Performance Evaluation of Electrocoagulation Technique for Removing Groundwater Hardness of Tikrit University

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Received on: 29/12/2011 & Accepted on: 7/6/2012

ABSTRACT
The performance of Electrocoagulation (EC) process for removal of hardness in groundwater (GW) of Tikrit University in Salahaddin province, north of Baghdad in Iraq has been studied using aluminum (Al) electrodes with both bipolar and monopolar configurations. The effect of initial pH, applied voltage (U), electrolysis time (t), and electrodes configurations on the performance of EC has been investigated. It was found that the best initial pH value to remove hardness (HD) was 9.5. The results indicated that increasing U and t had a positive effect on the hardness removal efficiency (% \( F_{\text{hardness}} \)) to reach 90.4% and it was influenced by the electrodes configuration in which % \( F_{\text{hardness}} \) was 83.5% in bipolar connection compared with 66.2% in monopolar connection. Also, the electrical energy consumption (\( W_{\text{exp}} \)) and (\( W_{\text{theo}} \)) were calculated. It was absorbed, as determined, that there is no significant difference between pseudo-first and second-order kinetic model except at 40 volt that the pseudo-second-order kinetic model fits better than the first-order kinetic model with the data of the electrocoagulation process. Finally, the cost at the most favorable conditions for EC process was calculated resulting that the electrocoagulation process is successfully applied to remove the hardness rate from groundwater with high performance.

Keywords: Groundwater, Hardness, Electrocoagulation, Electrode configuration

دراسة وتقييم كفاءة عملية التخلير الكهربائي في إزالة العسرة من المياه الجوفية الموجودة في منطقة جامعة تكريت

في هذه البحث تم دراسة كفاءة عملية التخلير الكهربائي لإزالة العسرة من المياه الجوفية الموجودة في منطقة جامعة تكريت في محافظة صلاح الدين باستخدام أقطاب الألمنيوم بطرق ربط مختلفة (U), (% \( F_{\text{hardness}} \)), وطريقة ربط الأقطاب على آلة عملية التخلير الكهربائي، وقد جرى أن درجة الحموضوية الأولية المثلثة لإزالة العسرة (9.5) كانت. وأظهرت النتائج إلى (U) % 90,90 وكذلك هي تتأثر بطرق ربط الأقطاب الكهربائية في الخلية حيث كانت نسبة إزالة العسرة %83.5 في الربط ثنائي الطبقات في حين وصلت في الربط حادي الطبق إلى %66.2، كما حسب حساب استهلاك الطاقة الكهربائية في المختبر (% \( W_{\text{exp}} \)) ونظريا (% \( W_{\text{theo}} \))، تم حساب نمذجة وحركة عملية التخلير الكهربائي، وتبين أن درجة حرارية عملية التخلير الكهربائي من الدرجة الثانية كان أفضل واقفاً مما في الدرجة الأولى تبعاً للبيانات المستحقة من التجارب العملية. وخيراً

https://doi.org/10.30684/etj.30.18.12
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INTRODUCTION

Suitable and available water for human consumption is highly limited and likewise, available drinking water has been reduced because of the pollution created naturally and artificially. Among water quality parameters, hardness has always been investigated as an important factor. Hardness which is one of the chemical characteristics of water is caused mainly by the presence of calcium and magnesium \[ \text{Ca}^{2+} \text{and Mg}^{2+}. \]

Hardness of waters varies considerably from place to place. In general, groundwater is harder than surface waters and water gets harder if the amount of calcium and magnesium is increased. Hardness can be expected in regions where large amounts of limestone are found. Hardness ions create a lot of problems for life and industry, therefore knowing the hardness of water is important in evaluating its use as a domestic or industrial water supply.

There are various techniques for the removal of water hardness, such as using electromembrane processes, nano-filtration, chemical or natural substances, ion exchange resins, and electrodialysis. One of these techniques is electrochemical technology such as electrocoagulation process, which is being used for the removal of colloidal and suspended particles, ions, heavy metals, dyes, organic matters and oily matters from water and aqueous environments.

Electrocoagulation is a complex process occurring via serial steps such as; electrolytic reactions at electrode surfaces, formation of coagulants in aqueous phase, adsorption of soluble or colloidal pollutants on coagulants which are removed by sedimentation or flotation.

A simple electro-coagulating batch reactor is made up of anode and cathode with monopolar or bipolar configuration. When a potential is applied from an external power source, the anode material undergoes oxidation, while the cathode will be subjected to reductive deposition of elemental metals.

