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Removal of Nickel from Drinking Water by Electrocoagulation Technique Using Alternating Current

S.E. Mansour and I.H. Hasieb

Department of Chemistry, Faculty of Science, Omar Al-Mukhtar University, B.O. 919, Al-Bayda, Libya

Corresponding Author: S.E. Mansour, Department of Chemistry, Faculty of Science, Omar Al-Mukhtar University, Box 919, Al-Bayda, Libya Tel: +218913767134

ABSTRACT

The main objective of this study is to investigate the removal of Ni(II) ions from water by electrocoagulation method under the effect of alternating current. AC is used in the electrocoagulation process in order to reduce the disadvantages of DC and to facilitate the effective current transfer between the anode and cathode. This process involves applying an alternating electric current to sacrificial aluminum electrodes inside a reactor tank. The coagulating Al ions generated from the electrodes react with Ni(II) ions in water forming flocs which precipitate. Several factors affecting efficiency of electrocoagulation are investigated. They include initial Ni(II) ion concentration, applied current density and inter-electrode distance. The power consumption for electrocoagulation increased proportionally when the current density, electrode gap and concentration of electrolyte in the solution were increased. The metal ions removal efficiency was reported to be between 86.80 and 99.50%.

Key words: Electrocoagulation process, alternating current, sacrificial aluminum anode, electrochemical treatment, nickel removal, drinking water

INTRODUCTION

Nickel is one of many heavy metals widely distributed in the environment. Drinking water generally contains nickel at concentrations less than 10 µg L^{-1}. Elevated nickel levels may exist in drinking water as a result of the corrosion of nickel-containing alloys used for pipes and other components in the water distribution system as well as from nickel-plated fittings. However, nickel may also be present in some ground waters as a consequence of dissolution from nickel ore-bearing rocks.
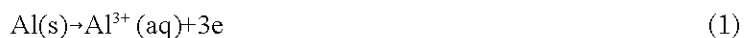
Nickel may cause health problems if found in amounts greater than the health standard set by the United States Environmental Protection Agency (USEPA, 2003; WHO, 2006; EC, 1998). The most common adverse health effect of nickel in humans is an allergic reactions such as skin rashes. It can also be dangerous when the maximum tolerable amounts are exceeded. An uptake of very high levels of nickel has higher chances of developing lung cancer, nose cancer, larynx cancer and prostate cancer. It can also cause heart disorders, asthma and chronic bronchitis (Meena *et al.*, 2005). This creates a need for the cleanup of nickel pollution. So it is very essential to remove Ni from soil, industrial wastewater, effluents and drinking water.

The conventional methods which are commonly used for the removal of nickel from water are physico-chemical methods, such as chemical coagulation, chemical oxidation or reduction, active
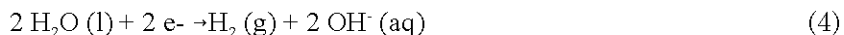
carbon adsorption, filtration, ion exchange and membrane technologies. These processes may be either ineffective or expensive, especially when the levels of heavy metal ions in the contaminated media are high.

Electrocoagulation-electroflotation (ECF) technology provides an alternative technique for removing pollutants from water and waste water. The method was found to be highly efficient and relatively fast compared to conventional techniques. Electrocoagulation is a process of destabilizing suspended, emulsified or dissolved contaminants in an aqueous medium by introducing electrical current into the medium without having to add chemical coagulants. Electrocoagulation consists of pairs of metal electrodes, either aluminum or iron plates, or a combination of both, that are arranged in pairs of two—anodes and cathodes. The electrical current provides the electromotive force causing the chemical reactions. In the case of aluminum, main reactions that take place on the electrode surfaces during electro-coagulation and electro-flocculation are as follows (Daneshvar *et al.*, 2006; Babu *et al.*, 2007).

