

## Mercury Removal Using Al – Al Electrodes by Electrocoagulation

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**ABSTRACT:** The attempt has been made to remove mercury from mercury containing waste water prepared synthetically by using sodium chloride as an electrolyte using sacrificial aluminum anode in a batch wise by electrocoagulation cell. The affect of applied potential, initial pH, initial concentration of solution, agitation, electrolyte concentration and energy consumption on percent removal of mercury have been investigated. The removal efficiency of mercury was achieved 98.5% under optimum condition in which solution pH was 4.5, applied potential 9 V, initial solution concentration 50 ppm, electrolyte concentration 1.333 g/lit. With a stirring speed 400 rpm.

**Keywords:** Aluminum chloride, applied potential, Electrocoagulation, Energy consumption, Optimization

### I. Introduction

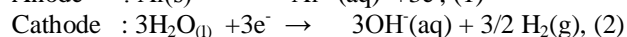
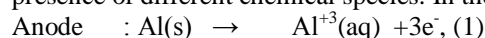
Electroplating or acid mine waste water generate different toxic substances like cyanides, alkaline cleaning agent, degreasing solvents, oil, fats and metals [1, 2]. Successful empirical studies [3, 4, 5, 6, 7, 8] show the successful treatment of waste water. Different techniques like adsorption, ion exchange, reverse osmosis and precipitation have been employed for the treatment of heavy metals. Effluent from metal plating and surface finishing generate higher concentration of metals in effluents. Due to high toxicity, discharge of waste water containing heavy metals are strictly regulated in the environment.

Since more than a century, electrocoagulation technique has been available. In last decades electrocoagulation [9, 10, 11, 12] consider an efficient method for the treatment of heavy metal containing solutions. Electrocoagulation is able to eliminate chemical oxygen demand [4,5,6,13], color [7], restaurant waste water [14], electroplating wastewater [3] and textile wastewaters [15,16,17,18]. The main objective of this study was examining some fundamental mechanisms and different aspects of electrocoagulation process influencing removal of mercury by electrocoagulation

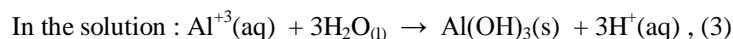
### II. Electrocoagulation

Electrocoagulation is an efficient method for water and waste water to destabilize the finely dispersed particles. Electrocoagulation consist of simple with reduction of equipments, easy operation and also decreased of sludge generation [19]. Different mechanisms like coagulation, absorption, adsorption, precipitation and flotation play an important role in the electrocoagulation process. The most common electrode material are aluminum and iron which are readily available, cheap and effective [20]. When direct current is applied, due to dissolution of aluminum electrode which act as a sacrificial anode, situ generation of  $Al^{+3}$  ions which act as a coagulants took place. These coagulants are responsible for destabilize finely dispersed particles in the vicinity of the anode surface and form flocks. Meanwhile, tiny bubble of  $H_2$  produced at cathode.

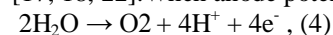
Due to dissolution of sacrificial electrode, various species generated [21] depending upon the pH of the solution and presence of different chemical species. In the case of aluminum, the main reactions are:



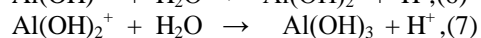
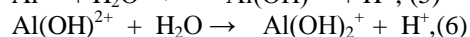
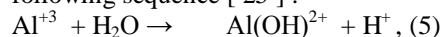
As per reaction (1) and (2),  $Al^{+3}$  and  $OH^{-}$  ions generated due to electrolytic dissolution of anode and form various monomeric species which follows complex precipitation kinetics and finally transform into  $Al(OH)_3(s)$ .



These "sweep flocks"  $Al(OH)_3$  have large surface area and amorphous in nature. So quick adsorption of soluble organic compounds and trapping of colloids particles forms. Finally, these flocks eliminated due to sedimentation or  $H_2$  floatation [17, 18, 22]. When anode potential is sufficiently high [3, 5, 20, 23, 24] secondary reaction may occur with oxygen evolution:



Due to spontaneous hydrolysis reaction, aluminum ion ( $Al^{+3}$ ) as per eq. (1), generated different monomeric species as per following sequence [ 25 ] :



According to eq.(5), (6) and (7) anode vicinity appears as acidic while due to generation of  $H_2$  as per eq.(2) cathode electrode vicinity became alkali.