Following equations, describe EC process in relation to Al electrode:

Anode:
\[ \text{Al} \rightarrow \text{Al}^{3+} + 3\text{e}^- \quad \text{....(1)} \]

Cathode:
\[ 3\text{H}_2\text{O} + 3\text{e}^- \rightarrow \frac{3}{2}\text{H}_2 + 3\text{OH}^- \quad \text{....(2)} \]

On the other hand, at high pH values, both cathode and anode may be chemically attacked by \( \text{OH}^- \) ions
\[ 2\text{Al} + 6\text{H}_2\text{O} + 20\text{OH}^- \rightarrow 2\text{Al}((\text{OH})_4)^- + 3\text{H}_2 \quad \text{....(3)} \]

\( \text{Al}^{3+} \) and \( \text{OH}^- \) ions generated by electrode reactions(1) and (2) react to form various monomeric species such as \( \text{Al}((\text{OH})_4)^- \), \( \text{Al}((\text{OH})_3)^- \), \( \text{Al}_2((\text{OH})_4)^- \), \( \text{Al}((\text{OH})_3)^- \), and \( \text{Al}((\text{OH})_4)^- \).
and polymeric species such as $Al(OH)_{15}^{3+}$, $Al(OH)_{17}^{3+}$, $Al(OH)_{20}^{5+}$, $Al(OH)_{22}^{3+}$, $Al(OH)_{34}^{5+}$, which transform finally into $Al(OH)_{x+y}$ according to complex precipitation kinetics\textsuperscript{[13,14]}.

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+ \quad \ldots (4)$$

Freshly formed amorphous $Al(OH)_{x+y}$ “sweep flocs” have large surface areas which are beneficial for a rapid adsorption of soluble organic compounds and trapping of colloidal particles. Finally, these flocs are removed easily from aqueous medium by sedimentation or $H_2$ flotation\textsuperscript{[15]}. EC technology, compared with other techniques, enjoys some advantages like plain equipment, easy functionality, short resistance time, no need of chemicals, low sludge production, sludge stability, suitable sedimentation of sludge, dewatering and environmental compatibility\textsuperscript{[15]}.

The aim of this study was to investigate EC process performance to remove HD from GW using Al electrodes as a substitute for other water softening techniques and determining the optimal pH, $U$, $t$ and electrode configuration, also calculate the % $E_{\text{hardness}}$, $W_{\text{exp}}$ and $W_{\text{theo}}$. In addition to determinethe order and the kinetic model ofthe obtained data of EC process and its cost.

**MATERIALS AND METHODS**

The GW was collected from many wells located at Tikrit University in Salahaddin Province in the north of Baghdad in Iraq, and its principle characteristics are listed in Table 1. Water samples were taken from the underground hadan average concentration about 2070 mg/l of total hardness rate.

Fig.(1.a,b) shows an overview of EC equipments which include DC power supply (Type SDR 4010, SODILEC Model; 40V, 10A, Japan) with separated sheets of Al electrodes of (100x35x8) mm of 35cm$^2$ an active area and an inter distance ($d$) of 5 and 2 cm in monopolar and bipolar configuration respectively were putting into a plexiglas reactor tank of 170×120×80 mm and 0.5L volume of solution. pH of the sample was adjusted using sulphuric acid and normal sodium hydroxide; the reactor was tested with water samples of different pHs (4.4, 7.2 and 9.5) under four applied voltages (10, 20,30 and 40v).

Under each testing conditions, four reaction durations were tested: 5, 15, 30 and 50 min. Samples were (25 mL) from the middle of the reactor, then filtered in order to remove the formed flocs. Finally, filtered samples were analyzed by atomic absorption spectrophotometer concerning and determined the total hardness removal efficiency % $E_{\text{hardness}}$. At the end of each run, the electrodes were washed thoroughly with dilute acid and water to remove any solid residues on the surfaces, dried and re-weighted. pHsolution was also analyzed at the end of the experiment. In the present study, the% $E_{\text{hardness}}$ by EC process has been evaluated at different condition pH, $U$, $t$ and different electrodes configuration.

$$% E_{\text{hardness}} = \frac{HD_o - HD_f}{HD_o} \times 100\% \quad (5)$$
Where $HD_o$ and $HD_i$ are the concentration of hardness rate before and after EC process respectively. Electrical energy consumption $EEC$ is a very important economical parameter in the electrocoagulation process. The $EEC$ was calculated using the following equation:

$$EEC = \frac{U \cdot I \cdot t}{V}$$  \hspace{1cm} (6)

