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Electrocoagulation for Treatment of Simulated Blowdown Water Of Cooling Tower

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ABSTRACT

This study investigates the results of electrocoagulation (EC) using aluminum (Al) electrodes as anode and stainless steel (grade 316) as a cathode for removing silica, calcium, and magnesium ions from simulated cooling tower blowdown waters. The simulated water contains (50 mg/l silica, 508 mg/l calcium, and 292 mg/l magnesium). The influence of different experimental parameters, such as current density (0.5, 1, and 2 mA/cm^2), initial pH(5,7, and 10), the temperature of the simulated solution(25^{0}C and 35 ^{0}C), and electrolysis time was studied. The highest removal efficiency of 80.183%, 99.21%, and 98.06% for calcium, silica, and magnesium ions, respectively, were obtained at a current density of 1 mA/cm^2, initial PH=7, the temperature of 25^{0}C and treatment time 60 min. The results have shown the ability of the EC process to remove silica and hardness ions from CTB water.

Keywords: Cooling towers blowdown water, Electrocoagulation, Silica removal, Calcium, and magnesium ions removal.

التخثير الكهربائي لمعالجة مياه التصريف المحاكاة لبرج التبريد

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الخلاصة

تبحث هذه الدراسة في نتائج التخثير الكهربائي باستخدام قطب انود من الالمنيوم وقطب كاثود من الحديد المقاوم للصدأ لإزالة أيونات السليكا وايونات الكالسيوم والمغنيسيوم من مياه التصريف المحاكاة لبرج التبريد. تحتوي المياه المحاكاة على (50 ملغم / لتر من السليكا, 508 ملغم / لتر من الكالسيوم و 292 ملغم / لتر من المغنسيوم). تمت دراسة تأثيرالظروف التجريبية المختلفة، مثل كثافة التيار (0.5 و1 و 2 مللي أمبير / سم^2) ، الأس الهيدروجيني الابتدائي (5،7 و 10) ، ودرجة حرارة المحلول (25 درجة مئوية و 35 درجة مئوية) وزمن التحليل الكهربائي، اعلى كفاءة إزالة 80.183 ٪ ،99.21 ٪ ، و 98.06 ٪ لأيونات الكالسيوم والسليكا والمغنيسيوم ، على التوالي تم الحصول عليها عند كثافة التيارمساوية1 مللي أمبير / سم ^{2} ،والقيمة الابتدائية

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للاس الهيدروجيني = 7 ، درجة حرارة 25درجة مئوية ووقت المعالجة 60 دقيقة. أظهرت النتائج قدرة عملية التخثير الكهربائي على إزالة أيونات السليكا وايونات العسرة الكالسيوم والمغنيسيوم من مياه التصريف المحاكاة لأبراج التبريد.

الكلمات الرئيسية: مياه التصريف لأبراج التبريد، التخثير الكهربائي ، إزالةالسليكا ، إزالة أيونات الكالسيوم والمغنيسيوم.

1. INTRODUCTION

Along with growing water contamination, and the severe shortage of water, the reuse of water has gained attention from the managers in diverse industries. Astonishingly, 70% of industrial water is utilized as cooling water in cooling towers (**Zhi et al., 2013**; **Koeman-Stein *et al.*, 2016).** Cooling towers are large construction to extract heat via evaporation of waters. Their applications were widespread in many industries, for example, petrochemical plants, oil refineries, power plants, food industries, and textile plants, etc. (**Hafez et al., 2018).**

Water sources involve various concentrations of suspended and dissolved solids (**Abdel-Shafy et al., 2016**). However, continuous evaporation of circulating water inside the tower causes an increase in the concentration of solids and insoluble mineral salts and leads to scale formation on heat transfer surfaces. Silica and hardness ions Ca^{+2} and Mg^{+2} are the most ones that contribute to the formation of deposition and scaling in cooling towers (**Lieet al., 2011).**

To improve the heat transfer and protect the cooling system from scaling, an amount of circulating water is withdrawn continuously from the tower as blowdown water and simultaneously replaced by fresh water, as demonstrated in **Fig.1**. The quantity of blowdown discharged is controlling the levels of dissolved and suspended solids in the system. Decreasing the blowdown rate increases the solids and vice versa (**Hafezet al., 2018).**

