Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2015.1015308

56 (2015) 2672–2681
December

Taylor & Francis
Taylor & Francis Group

Optimization of the electrocoagulation process for the removal of lead from water using aluminium as electrode material

Wided Bouguerra[a,*], Afef Barhoumi[b], Nesrine Ibrahim[b], Khaled Brahmi[a], Limam Aloui[b], Béchir Hamrouni[a]

[a]*Faculté des Sciences de Tunis, Département de Chimie, U.R. Traitement et Dessalement des Eaux, Manar II, Tunis 2092, Tunisia, Tel./Fax: +216 71871282; email: bg_wided@yahoo.fr (W. Bouguerra)*
[b]*Faculté des Sciences de Gafsa, Département de Chimie, U.R. Matériaux, Environnement et Energie, Campus Universitaire Sidi Ahmed Zarroug, Gafsa 2112, Tunisia*

Received 24 February 2014; Accepted 29 January 2015

ABSTRACT

Lead is a priority substance in the framework of the European water policy (Water Framework Directive 2000/60/EC and Water Environmental Quality Standards Directive 2008/105/EC), since it presents a significant risk to biota and humans, given its persistence, toxicity and bioaccumulation characteristics. This study is devoted to focus on lead removal by means of electro coagulation (EC). The performance of EC process with aluminium electrodes for removal of lead on laboratory electrochemical cell was studied. The effects of various parameters such as electrochemical treatment time, solution pH, current density (*J*), electrolyte concentration and electrical energy consumption on the percentage of lead removal were investigated. The optimum conditions for EC process were identified as pH 5, current density of 2.67 mA cm^{-2} and electrolyte concentration of 0.5 g L^{-1}. Effect of EC reactor design parameters such as the surface-area-to-volume ratio and the distance between electrodes were investigated. The obtained experimental results showed that optimal lead removal was achieved with distance between electrodes of 0.5 cm and surface-area-to-volume ratio (S/V) of 11.2 m^{-1}. Under optimal conditions with 30 min treatment, the lead removal efficiency was about 99%. The optimal operating conditions can achieve efficient removal in a relatively short reaction time and low energy consumption. In the light of these results, EC could be regarded as a potential technique for the treatment of industrial wastewater containing lead.

Keywords: Electro coagulation; Lead removal; Aluminium electrodes; Chemical parameters; Reactor design parameters; Water treatment

---

*Corresponding author.

Presented at the 4th Maghreb Conference on Desalination and Water Treatment (CMTDE 2013)
15–18 December 2013, Hammamet, Tunisia

1944-3994/1944-3986 © 2015 Balaban Desalination Publications. All rights reserved.

1. Introduction

Heavy metal pollution has become one of the most serious environmental issues [1]. The treatment of heavy metals is of special concern due to their toxicity and persistence in the environment. Heavy metals are discharged from a variety of sources and can be readily oxidized into ions when dissolved in water. Among the heavy metals, lead (Pb), cadmium (Cd), mercury (Hg), nickel (Ni), copper (Cu) and zinc (Zn) are the most hazardous [2]. As one of the heavy metals, lead is used in some industries such as battery, paint, pigments, ammunition, petrol, cable, alloy, steel, plastics and glass [3]. Lead is a priority substance in the framework of the European water policy (Water Framework Directive 2000/60/EC [4] and Water Environmental Quality Standards Directive 2008/105/EC [5]), because it presents a substantial risk to biota and humans, given its persistence, toxicity and bioaccumulation characteristics [6]. Lead ranks first in the list of prioritized hazardous substances issued by the Agency for Toxic Substances and Disease of the United States [3,7]. Once lead is taken into the human body, it causes severe damages to the kidney, nervous system, reproductive system, liver and brain. Long time exposure to lead can induce sterility, abortion, stillbirths and neonatal deaths [3,8]. Therefore, lead-contaminated wastewater has to be treated imperatively before being discharged into the sewage system or into the aquatic environment. Several methods were used to remove heavy metals and particularly lead [9–14].

Lead was efficiently removed by means of adsorption [15–21], vacuum distillation [22], UV/TiO_2/H_2O_2 process [23] and combined nanomembrane technology [24]. Conversely, adsorption has been recognized as an effective and economic method for low-concentration heavy metal wastewater treatment and the major drawback of membrane filtration technology is the high power consumption, and the restoration of the membranes [25].

Alternatively, electrochemical lead contaminated wastewater treatment techniques are regarded as rapid and well controlled that require less chemicals, afford good reduction yields and produce a reduced amount of sludge [25].

EC using sacrificial anode, generally made of aluminium or iron [26,27], is known as a reliable and mainly cost-effective wastewater treatment process [28,29]. It is characterized by simple and easy operated equipment, short operation time, none or negligible amount of chemicals and low sludge production. The flocs formed by EC are relatively large and contain less bound water. They are also more stable [29–31]. EC has been applied successfully to treat potable [32] and various wastewaters [33–41]. The effect of EC electrochemical parameters on the treatment of wastewater charged with heavy metals has received an eminent consideration in the existing literature dealing with EC technology [26,42–49]. EC using steel and aluminium electrodes [26,48] has been evaluated as a treatment process for lead removal. Nevertheless, removal efficiency at different conditions in various times and energy consumption estimation have not been reported.

The aim of this work was to throw more light on the removal process of lead ions from an aqueous solution by using EC. Aluminium was used as electrode materials due to the high coagulation efficiency of Al^{3+} [50]. This study was designed to investigate the effect of some chemical and electrochemical variables in order to improve as much as possible lead removal and significantly reduce the cost of EC process as well. The effect of pH, current density, treatment time and supporting electrolyte on EC feasibility was investigated. Moreover, EC reactor design parameter such as the surface-area-to-volume ratio and the distance between electrodes were studied in detail. Optimization of both electrochemical and reactor design parameter using aluminium electrodes was carried out in term of process performances and energy consumption. Eventually, this research will assess the suitability of EC for industrial applications on the treatment of wastewater containing lead.

