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Treatment Method with Electrocoagulation in Wastewater that is Dominant with Zinc

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ABSTRACT: *The electrocoagulation method was selected for the removal of* Zn^{+2}*. The effects of the parameters such as current density, pH, and supporting electrolyte concentration on this method were studied. The* Zn^{+2} *concentration, mixing speed, and temperature were 250 mg/L, 150 rpm, and 293 K in the determination of the optimum pH the results obtained showed that a pH of 6 provided the highest* Zn^{+2} *removals. A pH of 6 was taken to be a constant optimum value while studying the effects of current density and supporting electrolyte concentration on removal. Current density values were chosen as 0.25, 0.50, 1.00, and 1.50 mA/cm*^{2}*. Increasing current density increased* Zn^{+2} *removals significantly. Removal of 48.86%, 71.03%, 84.12%, and 97.39% were found for current densities of 0.25, 0.50, 1.00, and 1.50 mA/cm*^{2} *with an initial concentration of 250 mg/L with a reaction time of 30 minutes, respectively. An increase in current density caused an extreme increase in energy consumption. Energy consumption was 1.06 kW-h/m*^{3} *for a current density of 0.25 mA/cm*^{2} *with a reaction time of 30 minutes while it was 1.98, 3.46, and 5.31 kW-h/m*^{3} *for a current density of 0.50, 1.00, and 1.50 mA/cm*^{2} *at a pH of 6, respectively. It was found that the effect of supporting electrolyte concentration on removal efficiency was negative. Aluminum anodes were used in electrocoagulation processes. As supporting electrolyte concentration increased, removal efficiency decreased, and the energy consumption rate increased. It was determined, as the result of the experiments, that* Zn^{+2} *ions can be removed at the rate of 84.12% with a pH of 6, a 250 mg/L* Zn^{+2} *concentration, a 150 rpm mixing speed, a temperature of 293 K and a current density of 1.50 mA/cm*^{2} *in an aqueous solution.*

KEYWORDS: *Electrocoagulation; Zinc; Heavy metal; pH; Current density; Initial concentration.*

INTRODUCTION

Water is the most basic resource necessary for life. A life without water is unthinkable. Water is indispensable for the continuation of its living existence. Clean water is needed for a sustainable life. In addition to the natural water cycle, wastewater treatment is also important in the meeting on clean water. Wastewater; water consisting of industrial and domestic use of water. The recovery of wastewater is necessary for the contamination and

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sustainability of water resources. There are several methods of recovery of wastewater. The treatment process takes place electrochemical [1], physically, chemically [2], and biologically [3]. Wastewater may contain organic and inorganic pollution, heavy metals, suspended solids, oil grease, pathogens, and pesticides [4]. Heavy metals: Chemical elements with a specific weight greater than 4 are called heavy metals. Heavy metals are elements such as zinc, nickel, copper, aluminum, chromium, iron, and manganese, which are very, very little needed in the body, and metals such as lead, mercury, arsenic, and cadmium that the body should never have [5]. When heavy metal is mentioned, toxic effective elements taken with industrial and industrial products that the body does not need are mentioned [6]. Zinc is a fragile metal of bluish-light gray color. It is included in the group of transition elements in the periodic ruler. It has a low boiling temperature. This value is a very decisive factor, especially in the production of pyrometallurgic metals. It is hard and fragile in a spilled state. It can be shaped at 120°C. Zinc comes after iron, aluminum, and copper in terms of annual use in the world [7]. Zinc: for corrosion protection purposes, galvanizing other metals such as steel, brass, nickel silver, different soldering, German silver, etc. in the construction of alloys, usually in casting molds in the automotive industry, in the construction of the bodies of batteries. Zinc oxide is used as a white pigment in watercolors and as an activator in the tire industry [8]. It is contained in some ointments. It is protective against sunburns in summer and cold burns in winter [9]. It is also used in the treatment of eye diseases. Zinc chloride is used in deodorants and as a wood protector. Zinc sulfur is used in scorpions and windmills of watches as a glow-in-the-dark pigment. Zinc methyl ($Zn(CH_3)_2$) is used in the synthesis of many organic substances. Zinc is a component of many daily vitamin and mineral drugs [10].

