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Novel method for electrocoagulation removal of lead from simulated wastewater by using concentric tubes electrodes reactor

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ABSTRACT

Heavy metals pollution has become the most effective environmental problem in recent years as a result of releasing hazard materials into the environment. Various kinds of techniques are used for the treatment of these toxic materials such as electrocoagulation process. Electrocoagulation process is an attractive method for the treatment of wastewater because it is considered as rapid and well-controlled process that requires fewer chemicals, provides good reduction and yields and produces less sludge. The objective of the present study is to investigate the effect of electrodes configuration that may affect the efficiency of heavy metal removal from wastewater during a period of time under constant values of other parameters such as the initial concentration of lead metal, the current, the voltage applied, stirring speed, electrodes metal type, active surface area, the distance between electrodes and pH. It has been found that the best removal efficiency occurs when a cathode electrode is concentric between other two anode electrodes.

Keywords: Heavy metals; Wastewater treatment; Electrocoagulation; Electrodes configuration

1. Introduction

Because of the continuous development of industries such as metal plating, mining, fertilizer, batteries, paper and pesticides, etc., heavy metals wastewaters are increasingly being discharged into the environment directly or indirectly. Heavy metals are elements having atomic weights between 63.5 and 200.6 and a specific gravity greater than 5 [1]. In general, organic contaminants are biodegradable, while heavy metals are not. Heavy metals tend to accumulate in living organisms and their ions are known to be toxic or carcinogenic [2]. Copper, nickel, mercury, zinc, cadmium, lead and chromium are the most toxic heavy metals that are present in industrial wastewaters and must be treated to protect the environment [3–5]. In recent years, a variety of techniques are used for heavy metals removal from water and wastewater which include ion exchange, adsorption, chemical precipitation, membrane filtration, flocculation, coagulation, flotation and electrochemical methods [6–8]. Electrocoagulation is a clean electrochemical process, which uses an applied voltage (i.e., electrical current) to remove metals from solution. In addition, electrochemical method is also effective in removing suspended solids, dissolved metals and dyes. The electrocoagulation system is an effective method for the treatment of several kinds of industrial wastewaters, by the fact of various benefits including environmental capability, versatility, energy efficiency, safety, selectivity and cost effectiveness rather than chemical coagulation technique which is not efficient [9]. The contaminants present in wastewater such as lead ions are maintained in solution by electrical charges [10]. When these ions and other charged particles are neutralized with ions of opposite electrical

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charges provided by electrocoagulation system, they become destabilized and precipitate in a stable form. Electrochemical methods are simple, fast, inexpensive, easily operable and eco-friendly in nature [3,11]. Besides, purified water is potable, clear, colorless and odorless with low sludge production. There is no chance of secondary contamination of water in these techniques. Whereas, other treatment methods such as ion exchange, adsorption, membrane filtration and chemical coagulation which may cause a large amount of sludge as in chemical precipitation and chemical coagulation techniques or the serious secondary pollution due to the regeneration of ion-exchange resins and so on for other techniques [5,12]. The electrocoagulation process has the ability to eliminate the drawbacks of the classical treatment techniques to achieve a sustainable and economic treatment of polluted industrial wastewater.

Several parameters affect the efficiency of electrocoagulation process such as initial concentration of the metal, contact time, pH, current density or current, cell voltage, anode–cathode materials (mild steel, stainless steel, steel, titanium, iron, platinum, copper, carbon steel electrodes), batch or continuous, flow rate, inter-electrodes distance, conductivity of solutions, energy consumption and the electrodes configuration. The electrode material and the connection mode of the electrodes play a significant role in the cost analysis of the electrocoagulation process [13].

Many types of electrodes geometries have been considered for laboratory investigations of electrode kinetics. The shape of the electrodes affects the pollutant removal efficiency in the electrocoagulation process.

