Modeling and Simulation of Wastewater Electrocoagulation Reactor

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Huda Saeed. Al-Barakat[1], hudawww1372@gmail.com
Falah Kaify. Matloub[2], fa1117@yahoo.com
Sata Kathum. Ajjam[3] Satajam58@yahoo.com
& Tahseen Ali. Al-Hattab[4] alhattab.t@gmail.com

Chemical Eng. Department, College of Engineering, University of Babylon, Iraq

Abstract. Electro-Coagulation (EC) method is the most effective electrochemical treatment of the wastewater that contains heavy metals as pollutants such as (lead compounds). It is consumed less energy compared with the other conventional methods. In this work, the Computational Fluid Dynamics (CFD) modeling approach for the simulation of the EC unit was studied. Transport of species, current-voltage distribution, and fluid flow were simulated using the diffusion, Nernst-Planck, and Navier-Stokes equations, respectively. The commercial software (COMSOL 5.4) was used to solve the system of equations that govern this problem. The influence of different parameters such as Hydraulic Retention Time (HRT) (15-45 min), applied voltage (2-10v) rotating speed of electrode (0-150 rpm) on the EC reactor also has been studied. The extracted results from the simulation show how the voltage, current, velocity and concentration of the pollutant and coagulant are distributed in the reactor. Besides, the transient behavior of the lead removal from the water was demonstrated in this work. The obtained results from this study were compared with experimental results. The theoretical results showed a good prediction of lead removal from water by electrocoagulation system and they showed good agreement between predictive and experimental data.

Keywords: COMSOL, Electrocoagulation, Lead Removal, Mathematical mode.

1. Introduction

Electrocoagulation is a clean electrochemical process, that uses an applied voltage to remove metals from wastewater. The contaminants present in solution such as lead ions are held in solution by electrical charges. Neutralization these ions and other charged particles with opposite electrical charges of ions provided by EC system, causes to be destabilized and then precipitate in a stable form [1]. The EC process operates on producing cations (Fe and/or Al anodes) which is causes to increasing the coagulation of the pollutants from an aqueous medium. Dissolution of Al or Fe electrodes produce ions that are hydrolyzed immediately into aluminum hydroxide or polymeric ions [2].

Producing polymeric hydroxides close to the anode is the main function of sacrificial anode. These polymeric hydroxides are act as an excellent coagulating agent. The negative ions that are created at the cathode transferred to the anode because of electrophoretic action. These negative ions react with metal cations and convert to the coagulation. Water is electrolyzed due to continuous electrolysis. Small bubbles of O_2 are produced at the anode and at the cathode, small bubbles of H_2 are generated which are responsible

of electrolysis of water. These bubbles attract the flocculated particles and because of natural buoyancy, float towards the surface [2].

Therefore, the distribution of the ionic metal species in EC reactor, is the determining factor for its performance. Moreover, after the flocculation, the contaminants can be take out by flotation, sedimentation or filtration. The mathematical models that describe the physical system, help to better understand the phenomena involved in the EC process. However, because of the complexity of the process, the mathematical models cannot to describe all the mechanisms involved in the reactor. Therefore, the use of lumped-parameter models, where the process variables are independent of the position, depending only on the time could be an alternative [3].

The following work simulate and modeling the experimental apparatus of EC reactor in Comsol's software to predict the amount of lead removal with time. Also to simulate the distribution of voltage, current , velocity and concentration in EC reactor. And studding the effect of various parameter such as the effect of applied voltage, HRT and rotating anode speed on the process. This work contains two results, experimental and theoretical. The experimental results that studied in [4] and the theoretical results are obtained in this work. The aim of this study is to compare between experimental and theoretical results.

2.Materials and the method that used in the experimental work

Many chemical materials were used in the experimental work. This chemical materials taken from University Of Babylon /College of Engineering/Chemical Engineering Department. The chemical materials, formula and purity % are given in Table 1.[4]

Table 1. List of chemical used

NO.	Chemical	Formula
1	**Lead nitrate**	$Pb(NO_3)_2$:: purity equal to 99.5%wt, produced by: HiMedia Laboratories Pvt. Ltd., India).
2	**Nitric acid**	HNO_3: : purity equal to 72%wt, produced by: HiMedia Laboratories Pvt. Ltd., India).
3	**Sulfuric acid**	H_2SO_4:: purity equal to 98%wt, produced by: HiMedia Laboratories Pvt. Ltd., India).
4	**Sodium chloride**	$NaCl$: purity equal to 99.5%wt, produced by: HiMedia Laboratories Pvt. Ltd., India).
5	**Sodium hydroxide**	$NaOH$:: purity equal to 97.5 % wt, product of loba chemie , India)
6	**Hydrochloric acid**	HCl: purity equal to 38%wt, produced by: HiMedia Laboratories Pvt. Ltd., India).
7	**Ethanol**	CH_3CH_2OH : purity equal to 99.9 %wt, produced by: HiMedia Laboratories Pvt. Ltd., India.

2.1. Preparation of Aqueous polluted water

A stock solution of 1000 ppm lead was prepared by dissolving lead nitrate $Pb(NO_3)_2$ in distilled water. Lead stock solution of 1000 ppm was prepared by dissolving 1.598 g of lead nitrate. Two or three drops of nitric acid must be added to the Lead nitrate to dissolve the lead molecules .After mixing, one letter of the distilled water added to the solution for the complete mixing of lead nitrate. The required concentration is 200mg-lead/l that prepared from the stock solution by dilution equation[4]:

$$V_1 * C_1 = V_2 * C_2 \quad (1)$$

And then added 0.5g/l of NaCl for sample.

