

## Removal of lead ions ( $Pb^{2+}$ ) from a synthetic wastewater by electrocoagulation using aluminum (Al) as a rotating electrode

Rawaa Zahd Jafat<sup>1</sup> Sata Kathum Ajjam<sup>2</sup>

<sup>1,2</sup>Chemical Engineering Department, University of Babylon, Babylon, Iraq.

**Abstract :** The performance of the lead electrocoagulation process was tested for five major factors that affected the process which were: different initial lead concentration (200,300,400,500)mg/different applied voltage(2.5,5,7.5,and10)V, and distance between the electrode (2.3,2.8)cm. different rotational velocity of anode (0, 50,100, and 150) rpm, variable pH (5, 7, 9, and 11) and time(5,10,15,20,25,30).The results showed that the removal rate of lead (removal efficiency) decreased with increasing concentration at concentrations (200, 300,400and 500) mg/l. Also removal efficiency, increased with increased applied voltage and reach to a maximum efficiency value at 10V, but decreased with increasing distance ,while rotating anode velocity doesn't fix that is a stat with increased with low velocities (0-50)rpm and reached to higher efficiency at (100)rpm, while at high velocity(150-200)rpm the removal efficiency start in decrease gradually due to the destabilization of flocks that is formed While pH shows peak performance curve. It shall be the highest removal efficiency rate of acidic (7). The optimum removal efficiency of 99 % was achieved at concentration 200mg/l and at a voltage of 10V with 2.3cm spacing between the electrodes and rotational velocity= 100rpm and pH = 7,using (Al/St. St. ) electrodes, within 20 min of operating time. The lead removal data has been used to find adsorption isotherm.

**Keywords:** Electrocoagulation, lead removal efficiency, Wastewater, rotating anode.

### Introduction

The major challenges for the 21st century are water and energy. Due to increased pollution from point and non-point sources quality of the water become a crucial problem, particular for the Third-World Countries<sup>1</sup>. Wastewater can be defined as the flow of used water discharged from homes, businesses, industries, commercial activities and institutions which are directed to treatment plants by a carefully designed and engineered network of pipes. There are many sources of water pollution, but two general categories exist: direct and indirect contaminant sources. Direct sources include effluent outfalls from industries, refineries, waste treatment plant, etc. Indirect sources include contaminants that enter water supply from soil/groundwater system and form atmosphere via rain water<sup>12</sup>. Due to the discharge of large amounts of metal-contaminated wastewater, industries bearing heavy metals, such as Cd, Cr, Cu, Ni, As, Pb, and Zn, are the most hazardous among the chemical intensive industries<sup>3</sup>. Heavy metals are elements having atomic weights between 63.5 and 200.6, and a specific gravity greater than 5.0. Heavy metals are the environmental priority pollutants and are becoming one of the most serious environmental problems. So these toxic heavy metals should be removed from the wastewater to protect the people and the environment<sup>4</sup>.Water pollution by heavy metal ions has become one of the world wide environmental problems due to population explosion, urbanization and industrialization<sup>5</sup>.

## Electrocoagulation Mechanism

Electrocoagulation involves three major mechanisms<sup>6,7</sup>

1. Formation of the coagulants, by dissolving electrically the consumable electrode.
2. These coagulant reagents destabilization of the contaminants, particulate suspension, and breaking of the emulsions.
3. The turbulence generated by the oxygen and the hydrogen evolution generates a soft mix that helps the destabilized colloids to flocculate (to link together and to generate bigger particles (flocks)).

The destabilization mechanism of the contaminant, particulate suspension, and breaking of emulsion may be summarized as follows<sup>6</sup>.

1. Compression of the diffuse double layer around the charged species by the interactions of ions generated by oxidation of the sacrificial anode.
2. Charge neutralization of the ionic species in the wastewater by counter ions produced by the electrochemical dissolution of the sacrificial anode. The counter ions reduce the electrostatic inter-particle repulsion to the extent that Van der Waals attraction predominates, thus causing coagulation. In this process a zero net charge results.
3. Flock formation: a sludge blanket is created from the flock that formed as a result of the coagulation process<sup>8</sup>.

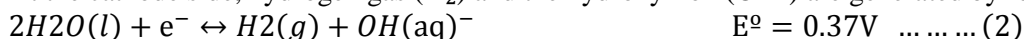
## Reactions that occurs at the electrodes surface

Electrocoagulation electrochemically introduces metal cations in situ, usually aluminium.<sup>9</sup>

At the anode the reaction is



At the cathode side, hydrogen gas (H<sub>2</sub>) and the hydroxyl ion (OH<sup>-</sup>) are generated by reducing the water.