It is more negative than iron in the electrochemical potential sequence. Thus, zinc finds an important use in cathodic corrosion protection as an anode. Galvanizing is one of these applications. Heavy metal removal in wastewater can be carried out by methods such as ion replacement, chemical precipitation [11], reverse osmosis, membrane filtration, evaporation, and biosorption [12, 13].

Heavy metal removal is also possible by electrocoagulation method [14]. Electrocoagulation is a process of destabilizing substances that are suspended in water, liquid-liquid heterogeneous mixture, or dissolved environment by giving electric current to the environment [15]. The electrocoagulation process is a combination of oxidation, flocculation, and flotation processes. In the electrocoagulation process, anode and cathode assembly are created inside a reactor. The system is supplied with the required electric current from the power supply [16]. Wastewater is discharged into the reactor. To eliminate the pollution in the wastewater, positive and negative loads are separated by the current installed in the system, which clings to the anode and cathode surface area. The purpose of electrocoagulation is to provide treatment process by forming metal hydroxide flocs in wastewater with the solubility of the anode after the electrolysis process [17, 18].

Electrocoagulation

The most preferred electrochemical method is electrocoagulation. One of the most important conditions in an electrochemical process is the type of electrode. Aluminum (Al^{+3}) and iron (Fe^{+3}, Fe^{+2}) electrodes are widely used in electrocoagulation. These electrodes react with water during the operation phase of the process, forming metal hydroxides such as $Al(OH)^3$, $Fe(OH)^2$ and $Fe(OH)^3$ [19]. Treatment begins with the formation of metal hydroxides. Metal hydroxides with very high adsorption capacity are removed from the water by adsorbing different pollutant parameters in the water and creating precipitation. This refinement method is available in many places today. During this process, very small gas bubbles begin to emerge from the water because of electrochemical processes formed in the electrodes. Considering that these gas bubbles form the basis of electroflotation, it can be said that some pollutants will be removed from the water by the electroflotation method during electrocoagulation. Since a certain degree of electroflotation also occurs, the yield of pollutant removal is high. This efficiency can be further increased by optimizing various operating conditions. There are usually 3 basic processes in electrocoagulation [20, 21].

a) Electrolytic reactions on the surface of the electrodes

b) Formation of coagulations (metallic ions) in the liquid phase

c) Removal of soluble or colloidal contaminants by adsorption, coagulant, sedimentation, or flotation mechanisms.

With electrocoagulation, liquid-liquid heterogeneous substances, suspended solids, and colloidal substances are stabilized. Therefore, in electrical applications, when the particles are properly contacted with electrodes, the particles are neutralized and the different particles form large flocs in combination [22, 23].

Aluminum or iron electrodes are usually used in the electrocoagulation process. This is because metallic ions are a good coagulant because of their high adsorption capacity. Dispersed particles can be purified from water by electrocoagulation process and stable sludge can be obtained. In one study, it was concluded that electrocoagulation can be shown as an alternative to chemical treatment, since flock formation is achieved in an accelerated way by electrocoagulation [24, 25].

Although the electrocoagulation method is like traditional coagulation, electrocoagulation has many advantages.

a) Electrocoagulation is more effective in stabilizing and removing small colloidal particles than conventional coagulation.

b) Less and more stable sludge is formed in electrocoagulation.

c) Electrocoagulation equipment is easy to use and operate.

d) There is no need to add chemicals in electrocoagulation. That's why it's easier to operate and maintain.

Electrocoagulation is the most common method of use in electrochemical treatment methods due to both effective efficiency and easy operating conditions [26, 27].