In electrocoagulation method, water is electrolyzed in a parallel reaction when a voltage potential is applied from a DC power supply [14]. Oxidation and reduction operations occur on anode and cathode electrodes, respectively, as explained below:

- At the anode electrode with metal M:

$$M_{(S)} \rightarrow M^{+n}{}_{(aq)} + ne^- \quad (1)$$

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \quad (2)$$

- At the cathode electrode:

$$M^{+n}{}_{(aq)} + ne^- \rightarrow M_{(S)} \quad (3)$$

$$2H_2O + 2e^- \rightarrow H_{2(g)} + 2OH^-{}_{(aq)} \quad (4)$$

In the present study, aluminum electrodes were used in two types of configuration to examine the effects of that novel design under specified parameters on the contaminant removal efficiency.

2. Experimental work

2.1. Apparatus

The schematic of electrocoagulation cell shown in Figs. 1 and 2, which consist of three concentric aluminum tubes with different diameters and thicknesses as given in Table 1 with an active area of approximately 285 cm^2. Other tools as follows:

1. Digital DC power supply (SYADGONG company-305D, China); 0–30 volt and 0–5 A.

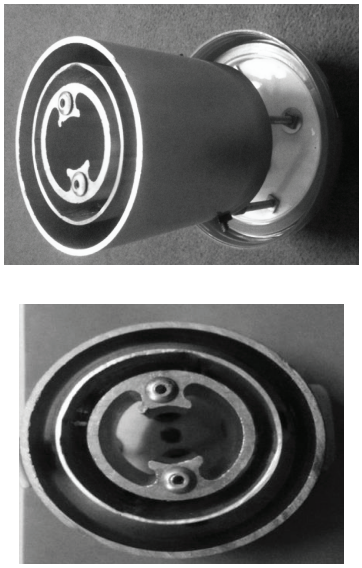

Fig. 1. Concentric electrodes.

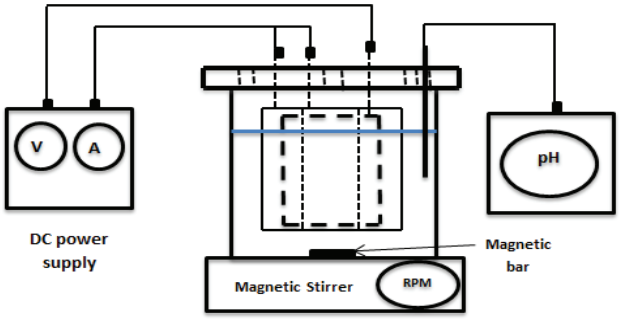

Fig. 2. The schematic of electrocoagulation reactor system.

Table 1
Description of aluminum electrodes

	Height (cm)	Wet height (cm)	Electrode thick (cm)	Outer diameter (cm)	Inner diameter (cm)	Distance in between (cm)
Outer electrode	9.7	4	0.2	7.5	7.3	1.6
Mid electrode	8.5	4	0.15	5.7	5.55	1.55
Inner electrode	7.1	4	0.3	4	3.7	

2. Digital balance (500 g × 0.01 g) (PROF company, China).
3. Magnetic stirrer (ALFA company, Iran: HS-860); 0–1,000 rpm.
4. pH meter (ATC company, China).
5. Digital timer (Sewan company, China).
6. Aluminum tubes with different diameters and thicknesses.

Tables 1 and 2 explain the description and configuration of the concentric electrodes that is made of aluminum tubes which consist of the height of electrodes, wet height, outer and inner diameters, distance in between and electrodes thicknesses individually.

The batch electrocoagulator is made of plexiglass with the volume of 1,000 mL. The parameters selected in the present experiments were described as follows in Table 3.

2.2. Materials

Simulated wastewater samples with an initial concentration of lead were prepared by dissolving ion nitrate $Pb(NO_3)_2$ in distillate water, where the required mass of this salt could be measured according to the following equation:

$$\text{Weight of salt (g)} = \text{Volume of solution (L)} \times \text{initial concentration of lead ions in solution (ppm)} \times (\text{M. wt of the lead nitrate/atomic weight of lead}) \quad (5)$$

In order to prevent the formation of an oxide layer on the anode electrode and to increase the conductivity of the simulated solution, an amount of sodium chloride was added.

Hydrochloric acid (0.1 N) and sodium hydroxide (0.1 N) were used to adjust the value of pH to obtain a neutral solution.