At anode:

$$Al(s) \rightarrow Al^{3+}(aq) + 3e \qquad (1)$$

$$2H_2O\ (l) \rightarrow O_2\ (g) + 4H^+ + 4e^- \qquad (2)$$

At cathode:

$$2\ H\ (aq) + +2\ e^- \rightarrow H_2\ (g) \qquad (3)$$

$$2\ H_2O\ (l) + 2\ e- \rightarrow H_2\ (g) + 2\ OH^-\ (aq) \qquad (4)$$

$$Al^{3+} + H_2O \rightarrow Al\ (OH)^{2+} + H^+ \qquad (5)$$

$$AlOH^{2+} + H_2O \rightarrow Al\ (OH)^{2+} + H^+ \qquad (6)$$

$$Al\ (OH)^{2+} + H_2O \rightarrow Al\ (OH)_3 + H^+ \qquad (7)$$

$$Al\ (OH)_3 + H_2O \rightarrow Al\ (OH)_4^- + H^+ \qquad (8)$$

$$2\ H_2O \rightarrow 4\ H^+ + 4\ e^- + O_2\ (g) \qquad (9)$$

Aluminum hydroxides, aluminum poly-hydroxides or monomeric or polymeric forms of: Al $(OH)_2^+$, $Al_2\ (OH)_2^{4+}$, $Al_6\ (OH)_{15}^{3+}$, $Al_{13}\ (OH)_{34}^{5+}$ produced in water were found to increase the removal efficiency (Jiang *et al.*, 2002; Bazrafshan *et al.*, 2007; Drouichea *et al.*, 2008).

This technique has been applied successfully to the treatment of different types of wastewaters such as textile effluents (Szpyrkowicz *et al.*, 2001; Chen *et al.*, 2003; Daneshvar *et al.*, 2006), landfill leachate (Lopez *et al.*, 2004; Wu *et al.*, 2004; Feng *et al.*, 2007), olive oil wastewaters (Israilides *et al.*, 1997; Gotsi *et al.*, 2005; Kotta *et al.*, 2007), domestic sewage (Vlyssides *et al.*, 2002) and waste water containing metal ions (Cu^{2+}, Zn^{2+} and Cr (VI)) (Gao *et al.*, 2005).

The objective of this study was to evaluate nickel removal from synthetic nickel solutions by the electrocoagulation method under the effect of alternating current using 50 volt at 50 Hz and to determine the effects of various parameters like applied current density, initial metal ion concentration and inter-electrode distance on the removal efficiency.

MATERIALS AND METHODS

The experiments in batch were carried out on the electrochemical cell or EC-chamber consisted of perspex tank (internal volume is approximately 5.8 L), electrode assembly and power supply (variable transformer was used to control the current and potential). The cell contained two parallel aluminum electrode plates (geometric area of 225 cm^2 and 4 mm thickness) placed vertically in the bottom of the reactor. Aluminum was used in this work as the sacrificial anode, rather than iron, because the residual ferrous ions are easily oxidized by oxygen. Inside the cell, the gap between the anode and cathode was maintained at various distances (ranging from 0.005-0.002 m).

Voltage and current were measured by a digital voltameter (max 2) and digital ammeter (DT9201A). The conductivity of the solutions was measured by means of Philips digital conductivity meter (PW 9526). The pH was measured using a Hanna Instrument Check Temp (HI8314) pH meter. Desired concentrations of the nickel solution were prepared by mixing proper amount of nickel chloride (Aldrich, A.C.S. reagent) with tap water with conductivity and pH of 13 S/m and 7.5, respectively.

Before starting-up the process, the electrodes were cleaned with 1 M HCl and rinsed with de-ionized water to eliminate impurities from the surface of the electrodes. The electrolysis cell was filled with 1 L of Ni (II) ion solution (of initial concentrations ranging from 5-25 mg L^{-1}) and various constant currents(ranging from 0.01-0.04 A m^{-2}) were applied between the electrodes. During electrolysis, a 1 mL aliquot of filtrate sample were taken from the EC solution at different time intervals (up to 35 min). The concentration changes of metal ions were measured by a Beckman (DU 800) spectrophotometer with an absorption maximum at λ_{max} = 490 nm.

