



Modelling and Simulation for Removal of Lead ions (Pb^{+2}) from a Synthetic Wastewater by Electrocoagulation Using Aluminum (Al) as a Rotating Electrode in a Batch Process by Using COMSOL Multiphysics

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Abstract

There are some technique that utilized for wastewater treatment, electrocoagulation (EC) is one of them. This method is useful for wastewater treatment, which has heavy metals (lead compounds) as contaminants. Computational Fluid Dynamics (CFD) modeling was utilized to simulate the batch (EC) unit in this study. To model species transport, current-voltage distribution and fluid movement, the diffusion, Nernst-Planck, and Navier-Stokes formulas have been employed. The system of formulas that governs this issue has been solved utilizing commercial software (COMSOL 5.4). Three main variables that impacted the performance of the lead EC process were tested: different starting lead amounts (200, 300, 400, 500 ppm), various applied voltage (2.5, 5 V), and various anode rotating velocities (50, 100, and 150 rpm). The removing rates of lead (removing effectiveness) reduced as the original lead level increased, according to the findings. And removing efficacy, which improved with increasing applied voltage, but not with increased rotational anode velocity, which climbed from 50 to 100 rpm before beginning to drop at 150 rpm. Since the instability of the produced flocks, the eliminating effectiveness progressively decreases. Utilizing (Al/St.St) electrodes, the best removal efficacy has been obtained at 200 ppm, 5 volts, and 100 rpm rotating velocity.

Keywords: COMSOL, EC process, lead removing efficacy, mathematical model.

1. Introduction:

Water and energy are the two most pressing issues facing the twenty-first century. Water quality has become a critical issue as a result of increasing contamination from point and non-point sources, particularly in Third-World countries [1]. Industries containing heavy metals, including Pb, seem to be the most dangerous among chemical intensive industries since the discharge of huge quantities of metal-contaminated wastewater [2]. Heavy metals are indeed a key contaminant in the environment and are rapidly being one of the most severe environmental issues. To safeguard humans and the environment, these hazardous heavy metals must be eliminated from wastewater [3]. Throughout the last two decades, electrochemical technologies have gained prominence globally, and processes including electrochemical metal recovery, electrocoagulation (EC), electro-flotation (EF), and electro-oxidation (EO) may now be considered established

technologies. Electrocoagulation is one of the most common uses of electrochemical reactor technology for the purification of water in the environmental sector. EC is a complex process that involves a number of chemical and physical processes and relies on consumable electrodes to deliver ions to the water stream [4].

Three main mechanisms are involved in electrocoagulation. [5,6]

1. Coagulants are formed by electrically dissolving the consumable electrode.

2. Destabilization of impurities, particle suspension, and emulsion cracking are all catalyzed by these coagulant reagents.

3. The turbulence created by the oxygen and hydrogen development creates a soft mix that aids in the flocculation (linking together and generating larger particles) of destabilized colloids (flocks). Electro flocculation is a term utilized to describe this procedure. Eventually, the contaminants are removed from the waste via sedimentation, filtering, or

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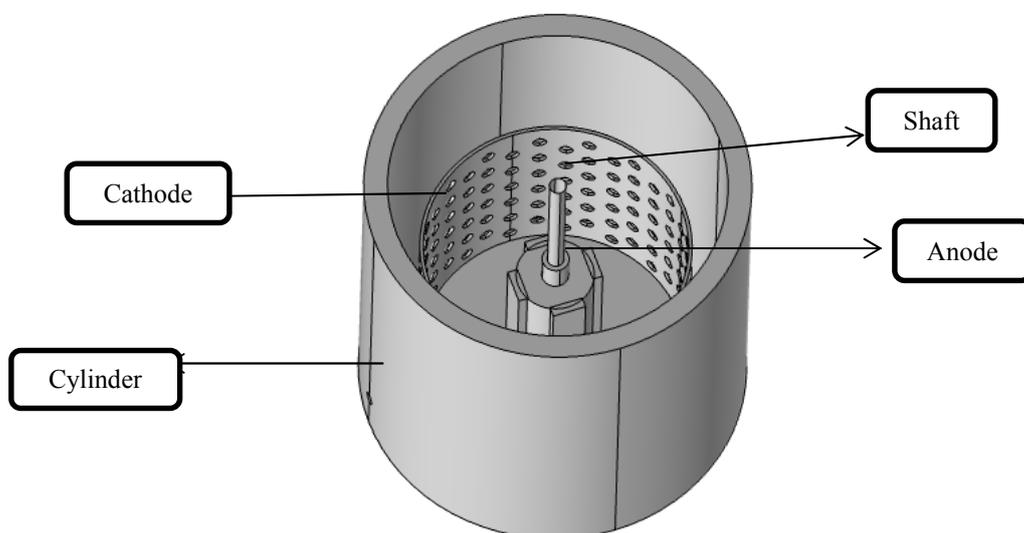
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flotation. The formation of bubbles in the oxygen, and especially in the hydrogen evolution, may aid in improving the efficiency of the latter process (electro flotation).

The following project simulate a mathematical model of the batch EC processing that able to forecast the lead removing from wastewater by utilizing commercial software (COMSOL 5.4). And also investigated the impact of various parameters including initial lead amount, applied voltage and various rotational velocity on its performance.