The simulated wastewaters of the experiments were prepared by dissolving $Pb(NO_3)_2$ having 99.99% of purity (BDH, England) in 500 mL of distilled water. The value of pH was adjusted by using 0.1 N HCl and 0.1 N NaOH. Electrical conductivity and decreasing passivation enhance removal efficiency by using 0.5 g/L of NaCl.

When the electrodes of the concentric tubes were immersed in the simulated wastewater, the DC current was switched on to supply 1.5 A (53 mA/cm^2) to the cell. Samples were collected from the treated simulated wastewater every 15 min and filtered by the cellulose Glass Microfiber discs (Grade: MGC; pore diameter was 0.47 μm, Munktell, Germany) before the analysis by the atomic absorption spectroscopy (type-AA-7000F, Shimadzu, Japan) for measuring quantities of lead ions present in the samples. At the end of each experiment, electrodes were washed one time with 0.1 N HCl and more than one time with water to ensure it was cleaned well. The same procedure was repeated for the next experiment according to the second configuration.

Table 2
Effects of electrodes configuration on removal efficiency

Configuration type	Electrodes	
	Anode electrode(s)	Cathode electrode(s)
Type 1: one anode – two cathodes	Mid only	Outer and inner
Type 2: one cathode – two anodes	Outer and inner	Mid only

Table 3
Experimental parameters

Parameters	Range or constant value
Initial lead concentration (ppm)	155
pH	7
Current or current density (A or mA/cm^2)	1.5 or 53
Stirring speed (rpm)	150
Contact time (min)	0–60

3. Results and discussion

Effects of the change of electrodes configuration as (type 1: one anode – two cathodes; type 2: one cathode – two anodes) under a period of time (5 – 60 min) and constant values of other parameters as shown in Table 2 were explained below.

3.1. Removal efficiency

From experiments results that consist of two different types of electrodes configuration (type 1: one anode – two cathodes; type 2: one cathode – two anodes). The efficiency of contaminate removing was larger when the cathode electrode concentrated between two other anode electrodes with restricted distance in between (i.e., type 2: one cathode – two anodes). Table 4 and Fig. 3 show that result.

Table 4
Effects of electrodes configuration on removal efficiency

Configuration type	Removal efficiency (%)	
	After 15 min	After 30 min
Type 1: one anode – two cathodes	82.34	99.31
Type 2: one cathode – two anodes	99.62	99.91

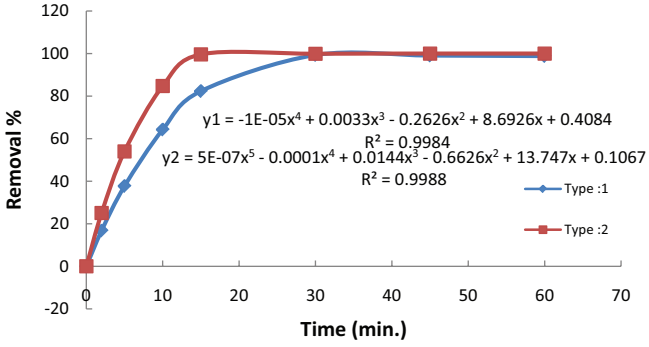

Fig. 3. Effecting of electrodes configuration on removal efficiency.

3.2. Temperature variation

Since the initial temperature is approximately 25°C, both types of configuration caused rising the temperature of the reactor with time because of continuous current supplied and ohmic drop between electrodes. The final temperature was the same in both experiments as shown in Table 5 and Fig. 4.

3.3. Energy consumption

This is an effective parameter for this kind of wastewater treatment techniques. In the present study, the first type of configuration (i.e., type 1: one anode – two cathodes) consumed a little more energy during the same period of its experiment than the other type, Table 6 and Fig. 5 explained that clearly.

3.4. Electrode consumption

In both types of configuration, some weight of electrodes was consumed, as shown in Table 7, Figs. 6 and 7, to complete the aim of electrocoagulation process according to previous Eqs. (1)–(4).