The calculation of Ni(II) removal efficiencies after electrocoagulation treatment were carried out using this equation (Daneshvar *et al.*, 2006):

$$R\% = [C_o\text{-}C \text{ / } C_o] \times 100 \tag{10}$$

where, C_o and C are concentrations of Ni(II) solutions before and after electrocoagulation in mg L^{-1}, respectively. The viscosity was measured by means of Ostwald type viscometer thoroughly cleaned, dried and calibrated with triply distilled water and purified ethanol. After attainment of thermal equilibrium, the efflux times of flow of liquids were recorded with a stopwatch correct to±0.1 sec. Viscosity of the solution, η, is given by the following equation:

$$\eta = (Kt\text{-}l \text{ /t) / } \rho \tag{11}$$

where, K and l are the viscometer constant and t and ρ are the efflux time of flow in seconds and the density of the experimental liquid, respectively. The uncertainty in viscosity measurements was within±0.003 m.pa.s.

The pK values of electrolytic solutions were calculated using the following equation:

$$pH = pK + \log C/C_o \tag{12}$$

where, C is the concentration of solution in each run and C_o is the initial concentration.

The effects of the reaction time, the applied current density, initial Ni(II) ion concentrations and inter-electrode distances were investigated.

RESULTS AND DISCUSSION

Effect of electrolysis time and current density: Current density (I) is the current delivered to the electrode divided by the active surface area of the electrode. In order to evaluate the effect of current density on the percentage removal of Ni(II) a number of measurements were carried out at different current densities 0.01, 0,02, 0.03 and 0.04 A m^{-2}, with the same initial Ni(II) concentration of 25 mg L^{-1} at pH = 7.8 using AC. According to the results shown in Table 1 the removal rates of the studied metal increased, as expected, with increasing current density. This was attributed due to the fact that at high current densities, the extent of anodic dissolution increased and the amount of hydroxo-cationic complexes resulted in increase of the removal efficiency.

These findings are consistent with the results of arsenic removal study by electrocoagulation method (Kumar *et al.*, 2004) and the results of the capability of electrocoagulation method with Aluminum electrodes in Cr^{6+} removal study (Kumar *et al.*, 2004; Bazrafshan *et al.*, 2007). This study also ascertained the maximum Ni(II) ion removal of 99.5% using Al electrodes at the 0.04 A m^{-2} for 35 min from the initial concentration of 25 mg L^{-1} of Ni(II) ion.

Effect of pH: It has been recognized (Chen *et al.*, 2000; Mollah *et al.*, 2004), that pH has a considerable effect on the efficiency of the EC process. Also, as observed by other investigators, the pH of the medium was changed during the process. This change depends on the type of electrode material and initial pH and alkalinity. In addition, pH measurements were found to be very important in showing the type of dominant reactions at the anode and cathode electrodes. experimental findings related to removal efficiency of metal ions as a function of initial pH (Adhoum *et al.*, 2004; Vasudevan *et al.*, 2009) have accounted for the decrease in removal efficiency of metal ions observed at extreme pH values. This decrease was attributed to the formation of soluble Al^{3+} cations at strong acidic pH and to the formation of monomeric anions Al(OH)$_4^-$ at strong alkaline pH. Therefore, the formation of these species are not constructive for water treatment process.

Present results illustrated in Table 2 showed, that the pH in a slightly alkaline medium (pH around 8) without addition of oxidants is nearly constant with time. This stabilization of solution pH at nearly constant value around 8.2 may be ascribed to the buffering capacity of complex

Table 1: Variation of percentage removal of Ni (II) with time at different current densities

Time (min)	Current density (A m^{-2})			
	0.01	0.02	0.03	0.04
5	66.60	67.10	70.60	73.30
10	71.70	72.50	74.90	77.70
15	76.30	77.30	79.40	82.10
20	79.10	80.70	84.20	85.80
25	81.40	82.90	86.10	88.60
30	82.70	84.50	87.20	94.70
35	84.00	87.60	93.70	99.50

Inter electrode distance: 0.005 m, Iinitial Ni (II) concentration: 25 mg L^{-1}, pH: 7.8, Conductivity: 12.5 S/m at 50 Hz and 50 volt using AC

Table 2: Variation of pH of Ni (II) with time at different current densities

Time (min)	Current density (A m^{-2})			
	0.01	0.02	0.03	0.04
5	7.82	7.85	7.88	7.91
10	7.84	7.87	7.90	7.94
15	7.86	7.89	7.93	7.97
20	7.88	7.91	7.95	7.99
25	7.92	7.93	7.97	8.05
30	7.95	7.96	7.99	8.09
35	8.00	8.03	8.07	8.15

