

**ELECTROCOAGULATION PROCESS USING RAW  
SEWAGE FOR TREATMENT AND ENERGY  
PRODUCTION**



**MASTER OF ENGINEERING (CIVIL ENGINEERING)**

**UNIVERSITI MALAYSIA PAHANG**

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**ELECTROCOAGULATION PROCESS USING RAW SEWAGE FOR  
TREATMENT AND ENERGY PRODUCTION**



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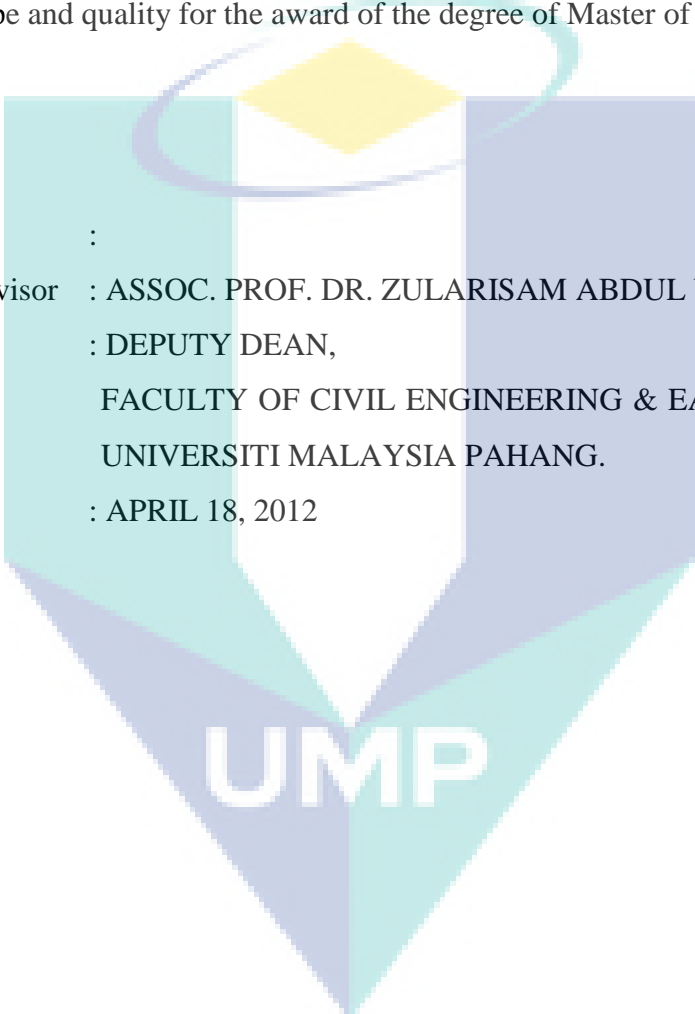
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For the award of the degree of  
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**UMP**  
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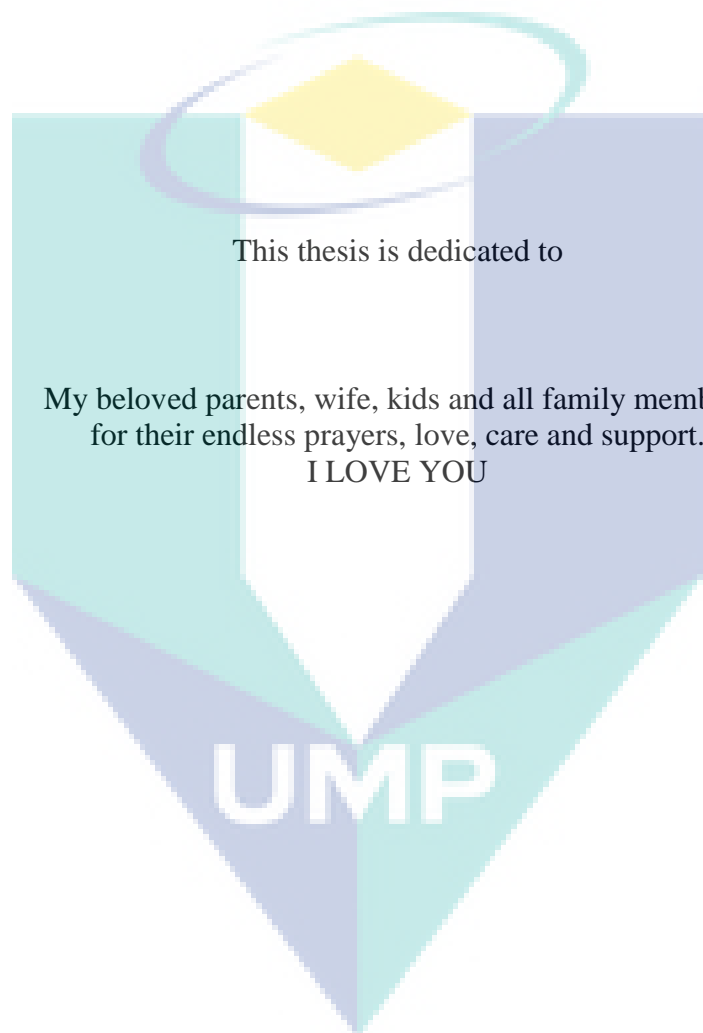
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## ABSTRACT

Due to the massive growth of Malaysian population, sewage water has become one of the major water pollution in Malaysia. Various physiochemical and biological technique have been employed to treat sewage water such as aerobic and anaerobic biological treatment, membrane filtration, adsorption, chemical coagulation and many others methods but either they took a long period of time or needed a large amount of chemical substance, both ways are less effective in treating sewage. The removal of pollutants from effluents by electrocoagulation (EC) has become an attractive method in recent years. This electrochemical process offer some distinctive advantages, including effects due to the production of polyvalent cations from the oxidation of corrodible anodes (like  $\text{Fe}^{2+}$  and  $\text{Al}^{3+}$ ) as a coagulant agent and the production of electrolysis gases. The gas bubbles can carry the pollutant to the top of the solution where it can be more easily collected and removed. Therefore, EC process has been studied to treat sewage water and to determine the effect of various operating parameters such as electrode types, operating time, current intensity, inter-electrode spacing and pH to evaluate the possible treatment efficiency. It was found that the application of high current intensity (20 A, 25A and 30 A) is more effective in treating sewage than using low current (1 A, 2A and 3 A). Aluminum hydroxypolymeric species formed during an earlier stage of the operation efficiently remove sewage molecules by precipitation, and in a subsequent stage,  $\text{Al}(\text{OH})_3$  flocs trap colloidal precipitates and make solid-liquid separation easier during the flotation stage. A Response Surface Method (RSM) showed that the optimum condition of COD, BOD and SS removal was obtained 29.09 A of current, 29.86 minutes of operating time, 10.71 mm of inter-electrode distance and pH value of 7.04. Light microscopy result shows more aggregation of sewage particles occurs with longer treatment period and higher current applied to the EC system. High production of hydrogen gas obtained from experiment in 60 seconds by high current application. The study is important to establish a sustainable clean energy in order to implement low cost treatment in sewage treatment.



