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RESEARCH ARTICLE

Electrocoagulation for the Removal of Copper and Zinc Ions from Water Using Iron Electrodes

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Abstract:

Background:

Many methods have been suggested for the removal of heavy metals from water to protect human health and the environment. Methods like precipitation and adsorption were proposed for this purpose.

Objective:

Electrocoagulation involves the generation of coagulant by the action of electricity on two metal electrodes (iron or aluminium) to aid the process of water decontamination.

Methods:

Electrodeposition cell was made with iron electrodes and application of voltage from the power supply (5-25 V) dipped in the working solution (Cu and Zn) at various concentrations (10-50 mg.mL⁻¹) for 30-150 min. Samples were drawn and analysed by atomic absorption spectrophotometry.

Results:

The work indicated efficient removal of the metal ions. The dependence of removal efficiency on the three parameters was studied. The behaviour of the two metal ions was not identical. At low initial concentration, the electrolysis voltage was very important in the removal of Zn and Cu ions. Electrolysis time is essential in the removal process and shows a polynomial dependence of removal efficiency on time. Electrolysis time of 150 min resulted in almost complete removal (94-97%) regardless of the initial concentration. Both co-precipitation and adsorption mechanisms may be involved.

Conclusion:

The removal efficiency was directly dependent on the initial metal ion concentration and electrolysis time. The process gave removal efficiency for copper that is higher than that of the zinc.

Keywords: Electrocoagulation, Iron electrodes, Removal of copper, Removal of zinc, Water, Optimum conditions.

Article History

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1. INTRODUCTION

The occurrence of heavy metals (HM), in the environment, represents a real challenge. Hence, they must be removed efficiently from surface water to reduce their adverse effects on aquatic life. Any action towards preventing the danger caused by heavy metals to human health and the environment, must involve their removal. This is because they cannot be transfor-

med into another safe species nor being biodegradable. They tend to accumulate in living organisms (Saha and Sanyal, 2010 [1]). Many methods have been suggested for the removal of heavy metals, depending on the nature and chemistry of the metal ions themselves and their interaction with the environment. The reaction with MgO was suggested to initiate the precipitation reactions and consequently to isolate the heavy metal ions from water in an easily separable species [Barbooti, *et al.*, 2011 [2]]. Adsorption receives a wide interest where the heavy metal ions attach themselves to a solid through

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electronic interaction with active centres on the adsorbent particle surfaces. A variety of single and composite materials have been studied and approved as potential adsorbents for the removal of heavy metals from water. Barbooti *et al.*, 2019 [3] introduced kaolin- polyacrylic acid and polyacrylamide composites as adsorbents for the removal of nickel ions from water. Clay minerals like Kaolin, montmorillonite were also used to adsorb heavy metal ions like Pb, Cr, and Ni from water [3 - 6]. Nano-materials were another category of adsorbents that have been utilized for the removal of heavy metals. However, costly adsorbents need to be regenerated and reused to assist the economy of the process [7].

Electrocoagulation, EC, was a recently introduced technique for the purification of water (Barrera-Díaz *et al.* 2012 [8]). The method is based on the co-precipitation and inclusion of the target materials within the in-situ formed coagulants like Al and Fe hydroxides (Naje, *et al.*, 2016 [9]). Roa-Morales, *et al.* [10], employed the method for the removal of color and chemical Oxygen Demand by coupling with Ozone treatment. The EC involves the formation of coagulants by electrolytic oxidation of the anode. The emulsion will be broken and the suspension of particulate and pollutants is weakened (Chaturvedi 2013 [11]). Flocs will then form through the coagulation and produces a sludge blanket, which capitulates and bridges the colloidal species in the aqueous phase. The solid oxides, hydroxides, and oxyhydroxides generate the active surfaces for the adsorption of the contaminants.

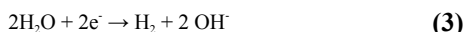
The potential of iron anode and cathode is based on the production of Fe(III) at the anode and reduction to the Fe(II) at the cathode (Vepsäläinen 2012 [12], Moreno *et al.* (2009 [13] and Gomes *et al.* 2007 [14]). Neutral to slight alkaline pH medium is typical for the oxidation into Fe(III), as in Equation (1), (Barrera Diaz *et al.* 2012 [9], Sasson *et al.* 2009 [15]).