The common methods for removal of silica and hardness ions practically from industrial water comprise the addition of antiscalant such as (amino trimethylene phosphonic acid, hydroxyl ethylidene, 1,1-diphosphonic acid, diethylene triamine pentane, methylene phosphonic acid, etc.), which also contribute to contaminating the water through inhibiting scaling (**Hakizimana et al., 2016; Neveux et al., 2016).** Reverse osmosis and ion exchange are also used, which produce high amounts of brine streams needing disposal **(Jiang et al., 2017; Tong et al., 2017).** In lime softening processes, high quantities of lime sludge streams are discharged **(Stewart et al., 2011).** Chemical coagulation treatment which utilizes conventional coagulant such as alum and ferric chloride ($Fecl_3$); these salts produce undesired ions for cooling water like chloride (corrosive) and sulfate (scalene) **(Gelover – Santiago et al., 2014).**

One of these conventional processes to defeat the drawbacks of traditional water and wastewater treatment is electrocoagulation. EC is a green technology that mixes the responsibilities and advantages of electrochemistry, next to conventional chemical treatment **(Demirbas** and **Kobya, 2017**). EC technology has gained interest in researches due to its simplicity, no addition to any chemicals, high removal efficiency, low sludge production, and no formation of any brine stream that needs removal. So the cost of treatment declines (**Khandegar** and **Saroha, 2013; Naje et al., 2017; Hakizimana et al., 2017).**

Few studies have proposed the treatment of scaling ions formation like Ca^{+2}, $Mg^{+2,}$ and silica ions in the CTB water via electrocoagulation. **(Lio et al. 2009)** study includes the effect of pH value, different antiscalant compounds, and coagulant aids on the performance of EC using aluminum (Al) and iron (Fe) electrodes for treatment of CTB water containing silica, calcium, and magnesium ions. (**Schulz et al. 2009**) investigated the effectiveness of the EC process utilizing Al and Fe electrodes for removing silica, $Ca^{+2,}$ and Mg^{+2} from CTB water. **(Hafez et al., 2018)** used three different electrodes Al, Fe, and Zinc, in the EC system for the treatment of CTB water to remove silica and hardness ions (calcium and magnesium) and characterized the slug production. However, the effect of temperature on scale forming species removal by EC was never studied previously.

The aim of this work is to show the efficiency of the EC process with aluminum electrodes for removing silica, calcium, and magnesium ions from cooling tower blowdown water. Also, study the effect of operational parameters on the removal efficiency.

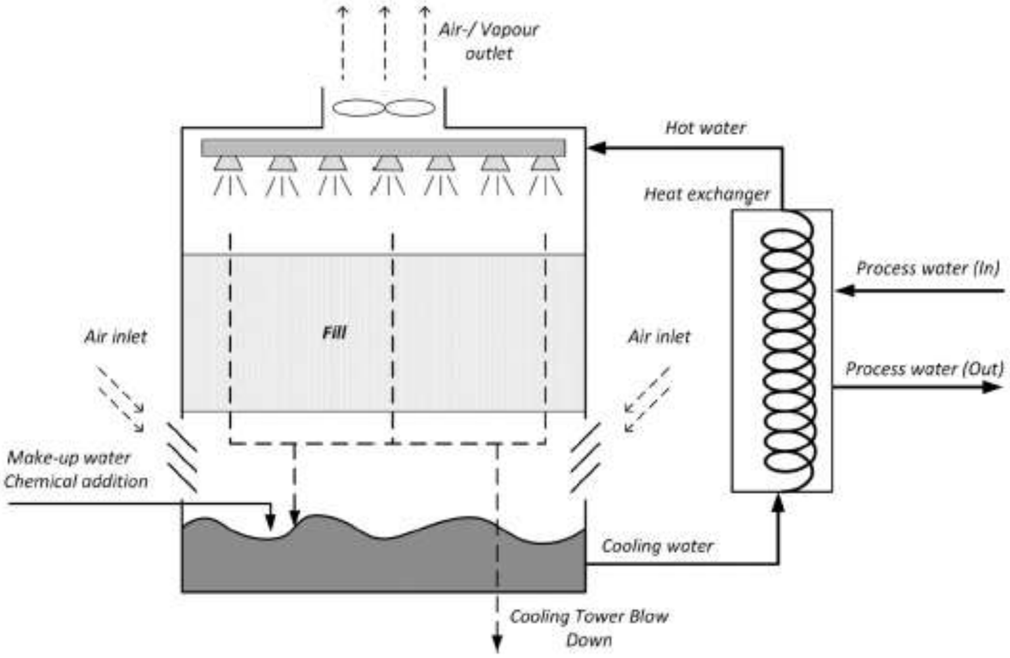


Figure1 . Schematic representation of the operation of the cooling tower **(Löwenberg et al., 2015).**