## ABSTRAK

Disebabkan oleh percambahan besar penduduk Malaysia, air kumbahan telah menjadi salah satu daripada pencemaran air utama di Malaysia. Pelbagai teknik fisiokimia dan biologi telah digunakan untuk merawat air kumbahan seperti rawatan aerobik dan anaerobik secara biologi, penapisan membran, penjerapan, kaedah kimia dan banyak lagi kaedah lain tetapi kaedah tersebut mengambil tempoh masa yang panjang dan memerlukan sejumlah besar bahan kimia membuatkan kaedah tersebut kurang berkesan dalam merawat air kumbahan. Penyingkiran bahan cemar daripada air kumbahan oleh kaedah elektrokoagulan (EC) telah menjadi satu teknik yang menarik dalam beberapa tahun kebelakangan ini. Proses elektrokimia ini menawarkan beberapa kelebihan, contohnya pengeluaran kation daripada pengoksidaan oleh anode (seperti  $\text{Fe}^{2+}$  dan  $\text{Al}^{3+}$ ) sebagai agen pengentalan dan pengeluaran gas melalui elektrolisis. Buih gas boleh mengapungkan bahan tercemar ke atas permukaan membolehkan ia lebih mudah terkumpul dan dikeluarkan. Oleh itu, proses EC ini telah dikaji untuk merawat air kumbahan dan juga untuk menyiasat kesan parameter-parameter yang digunakan contohnya jenis elektrod, masa operasi, keamatan arus, jarak antara elektrod dan juga pH terhadap keberkesanan rawatan. Telah didapati bahawa penggunaan arus yang tinggi (20 A, 25A dan 30 A) adalah lebih berkesan dalam merawat air kumbahan daripada menggunakan arus yang rendah (1 A, 2A dan 3 A). Aluminium spesies hidroksipolimerik terbentuk di peringkat awal operasi dan cepat mengeluarkan molekul kumbahan melalui mendapan, dan di peringkat yang berikutnya,  $\text{Al}(\text{OH})_3$  memerangkap mendapan koloid lalu mudah untuk diapungkan. Response Surface Method (RSM) menunjukkan bahawa keadaan optimum bagi penyingkiran COD, BOD dan SS didapati pada penggunaan arus sebanyak 29.09 A, masa operasi selama 29.86 minit, jarak antara elektrod sejauh 10.71 mm dan nilai pH pada 7.04. Gambar hasil daripada penggunaan mikroskop cahaya menunjukkan lebih banyak pengumpulan zarah-zarah kotoran terhasil jika lebih panjang tempoh rawatan yang diberikan dan arus yang lebih tinggi digunakan untuk sistem EC tersebut. Pengeluaran gas hidrogen yang banyak diperolehi dengan menggunakan arus yang tinggi daripada satu eksperimen dalam 60 saat. Kajian ini adalah penting untuk merawat air kumbahan dengan cekap dan dalam masa yang sama dapat menghasilkan tenaga yang bersih dengan berlanjutan.

## TABLES OF CONTENTS

	<b>Page</b>
<b>THESIS CONFIDENTIAL STATUS</b>	ii
<b>SUPERVISOR’S DECLARATION</b>	iv
<b>STUDENT’S DECLARATION</b>	v
<b>DEDICATION</b>	vi
<b>ACKNOWLEDGEMENTS</b>	vii
<b>ABSTRACT</b>	viii
<b>ABSTRAK</b>	ix
<b>TABLES OF CONTENTS</b>	x
<b>LIST OF TABLES</b>	xiv
<b>LIST OF FIGURES</b>	xv
<b>LIST OF ABBREVIATIONS/SYMBOLS</b>	xvii
<b>CHAPTER 1            INTRODUCTION</b>	<b>1</b>
1.1            Introduction	1
1.2            Background of Study	1
1.3            Problem Statement	3
1.4            Objective of the Study	4
1.5            Scope of the Study	5
1.6            Significant of the Study	5
<b>CHAPTER 2            LITERATURE REVIEW</b>	<b>7</b>
2.1            Introduction	7
2.2            Wastewater	7

2.2.1	Definition of Wastewater	7
2.2.2	Characteristic of Wastewater	8
2.3	Sewage	8
2.3.1	Characteristic of Sewage	9
2.3.2	Quality Parameter of Water	10
2.4	Introduction of Electrochemical Process	11
2.5	Coagulation	11
2.6	EC Process	14
2.6.1	Theory of EC	19
2.6.2	Optimization of EC	20
2.6.3	EC Kinetic	20
2.7	Application of EC Process	21
2.7.1	Domestic Wastewater Treatment	21
2.7.2	Industrial Wastewater Treatment	22
2.8	Design of Experiment	22
2.8.1	Respond Surface Methodology	23
2.8.2	Central Composite Design	24
2.8.3	Analysis of the Data	26
2.9	Production of Hydrogen by EC Process	28
<b>Chapter 3</b>	<b>METHODOLOGY</b>	<b>30</b>
3.1	Introduction	30
3.1.1	Preparation of EC Process	32
3.1.2	Preparation of Electrochemical Cell	32
3.1.3	Preparation of Sewage Sample	33
3.2	Determine the Characteristic of Sewage	34
3.3	Effect of Electrode Material	35
3.4	Effect of Current and Contact Time	35
3.5	Effect of Inter-Electrode Distance	36
3.6	Effect of pH	36
3.7	Design of Experiment: Respond Surface Method	37

3.8	Determine the Production of Hydrogen by EC Process	39
3.9	Laboratory Test and Analysis	40
3.9.1	Light Microscopy	40
<b>CHAPTER 4</b>	<b>RESULT AND DISCUSSION</b>	<b>43</b>
4.1	Introduction	43
4.2	Characteristic of Sewage Water	43
4.3	Effect of Electrode Material	44
4.4	Effect of Current and Contact Time	47
4.4.1	Observation of Organic and Inorganic Removal in EC Process	47
4.4.2	Explanation of Organic and Inorganic Removal in EC Process	52
4.5	Effect of Inter-Electrode Distance	56
4.6	Effect of pH	59
4.7	Experimental Design for RSM	63
4.7.1	Development of Empirical Model Equation for EC Process Preparation	65
4.7.2	Effect of Single Process Variables	71
4.7.2.1	Current Application ( $x_1$ )	72
4.7.2.2	Operating Time ( $x_2$ )	73
4.7.2.3	Inter-electrode Distance ( $x_3$ )	74
4.7.2.4	pH ( $x_4$ )	75
4.7.3	Combine effect on COD, BOD and SS	76
4.7.4	Optimization of experimental parameters	81
4.8	Hydrogen Gas Production by EC Process	85
4.9	Light Microscopy Results	87
4.10	EC Kinetic	90
<b>CHAPTER 5</b>	<b>CONCLUSION AND RECOMMENDATION</b>	<b>95</b>