The amount of metal cations dissolved during the reactions at the anode is calculated using Faraday's law [Eq. (2)] (Chaturvedi 2013 [11], and Khaled *et al.* 2015 [16]):

$$M = (ItM_w)/zF \quad (2)$$

where I is the current (A), t is the operation time (s), M_w the calculated value according to Faraday's law under is the molecular weight of the substance (g/mol), F is Faraday's constant (96,485 C/mol), z is the number of electrons involved in the reaction is 3 for Fe and Al), and m is the quantity of metal dissolved (g). Hydroxyl ions are produced at the cathode (Eq. 3), or by the consumption of the hydronium ions (Eq. 4) (Chaturvedi 2013 [12]):



Oxygen is formed on the anode (Khaled *et al.* 2015 [16]) under typical conditions (electrochemical potential) of EC systems when the anode dissolution follows Faraday's law (Mouedhen *et al.* 2008 [17]). The current density directly changes, affecting hydrogen production. In comparison with

chemical coagulation, EC surges the pH of the solution if acidic, neutral, or slightly alkaline and declines when it is extremely alkaline. This ultimately affects the speciation of aluminum and iron hydroxides. At highly acidic systems (pH 2), the EC is not adequate to raise the pH of the solution by the formed OH ions. Meanwhile, pH value shows some increase for solutions with pH 3 and higher (Mouedhen *et al.* 2008 [17]). The pH change is then affected by the anion concentration in the solution. The electrodes, especially aluminum, may be activated with Mg, In and Zn alloying elements to improve the performance [18]. Heidmann and Calmano (2008 [19]) reported that Zn(II), Cu(II), Ni(II), and Ag(I) can be eliminated from the solution by hydrolyzation and co-precipitation. However, Cr(VI) may be reduced to Cr(III), and precipitated as hydroxide. Iron electrodes produced crystalline phases, such as magnetite, and poorly crystalline phases, such as iron oxyhydroxides and lepidocrocite. Un and Aytac (2013 [20]) investigated the sludge produced after EC process with iron electrodes for textile wastewater treatment and described it by X-ray diffraction (XRD) to determine the main elimination pathway in the reactor. Electrode material determines the type of electrochemical reactions taking place in the EC system. Iron dissolves as Fe(II) and is oxidized in bulk solution to Fe(III) in the presence of oxidants like oxygen (Un and Aytac 2013 [21], Roopashree and Lokesh 2014 [[21]). Fe(II) is a weak coagulant compared to Fe(III) because of the higher solubility of hydroxides and lower positive charge (Un and Ocal 2015) [22]. Ideal material selection depends on the type of contaminants and the chemical characteristics of the electrolyte. The method was optimized for the treatment of metal plating wastewater using iron electrodes [23]. Linares-Hernández *et al.* (2009 [24]) reported high efficiency in reducing chemical oxygen demand (COD) from industrial wastewater for both color and COD reduction, a combination of iron and aluminum electrodes. Similar results were obtained when paper mill wastewaters were treated with various aluminum and iron electrode combinations (Katal and Pahlavanzadeh 2011 [25]). Similar results were obtained for copper, chromium and nickel removal from metal plating wastewater (Heidmann and Calmano 2010 [19]). Brahmi, *et al.*, 2019, investigated the electrocoagulation reactor design parameters effect on the removal of cadmium ions from synthetic and phosphate industrial wastewater [26, 27]. The application of the method was further simplified by utilizing solar photovoltaic cells to supply the needed electrical power to feed the process. Khan *et al.* [28], employed a column EC reactor to remove hexavalent chromium from water.

In the present work, an electrocoagulation system is constructed with iron electrodes and used in the removal of copper and zinc ions from water. The effect of working parameters are investigated on the removal efficiency of the ions

2. MATERIALS AND METHODS

2.1. Apparatus

The copper and zinc concentrations were measured on AA-7000 supplied from (SHIMADZU) atomic absorption spectrophotometer using the operating conditions

recommended by the manufacturer with the proper dilution of the samples as required.

2.2. Chemicals and Reagents

The standard stock solution of zinc (1000 mg.L^{-1}) was prepared by dissolving the analytical grade metal powder from (BDH) in hydrochloric acid and diluted to the mark with nano grade deionized water. For copper, the stock solution (1000 mg.L^{-1}) was prepared by dissolving the analytical grade copper sulfate from (BDH) in deionized and further dilution to the mark of the calibrated flask nano grade deionized water. The preparation of the working solution ($10\text{-}50 \text{ mg.L}^{-1}$) was done by placing the calculated volume with water to make 500 mL. The electrode material was commercially available steel slabs with dimensions of $3 \times 50 \times 140 \text{ mm}$.