2. EC reactor geometry

The batch EC reactor model is made out of a cylinder with measurements of (r=57mm, h=117mm). The cathode and anode are utilized in this cylinder. The cathode is indeed a hollow stainless steel cylinder with holes on its surface, with measurements of (r=40mm, h=68mm). The anode is a cylinder made of



aluminum that measures (r=11.5 mm, h=45mm) and seems to have four 2mm thick grooves. It was attached to a shaft that rotated the anode. Fig.(1) depicts the model's batch EC system topology.

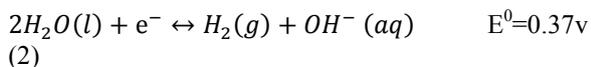
3. Reactions that happens at the surface of electrodes

Metal cations are electrochemically introduced in situ during electrocoagulation. [7].

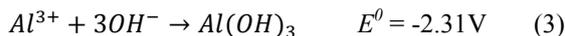
The reaction of anode is



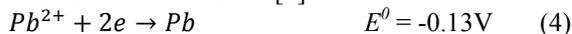
By decreasing the water on the cathodes sides, the hydroxyl ion (OH) and hydrogen gas (H₂) have been produced.



As well as in the bulk solution, the Al(OH)₃ have been produced.



Lead electro-deposition may happen as a secondary reaction at the cathodes. [8]



The cathodes generates OH⁻, which reacts with Pb²⁺ and Al³⁺ ions. The resulting amorphous Al(OH)₃ have high surface areas, which are helpful for adsorption and trapping of Pb²⁺ ions. The Pb²⁺ ions adsorption on Al(OH)₃ has been described by the following process.



4. Governing formulas

The governing formulas on the EC process are the Navier-Stokes, Nernst-Planck, and diffusion formulas. These formulas are utilized to describe the transport of fluid flow, current-voltages distribution, and species of the wastewater in the EC unit model.

4.1. Momentum transfer

The continuity formula and incompressible Navier-stokes (NS) formulas is utilized to express the EC reactor flow behavior [9].

-Continuity formula

$$\frac{\partial u}{\partial x} + \frac{\partial u}{\partial y} + \frac{\partial u}{\partial z} = 0 \quad (6)$$

-General momentum formula

$$\rho \frac{\partial u}{\partial t} + \rho(u \cdot \nabla)u = \nabla \cdot [-pI + \mu(\nabla u + (\nabla u)^T)] + F \quad (7)$$

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

4.1.1. Situation of boundary

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

$$u = u_w = \left(\frac{\partial x}{\partial t} \right)_x \quad (8)$$

$$dx = dx(r_{bp}, \omega, t) \quad (9)$$

$$\frac{d\omega}{dt} = w \quad (10)$$

At the surface wall (cylinder) and cathodes, the velocity=0 (u=0)

4.2. Ionic species' transferred mass

In EC system, There were three transferred flux mass kinds, which are electro-migration ($Z_i u_{m,i} F C_i \nabla \varphi$), diffusion ($D_i \nabla C_i$) and convection ($C_i u$). The species of transferred flux mass i could be obtained by Nernst-plank formula.

$$N_i = -D_i \nabla C_i - Z_i u_{m,i} F C_i \nabla \varphi + C_i u \quad (11)$$

Whereas: electrical potential is φ , constant of Faraday is F, velocity is u , mobility of species i is $u_{m,i}$, the charge number is Z_i , the average amount is C_i , D_i is the coefficient of diffusion and N_i is the chemical species flux since convection.

The conservation of charge and species is given by:

$$\frac{\partial C_i}{\partial t} = -\nabla N_i + R_i \quad (12)$$

Whereas R_i is the reaction rate of species i [9].

4.2.1. Situation of boundary

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

$$\text{At anode} \quad N_i = \frac{i}{ZF} \quad (13)$$

Whereas i is the current density value of electrolytes and Constant of Faraday is F

The initial coagulant amount and contaminant were:

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

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

4.3. Current distribution

the secondary current distribution utilized to describe the current distribution in the theoretical model.

$$\nabla \cdot i_l = Q_l, \quad i_l = -\sigma_l \nabla \varphi_l \quad (14)$$

$$\nabla \cdot i_s = Q_s, \quad i_s = -\sigma_s \nabla \varphi_s \quad (15)$$

Whereas 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).

4.3.1. Situation of boundary

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

$$\text{At anodes} \quad \varphi_{s, \text{ext}} = E_{\text{cell}}$$

$$\text{At cathodes} \quad \varphi_{s, \text{ext}} = 0$$

And when specific electrical current flow in the electrolytic cell, the Al mass is dissolved from the anodes, and it quantified by law of faradays [10].

$$W = \left[\frac{ItM}{ZF} \right] \quad (16)$$

Whereas: the constant of Faradays is F (96485 C/mol), the electrons number is Z included in the reaction, the specific molecular electrodes weight is M (g/mol), the electrolysis duration is t, the current is I (A), the dissolved material of anode amount is W(g).