2. ELECTROCOAGULATION MECHANISM

EC process is a combination of chemical and physical phenomena utilized for treating wastewater. It consists of an electrochemical cell with two electrodes or more. Aluminum or iron is usually used as anode and cathode, immersed in wastewater, and connected to a digital power supply. During the electrolysis, and with the current flow, the essential chemical reactions occur, including the dissolution of metal electrodes at the anode by oxidation to produce cations (Al^{+3}, Fe^{+3}, etc.). This destabilizes pollutants and neutralizes charges and generates hydroxyl and hydrogen due to water reduction at the cathode, as explained in **Fig.2**. The amounts of cation formed due to anode metal dissolution are following Faraday's law **(A-Mohammed, 2007), Eq.1**

$$m=\frac{I\times t\times M}{Z\times F} \qquad (1)$$

Where: m is the amount of anode metal consumption (gram), *I* is the applied current (Ampere), *t* is the treatment time (sec), *F* is Faraday's constant (96,485 C/mole), M is the molecular weight of anode electrode (g/mole) and ***Z*** the number of electrons released from anode metal.

The reaction at the electrodes is described in **Eq. 2.** and **3 (Kamaraj** and **Vasudevan, 2015)**:

At the anode:

$$Al_{(s)} \longrightarrow Al^{+3}_{(aq)} + 3\,e^{-} \tag{2}$$

At the cathode**:**

$$2H_2O + 2e^{-} \longrightarrow H_2\,(g) + 2\,OH^{-} \tag{3}$$

These cations like (Al^{+3}) or other types of metals cations also yield a monomeric or polymeric hydroxide complex. Many monomeric species like $Al(OH)^{2+}$, and $Al(OH)^{4-}$and polymeric species like $A_{l6}(OH)_{15}^{3+}$ $Al7(OH)_{17}^{4+,}$ $Al8(OH)_{20}^{4+}$change into $Al(OH)_3$. These insoluble $Al(OH)_3$ stay in the aqueous medium and destabilize contaminants by electrostatic attraction followed by coagulation**(Nayak, 2018)**as shown in **Eq.4**:

$$Al^{+n}_{(aq)} + n\,OH^{-}_{(aq)} \longrightarrow Al(OH)^{n}_{(s)} \tag{4}$$

The bubbles of hydrogen at the cathode and oxygen at the anode contribute to improving separation and rise of pollutant to the surface of the solution by flotation.

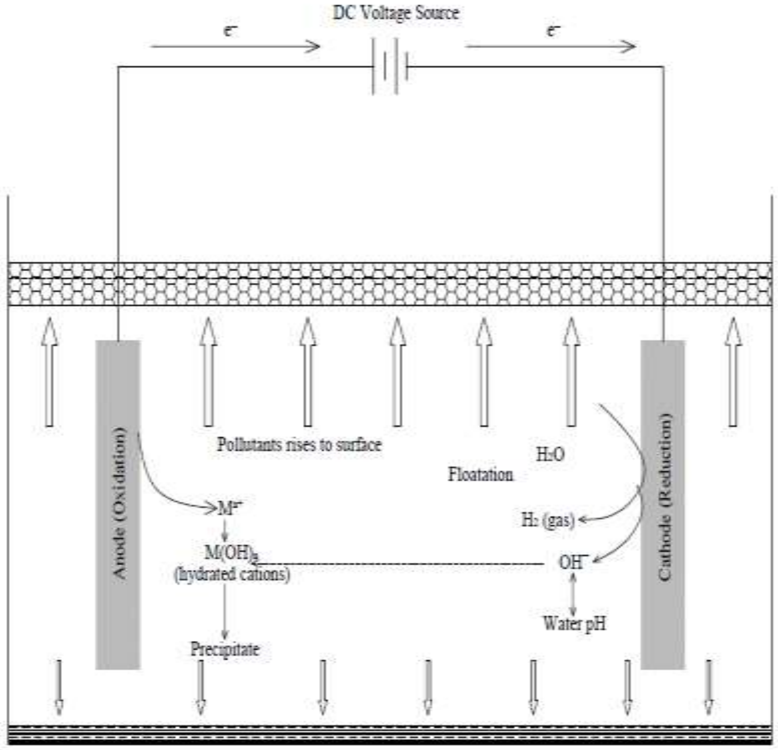


Figure 2. Reactions inside the electrocoagulation cell **(Bharath et al., 2018).**