2. Materials and methods

2.1. Electrocoagulation tests

The electrolytic cell used to conduct the experiments consists of a glass beaker. Electrocoagulation experiments were carried out using two parallel rectangular aluminium plates ($25 \times 8 \times 0.2$ cm) and 1,000 mL of electrolyte. Both sides of electrode surface area (8×7 cm) were effective corresponding to active electrode surface $S_a = 112$ cm^2. In order to avoid a passivation film, these electrodes were cleaned before use by treating them with NaOH and HCl aqueous solutions. A gentle agitation was made using a magnetic mixer. The applied current density was maintained at the ranges of 0 to 3 A and 0 to 30 V using a regulated direct current (DC) AFX 2930 SB DC power supply, and the voltage cell was continuously recorded. The conductivity and pH of the solution were measured during the experiments using a conductivity meter Jenway 4510 (Ω Metrohm) and a pH meter pH Cyber Scan 510 (WDW, Germany), respectively. The current intensity between the electrodes and the resulting voltage was controlled using the generator.

All solutions were prepared from analytical grade reagents and used without any further purification. Deionized water was used in all the experimental runs. The lead stock solution is prepared by dissolving a suitable amount of $Pb(NO_3)_2$. The initial pH of the solutions was adjusted by adding either HNO_3 (0.1 M) or NaOH (0.1 M). All experiments were performed at around 25˚C and duplicated. To follow the progress of the treatment, aliquots were periodically taken then filtered and acidified for analysis. The experimental error was around 5%.

A schematic diagram of electrochemical cell is shown in Fig. 1.

2.2. Analytical method

The lead concentration was determined by atomic absorption spectroscopy with flame (AASF) method. This method was the most feasible one and could be adequately adapted to higher concentration of the measurement of more than 30 elements [51], the residual concentration of aqueous Pb(II) was determined by atomic absorption spectrometry (Analytik Jena Nova 400) after nitric acidification and suitable dilution of samples. The Mohr method was used to determine the chlorine concentration [51].

To evaluate the experimental results statistically, several tests were done to calculate the linearity, fidelity (reproducibility and repeatability) and limits of both instrumental method detection and quantification. The result of the experimental statistical calculation for the atomic absorption analytical method showed that both repeatability and reproducibility coefficient's variations are less than 5%. The method for lead determination by AASF without background correction is a valuable method with a detection limit of $0.0441783\ \mathrm{mg\,L^{-1}}$ and quantification limit of $0.147261\ \mathrm{mg\,L^{-1}}$.

The process performance was measured in terms of removal efficiency (% removal). The mathematical expression for their calculation was:

$$\text{removal} = \frac{(C_i - C_f)}{C_i} \times 100 \tag{1}$$

where C_i = initial lead concentration ($\mathrm{mg\,L^{-1}}$) and C_f = residual lead concentration ($\mathrm{mg\,L^{-1}}$).

3. Results and discussion

3.1. Effect of pH on lead removal

As it has been well recognized, the pH of the treated medium has an important role in electro coagulation process. Both the nature and the efficiency of the involved electrochemical and chemical reactions are intimately correlated to the pH level in the system [44].

In order to examine the effect of the initial pH of the solution on initial lead concentration, different pH values ranging from 1 to 10 were retained and the initial lead concentration was measured after pH adjustment. Fig. 2 shows that for pH up to 5, lead concentration decreases sharply until a minimum value.

This decrease could be attributed to lead precipitation as $PbCO_3$, $Pb_3\,(CO3)_2\,(OH)_2$ and $Pb\,(OH)_2$. Indeed, for an initial pH up to 5, the appearance of a precipitate was observed in the solution. Subsequently, optimization

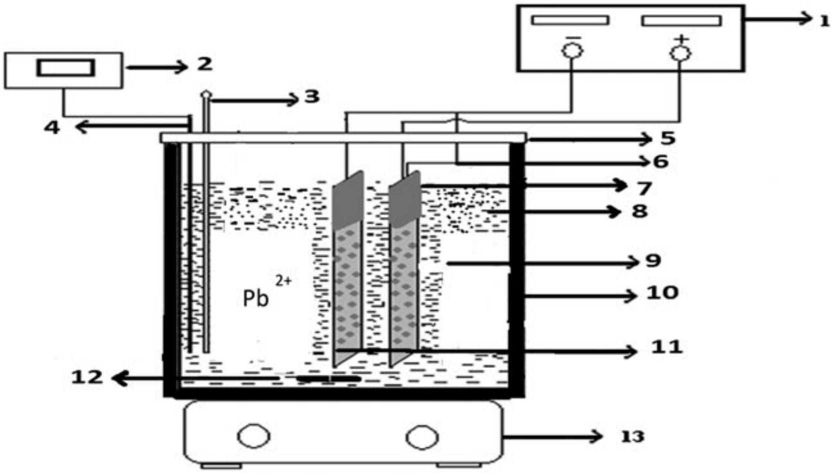

Fig. 1. Laboratory scale cell assembly: 1—DC power supply; 2—pH meter; 3—thermostat; 4—pH sensor; 5—support of electrochemical cell; 6—conductive cable; 7—anode; 8—flocculent; 9—electrolyte solution; 10—baker glass (Pyrex); 11—cathodes; and 12—magnet bar.

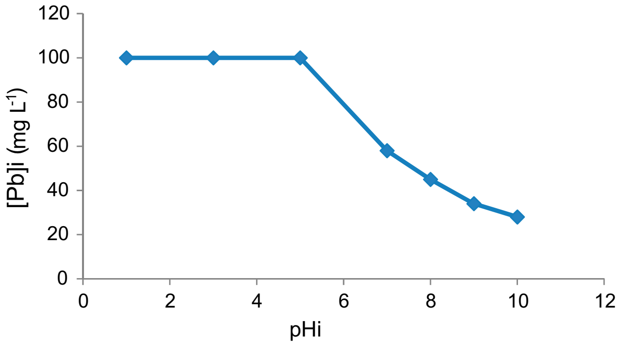

Fig. 2. Evolution of initial lead concentration with initial pH ([NaCl] = 3 g L^{-1}, $t = 0$ min and stirring speed = 300 rpm).

of pH for lead removal by electrocoagulation was done by carrying out a series of experiments with an initial pH varying in the range of 1 to 5 for an initial Pb(II) concentration of 100 mg L^{-1}, a current density of 2.67 mA cm^{-2}, electrolysis time of 60 min and an electrolyte concentration of 3 g L^{-1}.