5. Adsorption isotherm model

Faraday's law may be used to determine the quantity of coagulant produced. The adsorption phenomena

may be used to simulate contamination decrease. One of the adsorption isotherm models used in this EC modeling is the Freundlich isotherm [11]. Below are the Freundlich isotherm models:

Freundlich isotherm:

$$q_e = K_F C_e^{1/n} \quad (17)$$

Whereas q_e is the adsorbed molecules amount per adsorbent amount at equilibrium, C_e is the equilibrium adsorbate amount in water, K_F and n are indicating Freundlich constant and intensity respectively. So the rating of reaction for contaminant removing by utilizing Freundlich isotherm of adsorption is:

$$-\frac{dC_t}{dt} = \varepsilon_M \cdot \varepsilon \frac{I}{Z F V} K_F C_e^{1/n} \quad (18)$$

Whereas ε_M is the hydro- contaminant-aluminum creation efficacy, ε is the current efficacy, I is the applied current, Z is the electrode metal valence, F is the constant of Faradays, V is the solution volume.

6. Results and discussion

After entering all the formulas that need to solving the problem in the theoretical model the extracted findings from the COMSOL software demonstrate how the velocity, voltage, current density and amount are distributed in the EC reactor.

6.1. Velocity, voltage, current density and concentration distribution

Fig.2 demonstrate the velocity distribution on the batch EC reactor. Since there are numerous tiny holes on the cathodes' surface and the electrolyte rapidly passes from the small surface area, the greatest velocity occurs at the cathode's surface at different rotating speeds of the anodes, as illustrated in this Fig. (holes) from the cathodes to the anodes, and finally to the top of the cylinder, the velocity steadily diminishes. The maximal velocity for these rotational speeds is (0.2, 0.4, and 0.6 m/s) once the applied velocities on the reactor is (50, 100, and 150 rpm).

Fig.3 illustrates the current density and voltage distribution on the batches EC reactor at various applied voltages (2.5, 5V). The highest voltage occurs on the anode surface, then gradually declines until it approaches zero at the surface of the cathode, and the lowest voltages have been at the cathodes and the cylinder surface, as illustrated in Fig.3(a). As demonstrated in Fig.3(b), increasing the applied voltages increases the current density, which the current density is (250A/m²) when the applied voltages has been 2.5V and 500A/m² once the applied voltages have been 5V. The greatest current density occurs in the extremities of the anode, and it gradually decreases to the middle of the anode surface and to the cathodes, as demonstrated in Fig.3(b). And it reaches zero at the cylinder's surface.

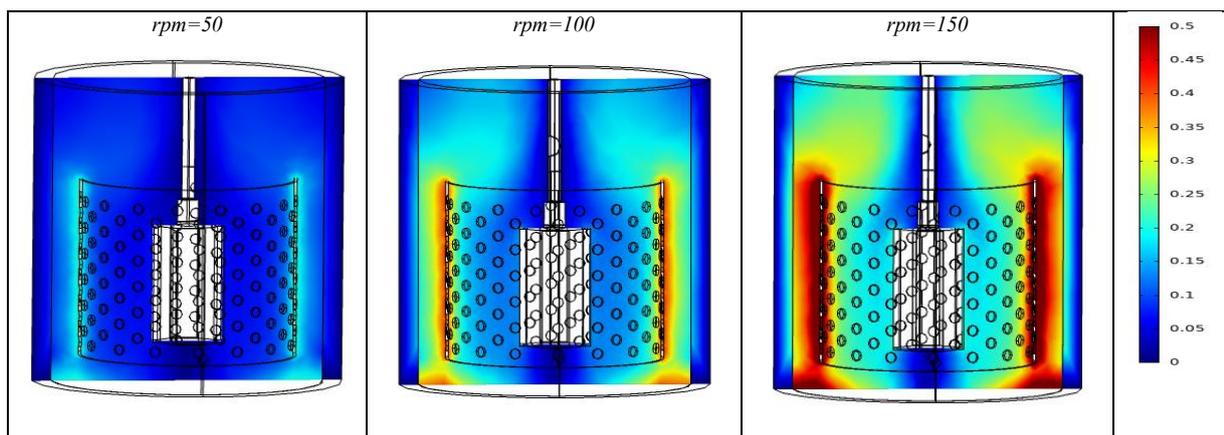


Fig.2 Distributions of the fluid velocity in the EC reactor at various Rotational anode speed.

