



Practical assessment of electrocoagulation process in removing nickel metal from aqueous solutions using iron-rod electrodes

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Received 16 August 2010; Accepted 18 December 2011

ABSTRACT

The aim of this study was to investigate the practical application of electrocoagulation (EC) process in removal of nickel from aqueous solutions by iron-rod electrodes. This experimental research was carried out as a pilot scale using forty bipolar iron-rod electrodes in a glass container with a 1.5 l capacity which was connected to an electrical source. The removal efficiency of nickel from synthetic solutions was measured with initial nickel concentrations of 5 and 500 mg l⁻¹, at pH of 3, 7 and 10, reaction times of 20, 40, 60 and 80 min and electrical potentials of 20, 30 and 40 V. Results showed that by increasing pH, nickel removal efficiency for each concentration has increased, as much as 99.9% and 99.8% for 500 and 5 mg l⁻¹ concentrations respectively. The optimum removal efficiencies at nickel concentrations of 500 and 5 mg l⁻¹ were reached at 20 and 40 min of reaction time and 20 V of potential difference respectively. The final pH of treated solutions has also increased which was due to rise in acidic pH and decrease in alcoholic pH. The results represent that EC process could be introduced as a promising technology and as an alternative method instead of other procedures in removing nickel from tainted aqueous solutions.

Keywords: Electrocoagulation; Industrial wastewater; Nickel removal; Heavy metal; Aqueous solutions; Iron electrode

1. Introduction

From general health and pollution control point of view, removal of these pollutants from aqueous environment is highly recommended [1,2]. Nickel ion compared to other heavy metal ions is more resistant pollutant [3]. Nickel removal to the permitted concentration limit is very important because its carcinogenicity in mammals has been well proved and it can also cause serious problems

such as dermatitis, allergy, damage to the neural and respiratory systems in human beings [4]. Nickel is found in industrial wastewaters such as paint making, non-iron metal processing's, petrochemical, power generation plants and especially electroplating industries [4,5]. For the time being, various techniques including chemical precipitation, ion-exchange, membrane separation, adsorption, evaporation, electrochemical reduction, coagulation and flocculation, flotation and biological treatment have been applied for removal and recycling of heavy metals from water and wastewater

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[6,7]. Major limitations of present treatment technologies consist of removal of high concentrated solutions, high electricity requirements, high per capita exploitation and maintenance expenses, disposal of large sludge volume, and inefficient removal requiring expensive equipments, and addition of undesirable chemicals [5,8,9]. In recent years, several studies have been conducted on replacing these technologies with modern technologies for removal of heavy metals from water and wastewater [9]. One of these technologies is EC process [8,10,11]. EC process is basically an electrolytic process including destabilization of suspended, emulsion or solved pollutants in aqueous media through using electrical current [12]. Three main steps which occur in EC processes are as below:

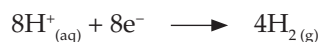
1. The reaction of electrolyte on anode.
2. Formation of coagulants in water phase.
3. Adsorption of colloids or soluble pollutants into coagulants and removal through settling or flotation process [13].

Applied electrodes in EC process are usually iron or aluminum [14]. It should be noted that in water treatment processes which Fe^{3+} being used as a floc forming material, has significant benefits and it is harmless [15], compared to Al^{3+} which has some toxicity effects (causing Alzheimer disease). In the present study, iron electrodes have been used. Iron oxidation in electrolytic systems produces ferrous hydroxide according to the following mechanism [15]:

(1) In anode



(2) In cathode



(3) Overall



This process has been widely used for the removal of pollutants such as organic matters, suspended substances, nutrients (nitrogen and phosphorus), heavy metals, turbidity, pathogens and etc. [11]. EC technology compared to other treatment techniques has some advantages such as simple equipments, convenient exploitation, avoiding chemicals, less sludge production, low residence time, rapid sedimentation of produced

flocs, simple de-watering of the produced sludge and low investment and exploitation expenses [16,17]. The aim of the present study was to evaluate the applicability of EC process by using iron-rod electrodes as a new innovative technology in removal of nickel from water and wastewater and to study the effects of electrical potential, amount of coagulant produced, initial pH, reaction time, initial concentration of nickel and the amount of electrode consumed on the removal efficiency.