The effect of pH on the removal of Pb(II) by electrocoagulation is shown in Fig. 3.

Fig. 3 illustrates the variation of lead removal vs. electrolysis time for aluminium electrodes at initial pH values: 1, 3 and 5. As shown, lead was well removed for initial pH of 3 and 5. The highest lead removal speed was obtained at pH 5 and the lowest at pH 1.

The percentage of lead removal increased with increase in pH. This result could be explained by the increase in the quantity of the coagulant $AlOH_3$. Indeed, at pH 5.0–8.5 interval, the dominant Al(III) species is in the form of $Al(OH)_3(s)$ [52]. The percentage of lead removal could be high if the electrocoagulation time was long enough even in the low pH conditions. In fact, the pH of the solution increased through the electrolysis (Fig. 4) making it possible to reach the optimal pH interval for $Al(OH)_3(s)$ formation. Lead removal efficiency increases as the ratio of the coagulant amount to Pb(II) amount increases. The maximum removal of Pb(II) was obtained at pH 5. The initial Pb(II) concentration of 100 mg L^{-1} was significantly reduced with a removal efficiency of 99% after 30 min of electrolysis time.

The use of soluble anodes causes a change in the pH of the solution during electrocoagulation. The evolution of pH during electrolysis depends on both the initial pH and buffer capacity due to the production and consumption of OH^- ions during the electro coagulation. Furthermore, the electrolysis with aluminium electrodes acted as pH neutralization [29].

As shown in Fig. 4, under all the pH conditions tested, the pH increases as the reaction time increased to reach stable values within 30 min of electrocoagulation. The pH variation currently observed is for the most part attributed to hydrogen evolution at the cathode:

$$H_2O + 1\ e^- \leftrightarrow 1/2H_{2(g)} + OH^- \tag{2}$$

This pH change can be also explained by the transfer of CO_2 which is oversaturated in acidic aqueous electrolyte and can release from the medium owning to H_2 bubble disturbance resulting in a pH increase [40].

As shown in Fig. 4, for initial pH of solution slightly acidic (pHi 3 and 5), lead removal by precipitation could be supported by the lower pH increase during EC indicating further OH^- consumption by Pb (II). It is to be noted that the same pH evolution was observed by Kim et al. when considering Pb(II) and

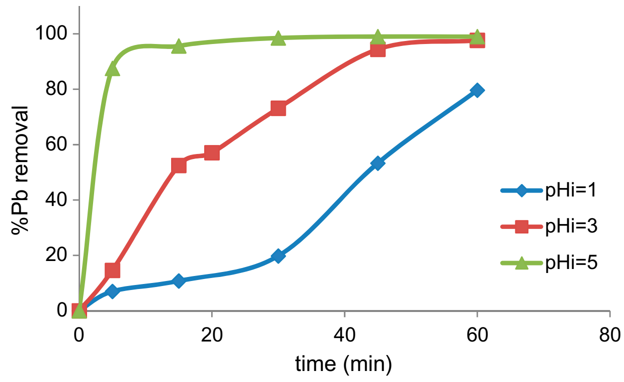

Fig. 3. Effect of initial pH on the removal of Pb(II) by electrocoagulation ($d = 1.5$ cm, [NaCl] = 3 g L^{-1}, $J = 2.67$ mA cm^{-2} and stirring speed = 300 rpm).

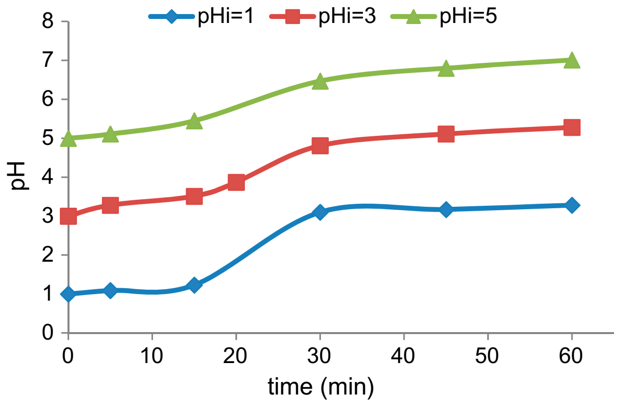

Fig. 4. Evolution of pH during electrolysis ($d = 1.5$ cm, [NaCl] = 3 g L^{-1}, $J = 2.67$ mA cm^{-2} and stirring speed = 300 rpm).

Cu(II) removal by electrocoagulation [53]. Electrolysis with aluminium electrodes offers to EC process significant potential for removing soluble heavy metals by electroprecipitation which acts synergistically with usual coagulation phenomena to remove pollutants from effluents containing ionic metallic species [29].

3.2. Effect of electrolysis time on lead removal

To highlight the effect of the electrolysis time on lead removal, a series of electrocoagulation tests were performed by tracking the residual lead concentration for different electrolysis time spans for initial pH of 5, initial Pb(II) concentration of 100 mg L^{-1}, constant current density of 2.67 mA cm^{-2}, electrolyte concentration of 3 g L^{-1} and distance between electrodes of 1.5 cm.

Fig. 5 shows the variation of the residual lead concentrations and pH vs. electrolysis time.

According to Fig. 5, residual lead concentration decreased significantly during the first stage of electrolysis (5 min) afterwards, it reached an unvarying value. Simultaneously, during the initial period of electrolysis, pH variation was not significant and the alkalinity produced during electrolysis was not sufficient to increase pH of the solution. This could be attributed to lead carbonate and/or hydroxide precipitation. Furthermore, the pH during the second stage of Pb(II) removal (20–60 min) was higher than 5.5 so the dominant Al(III) species would be in the form of $Al(OH)_3(s)$ [52]. As shown in Fig. 5, within 15 min and 30 min of electrolysis lead concentration decreased to 4.39 mg L^{-1} and 1 mg L^{-1} corresponding to removal of 95.61 and 99.00%, respectively. Similar Pb(II) removal efficiency has been reported with an Fe electrode [53].