3. EXPERIMENTAL WORK

3.1. Wastewater preparation

Simulated CTB water with the initial concentration of each ion is prepared by dissolving an appropriate amount of sodium metasilicate nonahydrate ($Na_2SiO_3 . 9H_2O$), $MgCl_2$ and $CaCl_2$ salts in deionized water. The needed mass of each salt could be calculated according to **Eq.5 (Mohammed and AlJaberi, 2018),** and weighted using a digital balance (BL210s – Sartoruis – 4 digits).

$$W= V \times Ci \times \left(\frac{M}{Aw}\right) \tag{5}$$

where:
W: Weight of salts, (g)
V: Volume of simulated CTB (liter)
Ci: Initial ions concentration (mg/l)
M: Molecular weight of salts (g/mole)
Aw: Atomic weight of ions (g/mole)

3.2. Experimental set-up and procedure

Fig. 3 illustrates the experimental electrocoagulation unit. The EC cell is made of glass having dimensions of (16×16×12) cm. Two electrodes of aluminum act as an anode. The other is stainless steel electrode as a cathode was placed between the two aluminum electrodes of the same size 13cm ×13cmx0.1mm, with a total effective surface area of 260 cm^2. They are vertically situated in the cell at 1.5cm distance between electrodes and connected to the digital power supply (SYADGONG Company- model: PS-305D). In each run, 2800 ml of the sample is placed into the electrolysis cell and stirred at 250 rpm using a magnetic stirrer (Stuart, CB 162). Then the pH of the simulated solution was adjusted by the addition of 0.1M HCL solution of 0.1M NaOH solution and measured using digital pH meter (ATC Company). The electrodes are immersed in the solution and connected to the power supply and connected with ammeter and voltmeter (type, Proskit, MT-1707). On switching the power supply, the current was adjusted to the desired value. Total treatment time of 60 min and 600 ml of samples were taken from the middle of the reactor by pipette at each 12 min interval, then analyzed. At the end of each experiment, the cell of EC was cleaned, the electrode was polished with sandpaper and dipped in 0.1M HCl solution for 2 min to remove any impurities.

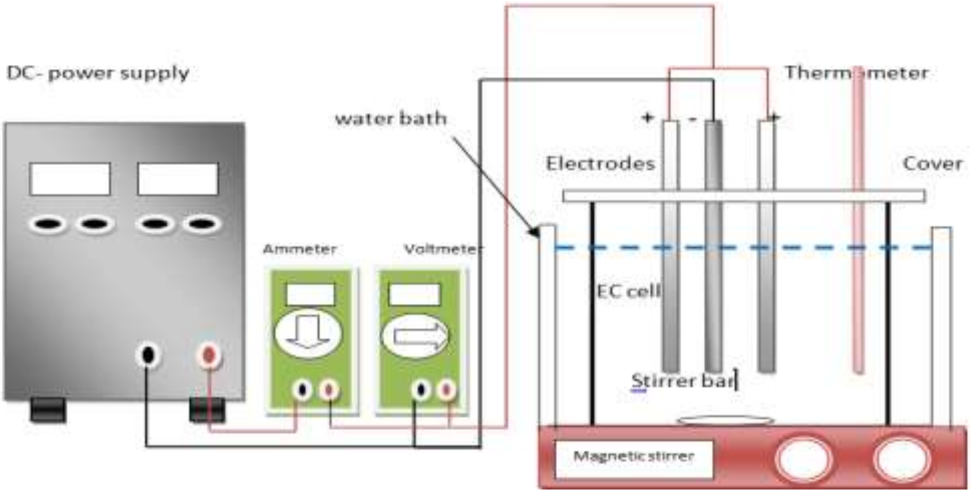


Figure 3. Schematic diagram of Experimental set-up.

3.3 Analytical Methods

The silica concentration was found using an ultraviolet (UV) spectrophotometer (CECDIL, England) at 815nm in Al-Dura Thermal Power Plant, Baghdad, Iraq.

The concentration of calcium and magnesium were determined using the atomic absorption spectroscopy (Type SHIMADZU-7000 F) in the Central Services Laboratory/Ibn Alhaithem College.

The removal efficiency of each measured ions are calculated according to **Eq.6:**

$$\text{Removal efficiency (\%)} = \frac{Ci-Cf}{Ci} \times 100 \qquad (6)$$

Where: **C*i*** and **C*f*** are the initial and final ions concentration (mg/l), respectively.