The previous results show that type two (i.e., one cathode – two anodes) configuration is more practical and efficient than the first type (i.e., two cathodes – one anode). The following table explains the summary of models that correlate each of responses with time at specified values of other experimental parameters listed in Table 3.

3.5. Simplified kinetic approach

Finally, a preliminary kinetic modeling for the best configuration (i.e., one cathode – two anodes) is provided to evaluate the reaction rate constants of electrocoagulation process. The general kinetic rate equation for representing the removal rate of lead concentration from the simulated water is described as follows:

$$\frac{dC_t}{dt} = -kC_t^n \tag{6}$$

where C represents the lead concentration, n is the order of reaction, k is the reaction rate constant, and t is the time.

Table 5
Effects of electrodes configuration on temperature rising

Configuration type	Maximum temperature (°C)
Type 1: one anode – two cathodes	45
Type 2: one cathode – two anodes	45

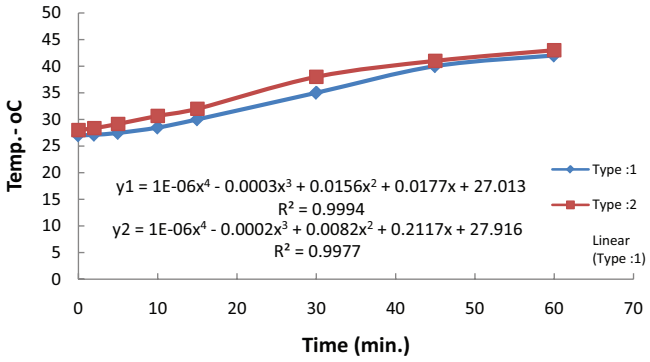

Fig. 4. Effecting of electrodes configuration on temperature rising.

Table 6
Effects of electrodes configuration on energy consumption

Configuration type	Energy consumption value (kWh/m³)
Type 1: one anode – two cathodes	20.175
Type 2: one cathode – two anodes	19.313

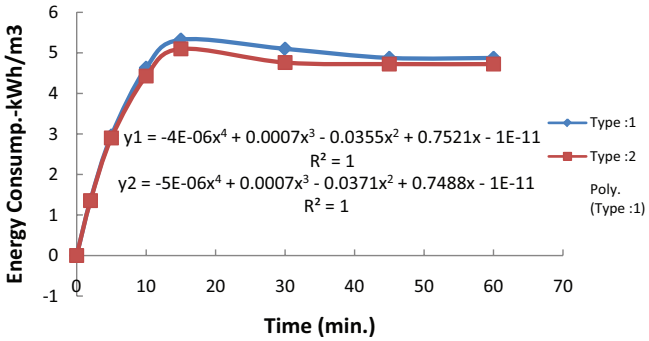

Fig. 5. Effecting of electrodes configuration on energy consumption.

Table 7
Effects of electrodes configuration on electrodes consumption

Configuration type	Electrodes consumption value (g)
Type 1: one anode – two cathodes	0.84
Type 2: one cathode – two anodes	0.93

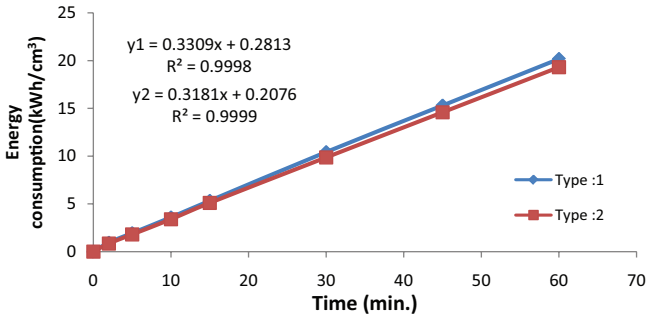

Fig. 6. Effecting of electrodes configuration on accumulated energy consumption.

