

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

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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 (% $E_{hardness}$) to reach 90.4% and it was influenced by the electrodes configuration in which % $E_{hardness}$ was 83.5% in bipolar connection compared with 66.2% in monopolar connection. Also The electrical energy consumption % $E_{hardness}$ and the experimentally and theoretically electrodes consumption (W_{exp}) and (W_{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 التخثير الكهربائي في إزالة العسرة من المياه الجوفية الموجودة في منطقة جامعة تكريت

الخلاصة

في هذه البحث تم دراسة كفاءة عملية التخثير الكهربائي لإزالة العسرة من المياه الجوفية الموجودة في منطقة جامعة تكريت في محافظة صلاح الدين باستخدام أقطاب الألمنيوم بطرق ربط مختلفة أحادية وثنائية القطب. وقد تم دراسة تأثير بعض المعايير مثل الرقم الهيدروجيني الأولي pH، والجهد المزدور (U)، والوقت (t)، وطريقة ربط الأقطاب على أداء عملية التخثير الكهربائي. وقد وجد أن درجة الحموضة الأولية المثلى لإزالة العسرة (HD) كانت 9.5. وأشارت النتائج إلى أن زيادة U و t كان لها أثر إيجابي على كفاءة إزالة العسرة لتصل إلى 90.4%، وكذلك هي تتأثر بطرق ربط الأقطاب الكهربائية في الخلية حيث كانت نسبة إزالة العسرة 83.5% في الربط نوع ثنائي القطبين في حين وصلت في الربط أحادي القطب إلى 66.2%. كما تم حساب استهلاك الطاقة الكهربائية واستهلاك الأقطاب تجريبيًا (W_{exp}) ونظريًا (W_{theo}). كما تم حساب نموذج وحركية عملية التخثير الكهربائي، وتبين أن درجة حركية عملية التخثير الكهربائي من الدرجة الثانية كان أفضل وأدق مما في الدرجة الأولى تبعاً للبيانات المستحصلة من التجارب العملية. وأخيراً

تم حساب الكلفة الاقتصادية لهذه العملية والتي أشارت إلى نجاح عملية التخثير الكهربائي في إزالة العسرة من المياه الجوفية.

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^[1]. 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^[2]. Hardness ions create a lot of problems for life and industry^[3], therefore knowing the hardness of water is important in evaluating its use as a domestic or industrial water supply^[2].

There are various techniques for the removal of water hardness, such as using electromembrane processes, nano-filtration^[4] chemical or natural substances^[5], ion exchange resins^[6], and electrodialysis^[7].

Recently, growing demand for high quality water has justified the development of modern and low cost technologies for hard and very hard water softening^[4]. One of these techniques is electrochemical technology such as electrocoagulation process, which is being used for the removal of colloidal and suspended particles, ions^[7] heavy metals^[8], dyes,^[9] organic matters^[10] and oily matters^[11] 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^[3].

A simple electro-coagulating batch reactor is made up of anode and cathode with monopolar or bipolar configuration^[12]. 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^[13].

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

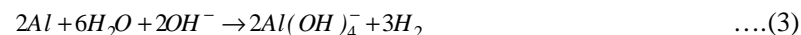
Atanode:



Atcathode:



On the other hand, at high pH values, both cathode and anode may be chemically attacked by OH⁻ ions



Al³⁺ and OH⁻ ions generated by electrode reactions (1) and (2) react to form various monomeric species such as Al(OH)²⁺, Al(OH)₂⁺, Al₂(OH)₂⁺, Al(OH)₄,

and polymeric species such as $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$, $Al_{13}(OH)_{34}^{5+}$, which transform finally into $Al(OH)_3(s)$ according to complex precipitation kinetics^[13,14].



Freshly formed amorphous $Al(OH)_3(s)$ "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^[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^[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_{hardness}, W_{exp} and W_{theo}. In addition to determine the order and the kinetic model of the 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 had an 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 35cm² 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 170x120x80 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_{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-weighed. pH solution was also analyzed at the end of the experiment. In the present study, the %E_{hardness} by EC process has been evaluated at different condition pH, U, t and different electrodes configuration.

$$\%E_{hardness} = \frac{HD_o - HD_t}{HD_o} \times 100\% \quad (5)$$

Where HD_o and HD_t 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:

$$EEC = \frac{U \cdot I \cdot t}{V} \quad (6)$$

Where; EEC is the electrical energy consumption (Wh/m³), U is the applied voltage (volt), I is the current intensity (Ampere), t is the electrocoagulation time (hr) & V is the volume (m³) of the treated water. Also, the amount of electrode dissolved was calculated theoretically by using Faraday's law and compared with the experimental amounts.

$$W_{theo} = \frac{M \cdot U \cdot t_{EC}}{N \cdot F \cdot V} \quad (7)$$

Where W_{theo} (kg Al electrode/m³ 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 (96485C/mol) and V is the volume (m³) 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].