5.1	Introduction	95
5.2	General Conclusion	95
5.3	Recommendation	98
<b>REFERENCES</b>		99
<b>APPENDICES</b>		106
A1	Removal percentage of COD, BOD and SS at 1A, 10 mm inter-electrode distance, pH 7 by using aluminum electrode	106
A2	Removal percentage of COD, BOD and SS at 2A, 10 mm inter-electrode distance, pH 7 by using aluminum electrode	107
A3	Removal percentage of COD, BOD and SS at 3A, 10 mm inter-electrode distance, pH 7 by using aluminum electrode	108
A4	Removal percentage of COD, BOD and SS at 20A, 10 mm inter-electrode distance, pH 7 by using aluminum electrode	109
A5	Removal percentage of COD, BOD and SS at 25A, 10 mm inter-electrode distance, pH 7 by using aluminum electrode	110
A6	Removal percentage of COD, BOD and SS at 30A, 10 mm inter-electrode distance, pH 7 by using aluminum electrode	111
B1	Electrochemical cell model	112
B2	Incubator	113
B3	Spectrophotometer HACH DR 5000	114

## LIST OF TABLES

Table No.	Title	Page
2.1	Acceptable conditions for sewage discharge of Standards A and B	10
2.2	Central composite designs	25
3.1	Sewage strength in terms of COD and BOD <sub>5</sub>	34
3.2	Experimental design matrix	38
4.1	Characteristic of raw sewage water	43
4.2	The effect of different types of electrodes material on sewage's parameters	44
4.3	Value of COD (mg/L)	47
4.4	Value of BOD (mg/L)	49
4.5	Value of SS (mg/L)	51
4.6	Weight dissolution of aluminum by experiment	53
4.7	Weight dissolution of aluminum by theory	53
4.8	Faradic efficiency of aluminum electrode dissolution	54
4.9	The effect of inter-electrode distance on sewage's parameters	57
4.10	The effect of pH on sewage's parameters	59
4.11	Complete design matrix of EC process	64
4.12	ANOVA for response surface quadratic model for COD removal	67
4.13	ANOVA for response surface quadratic model for BOD removal	68
4.14	ANOVA for response surface quadratic model for SS removal	69
4.15	Optimization result of variables and responses	81
4.16	Hydrogen gas production by different current applied	85

## LIST OF FIGURES

<b>Figure No.</b>	<b>Title</b>	<b>Page</b>
2.1	Destabilization and aggregation of particles.	12
2.2	Three type of central composite design	26
3.1	Flow chart of research framework	31
3.2	Electrochemical cell setup	33
3.3	Incoming flow of the raw sewage	34
3.4	Type of electrodes material	35
3.5	Water displacement method	40
3.6	Meiji Techno TC5200 Brightfield Inverted Microscope, Japan	41
4.1	Sewage sample	44
4.2	Comparison of sample sewage water treated by different electrode	46
4.3	Percentage of COD removal	48
4.4	Percentage of BOD removal	50
4.5	Percentage of SS removal	52
4.6	Experimental versus theoretical sacrificial electrode	55
4.7	Specific area decreases with the diameter of gas bubbles	56
4.8	Effect of inter-electrode distance on removal efficiency	58
4.9	Effect of pH on removal efficiency, using 30 A current applied at 30 minutes	61
4.10	pH change during EC process	61
4.11	Predicted versus experimental percentage of COD removal	70
4.12	Predicted versus experimental percentage of BOD removal	71
4.13	Predicted versus experimental percentage of SS removal	71
4.14	Individual effect of current applied	72
4.15	Individual effect of operating time	73
4.16	Individual effect of electrode distances	74
4.17	Individual effect of pH	75

4.18	3D surface plot for COD removal efficiency	78
4.19	3D surface plot for BOD removal efficiency	79
4.20	3D surface plot for SS removal efficiency	80
4.21	Optimization in 3D plot of COD removal efficiency	82
4.22	Optimization in 3D plot of BOD removal efficiency	83
4.23	Optimization in 3D plot of SS removal efficiency	84
4.24	Microscopic images (20X) of sewage particles before treatment, 5 minutes and 30 minutes of treatment at 30 A	87
4.25	Microscopic images (20X) of sewage particles at 30 minutes at variety of current applied	89
4.26	Effect of current on rate constant, k	91
4.27	Effect of electrode distance on rate constant, k	92
4.28	Effect of pH on rate constant, k	92
4.29	Pseudo-first order kinetic of EC, current 30 A; pH 7; Electrode distance 10 mm	93
4.30	Pseudo-second order kinetic of EC, current 30 A; pH 7; Electrode distance 10 mm	94

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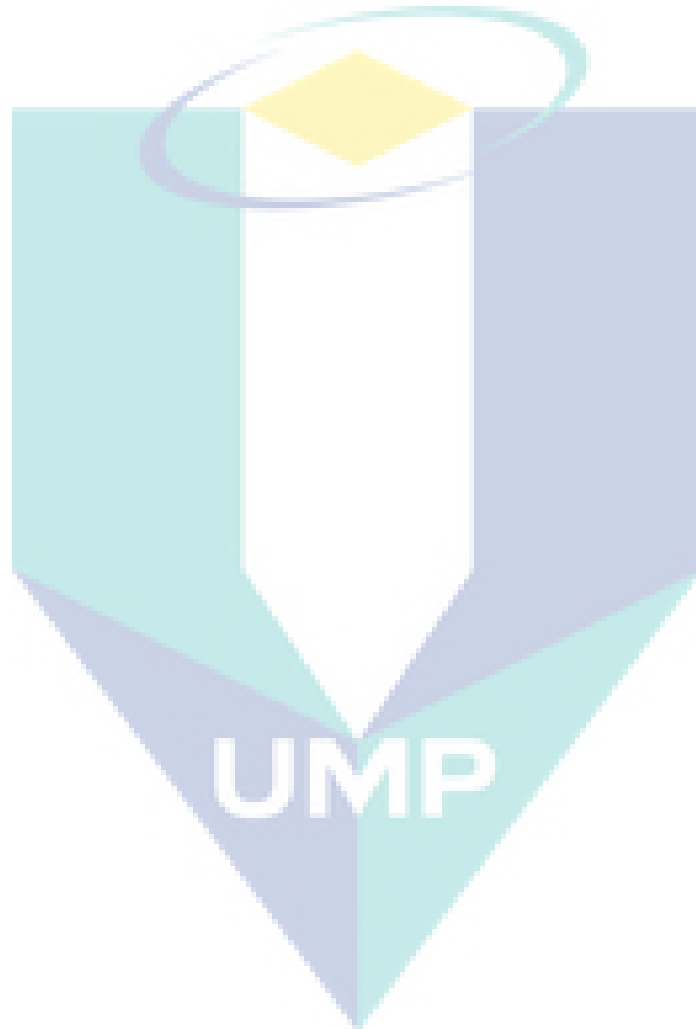
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**LIST OF ABBREVIATIONS/SYMBOLS**

