Effect of mono and bipolar connection modes on the electrocoagulation removal efficiency of multi-heavy metals from simulated wastewater

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ABSTRACT

Electrochemical treatment methods are frequently used to remove a wide range of pollutants from wastewaters generated by domestic and industrial operations. This work aims to investigate the impacts of using monopolar and bipolar connection modes of an electrocoagulation reactor (ECR) used to remove multi-toxic metals from synthetic wastewater. The present design of the ECR involves concentric-multi-cubic (CMC) aluminum electrodes with an activated area of 360 cm^2. The anode electrodes are perforated to be light-weight and decrease the amount of anode consumption as well as the increase of oxygen bubbles released that were assisting the buoyancy process of light-pollutants toward the surface of the solution in addition to the hydrogen bubbles are released from the plane electrodes of the cathode. The synthetic wastewater contains 100 ppm of each Pb, Cd, and Cu ion under the effects of pH of 7, applied current of 1.4 A (which equals remove to 3.88 mA/cm2), NaCl of 2 g, 300 rpm of stirring speed, and reaction time of (0-90 min). The core results proved that the bipolar connection mode (BCM) was more effective than the monopolar connection mode (MCM) in toxic metal-wastewater treatment. After 60 min of the reaction time, the highest removal efficiencies of these metals After a reaction timeof 60 min, the highest removal efficiencies of Pb, Cd, and Cu metals obtained via the BCM system were : 99.91%, 99.68%, and 99.14%, respectively 99.91%, 99.68%, and 99.14%, respectively. While they achieved 60.55%, 64.24%, and 89.55% via the MCM system , . re spectively The present new design of electrodes using the bipolar system was more reliable in wastewater treatment containing toxic metals with significantly low values of electrical energy consumption, electrode consumption, and cost-effectiveness

1. Introduction

In the modern world, large amounts of wastewater are discharged from industrial activities to the environment containing several types of pollutants that can be classified as degradable or non-degradable pollutants[1-3]. Domestic wastewater contains various organic materials and inorganic materials [4]. Industrial polluted water contains several types of toxic pollutants such as cyanide, pesticides, and heavy metals . Metals with a specific gravity greater than five are classified as heavy metals such

as lead, cadmium, zinc, chromium, copper, and mercury [5]. Excessive use of water results in the production of large quantities of wastewater laden with impurities in the water system and must then be treated through treatment processes. The industrial activities in Iraq, such as the petrochemical and chemical industries, in 2014 alone consumed more than (44,300) cubic meters of fresh water per day and produced more than (17,500) cubic meters of polluted water per day which contain different

organic and inorganic pollutants [6]. One of the most important resources is water.. As a result of the great industrial development, the increase in the population and the use of water in various matters, such as in homes and industries [7]. There fore , wastewater must be treated and pollutants removed for economical and environmental development [8], using effective methods such as chemical precipitation [9] ,ion exchange[10], adsorption [11], membrane filtration [12], nanomaterial [13], and electrocoagulation [14]. During the last twenty years, electrochemical technologies are developed in water and wastewater treatment with higher efficiency than other conventional methods because of their eco-friendliness by controlling pollution, versatility through the flexibility to treat a spread of pollutants via redox reactions, energy efficiency due to the minimizing of non-homogeneously in current applied distribution then the drop, safety, selectivity, and price effectiveness. Moreover, electrochemical technologies are simple, easily operable, and low amount of sludge produced without the generation of secondary pollution. Electrocoagulation is a distinct process based on the concept of (electrochemistry) using electric current to remove toxic metals from solutions containing waste discharged from various industrial uses such as metal plating wastewater [15], oily wastewater [16], polluted groundwater [17], wastewater from paper industries [18], olive mills-wastewater[19],municipal waste water [20] , etc .Electrocoagulation (EC) is one of the emerging processes for heavy metal-contaminated wastewater treatment [1] . The EC method is more economical and reliable when compared to other treatment techniques. (for its ability to make many contributions to the environmental treatment, recycling and ,control [21-23], Fig. 1.
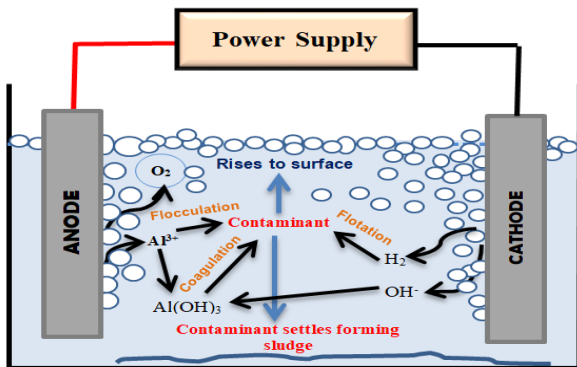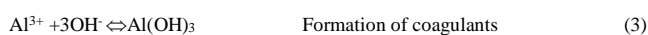

