Efficient Removal Of Bromate From Aqueous Solution Through Electrocoagualtion Using Aluminum Electrodes

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Abstract
In this study the removal efficiency of bromate from aqueous solution using the electrocoagulation process was investigated. The effects of operational parameters such as initial pH, initial bromate concentration, current density, type electrode, salt concentration, and temperature on the removal efficiency have been studied. With an initial pH of 3.7, an initial bromate concentration of 100 mg/L, (current density 22.7 mA/cm2 using Al electrodes, salt concentration of 1.5 g/L NaCl and temperature of 25° C, The results showed that the removal percentage for bromate was 97% using Al/Al electrodes at 10 min. It can be concluded that electrocoagulation process by Al electrode is very efficient and clean process for bromate removal from Aqueous solution.

***Keywords**: Electrocoagulation; Aluminum; Iron; bromate; Pesticide and Aqueous solution.*

I. INTRODUCTION

Water is an important and essential component of this universe and plays a vital role in the proper functioning of the Earth's ecosystems. In spite of this, safe drinking water is not available in some parts of the world. The quality of water resources is deteriorating exponentially because of their contamination. Both point and non-point sources are polluting our water resources because of tremendous population growth, modern industrialization, civilization, domestic and agricultural activities and other geological, environmental and global changes. Nowadays, water pollution is a serious issue because it affects our lives [1] With respect to human health, the most direct and most severe impact is the lack of improved sanitation, and related to it is the lack of safe drinking water, which currently affects more than a third of the people in the world. Additional threats include, for example, exposure to pathogens or to chemical toxicants via the food chain (e.g., the result of irrigating plants with contaminated water and of bioaccumulation of toxic chemicals by aquatic organisms, including seafood and fish) or during recreation (e.g., swimming in polluted surface water).The water treatment and wastewater a necessary for human survival, health and prosperity. Almost 900 million people lack access to an improved water supply and 2.6 billion to basic sanitation. WHO and UNICEF consider improved water supplies to be those technologies that provide water of an acceptable quality and quantity [2] According to the World Health Organization, more than 1.2 billion people lack access to clean water, and more than 5 million people die every year from contaminated water or water-related diseases [3]

So , reducing untreated wastewater and protecting aquatic ecosystems are targets of the Sustainable Development Goals set by the United Nations to be reached by 2030[4]Potassium bromate ($KBrO_3$), a white crystalline solid and a widely reactive food additive [5], it is often used in bakeries as flour improver yielding higher bread volume and used as a dough conditioner for flour [6-7] . On other hand bromate is generated as a contaminant in drinking water due to conversion of bromide found naturally in water to bromate by ozone which is used as disinfectant [8]. Studies have also shown that it possessed the potential of inducing cancer, kidney failure, deafness, redness and pains of the eye and skin [9].In the United States and the European Union (EU), the maximum acceptable contaminant level of $BrO3^-$ is 10 µg/L (Haag &

Holgné, 1983) and the LD50 values were higher for females than for males in all species and ranged from 280 mg/kg (male mice) to 495 mg/kg (female rats) [10-11].In recent years, novel methods for water purification have been developed including chemical, electrochemical and photochemical processes [12,13]. Electrochemical processes (electrolysis and electrocoagulation) have been successfully demonstrated for removing pollutants in various industrial wastewaters [14,15]. In this scenario, the electrocoagulation (EC) is an electrochemical has attracted increasing interest as a promising powerful method for efficiently removing of different types of pollutants from water [16]. The aim of this study is to conduct an experimental investigation on the removal of a bromate from the aqueous solution using the electrocoagulation method.