2.2. Experimental setup

A continuous flow EC reactor was made from Perspex glass of 1000 ml in volume. The reactor has three holes ,one of them located in the bottom of the glass to input the polluted water and two in the top to output the effluent . The electrodes used in the continuous EC process were stainless steel as the cathode and aluminum as the anode. The anode that used in the continuous EC process rotated in the reactor. Theanode is the cylinder with (2.3 cm) external diameter and 4.5cm long which have four grooves at its surface with thick (2mm), the effective area of the Aluminum electrode was 43.87cm².

All tests were carried out at a pH of 7±0.1 and room temperature (25±4 °C) and(200 ppm) concentration of lead. Before each experiment, the conductivity was detected by addition 0.5 g/l NaCl while the pH of the electrolyte was detected with addition drops of H_2SO_4 or NaOH solution [4].The relationship between the flow rate and the hydraulic retention time (HRT) during the operation is given by the following equation:

$$\text{HRT} = \frac{V_R}{Q} \quad (2)$$

(V_R)is the reactor volume (m^3) and(Q) is the flow rate (m^3/s).

In this experiment, flow rate was used at range from (3.2 l/h) to (1.06 l/h) corresponding to HRT of 15 to 45 min, respectively. The associated schematic diagram of the equipment is shown in figure1.

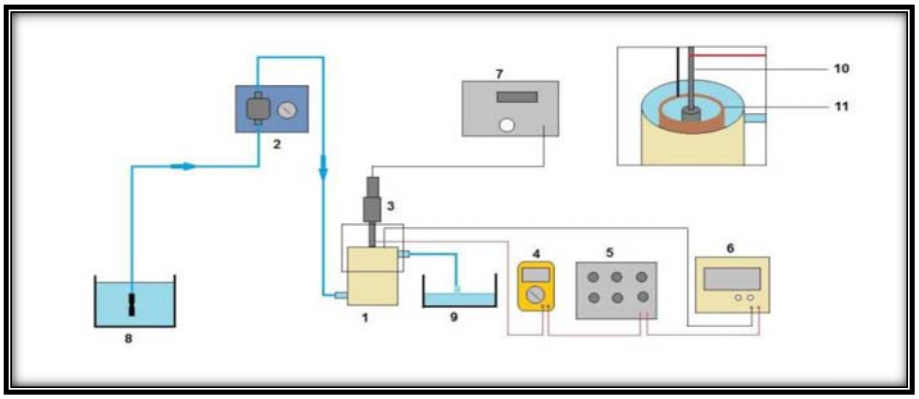


Figure1. Schematic Diagram center of Electrocoagulation (EC) Rotating Anode System 1.electrochemical cell 2.dosing pump 3.motor 4.digital multi ammeter 5.multi range resistor 6. Dc power supply 7.speed controller 8. Inlet tank 9.effluent tank 10. Anode 11. Cathode.

Applied voltage was varied using a transformer which converts AC to DC and the current was measured with multi-meters during the experimental run. Each run was timed starting with the rotating of the electrode (anode) to make mixing of the solution and DC power supply switching on. Also using the anode electrode as mixing is very important factor that affected the release of coagulants in the solution, mass transfer, bubble type and the flotation process. The rotating electrode (anode) was placed in the middle of the fixed cylinder(cathode). The cathode and anode electrodes were placed together in middle of the reactor . During the continuous EC experiments, waste water was pumped at a known flow rate to electrochemical coagulation tank, anodic dissolution occurred and hydrogen gas was produced at the cathode. After each hydraulic retention time according to intervals(15,25,35 and 45) min by using stop watch ,Effluent samples of 40 ml were taken at different times during the experiment to be filtered by filter paper (small pores ,small fast filtering) to get clear solution and then analyzed by atomic absorption spectrometry. After each run, the electrodes were washed and brushed then cleaned by ethanol or by hydrochloric acid to remove any solids accumulated on the electrode surface. The rotating electrode(anode) operated at various rotational velocity (0,50,100&150)rpm and it was used as mixer to maintain uniform distribution.[4]

The theoretical part

3. Geometry of the EC reactor

The geometry of the EC reactor consists of a cylinder (beaker in laboratory) with dimensions (r=5.7cm, h=11.7) . There are two holes at the surface of the cylinder; which are for entering the wastewater at one side and exiting from the other side (continuous system). The entering hole is at high 1.5cm and the existing hole is at high 8.6cm of the cylinder.

This cylinder contains the cathode and anode. The cathode is a hollow cylinder which made from stainless steel with dimensions (r=4cm, h=6.8cm) and has holes on its surface.

The anode is a cylinder which made from aluminum with dimensions (r=1.15cm, h=4.5cm) and has four grooves with 2mm thick. It connected to a shaft for rotating the anode. Figure 1 shows the structure of continuous EC system in model.

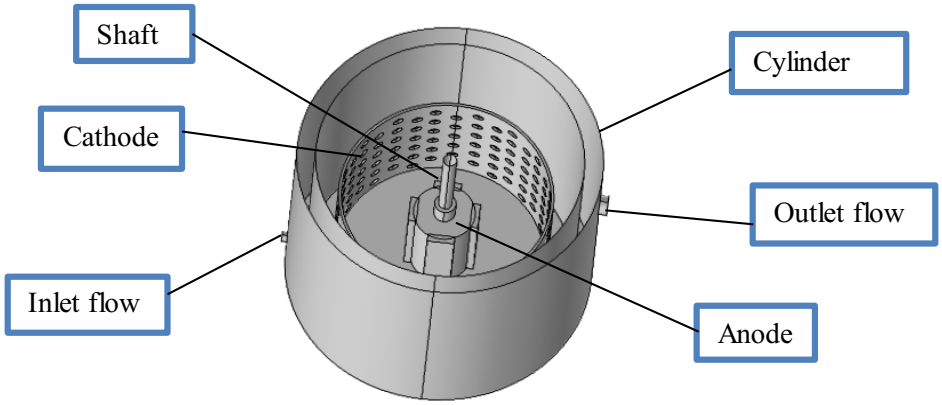


Figure2. Illustration of continuous EC reactor model

4. The reactions involved in EC reactor

Al and Fe electrodes are commonly used for the EC process. The type of coagulant is determined by electrode materials. The nature of the coagulant affects the coagulation and the efficiency processes. At the cathode the evolution of hydrogen occurs and this causes to an increment in the pH of the solution at the cathode [5].

