Performance Evaluation of Electrocoagulation Process for Removal of Sulphate from Aqueous Environments Using Plate Aluminum Electrodes

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Abstract: Sulphate is one of the main ions available in natural and wastewater. High rate of sulphate in drinking water causes health problems such as irritation and digestive problems and makes water bitter. The purpose of this study is to assess the efficiency of sulphate removal from drinking water through Electrocoagulation process (EC) using plate aluminum electrodes. This study is an experimental pilot one in which a glass tank in the volume of 1.3 liter containing 6 plate aluminum electrodes was used. These electrodes were attached to a power supply in a mono-polar and parallel arrangement in order to switch the alternating current to the direct one. That is, each electrode was attached to positive and negative poles directly and alternately. The tank was filled with synthetic water containing sulphate with the concentration of 350 & 700 mg/L. Percentage of sulphate removal in potential range of 10, 20 & 30 V, reaction times of 40, 20 & 60 min and pH 7.0, 3.0 & 11.0 was measured. The results demonstrate that the maximum efficiency of sulphate removal is in the electrical potential of 30 V, reaction time of 60 min and pH 11.0 and increasing the concentration of ion causes the increase of time needed to achieve a suitable efficiency of removal. It indicates the direct effect of pH and difference of electrical potential on removal of sulphate through EC process. Totally, the results obtained in this research show that Electrocoagulation technology can be introduced as a suitable and promising technique to remove sulphate from aqueous environments using plate aluminum electrodes.

Key words: Electrocoagulation process • Sulphate ion • Aluminum electrodes • Aqueous environments

INTRODUCTION

Sufficient water of desired quality is essential for survival and supplying clean water is one of the fundamental problems of human [1]. Sulphate is one of the main ions available in natural and wastewater [2-4]. Chemically, sulphate is a neutral, nonvolatile and nontoxic compound [3]. Main natural source of sulphate is underground and ground water, chemical weathering processes and analysis of minerals containing sulphate such as Pyrite, sulphide oxidation, sulphur and analysis of plant and animal remains [5-7]. Industrial wastewaters such as wastewater from paper, textile, fertilizer manufacturing, tanning and plastic industries are responsible for the most artificial spread of sulphate which may consist of thousands million gr/L of sulphate [2, 6, 8, 9]. High rate of sulphate in drinking water causes health problems such as irritation and digestive problems and gives water a bitter taste [6]. Sulphate is considered as an important ion both in general and industrial usage as well as formation of dregs in boilers and radiator [10]. Furthermore, it is of great attention regarding corrosion of waste pipes, creating serious problems in wastewater anaerobic treatment processes like reduction of sulphate to sulphide and as the strong inhibitor of microbial activities and plant growth [9, 11]. Although health effects of sulphate are relatively short term, it is acute and should be reduced in drinking water up to the recommended concentration [12]. Recommendation of WHO for the concentration of sulphate in drinking water is 250 mg/L [10]. Effective removal of this ion from water is a complicated issue due to its solubility and high
stability [6]. Various techniques have been employed to remove this ion from water such as ion exchange, reverse osmosis, nanofiltration, electrodialysis, chemical precipitation, adsorption, crystallization and biological techniques [2, 3, 6, 9, 13]. These techniques have some problems like increase of salts such as sodium, high exploitation and yield cost, need of final treatment of sewage and sludge removal [3, 6]. On the other hand, some promising techniques have been developed based on electrochemical techniques lack the aforesaid problems from which Electrocoagulation (EC) process can be mentioned [1]. In this process, the coagulant is produced in the place through electrolytic oxidation of the sacrificial electrode (anode) [14, 15]. Produced ions in anode electrode (of iron and aluminum type) react with hydroxide ions produced in cathode electrode and create metal insoluble hydroxides while react with ion species, colloids, suspended solids, etc. and sediment [1, 15]. Removal mechanism of EC process includes coagulation, surface absorption, precipitation and flotation [16]. This is the way EC process is expressed [15]:

1. Anode: \[ M - M^{n+} + ne \]
2. Cathode: \[ aH_2O + ne \rightarrow aH_2 + aOH \]
3. Metal hydroxide formation: \[ M^{n+} + nOH \rightarrow M(OH)_n \]

The main reactions occurring at the aluminum electrodes are:

1. Anode: \[ 2Al - 6e^{-} \rightarrow 2Al^{3+} \]
2. Cathode: \[ 2Al^{3+} + 3H_2O \rightarrow 2Al(OH)_3 + 3H_{2g} \]

EC technology, compared with other techniques, enjoys some advantages such as simple equipment, easy exploitation, short resistance time, no need of chemicals, low sludge production, effective sedimentation, capability of suitable sludge dewatering and environmental adaptation. Moreover, this process can cause oil, color, suspended particles, chemical waste, organic materials, synthetic detergents, heavy metals, phenol, surfactants and nitrate removal from water and wastewater [17-21]. This technique has enormous potential for removing the defects of old treatment techniques [1]. The purpose of this study is to investigate the efficiency of using Electrocoagulation process in removing sulphate from aqueous environments in order to reduce dangers of sulphate in drinking water in human health, achieving the quality standards of drinking water and developing a new treatment technology to remove sulphate from aqueous environments.