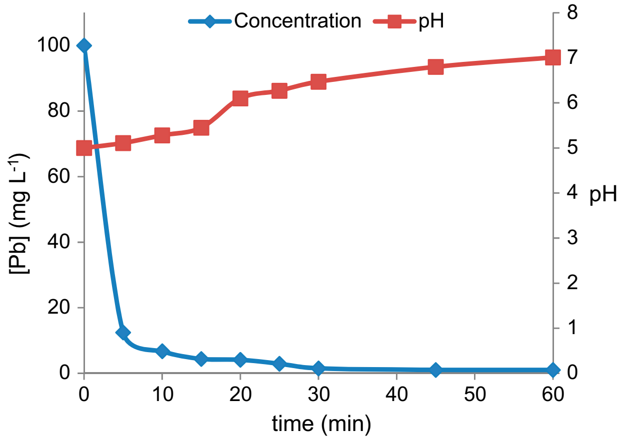

Fig. 5. Effect of electrolysis time on residual lead concentrations and pH variation ($d = 1.5$ cm, [NaCl] = 3 g L^{-1}, $J = 2.67$ mA cm^{-2} and stirring speed = 300 rpm).

Various electrochemical and chemical reactions were involved during EC. The determination of residual chloride concentration reveals a decrease of 133.125 mg L^{-1}. In fact, when some anions such as Cl^- and SO_4^{2-} are present in the electrolyte, they can exchange partly with OH^- in $Al(OH)_3$ to free OH^-, which also causes a pH increase [40]. Besides, some authors [53] suggested that chloride from background electrolyte contributes to heavy metal precipitation, as evidenced by the existence of chlorinated metals in the solid phase. The existence of Pb(OH)Cl has been also identified. The encapsulation of Pb(II) has also been evidenced indicating that precipitation plays an important role in Pb(II) removal [53]. The lower pH variation and the decrease of chloride concentration during EC would confirm that process efficiency is due to both adsorption and precipitation of lead ions.

3.3. Effect of supporting electrolyte on lead removal

The addition of electrolyte promotes the electrical transport ensuring better chemical dissolution of aluminium. The presence of the chloride ions in solution has been reported to decrease passivation of the aluminium surface and thereby increase the efficiency of electrocoagulation processes [54].

In order to examine the effect of chloride ion concentration on lead removal, different doses of sodium chloride (NaCl) were retained. Electrolyse experiments were carried out at 2.67 mA cm^{-2} after pH adjustment of the electrolytes at 5. During the course of electrolysis, residual lead concentrations were determined. Table 1 and Fig. 6 shows, respectively, the evolution of lead removal during electrocoagulation tests and the variation of lead removal as a function of NaCl doses.

The results reveal that whatever the supporting electrolyte concentration considered, lead removal is effective (over 90%) after 30 min of electrocoagulation.

Table 1
Evolution of lead removal as a function of electrolysis time for different electrolyte (NaCl) doses (pHi = 5, $J = 2.67$ mA cm^{-2} and $d = 1.5$ cm)

% Pb removal	Time (min)				
[NaCl] (g L^{-1})	0	5	10	15	30
0.15	0.00	64.05	81.03	89.31	91.37
0.30	0.00	75.21	90.53	91.27	95.64
0.50	0.00	84.87	91.26	96.55	96.61
1.00	0.00	85.50	91.75	97.82	98.77
1.50	0.00	92.00	94.50	96.47	99.84
3.00	0.00	91.52	96.57	98.19	99.99

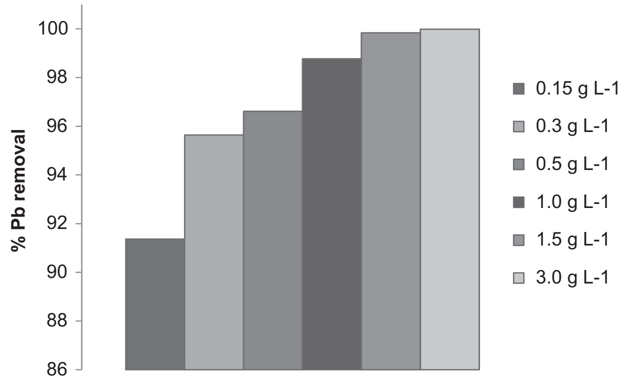

Fig. 6. Variation of lead removal as a function of NaCl concentration (pHi = 5, $J = 2.67$ mA cm^{-2}, $d = 1.5$ cm and $t = 30$ min).

On the other hand, if the percentages of removal with different electrolyte concentrations w ithin a certain treatment time were compared, electrolyte concentration was found to have significant effects on lead removal speed. For example, with 10 min electrolysis treatment, the highest removal was obtained with the electrolyte concentration 3.00 g L^{-1}, and the percentage of lead removal was 96.57%.

Increasing the concentration of NaCl and consequently the conductivity of the solution resulted in a slight enhancement of lead removal efficiency by virtue of better chemical dissolution. This result has its roots in the decrease of passivation of the aluminium surface due to the presence of the chloride ions in solution [29].

The effect of electrolyte concentration on the energy consumption was brought to light by recording, electrolysis voltage during each experiment. The electrocoagulation electric energy consumption W (kWh m^{-3}) is a parameter of great magnitude that can be calculated as:

$$W = \frac{U.I.t}{V} \tag{3}$$

where U is voltage cell (V), I is current (A), t is the time of electrolysis (h) and v is the volume (m^3) of the solution.

Table 2 shows the effect of NaCl concentration on electrolysis voltage.

As NaCl concentration increases from 0.15 to 3.00 g L^{-1}, the cell voltage decreases rapidly from 14.9 to 3 V. The decrease of electrolysis potential difference which would be beneficial in terms of energy consumption can be related to ohmic potential drop of the solution and/or to a decrease of the anode over potential [29]. But, there was an overconsumption of aluminium electrodes due to corrosion of elevated electrolyte concentration. Thus, the optimum concentration of NaCl is around about 0.5 g L^{-1}. It results in nominal energy consumption (less than 1 kW h m^{-3}) and an elimination of lead exceeding 96%.