Fig. (1): Schematic diagram of electrocoagulation set up, 1, Magnetic stirrer, 2, Working solution, 3, Iron electrodes, 4, Plastic bar, 5, Power supply, 6, connection wires.

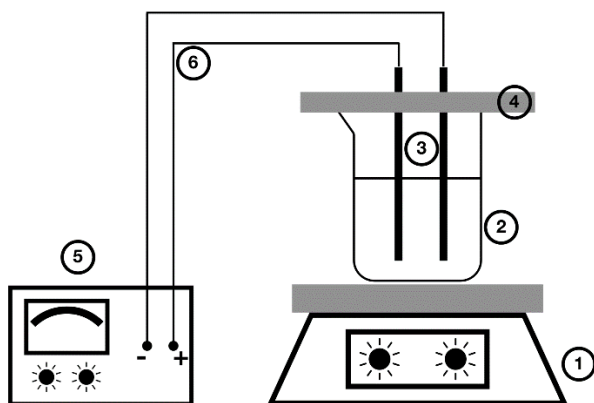


Fig. (1). shows the schematic diagram of the electrocoagulation system. In an 800-mL beaker, the calculated volume of the standard zinc or copper stock solution was placed together with deionized water to make 500 mL of the working solution of ($10\text{-}50 \text{ mg.L}^{-1}$) initial concentration. Two electrodes were made from commercially available steel slabs with dimensions of $3 \times 50 \times 140 \text{ mm}$. The electrodes were suspended and fixed in place with the aid of a perplex bar with rectangular holes of the dimensions of the steel slabs and placed on the beaker lid. Clamps were employed to ensure electrical contact with the power supplier. Almost 5.5-cm of the electrode length was dipped in the working solution. This made a working area of 27.5 cm^2 .

2.3. Electrocoagulation Setup

2.4. Procedures

Before each treatment, the electrodes (anode and cathode) were rubbed with sandpaper then cleaned with NaOH (2 M) and HCl (2 M) aqueous solutions to avoid a passivation film (Khaled *et al.*, 2015 [16]). The homogenization of the working

solution was done by mixing with a magnetic stirrer to form and float the flocs. Fast stirring is beneficial at the beginning to allow the water component to mix with coagulants and to disperse chemicals in the wastewater stream (Vepsäläinen 2012 [12]). The solution was kept without pH adjustment as almost neutral pH gave efficient removal of copper [29]

3. RESULTS AND DISCUSSION

3.1. Effect of Electrolysis Voltage

The effect of electrolysis time on the removal of Cu and Zn from water by electrocoagulation using various voltages was studied, and the residual concentration of the metal ion was measured to estimate the efficiency of removal. Fig. (2) shows the removal efficiency of copper as a function of time using three electrolysis voltages (5, 15, and 25 V). At the low time values, there was an appreciable improvement in the removal efficiency (41 to 59%) by the increase of voltage from 5 to 25 V, respectively. For the rest of the electrolysis time values, however, the electrolysis voltage does not seem critical for the removal of copper ions from water, although a slight increase is recorded. Thus, beyond one hour of electrolysis, the effect of voltage was minor on the overall removal efficiency.

Linear correlation can be observed for the increase of efficiency with voltage. The slope of the straight line decreases with the increase of electrolysis time, and rather, constant removal efficiency is established for the various voltages at the highest electrolysis time (150 min.).

For zinc removal, the results of the effect of time on the removal efficiency are given in Fig. (3) for various electrolysis voltages. There was no appreciable removal prior to one hr of electrolysis when the voltage was 5V. Higher electrolysis voltage resulted in good removal from the first 5 min of electrolysis. The removal efficiency exhibited a linear increase with the electrolysis time at 15 V. The efficiency increase with time at 25 V followed a polynomial relation. The higher voltage of electrolysis and, thus, the resultant current density will be higher. Such a condition improves the removal efficiency of Zn and Cu, as reported by Chen *et al.*, 2018 [30]. The difference in the behavior of zinc and copper lies in the electrochemical properties of the two metal ions in the solution.