$$2H^+ + 2e^- \rightarrow H_2 \quad \text{acid} \tag{3}$$

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad \text{neutral/alkaline} \tag{4}$$

For Al anodes, the chemical reaction occur at the electrode, and in the bulk the $Al(OH)_3$ is generated, which are given as follows [5]

$$Al \rightarrow Al^{3+} + 3e^- \tag{5}$$

$$Al^{3+} + 3OH^- \rightarrow Al(OH)_3 \tag{6}$$

At the cathode the electro-deposition of lead can occur as a secondary reaction [6];

$$Pb^{2+} + 2e^- \rightarrow Pb_{(s)} \tag{7}$$

At the cathode the OH^- produced, and it react with Al^{3+} and Pb^{2+} ions. The amorphous $Al(OH)_{3(s)}$ that formed have large surface areas, that are useful for the trapping and adsorption of Pb^{2+} ions. The following reaction explained The adsorption of Pb^{2+} ions on $Al(OH)_{3(s)}$.

$$Al(OH)_3 + Pb^{2+} \rightarrow Al(OH)O_2Pb + 2H^+ \tag{8}$$

when specific electrical current flow in the electrolytic cell, the mass of Al or Fe is dissolved from the anode, and it quantified by faradays law [7].

$$W = \left[\frac{ItM}{ZF}\right] \tag{9}$$

Where w is the amount of anode material dissolved (g), I is the current (A), t is the electrolysis time, M is the specific molecular weight of electrode (g/mol), Z is the number of electrons involved in the reaction, F is the faradays constant (96485 C/mol)

5. Governing equations

Simulation of the phenomena in an EC process includes the numerical solution of motion equations (mass, momentum), current distribution, electric potential and energy in the flow geometry, with other sets of equations related to the problem. These equation are used usually to explain the transport mechanics like reaction of chemical species, migration and diffusion in the presence of an electric field [8].

5.1. Momentum transfer

The continuity equation and incompressible Navier-stokes (NS) equations is used to describe the flow behavior in EC reactor [9].

- Continuity equation

$$\frac{\partial u}{\partial x} + \frac{\partial u}{\partial y} + \frac{\partial u}{\partial z} = 0 \tag{10}$$

- General momentum equation

$$\rho\frac{\partial u}{\partial t} + \rho(u \cdot \nabla)u = \nabla \cdot [-pI + \mu(\nabla u + (\nabla u)^T)] + F \tag{11}$$

Where p is the pressure (pa), ρ is density (kg/m3), F is the volume force vector (N/m3), μ is dynamic viscosity (pa.s), u is the mean velocity (m/s) and I is the current (A).

5.1.1. Boundary condition

In EC reactor the fluid flow in the reactor with initial velocity ($u_0 = u_{in}$)

At the anode the velocity is defined rotational frequency (angular velocity)

$$u = u_w = \left(\frac{\partial x}{\partial t}\right)\Big|_x \quad \& \quad dx = dx(r_{bp}, \omega, t) \quad \& \quad \frac{d\omega}{dt} = w \tag{12}$$

At the cathode and wall surface (cylinder) the velocity equals zero (u=0)

5.2. Mass transfer of ionic species

There are three types of mass transport flux in EC system that are convection ($C_i u$), diffusion ($D_i \nabla C_i$) and electro-migration ($Z_i u_{m,i} F C_i \nabla\varphi$). The mass transfer flux of species i is given by Nernst-plank equation[9].

$$N_i = -D_i \nabla C_i - Z_i u_{m,i} F C_i \nabla \varphi + C_i u \quad (13)$$

Where N_i is the flux of the chemical species due to convection, D_i is the diffusion coefficient, C_i is the average concentration, Z_i is the charge number, $u_{m,i}$is mobility of species i, u velocity, F faraday constant and φ is electric potential.

The conservation of charge and species is given by:

$$\frac{\partial C_i}{\partial t} = -\nabla N_i + R_i \quad (14)$$

Where R_i is the reaction rate of species i.

5.2.1. Boundary condition

The initial mass transfer flux at the anode and the cathode equals zero. At anode the amount of aluminum liberated is compute by faraday's law:

At anode $$N_i = \frac{i}{ZF} \quad (15)$$

Where i is the electrolyte current density magnitude and F faraday constant

The initial concentration of coagulant and pollutant are:

$$C_{i,Aluminum\ hydroxide} = 0$$

$$C_{i,pollutant} = C_0$$

5.3. Current distribution

There are three types of current and potential distribution (primary, secondary and tertiary current/potential distribution). The primary current distribution doesn't consideration the charge transfer on the electrode because of the negligible overpotential and concentration gradient. Secondary current distribution considers the activation overpotential by take place charge transfer on electrode surface. The current density is still low in comparison to the limiting current, so the concentration overpotential is neglected. Because of increasing the current above the limiting current, there is a concentration gradient nearby the electrodes, here the tertiary current distribution is obtained [8]**.** In this research the secondary current distribution were studied.

$$\nabla \cdot i_l = Q_l \quad , \quad i_l = -\sigma_l \nabla \varphi_l \quad (16)$$

$$\nabla \cdot i_s = Q_s \quad , \quad i_s = -\sigma_s \nabla \varphi_s \quad (17)$$

Where σ_l and σ_s is the electrolyte and electric conductivity(S/m) respectively ,φ_l and φ_s is the electrolyte and electric potential(V).