BOD	Biological Oxygen Demand
COD	Chemical Oxygen Demand
CCD	Central Composite Design
CCF	Central Composite Face Centered
CCI	Central Composite Inscribed
CCR	Central Composite Rotatable
DoE	Design of Experiment
EC	Electrocoagulation
IWK	Indah Water Konsortium
R.S.D	Relative Standard Deviation
RSM	Response Surface Methodology
SS	Suspended Solid
STP	Sewage Treatment Plant
A	Active anode surface
g	Gram
<sup>0</sup> C	Degree Celsius
F	Faraday's Constant
I	Current (A)
$\kappa$	specific conductivity
k	Rate Constant
L	Liter
M	Molecular Weight
mg	Miligram
mL	Mililiter

min	minute
$\mu\text{m}$	Micrometer
sec	Second
t	Time
w	Weight



## **CHAPTER 1**

### **INTRODUCTION**

#### **1.1 INTRODUCTION**

This chapter explains the important of water, sewage nature, the harmful of sewage water to the environment, recent treatment method for sewage, EC process and the advantages of treatment method. The problem statement, objectives the scopes of study and the significance of study were also described.

#### **1.2 BACKGROUND OF THE STUDY**

Pollution in its broadest sense includes all changes that curtail natural utility and exert deleterious effect on life. The crisis triggered by the rapidly growing population and industrialization with the resultant degradation of the environment causes a great threat to the quality of life. Degradation of water quality is the unfavorable alteration of the physical, chemical and biological properties of water that prevents domestic, commercial, industrial, agricultural, recreational and other beneficial uses of water (Ahmed, 1985).

Industrial discharges, agricultural run-off, municipal waste and oil spills are all contribute to decline in water quality. One important source of water pollution is human sewage discharge which enters the Malaysia's coastal waters from several sources. Sewage

is mainly composed of human faecal material, domestic wastes and industrial wastes. If these are discharged into river or sea without proper treatment, this may cause threat to the aquatic life and other consumers (Luthfaera, 1995). Everyday a large volume of sewage is being discharged into various rivers without proper treatment. Malaysia will face a severe water crisis sooner or later, unless water management practices are changed.

Sewage usually discharge from kitchen, bathroom, toilets, sinks, garbage-grinding and laundry sources. It usually contains a wide variety of contaminants, such as dissolved and suspended impurities, nutrients, biodegradable organics, refractory organics, pathogens and heavy metals. It amounts to a very small fraction of the sewage by weight, but it is large by volume and contains many impurities. Improper discharge of untreated sewage into the natural water bodies is definitely undesirable, as it would seriously deteriorate the water quality by the decomposition of the organic waste.

A common problem is the ability to control high BOD/COD levels in a system. Traditional methods trend to either increase the oxygen levels (dissolved oxygen) and or increase the retention capacity. This is typically the engineering solution and always not a solely problem solving. Sewage-contaminated water can cause eutrophication in open environment. Eutrophication is a serious problem which increase the concentration of chemical elements required for life. The nitrates, phosphates, and organic matter found in human waste serve as a food for algae and bacteria. This causes these organisms to overpopulate to the point where they use up most of the dissolved oxygen that is naturally found in water, making it difficult for other organisms in this aquatic environment to live especially in stagnant waters. The bacteria are basically strangling the other organisms. Some of the organisms that do overpopulate from this can also be disease-causing microorganisms.

These days, various physiochemical and biological technique have been employed to treat sewage water such as aerobic and anaerobic biological treatment, membrane filtration, adsorption, chemical coagulation and many others methods. Although the treatment techniques that mentioned above can be applied as the efficient methods, either

they took a long period of time or needed a large amount of chemical substance, both ways are less effective in treating sewage water where it requires a short time treatment and an environmental compatibility with free chemical effect.

Electrocoagulation (EC) process is one of electrochemical application that has been attracted a great attention in treating wastewater because of the versatility and the environmental friendly as it shall not create corrosion or any pollutants (Akyol, 2012). A successful application of EC technique for the removal of COD, BOD and SS from wastewater would address the environmental needs of reduction in the operational costs and potential saving in processing unit. Among different physical and chemical methods of water and wastewater treatments, this method offers a special attraction due to its safety, simplicity and low operating costs (Sohaili. J, 2004). This study has been done to treat sewage water by using EC process as this technique was proven can treat wastewater effectively.

Moreover, EC process also can produce fuel as a renewable energy which is hydrogen. People are using more and more nonrenewable resources for energy such as fossil fuels which may not long lasting. So, other sources must be found to at least partly replace fossil fuels. Electrochemical process is one of the simplest and easiest ways to produce hydrogen. Hydrogen is the future fuel due to its high conversion efficiency, recyclability and nonpolluting nature.

### **1.3 PROBLEM STATEMENT**

There are many challenges involves in the process of treating sewage, not only focus on the technical point of optimizing the water quality and solid/liquid separation. In developing a variable wastewater treatment method, several factors must be included such as environmental impact, ease of installation and operation, usefulness in various industrial applications, energy efficiency, cost-effective and widely applicable to the various pollutant ranges.

Nowadays, enormous technology of sewage water treatment units are installed all over the world and each having its advantages and disadvantages. This study aims to explore the use of EC, a process that was developed over a century ago for treating wastewater. Conventional treatment methods often induce a chemical reaction through the use of coagulants, flocculants, and other additives that aid in the removal or sedimentation of contaminants. In physical-chemical treatment units, sludge conditioning also requires the addition of polymers which change the properties of the sewage water and might decrease biodegradability (Bratby, 2006). Besides, by using these conventional treatment methods result in high sludge excess, which in turn would increase the treatment cost. Some of the treatments require extra land acquisition that significantly increases the overall cost.

Therefore a small, localized water treatment technology is required to reduce the consumption of energy as well as cost. EC is the possible best solution to treat sewage and it is stand-alone portable treatment with the ability to remove a wide range of pollution. Other than low management cost, this technology also safe and natural environmentally friendly. Therefore, it makes economic and practical sense to use it in treating the province's wastewater, in lieu of using chemicals that require storage, transportation, specific handling practices and in addition it also producing secondary pollution.

#### **1.4 OBJECTIVE OF THE STUDY**

This research project is based on these objectives:

- a) To determine the effect of electrocoagulation (EC) parameters such as current intensity, electrode type, treatment time, inter electrodes distance and pH on the sewage characteristics.
- b) To determine the optimum operational EC performance Method (RSM) using Response Surface analysis.
- c) To determine the effect of EC process on hydrogen production.