II. MATERIALS AND METHODS

Chemicals

Bromate representative pollutant was selected for the synthetic pollutant aqueous solution where the manufacture of the Bromate (Manufacturer company: Mkhchim), were purchased from (Company Abu Halima - Gaza) commercial.Sodium chloride (NaCl), potassium chloride (KCl), sodium sulfate (Na2SO4), sodium carbonate (Na_2CO_3), potassium iodide (KI), calcium chloride ($CaCl_2$), sodium fluoride (NaF), sodium hydroxide (NaOH) and sulfuric acid (H_2SO_4), were of analytical grade and purchased from Merck. Distilled water was used for the preparation of solutions.. Electrodes design is one of the most important factors that affect the electrocoagulation process. Electrode design affects the release of coagulants in the solution and the bubble type. Thereby influencing pollutant flotation, mixing and mass transfer. In this study iron , aluminum and stainless steel (304 L grade) electrodes were used as anode and cathode respectively.

Equipment's and procedures

Stock solution of Bromate (1000 mg/L) were prepared by dissolving an accurate quantity of each Bromate in distilled water and suitably diluted to the required initial concentrations. The amount of Bromate required to prepare the stock solution was calculated using the following equation:

$M1 \times V1 = M2 \times V2$ (1)

The electrocoagulation unit consists of an 100 ml electrochemical reactor with an effective surface area of 4 cm^2 . The electrodes were 2 cm x 2 cm × 0.2 cm. The electrode were positioned vertically and parallel to each other using aluminum or iron or stainless steel electrode as anode or cathode. The anode is connected to the positive pole and the cathode to the negative pole of the direct current power supply show figure1.

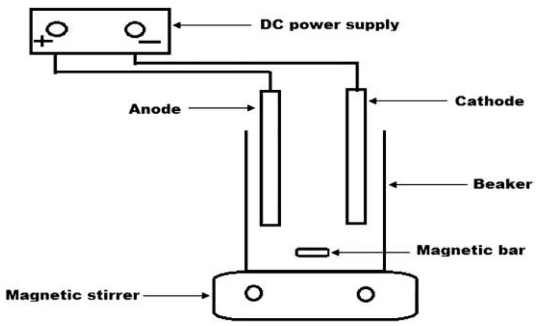

Fig 1. Experimental setup included magnetic stirrer, magnetic bar, beaker, cathode, anode and DC power supply.

The electrodes were positioned vertically and parallel to each other. The current density was maintained constant by means of a precision DC power supply; model (DZ040019) EZ Digital CO. Ltd. (Korea). The pesticide concentration was determined using a double - beam UV-Vis spectrophotometer, model UV 1601 is from Shimadzu (Japan) at 284 nm. Hot Plate, model (HB502), Bibby Sterilin Ltd (UK). A pH meter model AC28, TOA electronics Ltd., (Japan). Water bath model SB-650, Tokyo Kikakkai CO. Ltd. (Japan).

III. ANALYSIS

Two main parameter were measured to evaluate the electrochemical treatment efficiency, the remaining pollutant concentration (mg/L) and the COD (mg O2/L). Remaining pollutants of (Bromate) concentration were measured with the double-beam UV-Visible spectrophotometer at λmax= 284 nm, and for Bromate, using calibration curve with standard error ± 0.5%. The equation used to calculate the pollutants removal efficiency in the treatment experiments was:

$$\%E = [(Ao - A) / Ao] \times 100 \quad (3.2)$$

Where Ao and A are absorbance values of pollutant solutions before and after treatment with respect to their λmax [17]

Effect of the conductivity (NaCl dose):

In general NaCl is used to obtain conductivity in electrocoagulation process. The conductivity of the bromate solutions are adjusted to the desired levels by adding an appropriate amount of NaCl [18]. The effect of NaCl concentration on the removal % of the bromate is shown in Fig.2