Figure 1. General schematic of electrocoagulation technique (AlJaberi, 2022)

The chemical reactions proceed in the EC reactor are shown in the follwing Eqs. 1 to 3, [23]:

$$Al_{(S)} \Rightarrow Al^{3+}{}_{(aq)} + 3e^- \qquad \text{At the anode} \qquad (1)$$

$$2H_2O + 2e^- \Rightarrow H_{2(g)} + 2OH^-{}_{(aq)} \qquad \text{At the cathode} \qquad (2)$$

$$Al^{3+} + 3OH^- \Leftrightarrow Al(OH)_3 \qquad \text{Formation of coagulants} \qquad (3)$$

Several studies have used electrocoagulation method to remove heavy metals from wastewater by performing different configurations of electrodes.AbdulRehman, et al. (2015), [24] used bipolar aluminum electrodes in a batch electrocoagulation reactor to remove heavy metals such as lead from wastewater under the effect of the electrolysis, current density (,33 A/m2) and ,reaction time(30min), lead concentrations (5-15ppm), and solution pH (7)achieving 94% of lead removal efficiency.

Abdul Rehman, et al. used iron and aluminum plane-electrodes in a bipolar connecting mode to remove (105 ppm Ni ion,110 ppm Cu ion, and 63 ppm Pb ion) in a continuous electrocoagulation system under the impact of pH (3-9), inter-electrode spacing (4-24mm), hydraulic retention time (20-12sec.), and current density (0.007-0.04)A/cm2. , optimal experimental conditions (actual pH 6.32, current density 0.026 A cm-2), the removal efficiency of heavy metals from artificial wastewater was higher than 95 % Bernard, et al. (2013) [25] investigated the ability of an EC for simultaneous Ni, Zn, and Mn removal from sewage wastewater utilizing monopolar iron electrodes. The experiments were done under the impact of current density (from 2 to 25 mA/cm2) , Initial pH (3, 5.68, 8.95) and initial metal concentration (50 to 250 mg/L) . They found that higher pH levels are more suitable for metal removal by EC treatment. Except for Mn, which obtained a 72.6 percent removal value at a current density of 25 mA/cm2 and a total energy usage of 49 kWh/m3, all metals investigated achieved better than 96 percent removal efficiency1 . Tubular and finned shapes of electrodes are invented and performed in previous studies such as [26-27] in wastewater treatment.

This study aims to investigate the ability of a new configuration of EC reactor involving concentric-cubic-electrodes on the removal efficiency of multi-heavy metals from synthetic wastewater via the compare is on between two connection modes which were the bipolar connection mode (BCM) and the monopolar connection mode (MCM). The operational parameters studied were the initial concentrations of Pb, Cd, and Cu metals, pH, reaction time, applied current, NaCl concentration, and the stirring speed. The anode electrodes are designed as perforated shapes to be light-weight and to decrease the amount of anode consumption as well as the increase of oxygen bubbles released that were assisting the buoyancy process.

2. Experimental work

2.1. Apparatus

A 3 liter glass batch EC reactor was used to conduct the present experiments (Figures 2). It consists of four concentric aluminum cubes with different dimensions but the same thicknesses as given inTable1 with an active area of approximately (360 cm2). The EC cell was connected to a DC digital power supply (Model: SYADGONG-305D Company, China) to provide the designed current. A constant mixing speed of 300 rpm was provided by using a magnetic stirrer (Model: ALFA company, Iran: D-500; 100–1,800 rpm). The value of solution pH was measured by using an electronic pH meter (Model: ATC company, China), [28]. The concentric cubic electrodes were aluminum containing two perforated anodes and two non-perforated cathodes, Fig. 3.