## II.Experimental

### Electrolytic Cell Design (Electrochemical Cell)

The experiments were performed in transparent Pyrex glass of 1000ml in volume, the electrolytic cell consisting of the following parts. See Figure (1) for electro-coagulation process.

The detail of the electrochemical technique consists of two electrodes:

#### Counter Electrode (rotational anode)

The anode was a rotating cylinder made of aluminum .

#### Working Electrode (cathode)

The two cathode made of stainless steel was used.

#### Batch mode of electrocoagulation operations .

The associated schematic diagram of the equipment is shown in Figure 1

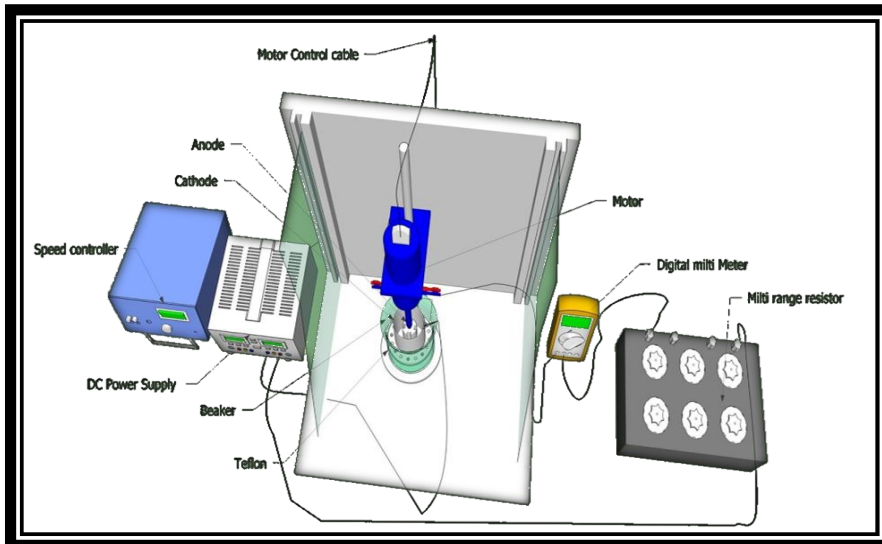


Fig.1: Schematic diagram of electrocoagulation (EC) rotating anode

### III.Results and Discussion

#### The effect of initial pH on the removal efficiency of Lead Ions

The pH of solution plays an important role in electrochemical and chemical coagulation process<sup>10</sup>. The influence of pH on electrocoagulation of lead is shown in figure (2– 5). The solutions were adjusted to the desired pH for all experiments using sulfuric acid or sodium hydroxide. Fig 6, this figure shows lead removal efficiency as a function of pH and various rotating velocity of anode, the maximum lead removal efficiency was obtained at optimum time of 20min, within pH of 7, and 100rpm, this it can be concluded that the majority of  $Al^{+3}$  coagulants are formed at this pH. So PH of 7 was used in other experiments. However, there is no significant effect of time after 20 min for pH of 7, so it is not feasible to use long time period of treatment because it leads to high power consumption. It can be also showed that for all concentration the rotational velocity not stable, and was seen that it is once low rotating velocity is best and once other that high velocity is preferred, and notes that 100rpm is the best velocity for all time, and all concentration.

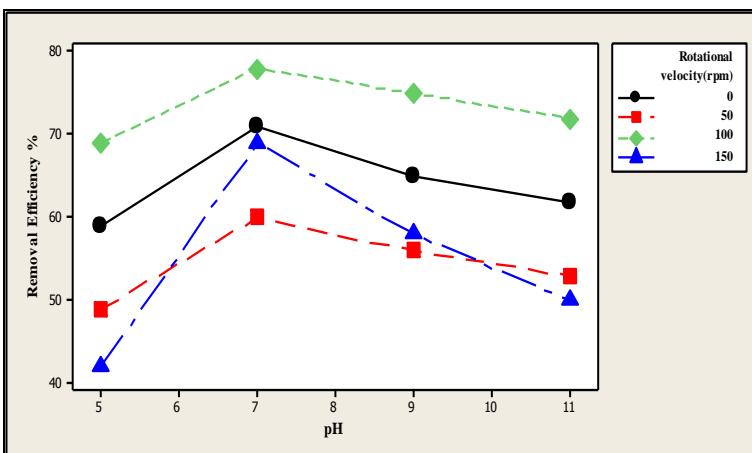


Fig 2: Effect of pH on the removal efficiency of lead at different Rotational (time, 5min)

