

## A COMPARISON BETWEEN ALUMINIUM AND IRON ELECTRODES IN ELECTROCOAGULATION PROCESS FOR GLYPHOSATE REMOVAL

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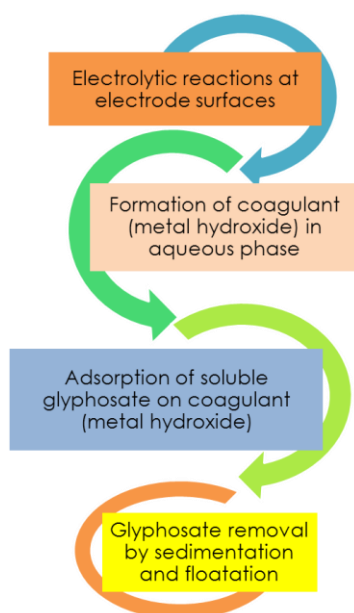
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### Graphical abstract



### Abstract

This study was intended to compare the performance of electrocoagulation process using aluminium and iron electrodes for glyphosate removal in aqueous solution. The effects of initial glyphosate concentration, electrocoagulation time and distance between electrodes, were discussed in detail. An electrocoagulation tank of 500mL with two metal plates electrodes, same in dimensions and metal types, was set up to perform batch mode laboratory experiment and the glyphosate in white powder was first diluted with deionized water. Production of metal cations showed an ability to neutralize negatively charged particles, which then encouraged to bind together to form aggregates of flocs composed of a combination of glyphosate and metal hydroxide. Compared with iron electrodes, aluminium electrodes were more effective for glyphosate removal, with a removal efficiency of over than 80%. This study revealed that electrocoagulation process using aluminium electrodes is reliable, especially designed for initial concentration 100 mg/L, electrocoagulation time 50 min, and distance between electrodes 6 cm. Finally, it can be concluded that electrocoagulation process using aluminium electrodes is efficient for glyphosate removal from aqueous environments.

Keywords: Electrocoagulation; glyphosate removal; percentage removal; aluminium electrodes; iron electrodes

### Abstrak

Kajian ini bertujuan untuk membandingkan prestasi proses electrocoagulation menggunakan aluminium dan besi elektrod untuk pembuangan glyphosate dalam larutan akueus. Kesan pembolehubah berterusan kepekatan glyphosate awal, masa electrocoagulation dan jarak antara elektrod, telah dibincangkan secara terperinci. Tangki electrocoagulation daripada 500mL dengan dua logam plat elektrod, sama dalam dimensi dan jenis logam. Ia telah ditubuhkan untuk melaksanakan mod kumpulan eksperimen makmal dan glyphosate dalam serbuk putih mula dicairkan dengan air teryahion. Pengeluaran kation logam menunjukkan keupayaan untuk meneutralkan zarah bercas negatif, maka, digalakkan untuk mengikat bersama-sama untuk membentuk agregat daripada flocs terdiri daripada gabungan glifosat dan logam hidroksida. Berbanding dengan elektrod besi, elektrod aluminium adalah lebih berkesan

untuk pembuangan glyphosate, dengan kecekapan penyingkiran lebih daripada 80%. Kajian ini mendedahkan bahawa proses electrocoagulation menggunakan elektrod aluminium boleh dipercayai, terutamanya direka untuk permulaan kepekatan 100 mg / L, masa electrocoagulation 50 min, dan jarak antara elektrod 6 cm. Akhirnya, dapat disimpulkan bahawa proses electrocoagulation menggunakan elektrod aluminium adalah berkesan untuk penyingkiran glyphosate dari persekitaran berair.

*Kata kunci:* Electrocoagulation; penyingkiran glyphosate; penyingkiran peratusan; elektrod aluminium; elektrod besi

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## 1.0 INTRODUCTION

Herbicide has been widely used since it was first marketed and application of herbicide in agriculture has further extended with increased usage for crops. In Malaysia, 172 herbicide products containing glyphosate with concentration range from 70% to 95%. Glyphosate or N-(phosphonomethyl)-glycine is a non-selective herbicide, has a function to control plants, including grasses, broad-leaves weeds, sedges and woody plants [1,2]. Its broad-spectrum and low toxicity compared to other herbicides make it quickly be adopted by farmers. Additionally, glyphosate is a phosphonomethyl derivative of the amino acid glycine. It is a white and odorless crystalline solid comprised of one basic amino function and three ionisable acidic sites. Herbicides were sold since the year 2007 in Malaysia in order to improve the quantity and quality of agricultural productions. This scenario indeed caused a very large amount of herbicide used.

