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 cm². 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 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 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. 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.

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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]. Therefore, 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.

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 (4-24nm), 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 efficiency. 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 lightweight 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 in Table 1 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 thickness (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              |

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

\[ \text{Al}(s) \Rightarrow \text{Al}^{3+} (aq) + 3e^- \]  
\[ 2\text{H}_2\text{O} + 2e^- \Rightarrow \text{H}_2(g) + 2\text{OH}^- (aq) \]  
\[ \text{Al}^{3+} + 3\text{OH}^- \Rightarrow \text{Al}(	ext{OH})_3 \]  

Several studies have used electrocoagulation method to remove heavy metals from wastewater by performing different configurations of electrodes. Abdul Rehman, 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.
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 washed by 0.1 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:

\[ \text{Electrical Energy Consumption (EEC)} = \frac{U \times I \times t}{V} \]  

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):

\[ TEC = \frac{I \times t \times M}{Z \times F} \]  

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 metals determined using Eq. (6):

\[ \text{Removal Efficiency} = \frac{C_0 - C_t}{C_0} \times 100 \]

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. Tables 3 to 5 and Figures 4 to 6 reveal these findings.

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.

![Figure 2. The schematic of the present EC reactor](image2)

![Figure 3. The configuration of the cubic electrodes](image3)

| Table 2. The values of experimental parameters |
|-----------------------------------------------|
| Parameters                  | Values |
| Pb ions (ppm)               | 100    |
| Cd ions (ppm)               | 100    |
| Cu ions (ppm)               | 100    |
| pH                          | 7      |
| Current (A or mA/cm²)       | 1.4 or 3.88 |
| NaCl (g)                    | 2      |
| Stirring speed (rpm)        | 300    |
| Reaction time (min)         | 0-90   |

| Table 3. Results of BCM and MCM experiments for Pb removal efficiency |
|---------------------------------------------------------------------|
| Reaction time (min) | BCM Experiment | MCM Experiment |
|                      | Pb Concentration | Pb Removal % | Pb Concentration | 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        |
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 Figure 7 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 4. Results of BCM and MCM experiments for Cd removal efficiency |
|-------------------------------------------------------------|
| **Reaction time** | **BCM Experiment** | **MCM Experiment** |
| (min) | Cd Concentration (ppm) | Removal % | Cd Concentration (ppm) | 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** | **BCM Experiment** | **MCM Experiment** |
| (min) | Cu Concentration (ppm) | Removal % | Cu Concentration (ppm) | 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 |

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The Cu ions Cd ions = −

The Cu ions Cd ions effective.

This observation is similar to that found observed BCMsystem. was higher through the MCM system compared to that measured via the MCM. The core findings proved that the total consumption of electrodes measured electrodes consumption Table 7 lists a summary of some previous studies which investigated the ability of electrocoagulation technology in wastewater treatment using bipolar and monopolar connection modes.

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 removal rate of each metal:

Table 8. Summary of some previous studies used 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                 |

3.3. Theoretical electrodes consumption

Table 7 and Figure 8 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 BCM system. 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.

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. Therefore, 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:

\[
\frac{dC_i}{dt} = -kC_i^n
\]  

(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.

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

Figure 6. Electrodes consumption for each metal using BCM and MCM systems
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 BCM configuration is more effective than CMC 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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