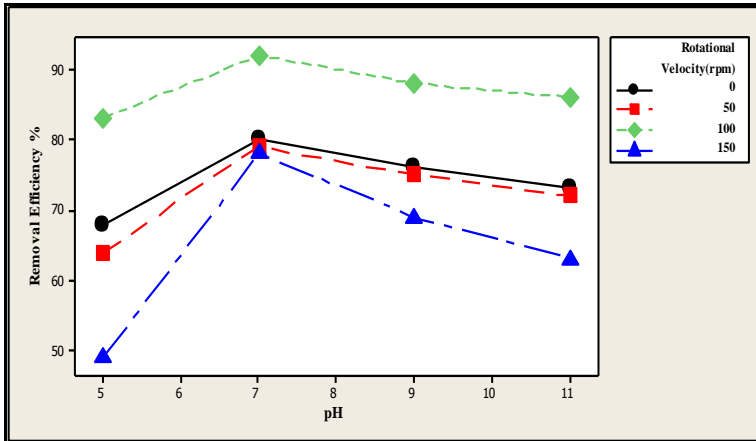


Fig 3: Effect of pH on the removal efficiency of lead at different Rotational velocity (time, 10min).

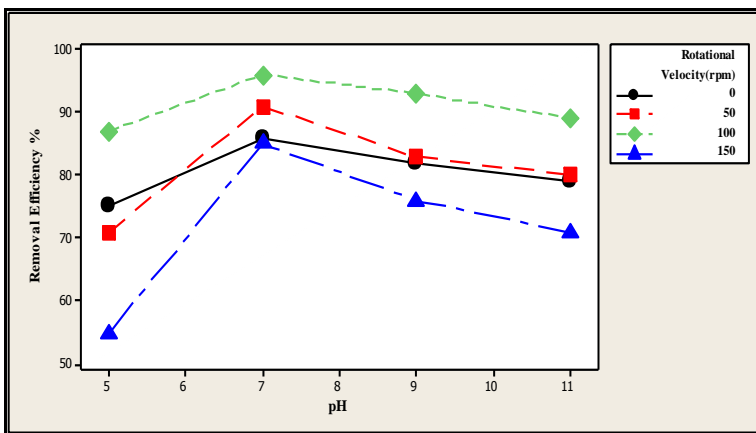


Fig 4: Effect of pH on the removal efficiency of lead at different Rotational velocity (time, 15min).

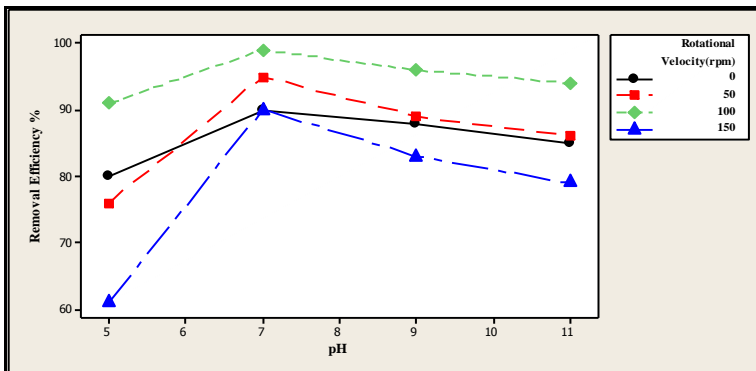
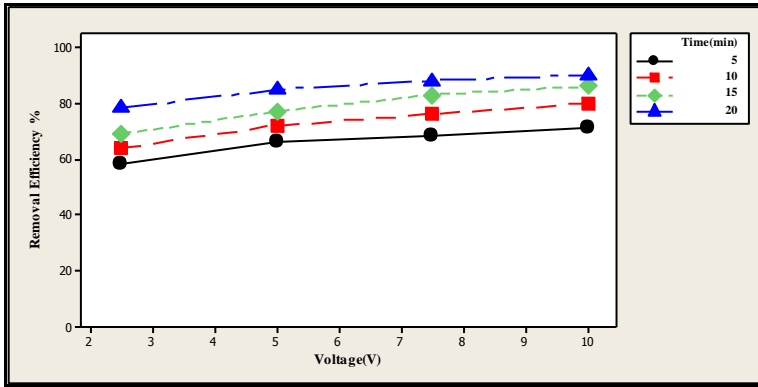


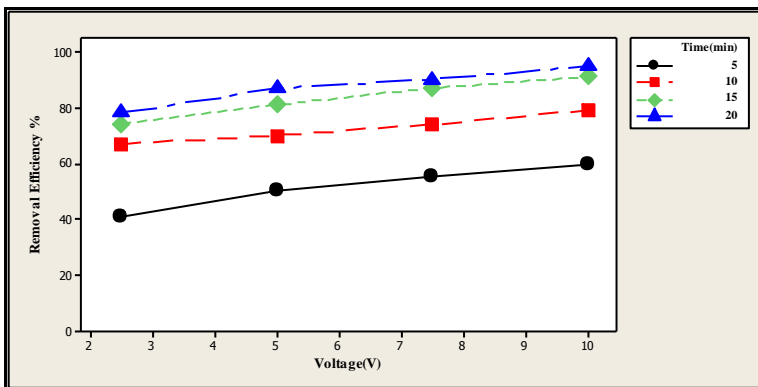
Fig 5: Effect of pH on the removal efficiency of lead at different Rotational velocity (time, 20min)