The widespread and enormous usage of herbicide can create a potential source of pollution and poses a significant threat to contamination of both surface water and underground water. It is due to discharge of herbicide wastes from the point or diffused sources, such as agricultural runoff. Additionally, consumption of water contains glyphosate can bring adverse effects to human health in short-term, long-term and carcinogenicity. [3]. Therefore, a treatment needs to be introduced to remove glyphosate from water.

Electrocoagulation process is susceptible to produce flocs of higher size and density, which facilitates pollutant removal by sedimentation in an aqueous solution. It was reported that aluminium plates as electrodes has been widely applied in electrocoagulation process to treat wastewater such as urban wastewater[4], fermentation wastewater [5], palm mill oil of effluent wastewater [6], cutting oil emulsion wastewater [7], textile wastewater [8] and dairy effluents wastewater [9]. In addition, researchers claimed that aluminum plates as electrodes in electrocoagulation process successfully removed heavy metals from wastewater such as mercury (III) [10], phosphate [11,12], arsenic [13,14], fluoride [15] and antimony [16].

Meanwhile, iron plates as electrodes has been used in electrocoagulation process to treat wastewater contained orange II azo-dye[17], methylene blue [18], polyvinyl alcohol [19], hardness [20], pesticide [21], natural organic matter (NOM) [22,23] and suspended solid [24]. Electrocoagulation process using iron plates as electrodes also can removed heavy metals such as lead and zinc [25], arsenic [26], indium ions [27] and chromium ions ( $Cr^{6+}$  &  $Cr^{3+}$ ) [28].

The objective of this study is to compare the performance of electrocoagulation process using aluminium and iron electrodes for glyphosate removal in aqueous solution; and to evaluate the effects of operational variables of initial glyphosate concentration, electrocoagulation time and distance between electrodes towards removal of glyphosate from aqueous solution.

## 2.0 EXPERIMENTAL

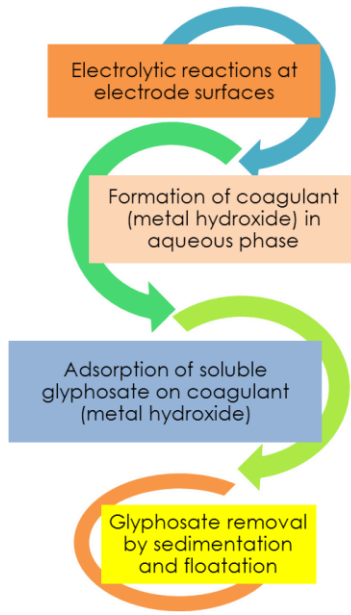
### 2.1 Mechanism

Oxidation and reduction process mechanisms occurred at both electrodes. Normally, during electrocoagulation process, there are four main processes which are electrolytic reactions at electrode surfaces, formation of coagulants in aqueous phase, adsorption of soluble or colloidal pollutants on coagulants and removal by sedimentation or floatation [29]. Figure 1 shows mechanism of electrocoagulation process in removing glyphosate.

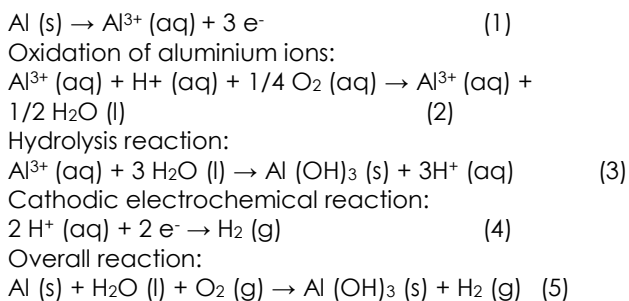
The success of electrocoagulation process is determined by the production of the bubbles [30]. Electrocoagulation process is intrinsically associated with electro-flotation since bubbles of hydrogen gas are produced at cathode electrode [31]. Some coagulated aggregates interact with the bubbles float to surface of aqueous solution and coagulated aggregates which are denser settle at bottom due to gravity attraction.