Table 8
Summary of calculations for kinetic rate equation estimation

Time (min)	Removal %; Type: 2	C_t	C_t/C_o	$-\ln (C_t/C_o)$	$1/C_t$
0	0	155	1	0	0.0064
2	0.2506	116.150	0.7493	0.2885	0.0086
5	0.5401	71.275	0.4598	0.7768	0.0140
10	0.8476	23.611	0.1523	1.8816	0.0423
15	0.9962	0.586	0.0037	5.5778	1.7064
30	0.9990	0.146	0.0009	6.9675	6.8493

Table 9
Summary of kinetic rate equations

Reaction order	General equation	Simulated equation	k (mol/m^3)$^{1-n}$	R^2
0	$C_{A0} - C_A = k_o t$	$C_A = -4.813\ t + 110.86$	–4.813	0.6853
1	$-\ln\frac{C_t}{C_0} = k_1 t$	$-\ln\frac{C_t}{C_0} = 0.2542t - 0.0442$	0.2542	0.9028
2	$\frac{1}{C_t} - \frac{1}{C_o} = k_2 t$	$\frac{1}{C_t} = 0.2322t - 0.9619$	0.2322	0.8840
nth-order: $n = 2.149$	Using the fractional life method with $F = 80\%$	$\log t_F = -1.1492\ (\log C_{A0}) + 2.7973$	0.012	0.9035

Table 10
Summary of fractional life method calculation

C_{A0}	$C_{A\,\text{end}}$ (=0.8C_{A0})	Time needed t_F (min)	$\log t_F$	$\log C_{A0}$
155	124	1.5	0.1761	2.1903
71.28	57.024	7	0.8451	1.8530
23.612	18.889	14	1.1461	1.3731

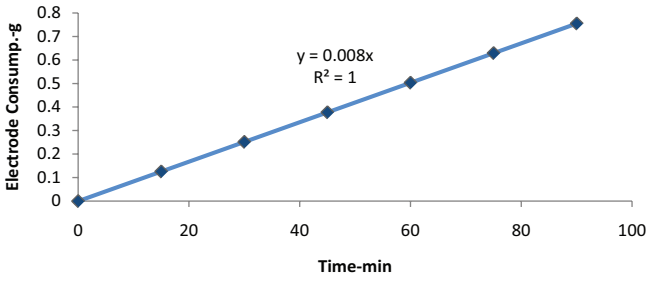

Fig. 7. Effecting of electrodes configuration on theoretical electrodes consumption.

Tables 8, 9 and 10 show the required calculation to estimate the kinetic rate reaction of electrocoagulation for lead removal.

Unfortunately, zero, first and second order do not give straight lines and their equations listed in Table 11 are approximated belong to their curves. Therefore, fractional life method t_f (i.e., Eq. (7)) with F = 80% is used to get the correct kinetic order of the reaction and its rate constant k (L/mg)$^{1-n}$ min^{-1} [15].

$$t_F = \frac{F^{1-n} - 1}{k(n-1)} C_{A0}^{1-n} \tag{7}$$

So, the rate equation that represents the electrocoagulation reaction is:

$$-r_{\text{Pb}} = \left(0.012 \frac{L^{1.149}}{\text{mg}^{1.149}\ \text{min}}\right) C_{\text{Pb}}^{2.149}, \quad \frac{\text{mg}}{\text{L min}} \tag{8}$$

4. Conclusions

Following are the results that were obtained from both electrocoagulation experiments data:

1. Configuration of electrodes in the reactor is the most effective factor to enhance the electrocoagulation reactor efficiency.
2. Configuration in the case of type two (i.e., one cathode – two anodes) is more efficient than type one (i.e., one anode – two cathodes) in removing lead from simulated wastewater.
3. Type one configuration (i.e., one anode – two cathodes) consumed a little more energy than type 2 (i.e., one cathode – two anodes).
4. Temperature rising of the solution in both types of configuration is approximately the same value.
5. Theoretical electrodes consumption for both types is the same value.
6. Moreover, the final value of pH is different from the initial value (i.e., pH = 7) as noticed.