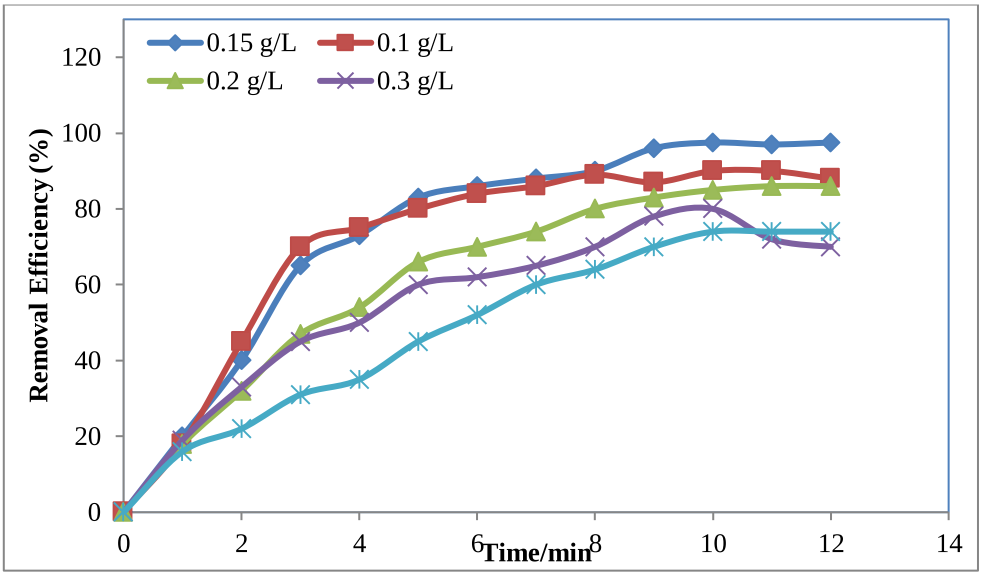

Fig 2. Effect of NaCl concentration change on the removal %

It can be seen from Fig. 2 that there is an increase in the removal % of the bromate up to 97% when the concentration of NaCl salt in the solution is 0.15 g/L. It was found that raising the conductivity of the solution has not a considerable effect on the removal % but it decreases the energy consumption. According to the obtained results, high removal% with low cell voltages and low energy consumption can be obtained in bromate solutions with NaCl of around 0.15 g/L. In this respect 0.15 g/L was used in the all following experiments.When the concentration of NaCl salt in solution increases, conductivity of the solution and the current density increase. The higher ionic strength will generally cause an increase in current density at the same cell voltage or, equivalently, the cell voltage decreases with increasing wastewater conductivity at constant current density [19]. This lead to products Cl_2 and OCl^- from anodic discharge when chlorides are present in the solution. So, added NaCl not only increases the conductivity but also contributes strong oxidizing agents [20]

Effect of pH on the bromate removal%

Fig.3 shows the effect of initial pH value on the removal % of the bromate under the following conditions: 200 ppm initial concentration of the bromate, current density of 22.7 mA/cm^2 , electrolysis time of 10 min., temperature of 25˚C. As can be seen from the plot of Fig. 3 that, there was a significant difference in the removal % of bromate when using Al-Al electrodes at different initial pH values. It seemed that the optimal pH value was ranged from 7-11. The $Al(OH)_{n(S)}$ formed in electrocoagulation remains in the aqueous stream as a gelatinous suspension at pH 7-11 , which can remove the pollutants from the water either by complexation or by electrostatic attraction, followed by coagulation [21].