## 1.5 SCOPE OF THE STUDY

Sewage wastewater was collected from Indah Water Konsortium (IWK) which located at Indera Mahkota, Kuantan, Pahang. Electrochemical cell was designed by using stainless steel cuboids box with monopolar electrodes (120 mm x 100 mm x 2 mm). In order to find the effect of electrode types on the treatment, aluminum, iron and stainless steel were used in this study. Two sets of current were used to find the effect of current intensity which was 1, 2, 3 A as low current and 20, 25, 30A as high current and to find their comparison. Time was varied every 5 minutes from 0 to 30 minutes to find the effect of time on treatment process and 60 seconds was used to determine the hydrogen production. The effect of Inter-electrode distance was carried out by varying the gap between electrodes every 5 mm from 5 to 30 mm in order to find the effect of inter-electrode distance on the treatment. pH effect was examined by varying pH value from 3 to 11. Response Surface Methodology (RSM) by using central composite design (CCD) was applied from a statistical software Design Expert software version 6.0.8 (STAT-EASE Inc., Minneapolis, USA).

## 1.6 SIGNIFICANCE OF THE STUDY

The study is to provide an alternative method for the treatment of sewage using EC process at high current intensity application. Application of EC technique is more environmentally friendly and could contribute to clean water supply for healthy human living and at the same time enable adoption of green technology in producing sustainable clean energy (hydrogen) for future energy requirement.

Although there have been a number of studies on EC process for wastewater treatment, the aspect of hydrogen recovery from the EC process has less been studied previously. Hydrogen is one of the important products generated from this technology. Hydrogen is a main by-product of the EC process as it is produced at the cathodes by water electrolysis. An electrochemical cell is a device capable of converting the chemical energy

contained in wastewater into hydrogen while reducing its organic load with an input of electricity. With an effective gas-liquid-solid separation process, high quality hydrogen can be recovered from the EC process and used as an energy source or as a reactant for industrial processes.

Hydrogen is considered as an energy carrier like electricity and produces no greenhouse gas or carbon dioxide when burnt in the presence of oxygen in related appliances including fuel cell or combustion engines. Hydrogen can be produced from different feedstock using a variety of techniques. Hydrogen is currently produced in large quantities from natural gas. Although, it is the cheapest way at present to produce hydrogen but the presence of carbon in methane is contributing to increase the global warming. A challenging problem in establishing  $H_2$  as a source of energy for the future is to establish the procedures to produce hydrogen in abundance without creating any environmental threats. This chapter will emphasize on the treatment of sewage water and simultaneous hydrogen production using EC process.

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## CHAPTER 2

### LITERATURE REVIEW

#### 2.1 INTRODUCTION

This chapter provides a review of wastewater, sewage and its characteristic, electrochemical process, coagulation and EC process and its application and energy production.

#### 2.2 WASTEWATER

Wastewater is any water that has been adversely affected in quality by anthropogenic influence. It comprises liquid waste discharge by domestic residences, commercial properties, industries, or agriculture and can encompass a wide range of potential contaminant and concentration. In the most common usage, it refers to the municipal wastewater that contains a broad spectrum of contaminant resulting from the mixing of wastewater from different sources (Salt *et al*, 1995).

##### 2.2.1 Definition of Wastewater

Generally wastewater is synonymously with sewage even though sewage is a more general term that refers to any polluted water including wastewater, which may contain

organic and inorganic substance, industrial waste, groundwater that happens to infiltration and to mix with contaminated water, storm, runoff, and other similar liquids (Miretzky *et al*, 2004). In other words, wastewater can be describe as using the water with dissolved or suspended solid, discharge from homes, commercial establishment, farms and industries (Rock, 1997).

### **2.2.2 Characteristic of Wastewater**

In general, wastewater is water that has been generated from domestic and industrial sources where throughout the world by dumping 10,000 new organic compounds each year. These compound needs to be properly handled and removed if they cause health problem. There are many industrial plants that have required pre-treating their wastewater before dumping in the wastewater system (Chandra, 2004).

### **2.3 SEWAGE**

Sewage is the main point-source pollutant on a global scale (Gijzen, 2002). There are a lot of the sewage produced in the world is released into the environment without any treatment (Bartone *et al*, 1994). Wastewater or natural water supplies into which wastewater has been discharged, are likely to contain pathogenic organisms similar to those in the original human excreta. Disease prevention programs have centered upon four groups of pathogens potentially present in such wastes: bacteria, viruses, protozoa and helminthes and these pathogenic organisms normally found in human excreta and sewage wastewater (Feachem *et al*, 1983). Simple, affordable, and efficient sewage treatment systems are urgently needed in developing countries because most of the conventional technologies currently in use in industrialized nations are too expensive and complex (Grau, 1996). Appropriate and sustainable sewage treatment technologies will help to preserve biodiversity and maintain healthy (freshwater) ecosystems, in order to provide clean water, flood control, abundant fisheries, and other services of vital interest to human societies.

### 2.3.1 Characteristic of Sewage and Domestic Wastewater

Sewage is characterized in terms of its physical, chemical and biological composition. The important physical characteristic of sewage is its total solid content, which is composed of floating matter, settle able matter, colloid matter, and matter in solution. Other important physical characteristic include odor, temperature, color, total solid and turbidity.

For the chemical properties typically classified by three constituents which are organic, inorganic and in gaseous state. Normally organic compounds of sewage contain of carbohydrates, fats, oil, grease, proteins, and surfactants. Other important elements, such as sulfur phosphorus, and iron, may also be present (Rock, 1997). Not only that, sewage water may contain small quantities of a large number of different synthetic organic molecules ranging from simple to extremely complex in structure. This may be due to sewage with high industrial waste element which consist of compounds and possibly include highly toxic chemicals. For example include surfactants, organic priority pollutant, agricultures pesticides and volatile organic compounds. The presence of these substances has complicated wastewater treatment since several have been found to be resistant to biodegradation (Snyder *et al*, 2004)

Meanwhile, inorganic constituents in water can be divided into individual elements such as Zinc (Zn), iron (Fe), chloride (Cl), and a wide variety of compound, such as, nitrate ( $\text{NO}_3$ ) and sulfate ( $\text{SO}_4$ ). Meanwhile, hydrogen sulfide, methane, oxygen are include in the gasses properties of sewage. Several inorganic components of wastewater are important in establishing and controlling wastewater quality. Concentration of inorganic constituents also increased by the natural process, which removes some of surface water and leaves the inorganic substance in wastewater (Llorens *et al*, 2000).

### 2.3.2 Quality Parameter of Water

In order to manage the water quality in Malaysia, The Environment Quality Act (1974), Environment Quality (Sewage) Regulation 2009 specifies two standard of effluent discharge from treatment plants to receiving waters. These take the form of acceptable upper limits for various effluent contaminants. Therefore, every sewage treatment plant (STP) shall be designed to meet either Standard A or B base on compliance to the proposed revised effluent standard in Table 2.1.