Table 1. The dimensions of the perforated (P) and non-perforated (NP) electrodes

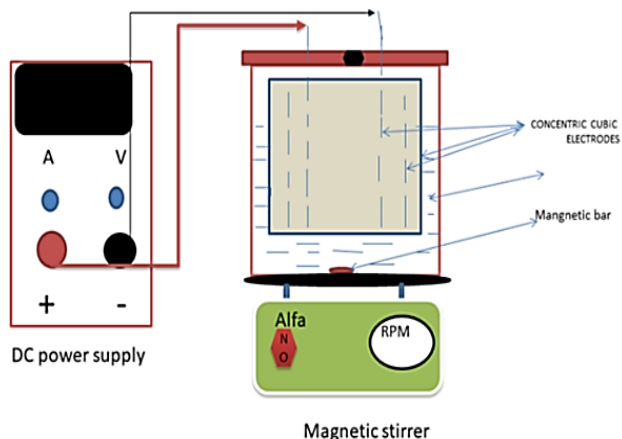| No.	Anode/ Cathode	P or NP	Dimensions (cm)	Electrode thick (cm)	Electrode Distance (cm)	Wet height (cm)
1	Anode	P	4 × 4 × 10	0.2	2	4
2	Cathode	NP	8 × 8 × 10	0.2	2	4
3	Anode	P	12 × 12 × 10	0.2	2	4
4	Cathode	Np	16 × 16 × 10	0.2	2	4

Figure. 2. The schematic of the present EC reactor
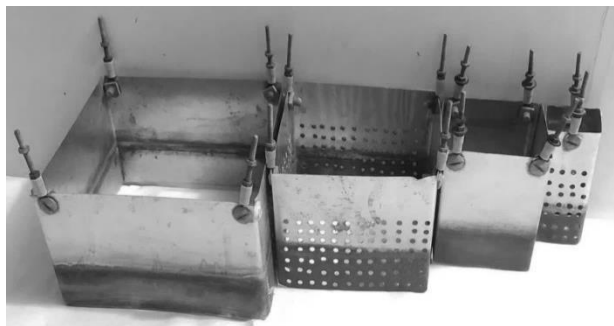
Figure.3. The configuration of the cubic electrodes

Table 2. The values of experimental parameters

Parameters	Values
Pb ions (ppm)	100
Cd ions (ppm)	100
Cu ions (ppm)	100
Ph	7
Current (Aor mA/cm2)	1.4or 3.88
NaCl (g)	2
Stirring speed (rpm)	300
Reaction time (min)	0-90

This study compared the ability of these electrodes in wastewater treatment between two operating systems which are the bipolar connection mode (BCM) and the monopolar connection mode (MCM). For the most part, electrochemical investigations have been used to investigate the operational parameters of this work.. Their values (Table 2) are selected based on a previous review paper [29].

2.2. Materials

A 2.5 liter synthetic wastewater was prepared by dissolving the required weights of Pb, Cd, and Cu salts in distilled water. The NaCl electrolyte was added to raise the electrical conductivity of the solution and prevent the passivation, i.e. the formation of an oxide layer, on the electrodes. The

required pH value was adjusted using NaOH (0.1 N) and HCl (0.1 N). When the electrodes immersed were in the simulated wastewater, the DC-current of 1.4 A(3.88 mA/cm²) was supplied to the electrochemical cell and the magnetic stirrer was turned on to agitate the solution at (300 rpm). The sampling of each experiment was conducted at the periods of (5, 10, 15, 30, 45, 60, 75, and 90 min). The analysis of the collected samples was done using atomic absorption spectroscopy(Model: AA-7000F, Shimadzu, Japan), [28] to determine the final concentrations of toxic metals present in the treated solution. After each experiment done for each configuration, these electrodes were washedby0.1 N HCl and water to ensure it was cleaned well.

The electrical energy consumed by the electrochemical cell is calculated using Eq.4 as follows:

$$Electrical\ Energy\ Consumption\ (EEC)\ = \frac{U \times I \times t}{V} \tag{4}$$

where: U is the applied voltage (volt), I is the electric current (A), t is the reaction time (h), and V is the solution volume of the contaminated water (m³).

The theoretical consumption of electrodes could be estimated from the following equation (Eq. 5):

$$TEC\ = \frac{I \times t \times M}{Z \times F} \tag{5}$$

Where: M is the molecular weight of electrodes metal, Z is the electrons number presented in the electrolysis reaction (for Al is 3), and F is Faraday's constant (96485.34 Col/mol).The removal efficiency of each toxic metalis determined using Eq. (6):

$$Removal Efficiency\ = \frac{c_0 - c_t}{c_0} \times 100 \tag{6}$$

where C_0 and C_t are the initial and final concentration of each metal (ppm).