Table 11
Summary of responses models

Responses	Configuration type	R^2	Model
Removal efficiency %	Type 1	0.9984	$y1 = -1E\text{-}05x^4 + 0.0033x^3 - 0.2626x^2 + 8.6926x + 0.4084$
	Type 2	0.9988	$y2 = 5E\text{-}07x^5 - 0.0001x^4 + 0.0144x^3 - 0.6626x^2 + 13.747x + 0.1067$
Temperature (°C)	Type 1	0.9994	$y1 = 1E\text{-}06x^4 - 0.0003x^3 + 0.0156x^2 + 0.0177x + 27.013$
	Type 2	0.9977	$y2 = 1E\text{-}06x^4 - 0.0002x^3 + 0.0082x^2 + 0.2117x + 27.916$
Energy consumption	Type 1	1.000	$y1 = -4E\text{-}06x^4 + 0.0007x^3 - 0.0355x^2 + 0.7521x - 1E\text{-}11$
	Type 2	1.000	$y2 = -5E\text{-}06x^4 + 0.0007x^3 - 0.0371x^2 + 0.7488x - 1E\text{-}11$
Electrodes consumption	Type 1	1.000	$y = 0.0084x + 2E\text{-}16$
	Type 2		
Accumulated energy consumption	Type 1	0.9998	$y1 = 0.3309x + 0.2813$
	Type 2	0.9999	$y2 = 0.3181x + 0.2076$

Symbols

C_o — Lead initial concentration (mol/m^3 or ppm)
C_t — Lead concentration at time t (mol/m^3 or ppm)
n — Order of reaction
k — Reaction rate constant
t_f — Fractional time
F — Fractional constant assumed

References

[1] N.K. Srivastava, C.B. Majumder, Novel biofiltration methods for the treatment of heavy metals from industrial wastewater, J. Hazard. Mater., 151 (2008) 1–8.
[2] F. Fu, Q. Wang, Removal of heavy metal ions from wastewaters: a review, J. Environ. Manage., 92 (2011) 407–418.
[3] C.C. Yang, Recovery of heavy metals from spent Ni–Cd batteries by a potentiostatic electrodeposition technique, J. Power Sources, 115 (2003) 352–359.
[4] G. Issabayeva, M. Kheireddine, Electrodeposition of copper and lead on palm shell activated carbon in a flow-through electrolytic cell, Desalination, 194 (2006) 192–201.
[5] A. Salman, Removal of Cadmium from Wastewater by Electrodeposition on Stainless Steel Concentric Tubes Bundle, PhD thesis, University of Baghdad, 2014.
[6] M.A. Barakat, New trends in removing heavy metals from industrial wastewater, Arab. J. Chem., 4 (2011) 361–377.
[7] A.A. Mohammed, Electrocoagulation of phenol for wastewater treatment, Iraqi J. Chem. Petrol. Eng., 9 (2007) 37–41.
[8] A.A. Mohammed, M.D. AlMureeb, Removal of lead from simulated wastewater by electrocoagulation method, J. Eng.,16 (2010) 5811–5821.
[9] Y. Oztekin, Z. Yazicigil, Recovery of metals from complex solutions by electrodeposition, Desalination, 190 (2006) 79–88.
[10] A.I. Adeogun, Electrocoagulation removal of anthraquinone dye Alizarin Red S from aqueous solution using aluminum electrodes: kinetics, isothermal and thermodynamics studies, J. Electrochem. Sci. Eng., 6 (2016) 199–213.
[11] I.A. Santos, I.O. Santos, L.V. Pontual, L.C. Monteiro, F.B. Mainier, Electrolytic removal of cadmium, lead and copper from wastewater, J. Environ. Prot., 7 (2016) 699–704.
[12] É. Fekete, Electrocoagulation: an electrochemical process for water clarification, J. Electrochem. Sci. Eng., 6 (2016) 57–65.
[13] E. Bazrafshan, L. Mohammadi, A.A. Moghaddam, A.H. Mahvi, Heavy metals removal from aqueous environments by electrocoagulation process – a systematic review, J. Environ. Health Sci. Eng., 13 (2015) 1–16. DOI 10.1186/s40201-015-0233-8.
[14] S. Bayar, Effects of operational parameters on cadmium removal from aqueous solutions by electrochemical coagulation, Desal. Wat. Treat., 51 (2013) 2635–2643.
[15] O. Levenspiel, Chemical Reaction Engineering, John Wiley & Sons, 3rd ed., 1999, pp. 49–50.