A/dm$^2$ and 2.11 A/dm$^2$. The slowness of the process is most likely related to the greater number of process stages when iron anodes are used.

## CONCLUSIONS

The efficiency of using electrocoagulation to remove oil from water-oil emulsions of different mineralization using aluminum and iron anodes was studied.

Electrocoagulation treatment of water-oil solutions provides 98–99% oil removal using these electrodes at an anodic current density of 0.57–2.11 A/dm$^2$ for highly mineralized waters and 0.34 A/dm$^2$ for fresh waters.

Water treatment during the first 15 minutes provides a reduction in oil concentration from 100 mg/dm$^3$ to values at the level of 1.55–2.93 mg/dm$^3$. Greater efficiency in highly mineralized waters is provided by the aluminum anode when extending the water treatment time to 45 minutes.

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