4. RESULTS AND DISCUSSION

4.1 Effect of current density

The current density is an important parameter for controlling the process of electrocoagulation by influencing the rate of anodic dissolution to produce coagulant (according to Faraday's law) and increase the rate formation of hydroxide flocs of metal so the removal efficiency of silica sand hardness ions increases **(Akyol et al., 2018)**. Current density also affects the rate and size of hydrogen bubble production, hence improves the flotation of H_2 **(Esgair2017; Mollah et al., 2001; Aouniet al., 2009).**

The effect of current density on the removal of scaling ions was investigated using three different values of current density, as shown in **Fig.4**. In this figure, the removal efficiency of silica, calcium and magnesium are increased from 91.54%, 33.622%, and 85.48%, respectively at a current density (0.5 mA/cm^2) to maximum value 99.21%, 80.182%, 98.06% at a current density of (1 mA/cm^2). That means increasing the current density, and also a large effective area of electrodes since the current density is a result of dividing electrical current by the effective area of anodes **(Fayad, 2018)**. So the removal efficiency of scale-forming ions or any pollutant increases due to the increase in the rate of aluminum hydroxide formation.

At a current density of 2 mA/cm^2, the removal efficiency of silica and hardness ions decreased, due to that the aluminum ions were in the form of Al $(OH)_4^-$-flocs remaining dissolved form, thus, dropping the removal efficiency of silica and hardness ions (**Garg** and **Prasad, 2016).**

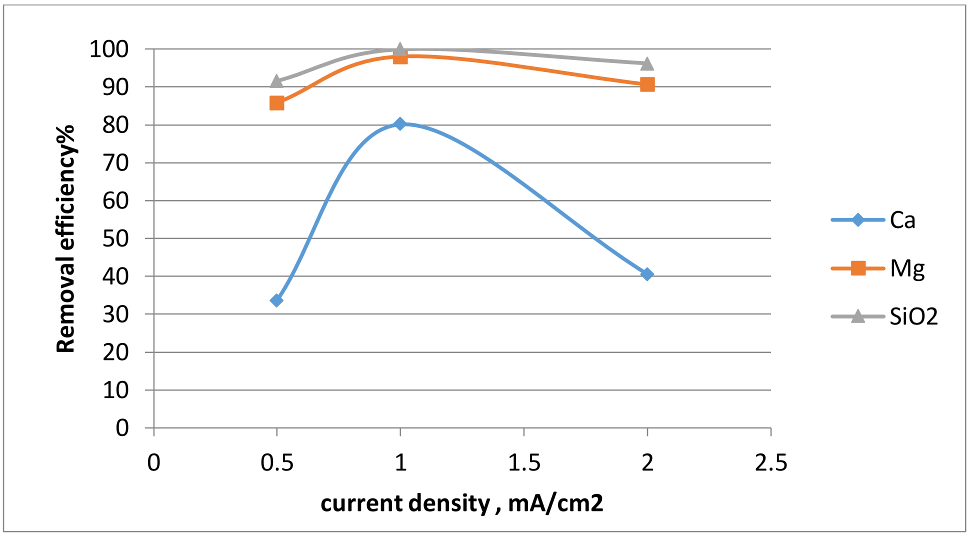


Figure 4. The effect of current density on the removal efficiency (pH=7, electrolysis time = 60 min and 25^{0}C).

4.2. The effect of initial pH

The initial pH value of the wastewater is regarded as one crucial factor in the EC process since it extensively influences the performance of the EC treatment method. The pH affects the dissolution of the anodes electrodes and the type of hydroxides (monomers and polymers) **(Zhi et al., 2013)**, as shown in **Fig.5**. At low initial pH value until to 6, the soluble cation of Al^{+3}, $Al(OH)^{+2}$, and $Al(OH)^{+2}$ was dominant, which were responsible for the formation of electro-coagulant flocks surface. The surface of the flocks became positively charged, which increase electrostatically and could attract the negatively charged ions and positively charged flocks sites. It might not reach the specified levels for the production of amorphous $Al(OH)_3$ with high-collecting capacity then the EC method will not be valuable **(Mohammed and AlJaberi, 2018; Esgair, 2017)**. When pH became high or more than 8.0, monomeric anions of $Al(OH)^{+2}$ will form, which is a weak coagulant **(Kobya et al., 2011).** At neutral pH, the hydrogen bubbles generated at the cathode are small, and that contributes to providing adequate surface area for gas-liquid-solid interfaces, and amalgamation efficiency to improve the aggregation of small destabilization particles and collides. To show the effect of initial pH on removal efficiency of scale-forming ions, three pH values were used in this study (5, 7, and 10) adjusted with 0.1M HCL and 0.1M NaOH solution. The removal efficiency of each ion with initial pH value is specified in **Fig .6;** the best result was attained at an initial pH value of 7.

