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A comparison of the effectiveness of electrocoagulation to coagulation processes using ferric chloride for the removal of cadmium from aqueous solution

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ABSTRACT

Cadmium removal from industrial wastewater due to its stability and accumulation properties is essential before discharge to the environment or a sewage collection network. The aim of this study was the comparison of the effectiveness of the removal of cadmium from industrial wastewater by electrocoagulation process, to a coagulation process. The effect of variables such as pH, concentration of ferric chloride coagulant and sedimentation time in a coagulation process and variables such as pH, retention time, voltage and distance of the electrodes in the electrocoagulation process by iron electrodes in the removal of cadmium were examined. All experiments were repeated three times. Optimal conditions were tested on real samples. The efficiency of each process was analyzed by using analysis of variance and regression. A cadmium removal efficiency 31.3% was obtained in the coagulation process in optimal conditions of pH 7, coagulant concentration 9 g/L and sedimentation time of 50 min. Alternatively, the cadmium removal efficiency of 99.12% was obtained in the electrocoagulation process in optimal conditions of pH 9, voltage difference 36 V and electrode distance 1.5 cm. In the real sample and optimal conditions, the cadmium removal for coagulation and electrocoagulation obtained was 21.72% and 88.2%, respectively. There was a significant difference between the two tested processes. By increasing pH, voltage and retention time, the electrocoagulation process had more efficiency in cadmium removal. Also, in the coagulation process increasing the coagulant concentration and the retention time in neutral pH showed less efficiency than the electrocoagulation process.

Keywords: Electrocoagulation, Coagulation, Cadmium, Ferric chloride, Electrode

1. Introduction

Heavy metals exist in natural form in water resources or enter by pollution [1]. Major human pollution sources are mining, disposal of untreated or semi-refined wastewater containing heavy metals, and the use of fertilizers containing heavy metals [2,3]. The maximum acceptable concentration of cadmium ions in drinking water according to World Health Organization guidelines is 3 μg/L [4].

Heavy metals such as cadmium are used widely in some industrials such as soldering and battery making [5]. Extensive research in the field of removal of heavy metals, industrial chemical materials, microorganisms, hardness and environmental pollutants by electrocoagulation process from aqueous solutions has been done [6–8]. In previous studies, several ways have been suggested to remove cadmium from industrial wastewater and aqueous solutions. Some popular processes to remove heavy metals such as reverse osmosis, ion exchange, oxidation, adsorption, etc. from industrial wastewater are applicable only for limited concentrations of pollutants because of high cost of

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treatment, need for additional treatment, formation of dangerous side products and low efficiency [9–12].

Interest in using the electrocoagulation process in water and sewage treatment is increasing and this technology is considered as a new method [13]. It can be noted to be environmentally friendly, with no need for special chemicals [14], having easy maintenance, with fully automatic and continuous exploitation, recycling and reuse of water, effective and rapid decomposition of organic matter with an efficiency close to 90%, and the production of colorless and odorless sewage being noted as benefits of the electrocoagulation process [15–17]. In the electrocoagulation process coagulant has been produced by anode oxidation [18]. Metal ions produced at the anode electrode react with hydroxide ions produced at the cathode and insoluble metal hydroxides are produced [19,20]. Lastly, formed coagulums on the basis of density will removed from the solution by using flotation or sedimentation processes [21,22]. The effectiveness of the electrocoagulation process has been investigated in several studies. Malakootian et al. [23,24] used electrocoagulation to remove hardness and paint from water [25].

Coagulation and flocculation processes can be used in the treatment of some wastewaters which contain heavy metals [26]. The importance of these processes in water and wastewater is clear, and research on the identification of suitable coagulants and determining their effectiveness in toxic removal is essential [27]. Torabian et al. [28] showed in their study that the use of coagulation, flocculation and chemical precipitation with sodium bisulfite regenerative material, and co-coagulant materials such as ferric chloride and lime, have suitable efficiency toward removing heavy metals from industrial wastewaters.

The aim of this study was to evaluate and compare performances of electrocoagulation process with coagulation process in cadmium removal from aqueous solutions. During the electrocoagulation process some parameters such as pH, retention time, voltage and electrode distance were evaluated; and during the coagulation process some parameters such as pH, retention time and concentration of ferric chloride coagulant were evaluated.

2. Experimental setup

This study is experimental and was done in the Environmental Health Engineering Research Center of Kerman University of Medical Sciences from September 2015 to March 2016. Stock solution with 1,000 mg/L concentration of cadmium was made from cadmium nitrate. Standard solutions of cadmium nitrate were made in the concentrations of 50, 40, 30, 20 and 10 mg/L. Removal of cadmium from an aquatic environment and real samples with an initial cadmium concentration of 50 mg/L were evaluated by using both electrocoagulation and coagulation processes.