The low voltage (5 V) did not cause any removal prior to one hour of electrolysis. Identical efficiency values were obtainable at 5 V after 30 and 60 min. The removal efficiency at the longest electrolysis time (150 min) at 5 V could be achieved after 60 min at 15 volts. The efficiency at 15 Volts after 2 hr was almost identical to that recorded after one hour of electrolysis at 25 volts. The removal efficiency linearly increases with the increase of the applied voltage for electrolysis times of 60-120 min. The increase follows a polynomial relation for 30 and 150 minutes of electrolysis time.

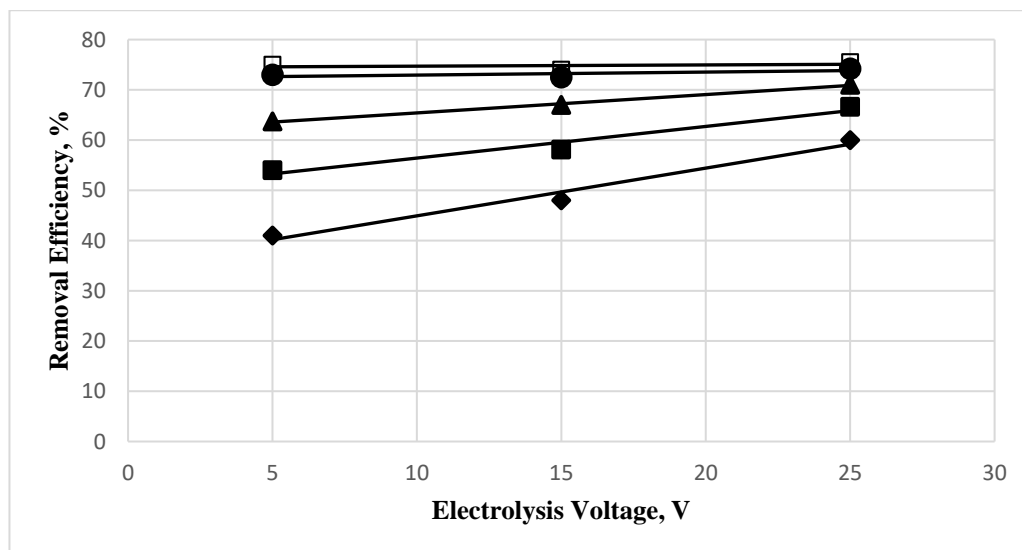


Fig. (2). Effect of electrolysis voltage on the removal efficiency of copper of 30 mg.L^{-1} initial concentration after electrolysis for 30, ♦, 60, ■, 90, ▲, 120, ●, and 150 min, □, at neutral medium.

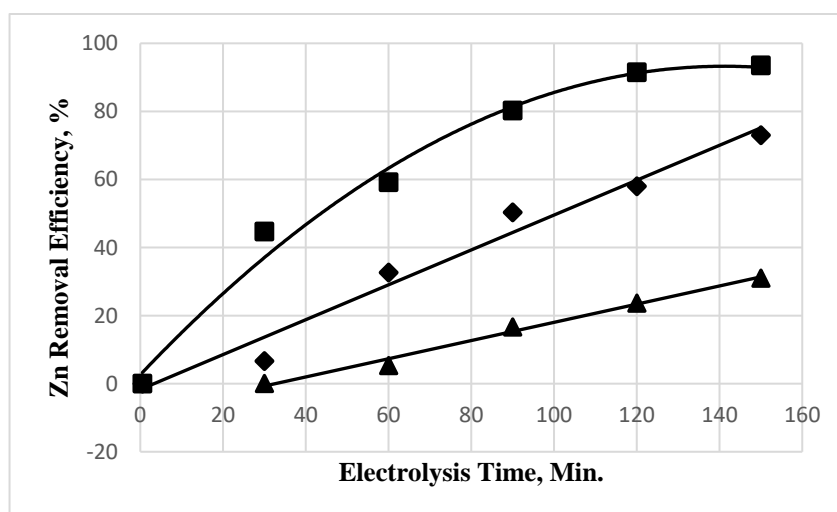


Fig. (3). Effect of electrolysis time on the removal efficiency of zinc of 30 mg.L^{-1} at electrolysis voltage of 5 V (▲); 15 V (♦) and 25 V (■), at neutral medium.