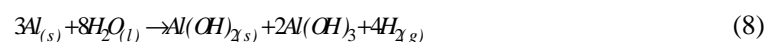
RESULTS AND DISCUSSION

Effect of pH on Hardness Removal

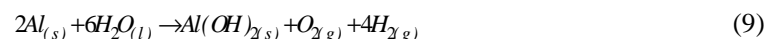
The pH of the solution is one of the most important parameters that govern the removal efficiency in the EC process. To examine this effect, the sample was adjusted to the desired pH for each experiment by using 0.1M NaOH solution and 0.1M H₂SO₄ solution. As showed in Fig.(2) at 20 volts, when pH increased from 4.4 to 9.5, the % $E_{hardness}$ 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.

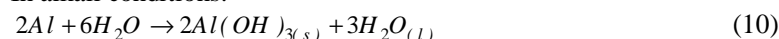
In neutral conditions:



In acid conditions:



In alkali conditions:



Here, $Al(OH)_3$ and $Al(OH)_2$ settled while, H_2 moved upward and caused flotation. As reactions showed, in acidity condition $Al(OH)_2$ and in alkali condition $Al(OH)_3$ were produced. Since $Al(OH)_3$ 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 $\%E_{hardness}$ by EC is shown in Fig.(3). Investigation was performed for softening 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 $\%E_{hardness}$ 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 $\%E_{hardness}$ of the 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 $\%E_{hardness}$ were studied at 10, 20, 30 and 40 volt. Fig.(4) illustrates the effect of these U on $\%E_{hardness}$ for various electrolysis time. As the electrolysis time increases, comparable enhancements in the $\%E_{hardness}$ were observed. Fig.(4) shows that at pH of 7.2, $\%E_{hardness}$ 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% $\%E_{hardness}$ 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^[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/m³ at 10 volt and 5 min to 4.667 kWh/m³ 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/m³ 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 a 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)^[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 GW is described by the following m^{th} order reaction kinetics:

$$\frac{dC_t}{dt} = -kC_t^m \quad \dots(11)$$

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:

$$\ln\left(\frac{C_t}{C_o}\right) = -k_1 t \quad \dots(12)$$

The slope of the plot of $\ln \frac{C_t}{C_o}$ 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_t is the concentration value in milligrams per liter at time t .

For a second-order reaction:

$$\frac{1}{C_t} - \frac{1}{C_o} = k_2 t \quad \dots(13)$$

The slope of the plot of $1/C_t$ versus time gives the value of the rate constant k_2 , lmg⁻¹min⁻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

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 volt, the first-order rate constant K_1 increased from 1.1074 to 2.4212 E-2 min⁻¹ while at 30 to 40 volt, the second-order rate constant K_2 increased from 5.6579 to 9.5354 E-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/m³ for an average final value of 200 mg hardness/L, and the sacrificial electrode loss is 27.9 g/m³ of aluminum as calculated in equation (7)^[27,28].

$$\text{Operating cost} = aC_{\text{energy}} + bC_{\text{electrode}} \quad \dots(14)$$

where C_{energy} (kWh/m³) and $C_{\text{electrode}}$ (kg Al/m³) 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/m³) is calculated using above values.

Operating Cost: 0.1337 US\$/m³ 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 soften 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.

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**Table (1) Characteristics of
Groundwater**

Characteristics	Value
T (°C)	22
pH	7.2
Total Hardness (mg/l)	2070
Ca ⁺² (mg/l)	640
Mn ²⁺ (mg/l)	89
Cl ⁻ (mg/l)	250
SO ₄ ⁻² (mg/l)	700
NO ₃ ⁻ (mg/l)	22
Turbidity (NTU)	2
Conductivity (μS/cm)	1210

**Table (2) Comparison of the Calculated Theoretical Metal Dissolution
W_{theo} with the Measured Experimental W_{exp} and the Energy
Consumption for Different Connections**

Time (min) Electrodes Connection	Voltage volt	HD%	W _{exp} kg/m ³	W _{theo} kg/m ³	EEC kwh/m ³
5 Mono-polar	10	18.9	0.59	0.56	0.0167
	20	24.5	2.1	2.13	0.1267
	30	40.5	2.5	2.8	0.2500
	40	54.6	3.2	3.92	0.4667
15 Mono-polar	10	32.4	1.72	1.68	0.050
	20	44.4	5.8	6.34	0.380
	30	58.6	8.56	8.39	0.750
	40	74.8	11.5	11.74	1.40
30 Mono-polar	10	38.7	3.6	3.36	0.1
	20	57	13.1	12.75	0.76
	30	77.1	17.2	16.78	1.5
	40	74.8	25	23.5	2.8
50 Mono-polar	10	54.2	6.1	5.6	0.1667
	20	77.3	20.8	21.3	1.270
	30	85.3	28.2	27.9	2.500
	40	90.3	27.2	39.1	4.700
15 bipolar	10	42	1.5	1.2	0.0334
	20	65	2.3	2	0.1208
	30	81.5	2.6	2.5	0.2204
	40	89.7	3.1	3.4	0.1336

Table (3) Pseudo-Kinetic Rate Constants with First-Order and Second-Order Models for Hardness Removal at Various Applied Voltages.

