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Removal of heavy metal ions from aqueous solution by electrocoagulation using a horizontal expanded Al anode

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ABSTRACT

The performance of a batch cell with a horizontal expanded Al-anode in removing Cu^{2+} and Cr from a hexavalent chromium solution by electrocoagulation from separate solutions was examined. The effect of electrolysis time, current density, pH, initial pollutant concentration and NaCl concentration on the removal efficiency of Cu^{2+} and Cr has been investigated. The results revealed that as current density increases the removal efficiency of Cu^{2+} increases, while in the case of Cr it increases up to a certain limit, then remains approximately constant. The optimum pH is from 4 to 6 and 3.5 for Cu^{2+} and Cr removal, respectively. The removal efficiency of Cu^{2+} increases as NaCl increases, but in the case of Cr it decreases. COD measurements showed that a 66.67% reduction of COD has been obtained at the optimum conditions of Cu^{2+} removal. Energy consumption was calculated for both heavy metals at different current density and NaCl concentration. At the optimum condition an energy consumption of 43 kWh is required for the removal of one kg of Cu^{2+} ions; a value of 145 kWh is required for the removal of one kg of Cr. The difference in behavior between Cu^{2+} removal and Cr removal was explained on the basis that cathodic reduction of Cr⁶⁺ to Cr³⁺ which controls the rate of Cr removal.

Keywords: Electrocoagulation; Heavy metals removal; Expanded horizontal anode; Aluminum electrode; COD

1. Introduction

Toxic heavy metal are released into the environment as a result of different activities such as battery manufacture, mining, pigment manufacture, metal finishing, leather tanning and agricultural activities [1]. These metals have a detrimental effect on human health and environment if not managed efficiently. For this reason, industrial wastewaters are strictly regulated and have to be treated before being discharged [2,3]. Various techniques have been employed for treatment of heavy metals, including chemical precipitation, adsorption, ion exchange and reverse osmosis [3,4]. Chemical precipitation is the most popular among these techniques and considered to be the most economical, but it produces a large amount of sludge that requires further treatment. Reverse osmosis and ion exchange can effectively reduce metal ions, but their use is limited due to a number of disadvantages such as membrane fouling, high operating costs in addition to the limited pH range for the ion-exchange resin [5]. Although adsorption on activated carbon is a recognized method for the removal of heavy metals from wastewater, the high cost of activated carbon limits its use in industry [1,6]. Recently, electrochemical techniques such as electro-

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oxidation, electrocoagulation and electro-deposition have received great attention as eco-friendly and cost effective pollution control processes [7–10].

Electrocoagulation is based on creating metallic ions and hydroxide flocks within the water by electrodissolution of the anode usually made of iron or aluminum. The resulting metal ions and hydroxides have the ability to break emulsions and colloidal solution and to remove metal ions and organic pollutants by adsorption [10].

Electrocoagulation has been used to remove organic pollutants and heavy metals from effluents resulting from different industries such as textile finishing [11–13], metal finishing industry [3,4], restaurant wastewater [14], olive mill [15], leather tanning [16], food process [17], pulp and paper mills [18] and oil bilge water [19]. In most of these studies a cell with vertical electrodes was used. In a recent study Fouad et al. [20] showed that horizontal cells are more efficient than vertical cells, the authors used an array of horizontal Al cylinders as anode.

The aim of the present work is to study the removal of Cu^{2+} and Cr from a hexavalent chromium solution by electrocoagulation using a horizontal expanded Alelectrode. The use of a horizontal expanded metal anode placed over a horizontal plate cathode would improve the mixing efficiency in the cell by virtue of the ability of the expanded metal anode to generate eddies [21] as the rising H₂-solution dispersion coming from the cathode passes through the mesh anode. Improving the mixing efficiency at the anode would reduce concentration polarization and increase the dissolution efficiency of the anode. The effect of initial pH, time of electrocoagulation, initial metal ions concentration, current density and NaCl concentration on the Cu²⁺ and Cr removal from separate solutions were investigated.