5.3.1. Boundary condition

The boundary of external electric potential at the anode and cathode are:

At anode $\varphi_{s,ext} = E_{cell}$

At cathode $\varphi_{s,ext} = 0$

6. Adsorption isotherm model

The amount of coagulant produced can be estimated by using Faradays law. Pollution reduction can be modeled by the adsorption phenomenon. The Langmuir isotherm is the most adsorption isotherm that used in EC modeling with the assumption of thermodynamic control [8].The Langmuir isotherm models are given below:

Langmuir isotherm: $q_e = q_{max} \frac{K_L C_e}{1+K_L C_e}$ (18)

Where q_e is the amount of adsorbed molecules per amount of adsorbent at equilibrium, C_e is the equilibrium adsorbate concentration in water, q_{max} is the adsorption capacity of adsorbed molecules per amount of metal cations, K_L is the Langmuir constant. So the reaction rate for pollutant removal by using Langmuir adsorption isotherm is:

$$-\frac{dC_t}{dt} = \varepsilon_M \cdot \varepsilon \frac{I}{ZFV} q_{max} \frac{K_L C_e}{1+K_L C_e} \quad (19)$$

Where ε_M is the efficiency of hydro-pollutant-aluminum formation, ε is the current efficiency, I is the applied current, Z is the valence of the electrode metal, F is the faradays constant, V is the volume of solution.

7. Mathematical model process

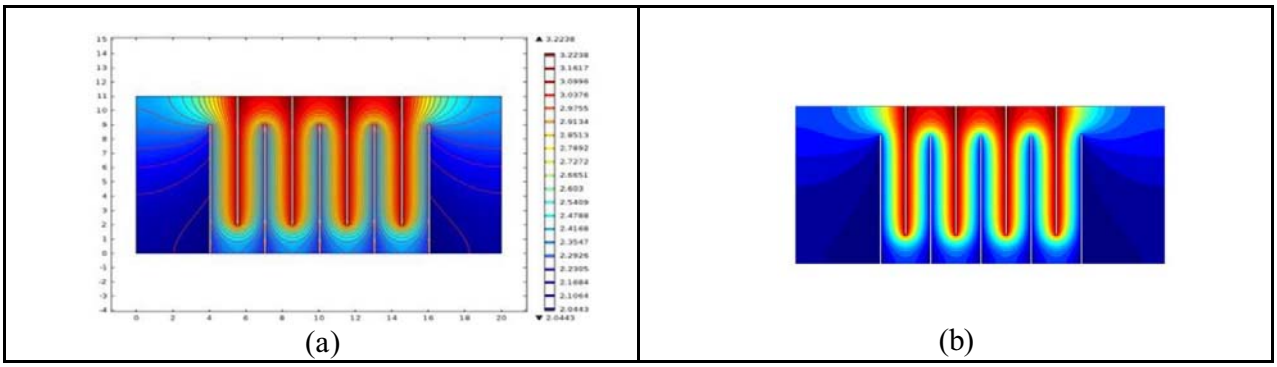
In this model the solution flow into the cylinder at the initial velocity. The three physics were used to incarnate all mechanisms occurred in EC reactor. The physics used in EC reactor are rotating machinery laminar flow for controlled the velocity of the solution, secondary current distribution for applied voltage or current on the anode and cathode, and transport of dilute species for diffusion of species in solution. The studies that used are stationary, frozen rotor and time dependent . Stationary is applied for secondary current distribution, frozen rotor for laminar flow and time dependent for transport of dilute species. After current applied in electrocoagulation, the aluminum ions are liberated at the anode and hydroxide ions are create by water electrolysis at the cathode. The Al^{3+} and OH^- will transfer into bulk solution and react together to form monomeric and polymeric hydroxides. To simplified the model assume that only amorphous aluminum hydroxide $Al(OH)_3$ (coagulant that adsorbed the pollutant) is considered.

8. Verification example

The research [8] studied the treatment of textile industry wastewater especially azo dyes by EC process. It simulated the EC system by Computation Fluid Dynamics (CFD). And it studied the distribution of the current and the velocity of the fluid flow on the cell

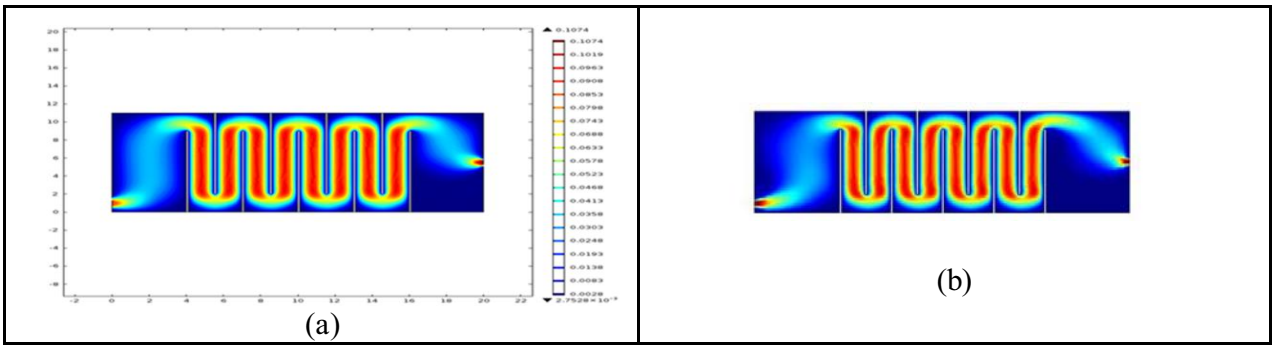
The EC system of this research consists of a continuous rectangular cell which contains five anodes and four cathodes. The cathodes are fixed on one side of the cell and the anodes in another side.

Applied this EC system in COMSOL software to show the obtain results. Figure(3,4) compared two results, as you see these results are the same.