3.2. Effect of Electrolysis Time

Fig. (4) shows the dependence of copper removal efficiency of the electrolysis time at electrolysis voltage of 25 V using various initial zinc concentrations. The removal efficiency as a function of time followed a logarithmic relation. Within the first 5 minutes, almost 50% of the total removal was performed for 40 and 50 mg.L^{-1} . The rest of the removal proceeded linearly with the increase of time. However, the voltage was an essential factor in the determination of removal efficiency. The increase of voltage from 15 to 25 V resulted in an increase in efficiency by 20%. At the early stages of the electrolysis, the initial concentration plays an important role in the removal of copper ions.

For zinc removal ions from water, the efficiency increases

with time following a polynomial function for various initial concentrations (Fig. 5). At short electrolysis time (30 min), the initial concentration was very effective, and many differences in the removal efficiency were recorded, being 8% at 10 ppm and 60% at the 50 ppm-level. At longer electrolysis time, the efficiency attained values ranging from 93 to 97.5% for the 10 and 50 ppm initial concentrations, respectively. There is a general increase in efficiency with the increase of initial concentration, especially at short electrolysis time (30 and 60 min). The process showed perfection as the initial concentration increases. However, the removal efficiency at the long electrolysis time values (150 min.) was almost independent of the concentration, and attained values in the range 95 to 98% for the 10 and 50 mg.L^{-1} after two hours of electrolysis.

3.3. Effect of Initial Concentration

Fig. (6) shows the dependence of removal efficiency of copper on the initial concentration for various electrolysis times using electrolysis voltage of 25 V. At the early stages of the electrolysis, the initial concentration plays an important role in the removal of copper ions, when almost 50% of the total removal was performed within the first 5 minutes. At the low concentration range (10 and 20 mg.L⁻¹), the efficiency was

highly dependent on the electrolysis time being higher with longer electrolysis time. The 10 mg.L⁻¹ values of 16% were recorded for 30 minutes and 93% after 150 mins. Respectively. The effect was lower for the high initial concentration (50 mg.L⁻¹), being 59.4 and 97% for 30 and 150 mins, respectively. The efficiency was almost independent of the initial concentration when 150 min time was used for the process.

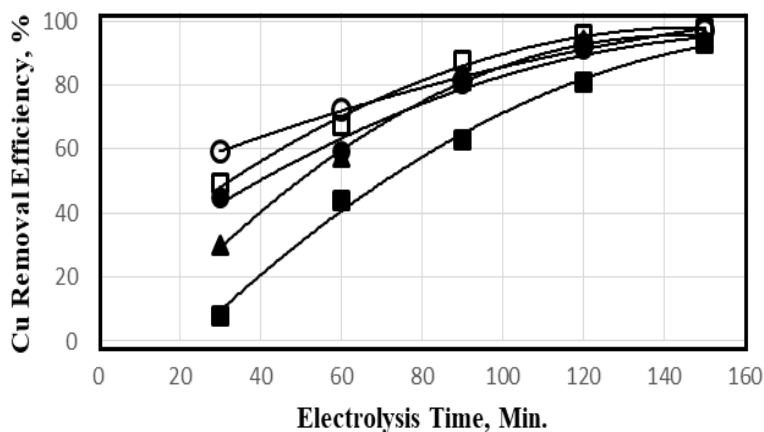


Fig. (4). Dependence of removal efficiency of Cu on the electrolysis time for various initial concentrations 10 (■), 20 (▲), 30 (●), 40 (◻), and 50 ppm (○) using a voltage of 25 V, at neutral medium.