2. Materials and methods

2.1. Electrochemical cell

In this study, closed electrochemical cell was made by using Plexiglas plates with thickness of 5 mm and dimensions of 140 mm × 120 mm × 120 mm as a pilot plant. The efficient volume of the reactor was 1500 cm³. Forty iron-rod electrodes with a diameter of 2 mm, active surface area of 49 cm² were prepared and a distance of 1 cm was set in between them. The Electrodes were connected to each other in a parallel arrangement and bipolar to the power source (connection of only outer electrodes to the positive and negative poles). The distance between electrodes and the reactor bottom was 2 cm for good mixing. The speed of mixing was 400 rpm. The diagram of cell is represented in Fig. 1.

2.2. Reagents and solutions

Chemical compounds required for this study were nickel chloride, sulfuric acid, sodium hydroxide and potassium chloride which were all supplied from Merck Company. For pH adjustment, 1N Sulfuric acid and sodium hydroxide solutions were used. To increase the electrical conductivity 1N potassium chloride solution was used.

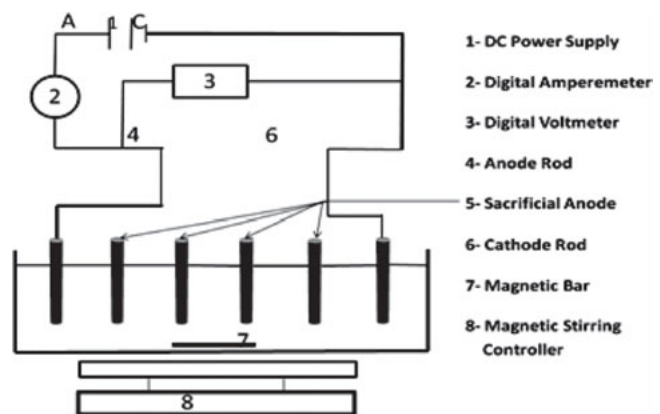


Fig.1. Bench-scale EC reactor with bipolar electrodes in parallel connection.

2.3. Experiments

Synthetic solutions of Nickel with concentrations of 5 and 500 mg l⁻¹ were made and injected into the electrochemical cell after pH adjustment at the range of 3, 7 and 10 and obtaining proper electrical conductivity conditions. Then, at electrical potentials of 20, 30 and 40 V, 50 mm samples were taken from the middle of the reactor at the time intervals of 20 min during 80 min reaction time. Then, samples were filtered through a 0.45 μm membrane filter for removal of produced flocs. Filtered samples were kept in 4°C and the concentrations of the residual nickel in the solutions were measured at the end. pH of the remained solutions were also measured.

3. Results and discussion

EC is completely a complicated process and is affected by several parameters such as current density, rate of coagulant production, initial pH, reaction time, concentration of pollutants, electrode spacing, the amount of consumed electrolyte and mixing. In order to increase the efficiency of this process, the effects of mentioned parameters are discussed [15].

3.1. The effect of current density and coagulant dosage on nickel removal

In all electrochemical processes current density is an important parameter for the control of reaction rate [17]. Current density, determines not only the coagulant dosage, but also the amount and size of bubbles produced. Therefore, this factor has major effect on the efficiency of pollutants removal. According to Faraday's law, the rate of produced ferrous and hydroxide ions is related to current density [17]:

$$m = \frac{ItM}{zF}$$

where I is the current intensity, t is the time, M is the molecular weight of aluminum or hydroxide ion (g mol⁻¹), z is the number of electrons transferred in the reaction (3 for aluminum and 1 for hydroxide) and F is the Faraday's constant (96,486 C mol⁻¹).

Moreover, with increase in current density the bubble size decreases, while bubble density and current increases. This is beneficial for separation process and results in more rapid removal of coagulant through flotation. Therefore, the probability of contact between coagulant and pollutant exists [17,18]. On the other hand, with increase in current density, the amount of oxidized iron resulting in production of high amounts of hydroxide flocs increases and thus increases the filtering expense [19]. For this reason, in order to achieve a

low filtration cost, current densities in low levels should be maintained [20]. Figs. 2–4 represent the efficiencies of nickel removal at 20, 30 and 40 V at pH of 3, 7 and 10. As it is shown, the highest removal efficiencies for nickel concentrations of 5 and 500 mg l⁻¹ are 99.8% and 99.9% respectively which are obtained at 20 V and reaction times of 20 and 40 min. These findings are in agreement with the results of researches done by Rantakumar et al. on Arsenic removal from water environments by using EC process, Bazrafshan et al. on the efficiency of EC with aluminum electrodes in removal of Cadmium, Rafati et al. on the removal of chromium(VI) from aqueous solutions using lewattit fo36 nano ion exchange resin, Koparal et al. on nitrate removal from water by EC/electrical

