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Research Article

The Investigation of Electrocoagulation Method for Removing Zinc and Chromium from Electroplating Industry Wastewater

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Abstract:

Electroplating wastewater generally contains high concentrations of heavy metals. This study used the electrocoagulation (EC) method with iron (Fe) electrodes to remove two heavy elements (Cr and Zn) from actual electroplating effluent at the same time. The effect of EC time and wastewater pH on removal performance was investigated. It was determined that optimum Cr and Zn removal occurred at a pH of 9 and after 30 minutes. It was discovered that the removal rates for Zn and Cr were 79% and 99%, respectively. The elimination of these heavy metal ions was compatible with a pseudo-first-order model, according to kinetic investigations. The removal of electroplating wastewater by the EC method occurs with low energy consumption, making the process economically viable and scalable. In the EC experiments using Fe electrodes, the electrode consumption was found to be 1.07 kg/m³, and the energy consumed was 25 kWh/m³.

1. Introduction

Steel surface treatment is frequently accomplished by electroplating. The steel is made smooth, glossy, and resistant to corrosion by electrochemically depositing metals such as Zn, Ni, Cr, Cu, Ag, and Au in a thin coating on the steel's surface [1]. Surface pre-treatment, electroplating, and post-treatment are the steps that make up electroplating, which produces a lot of effluent. One of the main chemical processes that produces a lot of toxic and hazardous wastewater effluents is electroplating. Electroplating effluent contains levels of heavy metals, cyanide, oil, grease, and suspended particles that are hazardous to the environment and could endanger human health [2].

Heavy metals are common contaminants found in many kinds of industrial wastewater, including that from mining and electroplating. Electroplating effluent often contains metals like zinc, copper, chromium, and nickel, which have been demonstrated to be very harmful in aquatic ecosystems [3-6]. Heavy metals are inherently highly toxic, even at very low concentrations [7]. Heavy metals are not degradable and can accumulate in human tissues, leading to the development of various chronic diseases [8]. Metals like copper, zinc, lead, and nickel are hazardous if released uncontrolled since they tend to accumulate in living things and are not biodegradable. In addition, a lot of ions of heavy metals are known to be harmful or carcinogenic. Industrial wastewater needs to be treated and closely monitored before being released because of its high toxicity [9].

Heavy metals are removed using a variety of methods. These include adsorption, precipitation, EC, ion exchange, and ion-exchange assisted membrane system [10, 11]. The most popular and regarded as the most cost-effective of these methods is precipitation. But this procedure produces a lot of settling sludge, which needs to be processed further [3, 5, 9].

Although membrane separation methods like reverse osmosis are successful at reducing metal ions, their application is restricted because to drawbacks such membrane fouling and expensive material and operating expenses [5]. Ion exchange methods provide fast and efficient removal of metals, but the pH of the wastewater can strongly influence this process. The most cost-effective techniques for removing heavy metals are known to employ a variety of adsorbents, including kaolinite, activated

carbon, and agricultural waste. Adsorbents must be renewed after usage, and their adsorption capacities differ depending on the type [5]. In addition to cleaning water for human consumption, innovative, cost-effective, and efficient methods are needed to clean industrial effluent before releasing it into other water systems. The EC technique shows promise as an alternative to the economical and effective removal of heavy metals, enabling the reuse of water in a variety of applications [5, 8, 9].

Since EC has shown effective in eliminating a variety of contaminants, it is seen as a possible substitute for chemical precipitation. These pollutants in wastewater include heavy metals, organic matter, and anions.