Table 2
Variation of energy consumption as a function of electrolyte concentration (pHi = 5, $J = 2.67$ mA cm^{-2}, $d = 1.5$ cm and $t = 30$ min)

[NaCl] (g L^{-1})	0.15	0.30	0.50	1.00	1.50	3.00
U (V)	14.90	9.70	6.60	4.50	3.60	3.00
W (kW h m^{-3})	1.96	1.23	0.84	0.62	0.53	0.42
% Pb removed	91.31	95.64	96.61	98.77	99.84	99.90

3.4. Effect of current density on lead removal

The current density is an effective parameter that controls the reaction rate in the electrochemical systems and it determines the amount of Al^{3+} ions released by the anode.

The effect of current density on lead removal was studied and the voltage U between the electrodes was recorded during the electrocoagulation tests. A series of tests were carried out for an initial Pb(II) concentration of 100 mg L^{-1}, an electrolyte concentration of 0.5 g L^{-1} and a current density ranging from 0.89 to 4.46 mA cm^{-2}. The results are displayed in Table 3 and Fig. 7.

Table 3 depicts the evolution of electrolysis voltage with current density. As shown, the electrolysis voltage increases with current density. Consequently, an increase of current density leads to an increase of power requirement.

This figure clearly shows that a significant increase in Pb(II) removal occurred just at the first stage of

Table 3
Variation of the electrolysis voltage between electrodes as a function of time for different current densities (pH = 5, [NaCl] = 0.5 g L^{-1} and $d = 1.5$ cm)

ddp (V)	Time (min)					
J (mA cm^{-2})	I(A)	0	5	10	15	30
0.89	0 .1	0.0	2.7	2.7	2.7	2.7
1.78	0.2	0.0	5.3	5.3	5.3	5.3
2.67	0.3	0.0	6.6	6.6	6.6	6.6
3.57	0.4	0.0	9.3	9.3	9.3	9.3
4.46	0.5	0.0	12.1	12.2	12.2	12.3

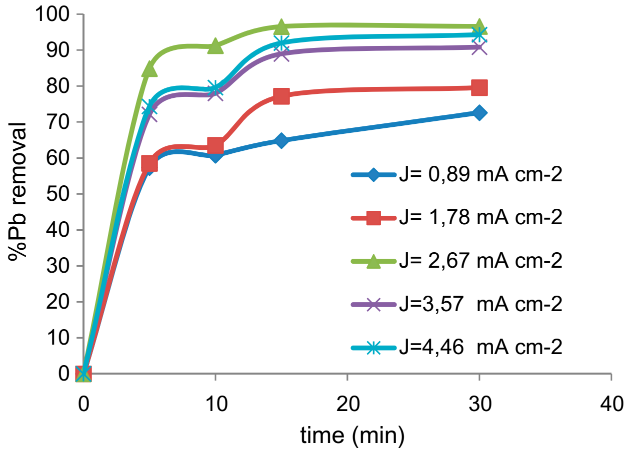

Fig. 7. Variation of lead removal percentage as a function of electrocoagulation time for different current densities (pHi = 5, NaCl = 0.5 g L^{-1} and d = 1.5 cm).

electrolysis for all the applied current densities and became slower afterwards. For current densities under 3.57 mA cm^{-2}, the required electrocoagulation time for lead removal decreased as the current density increases. This could be explained by the fact that at higher voltage the quantity of aluminium oxidized increased resulting in a higher quantity of coagulant and a better elimination. Similar results have been reported for Cu [26,48], Cd [26] and lead [3,26,48] removal by electrochemical processes using either aluminium or steel electrodes. Otherwise, Kim et al. [53] reported that Pb(II) is less sensitive to the floc amount than Cu(II) and Cd(II).

After 30 min of electrolysis, the removal efficiencies reached 72.61, 79.57 and 96.61% at the current densities of 0.89, 1.78 and 2.67 mA cm^{-2} respectively. Also, the results show that relatively high current densities (3.57 and 4.46 mA cm^{-2}) induce a slight decrease of lead removal but the removal efficiencies still over 90%.

The fact of rising the current density can cause fatal effects on the treatment efficiency of electrocoagulation process. Therefore, the current density determines not only the dose of the coagulant but also the rate of production of bubbles and the growth of the flocs [55]. Moreover, current density increase is accompanied by high energy consumption which is disadvantageous.

The effect of current densities on lead removal and energy consumption can be seen from Table 4.

To point out the current density and the optimum electrocoagulation time, conciliation between economically suitable energy and removal efficiency was required. Table 4 recapitulates the removal efficiencies and energy consumption as a function of time for the considered electrocoagulation current densities (2.67, 3.57, and 4.46 mA cm^{-2}). A current density of 2.67 mA cm^{-2} was effective enough to remove over 96% of lead at an electrolysis time of 15 min with an energy consumption of only 0.42 kW h m^{-3}.

3.5. *Effect of inter-electrode distance*

To highlight the effect of the inter-electrodes distance, various electrocoagulation tests were performed by varying the distances between anode and cathode in the range from 0.5 to 2 cm and keeping constant the initial Pb(II) concentration at 100 mg L^{-1}, initial pH at 5, electrolyte concentration at 0.5 g L^{-1} and current density at 2.67 mA cm^{-2}. The effect of inter-electrode

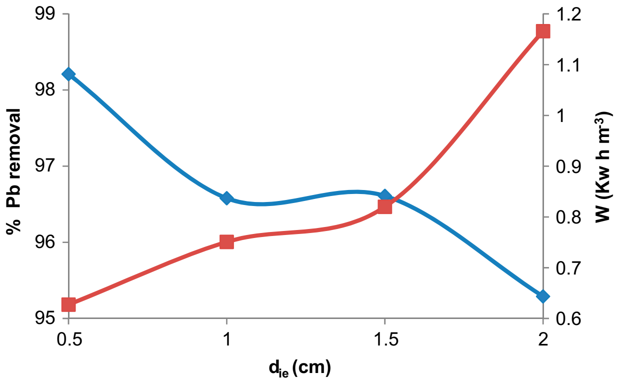

Fig. 8. Effect of inter-electrode distance on lead removal efficiency and energy consumption (pHi = 5, NaCl = 0.5 g L^{-1}, J = 2.67 mA cm^{-2} and t = 30 min).