**The effect of applied voltage on the removal efficiency of lead ions**

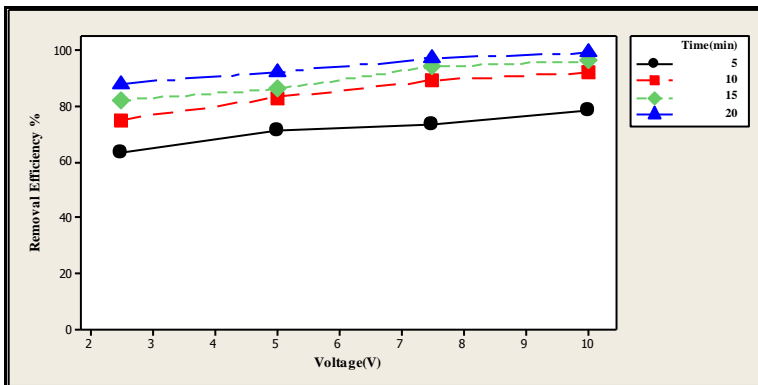
The influence of applied voltage on the removal efficiency of lead is shown in figure [ 6 to 9] for 200mg/l. The Current density increase with the voltage increase, Current density directly determines both coagulant dosage and bubble generation rates and strongly influences both solution mixing and mass transfer at the electrodes. From Fig.8, it can be seen that removals are increased when voltages applied increased; this is attributed with research of<sup>11</sup> that reported increase the voltage may increase the removal of heavy metal. When applied potential rate is increased from (7.5- 10)volt, lead removal efficiency increased from (97 - 99)% and then decreased from (5 - 2.5)volt about (92 - 88) % ,of the optimum value is 10Volt.



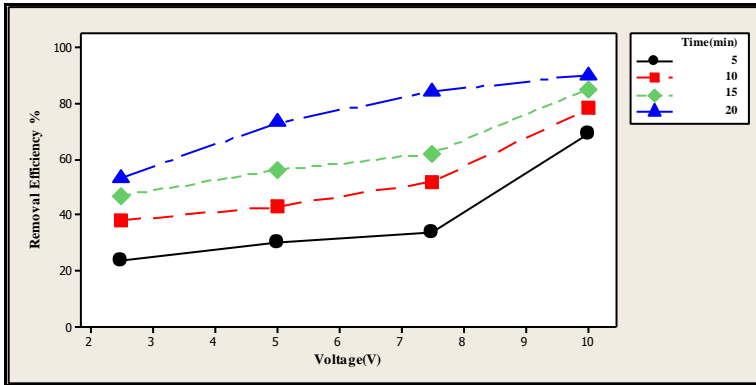
**Fig 6 :**Effect of applied voltage on the removal efficiency of lead at different time (rotating anode velocity, 0rpm).



**Fig 7:** Effect of applied voltage on the removal efficiency of lead at different time (rotating anode velocity, 50rpm).



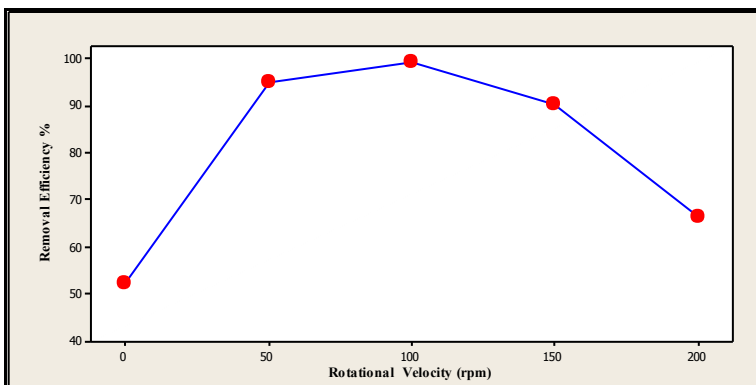
**Fig 8 :**Effect of applied voltage on the removal efficiency of lead at different time (rotating anode velocity,100rpm).



**Fig 9 :**Effect of applied voltage on the removal efficiency of lead at different time (rotating anode velocity,150rpm).