The electrochemical reaction with aluminium as an electrode may be summarized as follows:

Anodic electrochemical dissolution:

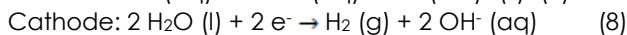
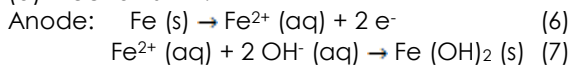


**Figure 1** Mechanism of electrocoagulation process in removing glyphosate

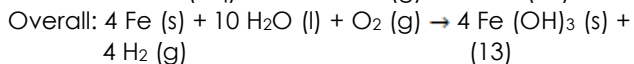
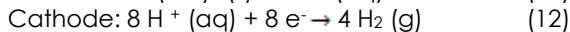
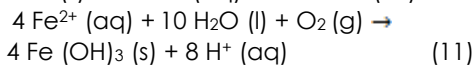
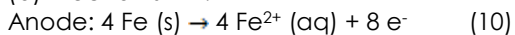


The electrochemical reactions with iron as the electrode may be summarized as follows:

(a) Mechanism 1:



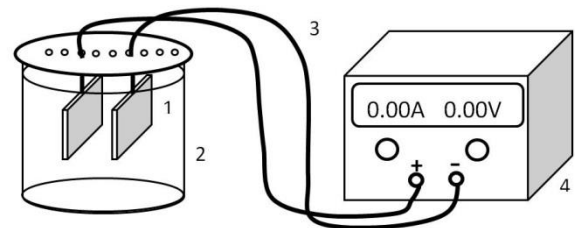
(b) Mechanism 2:



## 2.2 Methodology

Glyphosate aqueous solution was prepared by dissolving a certain amount of glyphosate powder within an electrolyte. The electrolyte chosen was sodium chloride (NaCl) solution. The conductivity was kept constant for every experiment by adjusting amount of NaCl added to the aqueous solution. Different amount of glyphosate powder was then

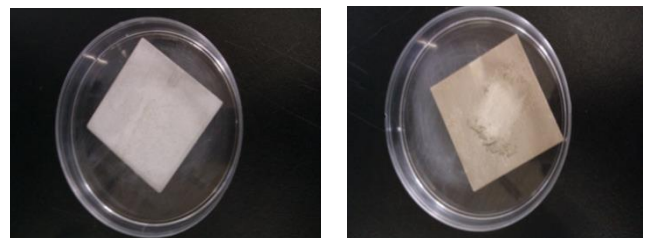
added to the aqueous solution, depending on required initial glyphosate concentration. The electrocoagulation unit was comprised of a 500 mL beaker of electrochemical cell and two plates of same type of metal with a size of 50 mm x 50 mm as electrodes (aluminum or iron) as illustrated in Figure 2. Figure 3 indicates the image of aluminium and iron plates electrode used in this study. The separation between the anode electrode and the cathode electrode was varied as well as electrocoagulation treatment time. Direct current power supply was then switched on to start electrocoagulation process at constant 40V of voltage supply. During the electrolysis process, free ions released from both electrodes; anode and cathode; which neutralize charges of particles and therefore initiate the coagulation process. The treated aqueous solution was then analyzed to determine the percentage of glyphosate removal. All the processes were handled at room temperature, 25°C.



Indicators:

1. Electrodes plates (both aluminium or both iron)
2. Plexiglass beaker reactor (electrochemical cell)
3. Copper wire
4. Digital D.C. Power Supply

**Figure 2** Schematic diagram of electrocoagulation cell with DC power supply and two electrodes



**Figure 3** Aluminium plate electrode and iron plate electrode

## 3.0 RESULTS AND DISCUSSION

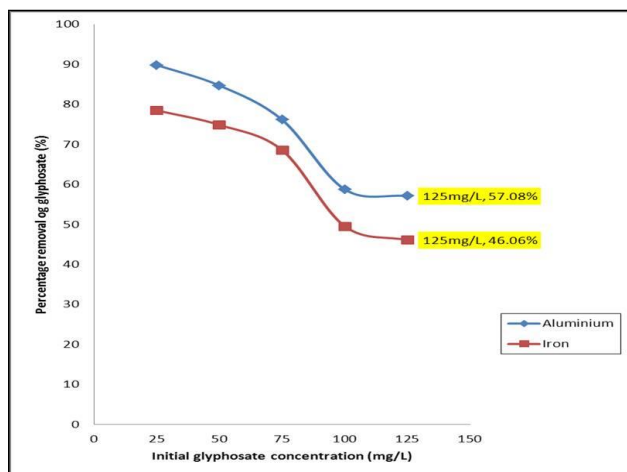
Generally, there is an electrodeposition mechanism in electrocoagulation process due to electrochemical reactions [31]. At the positive side (anode), it can be seen that there are some metals oxide deposited on the surface of electrode due to

