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Electrocoagulation process for the removal of Chromium and Cadmium from synthetic Industrial Wastewater

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Abstract

Electrocoagulation (EC) is an alternative and green technique for wastewater treatment due to of its high removal efficiency, lack of chemical requirements, use of simple devices, lack of secondary pollution, etc. The aim of the present work is to investigates optimum conditions for achieving simultaneous removal of chromium and cadmium ions from synthetic industrial wastewater by EC. The present work investigated the performance of EC in removal of chromium and cadmium via studying the effect of existence of anions (SO_4^{2-} and Cl^-), the existence of other cations (Ni^{2+} – Zn^{2+} – Cu^{2+}), electrode surface area and shape and reactor cross section. In addition, the use of two plates as anode or cathode has been investigated. Results showed that the larger the surface area of anode the better removal efficiency of Cr and Cd ions, however increasing the cathode area caused slight increase in the removal process. Having sulfate anion, the removal percent reached nearly 95% and 91% for Cr and Cd ions respectively within half an hour. The existence of Ni reduced the Cr removal efficiency decreased from 98% to 85% in existence of Ni ions , and from 98% to 75.30% in existence of Zn ions and from 98% to 70 % in existence Cu ions and 70%. Moreover, these cations minimized the Cd ion removal percent to the third. The best removal efficiency achieved was about 97 % and 88 % for Cr and Cd ions respectively. The used configuration was Fe/Al electrodes, 1.0A average applied current and solution with sulfate anions only after one hour, consequently. The study showed that use of EC is recommended for the removal of such heavy metals ions from industrial wastewater as chromium and cadmium.

1.Introduction

The world is entering into an age of sustainability. Nowadays, the freshwater available for human being represents only less than 1% while the salty water of seas and oceans is more than 97% of the total water on earth. Even worse, this small 1% part is now lessening due to of the growing pollution resulting from the industrial technologies, urban, agricultural and human activities, particularly in third world countries [1]. The wastewater, once generated, not only causes irreversible damage to the environmental balance but also contributing to the depletion of fresh water reserves, in this way creating a threat to the coming generations. Therefore, it is crucial to treat wastewater to achieve recovery and reuse. This wastewater contains a noteworthy concentration of heavy metal ions which has conducive to the accumulation of these toxic species in the ecosystem especially in living tissues [2].

Of these metals, Chromium waste Cr (VI), it is a kind of heavy metal that is deleterious to humans as well as to the environment as it is also more easily absorbed in the human body, especially the digestive tract in humans [3]. Several treatment processes including physicochemical techniques are performed such as adsorption [4], ion exchange [5], precipitation [6], membrane reserve osmosis [7]and electrocoagulation technology [8][9]. Furthermore, Cadmium produced from industrial effluents, like those from the smelting, mining, and electroplating industries, has boosted the severity of environmental issues due to its high concentration, bio toxicity and carcinogenicity [10]. Cadmium in solution is also highly water soluble and non-biodegradable in natural ecosystems. The permissible concentrations for chromium and cadmium discharged industrial wastewater that can be discharged in fresh water is less than 0.01 and 0.01 mg/L respectively, as proposed by The Egyptian Ministry of Health in law 1095 of 2011.

In the recent decades, several techniques, including chemical coagulation [6], biological degradation [11], adsorption [12][13], ion exchange [14], reverse osmosis [15], and membrane filtration [6] have been widely investigated in the treatment of toxic metals in wastewater. Nevertheless, these methods all have shortcomings like excessive cost, generation of severe secondary pollution, long run times and complexity.

Unlike the abovementioned methods, electrocoagulation (EC) is an alternative and green technique for various types of wastewater treatment due to its high removal efficiency, lack of chemical requirements, use of simple devices, lack of secondary pollution, etc.[10].

Coagulation is a process where the pollutants, suspended particles turn into sediment by collision with counter particles and get agglomerated to produce an insoluble agglomerate complex [2]. It is able to achieve by adding chemical coagulant like alum and iron compounds. The coagulant produced by the electrochemical process that is first proposed by Vik et al. is called electrocoagulation [16] .The most used electrodes in the process are iron, aluminum, mild steel and stainless steel. When these electrodes are subjected to the electric supply, the dissolution of anode results in coagulant formation in the system, this coagulant further leads to coagulation and flocculation. Thus effective pollutants removal becomes possible[2]. Treatment of wastewater by EC is 10 to 15% higher as compared with the conventional coagulation (for removal COD, turbidity, nutrients and TSS) [17] and also sludge generated from the EC is less as compared with the conventional coagulation [2]. Via this process, the coagulant dosage can be significantly decreased as compared with conventional methods [18], this process can also be used for reuse of wastewater and less sludge generation. The environmental compatibility, versatility, selectivity, energy efficiency, amenability to mechanization, safety and cost-effective are the add-on benefits of this process. EC is a potential technique for achieving these goals of sustainability [2].

