

# Estimating Energy Consumption and Cost for the Electrocoagulation of Arsenic-laden Water (ECAR) Using Iron Electrodes

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**Abstract.** In some areas in Pampanga, arsenic concentration from handpumps reaches up to 300 µg/L, 10 times higher than the safe limit for drinking water. An efficient way of reducing elevated arsenic concentration is through electrocoagulation (EC) process with the use of iron electrodes. However due to several factors, the efficiency of the technique is decreased. This study focuses on determining the energy consumption and cost through time. The cost per cycle was estimated through the power consumption and projecting its growth with time. One 600 L cycle costs around \$0.60 to \$1.10 which is approximately \$0.001 to \$0.002 per liter of water. This value increases through each cycle until half of the electrode is consumed (500 cycles) and is to be replaced. The current processing time was set at 30 mins, charge dosage of 150 C/L, applied current of 16.67 mA, and an electrode area of 6.6 cm<sup>2</sup>. One factor examined which may have caused the increase is the formation of passivation layer on the electrode surface. It was described using linear sweep voltammetry (LSV) and Tafel extrapolation method. The resistance due to charge transfer was determined to be increasing per cycle.

## 1 Introduction

In 2014 the Department of Health, in coordination with the Dutch Risk Reduction team, reported that Pampanga groundwater has elevated arsenic concentrations. The problem seems to be rampant particularly in certain areas in Lubao, with arsenic concentration reaching up to 300 µg/L–30 times higher than the Philippine National Standards for Drinking Water for arsenic [1].

Arsenic is a naturally occurring element and can contaminate water and soil and one possible reason for the contamination in Pampanga area is due to the Mt. Pinatubo eruption in 1991. Arsenic is a carcinogenic chemical and chronic exposure can eventually damage one's internal organs and cause skin diseases [2,3].

Seven out of nine drinking water samples from households of index cases have elevated arsenic concentrations between 10-600 µg/L from drinking water tests conducted in December 2014. Half the population of Lubao still uses handpump wells. From 2010 to 2014, 215 residents from 5 barangays in Lubao had consulted with similar dermatological symptoms, due to chronic exposure to the contaminated water. Some dermatologic symptoms are skin discoloration, skin rashes, lesions, etc [1].

Rural Bangladesh experiences the same situation, but arsenic levels were effectively reduced in the area using electrocoagulation process, from 100- 500 µg/L arsenic to below the maximum limit of 10 µg/L recommended

by the World Health Organization [4,5]. Electrocoagulation is an electricity-based treatment method wherein a sacrificial iron electrode is slowly electrochemically dissolved in arsenic-laden water [6]. Electrolytic dissolution of Iron (Fe-ED) is driven by applied current to the sacrificial iron anode as shown in Figure 1. The amount of electrons taking part in the process can be controlled based on the Faraday's Law, enabling the calculation of the iron dose from the applied current [7]. It is feasible due to its low treatment costs that offsets the maintenance, operation, and capital costs of an electricity source (usually small scale such as photovoltaics or battery packs).

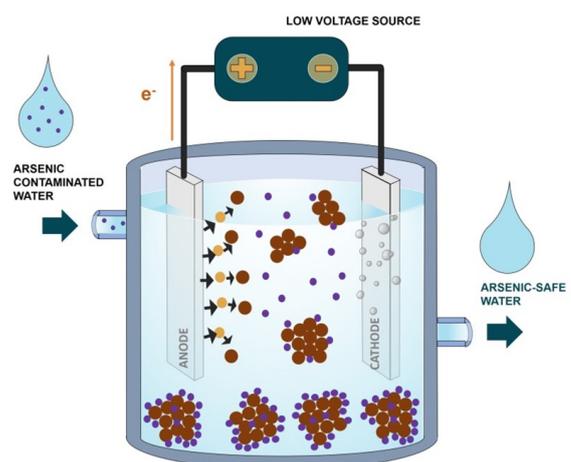


Fig. 1. Electrocoagulation process of arsenic-laden water

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There are several advantages of using electrocoagulation over conventional chemical coagulants. Usual chemical coagulants such as alum, usually sold as  $KAl(SO_4)_2 \cdot 12(H_2O)$ , and ferric chloride, sold as  $FeCl_3 \cdot 6(H_2O)$ , create additional counterions in the solution alongside the metal cations which serve as the main coagulant. With 1g dose of  $Al^{3+}$  or  $Fe^{3+}$ , additional 8.57 g and 1.91 g respectively of undesired counterions are created. Chemical coagulants also contain very little coagulating agent. 1 ton of Alum as  $KAl(SO_4)_2 \cdot 12(H_2O)$  only contains 114 lb of  $Al^{3+}$  [8]. Other advantages of the ECAR process over other common technologies are listed in Table 1. A green, yellow, or red circle indicates the technology is favorable, average, or unfavorable, respectively, in the categories shown.