**Table 2.1:** Acceptable conditions for sewage discharge of Standards A and B

Parameter	Unit	Standard	
		A	B
(a) Temperature	°C	40	40
(b) pH Value	–	6.0-9.0	5.5-9.0
(c) BOD <sub>5</sub> at 20°C	mg/L	20	50
(d) COD	mg/L	120	200
(e) Suspended Solids	mg/L	50	100
(f) Oil and Grease	mg/L	5.0	10.0
(g) Ammoniacal Nitrogen (enclosed water body)	mg/L	5.0	5.0
(h) Ammoniacal Nitrogen (river)	mg/L	10.0	20.0
(i) Nitrate – Nitrogen (river)	mg/L	20.0	50.0
(j) Nitrate – Nitrogen (enclosed water body)	mg/L	10.0	10.0
(k) Phosphorous (enclosed water body)	mg/L	5.0	10.0

Source: The Environment Quality Act (1974),  
Environment Quality (Sewage) Regulation 2009

## 2.4 INTRODUCTION OF ELECTROCHEMICAL PROCESS

Electrochemical process is a technique where the electric current is charged through electrode resulting in different chemical reaction. The oxidation occurs at the anode where the oxygen gas is evolving and the reduction at the cathode with H<sub>2</sub> production. The anode also serve as a sacrificial electrode in that insoluble metal electrode is dissolve and soluble

metal ion emigrate to the bulk solution and act as coagulating agent. The liberated  $H_2$  gas bubbles serve as an excellent floatation agent and carry the suspended particles to the surface. Additionally,  $O_2$  gas produced at anode is able to oxidize organics (Fuad, 2008).

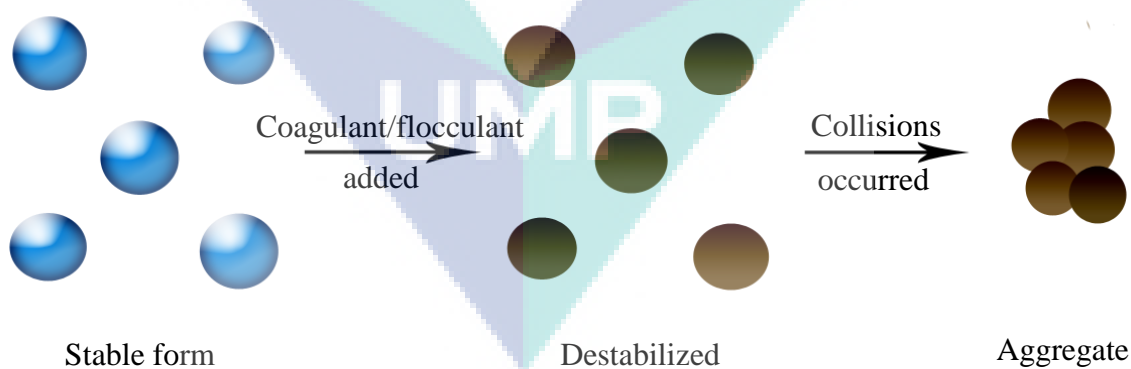
The expensive electricity supply and the huge capital investment always make the electrochemical treatment an insignificant choice (Kobya *et al*, 2003), but due to extensive research in US and USSR during the following half century, this technology has gained a large amount of knowledge (Rincon, 2011). By the stringent regulation regarding the wastewater discharge, the technology has regained its importance worldwide during the past two decades (Chen *et al*, 2000). By the latest technologies, reduction of electricity requirements, and miniaturization of the needed power supplies, electrochemical systems have now become a better choice of water treatment plants and industrial processes worldwide and also to the extent that they have bothered the market penetration of powerful chemical's coagulant manufacturing. Environmental compatibility, versatility, energy efficiency, safety, selectivity, amenability to automation, and cost effectiveness are the benefits of using electrochemical processes (Rajeshwar *et al*, 1994). Electrochemical based systems allow controlled and rapid reactions, smaller systems become viable and, instead of using chemicals and micro-organisms, the systems employ only electrons to facilitate water treatment.

## 2.5 COAGULATION

Coagulation is an essential physio-chemical process used in water treatment. Water clarification with coagulating agents has been practiced since ancient times, using a variety of substances. Crushed seeds being the most notable among the Egyptians, as early as 2000 BC, they used almonds smeared around a vessel to clarify river water. The early Romans were also familiar with alum, though it may not have been for water treatment (Bratby, 2006). In modern water treatment, coagulation and flocculation are still essential steps in the treatment processes. The need for a profound understanding of coagulation-flocculation processes is as important today as it has been in the past-even more so since today's waters

used for public supplies have an increased number of particulates and organics, further necessitating the effective removal of such substances.

Coagulation is the process of destabilizing, aggregating, and binding for ease of sedimentation by chemical or electrical methods. This is achieved by adding simple salts or by charge neutralization, resulting in a tendency in aggregation the particles to be small and dense. Flocculation is limited to the cases where polymer bridging is the dominant mechanism and aggregates (flocs) tend to be larger and more open in structure. It is natural that larger structure tends to be more open and less dense. The distinction between small, compact coagulation and larger, less dense flocculation is an inevitable result of the stronger interparticle binding in the case of polymers, causing to larger aggregates. In the area of water and wastewater treatment, coagulation refers to destabilization, by the dosing of appropriate additives, and flocculation refers the formation of aggregates, usually by some form of fluid motion. This corresponds to the two stages shown in Figure 2.6 and could be considered chemical and physical aspects of the aggregation process (Bratby, 2006).



**Figure 2.1:** Destabilization and aggregation of particles.

The additives used to cause destabilization of colloids may be called coagulants or flocculants, depending on their mode of action. Coagulants would be inorganic salts,

including those containing specifically adsorbing counter-ions, and flocculants would be long-chain polymers, which give bridging interactions. The mechanism of coagulation has been the subject of continual review. It is generally accepted that coagulation is brought about primarily by the reduction of the net surface charge to a point where the colloidal particles, previously stabilized by electrostatic repulsion, can approach closely enough for van der Waal's forces to hold them together and allow aggregation. The stability of a pollutant is determined by its physicochemical properties. Many pollutants are composed of similarly charged particles that repel each other, with the repulsive forces creating a stable, colloidal system. In order to maintain electroneutrality, oppositely charged ions, typically hydroxyl (OH<sup>-</sup>) or hydrogen ions (H<sup>+</sup>), are attracted to the (charged) pollutant particles. The attraction of counter ions to a negatively charged pollutant forms an electric double layer divided into a Stern and diffuse layer (Lyklema, 1978; Letterman *et al*, 1999).

Although there are potentially many different kinds of destabilizing agents the vast majority of those used in practice can be divided into two categories:

- a) Hydrolyzing metal coagulants
- b) Polymeric flocculants

Hydrolyzing metal coagulants are the main point of interest in the study. Alum [Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>] is such a chemical substance and one of the example of hydrolyzing metal coagulants, which has been widely used for ages for wastewater treatment. The precipitation of ions (heavy metals) and colloids (organic and inorganic) are mostly held in solution by electrical charges. By the addition of ions with opposite charges, these colloids can be destabilized. The coagulant is added in the form of suitable chemical substances.