Table 4
Variation of energy consumption as a function of current density (pHi = 5, [NaCl] = 0.5 mg L^{-1} and d = 1.5 cm)

J	2.67 mA cm^{-2}				3.57 mA cm^{-2}				4.46 mA cm^{-2}			
t (min)	5	10	15	30	5	10	15	30	5	10	15	30
% Pb removed	84.87	91.26	96.55	96.61	72.15	77.99	88.96	90.83	74.34	79.59	91.99	94.30
W (kW h m^{-3})	0.15	0.29	0.42	0.82	0.28	0.56	0.84	1.69	0.45	0.92	1.38	2.79

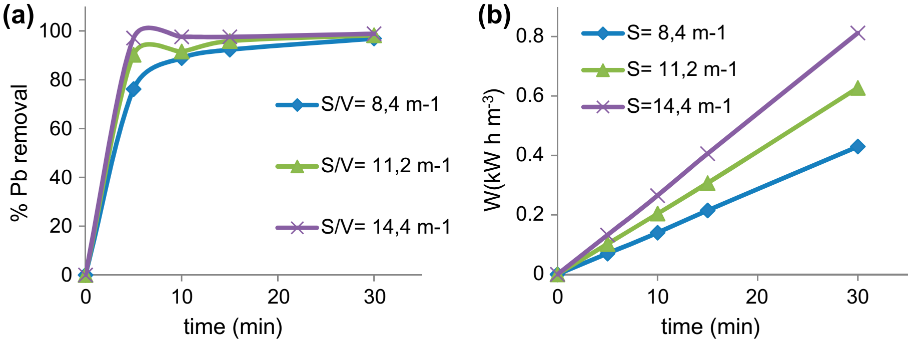

Fig. 9. Variations of lead removal percentage (a) and energy consumption (b) as a function of electrocoagulation time for different S/V ratios (pHi = 5, NaCl = 0.5 g L^{-1}, J = 2.67 mA cm^{-2} and d = 0.5 cm).

distance on removal efficiency and energy consumption is shown in Fig. 8.

Inter-electrode distance has an effect on the amount of electrical energy introduced into the system to generate an electric field and induce motion of ions [56]. The best lead removal percentage was achieved for the inter-electrode distance value of 0.5 cm. Within 30 min of electrolysis time, decreasing the inter-electrode distance from 2 to 0.5 cm resulted in a slight increase in lead removal efficiency from 95.29 to 98.21% and a reduction in energy consumption from 1.166 to 0.627 KWh m^{-3}. When the inter-electrode distance increased, the ohmic loss in relation to the anode and cathode over voltages and the resistance to mass transfer became larger; the kinetics of both charge transfer and the aluminium oxidation was slowed down [57]. So, further experiments were carried out at inter-electrode distance of 0.5 cm. Similar results were reported for boron removal by electrocoagulation using Zn anode [56].

3.6. Effect of the surface-area-to-volume ratio

The surface-area-to-volume ratio S/V is the ratio of the active surface area to the volume of the treated solution. It has been reported that the increase in the ratio S/V results in the reduction of the current density consumption [58]. The effect of S/V ratio on lead removal efficiency was studied by covering and deactivating either a part or all of the face of each electrode using durable water-resistant cellotape. A series of electrocoagulation tests was carried out for initial Pb(II) concentration of 100 mg L^{-1}, electrolyte concentration of 0.5 g L^{-1}, current density of 2.67 mA cm^{-2} and inter-electrode distance of 0.5 cm. Fig. 9 presents the removal efficiency of lead and energy consumption vs. reaction time at various S/V ratios.

For all studied values of ratio S/V, removal efficiency over 96% was achieved for an electrolysis time of 30 min but there was a huge difference in terms of energy consumption. The energy consumption varied linearly as the electrocoagulation time progressed. The increase in the S/V resulted in a rapid increase in energy consumption. This could be explained by the decrease of solution conductivity caused by lead precipitation. Increasing the S/V ratio from 8.4 to 14.4 m^{-1} resulted in an enhancement of lead removal efficiency from 76.17 to 97.08% and from 96.88 to 98.85% for electrolysis time of 5 and 30 min, respectively. Thus, increasing S/V ratio could be beneficial in term of treatment time. S/V ratio of 11.2 m^{-1} achieves removal efficiency of about 97% and low energy consumption within 15 min of electrolysis. Accordingly, 11.2 m was chosen as an optimum value for the lead removal.

4. Conclusion

In this study, the electrochemical removal of lead from aqueous media using aluminium as electrode materials has been investigated. The effects of electrochemical and reactor design parameters on lead removal efficiency in various times were examined. Energy consumption estimation was explored. The results obtained at varying pH conditions showed that removal efficiencies were significantly affected by initial pH. The highest removal of lead was obtained at pH 5 for an electrolysis time of 30 min. It seems that the removal efficiency obtained is the result of simultaneous adsorption on $Al(OH)_3$ precipitate and precipitation of lead ions. Increasing the electrolyte concentration resulted in an enhancement of lead removal. An increase of the current density notably reduced the electrocoagulation required time for the treatment. The optimum electrochemical operating parameters are electrocoagulation time of 15 min, current density of 2.67 mA cm^{-2}, electrolyte concentration of 0.5 g L^{-1} and pH 5. Inter-electrode distance decrease would be beneficial in terms of removal efficiency and energy consumption. Increasing the *S*/*V* ratio resulted in enhancement of lead removal efficiency and treatment time reduction. The results showed that inter-electrode distance of 0.5 cm and surface-area-to-volume ratio of 11.2 m^{-1} achieved removal efficiency of 97% and energy consumption of 0.307 kW h m^{-3} within 15 min of electrolysis. Under optimal conditions with 30 min treatment, the lead removal efficiency reached 99%. In view of that, electrocoagulation could be applied as a cost-effective process to remove lead from wastewater.

References

[1] H. Bessbousse, T. Rhlalou, J.-F. Verchère, L. Lebrun, Removal of heavy metal from aqueous solutions by filtration with a novel complexing membrane containing poly(ethyleneimine) in a poly(vinyl alcohol) matrix, J. Membr. Sci. 307 (2008) 249–259.