**Table 1.** Comparison of ECAR with common arsenic removal technologies for treating drinking water.

	Reliable (As III/V < 10 µg/L)	Low cost	Back-washing is not needed	No Media Replacement or Regeneration	Does not require a skilled Operator
ECAR	●	●	●	●	●
Ferric Salt Coprec/ZVI	●	●	●	●	●
Iron Based Media	●	●	●	●	●
Activated Alumina	●	●	●	●	●
Anion Resins	●	●	●	●	●
Reversed Osmosis	●	●	●	●	●
Piped Water Systems	●	●	●	●	●

The energy requirement and cost for the EC process depend on multiple factors such as the materials and equipment being used, cost of electricity, cost of labor and efficiency of the process. The power requirement necessary to effectively reduce the arsenic in water increases with time. This is due to the deterioration of the electrodes since they also serve as the source of the coagulants and are expected to pit. The pitting behavior of the electrodes lead to the formation of passivation layer [8,9]. As the passivation layer gets thicker, the overpotential required for the EC process also increases. At some time, the growth will hamper the process and deplete the efficiency of the process. The electrodes will soon have to be replenished or the current to be inverted to let the other electrode degrade. It is important to estimate the increase in cost to determine if the process is plausible and reproducible.

In the study of Gu et al., the energy consumption of electrocoagulation was estimated [10]. However, the effect of passivation layer formation was not included. Due to continued use of the iron electrodes, and possibly due to improper storage of the electrodes after the arsenic removal process, corrosion forms on the surface layer.

This study aims to determine the rate of increase in the power requirement of a 200 mL EC process and see

how it affects the cost per cycle of the ECAR process and try to explain the phenomenon through the determination of component resistances in the system. All are done in the premise of constant parameters including pH and water composition. These factors have been proven to have drastic effects in the EC process

## 2 Materials and Methods

### 2.1. Characterization of groundwater sample and preparation of synthetic Pampanga groundwater

Groundwater samples were collected from a number of handpumps in Pampanga. On site measurements of dissolved oxygen (DO), conductivity, resistivity, total dissolved solids (TDS), turbidity, temperature, and pH, were done. Arsenic, along with other cations present such as magnesium, sodium, and calcium present in the solution, were measured using Prodigy 7 Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES). Anions such as carbonate, phosphate, silicate, sulfate, and chloride were also characterized in the laboratory.

Determination of the arsenic speciation was done through Anodic Stripping Voltammetry method using 946 Portable VA Analyzer with scTRACE Gold sensor. The results from the various test analyses are shown on the Table 2.

**Table 2.** Groundwater Characterization Results

Ion	Conc. (ppm)	Analysis Method
Arsenic, $As_{total}$	0.057	ASV
Arsenate, $As[V]$	0.056	ASV
Arsenite, $As [III]$	$8.75 \times 10^{-4}$	ASV
Phosphorus, P	0.9	Stannous Chloride
Bicarbonate Alkalinity, $CaCO_3$	177	Titration
Sulfate, $SO_4$	3	Turbidimetric
Chloride, Cl	5	Argentometric
Silica, $SiO_2$	50	Molybdosilicate
Sodium, Na	66	Flame AAS
Magnesium, Mg	5	ICP-OES
Copper, Cu	0	ICP-OES

The procedure used and developed by Addy (2008) that makes use of relevant ions to produce the synthetic Bangladesh Groundwater in a similar study was followed in this study to produce the synthetic groundwater similar to the conditions of the groundwater from San Isidro in Guagua, Pampanga [6]. Table 3 lists the relevant ions, the reagents used, and the corresponding synthetic groundwater characteristics produced.