The reduction of the surface charge is a consequence of the decrease of the repulsive potential of the electrical double layer by the presence of an electrolyte having opposite charge. In the EC process, the coagulant is generated in situ by electrolytic oxidation of an appropriate anode material. In this process, charged ionic species - metals or otherwise - are removed from wastewater by allowing it to react with an ion having an opposite charge, or with floc of metallic hydroxides generated within the effluent.

## 2.6 EC PROCESS

Coagulants are either directly added as a salt like aluminum sulfate or produced by electrochemical reactions at the anodes. The former is called conventional coagulation and the coagulant added is called the chemical dosage. The latter is called EC, and coagulant produced is called electrochemical dosage. Electrochemical processes have been used in water and wastewater treatment since 1887, beginning in England, and then followed by other countries. In the processes in those times both aluminum and iron were used together as anodes which corroded during electrolysis. In recent years, however, smaller scale EC processes have found a niche in the water treatment industry, proving to be reliable and effective technologies, though they require greater technical understanding for their potential to be fully exploited (Holt *et al*, 2002).

EC is a complicated process involving many chemical and physical phenomena that make use of consumable electrodes to supply ions into the pollutant system. In an EC process the coagulating ions are produced “in situ” and it involves three successive stages:

- a) Formation of coagulants by electrolytic oxidation of the “sacrificial electrode”, such as aluminum and iron,
- b) Destabilization of the contaminants, particulate suspension, and breaking of emulsion,
- c) Aggregation of the destabilized phases to form flocs.

The destabilization mechanism of the contaminants, particulate suspension, and breaking of emulsions consists of many steps, and may be summarized as follows:

- a) Compression of the diffuse double-layer around the charged species by the interactions of ions generated by dissolution of the sacrificial anode.
- b) Charge neutralization of the ionic species present in polluted medium by the counter ions, produced during the electrochemical dissolution of the sacrificial anode. These



counter ions reduce the electrostatic interparticle repulsion to the extent that the van der Waals attraction predominates, thus leading coagulation.

- c) Floc formation; coagulation results in the formation of the floc, thus creating a sludge blanket that entraps and bridges colloidal particles still remaining in the aqueous medium (Mollah *et al*, 2004).

The solid oxides, hydroxides and oxyhydroxides provide active surfaces for the adsorption of the polluting species (Mollah *et al*, 2004). EC has been successfully employed in removing metals, suspended particles, clay minerals, organic dyes, oil and greases from a variety of industrial effluents. In this process, a potential is applied to the metal anodes, typically fabricated from either iron or aluminum, which causes two separate reactions:

- a) Iron or Aluminum is dissolved from the anode generating corresponding metal ions, which almost immediately hydrolyze to polymeric iron or aluminum hydroxide. These polymeric hydroxides are excellent coagulating agents. The consumable (sacrificial) metal anodes are used to continuously produce polymeric hydroxides in the vicinity of the anode. Coagulation takes place when these metal cations combine with the negative particles carried toward the anode by electrophoretic motion. Then they are removed by electroflotation, or sedimentation and filtration. Hence, instead of adding coagulating chemicals as in conventional coagulation process, these coagulating agents are generated in situ.
- b) Water is also electrolyzed in a parallel reaction, producing small bubbles of oxygen at the anode and hydrogen at the cathode. These bubbles attract the flocculated particles and, through natural buoyancy, float the flocculated pollutants to the surface (Mollah *et al*, 2004). In addition, the following physiochemical reactions may also take place in the EC cell (Paul, 1996 as cited in Mollah, *et al*, 2004):
  - Cathodic reduction of impurities present in the medium.
  - Discharge and coagulation of colloidal particles.

- Electrophoretic migration of the ions in solution.
- Electroflotation of the coagulated particles by O<sub>2</sub> and H<sub>2</sub> bubbles produced at the electrodes.
- Reduction of metal ions at the cathode.
- Other electrochemical and chemical processes (Mollah *et al*, 2004).

In an EC experiment the electrode or electrode assembly is usually connected to an external DC source. The amount of metal dissolved is dependent on the quantity of electricity passed through the electrolytic solution. Faraday's law can be written simply between current and the amount of substance dissolved.

$$w = \frac{ItM}{nF} \quad (2.2)$$

where,

$w$  = aluminum dissolved (g Al);

$I$  = current (A);

$t$  = time (s);

$M$  = molecular weight of Al ( $M = 27$ );

$n$  = number of electrons involved in the oxidation reaction ( $n = 3$ );

$F$  = Faraday's constant, 96484 C/mol.

An agreement is expected to be seen between the calculated amounts of substance dissolved as a consequence of definite quantity of electricity and experimental value. Usually a good agreement is obtained. However, significant error may be introduced, if sufficient attention is not given to the geometry of the electrode as well as the optimum operating conditions of the EC cell (Vik *et al*, 1984).

EC is the system that has been existed over a hundred years (Dieterich, 1906). The concept of using electricity as a source to treat water was first proposed in United Kingdom, England in 1889 (Strokach P.P., 1975), and then followed by other country.

Treatment of wastewater by EC has been practiced for most of the 20th century with limited success (Daneashvar *et al*, 2006). EC has been reported to offer various advantages over conventional coagulation, specifically in relation to hydraulic retention time, removal efficiency of ultrafine particles, coagulant dosage, sludge production, cost, and operating and maintenance simplicity (Vik *et al*, 1984). An extensive amount of literature has been published on EC in conjunction with other processes, such as dissolved air flotation (Pouet and Grasmick, 1995), electroflotation (Chen *et al*, 2000), high gradient magnetic filtration (Tsouris *et al*, 2001), and the use of a settling tank (Jenke and Diebold, 1984), to cope with various wastewater. Few works (Beck *et. al*, 1974; Rubach and Saur, 1997) have been reported on the EC process that directly caused colloids to be floated by hydrogen bubbles generated in the cathodes by the electrolysis of water.