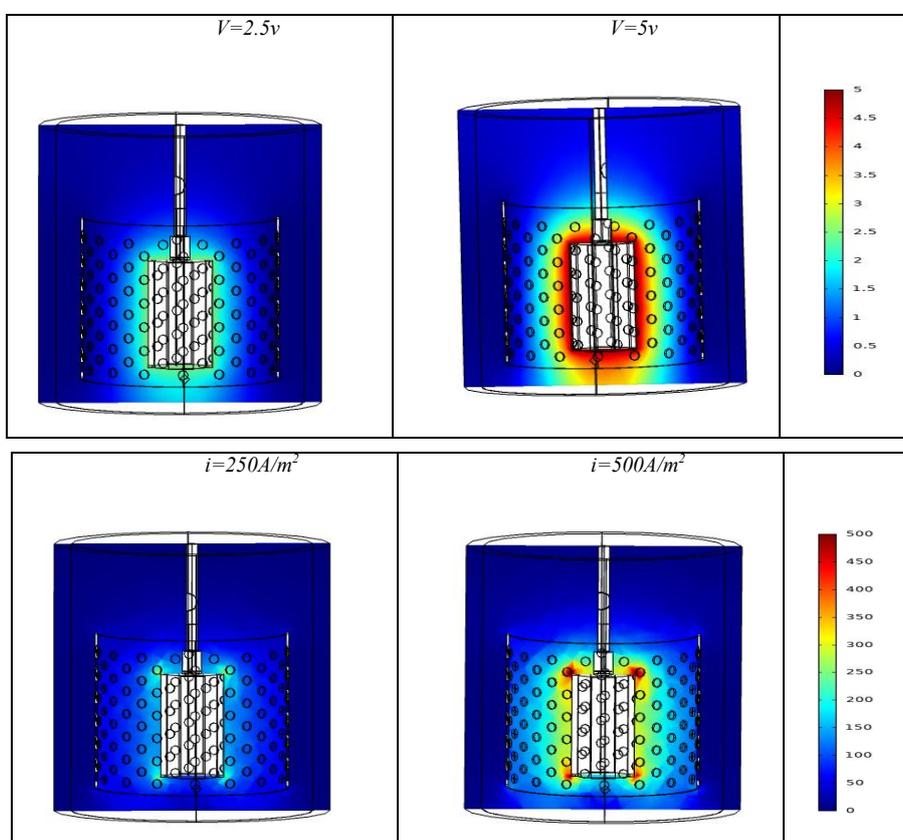


Fig.3 Distribution of the (a) potential and (b) current density in the EC reactor at various applied voltages

The rotational velocity is one of the significant factor that influence the performance of the EC processing. The primary function of the rotational velocity is to transfer the coagulant substance, that is produced by electrodes dissolution in efficient manner in the reactor. The Fig.4 demonstrates how the amount of the (a) coagulant and (b) contaminant (lead ions) are

distributed in the reactor when the rotational velocity is zero. As demonstrated in this figure the amount distribution is nonhomogeneous because the coagulant substance does not distribute in the reactor, therefore the regional change can be observed. But as demonstrated in Fig.5, when the anode rotates in the reactor the velocity of the solution increased. The