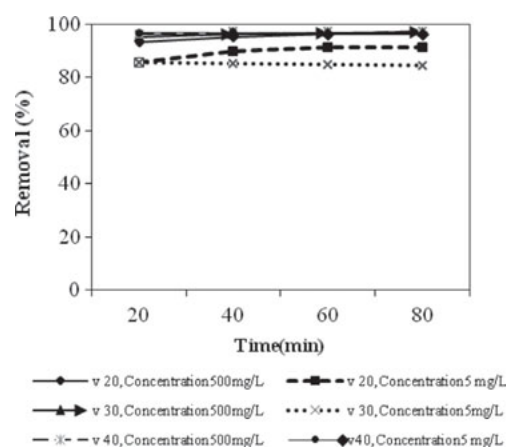


Fig. 2. Efficiency of Nickel removal during EC process using iron-rod electrodes (initial concentration for 5 and 500 mg l⁻¹ and pH = 3).

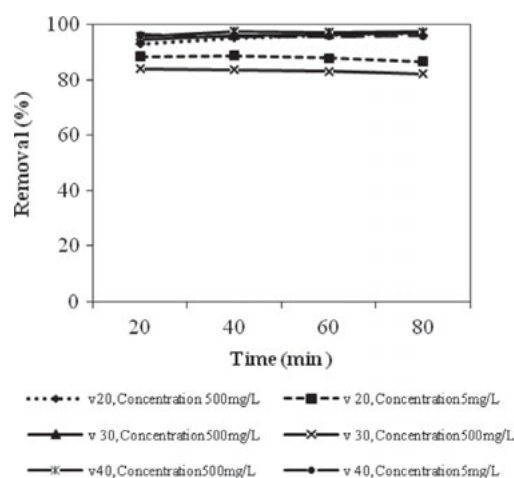


Fig. 3. Efficiency of Nickel removal during EC process using iron-rod electrodes (initial concentration for 5 and 500 mg l⁻¹ and pH = 7).

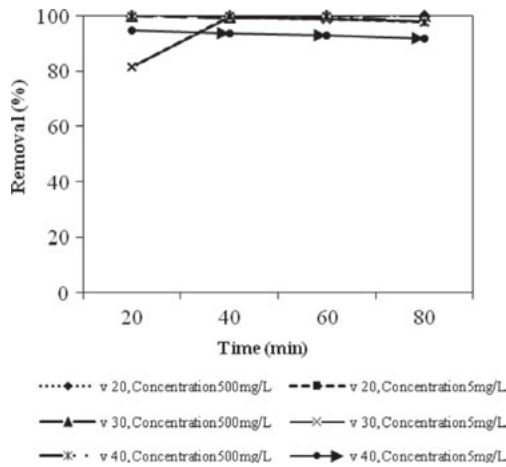


Fig. 4. Efficiency of Nickel removal during EC process using iron-rod electrodes (initial concentration for 5 and 500 mg l⁻¹ and pH = 10).

reduction, Njiki et al. on Mercury removal from water by EC process with Al and Fe electrodes and also Qiyian et al. on humic acid removal from underground waters by EC [21,22].

3.2. The effect of pH on nickel removal

As it has been proved, pH has an important effect on EC process [11,17]. In fact, the nature and efficiency of complex chemical and electrochemical reactions is completely related to the pH of system [23]. The effect of pH on nickel removal by EC process has been presented in Figs. 2–4. In the present study, the highest removal efficiency for 5 and 500 mg l⁻¹ nickel concentrations was attained in pH of 10. Final pH of synthetic solutions in both concentrations is presented in Fig. 5. The produced Hydrogen as one of the products of Redox reaction (oxidation–reduction) may remove soluble organic matters or any suspended substance through flotation. In spite of this, Fe³⁺ ions may be affected by Hydrogen and depending on the pH of the solution, under acidic conditions variants of Fe(OH)²⁺, Fe(OH)₂⁺ and Fe(OH)₃ are produced according to the following reactions [15]:



In alkaline conditions, Fe(OH)₆⁻ and Fe(OH)₄⁻ and also variants of Fe(OH)₂ and 2Fe(OH)₃ are produced according to the following reactions [24]:

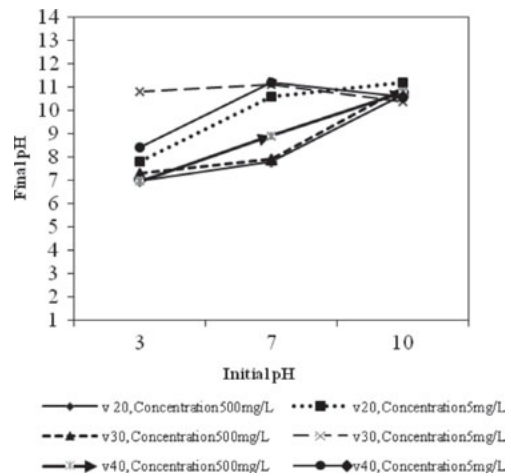
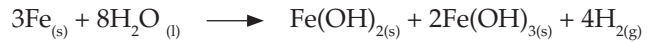


Fig. 5. pH variation after electrocoagulation.



If anode potential is sufficiently high, secondary reactions in anode such as oxidation of organic matters and water or chlorine ion in solution might occur [16,20]:



The produced chlorine causes the following reactions in high pH [20]:



Therefore, it is completely clear that in EC process the formation of anionic and cationic variants by using an iron plates or rods as sacrificing electrodes are possible [15]. The results showed that with increase in initial pH, the rate of nickel removal increases. These results are in agreement with the results of researches by Hyun-ki study on color removal, Sirajuddin study on electrolyte recycling of chrome salts from tanning wastewaters by EC, Ghosh et al. study on removal of Fe(II) from drinking water by EC technique, Escobar et al. study on optimizing EC process for removal of copper, lead and cadmium from natural waters and synthetic wastewaters and Tchamango et al. study on filtering of color wastewaters by EC with Al electrodes. All these studies suggest that EC process can act as a pH moderating factor [23,25].

3.3. The effect of reaction time on nickel removal

The effect of reaction time on nickel removal in EC process was studied. Figs. 2–4 represent this effect in initial concentrations of 5 and 500 mg l⁻¹. Theoretically, based on Faraday's law, duration of electrolysis affects the amount of released iron in a system with iron electrodes and determines the amount of produced Fe³⁺ from iron electrodes [26]. EC process includes two steps:

1. Destabilization
2. Accumulation

The first step is usually short and the second one is relatively long. With increase in reaction time, both energy and electrode consumption increases and this shows that reaction time is a very important parameter due to affecting the cost effectiveness of EC process in polluted waters [27]. In the present study, 95% of nickel was removed in the first minutes, while in later minutes the percentage of removal was low. This finding is similar to the results of researches done by J. Hu et al. on removal and recycling of chrome VI from wastewater by using nano-particles of magma, Ugurlu et al. on the removal of lignin and phenol from paper mill effluents by EC and Ayhansengil et al. on filtration of tanning wastewater by EC [28]. In 1 min Hydrogen bubbles formed from cathode were distributed in the aqueous environment and white foam was produced on the surface of solution. In 10 min the color of solution turned to yellow green. In 20 min the color turned to brownish green. In 60 min a little sediment was formed on anode. These findings are similar with the results of NafaaAdhoum on treatment of electrolyte wastewater containing copper, zinc and chrome VI by EC process, Caudhary on electrolytic removal of chromium VI from water environments, Lakshmipaathiraj study on removal of chrome VI by using electrochemical reduction and Ghernaout et al. on applying EC in *Escherichia coli* culture media and surface waters [29].

3.4. The effect of initial concentration on nickel removal

In order to investigate the effect of nickel ion concentration in water environments on its removal rate, two synthetic solutions with concentrations of 5 and 500 mg l⁻¹ were tested. Figs. 2–4 show the change in the rate of nickel removal based on the initial concentrations. As it was expected, the rate of nickel removal from the solution was increased with increase in nickel concentration. This fact has required higher electrical charges for reaching the remained metal concentrations to the permitted level for effluents to be discharged into wastewater collection system [20]. In optimum pH and electrical potential, the concentration of remained nickel with the initial

concentrations of 5 and 500 mg l⁻¹ decreased to 0.025 and 0.5 mg l⁻¹ respectively. It is obvious that in high initial concentration of nickel, the removal efficiency is high. In higher current density, with decrease in initial nickel concentration, the remained concentration increases. This fact is probably due to insufficient production of ferrous hydroxide [11]. It is clear that higher concentrations of pollutant require longer time for complete removal, but for initial high nickel concentration compared to low initial concentration a relatively short time was needed for complete removal. This can be explained by dilute solution theory. In dilute solutions, the formation of distributing layer around the electrode causes lower reaction speed but in concentrated solutions, distributing layer has no effect on distribution speed or immigration of metallic ions toward electrode surfaces [30]. This finding is in agreement with the results of researches done by Chaudary et al. on electrolytic removal of chrome VI from water environments, NafaaAdhoum et al. on treatment of electroplating wastewater containing copper, zinc and chromium VI by EC, Daneshvar et al. on discoloration of color solutions containing acid by EC, P. Gao et al. on Cr VI removal from wastewater by using EC process in combination with electroflotation process and Ghosh et al. on removal of Fe(II) from drinking water by EC [20,23,29].