2. Mechanism of electrocoagulation

The electrocoagulation process involves three successive stages:

- (a) Formation of coagulants by electrolytic oxidation of the sacrificial anode (either Al or Fe)
- (b) Adsorption of soluble or colloidal pollutants on coagulants
- (c) Aggregation of coagulants to form flocs, and they are removed by sedimentation.

The mechanism of electrocoagulation is highly dependent on the chemistry of the aqueous medium, especially conductivity. In addition, other characteristics such as pH, particle size and chemical constituent concentrations also influence the electrocoagulation process [10,22].

The electrochemical reactions with metal M as anode may be summarized as follows [13,23].

At the anode:

$$\mathbf{M}(s) \to \mathbf{M}_{(\mathrm{aq})}^{n+} + n \mathrm{e}^{-} \tag{1}$$

$$2H_2O_{(L)} \rightarrow 4H^+_{(aq)} + O_{2(g)} + 4e^-$$
 (2)

• At the cathode:

$$\mathbf{M}_{(aq)}^{n+} + \mathbf{n}\mathbf{e}^{-} \to \mathbf{M}(s) \tag{3}$$

$$2H_2O_{(L)} + 2e^- \rightarrow H_{2(g)} + 2\overline{O}H_{(aq)}$$

$$\tag{4}$$

The hydrogen gas released at the cathode would also help float the flocculated particles out of water [24]. If iron or aluminum electrodes are used, the generated $Fe_{(aq)}^{2+}/Fe_{(aq)}^{3+}$ (since Fe^{2+} may be oxidized to Fe^{3+} , which particularly depends upon the solution pH) or $Al_{(aq)}^{3+}$ ions will immediately undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides. The removal of Cu^{2+} and Cr ions by electrocoagulation in the present work was achieved by using aluminum electrode. The electrolytic dissolution of the aluminum anode produces the cationic monomeric species such as Al^{3+} and $Al(OH)_2^+$ then they are transformed to $Al(OH)_3$ and finally polymerized to $Al_n(OH)_{3n}$ in a wide pH range according to the following reactions [10]:

$$Al \to Al_{(aq)}^{3+} + 3e^{-} \tag{5}$$

$$Al_{(aq)}^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H_{(aq)}^{+}$$
(6)

$$n\operatorname{Al}(\operatorname{OH})_3 \to \operatorname{Al}_n(\operatorname{OH})_{3n}$$
 (7)

However, depending on the pH of the aqueous medium other ionic species, such as $Al(OH)_{2}^{+}$, $Al(OH)^{2+}$ and $Al(OH)_{4}^{-}$ may also be present in the system.

The gelatinous charged hydroxo cationic aluminum ion complexes can effectively remove pollutants (metal ions) by:

- (i) adsorption, as the hydroxide floc is relatively large and of less density, so it can be easily floated and separated
- (ii) co-precipitation, as the hydroxide ions formed at the cathode increase the pH of wastewater there by inducing precipitation of metal ions as corresponding hydroxides and co-precipitation with aluminum hydroxides:

$$M^{n+} + nOH^{-} \rightarrow M(OH)_{n(s)}$$
(8)

(iii) electrostatic attraction followed by coagulation [25–27].

As the hexavalent chromium ions are considered to be a strong oxidizing agent so its removal by the electrocoagulation method is achieved by the above mechanism or/and it can be reduced at cathode surface to trivalent chromium ions Cr^{3+} according to the following equation:

$$Cr_2O_7^{2-} + 6e^- + 7H_2O \rightarrow 2Cr^{3+} + 14^-OH$$
 (9)

in neutral medium, or

$$Cr_2O_7^{2-} + 6e^- + 14H^+ \rightarrow 2Cr^{3+} + 7H_2O$$
 (10)

in acidic solution.

The $(Cr)^{3+}_{(aq)}$ ions can be precipitated as $Cr(OH)_{3(s)}$ by raising the pH of the solution, or it can be adsorbed on the gelatinous flocs and removed by flotation.