EC offers several advantages, such as higher pollutant removal efficiency, energy efficiency, and environmental compatibility. The effective removal of heavy metals with high efficiency using EC has been documented in several studies [11-13]; operational characteristics have a significant impact on process efficiency [10]. During the EC process, in an electrochemical process, metal cations behave as coagulants. Iron or aluminum electrodes are typically used for the anode. When a current flows through the electrodes into the wastewater, the metal anode oxidizes and releases metal cations in solution. These metal cations have the ability to aggregate contaminants by forming metal hydroxide precipitates. Oxidation of water (H_2O) produces oxygen (O_2). Other oxidation processes may also take place if the electrolyte contains anionic species. For instance, chlorine gas (Cl_2) can be produced by oxidizing chloride ions (Cl^-). Hydrogen gas (H_2) and hydroxide ions (OH^-) are created at the cathode when water (H_2O) is reduced. Several processes, including adsorption, co-precipitation, oxidation, and reduction, are used to remove contaminants during EC [5].

Compared to conventional chemical coagulation, EC has the following benefits: (i) it is simple and easy to use; (ii) it requires fewer chemicals; (iii) it produces less sludge; and (iv) it may remove heavy metals in a single step without the requirement for reduction and precipitation stages.

Water-soluble oil wastes, textile effluent, and wastewater from petroleum refineries are just a few of the contaminants that EC has been used to address [8]. In this study, zinc and chromium removal from an electroplating facility's wastewater by the EC process was investigated. The impacts of wastewater parameters, beginning pH, and treatment time on removal efficiency were assessed in order to identify the ideal operating conditions.

2. Material and Methods

2.1 Wastewater characteristics

An electroplating facility in Konya, Turkey, provided the chromium and zinc electroplating effluent used in this investigation. The wastewater utilized in the investigations was transported to the laboratory from an electroplating pool's effluent stream. Its pH was 10.2, its zinc level was 48.4 mg/L, and its chromium content was 1.2 mg/L. Lower pH runs were adjusted with 0.1 M H_2SO_4. The samples taken were stored at 4^0C during the study period. In the study, Cr and Zn removal efficiencies were examined at different pH values and different retention times.

2.2 EC reactor design and the experimental method

The EC experiments were conducted within a 600 mL glass EC reactor. The reactor was operated in batch mode. Iron (Fe) plates with edge dimensions of 9 x 9 x 0.03 cm were used as electrodes. The electrodes were positioned vertically, fully submerged in the wastewater, and spaced 7 cm apart. A power supply providing 24 volts and 1 ampere of current was used in the reactor, resulting in a current density of 12.3 mA/cm². The effects of pH and reaction duration on removal efficiency were investigated. Figure 1 displays a schematic diagram of the reactor utilized in the investigation. Samples were taken every 10 minutes during the electrolysis time (30 minutes). ICP-MS (Inductively

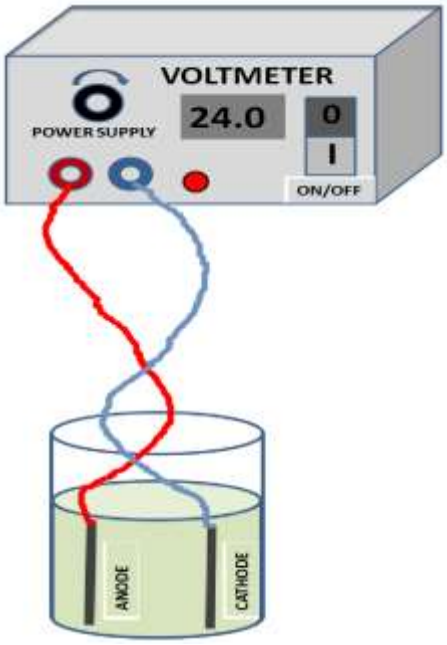

Figure. 1. *EC cell setup*

Coupled Plasma-Mass Spectrometry) was then used to examine the samples for the purpose of computing the percentage of heavy metal removed during the EC experiment and to ascertain the remaining metal content. All these experiments were carried out at room temperature. At the end of each experiment, the electrodes were rubbed and cleaned with distilled water to eliminate the oxide layer that had formed. The following formula (equation 1) was used to get the percentage removal:

$$Cr\ and\ Zn\ Removal\ Efficiency\ (\%) = \frac{C_0 - C}{C_0} x100 \quad (1)$$

For the dissolved Cr and Zn ions at time t, the initial and residual concentrations ($mg.L^{-1}$) were represented by C_0 and C, respectively.
The samples collected within the scope of the study were stored in 50 mL dark-colored glass bottles at a temperature of 4°C. pH measurements were conducted using a Hach Lange pH meter, and heavy metal analyses were performed using ICP-MS (Perkin Elmer / ICP MS ELAN DRC-E).