Parameters	Applied Voltage (volt)	1 st order model		2 nd order mode	
		$-k_1 \times 10^{-2} \text{ (min)}^{-1}$	r^2	$k_2 \times 10^{-2} \text{ (L/g.min)}$	r^2
Monopolar Connection	10	1.4074	0.9774	1.0519	0.9707
	20	2.4212	0.9918	2.4212	0.9881
	30	3.6648	0.9835	5.6579	0.9946
	40	4.4332	0.8731	9.5354	0.9437
Bipolar Connection	10	5.4473	0.9826	3.4983	0.9999

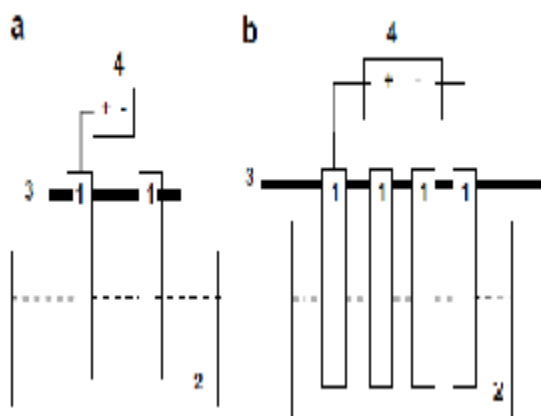
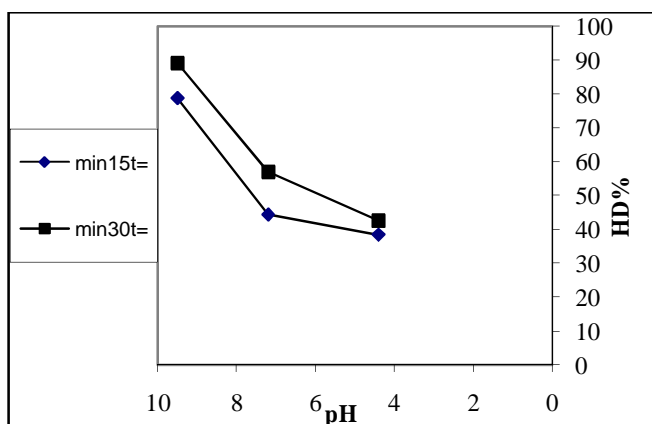
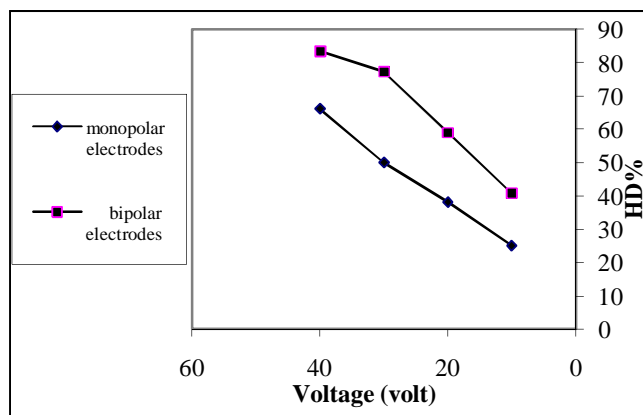


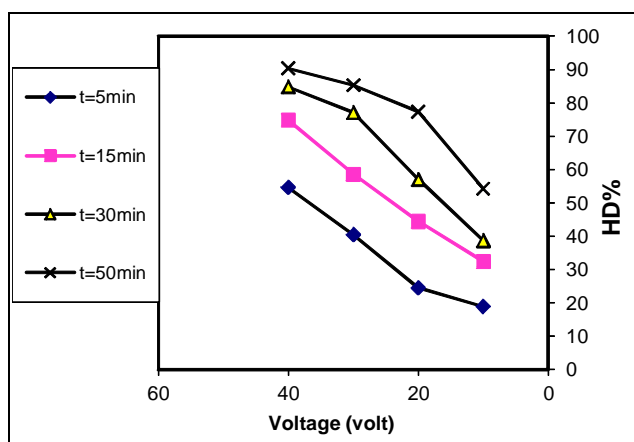
Figure (1) Schematic Diagram of EC Cell for the Groundwater Hardness Removal of a) Monopolar Connection and b) Bipolar Connection. (1) electrodes; (2) groundwater; (3) electrode support; and (4) DC power supply.



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.