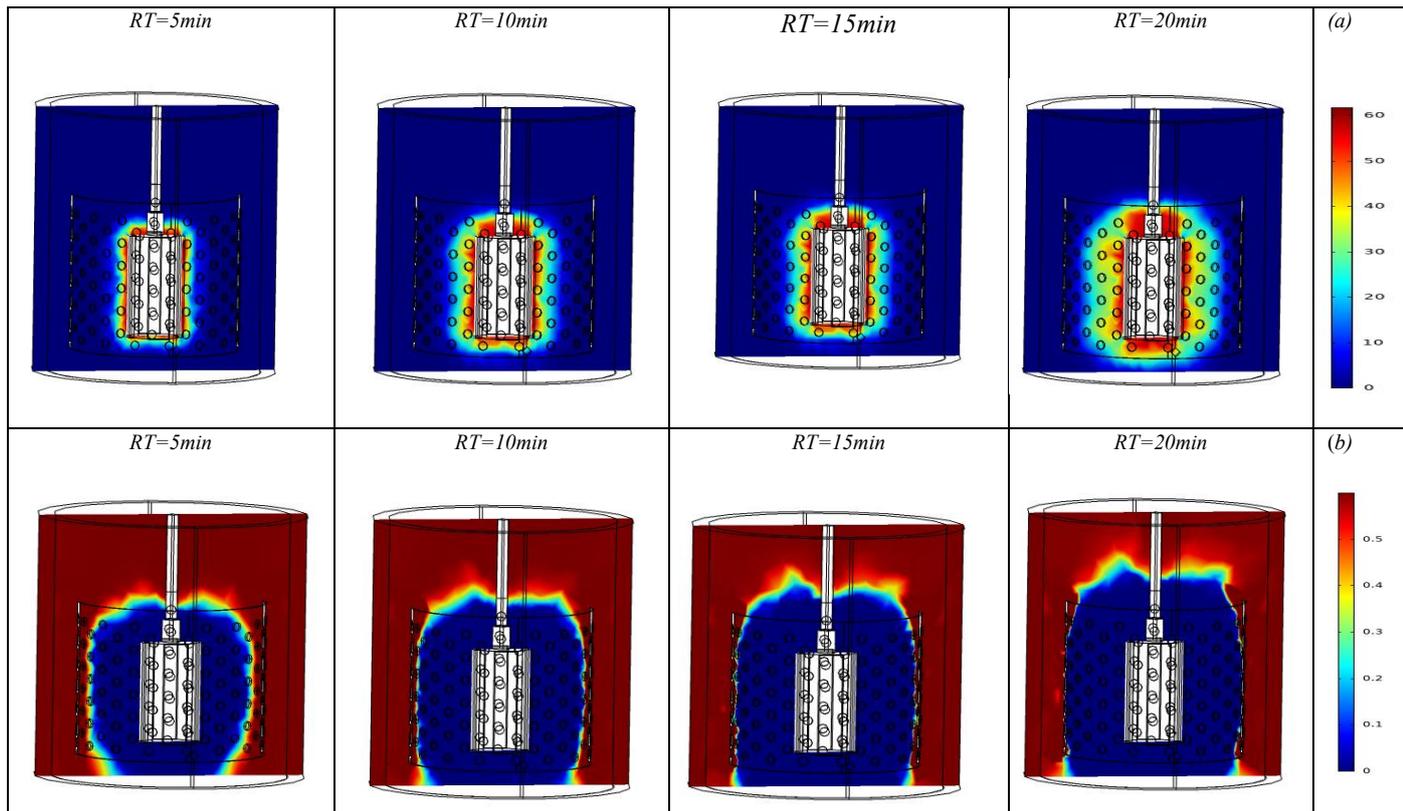
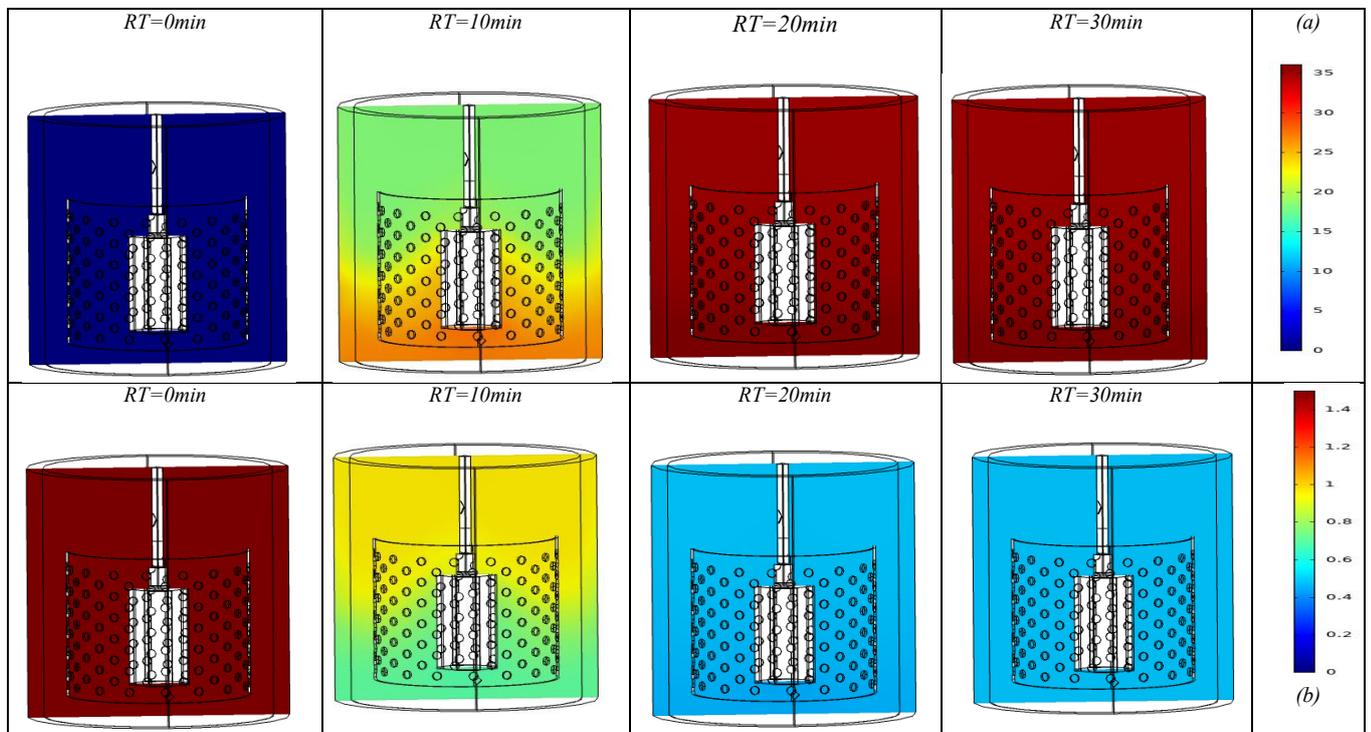


Fig.4 Distribution of the (a) coagulant and (b) lead amount in EC reactor for various residence time when ($C_0=200ppm$, $RPM=0$ and $V=5v$).

high velocity, make the amount homogeneous. So as demonstrated in Fig.5 the distribution of the coagulant production is the same with the distribution of the lead ions amount in the reactor. As

demonstrated in (Fig.5(a)) by increasing residence time the amount of coagulant product increased



F.5 Distribution of the (a) coagulant and (b) lead amount in EC reactor for various residence time when ($C_0=500ppm$, $RPM=100$ and $V=5v$).

therefore the amount of adsorbing lead ions increase (Fig.5(b)). This pattern of distribution is since the anode's rotational speed, which causes the coagulant to migrate, caulitizing the coagulant to diffuse in bulk solution efficiently. As a result of this occurrence, an equal quantity of contaminant is adsorbing in most of the reactor's regions.

In the theoretical model, the isotherm of Freundlich (formula 18) represents the adsorption phenomena. The coefficients of Freundlich, K_F and n , represent two factors in this formula that may be utilized in a theoretical model to regulate the adsorption mechanism. These two parameters have been obtained via COMSOL software optimization. The magnitudes of the Freundlich coefficients of isotherm obtained via optimization in the model are demonstrated in table1. The impact of voltage on these characteristics for various initial lead amounts is demonstrated in this table. The magnitudes of K_F get closer to each other when the applied voltage is increased, as seen in this table. Also, raising the initial lead quantity causes these magnitudes to progressively drop. And the n did magnitudes don't vary all that much.