EXPERIMENTAL SECTION

In this study with synthetic wastewater, stock solutions containing 1000 mg/L Zn^{+2} were prepared, and experimental studies were carried out by making appropriate dilutions. Electrocoagulation process is preferred from electrochemical methods. Electrocoagulation experiments were carried out in a reactive electrolytic cell. The electrocoagulation cell is made of plexiglass with dimensions of 15 x 8 x 10 cm. The total volume of wastewater for each experiment was about 1000 mL. The dimensions of aluminum (Al) plates used for victim electrodes are 7.5 cm high, 7.5 cm wide and 0.3 cm thick and arranged in monopolar configurations. During each experiment, ten plates were made in the electrochemical reactor, and the distance between the plates was fixed at about 5 mm. The current was held steady through a precise DC power supply (GW GPC-3060D), characterized by ranges of 0–6 A for current and 0–30 V for voltage. The wastewater samples used in the experiments were prepared using Merck brand 99.99% purity $ZnCl_2.H_2O$ chemical with a molecular weight of 154.3 g/mol. 2.36 g of $ZnCl_2.H_2O$ is dried at 105°C, some of it is dissolved in distilled water and made up to 1 L to obtain 1000 mg/L stock solution. Between experiments, the electrodes were sanded and washed with dilute HCl. Samples of 3 mL were periodically taken from the electrocoagulation cell to track the progression of treatment, then filtered to eliminate sludge formed during electrolysis (0.45 μm). Zinc analysis was performed using atomic absorption spectrometer (SHIMADZU AA6800) [28]. The studied experimental parameters and experimental setup are given in Table 1 and Fig.1.

Samples were taken against time and the removal efficiencies and energy consumptions from these samples were calculated with the help of the following equations.

Removal efficiency:

$$\eta\ (\%) = \left(\frac{C_0 - C_t}{C_0}\right) \times 100 \qquad (1)$$

Where C_0: Initial metal concentration (mg/L), C_t: Metal concentration at any time t (mg/L),

Electric energy consumption is a very important economic parameter in the electrocoagulation process. Electrical energy consumption was calculated using the following equation [28, 29]:

$$\mathrm{SEC} = \frac{l \times V \times t}{v} \qquad (2)$$

Here, you can see that

SEC= is specific energy consumption (kW-h/m^3), I =current intensity (A), V= is the potential difference applied (V), t =reaction time (h) and v =solution volume (m^3)

RESULTS AND DISCUSSION

Effect of Mixing Speed on Zinc Removal and Energy Consumption

Mixing speed can be considered as an important parameter in terms of flock formation and structure of flocks. The experimental conditions of this study were determined as J=1 mA/cm^2, C_0=250 mg/L, pH=5.0. Mixing speeds of 50 rpm, 150 rpm, 250 rpm and 400 rpm were selected for this experiment. As Fig. 2 shows,

Table 1: Experimental Parameters Studied.

Examined Parameter	Parameter Range	Fixed Variables
Mixing Speed (rpm)	50; 150; 250, 400	pHi≈5 (natural), J=1.00 mA/cm^2, T=20±1°C
Support Electrolyte Type	NaCl; $NaNO_3$; Na_2SO_4	pHi≈5 (natural), J=1.00 mA/cm^2, T=20±1°C, mixing speed =150 rpm
Support Electrolyte Concentration (NaCl, mM)	0, 10; 15; 20;	pHi≈5 (natural), J=1.00 mA/cm^2, T=20±1°C, mixing speed =150 rpm
(pHi) Wastewater İnitial pH	3; 4; 5 (natural); 6	J=1.00 mA/cm^2, T=20±1°C, mixing speed = 150 rpm
Current Density (mA/cm^2)	0.25; 0.50; 1.00; 1.50	pHi≈5 (natural), mixing speed =150 rpm, T=20±1°C