### The effect of the rotational anode velocity on the removal efficiency of lead ions

The mixing speed is an important operating factor influencing the performance of electrocoagulation process. To examine its effect on the removal of lead, the rotating anode speed was varied in the range of (0–200) rpm, were studied at a constant voltage for 10V for pH 7 and 2.3cm , different time .The influence of rotating anode velocity on the removal efficiency of lead is shown in Fig.10.



**Fig 10.** Effect of rotating velocity on removal efficiency of lead ion.

It is noted from this figure that increasing rotating anode velocity of the solution (0-50) rpm leads to the removal efficiency increased this speed did not supply a homogeneous mixture in the cell .And shows a maximum for rotating anode speed (50-100) rpm and then it decreases smoothly with increasing rotating anode velocity (100-150) rpm, coagulant matter formed of aluminum ions, attached together and disperse in the cell making the content of the cell homogenous. However, decrease in the efficiency at higher stirring rate (200) rpm may be due to the destabilization of flocks formed in the cell. So the best rotating anode velocity was obtained at 100 rpm. Graphical results shown the best rotating velocity is 100rpm for all concentrations, and this also showed that the flocks deposited between electrodes, because the flocks couldn't mix homogeneously and this deposition caused to the increment of cell resistance at low stirring speed. The increase in the cell resistance causes the increase of potential value in the systems where constant current density and this causes the increase of the amount of energy consumption per unit volume. The increased in rotating speed from (0-50-100), this confirms the fact that the removal efficiency is diffusion controlled, and the increase in stirring speed leads to increase in the intensity of turbulence and reduces the diffusion layer thickness at the electrode surface and Improves the mixing conditions in the electrolyte bulk ,This is attributed with the work of<sup>12</sup>.

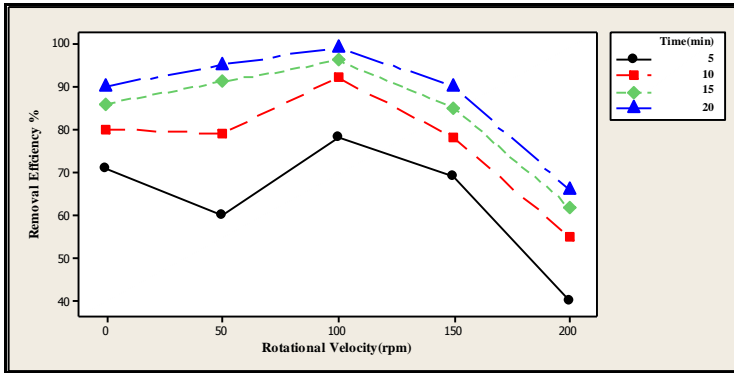


Fig 11: Effect of rotating anode velocity on the removal efficiency of lead with different times

**The effect of initial lead ions concentration on the removal efficiency**

The effect of initial concentration on electrocoagulation of lead ions Shown graphically in Fig.12 represented optimum condition for concentration effect. It is clear from this Fig, higher concentrations greater time is needed for the removal of lead ions at different rotational anode velocity, but higher initial concentrations of lead were reduced significantly in relatively less time compared to lower concentrations. This can be explained as follows: (1) from faraday’s law ( $W=I \cdot e \cdot t / n \cdot F$ ), where a constant amount of  $Al^{+3}$  passed into solution at the same current density and time for all lead concentrations,  $Al^{+3}$  was insufficient for solutions including higher lead concentration ,therefore the same amount of flocks would be produced in the solution.(2) Although the same amount of the coagulant produced in the electrocoagulation cell at the same current density for different lead concentrations, this amount of coagulant species. This result came in attributed with other researchers<sup>13</sup>, because of increasing lead concentration, the applied potential for the solution and the consumed energy was decreased as well.

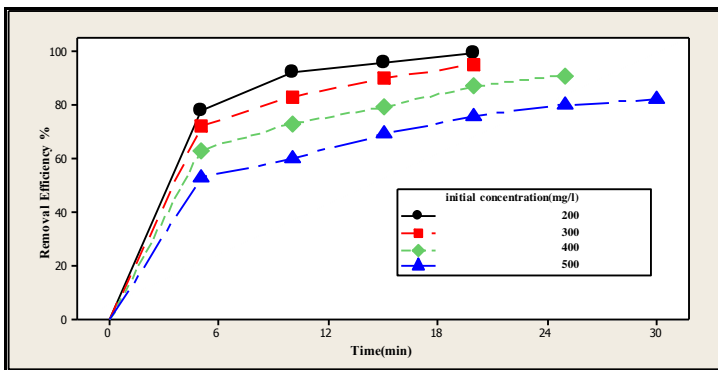


Fig 12 :Effect of initial concentration on removal efficiency of lead (rotating anode velocity, 100rpm)