Where; $EEC$ is the electrical energy consumption (Wh/m\(^3\)), $U$ is the applied voltage (volt), $I$ is the current intensity (Ampere), $t$ is the electrocoagulation time (hr) & $V$ is the volume (m\(^3\)) of the treated water. Also, the amount of electrode dissolved was calculated theoretically by using Faraday's law and compared with the experimental amounts.

$$W_{theo} = \frac{M \cdot U \cdot I \cdot t}{N \cdot F \cdot V}$$  \hspace{1cm} (7)

Where $W_{theo}$ (kg Al electrode/m\(^3\) of treated water) is the theoretical amount of ion produced by current intensity $I$ (Ampere) passed for a duration of operating time $t_{EC}$ (sec), $N$ is the number of electrons involved in the oxidation/reduction reaction; for Al $N_{Al}=3$, $M$ is the atomic weight of anode material ($M$ for Al = 0.02698 kg/mol), $F$ is the Faraday's constant (96485 C/mol) and $V$ is the volume (m\(^3\)) of the GW in the EC reactor. Hence, the amount of aluminum adsorption increased with the increase in adsorbent concentration, which indicated that the adsorption depended on the availability of binding sites for aluminum hydroxides flocs\(^{[16]}\).

**RESULTS AND DISCUSSION**

**Effect of pH on Hardness Removal**

The pH of the solution is one of the most important parameters that govern the removal efficiency in the EC process. To examine this effect, the sample was adjusted to the desired pH for each experiment by using 0.1M NaOH solution and 0.1M H\(_2\)SO\(_4\) solution. As showed in Fig.(2) at 20 volts, when pH increased from 4.4 to 9.5, the %Hardness increased from 38.4 to 78.8% at 15 min and from 42.5 to 89.2% at 30 min.

Since the effect of pH on coagulants depend on the produced reactions on different conditions.

In neutral conditions:

$$3Al_{(s)} + 8H_2O_{(l)} \rightarrow Al(OH)_{2(s)} + 2Al(OH)_2 + 4H_2(g)$$  \hspace{1cm} (8)

In acid conditions:

$$2Al_{(s)} + 6H_2O_{(l)} \rightarrow Al(OH)_{3(s)} + 3H_2O_{(g)}$$  \hspace{1cm} (9)

In alkali conditions:

$$2Al + 6H_2O \rightarrow 2Al(OH)_{3(s)} + 3H_2O_{(l)}$$  \hspace{1cm} (10)
Here, $\text{Al(OH)}_3$ and $\text{Al(OH)}_2$ settled while, $\text{H}_2$ moved upward and caused flotation. As reactions showed, in acidity condition $\text{Al(OH)}_2$ and in alkali condition $\text{Al(OH)}_3$ were produced. Since $\text{Al(OH)}_3$ had higher weight and density, it settled faster and had higher efficiency. Therefore, it acts better in enmeshment in a precipitate. Hence, based on the results of the present study and previous studies electrocoagulation process can act as pH moderator $^{[17,18]}$.

**Effect of Different Electrode Connections**

In order to improve the hardness removal efficiency from the ground water, electrode configuration can have a justified effect on the sludge formation as well as on the corrosion of the electrode. Effect of electrode configuration (monopolar and bipolar) for the $\% E_{\text{hardness}}$ by EC is shown in Fig.(3). Investigation was performed for softening $\text{GW}$ of 2070mg/l total HD with constant other parameters of (5cm) distance between electrodes, $t$ of (10min) and $U$ were maintained to be (10-40volt) during the experiments. It was observed that with the passage of time hardness concentration inside the EC bath was decreased for both electrode connections. It is also seen from Fig.(3) that the $\% E_{\text{hardness}}$ for actual acidity of 7.2 pH and for 10 min time at 10 to 40 volt in monopolar connection were 25.2% to 66.2%, whereas for bipolar connection, were 42 to 83.5% respectively. In bipolar connection, a higher surface area compared to that of monopolar connection favored the adequate anodic oxidation. As a result, with the same applied voltage or potential (current applied) for both kind of connection, the intensity is higher in the bipolar connection, therefore the $\% E_{\text{hardness}}$ of the $\text{GW}$ were found more than that was observed in monopolar electrode connection $^{[19]}$.