**Table 3.** Reagents used to produce synthetic Pampanga GW with corresponding characteristics

Ion	Reagent	Synthetic Pampanga GW
Arsenate, AS[V], µg/L	Na <sub>2</sub> HAsO <sub>4</sub> ·7H <sub>2</sub> O	55.77
Arsenite, As [III], µg/L	NaAsO <sub>2</sub>	0.875
Phosphate, PO <sub>4</sub> , mg/L	Na <sub>2</sub> HPO <sub>4</sub> ·7H <sub>2</sub> O	2.760
Bicarbonate, HCO <sub>3</sub> , mg/L	NaHCO <sub>3</sub>	107.9
Sulfate, SO <sub>4</sub> , mg/L	CaSO <sub>4</sub> ·2H <sub>2</sub> O	3.00
Magnesium, Mg, mg/L	MgCl <sub>2</sub> ·6H <sub>2</sub> O	23.567
Chloride, Cl, mg/L	NaCl	5.00
Calcium, Ca, mg/L	CaCl <sub>2</sub>	9.827
Silicate, SiO <sup>4+</sup> , mg/L	Na <sub>2</sub> SiO <sub>3</sub> ·5H <sub>2</sub> O	101.583

## 2.2 Determination of Power Consumption Through Electrocoagulation

Prior to electrocoagulation, the iron electrodes were submerged in 1M HCl solution for 30 minutes and finished with sandpaper, rinsed with deionized water while being finished with fine sandpaper, and then dried. The clean electrodes were attached to a 1.5 cm x 1.5 cm wood clamped and attached to an iron stand to ensure a constant spacing between the two electrodes and a constant depth throughout the electrocoagulation process.

The voltage source was connected to a multimeter (Fluke 117 True RMS Multimeter) and the iron counter electrode. The multimeter is connected in series with the working electrode using another electrical wire. Another multimeter is connected in parallel with the working and reference electrode, the four-point probe was used in order to monitor both the voltage and current simultaneously.

The electrodes were submerged in 200 mL of synthetic groundwater at a depth of 3.3 cm. Using a stirring plate and a magnetic stirrer, the stirring speed was kept constant at 100 rpm. A dosage rate of 5 C/L-min was used based on the optimum condition set by Addy in her study and the current that passed through the system was 16.67 mA, with current density 2.525 A/cm<sup>2</sup>. The voltage was adjusted from time to time to keep the current constant. The current processing time was set to 30 minutes.

Electrocoagulation process was done and repeated for 7 cycles. This was done to form the passivation layer on the surface of the anode with different number of cycles. The anodes were then air dried and stored in sealed plastic bags.

## 2.3 Quantification of the Resistances

The pitting behavior, passivity, and corrosion voltage of the iron working electrodes that underwent electrocoagulation were evaluated using linear sweep voltammetry (LSV) [11,12]. A three-electrode cell

connected to Metrohm Autolab PGSTAT 204 and controlled by Nova 2.0 software was used for data acquisition. Platinum counter (Pt) and Silver Chloride (AgCl) electrodes were respectively used as the auxiliary and reference electrodes submerged in 200 mL of synthetic groundwater. A spacer made from styrofoam was used. The electrodes are set at a distance of 1.5 cm from each other.

The electrochemical potential was scanned from -4.0V to +4.0V vs AgCICE at a scan rate of 0.5 mV/s. The polarization resistance, R<sub>p</sub>, was obtained by getting the slope of the LSV plot near the open circuit potential, where the voltage meets zero current. The charge transfer resistance, R<sub>ct</sub>, was computed by first obtaining necessary parameters through Tafel plots. Charge transfer resistance follows Faraday's law and can be calculated using the Butler-Volmer equation [6]:

$$i = i_0 \left( \frac{C_o}{C_o^*} \exp \exp \left( \frac{\alpha n F \eta}{RT} \right) - \frac{C_R}{C_R^*} \exp \exp \left( \frac{-(1-\alpha)n F \eta}{RT} \right) \right) \quad (1)$$

where  $i_0$  is the exchange current density,  $C_o$  is the concentration of oxidant at the electrode,  $C_o^*$  is the concentration of oxidant,  $C_R$  is the concentration of reductant at the electrode surface,  $\eta$  is overpotential,  $F$  is Faraday's constant,  $T$  is temperature,  $R$  is gas constant,  $\alpha$  is reaction order, and  $n$  is the number of electrons involved.

Assuming that the concentrations in the bulk and the electrode surface has the same value and the overpotential is very small that the system is approaching equilibrium, the charge transfer resistance can be reduced to a simple equation. It can be expressed as a function only of the resistance, overpotential, exchange current density and some constants (Eq. 2). Since the charge-transfer resistance is estimated under the premise of complete dissolution and even concentration, it is necessary for the electrolyte solution to be mixed well while measurements are being developed.

$$R_{ct} = \frac{RT}{nFi_0} \quad (2)$$

The ohmic resistance, R<sub>Ω</sub> was computed using the equation:

$$R_{\Omega} = \frac{L}{\kappa A} \quad (3)$$

where L is the distance between the anode and cathode,  $\kappa$  is the conductivity of the solution, and A is the area of the anode that is submerged in the solution. The conductivity of the solution was measured before and after each cycle.