The treatment of industrial wastewater containing heavy metals (Cr – Cu - Zn – Ni) was investigated by EC as the effect of current density, pH, electrode type and time was examined on the removal of these ions with COD. These study showed that iron electrode was the best for Cr and gave a relative adsorption for metal ions to be Cr>Cu>Ni>Zn[19]. In addition, other cations were removed using EC from hydrogen carbonate containing waters where the effect of HCO_3^- and Ca^{2-} ions on divalent iron and zinc removal was investigated using Al electrodes. The results were that treatment of HCO_3^- containing water is mainly due to adsorption of metal cations on $Al(OH)_3$ flocs formed; On the contrary , without this anion, OH^- generation raised pH profiles near the electrodes that gave additional removal of the metal cations by local formation of $Me(OH)_2$ solids[20].

As known in many studies the shape of electrodes was rectangular plates but there was cylindrical shape and these shapes were presented with some differences such as presenting the two shapes together or make openings in the electrodes so that they could reach the best removal efficiency, so we investigated the effect of increasing and changing anode and cathode area.

The aim of the present work was to investigat the effect of some anions (Cl^- - SO_4^{2-}) solution, the existence of some cations (Ni^{2+} - Zn^{2+} - Cu^{2+}) on the removal efficiency Cd – Cr from industrial wastewater. In addition some electrode characteristics, electrode surface area and shape and using more than one plate for electrode, have been examined. Finally, the reactor cross section shape has been investigated.

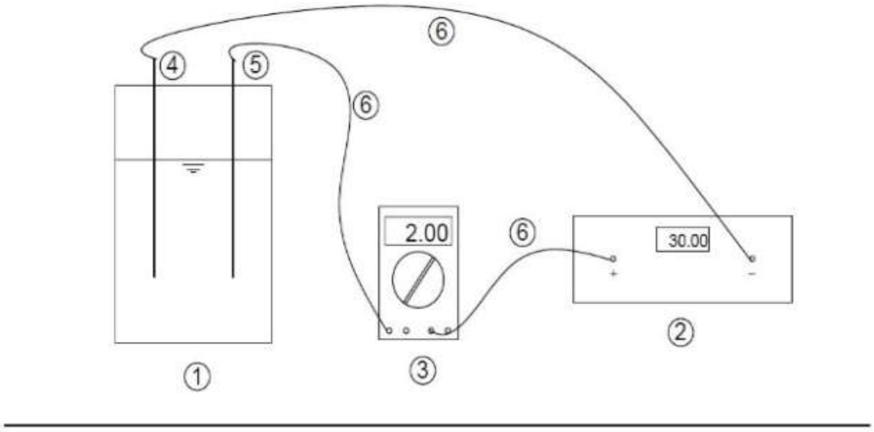
2. Material and Methods

2.1. Synthetic wastewater

The synthetic wastewater has been prepared dissolving stoichiometric amounts of $CrCl_3.6H_2O$ and $3CdSO_4.8H_2O$ in distilled water. These initial concentrations had been chosen to simulate real concentrations of heavy metals in industrial wastewater like tannery industries and dyeing industries as samples of chromium and cadmium was investigated with different concentrations, for chromium (560-280-140) ppm and cadmium (800-400-200) ppm, and the initial concentrations used in this paper were 280 for chromium and 400 for cadmium.

2.2.Electrocoagulation system

(Figure 1) shows the experiment set up which consists of a cell and electrical circuit. The cell consists of 12 L glass container (20*20*30 cm), plates with 105 mm width, 300 mm height and 1mm thick. The plates were chosen as iron and/or aluminum. The electrical source was 30-Volt D.C power supply with a voltage regulator and digital screen that display the voltage and the ammeter readings. The surfaces of the used electrodes were always cleaned with sand paper and submerged in diluted hydrochloric acid solution when the efficiency of the cell decreased to avoid the passivation of these electrodes and remove any impurities or adherent oxides on the surface of the electrodes.



1-Glass reactor
2- A 30-V digital DC power supply
3- Ammeter
4-Cathode electrode
5- Anode electrode
6- Connections

Figure1: Components of the electrocoagulation reactor.

Many parameters have been investigated for better understanding of the EC techniques to best remove heavy metal ions (chromium and cadmium). These parameters include existence of other cations (Ni^{2+} - Zn^{2+} - Cu^{2+}), type of anions (Cl^- – SO_4^{2-}). Also, the effect of surface area of electrodes, shape, the reactor cross area and using two plates as one electrode as anode or cathode have been investigated. Table 1 shows tests bases for different runs of the experimental works.