Inter electrode distance: 0.005 m, Iinitial Ni (II) concentration: 25 mg L^{-1}, pH: 7.8, Conductivity: 12.5 S/m at 50 Hz and 50 volt using AC

Table 3: Variation of extent of Ni (II) removal with time

Time (min)	Initial concentration of Ni(II) mg L^{-1}				
	5	10	15	20	25
5	0.835	2.090	3.405	4.800	6.875
10	0.370	1.070	2.040	3.760	4.675
15	0.115	0.530	0.945	1.580	2.625
20	0.055	0.140	0.375	0.680	1.075
25	0.025	0.050	0.225	0.340	0.525
30	0.015	0.030	0.060	0.160	0.350
35	0.000	0.016	0.026	0.040	0.150

Inter electrode distance: 0.005 m, Current density: 0.04 A m^{-2}, pH: 7.8, Conductivity: 12.5 S/m at 50 Hz and 50 volt using AC

nature of aqueous Al^{3+}/Al(OH)$_3$ system (Kobya *et al.*, 2006). Therefore, the results of our study and previous studies (Kim *et al.*, 2002; Bazrafshan *et al.*, 2007; Malakootian *et al.,* 2009) indicate that EC process can operate as a pH moderator.

The removal efficiency of the studied metal after 35 min of electrolysis time at the constant current density of 0.04 A m^{-2} reached very high values, 99.5% in this pH range. This means that cathodic activity is more dominant in this pH range and all of the generated OH$^-$ ions at the cathode are consumed by the generated Al^{3+} ions at the anode forming Al (OH)$_3$ flocs and more aggregates. Furthermore, our results indicated, that OH$^-$ ions can also combine with the Ni^{2+} ions to form the insoluble Ni(OH)$_2$, which is an important benefit of this method. For this reason our EC process was conducted using solutions of Ni^{2+} of 25 mg L^{-1} in the initial pH 8.

Effect of initial metal ion concentrations: To reveal the effect of initial Ni(II) concentration and the time required for its quantitative removal, a set of experiments were conducted with five different solutions containing initial concentrations of 25, 20, 15, 10 and 5 mg L^{-1} of the metal ion. The solutions were treated at a constant current density of 0.04 A m^{-2} and different times of electrolysis. Table 3 and Fig. 1 illustrate the variations of the initial concentrations of nickel ions with time. The results show, for all initial concentrations used in this study, that most of the Ni(II) ion has been removed in the beginning of process (ca. 5 min). This has also been found in study of the Arsenic removal efficiency (Kumar *et al.*, 2004) and in studies of Cr(VI) removal by EC process (Chaudhary *et al.*, 2003; Bazrafshan *et al.*, 2008). A complete removal is observed at the end of
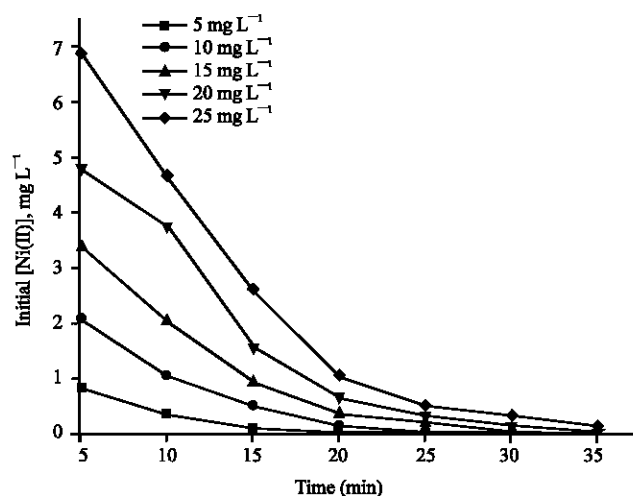
Fig. 1: Residual Ni(II) concentration versus time (min)

Table 4: Effects of inter-electrode distance on Ni (II) removal and current density at 0.04 A m^{-2}