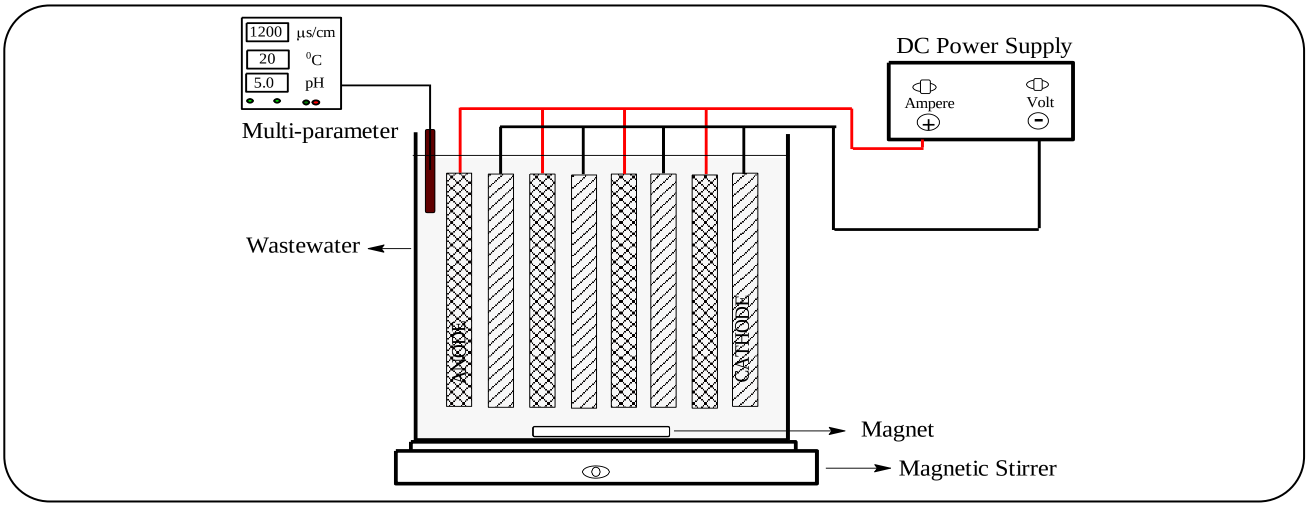

Fig. 1: Experimental Setup.

the mixing speed is ideally 150 rpm. This is because the resulting flocs do not dissipation at this speed. As the mixing speed decreases, the formation of flocks becomes difficult and is not preferred. When the mixing speeds are increased, the resulting flocs are dispersed, which is undesirable. In the studies where the effect of mixing speed on zinc removal yield was examined, the removal yields were calculated as 78.27%, 84.12%, 71.21% and 65.11% for 50 rpm, 150 rpm, 250 rpm and 400 rpm, respectively.[29].

As Fig. 3 shows, energy consumption is minimal when the mixing speed is in the desired state. When the mixing speed is increased over time, the amount of energy consumed increases. In this case, the amount of energy consumed increases depending on the state of formation of the flocs. While the mixing speed was 50 rpm, energy consumption was 4.17 kW-h/m^3, 3.46 kW-h/m^3 at 150 rpm, 5.29 kW-h/m^3 at 250 rpm and 6.33 kW-h /m^3 at 400 rpm [30].

Effect of Support Electrode Type on Zinc Removal and Energy Consumption

The choice of support electrode type is an important parameter in zinc removal. Zinc forms a precipitable compound while being removed from wastewater as heavy metal. NaCl, Na_2SO_4, and $NaNO_3$ were chosen as supporting electrolyte types. The experimental conditions of this study were determined as J=1 mA/cm^2, C_0=250 mg/L, pH=5.0 and Mixing Speed=150 rpm. As interpreted from Fig. 4, NaCl was determined as the most ideal supporting electrolyte type. The conductivity values change according to the solubility degrees of the supporting electrolyte type used. It was observed that the best soluble in water was NaCl, then $NaNO_3$ and Na_2SO_4.

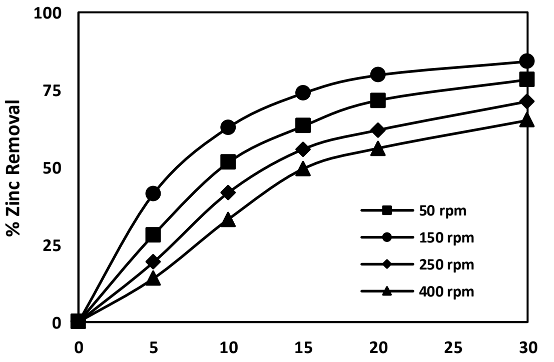

Fig. 2: Effect of Mixing Speed on Zinc Removal (J=1 mA/cm^2; C_0=250 mg/L; pH:5.0, without support electrolyte).