MATERIALS AND METHODS

The conducted study is of experimental pilot type. Chemical compounds used to produce synthetic water sample containing different concentration of sulphate and adaptation of sample pH in the mentioned rates include sodium sulphate, barium chloride, buffer solution, sulphuric acid and normal sodium hydroxide [10]. Sodium chloride was used for the purpose of increasing conductivity of the solution. Equipment related to EC include power supply (transformer of alternating current to the direct one), 6 plate aluminum electrodes with dimensions of 100×10×2 mm placed at a distance of 15 mm into a glass tank with dimensions of 110×100×150 mm in volume of 1.3 L [15]. Electrodes were attached to the power supply in a mono-polar and parallel arrangement. To be more precise, each electrode was attached to positive and negative poles directly and alternately [1].

Below figure shows an overview of the pilot:

Fig. 1: Bench-scale EC reactor with monopolar electrodes in parallel connection
Synthetic water samples containing sulphate with concentration of 350 and 700 mg/L were injected to the reactor after pH(3.7 and 11) adaptation. Mixing speed was 400 rpm [22]. Samples of 25 mL were selected after flowing electrical current with different voltage of 20, 10 and 30 from the middle of the reactor at the intervals of 40, 20 and 60 min. Then, selected samples were passed through a membrane filter with the pore size of 0.45 μm in order to remove the produced flocs. Finally, filtered samples were measured regarding the remained sulphate concentration using spectrophotometry instrument in wave length of 420 nanometer in conformity with the approved methods mentioned in the book of standard methods for water and wastewater experiments and final pH of the solution injected to the reactor was also measured after carrying the process.

RESULTS AND DISCUSSION

In present study, Electrocoagulation process is used through plate aluminum electrodes as a treatment technology to remove sulphate from aqueous environments and its removal efficiency has been evaluated regarding voltage, pH, initial concentration and different reaction times. Results of this study have been shown in graphs 1, 2, 3, 4, 5, 6 using Excel software. Graphs 1 & 2 show the removal efficiency of sulphate in stable pH 3.0, different voltages of 10, 20, 30, initial concentration of 350 & 700 mg/L and reaction times of 20, 40, 60 min. According to these graphs, voltage increase causes a slight increase in the efficiency of removal of sulphate so much that the maximum removal efficiency in the voltage of 30 and reaction time of 60 min in concentration of 350 & 700 mg/L was %51.46 & %40.3, respectively. Final pH of the product was measured too and demonstrated an increase from 3.0 to 8.4. The efficiency of removal of sulphate in stable pH 7.0 has been shown in graphs 3 & 4. As it is obvious, higher removal efficiency has been obtained in stable pH 7.0 and the maximum removal efficiency in the voltage of 30 and reaction time of 60 min in concentration of 350 & 700 mg/L was %58.75 & %44.2, respectively. Final pH of the product increased from 7.0 to 9.86. The efficiency of removal of sulphate in stable pH 11.0 has been shown in Graphs 5 & 6. In this pH, the maximum removal efficiency in the voltage of 30 and reaction time of 60 min in concentration of 350 & 700 mg/L was %79.7 & %55.8, respectively which was the highest efficiency achieved in this process using EC technology. Final pH increased up to 0.12.

Effect of Current Density: Density of electricity current is one of the most important control parameters of reaction speed in electrochemical processes like Electrocoagulation since it is not only the determiner of the coagulant dosage injected to the solution but effective on the rate of bubbles production, size and growth of produced flocs and consequently on the treatment efficiency of Electrocoagulation process [15, 23-26]. Studying the effect of electricity current density on removal of sulphate, it is observed that removal efficiency is increased with the increase of voltage. Rantakumar et al. achieved the same results in studying the removal of arsenic using Electrocoagulation process from aqueous environments. Likewise, Bazrafshan et al came to the similar conclusion in the study about surveying the application of Electrocoagulation process in removing heavy metal of chromium (VI) using aluminum and iron electrodes from aqueous environments [27, 28]. In this study, voltage 30 in the reaction time of 60 min showed the highest degree of sulphate removal, i.e. %79.7. The minimum removal efficiency was obtained in electrical potential of 10 volt. It indicates that oxidized aluminum has been increased in high voltage which results in the production of more amounts of sediments and hydroxide flocs with high absorption rate and consequently, increase of removal efficiency [15, 23, 24, 28]. On the other hand, high voltage causes the increase of aluminum release to water which is dangerous and not cost effective [15, 26]. Current increase leads to bubbles density increase and their size decrease and, therefore, causes more and rapid removal of pollutants [24, 27]. Effective level and resistance time of larger bubbles is more than the small ones, so the flotation efficiency is increased with the increase of density and decrease of bubbles size [22]. Decreasing the voltage, time needed for achieving the similar efficiencies is increased. These results are similar to the results of studies of G. Moudhen et al. regarding the electrochemical removal of chromium (VI) using iron and aluminum electrodes, Mohammad M. et al. regarding fluoride removal and Bazrafshan et al. about the chromium removal through Electrocoagulation process [15, 27, 29]. As a whole, voltage 30 is recommended to achieve the desired efficiency.