3. Results and discussion

The impact of changing the connection mode to the DC-power supply as BCM and MCM using the new configuration of electrodes, i.e. perforated and non-perforated concentric cubic-electrodes, was investigated in multi-heavy metals removal from synthetic wastewater under the influences of several parameters that listed in Table 2.

3.1. Removal efficiency

Based on the obtained results of the BCM and MCM experiments, the removal efficiencies of Pb, Cd, and Cu ions from simulated wastewater were higher in the case of BCM compared to that obtained in the case of MCM. Tables3 to 5 and Figures 4 to 6 reveal these findings.

Table 3. Results of BCM and MCM experiments for Pb removal efficiency

Reaction time (min)	BCM Experiment		MCM Experiment	
	Pb Concentration (ppm)	BCM-Pb Removal %	Pb Concentration (ppm)	MCM-Pb Removal %
0	100	0	100	0
5	10.2648	89.7352	88.30	11.696
10	9.2896	90.7104	81.19	18.81

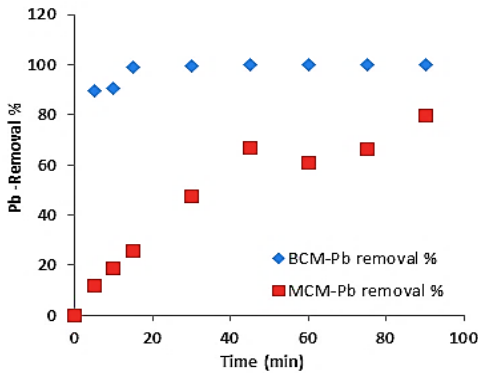| 15	1.1035	98.8965	74.63	25.373
30	0.4106	99.5894	52.70	47.296
45	0.0257	99.9743	33.21	66.789
60	0.0924	99.9076	39.45	60.553
75	0.231	99.769	33.67	66.327
90	0.1386	99.8614	20.28	79.722

Figure 4. BCM and MCM removal of 100 ppm Pb ions from simulated wastewater
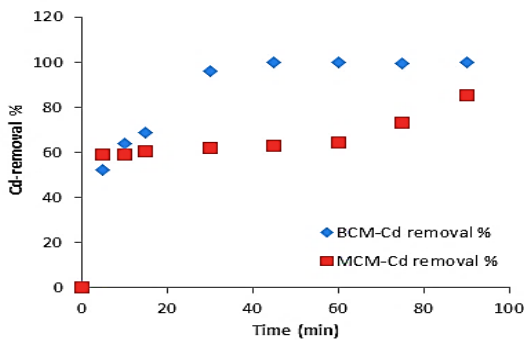

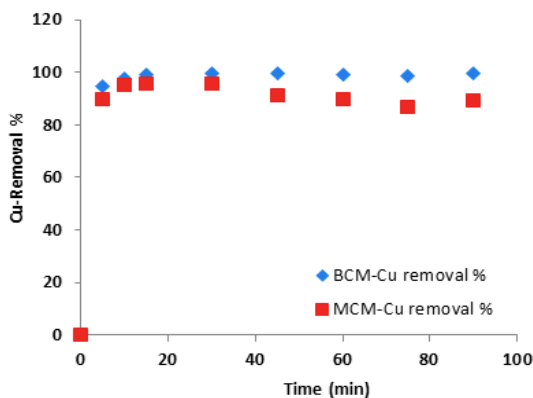**Figure 5. BCM and MCM removal of 100 ppm Cd ions from simulated wastewater**

Figure 6. BCM and MCM removal of 100 ppm Cu ions from simulated wastewater

Table 4. Results of BCM and MCM experiments for Cd removal efficiency

Reaction time (min)	BCM Experiment		MCM Experiment	
	Cd Concentration (ppm)	BCM-Cd Removal %	Cd Concentration (ppm)	MCM-Cd Removal %
0	100	0	100	0
5	47.785	52.215	40.9508	59.0492
10	36.431	63.569	41.0118	58.9882
15	31.2819	68.7181	39.8344	60.1656
30	4.1664	95.8336	38.3399	61.6601
45	0.3843	99.6157	36.9511	63.0489
60	0.3213	99.6787	35.7615	64.2385
75	0.4555	99.5445	26.9996	73.0004
90	0.002	99.998	14.8073	85.1927