3. Experimental

3.1. Chemicals and analytical

A stock solution of 1000 mgL⁻¹ of Cr⁶⁺ and Cu²⁺ were prepared by dissolving the required amounts of potassium dichromate and copper sulphate in deionized water. All chemicals were of analytical grade and supplied by Merck, Germany. In order to increase the conductivity of solutions, sodium chloride was added to the solutions. The pH of solutions were adjusted by using 0.1 M NaOH and/or 0.1 M HCl solution and measured by pH meter (Metrohm 654, Switzerland).

The concentration of metal ions (total Cr, Cu²⁺) in each sample was analyzed using Perkin–Elmer 2380 atomic absorption.

3.2. Batch experiments

Batch experiments were carried out in the electrocoagulation unit shown in Fig. 1. It consists of a rectangular Plexiglas container of dimensions 15×15×20 cm equipped with two parallel horizontal aluminum electrodes (15×15×0.4 cm).

The cathode was an Al plate placed horizontally at the bottom of the cell and was isolated from its back with epoxy resin. The anode was a horizontal expanded Al electrode (mesh no. = 6, wire diameter = 0.12 cm, specific area = $7.713 \text{ cm}^2/\text{cm}^3$, porosity = 0.769) placed 5 mm above the cathode in order to minimize the IR drop of the cell. This anode geometry would assist in the flotation of electrocoagulation flocs since it permits cathodic H₂ bubbles to pass through.

The electrodes were connected to a d.c. power supply (20 V, 10 A) with a multi-range ammeter connected in series and a voltmeter connected in parallel.

Before each run, the oxide and/or passivation layer on the electrodes surface were removed by dipping them for 15 min in 20% HCl solution. Two liters of metal solution were fed into the electrocoagulation unit. After adjusting the temperature, pH and current density to the desired value the experimental run was started, samples of 5 ml were periodically taken from the unit. The samples were left to settle for 1 h and then analyzed.



Fig. 1. Schematic view of the experimental system, (1) d.c. power supply, (2) anode, (3) reactor, (4) cathode, (5) electrode gap, (6) solution, (7) water bath.

4. Results and discussion

4.1. Effect of current density

Fig. 2 shows the effect of electrolysis time on % removal efficiency of a metal ions at different current densities (c.d) ranging from 12 to 96 A/m² and 24 to 96 A/ m² for Cu²⁺ and Cr, respectively. Preliminary experiments showed that at c.d of 12 A/m² no appreciable removal of Cr occurred thus a minimum value of 24 A/m² has been selected. As c.d and electrolysis time increase, % removal efficiency of Cu2+ increases linearly until no further removal is obtained as shown in Fig. 2a. The increase in the rate of Cu²⁺ removal with c.d is consistent with Faraday's law according to which the amount of generated aluminum hydroxide which adsorbs Cu²⁺ increases with increasing c.d. These results are in agreement with the results of other investigators [3]. Fig. 2a shows also that beyond 48 A/cm² there is no significance increase on % copper removal. It is also clear that as the time increases from 15 to 30 min, there is a slight increase on % copper removal and beyond 30 min there is no change on % removal, therefore the optimum time for electrocoagulation of copper is chosen to be 30 min for 48 A/cm².

Fig. 2b shows that in the case of Cr ions, c.d beyond 48 A/m² has a negligible effect on % removal efficiency.

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Fig. 2a. Effect of time on % removal efficiency at different current densities (Cu²⁺ concentration = 100 ppm, NaCl = 1 g/L, initial pH = 6.5).



Fig. 2b. Effect of time on % removal efficiency at different current densities (initial Cr^{6+} concentration = 50 ppm, NaCl concentration = 1 g/L, initial pH = 3.5).

This result may be explained as follows: as mentioned earlier Cr^{6+} is removed mainly by cathodic reduction to Cr^{3+} followed by precipitation as $Cr(OH)_3$ in the cathode vicinity which is strongly alkaline by virtue of the H₂ evolution according to Eq. (4).

