

# Performance Evaluation of Electrocoagulation Process for Removal of Chromium (VI) from Synthetic Chromium Solutions Using Iron and Aluminum Electrodes

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## Abstract

The performance of electrocoagulation with iron and aluminum sacrificial anode for removal of Cr(VI) was investigated. Several working parameters, such as pollutant concentration, pH, electrical potential, COD, turbidity, and contact time were studied in an attempt to achieve a higher removal capacity. Solutions of varying chromium concentrations (5-50-500 ppm) were prepared. To follow the progress of the treatment, samples of 25 ml were taken at 20 min intervals for up to 1 h and then filtered (0.45  $\mu$ ) to eliminate sludge formed during electrolysis. The pH of the initial solution was also varied to study their effects on the chromium removal efficiency. Results obtained with synthetic wastewater revealed that the most effective removal capacities of the studied metals could be achieved when the pH was kept at 3. In addition, the increase of electrical potential, in the range of 20-40 V, enhanced the treatment rate without affecting the charge loading required to reduce metal ion concentrations under admissible legal levels. The process was successfully applied to the treatment of electroplating wastewater where an effective reduction of Cr(VI) concentration under legal limits was obtained just after 20-60 min. The results of this study showed that the removal efficiency of chromium with iron electrodes is higher compared to aluminum electrodes. Also, it can be concluded that the electrocoagulation process has the potential to be utilized for cost-effective removal of heavy metals from water and wastewater.

**Key words:** Electrocoagulation, chromium removal, aluminum electrodes, iron electrodes

## Introduction

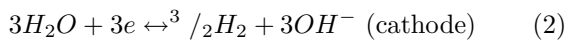
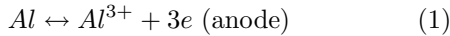
In recent years, there has been an increasing public awareness of the long-term effect of water containing dissolved heavy metal ions (Son et al., 2004). Heavy metals, such as chromium, cadmium, copper, zinc, and nickel, are contained in industrial waste water, which pollutes the environment and has negative effects on human health; the prevention of environmental pollution has been studied actively (Choi and Kim, 2003).

Toxic heavy metals are released into the envi-

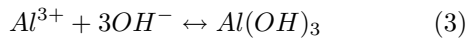
ronment from a number of industries such as mining, plating, dyeing, automobile manufacturing, and metal processing. The presence of heavy metals in the environment has led to a number of environmental problems. In order to meet the water quality standards for most countries, the concentration of heavy metals in wastewater must be controlled (Kim et al., 2005). Separation techniques of heavy metals, such as chromium, cadmium, copper, zinc, and nickel, from industrial wastewater include precipitation, ion exchange, adsorption, electro-dialysis, and filtration, but these techniques have limitations in

selective separation and they have problems associated with high investment and operation costs (Choi and Kim, 2005).

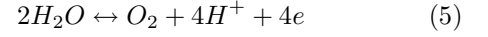
Chromium (VI) is released into aquatic environment from electroplating, metal finishing, chromate preparation, tannery, and fertilizer industries, and from industries that employ  $\text{Cr}^{6+}$  compounds as corrosion inhibitors (Gao et al., 2005; Adhoum et al., 2004). It is a potential carcinogen and its deleterious effects are well documented (Browning, E., 1969). The permissible limit of Cr(VI) for industrial wastewater to be discharged to surface water is  $0.1 \text{ mg l}^{-1}$  (Ranganathan, 2000). Hence it becomes imperative to remove Cr(VI) from wastewaters before discharging them into aquatic systems or on to land. Different methods, such as reduction and precipitation, ion exchange, electrodialysis, reverse osmosis, solvent extraction, electrochemical precipitation, and activated carbon adsorption have been suggested for the removal of hexavalent chromium (Gao et al., 2005). Most of them, however, are not efficient or cost effective. Electrocoagulation method using aluminum electrode has attracted significant attention for chromium removal process due to its operational simplicity. Electrocoagulation is a simple and efficient method where the flocculating agent is generated by electro-oxidation of a sacrificial anode, generally made of iron or aluminum. In this process, the treatment is performed without adding any chemical coagulant or flocculants, thus reducing the amount of sludge that must be disposed (Cenkin and Belevstev, 1985). On the other hand, electrocoagulation is based on the in situ formation of the coagulant as the sacrificial anode corrodes due to an applied current, while the simultaneous evolution of hydrogen at the cathode allows for pollutant removal by flotation. This technique combines 3 main interdependent processes, operating synergistically to remove pollutants: electrochemistry, coagulation, and hydrodynamics. An examination of the chemical reactions occurring in the electrocoagulation process shows that the main reactions occurring at the electrodes (aluminum electrodes) are:



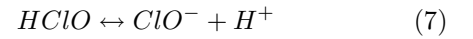
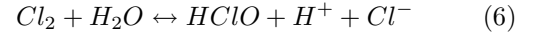
In addition,  $\text{Al}^{3+}$  and  $\text{OH}^-$  ions generated at electrode surfaces react in the bulk wastewater to form aluminum hydroxide:



If the anode potential is sufficiently high, secondary reactions may occur at the anode, such as direct oxidation of organic compounds and of  $\text{H}_2\text{O}$  or  $\text{Cl}^-$  present in wastewater:



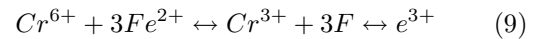
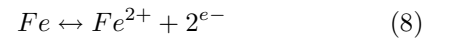
The produced chlorine undergoes a dismutation reaction at pH higher than 3–4:



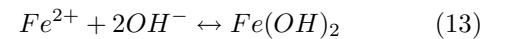
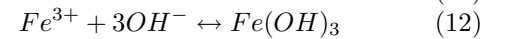
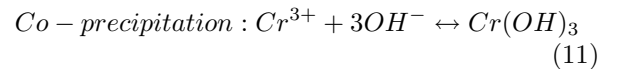
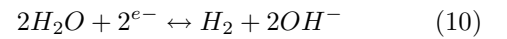
The aluminum hydroxide flocs act as adsorbents and/or traps for metal ions and so eliminate them from the solution. Furthermore, a direct electrochemical reduction of Cr(VI) in Cr(III) may occur at the cathode surface (Cenkin and Belevstev, 1985; Ogutveren et al., 1994). Simultaneously, the hydroxyl ions, which are produced at the cathode, increase the pH in the electrolyte and may induce co-precipitation of Cr(III) in the form of their corresponding hydroxides (Biswas and Lazarescu, 1991; Vik et al., 1984). This acts synergistically to remove pollutants from water.

Also, in the electrocoagulation (EC) cell, iron anodes dissolve and produce  $\text{Fe}^{2+}$ . This newly produced  $\text{Fe}^{2+}$  directly reduces  $\text{Cr}^{6+}$  to  $\text{Cr}^{3+}$  leading to the precipitation of  $\text{Cr}(\text{OH})_3$  and  $\text{Fe}(\text{OH})_3$ . Followings are the major reactions taking place in the EC cell (Kongsricharoern and Polprasert, 1995):

Anode (oxidation):



Cathode (reduction):



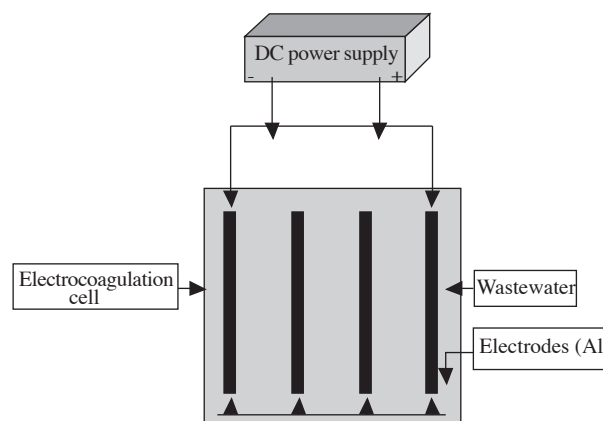
The object of this study was to evaluate chromium removal from synthetic chromium solutions by the electrocoagulation process method and to determine the effects of voltage, pH, COD, turbidity, and electrical conductivity on the removal efficiency.