## 2.4 Cost Estimation

The initial values for the costs were determined through canvassing, experimentation and adaptation from related literatures. The presents costs were projected to future values considering the inflation and purchasing power parity, sinking fund factor for the conversion of the future value to annual averages, etc. The list of requirements used for cost estimation is shown in Table 4.

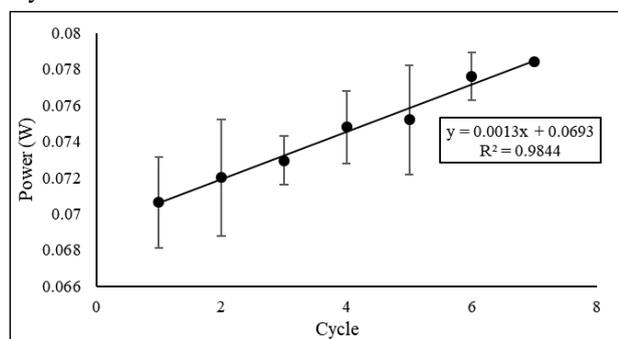
**Table 4.** List of cost estimation requirements and assumptions

Economic Consideration	Cost (Php)	Source	Unit Cost (Php/L)
<b>Capital Costs</b>			
Power supply and EC tank installation	280,996.00	[6]	0.0112
<b>Operating Cost</b>			
Maintenance and depreciation, per m <sup>3</sup>	0.26	[13]	0.0002
Electricity, per kWh	10.56	[14]	0.0037
Labor costs, per m <sup>3</sup>	5.22	Derived from DOLE daily rate [15]	0.0409
Iron electrode, per kg	31.56	Canvass of materials	0.0158
Chemicals (acid, salt, etc.), per m <sup>3</sup>	2.09	[11]	0.0017
Sludge disposal, per kg	0.63	[11]	$5.22 \times 10^{-6}$

### 3 Results and Discussion

#### 3.1 Power Requirement

It was determined that the power requirement increases with the cycles as shown in Fig. 2. This could be explained by the increase in thickness of the passivation layer.

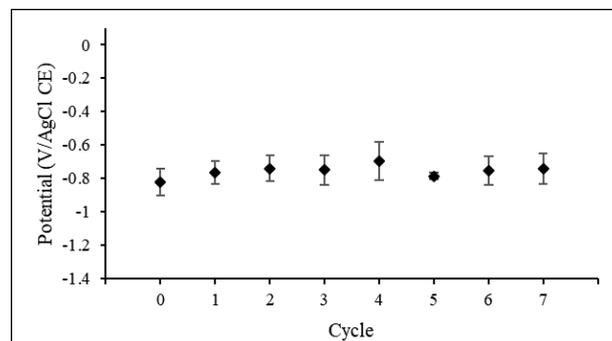


**Fig. 2.** Increase in power consumption per cycle

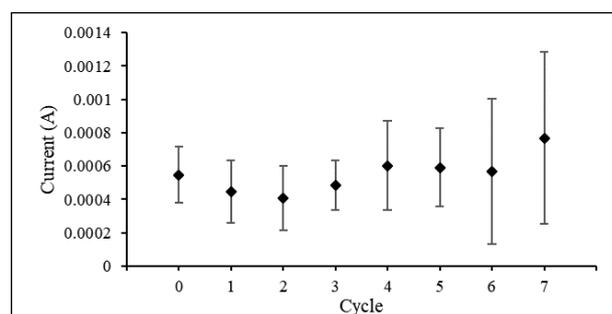
The power requirement of the ECAR process for a 200 mL setup starts from 0.0693 W. and increases per cycle by approximately 0.0013 W. Using this model, by the end of 100<sup>th</sup> cycle the approximate power requirement of the ECAR process is 0.1993 W, or equivalent to about 200% increase, which will later translate to increase in cost.

#### 3.2 Polarization and Charge Transfer Resistances

A linear sweep voltammetry is performed on the working electrodes using Metrohm Autolab and Nova 2.0 software to obtain the corrosion current and corrosion potential through Tafel extrapolation method.