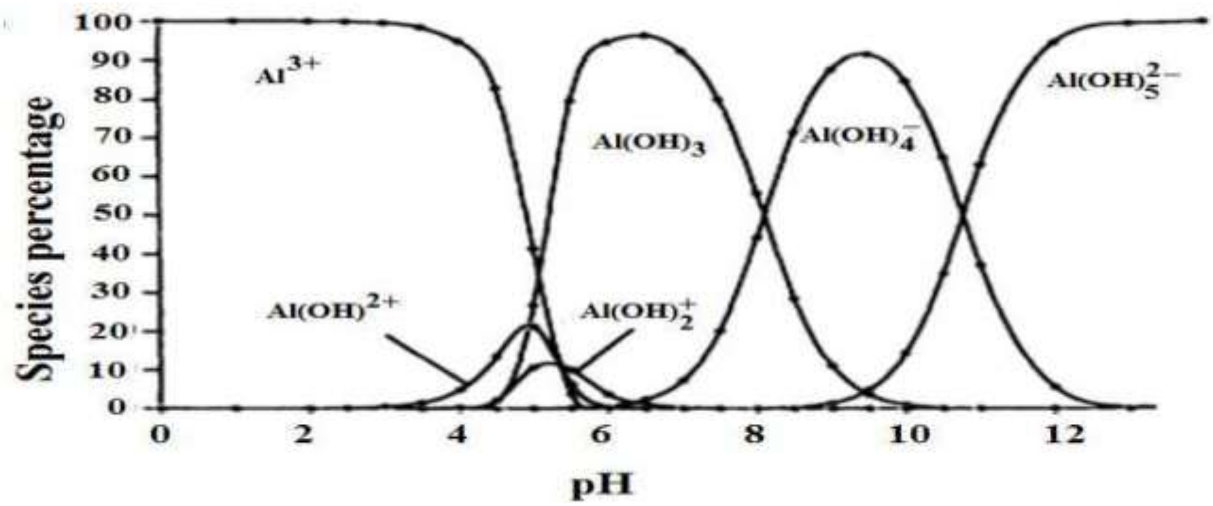


Figure 5. Aluminum hydrolysis species distribution as a function of pH **(Mohammed and AlJaberi, 2018).**

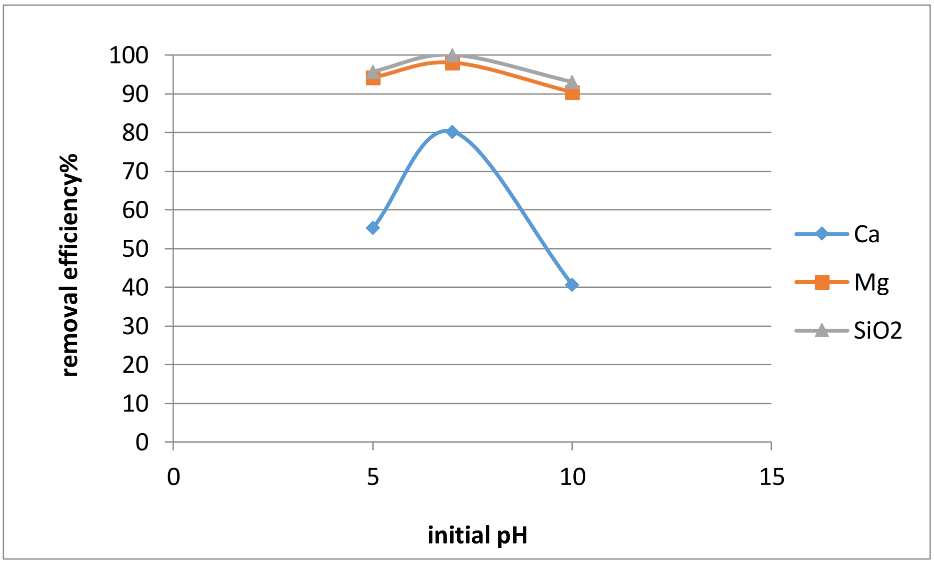


Figure 6. Effect of initial pH on removal ions efficiency (current density=1 mA/cm^2, time =60 min and 25^0.

