



## Removal of Total Arsenic in Simulated Irrigation Water Using Electrocoagulation

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**Abstract:** Arsenic (*As*) is one of the most toxic substances in the environment. Reports show that there is arsenic contamination in rice paddy soils in the Philippines. Some of these areas are in Bae, Laguna (15.49 ppm), Milaor, Camarines Sur (9.23 ppm) and Sipalay City, Negros Occidental (11.44 ppm). Irrigation water in Bae, Laguna contained 17 ppb *As* which is higher than the World Health Organization (WHO) standard for agricultural water. These demonstrate the need to prevent *As* contamination into the soil and groundwater which could be deposited and absorbed in food crop. Electrocoagulation is an efficient method used to remove dissolved heavy metals, such as *As*, in water. The solution is subjected to an electric charge through a set of electrodes causing the formation of a water-insoluble mass containing the heavy metal contaminant, thus allowing the separation of the contaminant from the solution. It has been found out that iron (*Fe*) electrodes are efficient due to the affinity of *As* to *Fe*. The focus of the study was on the effects of current density (3.0, 12.3 and 28.8 A/m<sup>2</sup>), electrode distance (16, 25 and 34 mm) and flow rate (0.4, 1.0 and 2.0 L/min) on the removal of total *As* using a bench-scale, 18-L, cylindrical, electrocoagulation reactor. Experimental runs were conducted based on the Taguchi Design of Experiments. Results show that electrocoagulation reduced the *As* concentration from an initial concentration of 20 ppb to 2 ppb. The highest percentage removal achieved was 87% at a flow rate of 1 L/min, electrode distance of 16 mm and current density of 28.8 A/m<sup>2</sup>.

**Key Words:** arsenic; rice paddy; irrigation water; electrocoagulation

### 1. INTRODUCTION

Arsenic contamination in water affects many countries around the globe, including the Philippines [2]. Some of the main sources of arsenic contamination are mining operations and the use of arsenic-containing fertilizers [3].

In the Philippines, a study on the investigation of the presence of heavy metals, including arsenic, in rice paddy soils was conducted [2]. Some of these areas are in Bae, Laguna (15.49 ppm), Milaor, Camarines Sur (9.23 ppm), and Sipalay City, Negros Occidental (11.44 ppm). Arsenic deposition in rice paddy soil has increased

as a result of the use of contaminated irrigation water. These arsenic compounds potentially can be taken up by food crop, resulting in bioaccumulation. In order to prevent this, the arsenic species must be captured before it reaches the rice paddy.

Electrocoagulation is one of the recent techniques used in removing heavy metal contaminants, such as arsenic, from water. The solution is subjected to an electric current through the use of electrodes to coagulate and facilitate the removal of arsenic in the solution. A study reported that using iron as electrodes can remove more than

99% of the total arsenic content from contaminated water [4, 5].

Studies have been conducted using groundwater and simulated groundwater in the removal of arsenic, but not on actual irrigation water and simulated irrigation water [11, 18]. Various studies have investigated varying the arsenic concentration, current density, pH, and nature of electrodes, but none have investigated the effect of varying the electrode distance and the flow rate [11, 19]. Also, majority of research in electrocoagulation was limited to beaker scale reactor, with the exception of Wei Wan et al. [6].

Therefore, this study investigated the effect of electrode distance and flow rate on the removal of arsenic using iron electrodes from simulated irrigation water in a bench scale electrocoagulation reactor.

## 2. METHODOLOGY

### 2.1 Simulation of Irrigation Water

An initial concentration of 20 ppb arsenic was used in the study based on the analysis of the irrigation water (Bae, Laguna, Philippines; 17 ppb) which had the highest concentration of arsenic in soil [2]. Arsenic from a stock standard solution was prepared by dissolving 1.320 g of  $As_2O_3$  in water containing four grams of NaOH diluted to one liter. The final ratio of arsenic to water in mg to mL is 1:1.

### 2.2 Parameters of the Study

The parameters used in the study are presented in Table 1. The electrode distance and flow rate were based on the optimum values obtained in the study of Lieu [7]. The minimum value for the current density was based on the study of Gomes et al. [8] and the maximum value was based on the study of Lieu [7]. The conductivity was based on the actual conductivity of the irrigation water samples. Conductivity will be adjusted with the addition of AR grade sodium chloride. The pH range was based on studies which found that that range was ideal for electrocoagulation [4, 5, 9]. In order to maintain the pH, HCl or NaOH was added to the solution.

The Taguchi Design of experiments was used to conduct the experimental runs of this study. The Taguchi Design of experiments is a simplified version of the full factorial design with the assumption that the variables are independent and do not interact with each other.