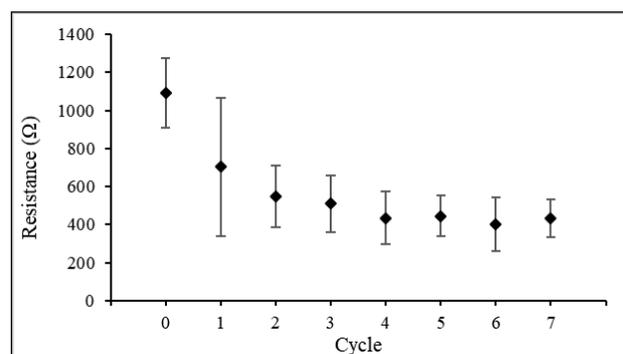
**Fig. 3.** Corrosion potential per cycle



**Fig. 4.** Corrosion current per cycle

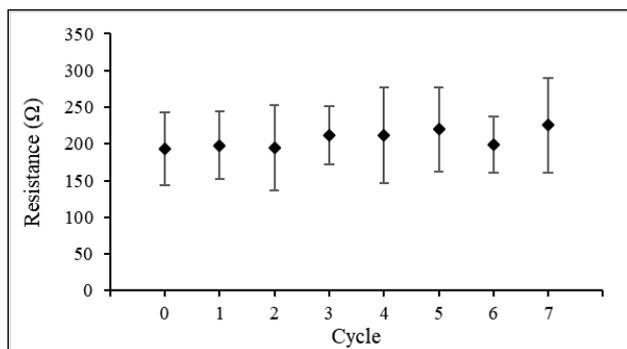
Figures 3 and 4 show the plot of the averages of the corrosion potential and the corrosion current for every cycle, respectively. The corrosion potential,  $E_{corr}$ , shows the state of a corroding metal, and as can be observed in the graph,  $E_{corr}$  generally increases with each cycle. On the other hand, the corrosion current,  $I_{corr}$ , reflects the instantaneous corrosion rate at the time of measurement. The general increase in corrosion current per cycle describes the increase in rate of corrosion of the anode per cycle.

The polarization resistance,  $R_p$ , is obtained by taking the inverse of the slope of the current potential curve at open circuit potential while the  $E_{corr}$  and  $I_{corr}$  are used in the computation of the charge transfer resistance,  $R_{ct}$  through Equation 2.



**Fig. 5.** Polarization resistance per cycle

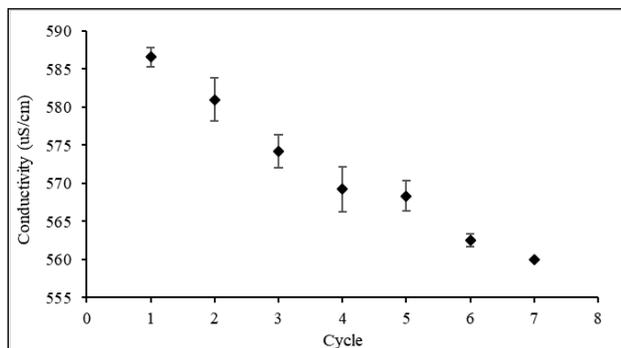
The averages of the  $R_p$  and  $R_{ct}$  in each cycle were plotted with their respective error bars. Figure 5 shows a decrease in polarization resistance in the working electrode through each cycle while Figure 6 gives an indication of an increase in the charge transfer resistance in the working electrode through each cycle, varying by approximately 10-20  $\Omega$ . The decrease in polarization resistance through each cycle implies low corrosion resistance of the specimen, which is expected due to the prolonged exposure to an external potential. The increase in charge transfer resistance on the other hand indicates that there is an increase in energy lost during charge transfer and an increase in the resistance necessary for the dosing of iron with each cycle. This proves that the growth of the passivation layer through time affects the total resistance of the system.



**Fig. 6.** Charge transfer resistance per cycle

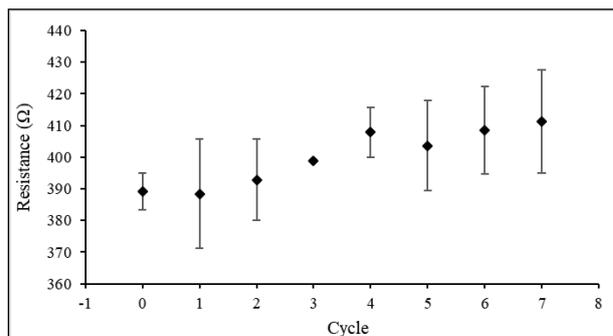
### 3.3 Conductivity

Conductivity of the synthetic groundwater before and after the electrocoagulation process is measured and plot of the averages of the conductivity per cycle is shown in Figure 7.