2.1. Coagulation process

The coagulation process was done by use of a Jar Test (VELP Scientifica, Italy) model 199,142. Samples in the fast mixing time of 1 min at 200 rpm and the slow mixing time of 20 min at 30 rpm were pretreated under conditions of pH (4–9), concentration of ferric chloride coagulant (3, 5, 7, 9, 11 and 15 g/L) and time (5, 10, 25 and 50 min). In order to measure remaining concentrations of cadmium, measurements were taken from 2 to 3 cm below the samples' surfaces.

2.2. Electrocoagulation process

In another part of the study, in order to prepare a reactor a cylindrical Plexiglass tank was produced with an effective volume of 250 mL. Two iron electrodes with dimensions of 11 × 2.5 cm, thickness of 0.3 mm, degree of purity more than 98.5% and effective surface area of 30 cm^2 were used. The electrodes in parallel state were connected to a power supply with a voltage of 5–40 V to convert alternating current to direct current. To measure the flow characteristics a multimeter model AKB-DT9208A was used. A schematic of the batch reactor used in the electrocoagulation process is shown in Fig. 1.

pH (4–9), reaction time (5, 10, 25 and 50 min), potential difference (8, 16, 23, 30 and 36 V) and electrode distance (1, 1.5, 2 and 3 cm) were examined. After each test period the electrodes were washed by hydrochloric acid (1 + 1) for 10 min and then by distilled water, in order to clean the surface of the electrodes [24]. The samples were taken from the middle reactor to measure the remaining concentration of cadmium. To adjust pH in both processes solutions of HCl 0.1 M solution and NaOH 0.1 M were used. pH in all samples before optimization was 7. The changing of pH was evaluated in each test period. The remaining cadmium nitrate concentration was measured by an atomic absorption spectrometer, model YOUNGLIN AAS 8020, equipped with system flame and stove. Before sample injection to the atomic absorption device, injection of standards calibration solutions was done from least concentration to the highest concentration, respectively. Optimum conditions were determined in both the coagulation and electrocoagulation processes in synthetic samples.

A comparison of the effectiveness of removal of cadmium from battery industrial wastewater by electrocoagulation

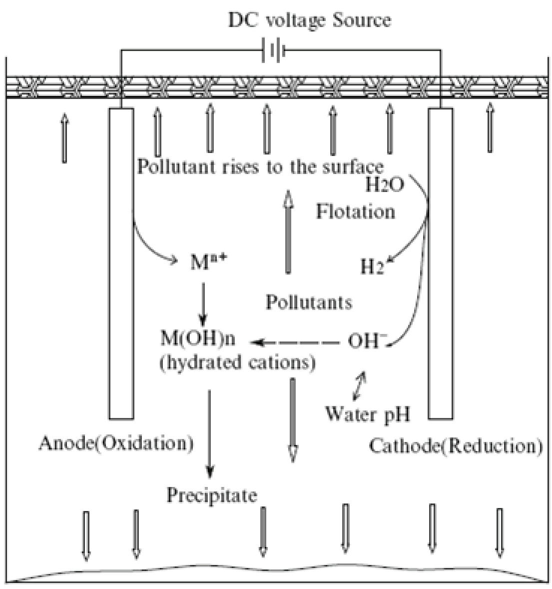

Fig. 1. Schematic of the electrocoagulation batch reactor.

and coagulation processes under optimum conditions, which previously was tested in the quality of pH, cadmium concentrations, biochemical oxygen demand (BOD), chemical oxygen demand (COD), and total dissolved solids (TDS) was done. Data were analyzed by SPSS software, and also the ANOVA and the regression tests were done.

3. Results and discussion

3.1. The effect of pH on cadmium removal

Coagulation processing depends on various parameters such as pH, retention time, concentration, the type of coagulant and the initial concentration of contaminant [29].

pH is an effective parameter in chemical reactions. In the primary pH environment, due to the processes used, pollutants are different [30]. The effect of pH on both results of the cadmium removal processes is shown in Fig. 2.