## Materials and Method

All chemicals including potassium dichromate ( $K_2Cr_2O_7$ , 99%), sodium hydroxide pellets, concentrated sulfuric acid, and potassium chloride were analytical grade. Desired concentrations of chromium solution were prepared by mixing proper amount of potassium dichromate with deionized water. In order to increase the conductivity of the solution to  $1.6 \text{ mS cm}^{-1}$ , potassium chloride (1 N) was added to the solution before injecting it into the apparatus. The chloride salt added to the solution can also prevent the formation of the oxide layer on the anode and therefore reduce the passivation problem of the electrodes. The pH of the influent solution was adjusted using sulfuric acid solution and sodium hydroxide (0.1 M).

Experiments were performed in a bipolar batch reactor (Figure 1), with 4 aluminum electrodes connected in parallel (bipolar mode). Also, a set of experiments was performed with 4 iron electrodes. Only the outer electrodes were connected to the power source, and anodic and cathodic reactions occurred on each surface of the inner electrode when the current passed through the electrodes. The internal size of the cell was  $10 \text{ cm} \times 13 \text{ cm} \times 12 \text{ cm}$  (width  $\times$  length  $\times$  depth) with an effective volume of  $1000 \text{ cm}^3$ . The volume (V) of the solution of each batch was 1 l. The active area of each electrode was  $10 \times 10 \text{ cm}$ . The distance between electrodes was 1.5 cm. A power supply pack having an input of 220 V and variable output of 0–40 V with maximum current of 5 A was used as a direct current source.

The temperature of each system was maintained at  $25 \pm 1 \text{ }^\circ\text{C}$ . The pH values in the influent and reactor unit were measured using a pH meter model E520 (Metrohm Herisau, Switzerland). A Jenway Conductivity Meter (Model 4200) was employed to determine the conductivity of the solution. Samples were extracted every 20 min and then immediately filtered through a mixed cellulose acetate membrane ( $0.45 \text{ }\mu\text{m}$ ). The residual chromium concentration was determined using the Atomic Absorption method according to the standard method (APHA, 1992). The amount of chromium ion removal was measured at pH 3, 7 and 10 and in electrical potential of 20, 30, and 40 V. Also at the end of each stage of the experiment, the volume of produced sludge was measured.



**Figure 1.** A schematic diagram of the experimental setup.

## Results and Discussions

The electrocoagulation process is quite complex and may be affected by several operating parameters, such as pollutant concentrations, initial pH electrical potential (voltage), COD, and turbidity. In the present study, electrocoagulation process has been evaluated as a treatment technology for chromium removal from plating bath wastewater. Chromium removal efficiency in different conditions (e.g. pH, electrical potential) at various times was evaluated. It has been established (Chen et al., 2000) that pH has a considerable effect on the efficiency of the electrocoagulation process. Also, as observed by other investigators, the pH of the medium changed during the process. This change depends on the type of electrode material and initial pH and alkalinity.

In this study, the pH was varied in the range 3–10 in an attempt to investigate the influence of this parameter on the removal of chromium. Removal efficiencies of chromium as a function of the initial pH with aluminum and iron electrodes are presented in Tables 1-6. As observed previously (Vik et al., 1984), a pH increase occurs when the initial pH is low ( $<7$ ). Vik et al. (1984) ascribed this increase to hydrogen evolution at cathodes. However, this was contested by Chen et al. (2000), who explained this increase by the release of  $CO_2$  from wastewater owing to  $H_2$  bubble disturbance.