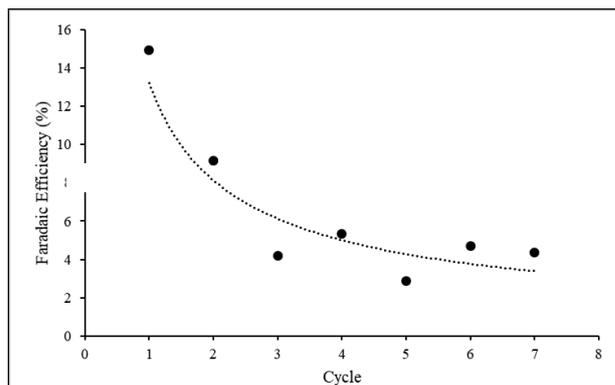
**Fig. 7.** Conductivity per Cycle

The conductivity of the system decreases with each cycle. This can be attributed to the decrease in dosing of iron due to the layer growth. The Ohmic resistance, which is indirectly proportional to conductivity, increases per cycle as shown in Figure 8. Through time, the solution resists the flow of electric current suggesting that the voltage must be increased in order to overcome it while keeping the current constant.



**Fig. 8.** Ohmic resistance per Cycle

### 3.4 Faradaic Efficiency



**Fig. 9.** Faradaic Efficiency for every cycle

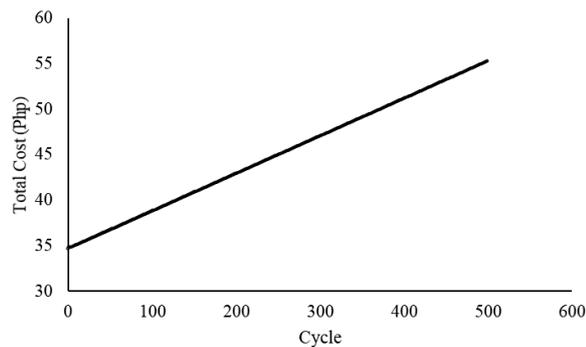
The dosing of the iron was determined to be decreasing with the cycles as suggested by the decreasing faradaic efficiency through each cycle as shown in Figure 9. There was an observed drastic drop in dosing at the first cycle and the trend continued a little less turbulent in the succeeding cycles.

### 3.5 Cost Estimation

The total cost per cycle of the ECAR process increases linearly. This is based on the power requirement model that was developed using a 200 mL bench scale ECAR reactor. By the time the electrode is already depleted halfway the increase in cost reaches around Php 20.

The total cost of the ECAR process is approximately Php 180,000 to 220,000 per year. The treated volume of groundwater is 2400 m<sup>3</sup> yearly and the electrode is expected to be changed eight times.

It was also observed that the labor fee (75%), the capital cost (19%) and the electricity cost (6%) contribute most to the cost of the EC process. This, however, could easily be subjected to change if the actual power usage will be used to compute. This should include all the power consumption of all other machines to be used in the actual process and taking into consideration their inefficiencies.



**Fig. 10.** Total cost vs cycle

The EC process costs around Php 0.06 at year one considering a fresh electrode. It increased to around Php 0.07 which is approximately 23.27% increase. As for the end of service life of the electrode, the cost per liter already hits around Php 0.12 which is already significantly high for the consumers as service charge.

### 3 Conclusion

The power requirement of the ECAR process for a 200 mL setup starts from 0.0693 W. and increases per cycle. After projecting the bench scale to a 600 L setup, the ECAR process approximately costs Php 0.06 at year one with a fresh electrode and increases to Php 0.07 which is approximately a 23.27 % increase. At the end of the assumed 10-year service life of the tank, the water treatment costs around Php 0.07 to Php 0.12 which is considerably high for a service charge.

The power consumption increases with time, possibly because of the build-up of the passivation layer. This is further supported by the quantification of resistances and calculation of efficiencies of the system.

The resistance due to the charge transfer was determined to be increasing with time. This could be a possible reason for the increase in power requirement. However, there was also an observed deposition of potentially impermeable oxide layers on the cathodes. This could also be a factor in the increase of power requirement. According to Eyvaz (2014), the cathode deposition decreases the ion transfer between the anode and cathode and the use of an alternating current was recommended to counteract the deposition through the occasional pitting due to the change in flow of electrons.

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