Table 1: The operating conditions taken for each parameter effect investigation

The conditions / The parameter	Applied Current	Average Voltage	Spacing between electrodes	Electrodes	Contact time	Initial Conc. (ppm)
The existence of other cations (Ni^{2+} - Zn^{2+} - Cu^{2+})	0.80 A	15 V	3.50 cm	Fe-Al	60 min	280 for Cr 400 for Cd 500 for all cations
Effect of the type of anions (Cl^- - SO_4^{2-})	1.0 A	20V	3.50 cm	Fe - Al	60 min	280 for Cr 400 for Cd
Effect of surface area of electrode	1.0 A	23 V	2.50 cm	Fe – Al	60 min	280 for Cr 400 for Cd
Effect of the cross section of reactor	12 mA/cm^2 (current density)	20V	3.50 cm	Fe – Al	60 min	280 for Cr 400 for Cd
Effect of using more than one plate as electrode	0.70 A	26 V	3.50 cm	Iron - Aluminum	60 min	500 for Cr 400 for Cd

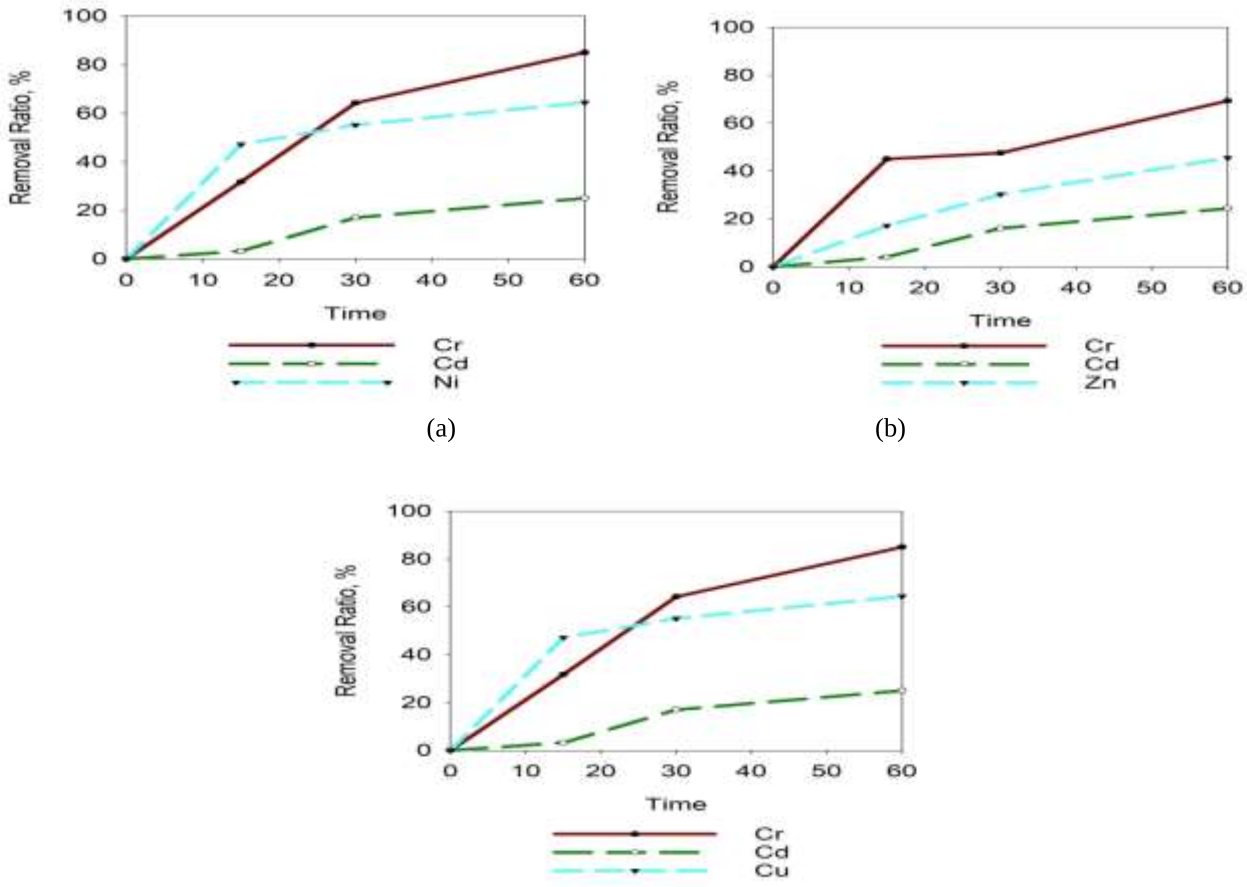
3. Results and discussion

3.1 Effect of the existence of other cations (Ni^{2+} - Zn^{2+} - Cu^{2+})

Industrial wastewater could involve numerous cations besides chromium and cadmium ions. In this part the effect of existence of other cations on the removal efficiency of Cr and Cd ions had been studied. These cations were nickel, zinc and copper ions. The experiments were implemented under the same conditions.

As presented in (Figure 2) , the results showed that the addition of another cation (Ni^{2+} - Zn^{2+} - Cu^{2+}) with chromium and cadmium affected the removal efficiency as the added cations in the solution with the same conditions consumed the coagulants and that decreased the removal ratios of chromium and cadmium ions, this clearly shown by comparing with (Figure 2) and (Figure 3) that shows removal efficiency of chromium and cadmium without any other cations.