Indeed, at low pH,  $CO_2$  is over saturated in wastewater and can be released during  $H_2$  evolution, causing a pH increase. In addition, if the initial pH is acidic, reactions would shift towards a pH increase. In alkaline medium (pH  $> 8$ ), the final pH did not vary very much and a slight drop was recorded. This

result is in accord with previously published works and suggests that electrocoagulation can act as a pH buffer. In this research, the influent pH did not affect the removal efficiencies significantly over a wide range. Therefore, pH adjustment before treatment is not required in practical applications. The highest efficiency of chromium removal (for both iron and

aluminum electrodes) observed in acidic medium (pH = 3) for the initial chromium concentration of 500 mg l<sup>-1</sup> and at lower concentrations, the removal efficiency is almost complete at all pH values. At high chromium concentration, however, the complete removal may require longer time.

**Table 1.** Percent of chromium removal during electrocoagulation process using iron electrodes (Initial concentration = 5 mg/l).

T = 60 min	T = 40 min	T = 20 min	Voltage	pH
99.8	99.6	99.4	20	3
99.8	99.6	99.4	30	
99.9	99.8	99.6	40	
99.6	99.0	98.8	20	7
99.8	99.2	99.0	30	
99.8	99.6	98.8	40	
99.4	99.0	98.4	20	10
99.8	99.4	99.2	30	
99.8	99.6	99.0	40	

**Table 2.** Percent of chromium removal during electrocoagulation process using iron electrodes (Initial concentration = 50 mg/l).

T = 60 min	T = 40 min	T = 20 min	Voltage	pH
99.98	99.90	99.88	20	3
99.98	99.96	99.90	30	
99.998	99.98	99.94	40	
99.96	99.86	99.82	20	7
99.96	99.92	99.84	30	
99.98	99.94	99.90	40	
99.94	99.90	99.78	20	10
99.94	99.90	99.84	30	
99.96	99.92	99.88	40	

**Table 3.** Percent of chromium removal during electrocoagulation process using iron electrodes (Initial concentration = 500 mg/l).

T = 60 min	T = 40 min	T = 20 min	Voltage	pH
95.920	87.360	0 71.64	20	3
99.984	95.680	83.780	30	
99.996	99.976	85.040	40	
74.220	66.320	58.900	20	7
90.520	71.240	47.400	30	
99.898	94.240	61.440	40	
70.160	65.680	52.700	20	10
88.160	73.360	56.280	30	
89.660	77.160	58.410	40	

**Table 4.** Percent of chromium removal during electrocoagulation process using aluminum electrodes (Initial concentration = 5 mg l<sup>-1</sup>).

T = 60 min	T = 40 min	T = 20 min	Voltage, (V)	pH
99.80	99.20	97.40	20	3
99.80	99.30	97.40	30	
99.90	99.60	97.80	40	
99.80	97.20	74.60	20	7
99.80	98.20	89.60	30	
99.80	99.00	96.60	40	
99.20	96.40	77.20	20	10
99.20	98.00	95.80	30	
99.60	98.40	97.20	40	

**Table 5.** Percent of chromium removal during electrocoagulation process using aluminum electrodes (Initial concentration = 50 mg l<sup>-1</sup>).

T = 60 min	T = 40 min	T = 20 min	Voltage, (V)	pH
98.62	94.78	83.76	20	3
98.74	95.64	85.64	30	
98.88	95.80	88.98	40	
98.40	89.76	82.76	20	7
98.44	91.72	83.14	30	
98.72	95.72	83.46	40	
92.00	90.80	64.60	20	10
97.64	92.18	77.00	30	
98.34	92.58	81.80	40	

**Table 6.** Percent of chromium removal during electrocoagulation process using aluminum electrodes (Initial concentration = 500 mg l<sup>-1</sup>).

T = 60 min	T = 40 min	T = 20 min	Voltage, (V)	pH
25.60	22.00	21.80	20	3
35.80	27.00	24.80	30	
83.00	71.20	51.80	40	
20.40	19.60	13.60	20	7
24.60	20.40	17.80	30	
80.80	64.60	41.00	40	
23.00	13.80	8.80	20	10
26.80	22.00	12.80	30	
52.00	41.20	32.00	40	

The pH variation of the solution after electrocoagulation process in various voltages showed that the final pH for pH 3 and 7 of experiments with aluminum electrodes is higher compared to the initial pH, which is in agreement with results obtained later,

but for the initial pH (pH = 10), the final pH was lower than 10 and the final pH for all of the experiments with iron electrodes were higher compared to the initial pHs, which is in agreement with results obtained later (Kobyas et al., 2003).