(a) (b)

Figure 3. The secondary current distribution in the cell.(a) in reference[8] (b) in this work



(a) (b)

Figure 4. The spatial evolution of velocity magnitude (m/s) at mid-height in the cell (a) in reference[8] (b) in this work

9. Results and discussion

9.1. The distribution of various parameter in the EC reactor

This study simulated the distribution of voltage, current, velocity and concentration in the EC reactor.

Figure (5,6) show the solution of equation (16) with its boundary condition at steady state. And explaine the distribuion of voltage and current on EC reactor at different applied voltage (2, 5, 7.5, 10 V). By increasing applied voltage the current dencity increased, which the maximum of current density is (200A/m^2) when the applied voltage is 2V and (500, 800 ,1000 A/m^2) when the applied voltages are(5, 7.5 ,10 V) respectively.

Figure(5) explains how voltage is distributed on the system which the maximum voltage exists on the anode surface then it decreases slowly until it reachs zero at the surface of the cathode. The voltage at the surface of the cathode and cylinder is zero because this study assumed that, the external voltage is applied at the anode only. Figure(6) shows how current is distributed on the EC reactor which the maximum current exists at the corners of the anode and it decreases slowly to the center of the anode surface and to the cathode. It reachs nearly zero at the cathode surface. And it becomes zero at the surface of the cylinder.

Figure(7) explains the solution of momentum equation (eq.(11)) with its boundary condition for different value of rotational anode speed (0, 50, 100 and 150 rpm) . And explains how the velocity distributed in the EC reactor. At (0 rpm), there isn't speed distribution in the reactor because the anode doesn't rotate. Except at the inlet of the reactor which refers to flow rate of the wastewater when input to the reactor. As shown in this case, the maximum velocity exist at the inlet of the reactor which is nearly (0.02m/s) and it gradually decrease in the reactor.

When the rotational speed of the anode are (50, 100 and 150 rpm) the distribution of the velocity changes in the reactor, because the anode begins to rotate in the reactor and causes to move the species. In these figures the maximum velocity exists at the cathode surface because the electrolyte or wastewater quickly pass from the holes. And the velocity decreases slowly from the cathode to the anode and to the top of the cylinder. But the velocity at the surface of the wall cylinder is zero. The maximum velocity for these rotating speed are (0.2, 0.4 and 0.6 m/s) when the applied velocity are (50, 100 and 150 rpm) respectively.

Figure(8) show the distribution of residual lead concentration when the anode rotated in the reactor with speed (50 rpm), at HRT=45min and applied voltage=2v . When the anode rotates in the reactor the velocity of the solution increased. The high velocity makes the concentration homogeneous. So in definite time the amount of residual lead concentration is approximately equal in the whole reactor. But in the inlet of the reactor flow and the place around it the amount of lead concentration is more than other places. because, the wastewater continuously flow to the reactor. If using batch reactor instead continuous reactor this region doesn't appear and the concentration becomes homogenous in the whole reactor. This manner of distribution is because of rotating speed of anode, which causes to move the coagulant, so the coagulant diffused in bulk solution. This phenomenon causes to adsorb equally amount of pollutant in most region of the reactor. The amount of lead concentration remaining in the reactor at a certain time is different, which decreases by the time. The amount of residual lead concentration are (0.55, 0.5, 0.4 and 0.37 mol/m^3) when the times are (10, 20, 30 and 40min) respectively.

Figure(9) show the distribution of coagulant production in the same conditions of figure(8). As explained previously, when the anode rotates in the reactor the velocity of the solution increased and causes to make the concentration homogeneous. So the distribution of the coagulant production is the same with the distribution of the lead concentration, but the distribution of the coagulant in the input flow and the place around it, is lower than other places. Figure(10) explains how the lead concentration decrease and coagulant increase with time at the same conditions of above section in the reactor.

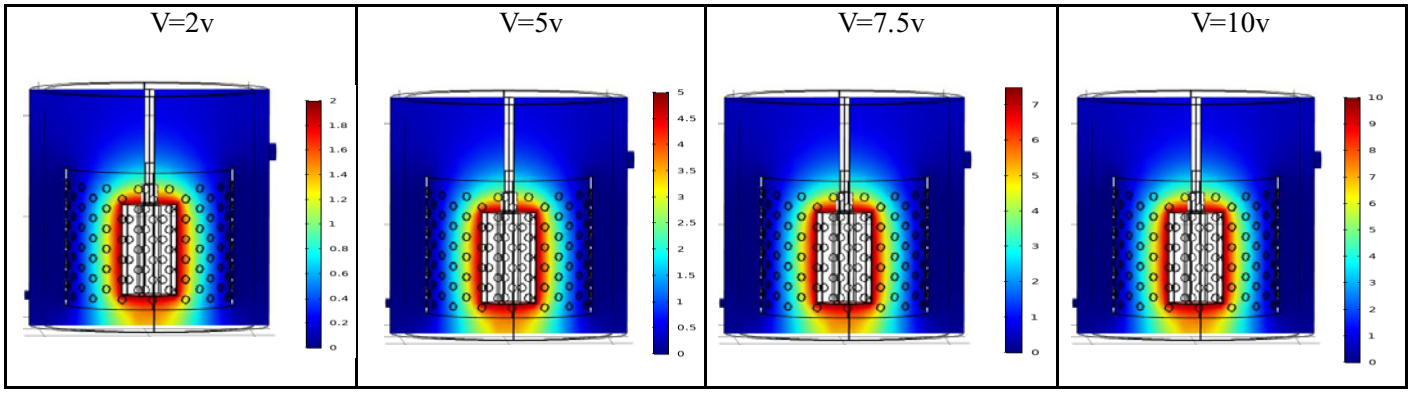


Figure 5. The distribution of the voltage in the EC reactor at different applied voltages.