6.2. The impact of applied voltages on the removing efficacy of lead ions

For various initial amounts of the contaminant, the impact of applied voltage on the removal effectiveness of lead ions is demonstrated in Fig.6 (200, 300, 400 and 500ppm). With increasing voltage, current density affects both coagulant dose and bubble formation rates, as well as having a significant effect on solution blending and mass transfer at the electrodes. As could be observed in this diagram, removing grow as the voltage applied increases. Furthermore, raising the initial lead amount reduced the removing efficacy. In addition, the Freundlich coefficient of isotherm is calculated in a theoretical model utilizing COMSOL program to estimate lead removal effectiveness in an EC reactor model. The magnitudes of the Freundlich coefficients of isotherm obtained via optimization in the model are demonstrated in table 2. For various initial lead amounts, this table illustrates the effect of rotational velocity on these parameters. As seen in the table, increasing rotational velocity brings the magnitudes of K_F closer together, and increasing the initial lead amount causes these magnitudes to progressively

decrease. And the n magnitudes don't vary all that much.

6.3. The impact of the rotational anode velocity on the removing efficacy of lead ions

Fig.7 depicts the effect of rotational anode velocity on lead removal effectiveness. It demonstrates that raising the solution's rotational anode velocity (50-100) rpm increases the removal effectiveness. Then, as the rotational anode velocity (100-150) rpm increases, coagulant matter composed of aluminum ions attaches to and disperses throughout the cell, making the cell's composition homogeneous. Higher stirring rates, on the other hand, may result in the instability of flocks produced in the cell, resulting in a reduction in effectiveness. As a result, the optimum rotational anode velocity was achieved at 100 revolutions per minute. The optimum rotational velocity for all quantities was 100rpm, but also revealed that the flocks deposited between electrodes since they couldn't mix evenly, and this deposition produced an increase in cell resistant at low stirring speeds. In systems with constant current density, a rise in cell resistance produces an increase in potential magnitude, which leads to an increase in energy usage per unit volume. The increase in stirring speed from 50 to 100 appears to confirm that the removing efficacy seems to be diffusion controlled, and the increase in stirring speed results in an increase in the intensity of turbulence, which decreases the diffusion thicknesses at the electrode surface and enhances the blending situations in the electrolyte bulk [12].

6.4. The impact of initial lead ions amount on the removing efficacy.

The optimal condition for amount effect was illustrated visually in Fig.8 by the effect of starting quantity on EC of lead ions. The removal of lead ions takes longer at higher concentrations, as seen in this graph, although higher starting concentrations of lead were decreased considerably faster than lower concentrations. This may be described in the following way: (1) According to Faraday's law, a consistent quantity of Al^{+3} went into solution at the same current density and duration for all lead levels, but Al^{+3} was inadequate for solutions with greater lead quantities, resulting in the same number of

Table 1. Freundlich coefficients isotherm at various C_0 and applied voltage for lead ions removing in model.

C_0 (ppm)	Voltage	Freundlich isotherm	
		$K_f(\text{mg/g})(\text{L/mg})^{1/n}$	N
200	2.5	8E-3	0.9
	5	2E-3	0.96
300	2.5	6E-4	0.975
	5	4E-4	0.98
400	2.5	4.5E-7	0.93
	5	1E-11	0.94
500	2.5	3E-6	0.93
	5	1E-12	0.97