3. Results and Discussions

Initial concentrations, current density, initial pH, and contact time are some of the operating factors that impact the EC process [9]. This study examined the use of the EC method to remove heavy metals (Cr and Zn) from wastewater in a reactor with a volume of 600 mL, employing Fe-Fe electrodes.
In the first phase, experimental studies were conducted at the original pH of the wastewater (10.2) with different electrodes and contact times. Research was conducted at a current density of 12.3 mA/cm² with different beginning pH values during the second phase of the tests.

3.1 Effect of EC Time

Reaction time is a crucial factor in an EC process that has a big impact on energy usage [14]. The efficiency of the system was assessed at 10-minute intervals over a 30-minute period of meticulous operation. As part of the investigation, the effectiveness of heavy metal removal was examined using Fe-Fe electrodes at the wastewater's initial pH of 10.2 with waiting periods of 10, 20, and 30 minutes. The removal rates of zinc and chromium were high for the first ten minutes, but eventually they started to decline. The oxidation reactions that cause electrode corrosion have resulted in the development of oxide layers on the surface of the anode electrodes. The EC cell's efficiency is decreased as a result of these layers [15]. After 30 minutes, the highest levels of heavy metal removal

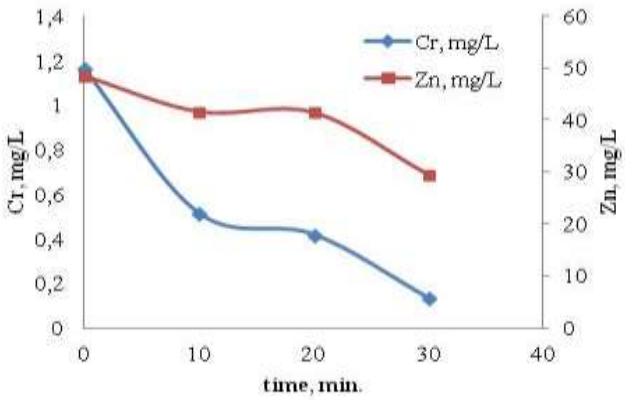

Figure. 2. *Change in heavy metal concentrations over time in the EC process using Fe-Fe electrodes. (pH: 10.2 (original pH), current density: 12.3 mA/cm², T: 25°C)*

were seen for Cr (88.6%) and Zn (39.2%). The graphical evaluation of the removal over time is displayed in figure 2.

3.2 Investigation of Heavy Metal Removal with pH Variability Using Fe-Fe Electrodes

One of the most crucial factors in EC is the initial pH. The pH of the solution significantly influences the EC performance and is also a parameter related to the conductivity of the solution and electrode dissolution. In addition, the solubility and speciation of coagulants are strongly dependent on the pH of the solution [10]. The dissolution of metallic electrodes [16] and iron ions in chemical form are extremely sensitive to the solution's pH [17]. At the 30th minute, which was identified as having the highest removal efficiency during the study time periods, changes were made to the pH, and the removal efficiency was evaluated. At this stage of the study, using Fe-Fe electrodes, the heavy metal removal efficiency was examined at pH levels of 5, 7, 9, and 10.2 (original pH) with a 30-minute waiting time. It was noted that having a pH of 5 or higher was effective in reducing metal concentrations and shortening the removal time [18]. The graphical evaluation of the pH changes during the 30-minute waiting time is displayed in figure 3. The removal efficiencies for Cr at pH 5, 7, 9, and 10.2 were found to be 99.6, 99.7, 99.0, and 88.6, respectively; for Zn, they were 68.6, 79.7, 79.0, and 39.2. Based on these results, it was determined that Cr was removed with a higher efficiency than Zn. At pH 7 and pH 9, high efficiencies were achieved, but due to being closer to the original pH value, pH 9 was chosen as the optimum pH, thus minimizing the cost associated with pH adjustment. Hydroxyl ion oxidation at the anode results in the generation of $Fe(OH)_4^-$ and $Fe(OH)_6^{3-}$ anions, which lowers the removal ability in an alkaline environment. Therefore, the removal