It was calculated that the zinc removal efficiency without supporting electrolyte was 84.12%, 86.93% when NaCl was added, 85.99% when $NaNO_3$ was added, and 85.26% when Na_2SO_4 was added [31].

NaCl, Na_2SO_4 and $NaNO_3$ were chosen as supporting electrolyte types. As can be seen from Fig. 5, the energy consumption decreases when the supporting electrolyte is added. According to the type of supporting electrolyte added, the energy consumed is the least, which dissolves best in water and increases conductivity. In this case, the energy consumption without supporting electrolyte was 3.46 kW-h/m^3, with NaCl addition 1.17 kW-h/m^3, with $NaNO_3$ addition 1.63 kW-h/m^3 and with Na_2SO_4 addition 2.54 kW-h/m^3 [32].

Effect of Support Electrolyte Concentration on Zinc Removal and Energy Consumption

Depending on the added NaCl concentration in zinc removal in wastewater, the removal decreases as the concentration decreases. The experimental conditions of this study were determined as J=1 mA/cm^2, C_0=250 mg/L, Support Electrolyte Type= NaCl, pH=5.0 and Mixing Speed=150 rpm. As the support electrolyte concentration, 20 mM NaCl, 15 mM NaCl, 10 mM NaCl, and without the support electrolyte were investigated. Starting from Fig. 6, since the conductivity does not increase when the supporting electrolyte is not in the environment, the removal takes place here at least. It was observed that the highest concentration of removal was when 20 mM NaCl salt was added. Thus, the removal efficiency without supporting electrolyte was calculated as 84.12%, when 10 mM NaCl was added, 85.92%, 15 mM was added as 86.93%, and 20 mM was added as 87.97% [33].

As observed in Fig. 7, it is planned to increase the conductivity by adding the support electrolyte concentration. When the conductivity is increased, the amount of energy consumed decreases. The amount of is determined as energy consumed without supporting electrolyte is 3.46 kW-h/m^3, when 10 mM NaCl is added 2.33 kW-h/m^3, when 15 mM NaCl is added 1.17 kW-h/m^3 when 20 mM NaCl is added 1.08 kW- h/m^3 [34].

Effect of pH on Zinc Removal and Energy Consumption

By decreasing the pH value, the environment becomes acidic. J=1 mA/cm^2, C_0=250 mg/L and Mixing Speed = 150 rpm. In this study, pH=3, pH=4, pH=5 and pH=6 was investigated.

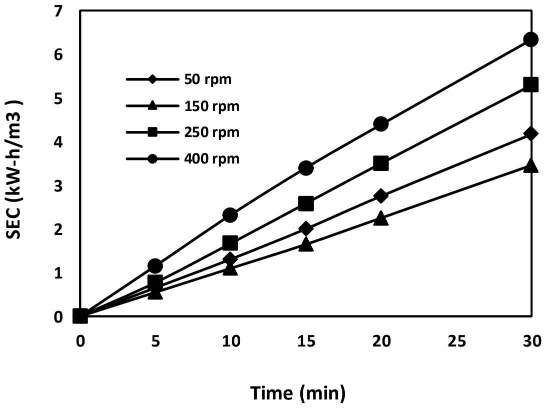

Fig. 3: Effect of Mixing Speed on Energy Consumption (J=1 mA/cm^2; C_0=250 mg/L; pH:5.0, without support electrolyte).

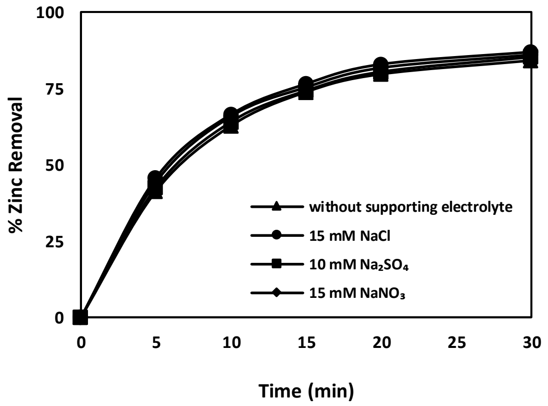

Fig. 4: Effect of Support Electrode Type on Zinc Removal (J=1 mA/cm^2; C_0=250 mg/L; pH:5.0; MS=150 rpm).