3.5. Effect of electrode spacing

The setups of electrode assembly are very important for required effective surface area of electrode and inter electrode intervals [23]. Removal percentages for nickel with various intervals between electrodes were investigated and shown in Fig. 6. It has been observed by increasing the inter-electrode intervals, the removal percentage of Ni decreases. In small intervals, the gas, was blown into the cathode compartment, and induces the

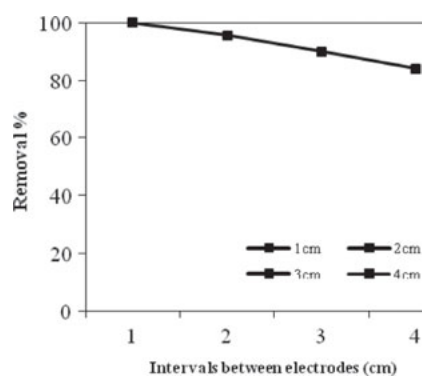


Fig. 6. Effect of intervals between electrodes on the removal efficiency of Ni ions with initial concentration 500 and 5 mg l⁻¹, current density 20 V, reaction time 20 and 40 min, pH 10.

phenomenon of formation of great floating complexes [31]. Also, at higher inter electrode intervals, the rate of aggregation of suspended particles as well as adsorption of contaminants would be low. This may be the reason behind the lower removal efficiency at higher inter electrode distance [23].

Our result is in accordance with data from other works like removing humic acid from groundwater waters using EC in China and Removal of Fe(II) from tap water by EC technique in India [23,31].

3.6. The amount of consumed electrode in nickel removal

One of the most important criterions for selection of an efficient process for removal of heavy metals is cost effectiveness of the process. In the present study after each series of experiments, iron electrodes were weighed and the results are presented in Fig. 7. With increase in current density, the amount of consumed electrode increased too. The highest weight loss of electrodes was at 40 V and in concentration of 5 mg l⁻¹, while the lowest weight loss was at 20 V and in concentration of 500 mg l⁻¹. These results are similar to the findings of research done by Adhoum et al. study on decolourization and removal of phenolic compounds from wastewater and treatment of electroplating wastewater containing Cu²⁺, Zn²⁺ and Cr(VI) by EC process [20].

3.7. Effect of mixing

Mixing is regarded as an important unit of water and wastewater treatment [32]. Using rod electrodes instead of flat equivalent, it was possible to put more electrodes inside reactor and had a better mixing condition; moreover, time needed to accomplish the operation problems would be reduced due to the production of more metal hydroxide flocs.

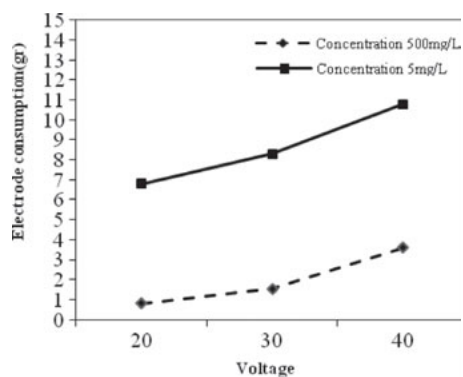


Fig. 7. Iron electrode consumption rate during electrocoagulation process.

4. Conclusions

The results of the present study show the applicability of EC process in removal of nickel from water environments. The most removal efficiency for high concentration (500 mg l⁻¹) was found at 20 V, pH of 10 and reaction time of 20 min, while for low concentration (5 mg l⁻¹), removal efficiency was at 20 V, pH of 10 and reaction time of 40 min. One of the major advantages of EC process is no need for adding chemical substances, and nickel containing solutions can be reused. The results of this study can be considered in treatment of industrial wastewaters, especially electroplating industries and polluted underground and surface waters.

Acknowledgements

This study was funded by the health research department of Zahedan University of Medical Sciences and was conducted in the chemical laboratory of school of public health, Zahedan University of Medical Sciences.

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