### Aluminum electrode

Preliminary laboratory testing of the electrolysis cell involved in the determination of the effect of applied voltage on the efficiency of chromium removal. It is well-known that electrical current not only determines the coagulant dosage rate but also the bubble production rate and size and the floc growth (Letterman et al., 1999; Holt et al., 2002), which can influence the treatment efficiency of the electrocoagulation. Therefore, the effect of current density or electrical potential (voltage) on the pollutant removal was investigated. As expected, it appears that, for a given time, the removal efficiency increased significantly with the increase in the current density. The highest electrical potential (40 V) produced the quickest treatment with >97% chromium reduction occurring after only 20 min (with initial concentration of 5 mg l<sup>-1</sup>) and the lowest chromium removal efficiency occurred in the lowest electrical potential (20 V) and initial chromium concentration of 500.0 mg l<sup>-1</sup>. This is ascribed to the fact that at higher voltage the amount of aluminum oxidized increased, resulting in a greater amount of precipitate for the removal of pollutants. In addition, it was demonstrated that bubble density increases and their size decreases with increasing current density (Khosla et al., 1991), resulting in a greater upwards flux and a faster removal of pollutants and sludge flotation. As the current decreased, the time needed to achieve similar efficiencies increased and the results of this research confirm this fact. This expected behavior is explained by the fact that the treatment efficiency was mainly affected by charge loading ( $Q = It$ ), as reported by Chen et al. (2000). However, the cost of the process is determined by the consumption of the sacrificial electrode and the electrical energy, which are the economic advantages of this method. These results suggest that 40 V is an optimal electrical potential for the treatment of effluents containing chromium since it ensures the quickest removal rate with the lowest cost.

A set of experiments was performed with different initial concentrations of chromium to determine the time required for removal under various conditions of electrocoagulation process. The results obtained at different electrical potentials showed that initial concentration of chromium may have an effect on the efficiency of the removal and for higher concentration of chromium, higher electrical potential or more reaction time is needed. On the other hand, if the initial concentration increases, the time required

should increase too. It is clear from Tables 1-6 that in higher concentrations, longer time is needed for removal of chromium, but higher initial concentrations of chromium were reduced significantly in relatively less time compared to lower concentrations. The time taken for reduction thus increases with the increase in concentration. This can be explained by the theory of dilute solution. In dilute solution, formation of the diffusion layer at the vicinity of the electrode causes a slower reaction rate, but in concentrated solution the diffusion layer has no effect on the rate of diffusion or migration of metal ions to the electrode surface (Chaudhary et al., 2003).

The time dependence of chromium removal by electrocoagulation process at different pH levels is shown in Tables 1-6. It can be seen from the tables that up to 98%-99% of the initial concentration (with iron electrodes) decreased within 20 min of processing (for initial concentrations 5 and 50 mg l<sup>-1</sup>) and the residual chromium concentration in effluent at the end of reaction time (60 min) reached to <0.01 mg l<sup>-1</sup>, which is the recent guideline value of WHO (WHO, 1984) so we can discharge treated effluents to the environment in safety.

### Iron electrode

Comparison of chromium removal efficiency with iron and aluminum electrodes (Tables 1-6) showed that removal efficiency of chromium with aluminum electrodes is lower than chromium removal efficiency with iron electrodes.

A set of experiments was performed with different initial concentrations of chromium (5, 50 and 500 mg l<sup>-1</sup>) to examine the effect of the presence of COD with concentration 100, 500, and 1000 mg l<sup>-1</sup> and various levels of turbidity (10, 50, and 200 NTU) in wastewater on the removal efficiency of chromium. The results obtained at the optimum condition (pH = 3, reaction time = 60 min, and voltage = 40V) showed that the removal efficiency for various concentrations of chromium was unchanged and hence electrocoagulation process can be applied efficiently for chromium removal in the presence of COD and turbidity.