In case of nickel, zinc and copper ions existence with chromium and cadmium ions, the removal of the chromium and cadmium ions declined but in varying proportions especially for chromium ions. The effect of Ni on Cr was less than Zn and Cu effect as the percentage of removal of chromium in existence of Ni decreases from [98% to 85%], from [98% to 75.30] % in existence of Cu and from [98%to 70 %] in existence of Zn .Also the results presented that the removal efficiency of Ni and Cu ions removal was better than Zn removal .The effect of Zn, Ni and Cu ions on cadmium ions was almost the same as the removal ratio dropped from [72 % to nearly 25 %]. The reason may be due to the similarity of the removal mechanism that could be adsorption, flocculation and co-precipitation. On the other hand, the priority of the removal via the contact time could be because of the solubility product constant (K_{sp}) of the metal hydroxides that determine the degree of solubility.



(a) (b)

(c)

Figure 2: Effect of the existence of other cation on with Cr and CdWith Ni (b) with Zn (c) with Cu

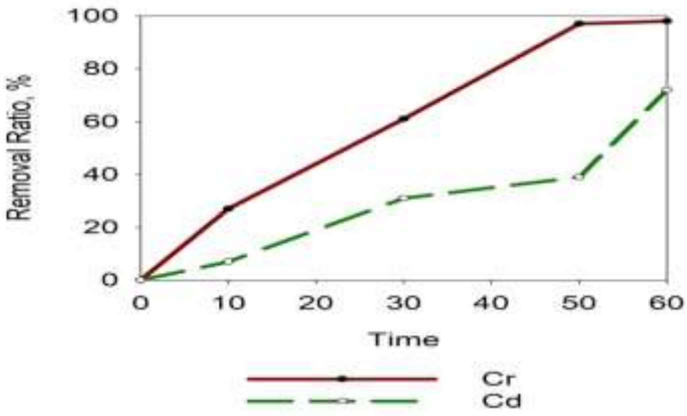
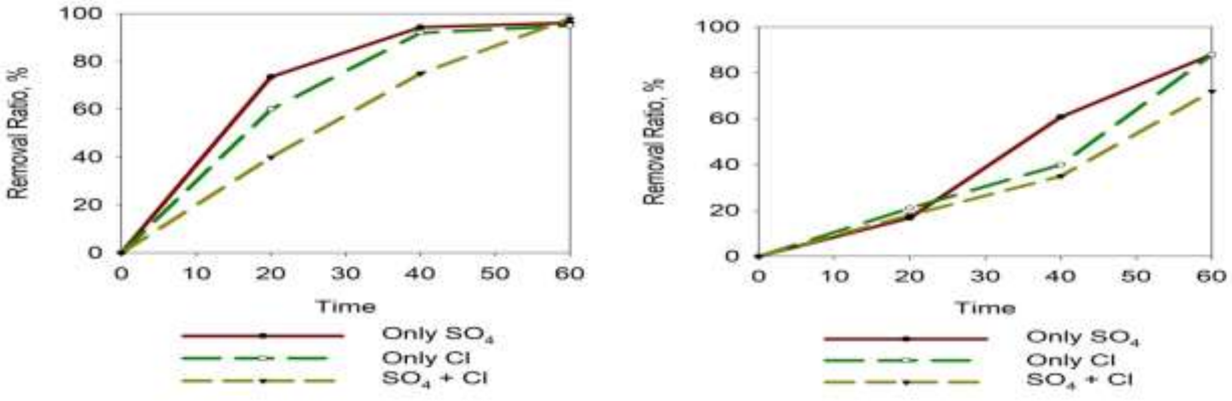


Figure 3: the removal efficiency for chromium and cadmium without any other cations

The higher value of (K_{sp}) is, the difficulty of aggregation between the cation and its hydroxides is, and hence that hindered the precipitation process and reduced the removal efficiency. As the values of chromium, copper, nickel, zinc and cadmium hydroxides are [$1*10^{-30}$, $4.80*10^{-20}$, $5.48*10^{-16}$, $3.0*10^{-17}$ and $7.20*10^{-15}$] respectively [21]. That means that the chromium has the priority to be removed as it has the minimum solubility product constant and cadmium always had the minimum removal ratio as it has the maximum solubility product constant and this appeared clearly in (Figure 2) as chromium ions had the higher removal efficiency in all cases and on contrary the cadmium had the lower efficiency. The reason of variation of the removal efficiencies of the metal hydroxides due to the different values of the solubility product constants that explained what happened here was the same between Cd and Zn ions removal at another study [22].