[2] M. Hofman, R. Pietrzak, Copper ions removal from liquid phase b polyethersulfone (PES) membranes functionalized by introduction of carbonaceous materials, Chem. Eng. J. 215–216 (2013) 216–221.

[3] Y. Liu, J. Yan, D. Yuan, Q. Li, X. Wu, The study of lead removal from aqueous solution using an electrochemical method with a stainless steel net electrode coated with single wall carbon nanotubes, Chem. Eng. J. 218 (2013) 81–88.

[4] Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 Establishing a Framework for Community Action in the Field of Water Policy.

[5] Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on Environmental Quality Standards in the Field of Water Policy.

[6] M. Bernardo, S. Mendes, N. Lapa, M. Gonçalves, B. Mendes, F. Pinto, H. Lopes, I. Fonseca, Removal of lead (Pb^{2+}) from aqueous medium by using chars from co-pyrolysis, J. Colloid Interface Sci. 409 (2013) 158–165.

[7] ATSDR, Toxicological Profile for Lead, United States Department of Health and Human Services, Atlanta, GA, 2007.

[8] R.A. Goyer, I.J. Chisolon, Lead: In Metallic Contamination and Human Health, first ed., Academic Press, New York, NY, 1972.

[9] H.A. Aziz, M.N. Adlan, K.S. Ariffin, Heavy metals (Cd, Pb, Zn, Ni, Cu and Cr(III)) removal from water in Malaysia: Post treatment by high quality limestone, Bioresour. Technol. 99 (2008) 1578–1583.

[10] J. Goel, K. Kadirvelu, C. Rajagopal, V.K. Garg, Removal of lead(II) by adsorption using treated granular activated carbon: Batch and column studies, J. Hazard. Mater. 125 (2005) 211–220.

[11] A. Gundogdu, D. Ozdes, C. Duran, V.N. Bulut, M. Soylak, H.B. Senturk, Biosorption of Pb(II) ions from aqueous solution by pine bark (Pinus brutia Ten.), Chem. Eng. J. 153 (2009) 62–69.

[12] S. Ahmed, S. Chughtai, M.A. Keane, The removal of cadmium and lead from aqueous solution by ion exchange with Na Y zeolite, Sep. Purif. Technol. 13 (1998) 57–64.

[13] M. Soylak, R.S. Cay, Separation/preconcentration of silver(I) and lead(II) in environmental samples on cellulose nitrate membrane filter prior to their flame atomic absorption spectrometric determinations, J. Hazard. Mater. 146 (2007) 142–147.

[14] T.N. Shilimkar, M.A. Anuse, Rapid extraction of lead (II) from succinate media with n-octylaniline in toluene, Sep. Purif. Technol. 26 (2002) 185–193.

[15] V.K. Gupta, S. Agarwalc, T.A. Saleh, Synthesis and characterization of alumina-coated carbon nanotubes and their application for lead removal, J. Hazard. Mater. 185 (2011) 17–23.

[16] M. Abdel Salam, Coating carbon nanotubes with crystalline manganese dioxide nanoparticles and their application for lead ions removal from model and real water, Colloids Surf., A 419 (2013) 69–79.

[17] M. Jiang, Q. Wang, X. Jin, Z. Chen, Removal of Pb(II) from aqueous solution using modified and unmodified kaolinite clay, J. Hazard. Mater. 170 (2009) 332–339.

[18] R. Rojas, Copper, lead and cadmium removal by Ca Al layered double hydroxides, Appl. Clay Sci. 87 (2014) 254–259.

[19] Y. Deng, Z. Gao, B. Liu, X. Hu, Z. Wei, C. Sun, Selective removal of lead from aqueous solutions by ethylenediamine-modified attapulgite, Chem. Eng. J. 223 (2013) 91–98.

[20] M.A. Martín-Lara, I.L. Rodríguez, G. Blázquez, M. Calero, Factorial experimental design for optimization the removal conditions of lead ions from aqueous solutions by three wastes of the olive-oil production, Desalination 278 (2011) 132–140.

[21] M. Bernardo, S. Mendes, N. Lapa, Ma. Gonçalves, B. Mendes, F. Pinto, H. Lopes, I. Fonseca, Removal of lead (Pb^{2+}) from aqueous medium by using chars from co-pyrolysis, J. Colloid Interface. Sci. 409 (2013) 158–165.

[22] X. Li, Y. Gao, H. Ding, Removing lead from metallic mixture of waste printed circuit boards by vacuum distillation: Factorial design and removal mechanism, Chemosphere 93(4) (2013) 677–682.

[23] M.R. Samarghandi, J. Nouri, S. Nasseri, Efficiency removal of phenol, lead and cadmium by means of UV/TiO_2 /H_2O_2 process, Int. J. Environ. Sci. Technol. 4(1) (2007) 19–25.

[24] I. Hajdu, M. Bodnár, Z. Csikós, S. Wei, L. Daróczi, B. Kovács, Z. Győri, J. Tamás, J. Borbély, Combined nano-membrane technology for removal of lead ions, J. Membr. Sci. 409–410 (2012) 44–45.

[25] F. Fu, Q. Wang, Removal of heavy metal ions from wastewaters: A review, J. Environ. Manage. 92 (2011) 407–418.

[26] C. Escobar, C. Soto-Salazar, M.I. Toral, Optimization of the electro coagulation process for the removal of copper, lead and cadmium in natural waters and simulated wastewater, J. Environ. Manage. 81 (2006) 384–391.

[27] G.H. Chen, Electrochemical technologies in wastewater treatment, Sep. Purif. Technol. 38 (2004) 11–41.

[28] C.Y. Hu, S.L. Lo, W.H. Kuan, Effects of co-existing anions on fluoride removal in electro coagulation (EC)

process using aluminium electrodes, Water Res. 37 (2003) 4513–4523.

[29] G. Mouedhen, M. Feki, M. De Petris Wery, H.F. Ayedi, Behaviour of aluminium electrodes in electro coagulation process, J. Hazard. Mater. 150 (2008) 124–135.

[30] N.S. Abuzaid, A. Bukhari, Z.M. Al-Hamouz, Ground water coagulation using soluble stainless steel electrodes, Adv. Environ. Res. 6 (2002) 325–333.