**The effect of distance between electrodes on the RE (removal efficiency)**

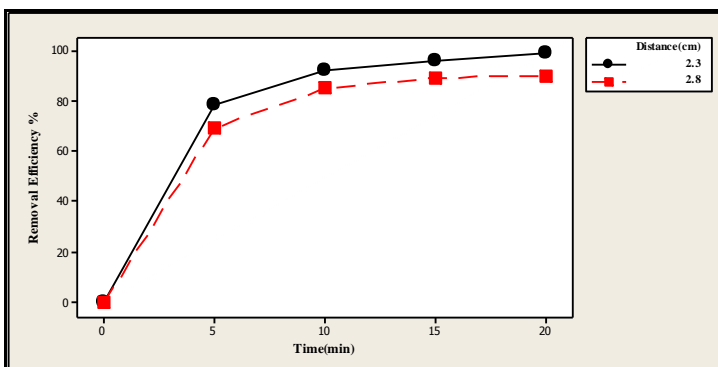


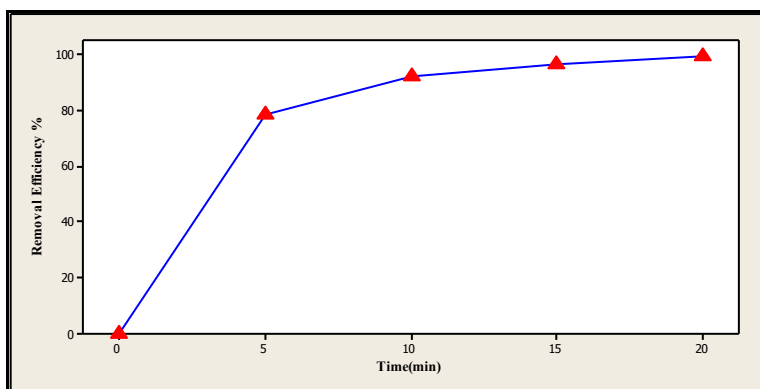
Fig 13: Effect of the distance on removal efficiency of at different time (rotating anode velocity=100rpm).

Fig.13 represented the preferred operating condition (100rpm, 200 mg/l, pH 7,and 10volt of applied voltage) for the effect of distance.

In the EC operation, the ions produced from the cathode and anode collects to form flocks. The travelling time of the participating ions in the EC reaction decreases with a decrease in the inter electrode distance, resulting in a higher removal efficiency for a lower duration electrolysis<sup>14</sup>.According to ohm’s law, the value of the electric current during a metal conductor in a circuit is directly proportional to the voltage impressed across it, for any given temperature.

**The effect of treatment time**

It was confirmed that the best condition for lead removal was at the sample original pH 7.0 with voltage 10volt , initial lead ions concentration 200 mg/l with distance between electrodes 2.3 cm and rotating anode velocity 100rpm form (5-20) min of operating time, Fig .14 illustrate Pb<sup>+2</sup> removal efficiency versus time for these conditions. It shows that when treatment time is increased, removal efficiency also increases.



**Fig 14 :Effect of time on the removal efficiency of lead.**

**Studies of Adsorption isotherms modeling**

The effect of pH, on the adsorption of lead ions by electrocoagulation technique. This was done at pH values, (5-11), for a particular study, a parameter was varied while the others were kept constant.

**Adsorption dependence on PH**

**Table (1) Isotherm Constant and correlation coefficients at different pH for lead ion removal.**

| pH | Langmuir Isotherm |              |        | Freundlich Isotherm     |      |        |
|----|-------------------|--------------|--------|-------------------------|------|--------|
|    | $Q_m$ (mg/g)      | $k_L$ (L/mg) | $R^2$  | $K_F(mg/g)(L/mg)^{1/n}$ | n    | $R^2$  |
| 5  | 312.5             | 0.0095       | 0.9607 | 10.8                    | 1.76 | 0.9472 |
| 7  | 769.39            | 0.0016       | 0.9345 | 1.303                   | 1.02 | 1      |
| 9  | 416.6             | 0.0202       | 0.9345 | 22.9                    | 1.9  | 0.9793 |
| 11 | 175.43            | 0.0867       | 0.9913 | 3.109                   | 1.28 | 0.9552 |

The  $R^2$  values indicating that the experimental data can be better explained by the Freundlich Isotherm than the langmuir. The  $K_F$  value as calculated from the Freundlich isotherm.Table (2) shows the  $R_L$  values against the initial lead concentration.