4.3 The effect of reaction time

The dissolution of anode metal and liberation of coagulation ion species occurs through the electrolysis process. The removal of silica and hardness ions depends on the concentration of coagulant ion produced from the metal anode electrode (**Hafez et al., 2018).** By expanding the treatment time, the quantity of metal ions and their hydroxide flocs raise (**Thakurand** and **Mondal, 2017).** The effect of electrolysis time at initial pH = 7 and current density =0.1 mA/cm^{2}on the silica and hardness ions was illustrated in **Fig .7**. As depicted in this figure, an increase in the treatment time from 12 to 60 min, produces an increase in the removal efficiency of silica from 95.43% to 99.21%, calcium ions from 17.404% to 80.1622% and magnesium ions from 90.667% to 98.06.

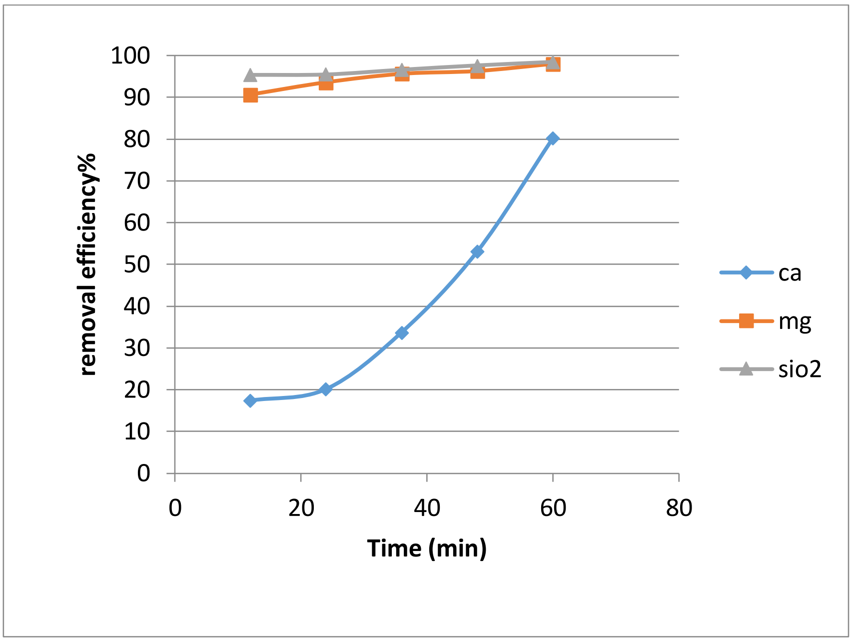
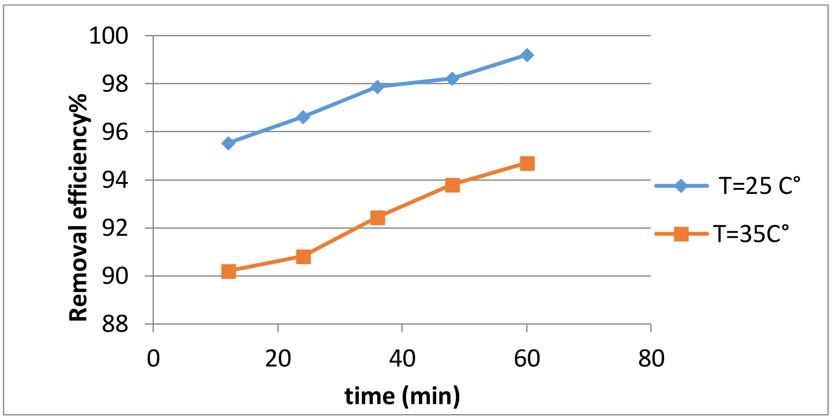


Figure 7. The effect of electrolysis time for removal of scale-forming ions at pH=7, T=25 ^{0}C, and current density=1mA/cm^2.