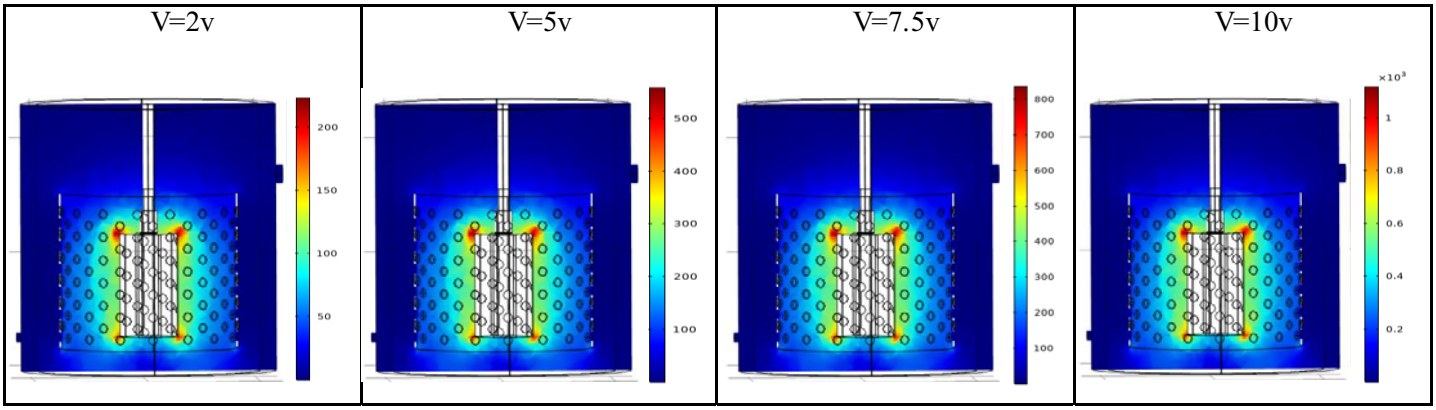


Figure 6. The distribution of the current in the EC reactor at different applied voltages.

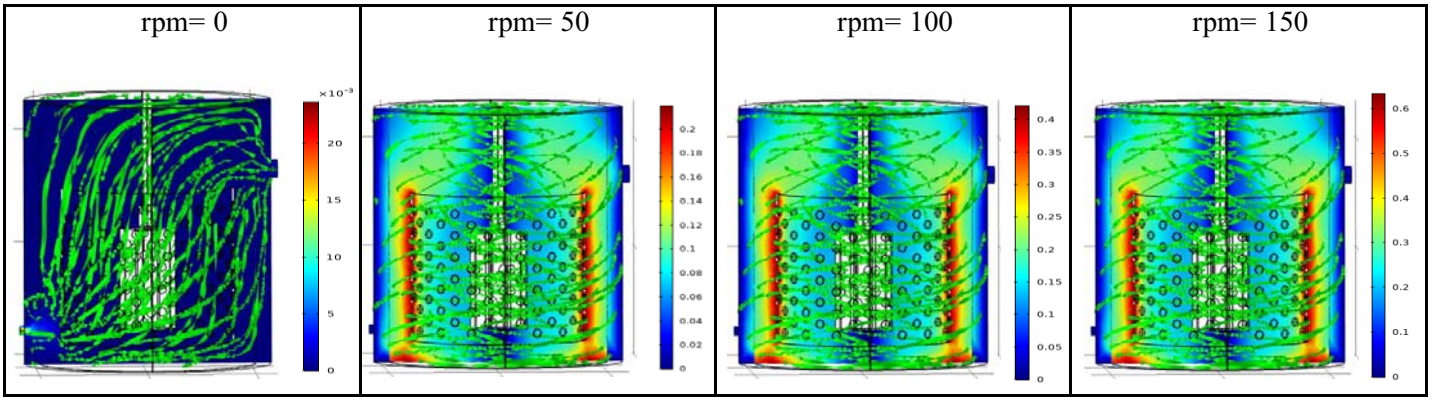


Figure 7. The Distribution of the Velocity of the Fluid in the EC Reactor at different Rotating anode Speed

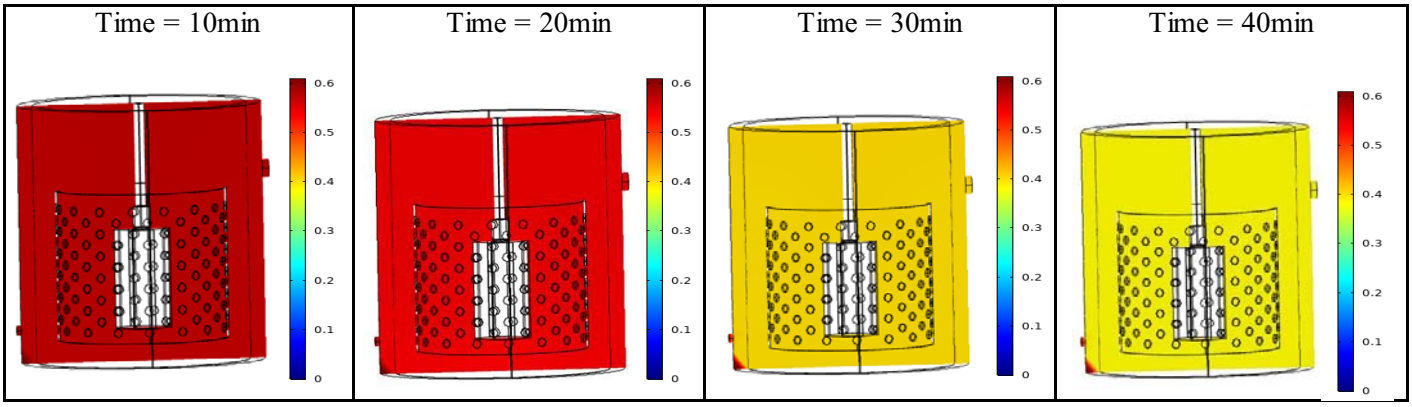


Figure 8. The Distribution of the lead Concentration in EC Reactor for definite time when (HRT= 45min, RPM= 50 rpm and V=2v).