**Effect of applied potential and Time**

The effect of $U$ on the $\% E_{\text{hardness}}$ were studied at 10, 20, 30 and 40 volt. Fig.(4) illustrates the effect of these $U$ on $\% E_{\text{hardness}}$ for various electrolysis time. As the electrolysis time increases, comparable enhancements in the $\% E_{\text{hardness}}$ were observed. Fig.(4) shows that at pH of 7.2, $\% E_{\text{hardness}}$ were achieved from 18.9 to 54.6% after 5min, 32.4 to 74.8% for 15min, 38.7 to 88.8% for 30min and 54.2 to 90.3% after 50min of electrolysis from 10 to 40 volt respectively.

As the applied voltage increased, the removal efficiency of hardness increased. Simultaneously, the sufficient current passing through the solution rise due to the increased applied voltage. Due to sufficient current through the solution, the metal ions generated by the dissolution of the sacrificial electrode were hydrolyzed to form a series of metallic hydroxide species. However, it took about 15 min to reach over 75% of hardness removal, and 30 min to reach 88.8 when the applied voltage was 40V while under 20 and 40 volt it took 50 min to reach 72% and 90.3% respectively. This is primarily due to an insufficient amount of electric power supplied at 10V for the complete destabilization of the suspended metallic hydroxide species in the solution. Both 30 and 40 volts are suitable for this electrocoagulation experiment; also 30 to 50 min was the requiring treatment times to reach over 88-90% hardness removal. Therefore the optimal reaction time is 30-50 min for this equipment considering the treatment cost and efficiency. As time progressed and dissolved coagulants at the aluminum electrode increased, there was
an increase in the removal efficiency, which could be explained by a sufficient amount of coagulant dissolving from the aluminum electrode to effectively reduce the layer of the suspended metallic hydroxide species to destabilize the metallic hydroxide species. In order to investigate the optimum applied voltage, the consumption of the specific energy for an applied voltage during electrocoagulation process was evaluated.\(^\text{[20, 21]}\)

**Electrodes Metal and Specific Energy Consumption**

The synthetic GW with high concentrations HD were softened in terms of % \(E_{hardness}\) and specific \(EEC\) by aluminum electrocoagulation. Table 2 shows that the energy consumption in EC cell of 7.2 pH solution increased from 0.0167 kWh/m\(^3\) at 10 volt and 5 min to 4.667 kWh/m\(^3\) at 40 volt and 50 min with increasing the removal efficiency from 18.4 to 90.3% in monopolar connection, in bipolar connection increased from 0.033 to 0.220 kWh/m\(^3\) at rate of removal efficiency from 42 to 89.7% through 10 min. Table 2 also represents a comparison between the theoretical and experimental aluminum amount that released to solution when voltage from 10 to 40 volts was applied in EC cell at pH 7.2 for various duration time from (5-50 min). Theoretical aluminum amount was calculated by Faraday’s law, equation (7). \(^\text{[22-24]}\)

**Kinetic Study of Hardness Removal**

The overall apparent kinetics of electrocoagulation process of hardness removal is described by a macro-kinetics model in which the rate constant depends on the applied voltage (or current density). This model provides preliminary data for evaluating the reaction rate constants. The kinetic rate equation for representing the removal rate of hardness concentration from the GWs described by the following \(m\)th order reaction kinetics:

\[
\frac{dC_i}{dt} = -kC_i^m
\]

where \(C\) represents the hardness concentration, \(m\) is the order of reaction, \(k\) is the reaction rate constant, and \(t\) is the time. For a first-order reaction:

\[
\ln\left(\frac{C_i}{C_o}\right) = -k_1 t
\]

The slope of the plot of \(\ln\left(\frac{C_i}{C_o}\right)\) versus time gives the value of the rate constant \(k_1\), min\(^{-1}\). Here, \(C_o\) is the initial concentration in milligrams per liter, \(C_i\) is the concentration value in milligrams per liter at time \(t\).

For a second-order reaction:

\[
\frac{1}{C_i} - \frac{1}{C_o} = k_2 t
\]

The slope of the plot of \(\frac{1}{C_i}\) versus time gives the value of the rate constant \(k_2\), lmg\(^{-1}\)min\(^{-1}\). The values of \(k\) with first-order and second-order models for hardness removal at various applied voltages were determined graphically and are shown in Table 3, respectively for monopolar and bipolar configuration. The conformity between experimental data and the model values was evaluated by the correlation values \((r^2)\). As can be seen in Table 3, regardless of the higher applied voltage, the
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The $r^2$ value for the second-order model was slightly higher than that for the first-order model. When fixing the other experimental conditions, but increasing the applied voltage from 10 to 20 volts, the first-order rate constant $K_1$ increased from 1.1074 to 2.4212 E-2 min$^{-1}$ while at 30 to 40 volts, the second-order rate constant $K_2$ increased from 5.6579 to 9.5354E-2 (L/g.min). It is very important to note that fast hardness removal took place at a short electrolysis time; this is considered a great advantage of using the electrocoagulation process [25, 26].