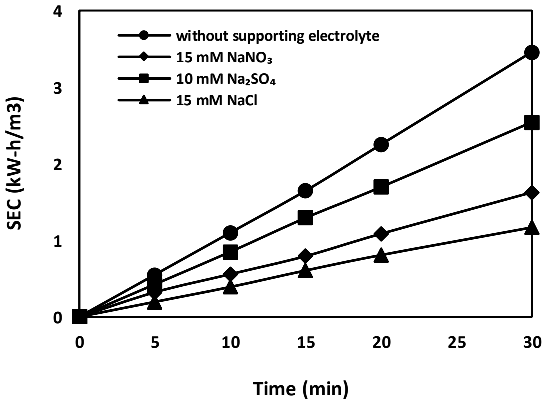

Fig. 5: Effect of Support Electrolyte Type on Energy Consumption (J=1 mA/cm^2; C_0=250 mg/L; pH:5.0; MS=150 rpm).

As indicated in Fig. 8, the maximum removal was observed at pH 6 of ideal zinc removal. The least removal occurred at pH 3. Respectively, zinc removal efficiencies were found to be 87.89% at pH=6, 84.12% at pH=5, 79.44% at pH=4, and 75.54% at pH=3 [35]. At high pH values, heavy metals precipitate as hydroxide. Therefore, pH values higher than 6 have not been studied to investigate heavy metal removal without precipitation [36].

As can be read from Fig. 9, energy consumption was determined as the highest pH = 5 and the lowest pH=3. Energy consumptions respectively 2.92 kW-h/m^3 at pH=6, 3.46 kW-h/m^3 at pH=5, 2.29 kW-h/m^3 at pH=4 and pH=3 and it was calculated as 1.79 kW-h/m^3 [37].

Effect of initial concentration on Zinc removal and energy consumption

The experimental conditions of this study were chosen as J=1 mA/cm^2, pH=5, and Mixing Speed=150 rpm. In this analysis, the initial. 10, the maximum removal was observed when the initial concentration was at 25 mg/L, while the minimum removal was observed at 1000 mg/L. However, although it was observed that the calculated yield value decreased with increasing concentration, the amount of zinc removed per unit of wastewater volume increased significantly. Namely, while 24.73 mg/L zinc was removed with 98.91% removal efficiency at 25 mg/L initial concentration, 433 mg/L zinc removal was achieved at 1000 mg/L initial concentration, 43.30% removal was achieved. Zinc removal efficiencies were calculated as 98.91% in 25 mg/L, 96.44% in 50 mg/L, 91.67% in 100 mg/L, 84.12% in 250 mg/L, 69.48% in 500 mg/L and 43.30% in 1000 mg/L, respectively [38].

As interpreted from Fig. 11, the highest energy consumption was observed as 25 mg/L and at least 1000 mg/L. The energy consumption amounts are 6.33 kW-h/m^3 in 25 mg/L, 5.50 kW-h/m^3 in 50 mg/L, 4.58 kW-h/m^3

Effect of current density on Zinc removal and energy consumption

Current density is also an important parameter for zinc removal. As the current supplied to the system increases, the current density increases, and the amount of removal increases. The experimental conditions of this study were chosen as pH=5, C_0=250 mg/L, and Mixing Speed=150 rpm. In this study, current densities were investigated by choosing 0.25 mA/cm^2, 0.50 mA/cm^2, 1 mA/cm^{2,} and 1.5 mA/cm^2.