### III. Experimental set up

Electrocoagulation was performed in a cylindrical electrochemical cell in a batch wise with a pair of aluminum electrodes displayed in fig. (1) With a two facing of 10 mm. The dimension of each electrode was 14 cm. x 1.8 cm. x 0.2 cm.

with a effective area of each electrodes was 6cm. x 1.8 cm. x 0.2 cm. Waste water samples used in the experiments were prepared synthetically using  $HgCl_2$  having 99.99 % of purity from Merck. All solutions used for electrocoagulation were prepared from analytical grade chemical reagent (Merck products). Experiments were conducted with temperature

around  $\pm 28^{\circ}\text{C}$ . The solution was stirred by an overhead stainless steel rod stirrer for homogeneity of the solution. After each run, the electrodes were washed thoroughly with dilute  $\text{H}_2\text{SO}_4$  solution to remove oxide then washed with distilled water to remove any solid residues on the surfaces then dried and reweighed. The parameters chosen in the experiments whose range given in table no. (1). Electrodes were connected to a direct current power supply. Current passing through the circuit and applied potential were measured by using two digital multimeters as ampere meter and voltmeter respectively. Sodium chloride as supporting electrolyte was added to increase ionic conductivity of the solution. Besides its ionic contribution also reduce the adverse effect of anions like  $\text{HCO}_3^-$ ,  $\text{SO}_4^{2-}$ . Electrolysis generated chlorine is also improved in water disinfection [26]. High precision pH meter (microprocessor based model 1012 – E) equipped with a combined glass electrode used for measurement of pH. Sulfuric acid or sodium hydroxide was used for changing the initial pH of the solution. A digital conductivity meter (model – 1601 E) equipped with an immersion measurement probe was used to measure the ionic conductivity of aqueous phase. The concentration of mercury determined by using a UV–vis spectrophotometer (Elco SL – 159) at 575 nm. Mercury removal efficiency is calculated as per following equation [27]:

$$\text{Mercury removal efficiency (\%)} = \frac{C_i - C_o}{C_i} \times 100, \quad (8)$$

$C_i$  and  $C_o$  were the initial and present concentration (mg/lit.) of the mercury in the solution respectively.

## IV. Results and discussion

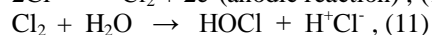
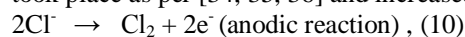
### 4.1 Effect of electrolyte concentration

Fig. (2) Shows the effect of without electrolyte concentration on removal efficiency of mercury with time. In the absence of electrolyte, conductivity of the solution and current density would be lower. So amount of the chloride ions in the solution decreased, which cause the increased of the total residence in electrocoagulation cell. As conductivity of the solution was less, generation of aluminum hydroxide was slower in the solution, which was responsible for lower removal efficiency of mercury. In the following table no. (2), Shows the effect of electrolyte concentration of absents to maximum concentration on removal efficiency of mercury with time.

Fig.(3) indicates the effect of NaCl concentration with respect to the time to achieved 98.5 % mercury removal. Low electrical conductivity of the solution cause maximum resistance between anode and cathode which is reduced on the base of high energy. By addition of sufficient quantity of NaCl in the solution overcome this problem and increase the conductivity and density of current would be increased as per [28]:

$$V = E_C - E_A - |\varepsilon_A| - |\varepsilon_C| - IR_{\text{cell}} - IR_{\text{circuit}}, \quad (9)$$

In addition of electrolyte increase of conductivity of the solution resulted into the reduction of the energy consumption [29, 30]. Also removal efficiency increase with increase in conductivity of the solution according [28, 31, 32]. According to reactions (10), (11) and (12) [33], when NaCl as electrolyte was added, formation of weak hypochlorous acid took place as per [34, 35, 36] and increased of chloride ion concentration as dissociation took place.



For lower concentration 0.6667 g/lit. There are not enough ions formed to conduct the current and probably lower efficiency of mercury removal observed [37]. As concentration of chloride ions increased 1.333 g/lit. in the solution, destroy the passivation layer and increase dissolution of electrode either by precipitation or incorporation of chloride ions into oxide film in the electrode dissolution [38]. As anodic dissolution of the electrode increased which cause blanketing oxide layer and attributed optimum 98.5 % removal rate of mercury? But at higher amount of  $\text{Cl}^-$  ions, solubility of mercury will be higher which reduced removal rate of mercury.