dissolution of anode, generation of metal ions (metal hydroxide) for coagulation. Meanwhile, negative side (cathode), the metal electrode surface was cracked because the release of too much hydrogen bubbles which help flocculated particles to float out of the aqueous solution.

There are three layers were formed right after the process. A very thin waxy layer of flocs formed at surface, clear supernatant at the middle and a thick layer of precipitated sludge at the bottom. Aqueous solution in electrocoagulation cell that used aluminium plates as electrodes become white, right after electrocoagulation process. Meanwhile, the treated aqueous solution using iron plates as electrodes become dark green after electrocoagulation process. It is due to metal corrosion and electrochemical reactions.

### 3.1 Effect of Initial Glyphosate Concentration

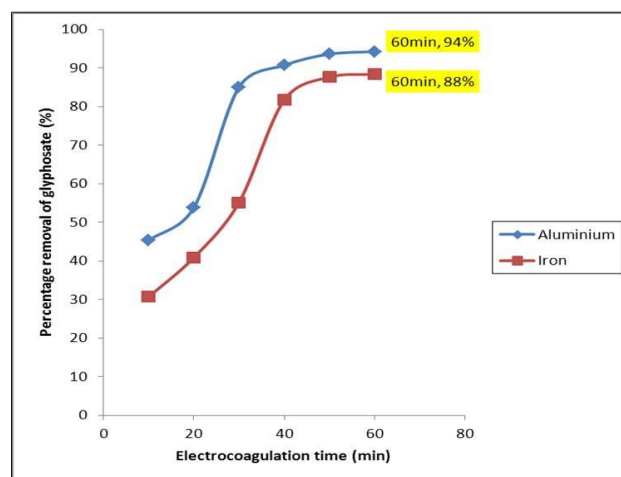
High removal efficiency could be achieved with less initial concentration [21]. The availability of metal hydroxide coagulant produced was greater due to lesser amount of glyphosate to be removed. The removal of glyphosate was limited by amount of metal hydroxide. The performance of electrocoagulation process has been slow down when initial glyphosate concentration is 100mg/L. Figure 4 shows the effect of initial glyphosate concentration towards percentage removal of glyphosate. The figure indicates that maximum potential of electrocoagulation process using aluminium electrodes, removed 57.08% of glyphosate meanwhile iron electrodes removed 46.06% of glyphosate when initial glyphosate concentration used is 125mg/L. Therefore, these percentage removal values are considered as optimum values because there is no more changes in percentage of glyphosate removal could be detected when higher initial concentration of glyphosate was implemented.



**Figure 4** Effect of initial glyphosate concentration (60 min electrocoagulation time and 6 cm electrode gap).

### 3.2 Effect of Electrocoagulation Time

Time of electrocoagulation is a time provided to the process to generate metal hydroxides and to complete coagulation of glyphosate. Highest removal efficiency has been achieved due to increment amount of metal hydroxide coagulant produced parallel with increment in time [8]. Besides, increment in treatment time contributed to higher rate of bubble generation, which helped to remove glyphosate and flocs of lower density and size by gas flotation [2,30]. The rate of bubbles generation also increased and the size of bubble decreased which induced a higher removal of glyphosate by hydrogen gas flotation. Figure 5 shows the effect of electrocoagulation time towards percentage removal of glyphosate. The figure revealed that 94% of glyphosate removed when electrocoagulation process using aluminium electrodes and 88% of glyphosate removed when using iron electrodes for 60min electrocoagulation time. These percentage removal values were induced by highest potential of electrocoagulation process. Thus, these percentage removal values are selected as optimum values of the process. Percentage removal of glyphosate would remain the same values even the process increase its electrocoagulation time longer than 60min.