3.1.1. The coagulation process

The remaining cadmium concentration in pH 4, 5, 6, 7, 8 and 9 was obtained at 35.86, 37.37, 37.06, 31.82, 37.90 and 35.67 mg/L, respectively. The maximum efficiency of cadmium removal in the coagulation process by the coagulant ferric chloride was obtained at pH 7. The pH in each test period was increased from 7 to 8.33, 8.42, 8.70, 9.23, 8.17 and 8.15, respectively. pH and the coagulant concentration are the most important factors for the coagulation process. The optimal pH of 7 had more efficiency in leading flock formation. According to the results the removal efficiency obtained was 36.34%. In the coagulation process, the pollutant isolates from wastewater by adsorption and load neutralization and density techniques. In this study, coagulation, adsorption, load neutralization and density are in highest level to forming iron hydroxide complexes in the neutral pH range. Studies done by Song et al. [31] in Syria indicated hexavalent chromium in pH was near to neutral in tannery industrial sediment. Boumechhour et al.'s [32] reviews in Algeria showed that pH in the range of 6–7 was the optimal pH to remove the waste leachate COD by ferric chloride coagulant. That confirms the results of this study.

3.1.2. The electrocoagulation process

The remaining cadmium concentration in pH 4, 5, 6, 7, 8 and 9 was obtained, at 40.21, 39.92, 38.18, 37.30, 31.28 and 10.63 mg/L, respectively. In this process, the highest cadmium removal was obtained at pH 9 with a removal efficiency of 78.72%. The pH in each test period was increased from 11.35 to 11.42, 11.80, 12.25, 12.28 and 12.81, respectively. In the electrocoagulation process, by reducing the acidity of samples from 4 to 9 removal efficiency increased considerably from 19.57% to 78.72%. In this process, cations will be produced by anode oxidation. Cations react with hydroxide produced at the cathode. The formation of insoluble hydroxide cations will happen in alkaline conditions. Therefore, in high pH, the cadmium removal efficiency will increase by cation hydroxide flocks. Finally, the produced flocks based on density will settle or will float by hydrogen gas generated from cathodes. Akhondi et al.'s [17] study in Tehran have shown that pH has an effect on the solubility of products.

3.2. The effect of retention time on the removal of cadmium

Results of the comparison of the effects of retention time on both cadmium removal processes are shown in Fig. 3.

3.2.1. The coagulation process

The remaining cadmium concentrations at 5, 10, 25 and 50 min were 39.30, 38.15, 37.43 and 34.50 mg/L, respectively. The maximum efficiency of cadmium removal in the coagulation process was obtained at 31% in retention time of 50 min, since more time for absorption and sedimentation clotting produces maximum removal rates. The increasing retention time had no effect on increasing cadmium removal. Coagulation and sedimentation directly depend on the concentration of pollutants, concentration of coagulant and laboratory conditions.

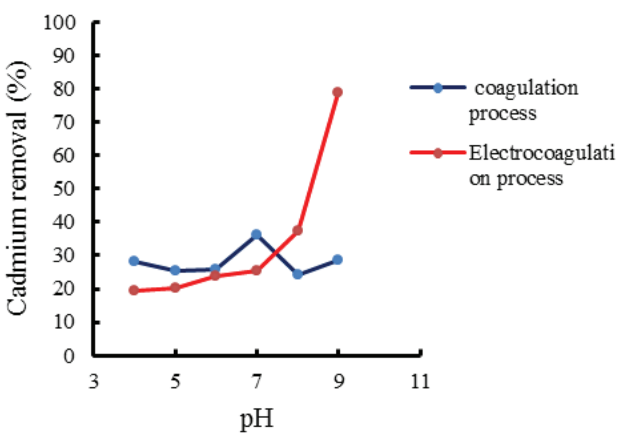

Fig. 2. Comparing the effect of pH on the cadmium removal in coagulation processes (0.3 g coagulant and retention time 20 min) and electrocoagulation processes (16 V, retention time 5 min and distance electrodes 1 cm) and initial cadmium concentration 50 g/L in both processes.

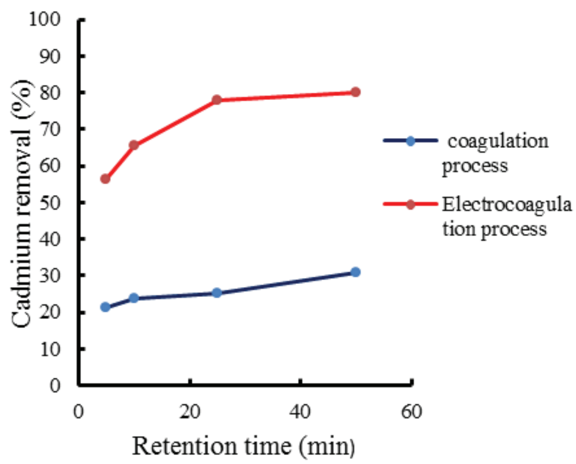

Fig. 3. Comparing the effect of retention time on cadmium removal by coagulation process (coagulant mass 0.3 g and optimal pH 7) and the electrocoagulation process (voltage 16 V, optimal pH 7 and the electrode distance 1 cm).