### 4.2 Effect of initial concentration

In the fig. (4), effect of initial concentration of mercury from 30 ppm to 100 ppm on removal efficiency with time is shown. Out of the different mechanisms for removal of mercury in electrocoagulation adsorption on to metallic hydroxide flocks has crucial important. Although at higher concentration there is certain limitation for adsorption capacity of flocks [37]. Concentration of ions and their flocks initiates at initial concentration – 30 ppm of solution turned towards into the removal rate of mercury. Optimization removal 98.5 % gain at 50 minute for 50 ppm concentration of mercury as adsorption capacity of flocks maximum. But above the concentration of 50 - ppm, adsorption capacity of flocks exhausted which was responsible for inactiveness of electrode effective surface. Meanwhile, at higher concentration of solution, generation of intermediate products increased which compete with mercury and water for active site on the electrodes [37] which leads to insolubility of aluminum hydroxide ions and declined mercury removal rate fall down. In the following table no. (3), Shows the effect of initial concentration on removal efficiency of mercury with time

### 4.3 Effect of applied potential

During electrocoagulation, applied potential is a crucial parameter on the performance of electrolytic cell. In this applied potential was varied by 6V, 9V, 12V at a constant pH 4.5, agitation 400 rpm, initial concentration 50 ppm, electrolyte concentration 1.333 g/lit. Shown in fig. (5). as per reaction (1) and (4) sacrificial metal dissolution and oxygen formation compete with each other respectively. According to faraday's law as per following eq. (14) [39]:

$$m = ItM, \quad (14)$$

ZF

m = amount of anode material dissolved (g)

I = current (A)

t = electrolysis time (s)

Z = no. of electrons involved in the reaction

F = Faraday's constant

The amount of dissolved sacrificial anode and generated hydroxyl ions are controlled by the applied current. After applied sufficient voltage, dissolution of sacrificial anode cause metal ions which were hydrolyzed and form a series of metallic hydroxide species. Electrostatic interparticles attraction increase enough as dispersed particles neutralized by these hydroxides species and encouraging agglomeration [40]. In the following table no.(4), Shows the effect of applied potential on removal efficiency of mercury with time. As applied potential increased for same initial pH, removal rate has increased according to [41, 42, 43, 44, 45]. In the early stage of electrocoagulation at voltage of 6V, suspended oxide particles were not able to destabilized and occur insufficient coagulation leads to slower removal rate of mercury. At critical voltage of 9V, optimum removal efficiency 98.5 % attributed as to surpass of effective  $Al^{+3}$  dissolution [46]. When voltage is exceed than this value, oxygen evolution as per eq. (4), lower the aluminum dissolution. Simultaneously, the oxygen promoted oxidation of Al to  $Al^{+3}$ . So, less or no aluminum ions are available [46] because abrupt decreased in mercury removal.

#### 4.4 Effect of agitation

Electrocoagulation performed at different stirring speed of 200 rpm, 400 rpm and 600 rpm at optimum parameters of initial pH – 4.5, initial concentration – 50 ppm., voltage – 9V, electrolyte dosage 1.333 g./lit. For a different time vary from 5 minute to 60 minute shown in fig.(6). In the following table no. (5), Shows the effect of agitation on removal efficiency of mercury with time. During electrocoagulation, generation of aluminum hydroxides, different polymers with its complexes took place according to different criteria In the beginning of the electrocoagulation from 200 to 400 rpm, generation of coagulants were negligible which turned into slower removal rate of mercury. As rpm increased during electrocoagulation, aluminum hydroxide as a coagulant formed which leads to improved contact between mercury and different aluminum hydroxides and complexes, formed in the vicinity of an electrode [47] which was responsible for optimum 98.5 % removal rate of mercury at 50 minute. But at higher agitation, excess temperature took place which destroyed the aluminum oxide film on the electrodes surface and compact  $Al(OH)_3$  flocks formed on the surface of the electrodes [48] and reduced the removal rate of mercury.