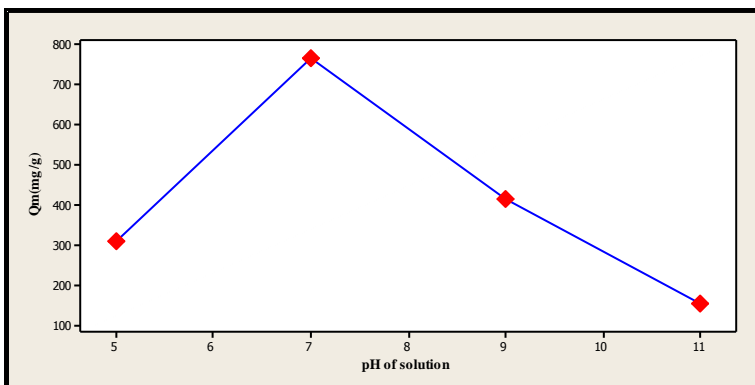


**Table (2) Essential Characteristics of Langmuir Isotherm at different pH.**

| Co(mg/l) | $R_L$  |        |        |         |
|----------|--------|--------|--------|---------|
|          | pH     |        |        |         |
|          | 5      | 7      | 9      | 11      |
| 200      | 0.3448 | 0.7496 | 0.1984 | 0.0545  |
| 300      | 0.2597 | 0.6662 | 0.1416 | 0.0370  |
| 400      | 0.2083 | 0.5995 | 0.1101 | 0.0280  |
| 500      | 0.1739 | 0.5449 | 0.0900 | 0.02254 |

$R_L$  values between 0 and 1 indicate that the sorption of lead ion is highly favorable, moreover, the sorption is favorable for both low and high initial Concentration as the values of  $R_L$  are very close to zero.

The initial pH of a solution is a very important factor to be considered in adsorption studies as it has been observed to play a major role in the adsorption of metal ions by various adsorbents<sup>15</sup>. The result illustrating the dependence of pH on adsorption of lead is presented in Fig.14. Maximum adsorption was obtained in the pH range of (5-11) and its increased as pH values increased from 2 to 7. Indeed, the Maximum adsorption,  $q_{max}$ , of Pb (II) ions was attained at a pH value of 7. Indicating very strong capacity for lead ions removal at pH equal to 7 that is alkali solution as compared to 9 and 11. This can be explained that at low pH values the solution is highly acidic. This led to the competition between lead ions and protons for the active sites resulting in a low adsorption capacity. Subsequently, as the pH of the solution increased, the number of protons decreased which reduced the competition between lead ions and protons for the active sites, hence more lead ions were adsorbed from solution<sup>16-25</sup>.

**Fig.15: Effect of pH on adsorption of lead (rotating velocity, 100rpm, applied voltage, 10volt).**

## V. conclusion

Experiments have been carried out to determine the best operating conditions

1. The effect of rotating velocity on lead removal also was studied and the result showed that the removal efficiency decreased when the rotating speed is high
2. In general, high applied voltage and high bath concentration, achieve the lead electrocoagulation efficiency. Because consumable of energy in high voltage and high running time is needed in high concentration.
3. The effect of distance on removal efficiency of lead was studied and the result Reported that the removal efficiency decreased when increased in the distance.

## VI- References

1. Chaturvedi, S. I., "a Novel Waste Water Treatment Method", International Journal of Modern Engineering Research (IJMER), Vol.3, Issue.1, Jan-Feb. PP.93-100, 2013.
2. Rao.D.G and Anthony Byrne .J. "Wastewater Treatment: Advanced Processes and Technologies", CRC Press co-published series, PP. 1-10, 2012.