3.2 Effect of the type of anions (Cl^- - SO_4^{2-})

Real wastewater contains various anions that primarily come from different types of industries. Chloride and sulfate are considered from the largest anions in smelting wastewater. The effect of these anions has investigated before in many studies. In the EC process the existence of anions affect the conductivity of the solution, cell voltage and the consumption of energy in the EC cell. Herein, the effect of chloride and sulfate on the removal efficiency of chromium and cadmium was investigated with following simulating wastewater with constant parameters. To evaluate the effect of chloride and sulfate, the doses of chloride as (NaCl) and sulfate as (Na_2SO_4) were added to the solution. The experiments were divided according to the type of anion in the solution to three stages; First, the only anion existed was chloride anion, second stage with sulfate anion only and the last stage mixture between the chloride and sulfate anions. According to (Figure 4), the results indicated that the existence of sulfate was more effective to remove Cr and cd anions within the most of the time contact. On the other hand, for chromium the binary system reached to the best removal efficiency but needed more contact time than the case of sulfate only. Charge neutralization, precipitation and electronic double-layer compression may be considered the mechanisms for what happened in the case of different anions as mentioned at [23].



(a) (b)

Figure 4: Effect of the existence anions on removal of (a) Chromium (b) Cadmium

As mentioned earlier, the priority of chromium ions removal appeared clearly in (Figure 3) as the removal rate of cadmium ions started to increase clearly after 20 minutes after the most of chromium had been removed in cases of SO_4^{2-} only and Cl^- only. At these cases the sulfate was better. As mentioned, the binary system had better removal efficiency but with increasing contact time for chromium and that affected adversely the cadmium removal. In case of the binary system, for cadmium ions the removal rate increased clearly after the most of chromium ions had been removed after 40 minutes and accordingly the binary system had better removal efficiencies than chloride system and lower removal efficiency than sulfate system and this result agreed with the study that investigated the effect of anions of Cd, Zn and Mn ions and sulfate was better for cadmium, then the binary and the chloride in the last [9] and other study presented that removal efficiency of cadmium ions was increased by increasing sulfate anions [24].

To understand what happened it is necessary to identify what sulfate and chloride did in the solution. First, the existence of sulfate anion in the solution causes positive and negative effects. For the positive side, the SO_4^{2-} anions in the solution generated more OH^- ions at the cathode in the solution according to the (Equation 3, 4). Also, due to the stronger charge neutralization of divalent SO_4^{2-} solution system, more proportioned Fe^{2+}/Fe^{3+} hydroxides were generated and turned into bigger specific surface area flocs than without sulfate concentration and that activated and accelerated the destabilization and precipitation process[25].

On the other hand, SO_4^{2-} an anion causes anode passivation that impedes the dissolution of the electrode then limited the removal efficiencies of the heavy metals ions. In addition, the reverse voltage dropped significantly as a result of electrode passivation. Also it was noticed an increase in current consumption, and that increased the energy consumption which agreed with what proved that sulfate anions in the solution which raises the voltage so that it can overcome the passive film and this leads to waste electrical energy [26].

$$SO_3^{2-}{}_{(aq)}+H_2O_{(l)}\rightarrow HSO_3^-{}_{(aq)}+OH^-{}_{(aq)} \quad (3)$$
$$HSO_3^-{}_{(aq)}+H_2O_{(l)}\rightarrow H_2SO_3^-{}_{(aq)}+OH^-{}_{(aq)} \quad (4)$$

With regard to chloride anions, it has an adverse effect for the passivation that sulfate anions cause as chloride anions are used as effective means against passivation in the EC process and converts it to soluble compounds (Equation 5, 6), then reduced the electrical power consumption [27] [28]–[30]. And it is worth mentioning that the current decreased through the contact time and also the flotation process was more than sulfate anion case as an indicator of chloride anion generation and that appeared clearly in the present work as the removal process by flotation was clearly more than sedimentation process.

$$Fe(OH)_{2(s)}+Cl^-\rightarrow Fe(OH)Cl_{(aq)}+OH^-{}_{(aq)} \quad (5)$$
$$Fe(OH)_{2(s)}+2Cl^-\rightarrow FeCl_{2(aq)}+2OH_{-(aq)} \quad (6)$$

Also, the existence of hypochlorite (OCl^-) produced in Cl^- ions (Equation 7- 9) solution contributed in the removal of metal [31].

$$2Cl^-{}_{(aq)}\rightarrow Cl_2(g)+2e^-{}_{(aq)} \quad (7)$$
$$Cl_{2(g)}+H_2O_{(l)}\rightarrow HOCl^-{}_{(aq)}+H^+{}_{(aq)}+Cl^-{}_{(aq)} \quad (8)$$
$$HOCl^-{}_{(aq)}\rightarrow OCl^-{}_{(aq)}+H^+{}_{(aq)} \quad (9)$$

On the other hand the existence of Cl- consumed OH- and decreased the pH of the solution according to the last equations and when the pH of the solution decreased the efficiency of the aluminum as a cathode decreased and this was on the contrary of the existence of sulfate. Also the high concentration of the Cl- had an adverse influence on the adsorption capacity of flocs as it contributed to generate weak stability products [27]. Finally, to take the most advantage of the existence of anions, it is necessary for chloride to exist with sulfate to minimize its adverse effects and gain better performance of EC process. The results agreed with other studies that examined the influence of anions existence in the solution on the removal efficiencies of heavy metal ions (for cadmium and zinc) [22], it was observed that both chloride and sulfate were better for zinc removal and sulfate was better for manganese removal but it was better to use chloride with sulfate to decrease negative effect of sulfate [32].