Cost of the EC process

Preliminary estimate of the cost of the removal of groundwater hardness by electrochemical process has been done considering the energy cost and the cost of electrode. From the kinetic constants obtained in the study, amount of electric power required for desired hardness reduction can be computed. The cost of electrical energy is variable in different parts of the world. Considering, as in Iraq, the cost of electrical energy about 0.02 US$ per KWh and the cost of electrode is 0.003 US$ per gram of aluminum metal. The cost of the process is determined by the sacrificial electrode loss and the electrical energy consumption. In the most favorable conditions 40 volt potential applied, 5 cm distance between the electrodes 7.2 pH and 50 min time, the maximum possible hardness reduction was reached 90.3%. Then, the power consumption expected from equation (6) would be 2.5 kWh/m$^3$ for an average final value of 200 mg hardness/L, and the sacrificial electrode loss is 27.9 g/m$^3$ of aluminum as calculated in equation (7) [27, 28].

Operating cost = $aC_{\text{energy}} + bC_{\text{electrode}}$  

where $C_{\text{energy}}$ (kWh/m$^3$) and $C_{\text{electrode}}$ (kg Al/m$^3$) are consumption quantities for the Al removal, which are obtained experimentally. “a” electrical energy price US$/kWh; “b” electrode material price US$/kg Al. Cost due to electrical energy (kWh/m$^3$) is calculated using above values.

Operating Cost: 0.1337 US$/m^3$ of solution

According to this value of cost, the electrocoagulation process is successfully applied to remove the hardness rate from groundwater with high efficiency.

CONCLUSIONS

Batch EC studies with various experimental parameters such as initial pH, applied voltage, operating time and electrodes configuration were performed to evaluate the influence of electrode connection modes on the removal of hardness from groundwater. The EC process at the optimum conditions was able to softening the groundwater to acceptable range of hardness rate by Al electrodes. The highest removal efficiencies of hardness rate is 90.3% at pH 7.2, 40 volt and 50 min. Values of kinetic rate constants for hardness rate removal at various applied voltages were calculated. The kinetic results show that there is no significant difference between pseudo-first and second-order kinetic model except at 40 volt that the pseudo second-order kinetic model matched satisfactorily with the experimental observations. The lowest operating costs for electricity consumption and Al electrodes consumption were also obtained. So, the
electrocoagulation process is successfully applied to remove the hardness from groundwater with high performance.

REFERENCES
Table (1) Characteristics of Groundwater

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<thead>
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<th>Characteristics</th>
<th>Value</th>
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<tbody>
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<td>Total Hardness (mg/l)</td>
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<td>Ca^{2+} (mg/l)</td>
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<td>Conductivity (µS/cm)</td>
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Table (2) Comparison of the Calculated Theoretical Metal Dissolution W_{theo} with the Measured Experimental W_{exp} and the Energy Consumption for Different Connections

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<tr>
<th>Time (min) Electrodes Connection</th>
<th>Voltage (volt)</th>
<th>HD%</th>
<th>W_{exp} kg/m³</th>
<th>W_{theo} kg/m³</th>
<th>EEC kwh/m³</th>
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Table (3) Pseudo-Kinetic Rate Constants with First-Order and Second-Order Models for Hardness Removal at Various Applied Voltages.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Applied Voltage (volt)</th>
<th>$1^{st}$ order model</th>
<th>$2^{nd}$ order mode</th>
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<td></td>
<td>$-k_1 \times 10^{-2}$</td>
<td>$r^2$</td>
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<td>Monopolar</td>
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<td>(min)$^{-1}$</td>
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</tbody>
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Figure (1) Schematic Diagram of EC Cell for the Groundwater Hardness Removal of a) Monopolar Connection and b) Bipolar Connection. (1) electrodes; (2) groundwater; (3) electrode support; and (4) DC power supply.
Performance Evaluation of Electrocoagulation Technique for Removing Groundwater Hardness of Tikrit University

Figure (2) Effect of pH on HD% removal.

Figure (3) Effect of Electrodes Connection on HD% removal.

Figure (4) The Effect of Voltage and Time on HD% removal.