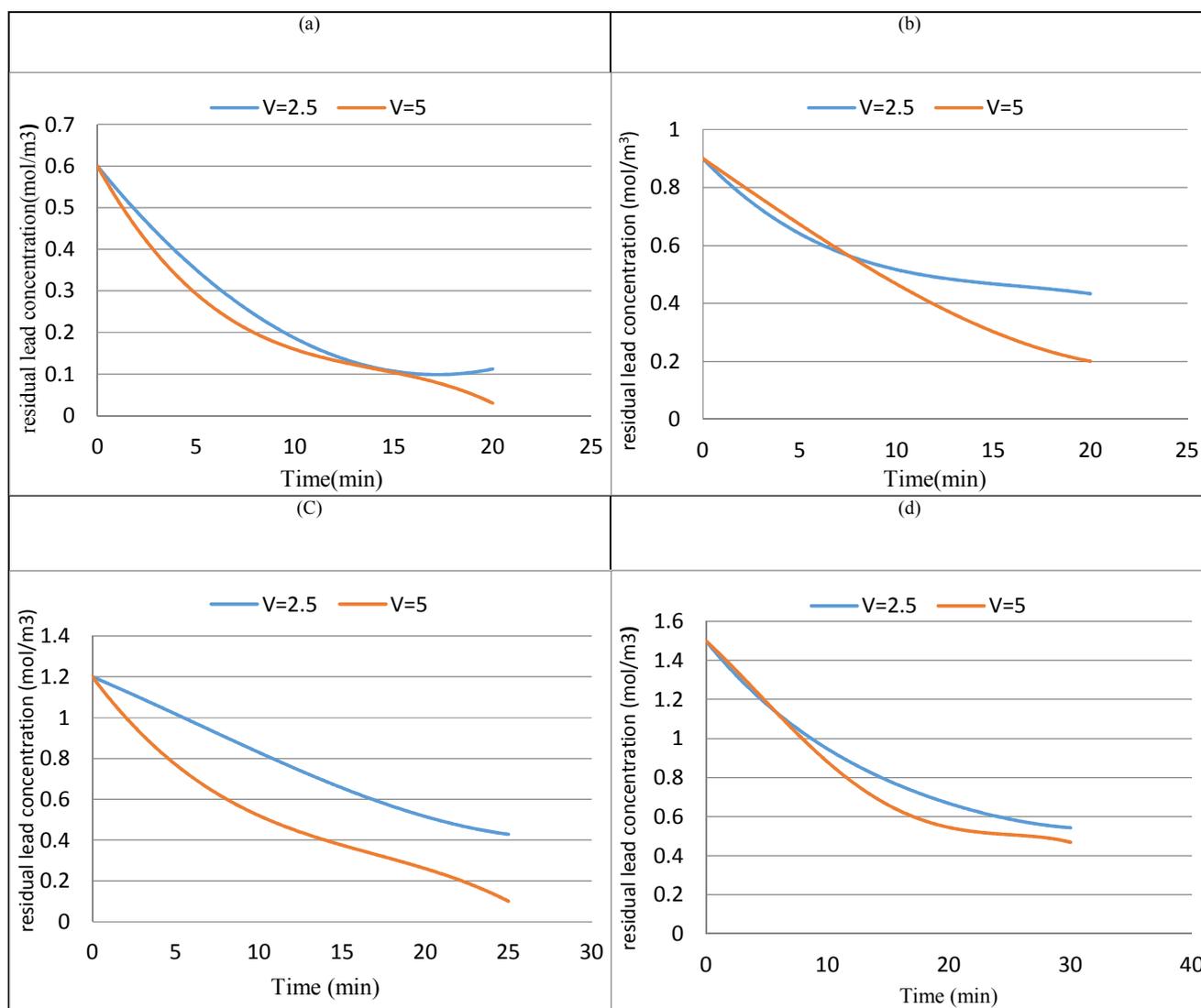
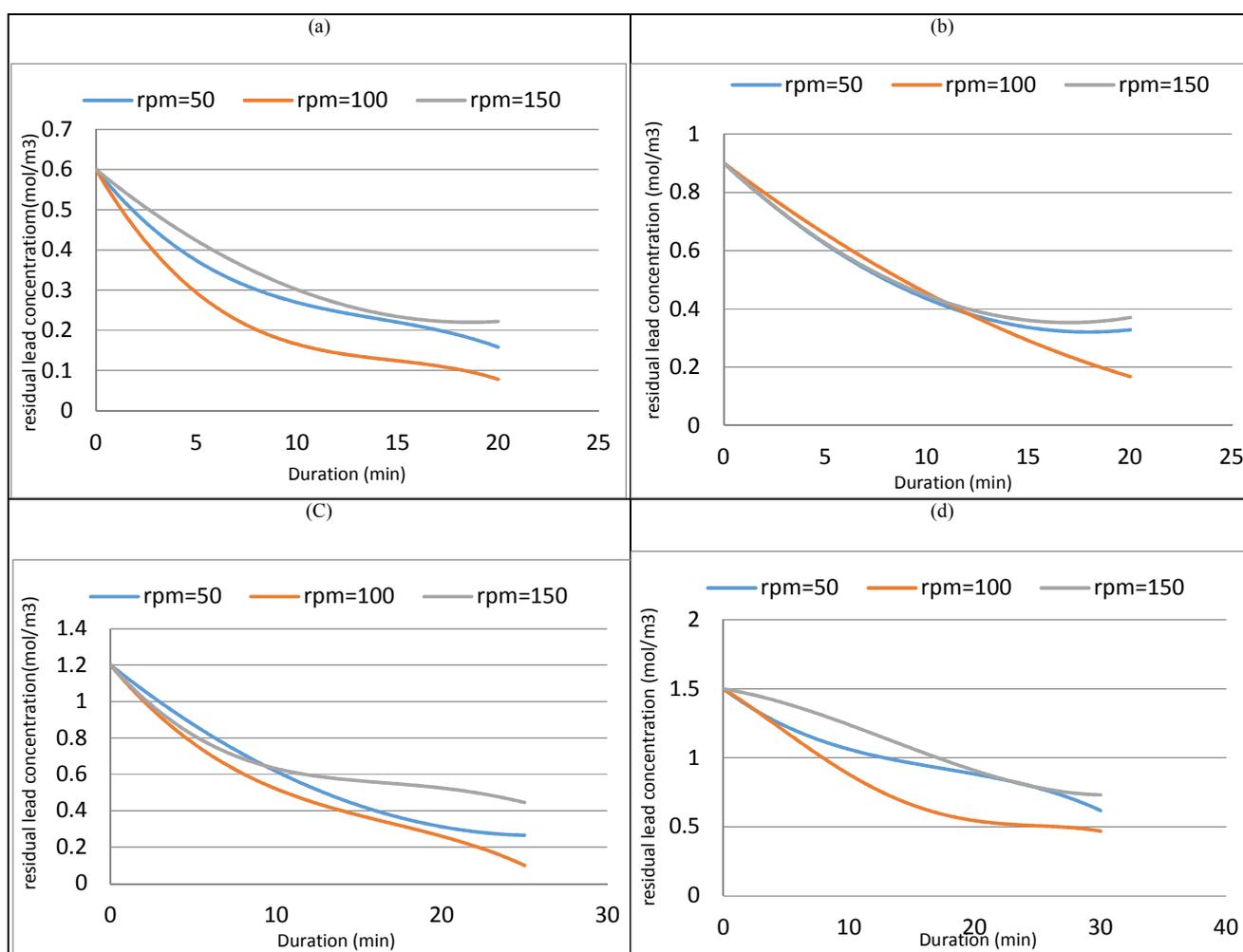