of heavy metal ions is reduced in higher basic medium. The cathode's conversion of protons in solution to hydrogen gas causes insufficient hydroxyl ion synthesis in an extremely acidic environment. Additionally, pH has an impact on EC performance by changing the solution's physicochemical characteristics, including electrical conductivity and the solubility of metal hydroxides. Iron (III) complex colloidal particle size is another element that has a significant reaction with heavy metal ions [19]. The elimination process involves adsorption of the molecule through electrostatic attraction and physical entrapment. Moreover, molecules can be eliminated via surface complexation or electrostatic attraction using iron's insoluble metal hydroxide. The molecule is thought to attach to the iron in surface complexation by reacting with water via a process that combines adsorption and precipitation [20].

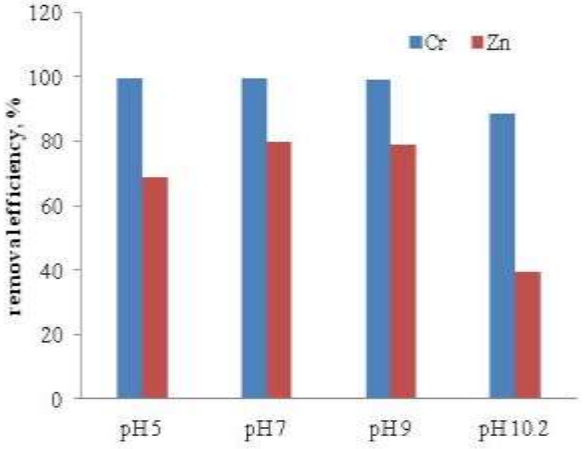

Figure. 3. *Effect of pH on the elimination of heavy metals in the EC process (t: 30 min, current density: 12.3 mA/cm², T: 25°C).*

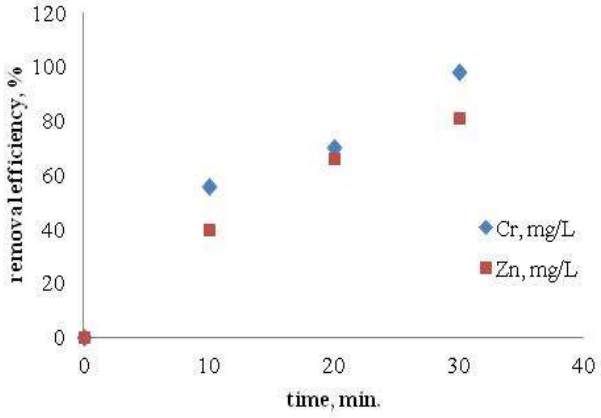

Figure 4. *Efficiency of removal achieved in the EC process using Fe-Fe electrodes (pH: 9, current density: 12.3 mA/cm², T: 25°C)*

3.3 Optimum removal conditions for the EC process

In the setup used, heavy metal removal efficiencies in wastewater were investigated at the determined optimum pH value (pH 9) during waiting times of 10, 20, and 30 minutes, using Fe-Fe electrodes. It was found that as the waiting time increased within the studied time, the removal efficiency also increased. Nevertheless, the EC process occurred in a short EC time (30 min). The heavy metal removal efficiencies are displayed in figure 4.