 $Cr(OH)_3$ is carried away from the cathode vicinity to the solution bulk by the rising H₂ bubbles which evolves at the cathode, in the solution bulk $Cr(OH)_3$ and free Cr^{3+} which dissolve from $Cr(OH)_3$ adsorb on aluminum hydroxide. The fact that the % Cr^{6+} reaches a limiting value beyond a c.d of 48 A/m² may be explained by the following

 (i) the reduction of Cr⁶⁺ to Cr³⁺ is diffusion controlled reaction in unagitated and slightly agitated solution [28], consequently current density has insignificant effect on the rate of Cr⁶⁺ reduction.

- (ii) As the current density increases the anode potential increases and may reach O₂ discharge potential with subsequent decrease of Al dissolution efficiency [29].
- (iii) As the current density increases the rate of H₂ bubble evolution increases with the consequent increase of pH in the vicinity of cathode. As the pH increases the rate of Cr⁶⁺ reduction to Cr³⁺ decreases consequently, % removal decreases. An electrolysis time of 50 min was chosen as optimum time for further experiments of Cr.

As seen from the above results both time and current have strong effect on % metal removal efficiency. To consider the combined effect of both time and current charge loading must be considered. Charge loading is defined as amount of electricity passes per unit volume of solution to be treated (As/L) [30].

It has been found that at a c.d of 48 A/m² the corresponding values of charge loading are 1800 and 3000 As/L for Cu²⁺ and Cr respectively, which indicates that the removal of Cu²⁺ is much more efficient than Cr also that results support the difference in mechanism between Cu²⁺, Cr⁶⁺ removal, which is consistent with other investigators [3].

4.2. Effect of initial metal ion concentration

Fig. 3 illustrates the effect of initial metal ion concentration on % removal efficiency for Cu^{2+} and Cr. It is obvious from Fig. 3 that % removal efficiency of Cu^{2+} and Cr decreases considerably with increasing the initial concentration of their ions. Similar results have been found by other authors [3,26,30] which can be attributed to the fact that at a constant current density the coagulant dosage and H₂ production rate are constant and do not depend on initial metal concentration, thus as metal ion concentration increases, the adsorption capacity of ge-



Fig. 3. Effect of initial metal ions concentration on % removal efficiency. (Cu^{2+} – c.d = 48 A/m², NaCl concentration = 1 g/L, initial pH = 6, time = 30 min; Cr^{6+} c.d = 48 A/m², NaCl = 1 g/L, initial pH = 3.5, time = 50 min).

latinous aluminum hydroxide compounds is consumed its maximum potential.

4.3. Effect of electrolyte

The presence of electrolyte in solution during electrolysis is essential to decrease IR drop within the cell and consequently decreases energy consumption. Sodium chloride is commonly employed as an electrolyte in electrocoagulation process for many reasons

- (i) decrease energy consumption
- (ii) Cl⁻ ions act as antipassive agents and destroy passive layer formed on anode during electrolysis especially in case of Al anode
- (iii) chlorine gas is evolved at the anode and may oxidize any organic matter present in aqueous solution and hence contribute to enhancement of electrocoagulation process [9].

The effect of NaCl concentration on the performance of electrocoagulation process of Cu²⁺ and Cr is shown in Fig. 4. It is noted that Cu²⁺ and Cr show different behavior with respect to NaCl concentration. The removal efficiency of Cu²⁺ increases by increasing NaCl concentration probably because excess Cl⁻ ions inhibit Al anode passivation with a consequent increase in the anode dissolution efficiency [31], while in the case of Cr the removal efficiency decreases by increasing NaCl which may be attributed to the fact that the presence of Cl⁻ ions enhances the solubility of Cr(OH)₃ and consequently decreases co-precipitation of Cr(OH)₃ and lowers its removal efficiency [32]. It is also possible that with increasing NaCl concentration interionic attraction between NaCl ions and K₂Cr₂O₇ ions becomes pronounced, accordingly,



Fig. 4. Effect of electrolyte concentration on % removal efficiency.($Cu^{2+} - Cu^{2+}$ concentration = 100 ppm, c.d = 48 A/m², initial pH = 6, time = 30 min; Cr^{6+} – initial Cr^{6+} concentration = 50 ppm, c.d = 48 A/m², initial pH = 3.5, time = 50 min).

the activity and diffusivity of Cr_2O_7 decrease [33] with a consequent decrease in the rate of the diffusion controlled cathodic reduction of Cr_2O_7 .