Table 5. Results of BCM and MCM experiments for Cu removal efficiency

| Reaction time (min)	BCM Experiment		MCM Experiment	
	Cu Concentration (ppm)	BCM-Cu Removal %	Cu Concentration (ppm)	MCM-Cu Removal %
0	100	0	100	0
5	5.2944	94.7056	10.309	89.691
10	2.3296	97.6704	4.8255	95.1745
15	0.8547	99.1453	4.47	95.53
30	0.2798	99.7202	4.175	95.825
45	0.3857	99.6143	8.9778	91.0222
60	0.8547	99.1453	10.4452	89.5548
75	1.195	98.805	13.2134	86.7866
90	0.4614	99.5386	10.846	89.154

As revealed that the removal efficiencies of heavy metals were improved when the concentric cubic-electrodes were arranged as bipolar connection mode (BCM) compared to those of the monopolar connection mode (MCM). After a reaction time of 60 min, the highest removal efficiencies of Pb, Cd, and Cu metals obtained via the BCM system were : 99.91%, 99.68%, and 99.14%, respectively. While they achieved 60.55%, 64.24%, and 89.55% via the MCM system, respectively .These results were agreed with [30-31].

3.2. Electrical Energy consumption

This is an effective parameter in wastewater treatment when any type of electrochemical technology is performed. Table 6 and Figure7 list the value of EEC (Eq. (1)) at the highest removal efficiency for each metal using BCM and MCM configuration systems. As observed that the total energy consumption of the BCM system is less than of the MCM system. The new configuration of the present electrodes assists the performance of the electrocoagulation reactor by minimizing the consumption of the electrical energy [32].

Table 6. Energy consumption for each metal using BCM and MCM systems

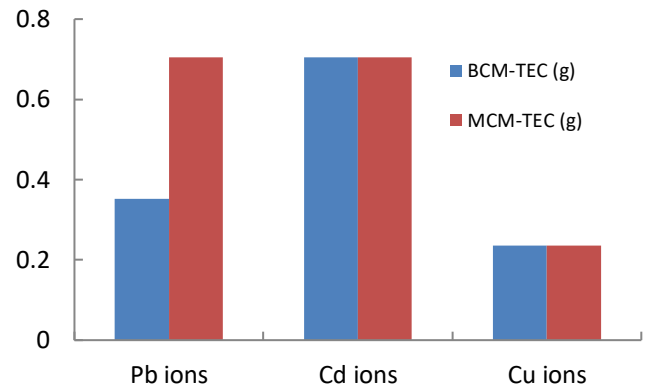| Heavy metals	BCM System			MCM System		
	Reaction time (min)	Removal %	EEC (Wh/m3)	Reaction time (min)	Removal %	EEC (Wh/m3)
Pb	45	99.974	12.6	90	79.722	25.2
Cd	90	99.998	25.2	90	85.193	25.2
Cu	30	99.720	8.4	30	95.825	8.4
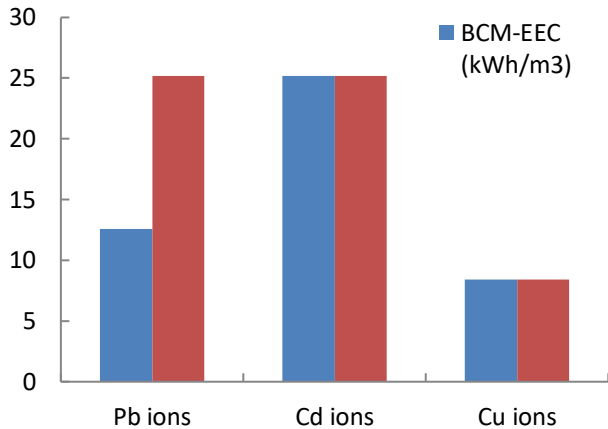
Figure 7. Energy consumption via the BCM and MCM system to remove multi-heavy metals from simulated wastewater

3.3. Theoretical electrodes consumption

Table 7 and Figure8 explain the amounts of the theoretical consumption of electrodes measured in (gram) using Eq. (2) via both systems of BCM and MCM. The core findings proved that the total consumption of electrodes was higher through the MCM system compared to that measured via the BCMsystem. In general, the small amount of electrodes consumption observed is due to the new configuration of perforated electrodes of the anode. This observation is similar to that found by [33] was 0.23 g for BCM system and 0.58 g for the MCM system which means that the BCM is cost-effective.