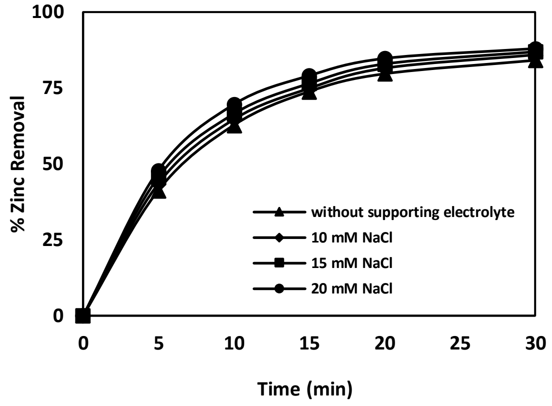

Fig. 6: Effect of Support Electrolyte Concentration on Zinc Removal (J=1 mA/cm^2; C_0=250 mg/L; pH:5.0; MS=150 rpm; SE=NaCl).

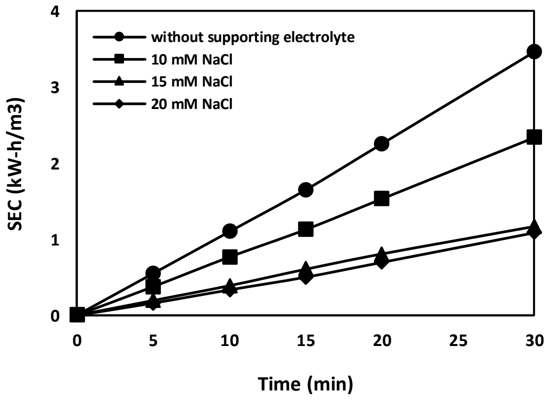

Fig. 7: Effect of Support Electrolyte Concentration on Energy Consumption (J=1 mA/cm^2; C_0=250 mg/L; pH:5.0; MS=150 rpm; SE=NaCl).

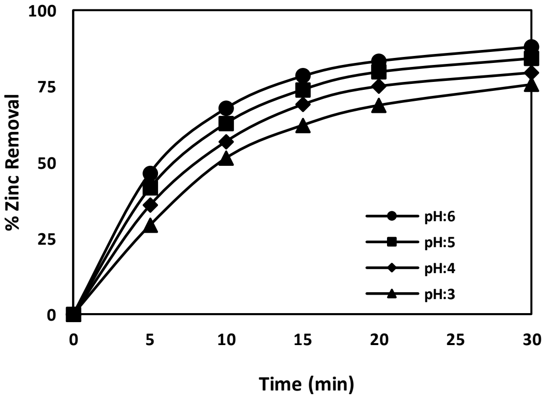

Fig. 8: Effect of pH on Zinc Removal (J=1 mA/cm^2; C_0=250 mg/L; MS=150rpm without support electrolyte).

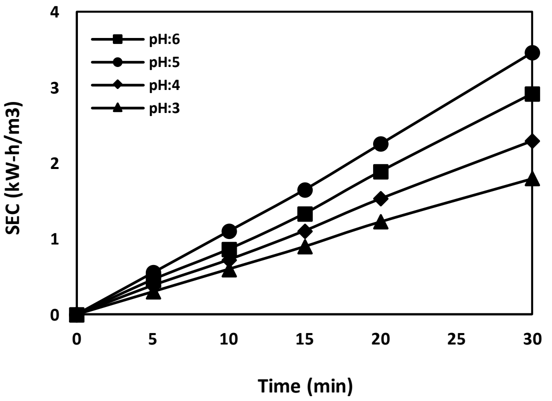

Fig. 9: Effect of pH on Energy Consumption (J=1 mA/cm^2; C_0=250 mg/L; MS=150 rpm without support electrolyte).

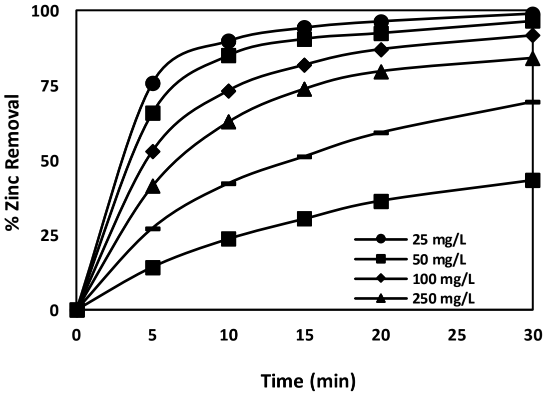

Fig. 10: Effect of Initial Concentration on Zinc Removal (J=1 mA/cm^2; pH:5.0; MS=150 rpm, without support electrolyte).