Inter electrode distance (10^2) m	Removal of Ni (II) %
0.5	99.40
1.0	95.30
1.5	92.50
2.0	88.85
2.5	86.80

Initial concentration of Ni (II): 25 mg L^{-1}, Time 35 min, pH: 7.8, Conductivity: 12.5 S/m at 50 Hz and 50 volt using AC

35 min of process. The higher initial concentrations needed longer operating time to be quantitatively removed. Indeed, when the initial nickel concentrations were higher, more aluminum hydroxides were needed to decrease the dissolved nickel concentrations. Nickel removal is consequently limited by the production rate of Al^{+3}. However, higher initial concentrations were reduced considerably in relatively less time than lower concentrations. Therefore, EC process can be considered more effective at the beginning when the concentration is higher than at the end of the operation when the concentration is low.

Effect of inter electrode distance: Reasonable expenditure of low-cost components and low cell voltage are among the advantageous parameters in the EC reactor design. The inter-electrode distance and effective surface area of electrodes are important variable when an operational costs optimization of a reactor is needed. To decrease the energy consumption (at constant current density) in the treatment of effluent with a relatively high conductivity larger spacing should be used between electrodes. But for effluent with low conductivity, energy consumption can be minimized by decreasing the distance between the electrodes (Vik *et al.*, 1984). As can be seen from Table 4, when the inter-electrode distances changed from 0.005 to 0.025 m under 50 Hz and 50 V, the removal (%) of the Ni(II) was decreased from 99.40 to 86.80%. As expected, it appears that conductivity had some effect on the removal efficiency of Ni(II) in the investigated range shown in Table 5. An increase in conductivity require larger spacing be used between electrodes. On the other hand, with increasing electrodes distance, under the same current density, the internal resistance (or the ohmic) drop between electrodes increases and that leads to the

Table 5: The values of resistance (Ω), equivalent conductance and viscosity at current density 0.04 A m^{-2} using 50 Hz and 50 volt with AC

Time (min)	Resistance (Ω)	Equivalent conductance cm^2/Ω.eq	Viscosity 10^{-3} η (Nm^{-2}S)
5	145	291.58	1.003
10	156	341.58	1.001
15	154	389.59	1.010
20	151	447.40	0.999
25	149	526.85	0.995
30	146	570.33	0.900
35	145	575.00	0.982

Table 6: The kinetic variables and pK values for removal of Ni (II) at various pH values

Time (min)	pH	pK	C_t (10^6)	t/c (10^{-5})	$1/C_t$ (10^{-6})
5	7.91	7.94	3.93	1.27	0.2544
10	7.94	8.52	3.42	29.20	0.2924
15	7.97	8.65	3.00	50.00	0.3334
20	7.99	8.79	2.66	75.00	0.3750
25	8.05	8.90	2.38	105.00	0.4201
30	8.09	9.05	2.22	135.00	0.45045
35	8.15	9.17	2.20	159.00	0.4545

Initial concentration of Ni (II): 25 mg L^{-1}, Inter electrode distance: 0.005 m, Current density: 0.04 A m^{-2}, Temperature : 303 K ,at 50 Hz and 50 volt using AC
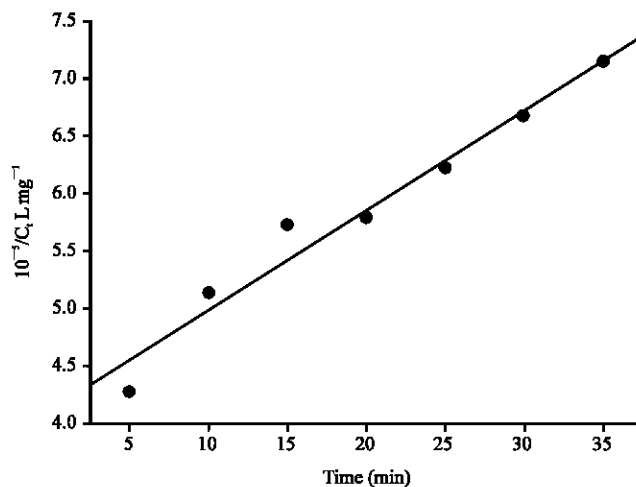

Fig. 2: 10^{-5}/ C_t (L mg^{-1}) versus time (min)

decrease of aluminum ions production and therefore a decrease in removal efficiency. The results of this study agreed with the findings of other researchers (Daida, 2005; Kim *et al.*, 2002).