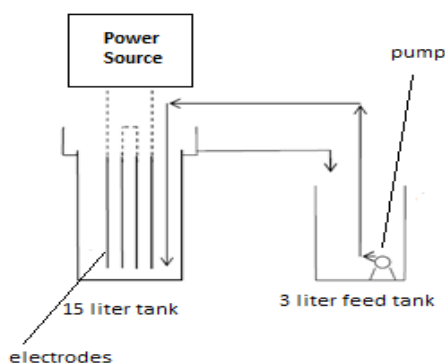
**Table 1. Parameters of the study**

<b>Constant Parameters</b>	
pH range	6-8
Volume of Water, L	18
Electrode Connection	Monopolar
Electrode Pairing	Parallel
Anode/Cathode	Fe/Fe
Initial Conductivity	600 $\mu$ S/cm
<b>Variable Parameters</b>	
Current Density, A/m <sup>2</sup>	3, 12.3, 28.8
Inner-Electrode Distance, mm	16, 25, 34
Flow Rate, L/min	0.4, 1, 2

### 2.3 Experimental Set-up and Procedures

A bench-scale electrocoagulation reactor with a capacity of 18 liters and an effective volume of 15 liters was used in this study. As shown in Figure 1, four vertical upright iron electrodes submerged completely in the simulated irrigation water were arranged in mono-polar parallel configuration with Fe both as the anode and the cathode. The simulated irrigation water was continuously re-circulated from the feed tank to the reactor by means of a submersible pump located inside the feed tank.

Sampling of water was done every 15 minutes, and these samples were taken to an accredited laboratory for arsenic analysis. The analysis was done using Graphite Furnace Atomic Absorption Spectrophotometry (GF-AAS).



**Figure 1. Electrocoagulation Reactor Schematic Diagram**

## 3. RESULTS AND DISCUSSION

The results obtained in this study are presented in the succeeding sections. The data presented were acquired through GF-AAS analysis

equipped with As-HCL and by in-situ instrument measurements. Data analysis was done using Minitab 16 and Microsoft® Excel® 2010.

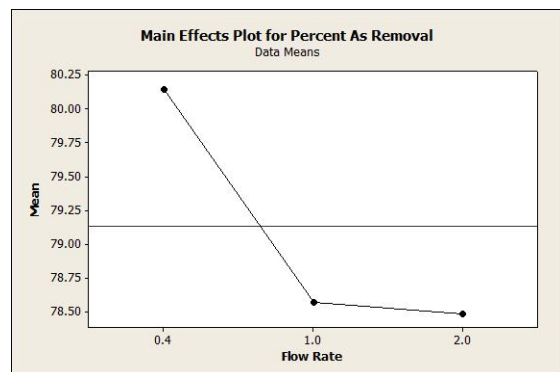
The combination of the three operational parameters and their corresponding percent removal are presented in Table 2.

**Table 2. Percent Removal of Arsenic**

Run	Flow Rate L/min	Electrode Distance mm	Current Density A/m <sup>2</sup>	Percent Removal %
1	1.0	16	28.8	87.17
2	0.4	25	12.3	83.66
3	2.0	34	3.0	77.65
4	2.0	25	28.8	83.74
5	1.0	34	12.3	69.97
6	0.4	16	3.0	79.05
7	0.4	34	28.8	78.75
8	2.0	16	12.3	74.06

The highest percent removal obtained was 87.17%. It can be seen that the percent removal achieved by the electrocoagulation process did not reach 99% unlike in other studies. This might be due to the use of an 18-liter bench scale reactor instead of the beaker scale reactor. In addition to that, the initial concentration used in this study is less than the initial concentration of other studies (ppm vs. ppb), whereas the percent removal would be higher with higher initial concentrations [6, 11, 18].

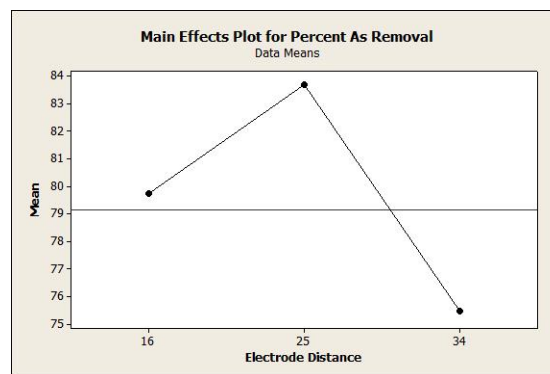
### 3.1 Effect of Flow Rate on Percent As Removal



**Figure 2. Effect of Flow Rate on Percent As Removal**

Results indicate that the percentage removal decreased with an increase in flow rate as shown in Figure 2. This was also observed in other studies and was attributed to the depletion of the coagulant produced by the application of current at the electrodes caused by the mixing process [11, 12]. According to Harif et al. [13], the floc structures formed are porous and fragile when subjected to high shear environment. In addition to that, Lieu [7] stipulated that high flow rates may have hindered the formation of flocs which resulted in lower removal of arsenic.