Table 7. Electrodes consumption for each metal using BCM and MCM systems

Heavy metals	BCM System			MCM System		
	Reaction time (min)	Removal %	TEC (g)	Reaction time (min)	Removal %	TEC (g)
Pb ions	45	99.974	0.352	90	79.722	0.705
Cd ions	90	99.998	0.705	90	85.193	0.705
Cu ions	30	99.720	0.235	30	95.825	0.235

Figure 7. Electrodes consumption via the BCM and MCM systems to remove multi-heavy metals from simulated wastewater

As observed that the bipolar connection mode (BCM) is more effective than the mono-polar connection mode to remove Pb, Cd, and Cu ions from synthetic wastewater with low values of electrodes and energy consumption. Table 8 lists a summary of some previous studies which investigated the ability of electrocoagulation technology in wastewater treatment using bipolar and monopolar connection modes.

Table 8. Summary of some previous studies used that electrocoagulation removal of multi-heavy metals from wastewater

References	Pollutants	BCM or MCM	Removal Efficiency%
Ahmad1, et al. [34]	Cu and Zn	BCM	96%
C. Escobara	Cu, Pb, and Cd	MCM	80
U. T.Un et al. [35]	Cd, Cu, and Ni	BCM	98
A.Rehman [31]	Cu, Ni, and Pb	BCM	95
Al Aji, et al. [34]	Cu, Ni, Zn, and Mn	MCM	96
Present study	Pb, Cd, and Cu	BCM	>99

3.4. Simplified kinetic approach

In general, the order of any reaction is based on several conditions such as the stoichiometry of the chemical reaction equation which is corresponded or does not with the rate equation.There fore, the order of reaction throughout every reactor should be studied. The kinetic modeling for the effective configuration (i.e., BCM) is provided to estimate the reaction rate constants of the electrocoagulation process. Eq. 7 shows the general rate equation that represents the removal rate of each metal:

$$\frac{dC_t}{dt} = -kC_t^n \tag{7}$$

where C is the concentration of each metal, k is the rate constant,n is the reaction's order, and t is the reaction time. Table 9 lists the summary of the kinetic study for each heavy metals throughout the electrocoagulation reactor using the effective configuration of electrodes, i.e. BCM system.

Table 9. summery of the kinetic study

Heavy Metals	Reaction order	General equations	Simulated equations	k(mol/m3)1-n	R2
Pb	1	$-\ln(C_t/C_i)=k_1t$	$y = 0.1731x + 0.8536$	0.1731	0.864
	2	$(1/C_t)-(1/C_i)=k_2t$	$y = 20.592x - 111.63$	20.592	0.842
Cd	1	$-\ln(C_t/C_i)=k_1t$	$y = 0.101x + 0.0063$	0.1010	0.964
	2	$(1/C_t)-(1/C_i)=k_2t$	$y = 0.1951x - 1.1432$	0.1951	0.776
Cu	1	$-\ln(C_t/C_i)=k_1t$	$y = 0.3021x + 0.5989$	0.3021	0.905
	2	$(1/C_t)-(1/C_i)=k_2t$	$y = 6.4166x - 20.00$	6.4166	0.635

As revealed in Table 9 that all heavy metals in this work obeyed in their behavior the first order reaction with the highest values of the regression coefficient (R^2) for each. The new design provides an additional advantage as Pb, Cd, and Cu removal from wastewater via a first order reaction which is fastest than the second-order reaction.

4. Conclusions

This study investigated the ability of an electrocoagulation reactor involving a new configuration of electrodes to remove multi-toxic metals from synthetic wastewater. Four concentric-cubic electrodes made of aluminum were performed as two-perforated anodes and two-non-perforated cathodes. A comparison of two connection modes (bipolar and monopolar, i.e. BCM and MCM) to the DC-power supply was conducted .The main conclusion points are listed below:

1. The new configuration of electrodes is significantly enhancing the electrocoagulation reactor efficiency.
2. The BCMconfiguration is more effective than CMC the configuration in removing Pb, Cd, and Cu from synthetic wastewater.
3. The BCM system consumes a low electrical energy compared to that observed by the MCM system.
4. The BCM system consumes a low theoretical consumption of electrodes compared to that of the MCM system.

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