3. Barakat, M.A., "New Trends in Removing Heavy Metals from Industrial Wastewater", *Arabian Journal of Chemistry*, Vol. 4, PP. 361–377, 2011.
4. Fu, F. and Q. W., "Removal of Heavy Metal Ions from Wastewaters: a Review", *Journal of Environmental Management*, Vol. 92, PP. 407-418, 2011.
5. Nwabanne, J. T., and Anthony C. Okoye," Treatment of Synthetic and Battery Industry Wastewater by Electrocoagulation", *Pelagia Research Library,Der Chemica Sinica*, Vol.6,No.4,PP.32-39,2013.
6. Essadki, A. H., "Electrochemical Probe for Frictional Force and Bubble Measurements in Gas-Liquid-Solid Contactors and Innovative Electrochemical Reactors for Electrocoagulation /Electroflotation", In: Y. Shao, ed. *Electrochemical Cells – New Advances in Fundamental Researches and Hazardous Materials*, No.1,Vol.158, pp.65-72, 2008.
7. Alkaim AF, Alqaragully MB. Adsorption of basic yellow dye from aqueous solutions by Activated carbon derived from waste apricot stones (ASAC): Equilibrium, and thermodynamic aspects. *Int. J. Chem. Sc.* 2013; 11(2): 797-814.
8. Davarnejad R, Panahi P. Cu (II) removal from aqueous wastewaters by adsorption on the modified Henna with Fe<sub>3</sub>O<sub>4</sub> nanoparticles using response surface methodology. *Separation and Purification Technology*. 2016; 158: 286-292.
9. Hu QH, Qiao SZ, Haghseresht F, Wilson MA, Lu GQ. Adsorption Study for Removal of Basic Red Dye Using Bentonite. *Industrial & Engineering Chemistry Research*. 2005; 45(2): 733-738.
10. Ghaedi M, Sadeghian B, Pebdani AA, Sahraei R, Daneshfar A, D, C. Kinetics, thermodynamics and equilibrium evaluation of direct yellow 12 removal by adsorption onto silver nanoparticles loaded activated carbon. *Chem. Eng. J.* 2012; 187: 133-141.
11. Zhu J, Lou Z, Liu Y, Fu R, Baig SA, Xu X. Adsorption behavior and removal mechanism of arsenic on graphene modified by iron-manganese binary oxide (FeMnOx/RGO) from aqueous solutions. *RSC Advances*. 2015; 5(83): 67951-67961.
12. Rioja N, Benguria P, Peñas FJ, Zorita S. Competitive removal of pharmaceuticals from environmental waters by adsorption and photocatalytic degradation. *Environmental Science and Pollution Research*. 2014; 21(19): 11168-11177.
13. Yilmaz, A.E., Boncukcuoglu, R., Kocakerim, M.M. and Keskinler, B., "The investigation of parameters affecting boron removal by electrocoagulation method ",*journal of hazardous materials* Vol.125,No.1, pp.160-165, 2005
14. Ahmed A.Mohammed and Muhanned Dhia Fadhil Al-Mureeb., "Removal of Lead from Simulated Wastewater by Electrocoagulation Method", *Journal of Engineering*, University of Baghdad, Vol. 16, No. 4,December ,pp. 5811-5821, 2010.
15. Anayur R.A., Sari A. and Tuzen .,"Equilibrium, thermodynamic and Kinetic Studies on Biosorption of pb and cd From Aqueous Solution by Mcrofungus (lactarius Scrobiculatus) Biomass", *chemecal. Engineering Journal*,151,pp.225-261, 2009.
16. Vimala, R. and Das, N., "Biosorption of cadmium (II) and lead (II) from aqueous solutions using mushrooms: a comparative study", *Journal of Hazardous Materials*, 168,No.1, pp.376-382, 2009.
17. Salman JM, Abdul-Adel E, Alkaim AF. Effect of pesticide glyphosate on some biochemical features in cyanophyta algae *oscillatoria limnetica*. *International Journal of PharmTech Research*. 2016; 9: 355-365.
18. Raheem RA, Al-gubury HY, Aljeboree AM, Alkaim AF. Photocatalytic degradation of reactive green dye by using Zinc oxide. *journal of Chemical and Pharmaceutical Science*. 2016; 9: 1134-1138.
19. Omran AR, Baiee MA, Juda SA, Salman JM, Alkaim AF. Removal of Congo red dye from aqueous solution using a new adsorbent surface developed from aquatic plant (*Phragmites australis*). *International Journal of ChemTech Research*. 2016; 9: 334-342.
20. Kamil AM, Mohammed HT, Alkaim AF, H. HF. Adsorption of Congo Red on Multiwall Carbon Nanotubes: Equilibrium isotherm and kinetic studies. *International journal of chemical sciences*. 2016; 13: 1657-1669
21. Alkaim AF, Sadik Z, Mahdi DK, et al. Preparation, structure and adsorption properties of synthesized multiwall carbon nanotubes for highly effective removal of maxilon blue dye. *Korean J. Chem. Eng.* 2015; 32(12): 2456-2462.
22. Aljeboree AM, Alkaim AF, Al-Dujaili AH. Adsorption isotherm, kinetic modeling and thermodynamics of crystal violet dye on coconut husk-based activated carbon. *Desalin. Water Treat.* 2015; 53(13): 3656-3667.

23. Kamil AM, Mohammed HT, Alkaim AF, H. HF. Adsorption of Congo Red on Multiwall Carbon Nanotubes: Equilibrium isotherm and kinetic studies. International journal of chemical sciences. 2016; 13(3): 1657-1669.
24. Aljeboree AM. Adsorption of crystal violet dye by Fugas Sawdust from aqueous solution. International Journal of ChemTech Research. 2016; 9(3): 412-423.
25. Aljeboree AM. Adsorption of methylene blue dye by using modified Fe/Attapulgitic clay. Research Journal of Pharmaceutical, Biological and Chemical Sciences 2015; 6(4): 778-788.

\*\*\*\*\*