### 3.2 Effect of Electrode Distance on Percent As Removal

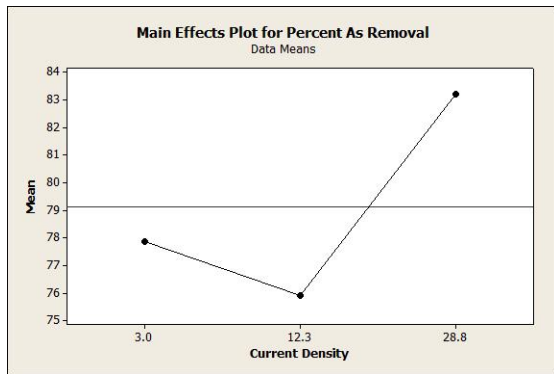


**Figure 3. Effect of Electrode Distance on Percent As Removal**

It was observed that from 16 to 25 mm, the percent removal increased from 79.8% to 83.6% while from 25 to 34 mm, the percent removal decreased from 83.6% to 75.4% as shown in Figure 3. Currently, no studies have been reported on varying the electrode distance as a parameter in the removal of arsenic. However, the closer the electrodes are, the lower is the electrical resistance between them which is more desirable in electrocoagulation [14]. On the other hand, Mohora et.al. [12] reported that at a close electrode distance hydrogen bubbles obstruct the mass transfer of the system lowering the removal of the contaminant. Thus, it may be considered that percentage removal with an electrode distance below 25 mm is affected by this obstruction. In addition to that, the accumulation of solid particles and bubbles in between the electrodes impedes the passage of current [16]. Hence, below 25 mm, the electrocoagulation process is mass-transfer limited. However, increasing the electrode distance to 34 mm also results in a decrease in percent removal;

this implies that at 34 mm electrode distance, the electrocoagulation process is limited.

### 3.3 Effect of Current Density on Percent As Removal



**Figure 4. Effect of Current Density on Percent As Removal**

It was observed that from 3 to 12.3 A/m<sup>2</sup> the percent removal decreased from 78.8% to 75.9% while from 12.3 to 28.8 A/m<sup>2</sup>, the percent removal increased from 75.9% to 83.5% as shown in Figure 4. Theoretically, as the current density increases, the more Fe<sup>2+</sup> ions are released and the more hydrogen gas bubbles are formed leading to an increase in the removal efficiency of the system. However, high current density is not desirable because of the increase in energy consumption and the increase of temperature due to excess energy [17].

The difference in these findings is mainly associated with the charge loading that was applied. According to Chen et al. [24] previous studies have investigated the effect of current density while varying the charge loading at the same time. This can also be observed in the study of Balasabrumanian [20] wherein the charge loading was not fixed, instead, an operating voltage range of 0-30 V was applied. It can be noted in both the studies of Chen et al. [24] and Kumar et al. [5], that while the current density was being investigated, charge loading was held constant and vice versa. This allows both studies to be able to conclude that current density has no effect on the total removal of pollutants [5, 24].

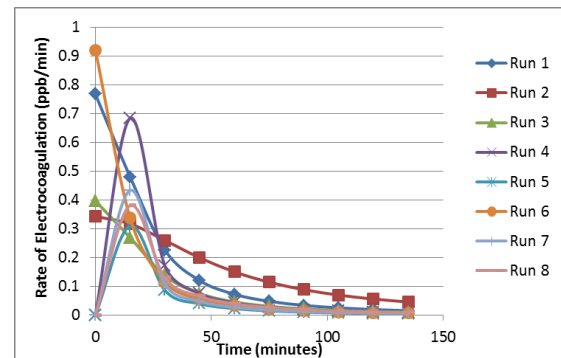
### 3.4 Electrocoagulation Rate

The electrocoagulation rate relates the change in concentration of arsenic in solution with time. The electrocoagulation rate was

approximated using Equation 1 which is an arbitrary inverse function with four degrees of freedom. The values of the four regression constants were determined using the Solver<sup>®</sup> function of Microsoft<sup>®</sup> Excel<sup>®</sup>.

$$R(t) = \frac{c_1}{c_2 + c_3t + c_4t^2} \quad \text{Eq. 1.}$$

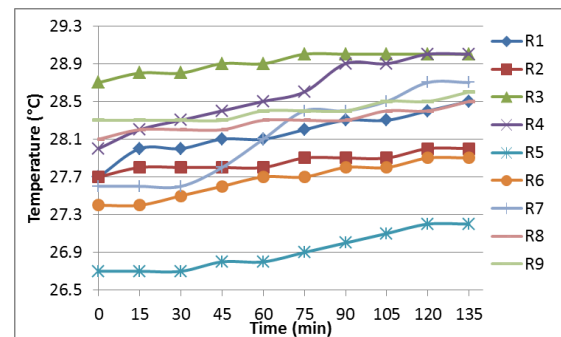
where:  $R$  = rate of electrocoagulation  
 $c_{1-4}$  = regression constants  
 $t$  = time