### V. Conclusion

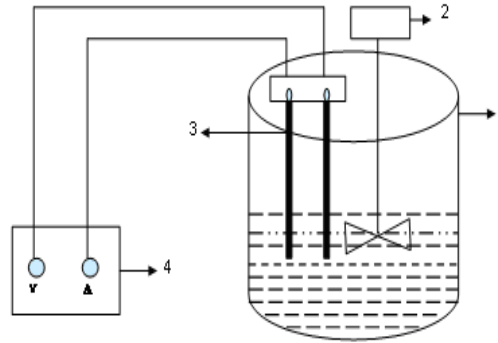
In this electrocoagulation, the behavior of aluminum electrodes has been find out based on the different parameters such as electrolyte concentration , electrolysis time, Initial pH, solution concentration, applied potential and agitation for removal of mercury containing wastewater prepared synthetically. Removal mechanism mainly due to the formation of hydroxyl ion as reduction of the cathodic surface. Also generation of aluminum ions based on the weight loss of electrodes. To destroyed the passive film at anode and by optimizing electrolyte concentration 1.333 g/lit., applied potential 9V, initial solution pH 4.5, initial solution concentration 50 ppm, agitation 400 rpm, removal efficiency of mercury achieved 98.5 % within 50 minutes. Thus the electrocoagulation method is efficient and faster method for removal of mercury at optimum conditions.

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Figures and Tables



Schematic process arrangement

- 1. Electrolytic cell
- 2. Stirrer
- 3. Aluminum - Aluminum electrode pair
- 4. Power supply (V, Voltage regulator and A, current regulator)

Fig. (1) Electrolytic cell

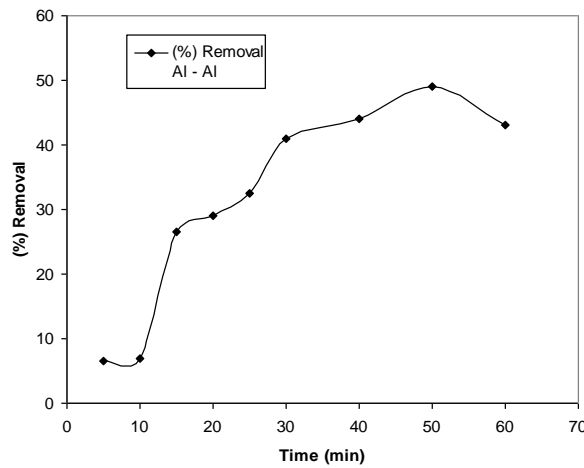


Fig. (2) Effect of without electrolyte concentration on the removal efficiency Of mercury with Time. Ppm – 50, Agitation – 400 rpm, Voltage – 9V, pH – 4.5

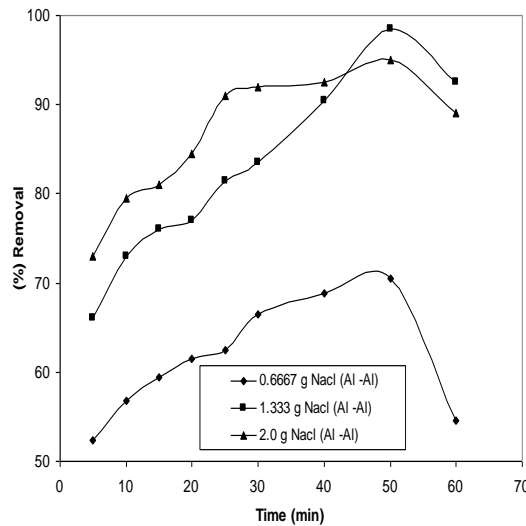
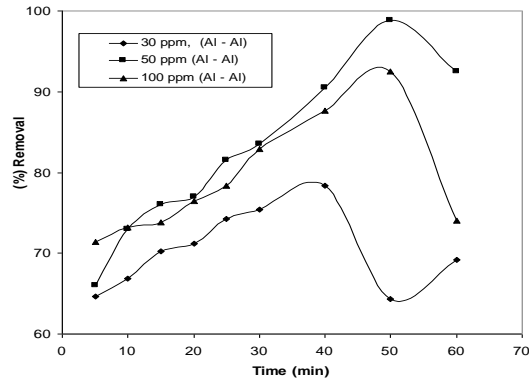
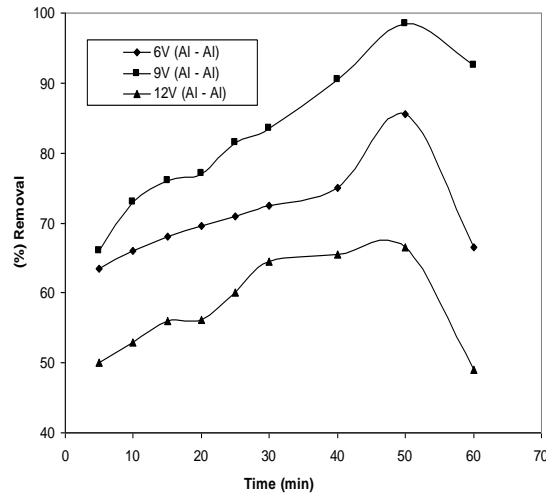