Movahedian and Salehi [33] showed that the cyanide chemical sedimentation process by ferrous sulfate needs 60 min.

3.2.2. The electrocoagulation process

The remaining cadmium concentrations in retention times of 5, 10, 25 and 50 min were obtained at 21.87, 17.26, 11.07 and 10.00 mg/L, respectively. The maximum efficiency of cadmium removal in the electrocoagulation process was obtained at 80% in retention time of 50 min. But the retention time of 25 min was chosen as optimal, with 77.84% cadmium removal. It can be concluded that this increased removal efficiency up to 25 min retention time was appropriately effective, with a direct and linear relationship. But a retention time of more than 50 min did not produce significantly more removal. Also, aside from consuming more energy, more electrons are consumed. The results of Mahvi et al.'s [34] study that evaluated electrocoagulation process efficiency in removing heavy metals copper, zinc and cobalt from landfill leachate in Tehran support this study's results. Akhondi et al. [17] studied the efficiency of the electrocoagulation process on the removal of cadmium heavy metal in aquatic environments. They succeeded at a retention time of 25 min with an efficiency of 96.7% cadmium removed. Increased removal efficiency by increasing the retention time in the electrocoagulation process was shown by Aoudj et al. [35] in the application of the electrocoagulation process in textile industry wastewater treatment examinations.

3.3. The effect of coagulant dose and voltage on cadmium removal

The results of the effect of coagulant dose on cadmium removal in the coagulation process and the effect of voltage in removal of cadmium by electrocoagulation process are shown in Fig. 4.

3.3.1. The coagulation process

The remaining cadmium concentrations in coagulant concentrations of 3, 5, 7, 9, 11 and 15 g/L were obtained at 38.40, 37.59, 37.53, 35.44, 37.39 and 42.81 mg/L, respectively.

The ferric chloride coagulant in concentration 9 g/L had the best efficiency of cadmium removal, at 29.11%, since higher concentrations have a negative effect on the effectiveness of the process of cadmium removal because of increasing of the opposed charge. In low concentrations, the only mechanism to destabilize colloidal particles is neutralization and other mechanisms such as sweep flocculation, because at low concentration formation of flocks will not be effective by using coagulation processes. Jonidi et al. [27] in Tehran reported a concentration of 7 g/L as the optimal concentration of ferric chloride for simultaneous cyanide and chromium removal. They concluded that higher concentrations than 7 g/L do not have an important effect on simultaneous cyanide and chromium removal.

3.3.2. The electrocoagulation process

The remaining cadmium concentrations at voltages of 8, 16, 23, 30 and 36 V were obtained at 18.80, 13.83, 13.36, 10.64 and 10.00 mg/L, respectively.

Voltage is one of the most important parameters in the electrocoagulation process [36]. So that, in addition to directly affecting efficiency, it is determinative in the amount of coagulant reaction [37]. In other words, in the electrocoagulation process coagulant is produced by oxidation of anodes. Iron electrodes by producing ferric hydroxide will cause cadmium removal and flock formation. The anode potential difference in the electrocoagulation process affects the rate of coagulant (ferric hydroxide) produced. In the electrocoagulation process potential difference was evaluated against the concentration of coagulant consumption in the coagulation process. The efficiency of cadmium removal with the potential difference equal to 36 V, 80% removal was obtained, more than at other potential differences. In high potential difference, the electrocoagulation process efficiency in cadmium removal was more, because the production of ferric hydroxide increased. With a change of voltage from 8 to 36 V removal efficiency increased from 62.40% to 80%. The result of Malakootian and Heydari [24] supports