**Figure 5. Electrocoagulation Rates**

From Figure 5, it can be seen that the highest electrocoagulation rate took place within the first 15 minutes of the process. This signifies that most of the arsenic was removed within few minutes of treatment [5]. As the treatment progresses, the concentration of arsenic in the solution is reduced causing the decrease in the electrocoagulation rate.

### 3.5 Temperature Profile



**Figure 6. Electrocoagulation Temperature Profile**

As shown in Figure 6, the temperature profile for all runs increased for only a small amount. The

highest increase was 1.1°C with a flow rate of 0.4 L/min, an electrode distance of 34 mm, and a current density of 28.8 A/m<sup>2</sup>. From theory, the temperature increases because of the passage of heat through a conductor. A small increase in temperature would mean that the energy supplied by the current is used for the coagulation process, but not to heat up the solution. In addition, Mission [23] reported that a large increase of temperature leads to inefficient contaminant removal.

### 3.6 pH Profile

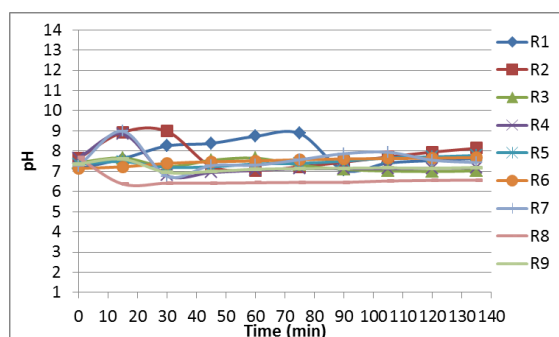


Figure 7. Electrocoagulation pH Profile

From Figure 7, the small increase in pH may be attributed to the production of H<sub>2</sub> bubbles which escape from the top of the solution. The loss of hydrogen ions gradually increases the pH of the solution. This was also observed by Kumar & Goel [4]. HCl was used to lower the pH of the solution and maintain a pH range of 6 – 8.

The highest pH measured was 8.98. The pH may be correlated with arsenic removal since that the hydroxyl ions are used to adsorb the arsenic. That phenomenon is evident in Run 1 which had high pH values resulting to a percentage removal of 87.17%.

### 3.7 Conductivity Profile

The conductivity during electrocoagulation remained constant as shown in Figure 8 except for Run 1 wherein the conductivity increased at time t = 105 minutes because of the addition of HCl. Theoretically, the conductivity is affected by the ions present inside the solution. The ions are produced by the electrodes and reduced by the formation of froth and sludge in the solution. A constant conductivity would imply an equal rate for both the ion production and froth and sludge formation. This phenomenon was also observed by Wan et.al [6] who also used a bench scale reactor

with a total volume of 50-Liters. However, Kumar & Goel [4], reported that conductivity increases as the process continues; the increase in conductivity was said to be caused by the dissociation of the anode to ferric or ferrous ions.

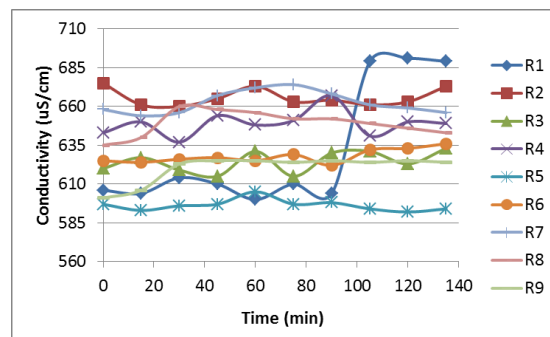


Figure 8. Electrocoagulation Conductivity Profile

## 4. CONCLUSIONS

The removal of arsenic from simulated irrigation water by electrocoagulation was investigated using an 18-L bench scale reactor. The effects of electrode distance and flowrate were investigated. Results show that decreasing the flowrate increased the arsenic removal of the system, and that an electrode distance of 25 mm has the highest average arsenic removal of 83.72%. Temperature of the solution was monitored to ensure that the current input to the system is used for the treatment process and not for heating. The pH of the solution was controlled at a pH range 6 - 8. The highest percentage removal of total arsenic is 87.17% at a flowrate of 1L/min flowrate, electrode distance of 16 mm, and current density of 28.8 A/m<sup>2</sup>.

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