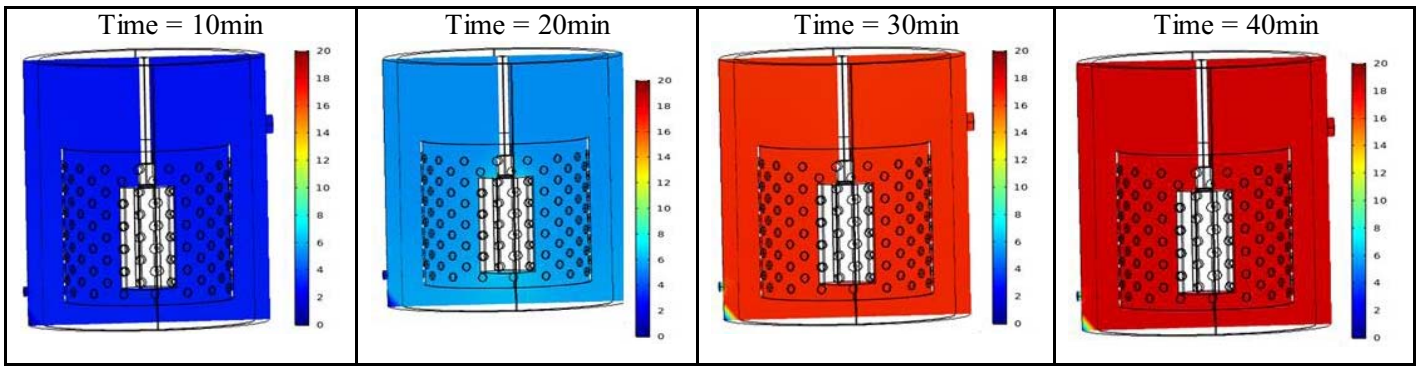


Figure 9. The Distribution of Aluminum ions production in the EC Reactor for definite time when (HRT=45min , RPM= 50 rpm and V=2v).

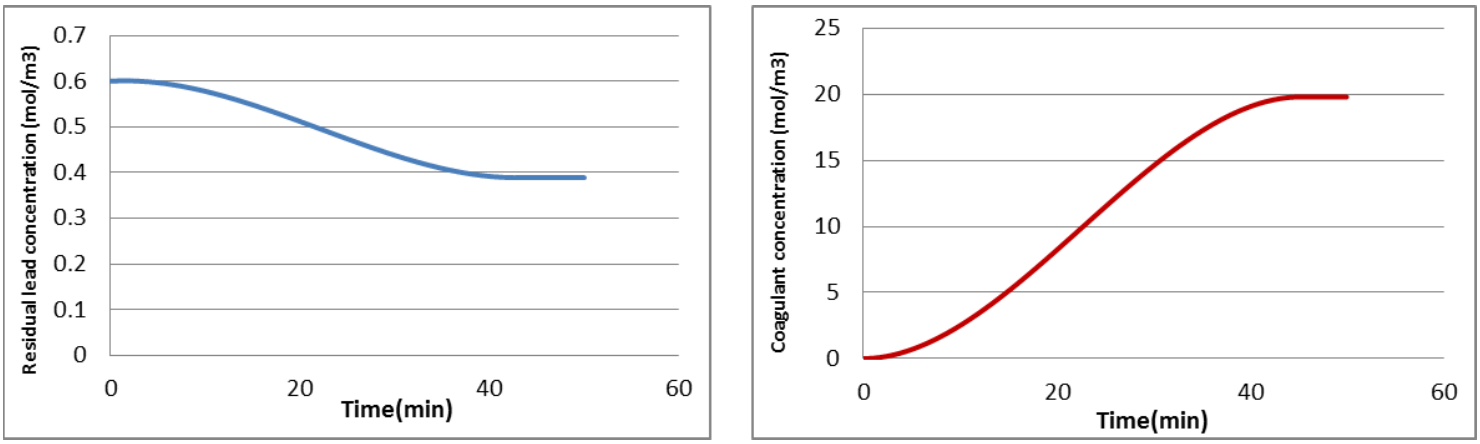


Figure 10. the residual lead and coagulant concentration in the reactor with time (a)residual lead concentration (b) coagulant concentration.

9.2. Langmuir isotherm coefficients obtain in optimization

As explains previously, the Langmuir isotherm equation eq.(18) represent the adsorption phenomenon in theoretical model. There are two parameters in this equation which are q_{max} and K_l that can be used in theoretical model to control the EC process. These two parameters were determined by optimization in COMSOL software.

Table (2) shows the values of Langmuir isotherm coefficients that determined by optimization in themodel. This table shows the effect of voltage on these parameters for different HRT. As shown in this table, by increasing applied voltage, the value of these parameters are near to each other. And also shows that by increasing the HRT these value are decreased gradually.

Figure(11) shows the prediction results which obtained when using optimization values in theoreticalmodel and compare them with experimental results. And show the effect of applied voltage on residual lead concentration with running time for different HRT (15, 25, 35 and 45 min).

As seen in these figures, at 2volt the prediction results is close to experimental results in all cases so the optimization values in these cases are correct. But at 5volt there are differences between them. Where the prediction result for HRT=35min(fig.11c) is differ from others, which the residual lead in the reactor decrease with time until reach to 30min and then increase slowly for 15min and again begin to decrease. This may be because the optimization value that used are incorrect or need to modification. In other cases at 5V the prediction results is decrease with the time as experimental results, but the prediction result at HRT=15min is far from the experimental result than other cases.