[31] H. Bergmann, A. Rittel, T. Iourtchouk, K. Schoeps, K. Bouzek, Electrochemical treatment of cooling lubricants, Chem. Eng. Process. 42 (2003) 105–119.

[32] E.A. Vik, D.A. Carlos, A.S. Eikum, E.T. Gjessing, Electrocoagulation of potable water, Water Res. 18 (1984) 1355–1360.

[33] U.B. Og˘u¨tveren, S. Koparal, Electrocoagulation for oil–water emulsion treatment, J. Environ. Sci. Health, A 32 (1997) 2507–2520.

[34] S.J.S. Do, M.L. Chen, Decolorization of dye-containing solutions by electrocoagulation, J. Appl. Electrochem. 24 (1994) 785–790.

[35] A. Gürses, M. Yalçın, C. Doğar, Electrocoagulation of some reactive dyes: A statistical investigation of some electrochemical variables, Waste Manage. Chem. 596 (2006) 7–12.

[36] Ü.B. Öğütveren, N. Gönen, S. Koparal, Removal of dye stuffs from waste water: Electrocoagulation of Acilan Blau using soluble anode, J. Environ. Sci. Health, Part A: Environ. Sci. Eng. A27(5) (1992) 1237–1247.

[37] S.H. Lin, C.F. Peng, Treatment of textile wastewater by electrochemical method, Water Res. 28 (1994) 277–282.

[38] M.F. Pouet, A. Grasmick, Urban wastewater treatment by electrocoagulation and flotation, Water Sci. Technol. 31 (1995) 275–283.

[39] G. Chen, X. Chen, P.L. Yue, Electrocoagulation and electroflotation of restaurant wastewater, J. Environ. Eng. 126 (2000) 858–863.

[40] X. Chen, G. Chen, P.L. Yue, Separation of pollutants from restaurant wastewater by electrocoagulation, Sep. Purif. Technol. 19 (2000) 65–76.

[41] R.S. Bejankiwar, Electrochemical treatment of cigarette industry wastewater: Feasibility study, Water Res. 36 (2002) 4386–4390.

[42] I. Heidmann, W. Calmano, Removal of Zn(II), Cu(II), Ni(II), Ag(I) and Cr(VI) present in aqueous solutions by aluminium electrocoagulation. J. Hazard. Mater. 152 (2008) 934–941.

[43] C.P. Nanseu-Njiki, S.R. Tchamango, P.C. Ngom, A. Darchen, E. Ngameni, Mercury(II) removal from water by electrocoagulation using aluminium and iron electrodes, J. Hazard. Mater. 168 (2009) 1430–1436.

[44] G. Mouedhen, M. Feki, M. De Petris-Wery, H.F. Ayedi, Electrochemical removal of Cr(VI) from aqueous media using iron and aluminium as electrode materials: Towards a better understanding of the involved phenomena, J. Hazard. Mater. 168 (2009) 983–991.

[45] A. Shafaei, M. Rezayee, M. Arami, M. Nikazar, Removal of Mn^{2+} ions from synthetic wastewater by electrocoagulation process, Desalination 260 (2010) 23–28.

[46] P.R. Kumar, S. Chaudhari, K.C. khilar, S.P. Mahajan, Removal of arsenic from water by electrocoagulation, Chemosphere 55 (2004) 1245–1252.

[47] J.R. Parga, D.L. Cocke, J.L. Valenzuela, J.A. Gomes, M. Kesmez, G. Irwin, H. Moreno, M. Weir, Arsenic removal via electrocoagulation from heavy metal contaminated groundwater in La Comarca Lagunera México, J. Hazard. Mater. 124 (2005) 247–254.

[48] W. Bouguerra, A. Barhoumi, B. Hamrouni, L. Aloui, Optimization of the electrocoagulation process for the removal of lead and copper ions from aqueous solutions using aluminium electrodes, Int. J. Eng. Res. Technol. 3(2) (2014) 2917–2922.

[49] M.M.S.G. Eiband, K.C. de, A. Trindade, K. Gama, J. Vieira de Melo, C.A. Martínez-Huitle, S. Ferro, Elimination of Pb through electrocoagulation: Applicability of adsorptive stripping voltammetry for monitoring the lead concentration during its elimination, J. Electroanal. Chem. 717–718 (2014) 213–218.

[50] F. Shen, P. Gao, X. Chen, G. Chen, Electrochemical removal of fluoride ions from industrial wastewater, Chem. Eng. Sci. 58 (2003) 987–993.

[51] J. Rodier, L'analyse physico-chimique de l'eau (physico-chemical analysis of water), ninth ed., Dunod, Paris, 2009, pp. 172–182.

[52] A.E. Yilmaz, R. Boncukcuoğlu, M.M. Kocakerim, M.T. Yilmaz, C. Paluluoğlu, Boron removal from geothermal waters by electrocoagulation, J. Hazard. Mater. 153(1–2) (2008) 146–151.

[53] D. Kim, R.J.S. Palacios, S. Ko, Characterization of sludge generated by electrocoagulation for the removal of heavy metals, Desalin. Water Treat. 52(4–6) (2014) 909–919.

[54] K. Bensadok, S. Benammara, F. Lapicque, G. Nezzal, Electrocoagulation of cutting oil emulsions using aluminium plate electrodes, J. Hazard. Mater. 152(1) (2008) 423–430.

[55] M.Y.A. Mollah, R. Schennach, J.R. Parga, D.L. Cocke, Electrocoagulation (EC)—Science and applications, J. Hazard. Mater. B84 (2001) 29–41.

[56] S. Vasudevan, J. Lakshmi, G. Sozhan, Electrochemically assisted coagulation for the removal of boron from water using zinc anode, Desalination 310 (2012) 122–129.

[57] D. Ghosh, H. Solanki, M.K. Purkait, Removal of Fe(II) from tap water by electrocoagulation technique, J. Hazard. Mater. 155 (2008) 135–143.

[58] D. Belhout, D. Ghernaout, S. Djezzar-Douakh, A. Kellil, Electrocoagulation of a raw water of Ghrib Dam (Algeria) in batch using aluminium and iron electrodes, Desalin. Water Treat. 16 (2010) 1–9.