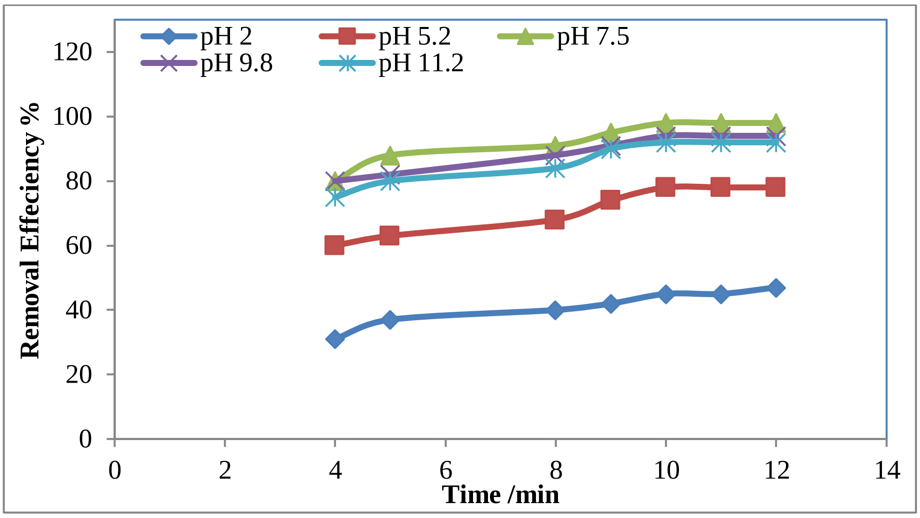

Fig 3. Effect of pH value change, keeping other variants constant

Effect of current density change

The current density determines the coagulant dosage rate. This parameter should have a significant impact on the removal efficiency of the bromate. The influence of the current density on the pesticides removal % during the electrolysis with the Al-Al electrodes is reported in Fig. 4.

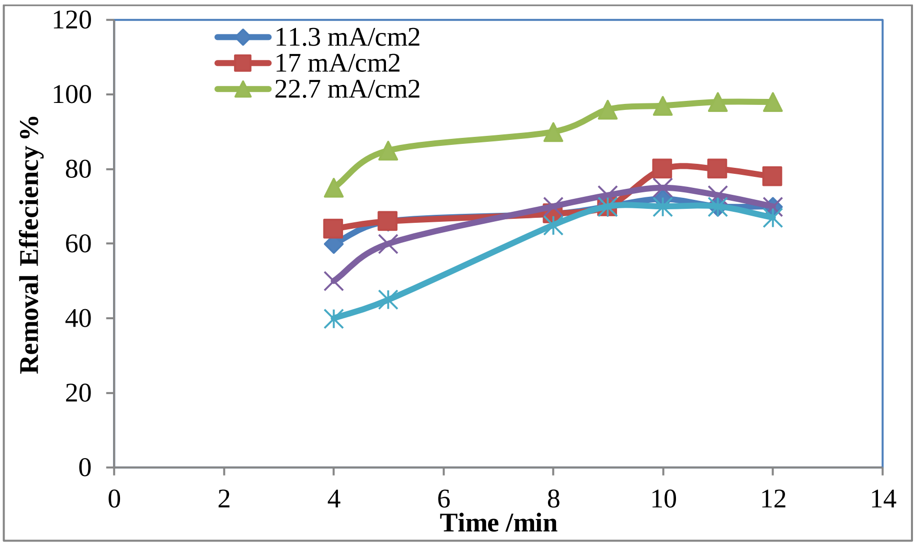

Fig 4. Effect of current density change on the removal %

The removal of the bromate at higher current densities than 22.7 mA/cm2 decrease. At a high current density, the extent of anodic dissolution of aluminum increases, resulting in a greater amount of Al $(OH)_n$(s). Moreover, the rate of bubble- generation increases and the bubble size decreases with increasing current density, both of these trends are beneficial in terms of a high-pollutant removal efficiency by H2 flotation [22].

Effect of electrolysis time

Electrolysis time determines the production rate of coagulant . Fig. 5 show the effect of electrolysis time on the removal % of the pesticides under the following operating conditions: 200 ppm initial dose of bromates, pH of 7-11, current density of 22.7 mA/cm^2 , temperature of 25˚C. It is indicated from the plot of Fig. 5 that the removal % increases with the increase of electrolysis time up to 10 minutes. No further increase in the removal % with the increase of the electrolysis time more than 10 minutes.