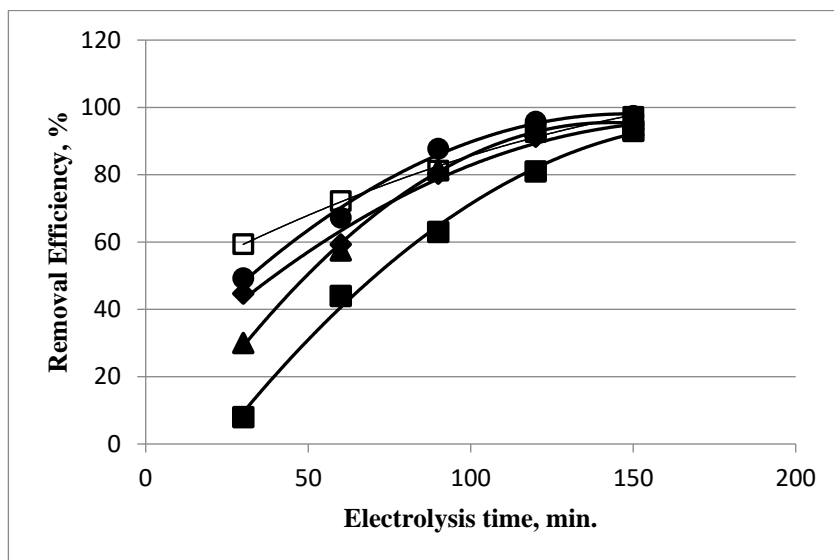


Fig. (5). Dependence of removal efficiency of Zn on the electrolysis time for various initial concentrations 10 (■), 20 (▲), 30 (◆), 40 (●), and 50 ppm (◻) using a voltage of 25 V.

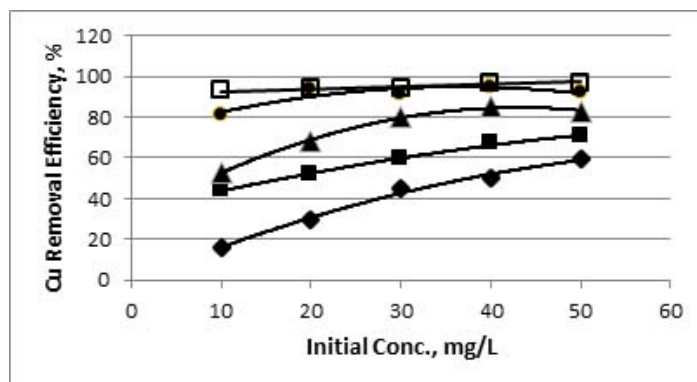


Fig. (6). The dependence of removal efficiency of copper on the initial concentrations for various electrolysis time at 25 V electrolysis voltage.

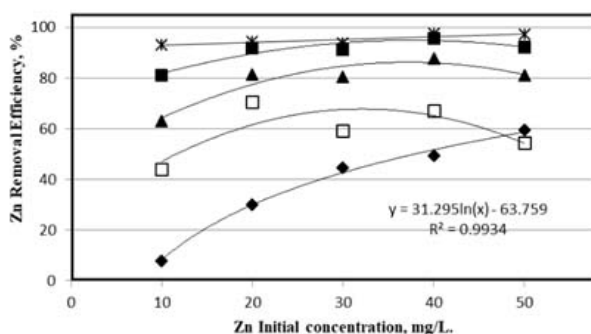


Fig. (7). Dependence of removal efficiency of zinc on the initial concentrations for various electrolysis time 30, ♦, 60, □, 90, ▲, 120, ■, and 150 min, x. The electrolysis voltage is 25 V, at a neutral medium.

Fig. (7) displays the dependence of removal efficiency of zinc with the initial concentrations for various electrolysis time at 25 V electrolysis voltage. At a short electrolysis duration (30 min), the initial concentration of zinc is highly effective on the removal efficiency. It follows a polynomial relationship with a reasonable correlation coefficient of ($R^2 = 0.9924$) as follows.

$$R_{\text{eff}} = -0.0132C^2 + 1.8623C - 1.2933 \quad (5)$$

Where R_{eff} is the removal efficiency and C is the initial concentration of zinc in the working solution. For other electrolysis time intervals, the effect of the concentration was minor, and the efficiency gets higher as the electrolysis time increases. The R_{eff} ranged from 93-97% for all concentrations when the electrolysis time was 150 min.

CONCLUSION

Electrocoagulation can be recommended as a simple and rather cheap method for the removal of heavy metals from water. The removal efficiency is dependent on the initial concentration only at short electrolysis times. Electrolysis voltages of 15 V and higher are preferred to give remarkable removal efficiency values regardless of the initial concentration for durations of 90 min and longer. At 25 V, the removal efficiency ranged from 93-97% for all concentrations when the electrolysis time was 150 min. Regarding the cost of the process, there are two main criteria for comparison of the

available method with the addition of alum to the water. The first is the omission of purchase of the alum. The second is the reduction in the equipment of the process.

CONSENT FOR PUBLICATION

Not applicable.

AVAILABILITY OF DATA AND MATERIALS

Not applicable.

FUNDING

None.

CONFLICT OF INTEREST

The authors declare no conflict of interest, financial or otherwise.

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