4.4. Effect of pH

The effect of initial pH on % removal of Cu²⁺ and Cr is shown in Fig. 5. The maximum removal efficiency of Cu²⁺ occurs at pH from 4 to 6 with % removal efficiency of 99.99–96.2%, respectively. It is well known that in the pH range from 4 to 8 Al dissolves at anode and forms insoluble monomeric and polymeric aluminum hydroxide species, such as $Al_{13}O_4(OH)_{24}^{7+}$. These polymeric species undergo different transformation until finally precipitate as $Al(OH)_{3(s)}$ which adsorb Cu^{2+} and are floated by the rising H₂ bubbles. In acidic solutions (pH = 2-3) Al³⁺ and $Al(OH)^+_2$ are the dominant soluble chemical species. They are ineffective coagulants. When pH is higher than 10, the soluble anion Al(OH)₄ increases and Al(OH)_{3(s)} decreases therefore % removal decreases [34]. Fig. 5 also shows that the maximum removal of Cr occurs at pH = 3.5, in which the reduction of Cr⁶⁺ to Cr³⁺ is maximum as the reduction of Cr⁶⁺ is favored in acidic medium [35]. In view of the fact that the reduction potential of Cr6+ decreases with acidity as indicated by applying Nernest's equation to reaction (10). As the pH increases the rate of Cr⁶⁺ reduction to Cr³⁺ decreases consequently, % removal decreases. This result agrees with previous studies [36]. In highly acidic condition (pH = 2), the reduction of Cr^{6+} to Cr^{3+} is efficient, however precipitation and electrocoagulation of Cr³⁺ are not effective as the librated OH- is not enough to increase pH and effect precipitation of Cr3+ which takes place at pH = 8.5 in the presence of Cl⁻ ions [37]. This agrees with the results obtained by Mouedhen et al. who followed pH change during electrolysis using Al anode at different initial pHs. They found that in highly acidic electrolyte



Fig. 5. Effect of initial pH on % removal efficienc ($Cu^{2+} - Cu^{2+}$ concentration = 100 ppm, NaCl = 1 g/L, c.d = 48 A/m², time = 30 min; Cr^{6+} - initial Cr^{6+} = 50 ppm, c.d = 48 A/m², NaCl concentration = 1 g/L, time = 50 min).



Fig. 6. Effect of initial pH on final pH after electrocoagulation of Cu^{2+} ions (Cu^{2+} initial concentration = 100 ppm, NaCl concentration = 1 g/L, c.d = 48 A/m², time = 30 min).

(pH = 2), the alkalinity produced during electrolysis was not sufficient to increase the pH of the solution [29]. It is obvious from Fig. 5 that within the pH range from 4 to 8 the % removal is approximately constant. It is noteworthy that the amount of Al electrodissolved depends on the pH. It has been found that a sharp increase of the amount of Al released occurs at pH 2–3. Beyond pH = 3, the variation of the amount of dissolved aluminum seems to be insignificant [29], which may explain the slight change on the % removal in the pH range 4–8.

Table 1

Effect of current density on % removal efficiency and energy consumption

Fig. 6 shows the final pH after electrocoagulation process vs. initial pH. It can be seen that final pH increases from 5 to 9.3 corresponding to the initial pH from 2 to 8 respectively. The increase of the pH is due to the liberation of OH⁻ at cathode. It is also clear that a slight drop in final pH occurs in highly alkaline medium (pH = 10). That pH drop is attributed to the dissolution reactions of Al(OH)₃ and the formation of Al(OH)⁴ anion which consumes OH⁻ group [29].

4.5. COD measurement

COD is one of the main parameters which could affect the performance of electrocoagulation process strongly. COD measurement has been conducted at optimum operating condition of Cu^{2+} removal. The results showed thast 66.67% COD reduction has been obtained.

4.6. Energy consumption

From Eq. (11) energy consumption (E.C) required to remove 1 kg of heavy metal was calculated.