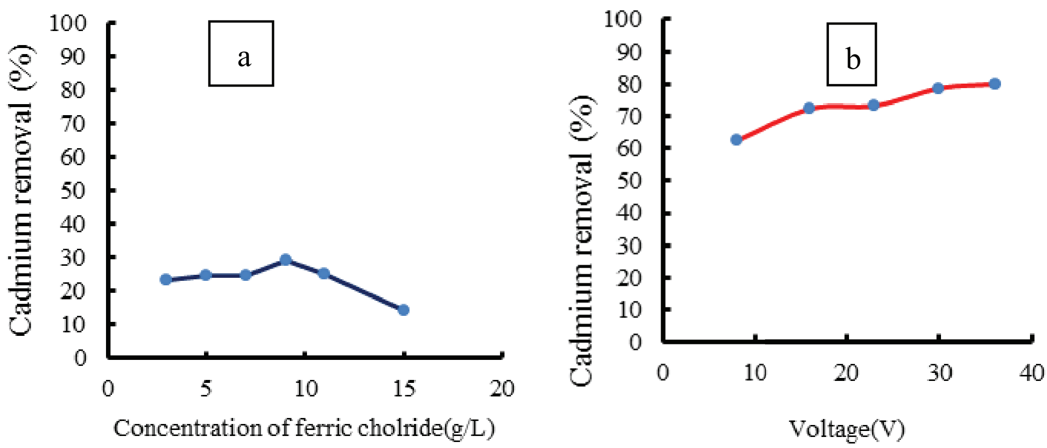

Fig. 4. (a) The results of the effect of coagulant dose on cadmium removal in the coagulation process (optimal time 50 min and optimal pH 7). (b) The results of the effect of voltage dose on cadmium removal in the coagulation process (optimal time 25 min, optimal pH 9 and the electrodes' distance 1 cm).

Table 1
The means of pH, Cd, BOD, COD and TDS in real samples of battery industrial wastewater

Samples	TDS (mg/L)	COD (mg/L)	BOD (mg/L)	Cd (mg/L)	pH
Raw wastewater	920 ± 16.32	376 ± 12.34	60 ± 1.07	52 ± 0.30	6.15 ± 0.27
After the coagulation process	303.6 ± 3.26	209.808 ± 2.30	36.89 ± 0.32	40.7056 ± 0.10	7.3 ± 0.20
After the electrocoagulation process	36.524 ± 3.21	20.68 ± 1.02	4.02 ± 0.02	6.0994 ± 0.20	10.28 ± 0.30

the results of this study. Emamjomeh and Sivakumar [21] in their study in Qazvin to evaluate nitrate removal by using an electrocoagulation process in an aquatic environment also achieved similar results.

3.4. Evaluation of optimal conditions in coagulation process

In the coagulation process under optimal conditions such as coagulant concentration 9 g/L, pH 7, sedimentation time 50 min, a maximum cadmium removal efficiency of 31.3% was obtained.

3.5. Evaluation of optimal conditions in the electrocoagulation process

In the electrocoagulation process under optimal conditions such as pH 9, potential difference 36 V, retention time 25 min and electrodes' distance 1.5 cm, a maximum cadmium removal efficiency of 99.12% was obtained.

3.6. Evaluation of optimal conditions for the real solution

The real quality of battery industrial wastewater regarding pH, cadmium concentration, BOD, COD and TDS is shown in Table 1.

The maximum cadmium removal obtained in real samples by electrocoagulation and coagulation processes in optimum conditions was 21.72% and 88.27%, respectively.

The maximum removal efficiencies of BOD, COD and TDS obtained in real samples by electrocoagulation in optimum conditions were 93.3%, 94.5% and 96.03%, respectively.

The maximum removal efficiencies of BOD, COD and TDS obtained in real samples by coagulation in optimum conditions were 38.5%, 41.3% and 67%, respectively.

In the coagulation process low turbidity and no primary cores to form clots were the reasons for the formation of weak larger clots and no sedimentation in considered time, which leads to a reduced process efficiency [27].

There was a significant difference between the two tested processes. By increasing pH, voltage and retention time the electrocoagulation process had more efficiency in cadmium removal. Also, in the coagulation process increasing the coagulant concentration and the retention time in neutral pH showed less efficiency than the electrocoagulation process.

4. The effect of distance of internal electrodes on cadmium removal

In the electrocoagulation process, a maximum cadmium removal rate was observed at 98.15% when the electrodes' distance was 1.5 cm. When the distance between the electrodes increases, polymer absorption of iron by the electrodes is reduced so that their movement becomes slower, leading to density in the clots. Therefore, when the electrode distance increases to more than 1.5 cm, interactions between the molecules and the clots become low, resulting in decreasing efficiency of removal [35]. Although by reducing the distance between the electrodes higher pollutant removal efficiency can be achieved, electrode distances of more than 1.5 cm causes increases in current density and makes a short circuit in the electrical current, and is not affordable [17]. There was a significant difference between the two processes tested. With the electrocoagulation process, by increasing the voltage, pH and retention time it showed a greater efficiency in eliminating cadmium in synthetic and real samples. Using the coagulation process, after increasing the concentration of coagulants and time the call was made at neutral pH and reaction time ($P < 0.05$).

5. Conclusion

Cadmium removal efficiency by using the electrocoagulation method is more efficient than the coagulation process; thus the electrocoagulation process for cadmium removal from the battery industry and other related industries is preferably recommended.

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