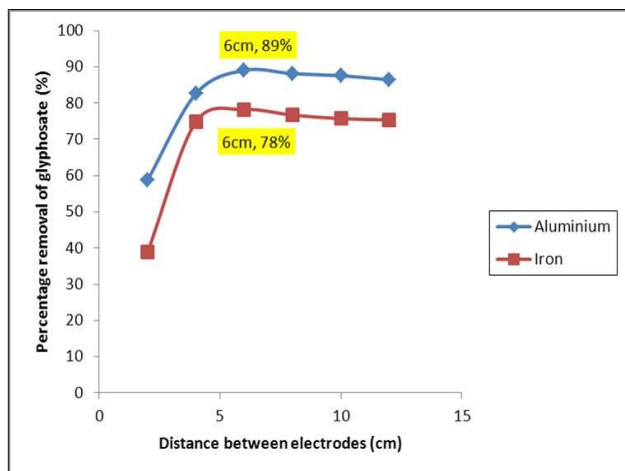


**Figure 5** Effect of electrocoagulation time (100 mg/L initial glyphosate and 6 cm electrode gap)

### 3.3 Effect of Distance between Electrodes

Increment in electrodes spacing reduced the treatment efficiency, meanwhile, shorter gap increases the removal efficiency for both type of electrodes. Highest removal efficiency has been achieved at the distance, 6 cm. Increment in distance between electrodes encouraged in less efficiently transfer of formed ions, facilitated their flocculation and was unlikely to coagulate with metal hydroxide [10]. Figure 6 shows the effect of

distance between electrodes towards percentage removal of glyphosate. The figure displays that electrocoagulation process using aluminium as electrode removed 89% of glyphosate and electrocoagulation process using iron as electrode removed 78% of glyphosate at 6cm distance between electrodes. From the graph, these values of percentage removal are pointed as optimum values for this process since the values indicate highest peak of each graph. In addition, these optimum values also interpreted maximum removal values that could be achieved by the process.



**Figure 6** Effect of distance between electrodes (60 min electrocoagulation time and 100 mg/L initial glyphosate)

### 3.4 Sludge Production

Electrocoagulation process produced aggregates of flocs which then settled at the bottom of the beaker as a precipitate of sludge due to gravity. At the end of the process, treated water became clear and isolated. Theoretically, the sludge contained glyphosate and polymeric metal which act as a coagulant in the process [32,33]. The percentage of metal hydroxide or metal oxide in sludge is normally greater than glyphosate [8]. The sludge was easily settled down at the bottom of the beaker because of the metal either aluminium or iron are denser than aqueous solution. High percentage removal of glyphosate induced high amount of sludge. As viewed in previous figures, initial glyphosate concentration, electrocoagulation time and distance between electrodes were affected the percentage removal of glyphosate, therefore it also encouraged the amount of sludge produced.

Electrocoagulation process using aluminium electrodes produced a sludge which is composed of aggregates of flocs contained glyphosate and polymeric aluminium such as aluminium oxide and aluminium hydroxide [34]. The sludge is in form of greyish white wax and easy to dispose when it is dry enough. Meanwhile, electrocoagulation process using iron electrodes produced sludge in form of brown powder when it is dry enough. The sludge

formed tends to be readily settlable and easy to dewater because it is composed of mainly ionic oxides or ionic hydroxides [34]. Since the sludge was composed of ionic compounds, the dried sludge has qualities and characteristics to produce beneficial products.

## 4.0 CONCLUSION

### 4.1 Conclusion

In conclusion, this study showed that electrocoagulation process has a potential to remove glyphosate from aqueous solution. Results showed that percentage removal of glyphosate from the aqueous solution using aluminium as electrodes always reached more than glyphosate removal using iron as electrodes for all three effects in electrocoagulation process. It can be concluded that electrocoagulation method is reliable and efficient for glyphosate removal which especially designed for initial concentration 125 mg/L, electrocoagulation time 60 min and distance between electrodes 6 cm. Percentage removal of glyphosate encouraged amount of sludge produced. High percentage removal of glyphosate induced high amount of sludge.

Hence, by removing glyphosate, it reduced the amount of glyphosate in water before the water is safe to be consumed. The toxicity of the water can also be reduced or totally eliminated. This project is considered a new innovation since the method of glyphosate removal using electrocoagulation process has not been discussed yet in any field even in agriculture, wastewater treatment or separation technology field.

### 4.2 Recommendations

There are many parameters that also can affect the results of electrocoagulation process such as temperature and mixing rate. By changing these parameters, the removal efficiency might be increased as well.

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