The removal efficiency in theoretical results is better than experimental results because the operating condition in the experience cannot keep constant and change with the weather. These figures show that, similar to experimental work by increasing applied voltage the efficiency of lead removal increased . And show by increasing HRT the efficiency of lead removal increased.

Table 2. Langmuir Isotherm coefficients at Different HRT for lead ions removal in model.

HRT(min)	Voltage(V)	Langmuir Isotherm	
		q_{max} (mg/g)	K_L (L/mg)
15	2	6.38E-01	5.75E-04
	5	5.50E-01	5.50E-04
25	2	5.00E-02	5.00E-05
	5	7.00E-02	7.00E-05
35	2	1.00E-03	1.00E-06

(b)		5	9.00E-04	2.00E-06 (a)
	45	2	1.00E-04	1.00E-07
(d)		5	7.00E-05	7.00E-08 (c)

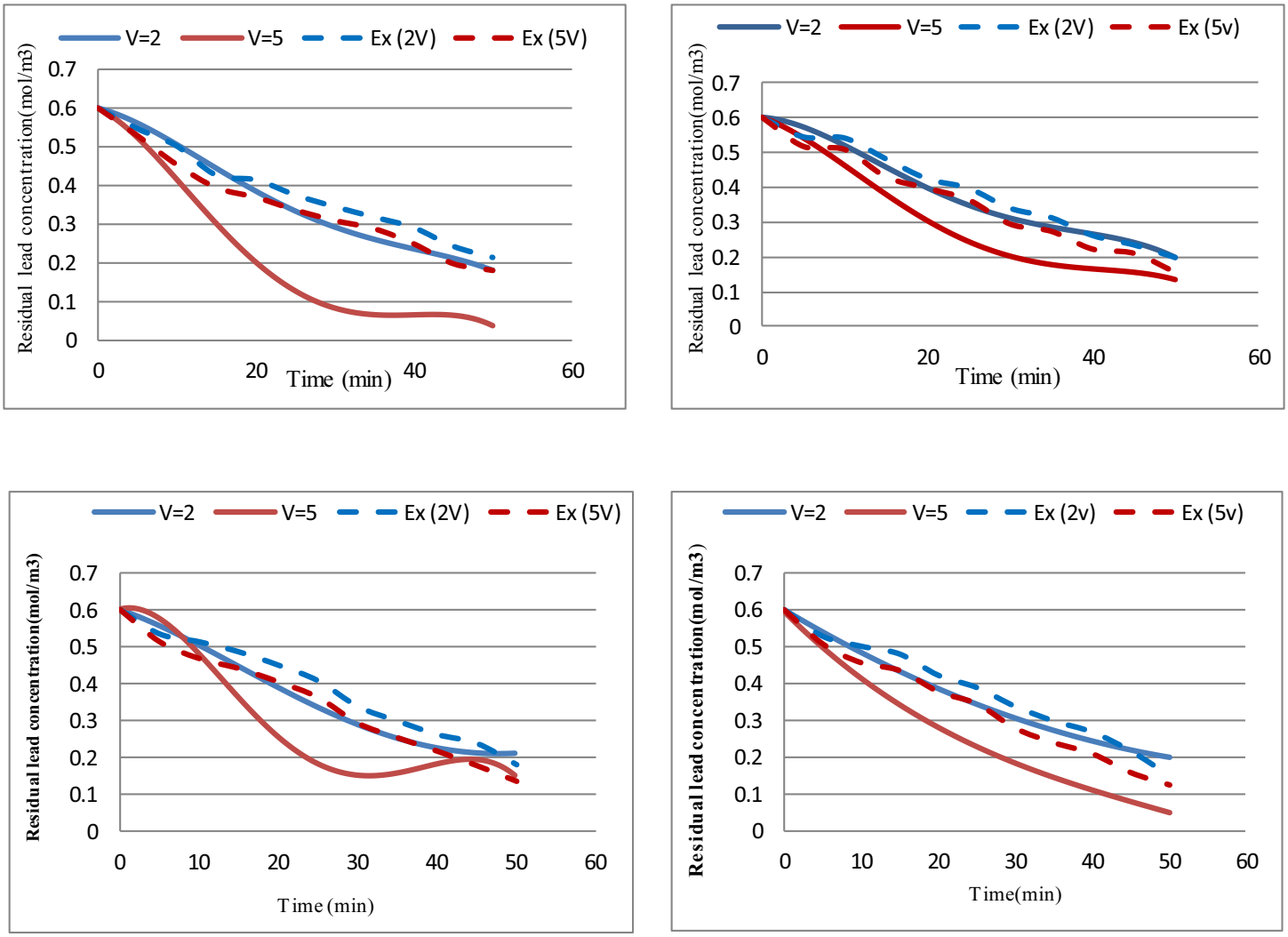


Figure11. The Effect of Applied Voltage on Residual Lead Concentration at different HRT ; a)15min , b)25min , c) 35min , d) 45min.

10. Conclusions

The following points can be concluded from this work

- A mathematical model was successfully established to simulate the treatment of the wastewater that contains lead in a continuous EC process.
- The distribution of the voltage, current, velocity and concentration in the reactor were modeled.
- The removal of lead are controlled by Langmuir isotherm coefficients that used in the simulated reactor. And these influenced on the process, which decreased by increasing HRT.
- The simulated results show that the mathematical model reasonably predict the EC operation performance for the removal of lead from water, then were compared them with experimental results.
- The results show that the removal efficiency increases with the increase of hydraulic retention time (HRT) and applied voltage. Also, the results indicate that the increase in the rotation of electrode improves the removal efficiency which reaches the maximum at (100rpm), while at higher velocity (150 rpm) the removal efficiency decrease gradually due to the destabilization of flocks that are formed.
- There was agreement between theoretical and experimental results.

Reference

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