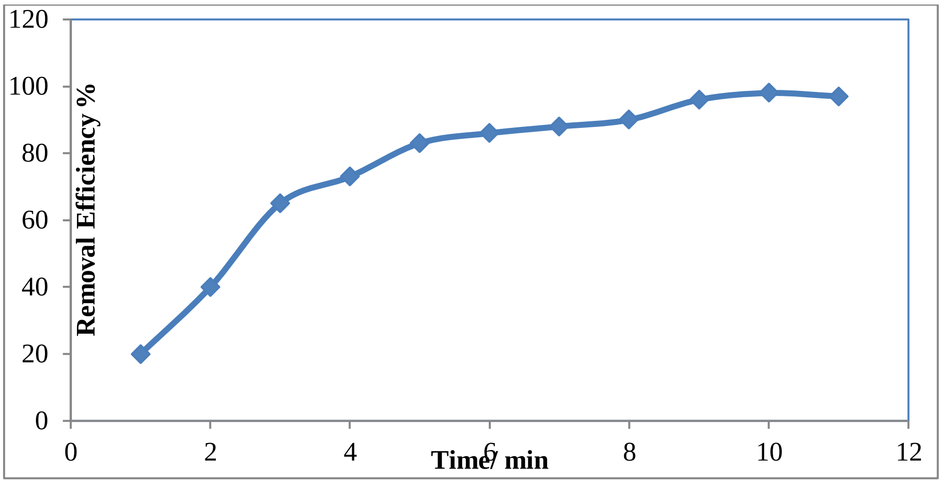

Fig 5. Effect of time electrrolysis change on the removal %

Effect type of electrolyte

Fig. 6 explain the effect of electrolyte types on the removal efficiencies of bromate .of. Electrolytes of 1.5 g/L of the following salts: NaCl, KCl, Na_3PO_4, NaF and Na_2SO_4 were studies by three electrodes. According to Fig 6. The bromate removal at NaCl, KCl, NaF were the most effective conductive electrolytes for the EC while Na_3PO_4, and Na_2SO_4 electrolytes show poor results**.**

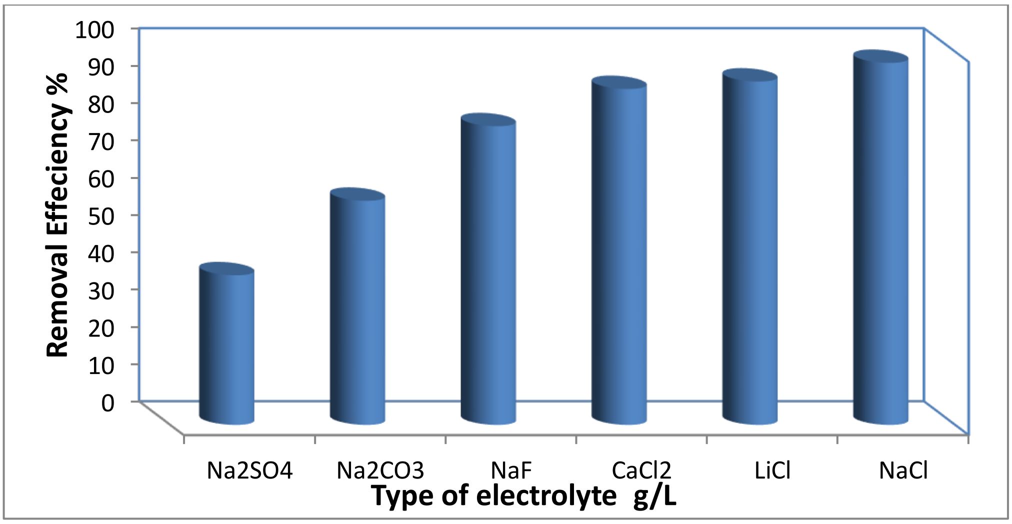

Fig 6. Effect of type of electrolyte change on the removal %

It can be seen from Figs. 6. that in the presence of chloride ions of NaCl, KCl, and $CaCl_2$ (which provide the effective Cl^-ion) electrolytes the removal efficiency of bromate was high due to formation of hypochlorite (OCl^-) and hypochlorous acid [23].

Effect of initial bromate dose

A series of batch experiments with different initial bromates concentration were carried out to study the effect of initial dose on the removal % during the electrocoagulation process, different initial concentrations in the range of 100–2000 mg/L were treated using (SS) and (Fe) electrodes in the optimum condition. The plot of Fig. 7 indicated that the removal decreased at high initial concentrations of the bromate. This is possibly due to the formation of insufficient number of aluminum hydroxide complexes by the electrode for a given conductivity and applied cell voltage to coagulate the excessive number of bromate molecules at higher concentration. It is therefore, quite clear that under the present experimental conditions the lower is the bromate concentration, the better is the removal efficiency.