3.3 Effect of surface area of electrode

In the present work the effect of the surface area of the electrode was investigated as many researchers studied the efficiency of EC on removal of various pollutants using rectangular electrodes and some studied cylindrical electrodes [33][34]. Herein, various shapes were used, solid cylindrical electrode and mesh cylindrical electrode. The effect of changing of surface area was investigated by in two steps first changing of cathode area with the same anode and second changing of anode area with the same cathode. The experiments were performed with constant conditions.

For the same anode that was cylindrical with surface area 125.68 cm^2, the cathode was cylindrical with areas (348.60 cm^2 for solid and 695 cm^2 mesh one) as shown in (Figure 5). The mesh was with 2.50 *2.50 cm opening. For the same areas but the charge of electrodes was reversed they were used to investigate the effect of changing anode area for the same cathode area.

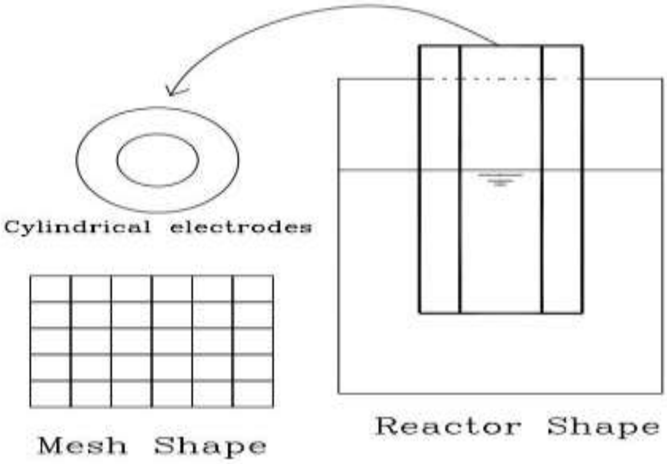
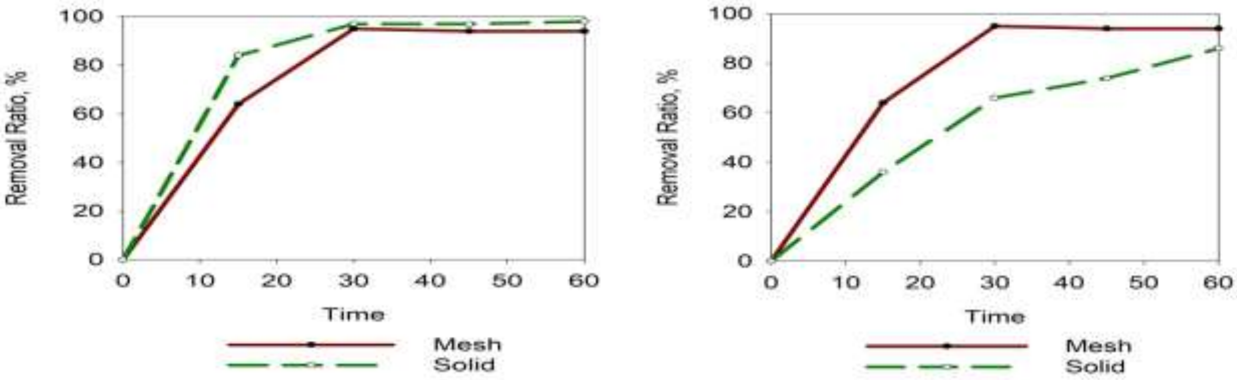


Figure 5: Schematic diagram of the cylindrical reactor.

The results shown in (Figure 6) presented the removal efficiency of chromium and cadmium ions in case of changing surface area of cathode with the same anode.