Fig. (3) Effect of electrolyte concentration on the removal efficiency Of mercury with Time. Ppm – 50, Agitation – 400 rpm, Voltage – 9V, pH – 4.5



“Fig. (4) Effect of initial concentration on the removal efficiency of mercury With Time. Voltage – 9V, Agitation – 400 rpm, Electrolyte concentration – 1.333 g/lit, pH – 4.5”



“Fig. (5) Effect of applied potential on the removal efficiency of mercury with Time. Ppm – 50, Agitation – 400 rpm, Electrolyte concentration – 1.333 g/lit., pH – 4.5”

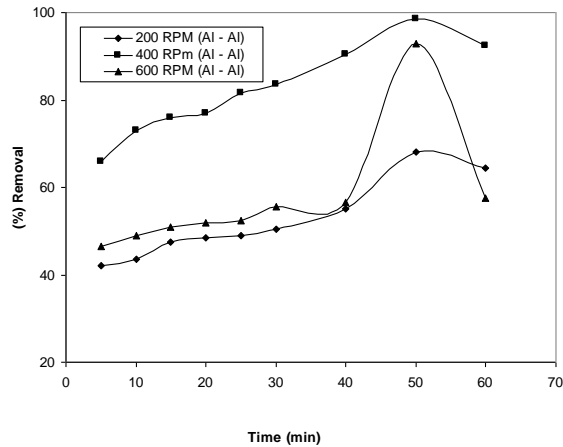


Fig. (6) Effect of agitation on the removal efficiency of mercury with Time. Ppm – 50, electrolyte concentration 1.333 g/lit., Voltage – 9V, pH – 4.5”

TABLE (1) Experimental parameters with Range

<i>Experimental parameters</i>	<i>Range</i>
Agitation (rpm)	0.6667, 1.333, 2.0
Concentration of supporting electrolyte (g/lit)	0.6667, 1.333, 2.0
pH	2.5, 4.5, 6.5
Mercury concentration (ppm)	30, 50, 100
Applied potential (V)	6,9,12
Agitation (rpm)	200, 400, 600

TABLE (2) Effect of electrolyte concentration on the (%) removal efficiency of mercury with Time.

<b>Time (min)</b>	<b>(%) Removal efficiency of mercury</b>			
	<b>Without NaCl</b>	<b>0.6667 g NaCl/lit.</b>	<b>1.333 g NaCl/lit.</b>	<b>2.0 g NaCl/lit.</b>
5	6.5	52.3	66	73
10	7	56.8	73	79.5
15	26.5	59.4	76	81
20	29	61.5	77	84.5
25	32.5	62.5	81.5	91
30	41	66.5	83.5	92
40	44	68.8	90.5	92.5
50	49	70.5	98.5	95
60	49	54.6	92.5	89

TABLE (3) Effect of initial concentration on the (%) removal efficiency of mercury with Time

<b>Time (min)</b>	<b>(%) Removal efficiency of mercury</b>		
	<b>30 ppm</b>	<b>50 ppm</b>	<b>100 ppm</b>
5	64.6	66	71.4
10	66.9	73	73.2
15	70.2	76	73.8
20	71.2	77	76.5
25	74.3	81.5	78.4
30	75.4	83.5	82.9
40	78.4	90.5	87.6
50	64.3	98.5	92.5
60	69.2	92.5	74

TABLE (4) Effect of applied potential on the (%) removal efficiency of mercury with Time

<b>Time (min)</b>	<b>(%) Removal efficiency of mercury</b>		
	<b>6 Volt</b>	<b>9 Volt</b>	<b>12 Volt</b>
5	63.5	66	50
10	66	73	53
15	68	76	56
20	69.5	77	56.2
25	71	81.5	60
30	72.5	83.5	64.4
40	75	90.5	65.5
50	85.5	98.5	66.5
60	66.5	92.5	49

TABLE (5) Effect of agitation on the (%) removal efficiency of mercury with Time

<b>Time (min)</b>	<b>(%) Removal efficiency of mercury</b>		
	<b>200 RPM</b>	<b>400 RPM</b>	<b>600 RPM</b>
5	42	66	46.5
10	43.6	73	49
15	47.4	76	51
20	48.5	77	52
25	49	81.5	52.5
30	50.5	83.5	55.5
40	55	90.5	56.5
50	68	98.5	93
60	64.5	92.5	57.5