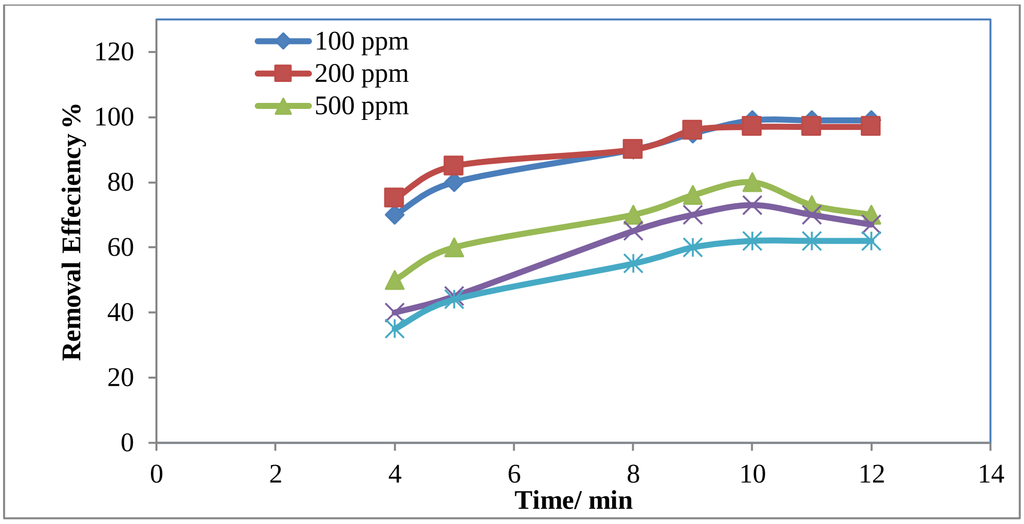

Fig 7. Effect of electrolyte concentration change on the removal %

Effect of temperature (°C)

The effect of temperature on the removal of pollutants through EC has been studied in a few articles. Effect of temperature from 5 to 50 °C has been studied for the removal efficiencies of bromate using Al-Al electrodes. Figure 8 indicate that the bromate removal efficiencies decrease with increasing temperatures above 25°C. Therefore, 25°C was fixed as optimal temperature under the same conditions mentioned, this is may be due to exist the inversely proportional between the adsorption and temperature.