Kinetics of the adsorption process: As seen in Fig. 2, a pseudo-second-order kinetic model provided a good fit to the experimental results given in Table 6 at 303 K and various pH values. It can be ascertained that a higher value of regression coefficient of r^2 (0.99) for the pseudo- second order kinetics confirms that the adsorption process follows the pseudo-second-order. The kinetic rate law for describing the decrease in Ni(II) concentration with time can be given as:

Table 7: Thermodynamic parameters of adsorption of Ni (II) in EC process

Temperature K	ΔG (kJ mol^{-1})	ΔH [10^3] 10^3 (kJ mol^{-1})	ΔS [10^3] (J/K. mol^{-1})	TΔS (J mol^{-1})
303	-7.310	-3.326	0.0240	7.310
313	-7.480		0.0238	7.480
323	-7.790		0.0241	7.790
333	-8.030		0.0241	8.030
343	-8.273		0.0241	8.273

Initial concentration of Ni (II): 25 mg L^{-1}, Inter electrode distance: 0.005 m, Current density: 0.04 A m^{-2},

$$- dC_t / dt = k_{obs} C_t^2 \qquad (13)$$

where, C_t is the residual concentration of Ni(II) in mg L^{-1} at the time t and k_{obs} (L mg^{-1} min) is the pseudo-second-order rate constant for the adsorption process. Integration of Eq. 12 with boundary conditions C_O at t = 0 to C_t at t and rearrangement results:

$$1/C_t = 1/C_O + k_{obs} t \qquad (14)$$

The slope of the plot 1/C_t versus t was used to calculate the pseudo-second-order rate constant (k_{obs} = 7.1679×10^4 L mg^{-1} min at 303K). Similar behavior have been observed in an electrocoagulation process for the removal of iron from drinking water with aluminum alloy as the anode and stainless steel as the cathode (Vasudevan *et al.*, 2009).

Thermodynamic parameters for the adsorption process: The heat of adsorption ($\Delta H°$) and entropy changes $\Delta S°$ were calculated using Van't Hoff Equation (Boonamnuayvitaya *et al.*, 2004). ln:

$$K_{ads} = -\Delta H / RT + \Delta S / R \qquad (15)$$

The values of $\Delta H°$ and $\Delta S°$ were calculated from the plot of ln K_{ads} versus 1/T and were used to obtain the Gibbs free energy of adsorption ($\Delta G°$) using:

$$\Delta G = \Delta H - T \Delta S \qquad (16)$$

The data given in Table 7 shows that $|\Delta H| < |T\Delta S|$ at all temperatures. This indicates that the adsorption process is dominated by entropic rather than enthalpic changes (Mpofu *et al.*, 2004). Also, the values of Gibbs free energy, enthalpy and entropy show that the adsorption of Ni(II) on metal hydroxides is feasible, spontaneous and exothermic in the temperature range of 303-343 K.

CONCLUSION

This study produced significant information regarding the optimal conditions of several variables that influence nickel removal by electrocoagulation using AC. It was found that Ni(II) removal was achieved mainly by adsorption on metal hydroxides. At a critical Al electrode geometric area of 225 cm^2 and operating time of 5 min, a maximum nickel removal rate of 80% was observed. Optimum electrode distance of 0.005 m under 50 Hz and 50 V was found. The removal efficiency of the studied metal after 35 min of electrolysis time at the constant current density of 0.04 A m^{-2} reached very high values, 99.5% at pH of 7.8. Thermodynamic parameters showed that

the adsorption of Ni(II) on the coagulated product was exothermic and spontaneous in nature in the temperature range of 303-343 K. For nickel removal in the pH range 7.9-8.15, the adsorption process appeared to follow pseudo-second-order kinetics, with a pseudo-second-order rate constant of 7.1679×10^4 L mg^{-1} min at 303 K.

The simplified approach used in this study provides only preliminary data and additional studies are necessary to verify these results and further refine parameters for industrial application.

ACKNOWLEDGMENT

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