Fig.6 Impact of applied voltages on remaining lead amount at rotational velocity 100rpm and at various initial amount; a)200ppm , b)300ppm , c) 400ppm , d) 500ppm.

Table 2. Freundlich isotherm coefficients at various C_0 and rotational velocity for lead ions removing in model.

C_0 (ppm)	Freundlich isotherm		Freundlich isotherm	
			$K_F(\text{mg/g})(\text{L/mg})^{1/n}$	n
200	50		9E-5	0.97
	100		2E-3	0.96
	150		9E-4	0.96
300	50		9E-7	0.96
	100		4E-4	0.98
	150		4E-4	1
400	50		5E-11	0.98
	100		1E-11	0.94
	150		3E-12	0.92
500	50		5E-10	1.2
	100		1E-12	0.97
	150		1E-11	0.98

**Fig.7** Impact of rotational velocity on residual lead amount at applied voltage 5V and at various initial amount; a)200ppm , b)300ppm , c) 400ppm , d) 500ppm.

flocks in the solution. (2) Despite the fact that the same quantity of coagulant was generated in the EC cell at the same current density for various lead

concentrations, the amount of coagulant species was the same. According to other studies [13], as the quantity of lead in the solution increased, the applied

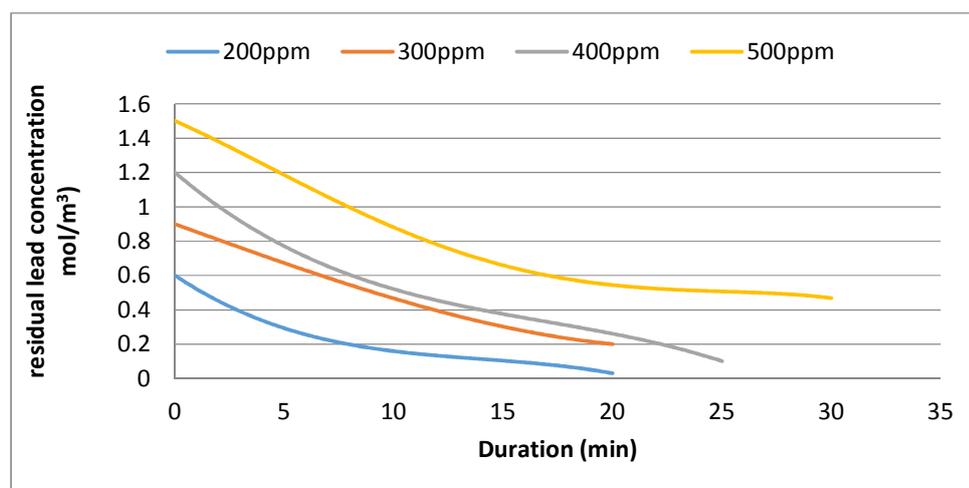


Fig.8 Impact of Initial lead ions amount on removing efficacy of Lead (Rotational Velocity= 100rpm, V=5v).

potential for the solution dropped, as did the amount of energy utilized.

7- Conclusion

EC is suitable way to wastewater treatment that contain various pollutant and heavy metals such as Hg, Pb, Cr, Cd, Cu, etc; because of its low cost, efficiency, simple, eco-friendly, versatile and its also nonspecific technique. Although this benefits of EC process it is difficult to simulate because of its complex process. CFD modeling is a good model that can simulate the EC process. This simulate helps to understand the phenomenon involved the EC system and to prediction the effect of different parameters on EC process without using the apparatus of experimental and this helps to save time and equipment consumption. In this study the CFD modeling used to simulate the batch EC process to identify the optimum operational situations which gave a maximum lead ions removing. The following facts could be concluded from the current investigation:

1. The CFD model was successfully simulate the EC process and its results show how the voltage, current, velocity and concentration distribute in the EC reactor.
2. The batch EC method was impactively utilized to eliminate lead ions from aqueous solution, as predicted by the theoretical model. The impactiveness of lead removing has been discovered to be affected by the initial lead amount, applied voltage, and rotational anode speed.

3. The best conditions for wastewater treatment (that contains lead ions) in the range of utilized parameters were initial amount = 200ppm, voltage = 5 volt and rotational anode speed= 100 within 20min operating time.

4. The impact of rotational velocity on lead ion removing had also been investigated, and the findings confirmed that when the rotational speed is extremely high 150rpm, the removing efficacy decreases since flock instability.

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