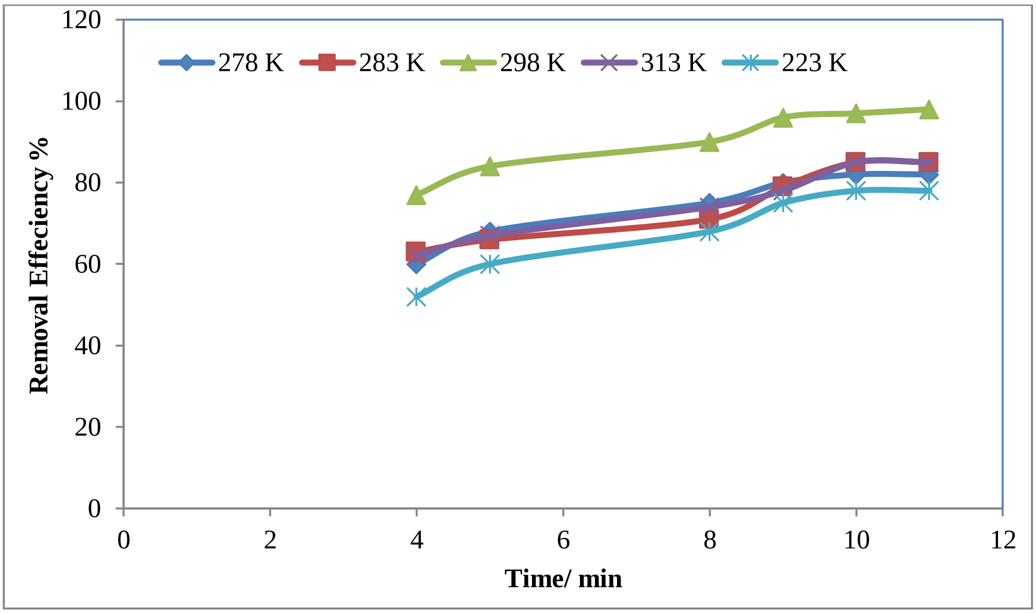

Fig 8. Effect of temperature change, keeping other variants constant.

Effect of types of electrodes

A series of batch experiments with different electrodes were carried out to study the effect of type of electrodes used in electrocoagulation on the percentage bromate removal. These experiments were carried out under the following operating conditions: current density of 22.7 mA/cm^2 , pH of 7-11, electrolysis time 10 min., and temperature of 25°C. Fig. 9 show the effect of type of electrodes used in electrocoagulation. This figure indicate that the bromate removal efficiencies change with different electrodes above 25°C. Therefore, Al-Al was fixed as optimal electrodes under the same conditions mentioned.

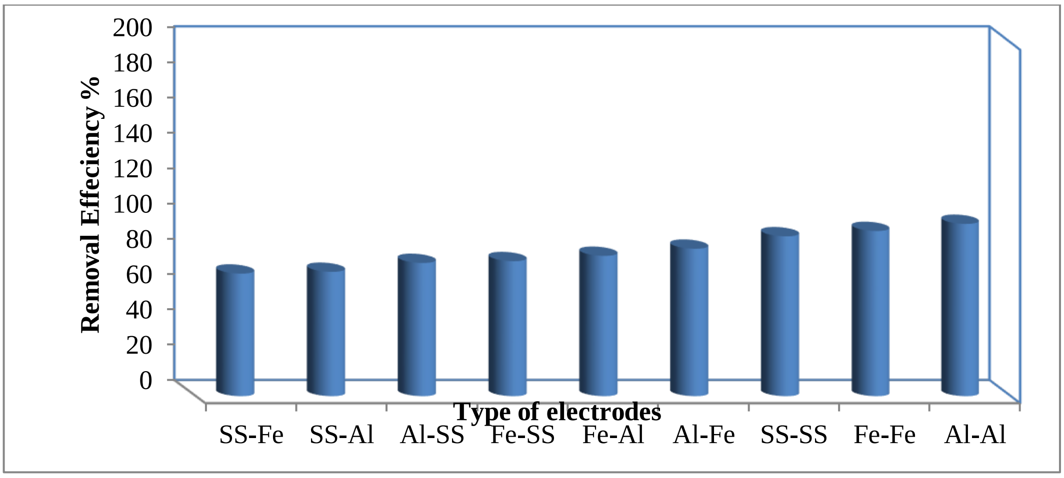

Fig 9. Effect of type of electrodes change on the removal %

IV. CONCLUSION

The removal efficiency of bromate from aqueous solution was examined by electrocoagulation using Al electrodes . The effects of initial pH, initial bromate concentration, current density, type electrode, salt concentration, and temperature were investigated on removal efficiency. It was observed that these variables significantly affected the bromate removal efficiency. The optimum bromate removal was obtained with typical operating conditions: an an initial bromate concentration of 100 mg/L, current density 22.7 mA/cm2 , salt concentration of 1.5 g/L and temperature of 25° C, the results showed that bromate removal was 97% using Al electrodes.