Effect of pH: Electrocoagulation process is significantly related to the solution pH which is increased during the process [15, 26, 30-32]. In this study, pH 3.0, 7.0 and 11.0 of the remaining solution has increased up to 8.4, 9.86 and 11.12, respectively. As aluminum hydroxide is a
Fig. 1: Efficiency of sulphate removal during EC process using AL electrodes (Initial concentration=350mg/L and pH=3)

Fig. 2: Efficiency of sulphate removal during EC process using AL electrodes (Initial concentration=700mg/L and pH=3)

Fig. 3: Efficiency of sulphate removal during EC process using AL electrodes (Initial concentration=350mg/L and pH=7)
Fig. 4: Efficiency of sulphate removal during EC process using AL electrodes (Initial concentration=700mg/L and pH=7)

Fig. 5: Efficiency of sulphate removal during EC process using AL electrodes (Initial concentration=350 mg/L and pH=11)

Fig. 6: Efficiency of sulphate removal during EC process using AL electrodes (Initial concentration=700mg/L and pH=11)
hydrophobic one, pH is considered as an essential factor for the formation of Aluminum hydroxide floes [15, 26]. Alternation range of pH depends on the type of the electrodes used and amount of initial pH of the solution [31, 33]. In this study, experiments have been conducted in three ranges of pH for the purpose of confirming the effect of pH on the sulphate removal efficiency and it is observed that the increase of pH increases sulphate removal. The maximum efficiency of removal was achieved in pH 11.0 because of different reactions occurred in different pH [28, 30].

1) Reaction 1 (acid pH): 2Al\(_2\)(OH)\(_4\) + 6H\(_2\)O \rightarrow O\(_2\) (g) + 4H\(_2\) + 2Al(OH)\(_3\)
2) Reaction 2 (neutral pH): 3Al\(_2\)(OH)\(_4\) + Al(OH)\(_3\) \rightarrow 2Al\(_2\)(OH)\(_3\)(aq) + 4H\(_2\)O
3) Reaction 3 (alkaline pH): 2Al\(_2\)(OH)\(_4\) + 6H\(_2\)O \rightarrow 2Al(OH)\(_3\)(aq) + 3H\(_2\)O

H\(_2\) produced in these reactions goes upward and causes flotation and Al\(_2\)(OH)\(_3\) & Al(OH)\(_3\) sediment. The results of this study are in conformity with the results of studies of Hyun Kim about color removal, Sirajuddin about electrolytically recycle of chromium salts from tanning wastewater and Bazaarshian about the removal of Cr VI through EC process and indicate that the EC process can act as a pH modifier [27, 34, 35].

**Effect of Resistance Time:** In present study, the effect of resistance time on the sulphate removal efficiency was also investigated. The results demonstrate that removal efficiency is increased by the increase of resistance time. Moreover, it is concluded that the maximum rate of removal was obtained at the onset of the experiment which is in conformity with the results presented by Nafaa Ahdoum considering the wastewater treatment containing copper, zinc and chromium VI through Electrocoagulation process, Chaudhry concerning the electrolytical removal of Cr\(^{VI}\) from aqueous environments and P. Lakshmpathi regarding the removal of Cr (VI) through electrochemical reduction [36-38]. The results of this research are shown in Graphs 1, 2, 3, 4, 5, 6. As you see, efficiency of sulphate removal is increased by increasing the electrical potential and pH and desired removal of sulphate is obtained in voltage 30, pH of 11.0 during 60 min.

**Effect of Concentration:** The results of the experiments reveal that the initial concentration of sulphate can affect the removal efficiency so much that in higher initial concentration, removal efficiency of sulphate ion has reduced. That is, higher voltage or longer reaction time is needed (Figures). These results are in agreement with the previous results of studies conducted by Daneshvar et al. regarding the efficiency of Electrocoagulation process in colour removal, Chaudhry et al. regarding the electrolytical removal of Cr\(^{VI}\) from aqueous solutions, Nafaa Ahdoum considering the wastewater treatment containing copper, zinc and chromium VI through Electrocoagulation process, Daneshvar et al. regarding colour removal from colour solutions containing acid using Electrocoagulation, P. Gao et al. about the removal of Cr VI from wastewater using EC process along with Electro-flotation process and Bazaarshian et al. about the removal of Cr VI from synthetic chromium solutions through EC process. This study and similar studies demonstrate that EC technology, compared with other techniques, enjoys some advantages such as simple equipment, easy exploitation, short resistance time, no need of chemicals, low sludge production, effective sedimentation of sludge, capability of suitable dewatering and biological adaptation [27, 37, 39, 40].

This and other related studies indicate that EC technology, compared with other techniques, enjoys some advantages such as simple equipment, easy exploitation, short resistance time, no need of chemicals, low sludge production, effective sedimentation, capability of suitable sludge dewatering and environmental adaptation [18-21, 36]. Studies show that in some part of Iran, concentration of nitrate and fluoride are high in groundwater and therefore this process is suitable for treatment of such polluted waters [41-44].

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