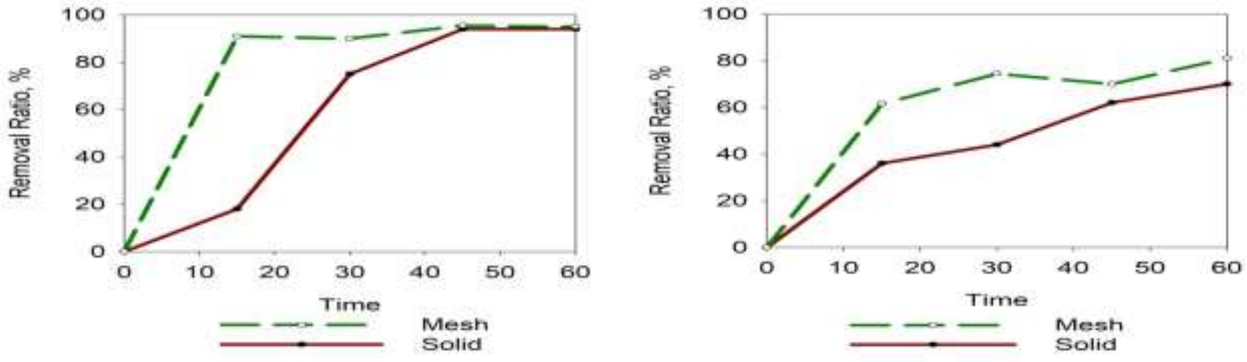
From the results it is clear that the increase of surface area of cathode increased the removal efficiency slightly and this may be due to the essential electrode in the removal process is the anode as anode is the responsible for coagulants generation and the anode was the same in the two cases. However, the slight increase in removal efficiency may be due to the increase of surface area where the reduction of water happened that caused the generation of hydroxides ions that helped metal hydroxides generation so it boosted the removal process slightly as shown in the results.



(a) **(b)**

Figure 6: Effect of the Changing Cathode area on removal of (a) Chromium (b) Cadmium

On the other hand, for investigating the changing of anode area, the results shown in (Figure 7) presented the removal efficiency of chromium and cadmium ions in case of changing surface area of anode with the same cathode.



(a) (b)

Figure 7: Effect of the Changing Anode area on removal of (a) Chromium (b) Cadmium

From the results it is clear shown that as the anode surface area increased, the removal percent increased due to generation of coagulants was according to (Equation (2)). Hence, high removal efficiency could be achieved faster. For mesh anode whose surface area was with high removal efficiency of Cr ions reached 90 % at 15 min and it became approximately constant after that.

On the other hand for cadmium it exceeded 60 % at 15 min better than the case of solid anode with 36 % removal ratio within the same period.

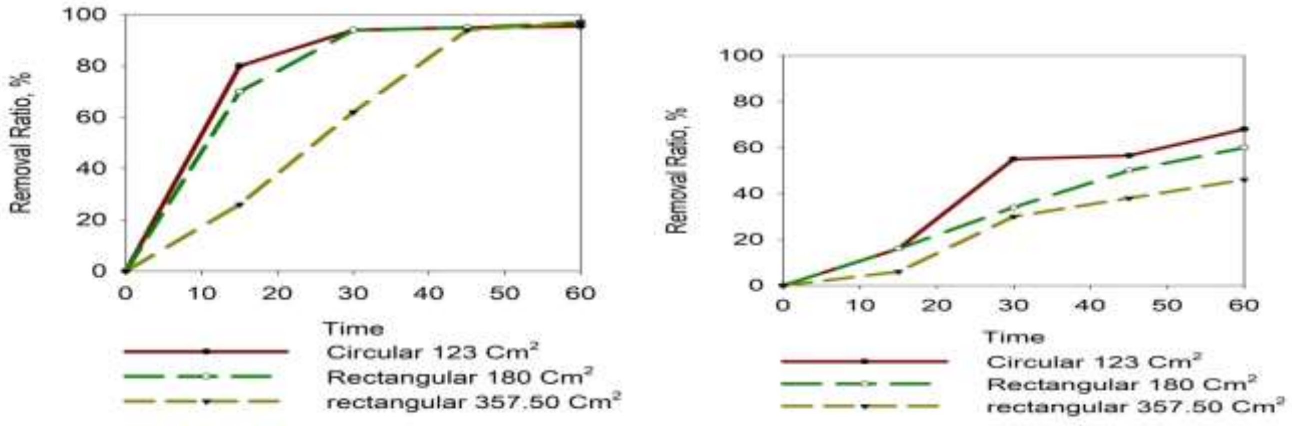
It is also obvious in the results that the way of removal of chromium and cadmium ions was approximately the same for two cases (mesh and solid anode) as the relation between removal efficiency and time is approximately the same with one different that was the removal rate which was related to the coagulants generation.

3.4 Effect of the cross section of reactor

The cross section of the reactor was investigated to determine its effect on the removal efficiency for chromium and cadmium ions. The shapes of the cross section tested were rectangular (surface area 357.50 and 180 cm^2) and circular (surface area 123 cm^2). The initial concentration of heavy metal and the current density were the same during the experiments. The results showed in (Figure 8) that the circular shape with smaller cross section area was better than rectangular shape as after 30 min.

the removal ratios were [95% and 55 %] for chromium and cadmium ions respectively in case of circular shape but were [61% , 95%] and [31%, 35 %] for chromium and cadmium ions respectively in case of rectangular one.