With regard to a series of tests conducted with different concentration of chromium in the solution, the weight of the electrode consumed with respect to different voltage of the pilot under study is given in Tables 7 and 8. It can be concluded that the higher voltage of the system applied, the more the weight of

the electrode consumed in the process is, and also the higher the concentration of the chromium in the solution, the higher consumption of the electrode would be. The electrodes consumed with 40 V in the process are much more than the process conducted with 20 V. As the chromium concentration in the solution increased to 500 mg l<sup>-1</sup>, the consumption of the electrode did not increase as much, but efficient chromium removal occurred, because much floc formation helped to sweep away chromium and there was no need for as much electrode consumption as before. For example, iron electrode consumption for the initial concentration of 5.0 mg l<sup>-1</sup> and voltage of 40 V was 9.01 g while for initial concentration of 500.0 mg l<sup>-1</sup> it was 7.70 g.

**Table 7.** Weight of iron electrode consumption during electrocoagulation process (g l<sup>-1</sup>).

Chromium Concentration, mg l <sup>-1</sup>			Voltage (V)
500.0	50.0	5.0	
3.07	3.91	3.01	20
3.60	4.50	4.27	30
7.70	9.07	9.01	40

**Table 8.** Weight of aluminum electrode consumption during electrocoagulation process (g l<sup>-1</sup>).

Chromium Concentration, mg l <sup>-1</sup>			Voltage (V)
500.0	50.0	5.0	
0.08	1.48	1.52	20
0.43	2.30	2.56	30
2.01	4.26	4.66	40

This study showed that consumed energy for removal of 1 g chromium at the electrical potential of 40 V (Tables 9 and 10), 5 mg l<sup>-1</sup> initial concentration of chromium and pH 3, 7, and 10 with aluminum electrodes was 15.16, 16.63, and 28.81 kWh, respectively, and consumed energy for removal of 1 g chromium at electrical potential of 40 V, 50 mg l<sup>-1</sup> initial concentration of chromium, and pH 3, 7 and 10 was 0.71, 2.01, and 2.11 kWh, respectively. Also, with 500 mg l<sup>-1</sup> initial concentration of chromium, with pH 3, 7, and 10, at the electrical potential of 40 V, the energy consumed for removal

of 1g of chromium was 0.32, 0.35, and 0.35 kWh, respectively. It can be concluded that the consumed energy decrease with an increase in chromium concentration, because the enhanced floc formation help sweep chromium away from the solution.

**Table 9.** Energy consumption during electrocoagulation process (kWh g<sup>-1</sup>), using iron electrodes at 40 V voltage.

Chromium Concentration, mg l <sup>-1</sup>			pH
500.0	50.0	5.0	
0.27	2.82	25.95	3
0.17	2.89	32.82	7
0.23	2.53	31.94	10

**Table 10.** Energy consumption during electrocoagulation process (kWh g<sup>-1</sup>), using aluminum electrodes at 40 V voltage.

Chromium Concentration, mg l <sup>-1</sup>			pH
500.0	50.0	5.0	
0.32	0.71	15.16	3
0.35	2.01	16.63	7
0.35	2.11	28.81	10

Finally, it can be concluded that electrocoagulation method is a reliable, safe, efficient, and cost-effective method for removal of chromium from industrial effluents, especially designed for pH = 3 and voltage = 40 V. On the other hand, in this study it was shown that electrocoagulation process achieves a fast and effective reduction of chromium (more than 98%) present in industrial effluents (such as plating bath wastewater). Indeed, the reported results show that electrocoagulation is an effective process for chromium removal from aqueous environments.

Nevertheless, further studies should be carried out to confirm the practical feasibility of this method for treating various wastewaters and with different conditions.

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