Energy consumption (kWh/kg) is calculated as follows

$$E.C = \frac{I * V}{1000(m)}$$
(11)

where *I* is the applied current (A), *V* is the cell voltage (V) and *m* is amount removed in (kg/h).

Table 1 illustrates the effect of c.d on E.C and % removal efficiency for Cu^{2+} , Cr respectively. As c.d. increases E.C increases for Cu^{2+} and Cr within the present range of

Cr ⁶⁺ (Constant cond NaCl conc. = 1 g/l)	itions: Cr ⁶⁺ conc. = 50	ppm, time = 50 min,	Cu ²⁺ (Constant conditions: Cu ²⁺ conc. = 100 ppm, time = 30 min, NaCl conc. = 5 g/l)		
Current density (A/m ²)	Removal efficiency (%)	Power consump- tion (kWh/kg)	Current density (A/m²)	Removal efficiency (%)	Power consump- tion (kWh/kg)
24	69	67.63	12	53.11	35.77
48	85	145	24	77.78	37.44
72	86	293.6	48	95.11	43.89
96	85.5	428.8	96	98.74	64.06

Table 2

Effect of NaCl concentration on % removal efficiency and energy consumption

Cr ⁶⁺ (Constant conditions: Cr ⁶⁺ conc. = 50 ppm, c.d = 48 A/m ² , time = 50 min)			Cu ²⁺ (Constant conditions: Cu ²⁺ conc. = 100 ppm, c.d = 48 A/m ² , time = 30 min)		
NaCl concentration (g/l)	Removal efficiency (%)	Power consump- tion (kW.h/kg)	NaCl concentration (g/l)	Removal efficiency (%)	Power consump- tion (kWh/kg)
1	85	145	0.5	95.11	42.11
2	81.3	120.9	1.25	98.57	25.39
3	79.5	113.9	1.75	100	24.8

c.d, E.C ranges from 35.7 to 64 kWh/kg for Cu^{2+} and from 67.6 to 428.8 kWh/kg for Cr. The optimum current density chosen for Cu^{2+} and Cr is 48 A/m² which corresponds to the maximum removal rate and the lowest E.C.

Table 2 shows the effect of NaCl on E.C and % removal for both Cu^{2+} and Cr. As NaCl concentration increases E.C decreases and % removal increases in the case of Cu^{2+} . However in the case of Cr as NaCl concentration increases, both E.C and % removal decrease. At the optimum condition an energy consumption of 43 kWh/kg is required for Cu^{2+} removal; however a value of 145 kWh/kg is required for Cr removal.

5. Conclusions

The use of a horizontal expanded electrode seems to be effective for the removal of heavy metals by electrocoagulation, since it offers a high specific area beside its ability to improve the mixing conditions in the cell by virtue of its ability to generate turbulence in the solution when H₂ bubble-liquid dispersion passes across it. The results showed that as current density increases, the removal efficiency of Cu²⁺ increases, however in the case of Cr, the removal efficiency increases up to a certain limit of 48 A/m², then it remains approximately constant. Optimum pH was found to be from 4 to 6 and 3.5 for Cu²⁺ and Cr respectively. As the initial concentration of Cu²⁺ and Cr increases, the removal efficiency decreases for both metal ions.

The removal efficiency of Cu²⁺ increases slightly as NaCl concentration increases; while the removal efficiency of Cr decreases slightly as NaCl concentration increases. Under optimum condition of Cu2+ removal, 66.67% of COD reduction is obtained. Energy consumption calculations reveal that at optimum operating conditions, for every one kg of Cu2+ ions removed 43 kWh is required, however for every kg of Cr ions removed a value of 145 kWh is needed. The difference in behavior between the investigated heavy metal ions is attributed to the difference in the mechanism of removal of Cu²⁺ from that of Cr. While Cu²⁺ removal takes place by adsorption on aluminum hydroxide and floatation, Cr is mainly reduced to Cr³⁺ at the cathode followed by electrocoagulation of Cr³⁺, besides the ionic charge difference between Cu²⁺ and Cr.

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