REFERENCES

[1] Imran Ali1 & V K Gupta, Advances in water treatment by adsorption technology , Nature Protocols 1 ,6 | 2006,1-5

[2] Guy Howard, Katrina Charles , Securing 2020 vision for 2030: climate change and ensuring resilience in water and sanitation services

[3] Nicholas L. Cain Peter H. Gleick , The global water crisis , Issues in Science and Technology · 1-5 , 2005

[4] Ying Zhua , Oliver R. Price, Drivers of contaminant levels in surface water of China during 2000–2030: Relative importance for illustrative home and personal care product chemicals , environment International 115 (2018) 161–169

[5] World Health Organization (WHO) 1996. Guidelines for drinking water quality. 2nd ed. Vol. 2. Health criteria and other supporting information. pp: 822-828

[6] Kurokawa Y, A Mackawa, N Takahashi and Y Hayeshi 1990. Toxicity and carcinogenicity of potassium bromate a new renal carcinogen. Environ. Health Perspect, 87: 309-355.

[7] Diachenko GW and CR Warner 2002. Potassium bromate in bakery products. In LEE, TC. and HO, CT. (Eds.). Bioactive Compounds in Foods. Washington: American Chemical Society, p.218.

[8] Ueno H, K Oishi, Y Sayato, and K Nakamuno 2000. Oxidative cell damage in Kat-Sod assay of oxyhalides as inorganic disinfection byproducts and their occurrence by ozonation. Arch Environ Contam Toxicol, 38: 1-6

[9] Mack RB 1988. Round up the usual suspects. Potasium bromate poisioning. N. Carolina Med J, 49: 243-245

[10] U.S. Environmental Protection Agency, Toxicological Review Of Bromate , 2001 , 1-56

[11] Haag, W.R., and Holgné, J. (1983). Ozonation of bromide-containing waters: Kinetics of formation of hypobromus acid and bromate. *Environ. Sci. Technol.* 17, 261–267.

[12] Centi G, Perathoner S (2003) Appl Catal B Environ 41: 15–29.

[13] Tajeddine L, Nemmaoui M, Mountacer H, Dahchour A, Sarakha M, et al. (2014) Clay and Soil Photolysis of the Pesticides Mesotrione and Metsulfuron Methyl. Applied and Environmental Soil Science.

[14] Vlyssides A, Karlis P, Loizidou M, Zorpas A, Arapoglou D (2001) Treatment of leachate from a domestic solid waste sanitary landfill by an electrolysis system. Environ Technol 22: 1467-1476.

[15] Naumczyk J, Szpyrkowicz I, De Faverri DM, Zilio- Grandi F (1996) Environmental Water: Advances in Treatment, Remediation and Recycling. Trans IChemE 74B: 59.

[16] Nasser M Ghalwa, Ahmed Z Musabeh and Nader B Farhat Performance Efficiency of Electrocoagulation Adsorption Process of Oxyfluorfen Herbicide from Aqueous Solutions Using Different Anodes , ***J Environ Anal Toxicol*** 2017, 7:3

[17] Carneiro PA, Fugivara CS, Nogueira FP, Boralle N, Zanoni VB, et al. (2003) A Comparative on Chemical and Electrochemical Degradation of Reactive Blue 4 Dye. Portugaliae Electrochimica Acta 21: 49-67.

[18] Chen G (2004) Electro chemical technologies in wastewater treatment. Sep. Purif. Technol 38: 11-41

[19] Vlyssides A, Karlis P, Loizidou M, Zorpas A, Arapoglou D (2001) Treatment of leachate from a domestic solid waste sanitary landfill by an electrolysis system. Environ Technol 22: 1467-1476.

[20] yssides AG, Karlis PK, Zorpas AA (1999) Environ. Int 25: 663.

[21] Ghosh D, Medhi CR, Solanki H, Purkait MK (2008) Decolorization of Crystal Violet Solution by Electrocoagulation. ***J environ Protect*** 2: 25 –35.

[22] Mollah MY, Morkovsky P, Gomes JA, Kesmez M, Parga J, et al. (2004) Fundamentals, present and future perspectives of electrocoagulation. ***J Hazard Mater*** 114: 199-210.

[23] H.S.Awad , N. Abo Galwa , Electrochemical degradation of Acid Blue and Basic Brown dyes on Pb/PbO2 electrode in the presence of different conductive electrolyte and effect of various operating factors , Chemosphere , 2005 .