When the pH values are evaluated together for chromium and zinc removal, it is observed that the optimum treatment efficiency is achieved at pH 9 after 30 minutes. Studies have reported optimal heavy metal removal under similar conditions around pH 9 [5]. Adsorption frequently removes metal cations from iron hydroxide flocs' surface. Iron(III) hydroxide precipitates must occur by co-precipitation, adsorption, and precipitation processes for metal removal to be effective. OH^- concentrations, however, are insufficient to produce metal hydroxide species in acidic environments. As a result, it is challenging to generate enough OH^- ions in an acidic environment to make metal hydroxides. On the other hand, metal hydroxides can easily develop in alkaline circumstances, which leads to the efficient heavy metals elimination. However, at conditions above pH 9, the solubility of $Fe(OH)_3$ is highest at pH 6.5, so metal hydroxides may re-dissociate into metal cations [5].

3.4 Kinetic study

EC has employed several kinetic models, including first-, second-, and pseudo-second order, to remove heavy metals. Operating factors, electrode material, and contaminants can all affect the kinetic model used for EC [5, 21]. Other metal ions have the ability to chemically absorb iron hydroxide species during EC, resulting in the formation of mixed bimetallic hydroxides. Co-precipitation processes can result in the formation of metal-OH and metal-O-metal linkages [5]. The heavy metal ion removal kinetics investigation was conducted in this experiment using a fixed wastewater volume of 600 mL and a current density of 12.3 mA/cm^2. For this type of EC batch process, the mass conservation of the heavy metal ion is (equation 2):

$$-\frac{dC}{dt} = (-r_D) \qquad (2)$$

where t is the EC time in minutes and $(-r_D)$ is the heavy metal ion removal rate in mg/L.min. The removal rate equations were described using first

(equation 3), second (equation 4), and pseudo-first-order models (equation 5). When the initial concentration $C(0) = C_0$, the integration of Eq. (2) yields the following conclusion utilizing the first-order model ($-r_D = k_1 C$):

$$C(t) = C_0 e^{-k_1 t} \qquad (3)$$

where the rate constant of the first order in min^{-1} is denoted by k_1. The time-dependent concentration for the second-order model ($-r_D = k_2 C^2$) is as follows:

$$\frac{1}{C(t)} = \frac{1}{C_0} + k_2 t \qquad (4)$$

The rate constant of the second order in $L/mg.min$ is donated by k_2. Furthermore, the integration of Eq. (2) yields the following when the pseudo first-order model, $-r_D = k^{app}(C - C_e)$, is dominant:

$$C(t) = C_e + (C_0 - C_e)e^{-k^{app}t} \qquad (5)$$

where k^{app} is the rate constant of pseudo-first order in min^{-1} and C_e is the equilibrium level of concentration. The least-squares approach was applied to ascertain the optimal values for the kinetic parameters. The correctness of the kinetic model was assessed using the squared correlation

coefficient, R^2 [19]. The kinetic parameters and the R^2 values are provided in table 1.

At a current density of 12.3 mA/cm², when evaluated based on the R^2 values for all three kinetic models, it was determined that both Cr and Zn exhibited pseudo-first-order removal kinetics with values close to 1. The R^2 values calculated for the pseudo-first-order kinetic model were higher than those for the other kinetic models. Therefore, the rate at which heavy metal ions are removed can be accurately used pseudo-first-order kinetics to model. It has also been demonstrated in certain research in the literature that heavy metal elimination is described by the pseudo-first-order kinetic model [19, 20].

Three methods can be used to eliminate heavy metals: (1) EC can generate a Me(OH) precipitate; (2) Adsorption of heavy metal ions is possible by the precipitate having a substantial specific surface area; and (3) flotation of the floc can remove heavy metal ions (at negligible levels) [16].