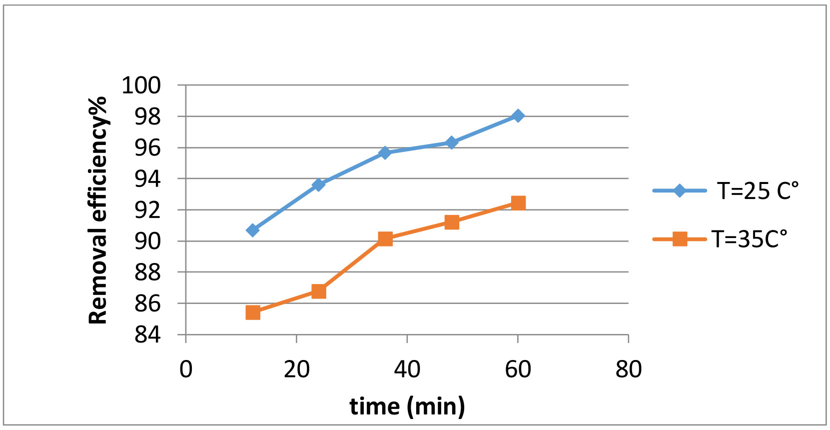
4.4 The effect of temperature

Few articles studied the effect of temperature on the pollutants removal by EC treatment **(Fayad, 2018).** Some of the researches showed that increased temperature leads to increase removal efficiency**;** they concluded that the rate of anode dissolution increases at high temperatures, so the removal efficiency increased **(Yilmaz et al., 2008; Vasudeven et al.,2009).** In contrast, there was a negative effect of increased temperature on the removal efficiency of the paper mill **(Katal and Pahlavanzadeh, 2011**).

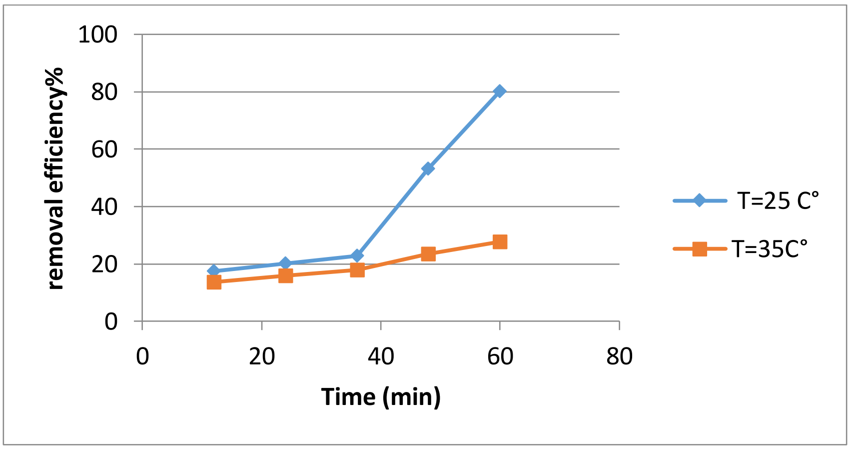
In this study, the effect of temperature on the removal of silica, Mg $^{+2,}$ and Ca^{+2} ions has been investigated at 25 ^{0}C and 35^{0}C, as shown in **Fig. 8(a), (b), and (c)** at pH=7 and current density of 1mA/cm^2, respectively. It was observed that the removal efficiency of each ion at 25^{0}C became higher than that at 35^{0}C due to the decrease in the rate of metal hydroxide formation with the increase of temperature. The temperature also affects the EC process by changing the rate of reactions, the solubility of metal hydroxide, and the conductivity of the solution **(Bharath et al., 2018**).



(a)



(b)



(c)

Figure 8. The removal efficiency of silica (a) magnesium (b) and calcium ions with time at a temperature of 25°C and 35ºC.

5. CONCLUSIONS

Batch EC process using aluminum electrodes experimental study has been evaluated in terms of silica, calcium, and magnesium removal efficiency from simulated cooling tower blowdown water. The conclusions can be summarized in the followings:

- The results show that the EC method is effective in removing hardness ion (Ca^{+2}, Mg^{+2}), and dissolved silica, which is responsible for scale formation on the cooling tower.
- The increase in temperature has a negative effect on calcium, magnesium, and silica ions removal.
- The increase of initial pH above 7 leads to a decrease in the removal efficiency of each ion for all current density values during EC treatment time.
- The high removal efficiency of 80.183%, 99.21%, and 98.06% was obtained for calcium, silica, and magnesium ions, respectively, at a current density of 1 mA/cm^2, treatment time of 60 min, the temperature of 25^0C and pH=7.

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NOMENCLATURE

m= mass of electrode dissolution, grams
I= current, Ampere
t=time, sec
M= molecular weight, g/mole
Z= number of electrons involved in the reaction
F= faraday's constant

C_i= initial ion concentration, mg/l
C_f= final ion concentration, mg/l
T= Temperature, ^{0}C
W= mass of salts, gram
V= volume of solution, liter
C= initial ions concentration, mg/l
Aw= atomic weight of ion,g/mole

Abbreviations

EC= electrocoagulation
CTB= cooling tower blowdown