The reason of that may be due to the reactor surface area ,as in case of circular shape , the surface area was 122.72 cm^2 but for rectangular one, it was 357.20 cm^2 that was larger and this means the pollutants in the solution were near from the electrodes in case of circular one and this maximized the ability to remove pollutants. In addition, the flotation process was high especially in the first stage of the process and this appeared clearly in this work. In the other hand, for rectangular shape when the cross section area became 180 cm2 the removal efficiencies achieved higher values and that confirms that smaller area around electrodes helped energizing removal efficiency. At 45 min, the current density in case of circular decreased to 8 mA/cm^2 due to decreasing the effective depth of the electrode in the solution due to the accumulation of floating pollutants around the electrodes due to the narrow surface area, but on the contrary the current density was still constant in case of rectangular shape. And that explained the results convergence in this part

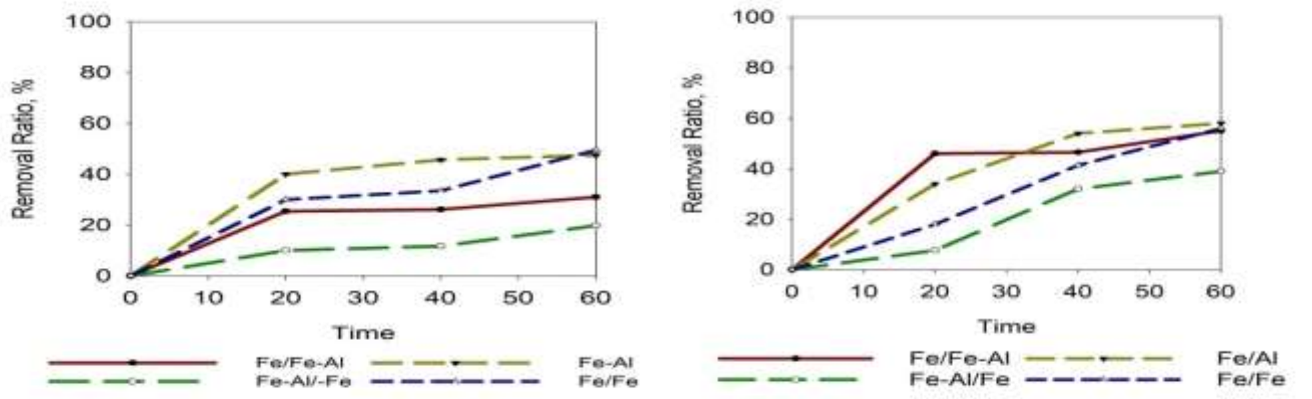


(a) (b)

Figure 8: Effect of the Changing reactor Cross section on removal of (a) Chromium (b) Cadmium

3.5 Effect of using more than one plate as electrode

In another context to examine using two plates with different material as one electrode, four runs were implemented. First run was anode (Fe) – cathode (Fe & Al). Second run was anode (Fe & Al) – cathode (Fe) as the two plates in the two trials were connected together using a nail. Third one was anode (Fe) – cathode (Al) and last one was anode (Fe) – cathode (Fe). The connection between plates can be performed with two types monopolar, bipolar as the first configuration is a monopolar electrode configuration, i.e. each electrode is connected to either the anode or the cathode, and the second one is bipolar and thus involves connecting only the first electrode to the anode and the last electrode to the cathode[35][36]. In the present work the configuration was monopolar but with two differences, first only one electrode with two plates and second was no distance between two plates used for the same electrode. The results in (Figure 9) showed that the best removal efficiency was by using single plate as anode and cathode that were Fe and Al for chromium and cadmium removal and that could be due to of high resistance of using two plates as one electrode that consume more energy and that appeared in the experiments as using two plates in anode or cathode consumed more current during the experiment.



(a) (b)

Figure 9: Effect of using two plates for one electrode (a) Chromium (b) Cadmium

Using more than one plate increased the removal ratio but needed high current density and therefore more energy consumption[36]. Also, it was found that iron as an anode was always the best when it was used alone for removal chromium and cadmium ions and using aluminum with it decreased the removal efficiency as it affected the coagulates concentration.

Conclusion

Based on the test results and limited to the study conditions and range of values of the parameters considered in this study, the following points can be drawn.

1. The existence of other cations (Ni^{2+} – Zn^{2+} – Cu^{2+}) in the wastewater reduced the removal efficiency as they consumed coagulants with chromium and cadmium ions.
2. The presence of sulfate anion in the solution was better for chromium and cadmium removal than chloride only or the existence of sulfate and chloride together but with electrode passivation that consumes more energy so, it is better to use chloride with sulfate to overcome its disadvantage.
3. Increasing the surface area of anode with the same cathode area increased the removal ratio significantly, but removal efficiency increased a little in case of increasing cathode area with the same anode area.
4. Smaller cross section of the reactor was better than larger one, as rapprochement of contaminants from anodes increased the removal efficiency.
5. With the same operating conditions, increasing the number of plates for one electrode achieved lower removal efficiency than using one plate for each electrode.

Finally, we can say that this study concluded that Electrocoagulation is an eco-friendly and effective method for treatment of industrial wastewater containing Cr and Cd ions; however, a similar study using real wastewater is needed to go forward towards its implementation at the industrial scale

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