3.5 Economic evaluation

The quantity of electrical energy utilized in the EC process is a crucial parameter for economic evaluation. The formula below was utilized to ascertain the electrical energy consumption [22]:

$$E = \frac{U.I.t}{V} \qquad (6)$$

where U is the applied voltage (V), I is the current intensity (A), t is the EC time (h), V is the treated wastewater volume (L), and E is the energy usage (kWh/m^3). Faraday's law was used to theoretically compute the amount of electrode used [3].

$$C = \frac{I.t.M}{Z.F.V} \qquad (7)$$

where I is the current intensity (A), t is the EC time (s), M is the anode's molecular weight (g/mol), Z is the chemical equivalency, F is the Faraday constant (96500 C/mol), V is the treated wastewater volume (L), and C (kg/m^3) is the electrolytic cell's Fe concentration.

For current densities of 12.3 mA/cm^2, the energy and electrode consumptions were calculated to be 25 kWh/m^3 and 1.07 kg/m^3 at 30 minutes of EC time. Gatsios, Hahladakis [23] found similar results for energy consumption in their studies. The removal efficiency, energy and electrode consumption amounts are presented in table 2 comparatively with previous studies.

4. Conclusions

The treatment of chromium and zinc containing wastewater from an electroplating plant by EC method was evaluated. It examined how factors like EC time and wastewater pH affected the removal of zinc and chromium. The findings show that EC is capable of efficiently reducing zinc and chromium. Adsorption of heavy metal ions is possible for a precipitate having a substantial specific surface area. With iron electrodes, 30 minutes was the ideal EC duration. After 30 minutes of electrocoagulation, 99% chromium and 79% zinc were removed by EC treatment using a Fe-Fe electrode pair at a current density of 12.3 mA/cm² at pH 9. At pH 9, the best rate of chromium and zinc elimination was achieved. The elimination of Zn and Cr was shown to follow a pseudo-first-order model in the kinetic investigation. The energy consumption and electrode usage in this investigation were found to be 25 kWh/m³ and 1.07 kg/m³, respectively. Due to its low energy consumption, the EC process utilized to remove heavy metals from electroplating effluent is both scalable and economically feasible. Consequently, EC shows that metal removal may be accomplished effectively using energy and that less sludge is produced. Particularly when treating industrial wastewater with significant levels of contaminants, the EC procedure might not be adequate. In this instance, more research is required to determine

Table 1. *Estimated values for zero, first- and second-order kinetics, and Zn and Cr removal rates with solution volume: 600 mL, pH: 9 and current density: 12.3 mA/cm².*

Heavy metal	First-order model $-dC/dt = k_1 C$ k_1 (min^{-1})	R^2	Second-order model $-dC/dt = k_2C^2$ k_2 ($L/mg.min$)	R^2	Pseudo first-order model $-dC/dt = k^{app}(C- Ce)$ k^{app} (min^{-1})	R^2
Cr	0.1212	0.8524	1.27	0.6387	0.0281	0.9872
Zn	0.0588	0.9486	0.0029	0.9332	0.0468	0.9986

Table 2. *Comparison of this study with previous studies*

Wastewater	Parameter	Electrode	Current densities (mA/cm^2)	pH	t (min)	η(%)	E (kWh/m^3)	C (kg/m^3)	References
Metal plating	Cu, Cr, Ni, Zn	Fe-Fe	4	9.56	45	> 97	6.25	1.31	[19]
Model wastewater	Cu, Ni, Zn, Mn	Fe-Fe	25	8.95	5-20	> 96	49	-	[9]
Smelting	Zn, Cd, Mn	Fe-Fe	15	6.9	120	99.9, 97.2, 85.5	14.76	2.09	[24]
Electroplating	Zn, Cr	Fe-Fe	12.3	9	30	79, 99	25	1.07	This Study

how EC can be used in conjunction with other oxidation treatment techniques to treat wastewater that contains significant levels of organic chemicals and heavy metals.

Author Statements:

- **Ethical approval:** The conducted research is not related to either human or animal use.
- **Conflict of interest:** The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper
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  HCG: Conceptualization, Methodology, Investigation, Writing.
- **Funding information:** The authors declare that there is no funding to be acknowledged.
- **Data availability statement:** The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

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