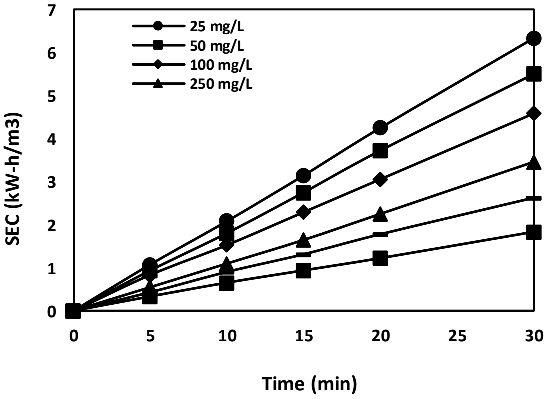

Fig. 11: Effect of Initial Concentration on Energy Consumption (J=1 mA/cm^2; pH:5.0; MS=150 rpm without support electrolyte).

n 100 mg/L, and 3.46 in 250 mg/L, respectively. kW-h/m^3 is calculated as 2.63 kW-h/m^3 in 500 mg/L and 1.83 kW-h/m^3 in 1000 mg/L [39].

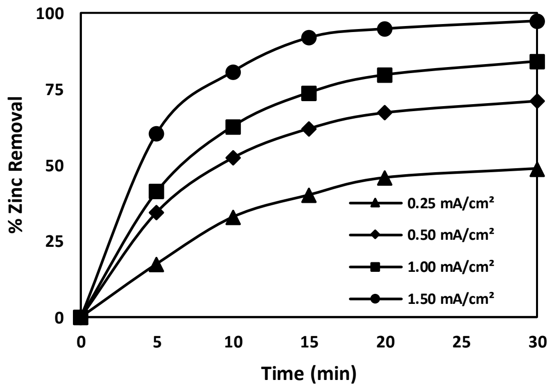

Fig. 12: Effect of Current Density on Zinc Removal (C_0=250 mg/L; pH:5.0, MS=150rpm, without support electrolyte).

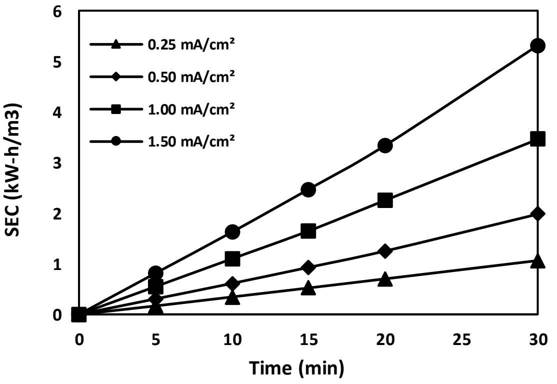

Fig. 13: Effect of Current Density on Energy Consumption (C_0=250 mg/L; pH:5.0, MS=150rpm, without support electrolyte).

As indicated in Fig. 12, as the current passing through the electrodes increases, the amount of ions retained in the electrodes increases. As can be seen from the graph, the maximum zinc removal was 1.5 mA/cm^2, and the minimum removal was 0.25 mA/cm^2. Zinc removal efficiencies were 97.39% at 1.5 mA/cm^2, 84.12% at 1 mA/cm^2, 71.03% at 0.50 mA/cm^{2,} and % at 0.25 mA/cm^2, respectively. It was determined as 48.86 [40].

As indicated in Fig. 13, it was observed that the energy consumption amount was at most 1.5 mA/cm^2 and at least 0.25 mA/cm^2. The greater the amount of current passing through the electrodes, the more electricity is consumed. The amount of energy consumed at 1.5 mA/cm^2 5.31 kW-h/m^3, the energy consumed at 1 mA/cm^2 3.46 kW-h/m^3, the energy consumed at 0.50 mA/cm^2 1.98 The energy consumed at kW-h/m^3, 0.25 mA/cm^2 is calculated as 1.06 kW-h/m^3 [41].

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