In EC process, the appropriate distance between anode and cathode has to be considered important in order to achieve the optimum treatment. The increase of inter-electrode distance will make the cell potential (V) increases which also increase the resistance and adversely affect the treatment. According to Ohm's law, the amount of electric current through a metal conductor in a circuit is directly proportional to the voltage impressed across it, for any given temperature. This relationship can be expressed as:

$$V = IR \quad (2.3)$$

Ohmic potential drop or IR drop can have a significant influence on electrochemical measurements. Ohmic potential drop is potential drop due to solution resistance. The difference in potential required to move ions through the solution. The variation in IR drop is governed by equation 2.4:

$$\eta_{IR} = I \cdot \frac{d}{A \cdot \kappa} \quad (2.4)$$

Where:

$I$  = current (A)

$d$  = distance between cathode and anode (mm)

$A$  = active anode surface ( $\text{mm}^2$ )

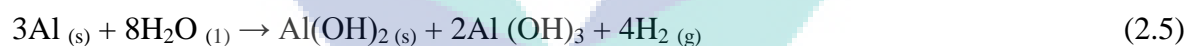
$\kappa$  = specific conductivity

(Vik *et al*, 1984)

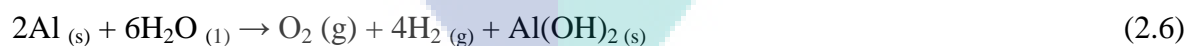
From the equation 2.4, infers that IR drop will increase by increasing the distance of the electrodes. During the experiment, the current suddenly drop after some time, so applied voltage has been increase in order to maintain the constant current. This situation occurs maybe due to the rising of Ohmic loss (IR drop) which lead to the rate of anodic oxidation inhibited. Therefore, the increase of IR drop by increase the distance between anode and cathode is not recommended in EC process.

EC is strongly depends on the pH of a solution. The effect of pH on coagulants depends on the produced reactions on different pH conditions.

In neutral conditions:



In acid conditions:



In alkali conditions:



As reactions show,  $\text{Al}(\text{OH})_2$  form in acidity condition but so in alkali condition,  $\text{Al}(\text{OH})_3$  are produced.  $\text{Al}(\text{OH})_3$  and  $\text{Al}(\text{OH})_2$  settle while,  $\text{H}_2$  moves upward and causes flotation. Since  $\text{Al}(\text{OH})_3$  has higher weight and density, it settles faster and has higher efficiency.

Therefore, it acts better in enmeshment in a precipitate where the flocs of Aluminum Hydroxide acts as a blanket and brings all the stable suspended solid down while settling.

### 2.6.1 Theory of EC

EC is a complicated process involving many chemical and physical phenomena that make use of consumable electrodes to supply ions into the pollutant system. It is an electrochemical technique whereby anodes (aluminum or iron electrodes) corrode to release active coagulants into solution. The generated metallic ions, for example  $Al^{3+}$  (aq) and  $Fe^{3+}$  (aq), then undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides. These hydroxides/polyhydroxides/polyhydroxy-metallic compounds have a strong affinity with dispersed/dissolved molecules as well as any dissolved ions to cause coagulation/adsorption (Cañizares *et al*, 2009).

The main advantages of EC over other conventional techniques such as chemical coagulation and adsorption are “in situ” delivery of reactive agents, no generation of secondary pollution, and compact equipment and it involves three successive stages:

- a) Formation of coagulants by electrolytic oxidation of the ‘sacrificial electrode’,
- b) Destabilization of the contaminants, particulate suspension, and breaking of emulsions,
- c) Aggregation of the destabilized phases to form flocs (Mollah *et al*, 2004)

There are so many studies have reported the potentials of EC in treating a variety of wastewater including removing suspended solids, removing dyes, heavy metals, complex organics, and removing bacteria, viruses and cysts and breaking oil emulsions in water. Although there have been a number of studies on EC for wastewater treatment, large-scale applications of this technology have been relatively few. One possible reason is the energy demand of the EC process.

### 2.6.2 Optimization of EC

By considering the response surface methodology, Körbahti and Tanyolac (2008) concluded that the optimum conditions for treating simulated textile dye wastewater with industrial component were at temperature of 30 degrees Celsius, 25 g/L electrolyte concentration, 8 V electrical potential, with a 35.5 mA/cm<sup>2</sup> current density where the color, COD, and turbidity removal of 99.6%, 61.6% and 66.4%. From Box-Behnken design for the treatment of biodiesel wastewater, Chavalparit and Ongwadee (2009) show that EC process using an aluminum anode and graphite cathode is effective in reducing COD, oil and grease, and suspended solid by 55.43%, 98.42%, and 96.59%, respectively, at the optimum conditions of pH 6.06, applied voltage 18.2 V, and reaction time 23.5 min. Koparal *et al* (2008) investigate the effect of initial pH batch removal of synthetically prepared wastewater having high concentration of humic substances by EC method using plate electrodes. While examining the effect of initial pH on the system performance, pH range has been chosen as 5–9.6. The removal efficiency decreases when initial humic substance concentration is relatively high ( $C_0 > 200 \text{ mg L}^{-1}$ ), due to the formation of gel layer on the electrode surface; to prevent the gel formation initial pH of the water was adjusted to 5.0 and decrease in removal efficiencies disappeared. The most favorable results are achievable when initial pH equals to 5.0, although at low concentrations ( $C_0 < 200 \text{ mg L}^{-1}$ ) there are no pH limitations. Chou *et al* (2010), investigated that the optimum applied voltage and solution temperature were found to be 20 V and 308 K, respectively of removing salicylic acid from aqueous solution and the generation of coagulant metal hydroxides for the Al/Al electrode pair was greater than that for the Fe/Fe electrode pair, which also enhanced the removal efficiency.

### 2.6.3 EC Kinetic

Chou *et al* (2010), concluded that the activation energy was 20.21 J mole<sup>-1</sup> based on pseudo-second-order rate constants from the Arrhenius equation, in salicylic acid abatement from aqueous solutions by EC process. Saravanan *et al* (2010) concluded that

under  $3 \text{ A/dm}^2$  of current density, pH of 6.5, and  $2 \text{ g/L}$  electrolytes concentration, was capable of removing 91% COD by using Acid Blue 113 with EC and determined that this relationship resembled a pseudo-first order kinetic model. Zaroual *et al* (2009) concluded that the treatment efficiency of 91% could be completed for treating chromium (III) with an energy consumption of  $3.536 \text{ KWh/m}^3$  by aluminum anodes for EC. A mathematical model by using a pH of 4.23, electrical potential of 9.14 V, 10 minute reaction time, and  $27.5 \text{ }^\circ\text{C}$  temperature was established using central composite design.

## 2.7 APPLICATION OF EC PROCESS

Nowadays EC technique has been applied in the treatment of a wide variety of wastewater. It successfully been used for the treatment of wastewater containing of dangerous heavy metal, heavy load of COD, oil and grease, colloid and hazardous/toxic chemicals such as phenol.

### 2.7.1 Domestic Wastewater Treatment

Illhan *et al.* (2008) took 15 minutes for heavy loaded ( $380 \text{ mg COD/L}$ ) and 8 minutes for weakly loaded ( $260 \text{ mg COD/L}$ ) to remove 60 and 70% of COD and SS respectively from domestic wastewater by using iron electrode. Sarala (2012) acquired  $60 \text{ mg COD/L}$  from initial  $260.4 \text{ mg COD/L}$  of domestic wastewater after 20 minutes of EC process by using iron electrode and then removed by filtration. Rodrigo *et al.* (2010) is capable of removing COD and ionic phosphorus, when using conductive-diamond electrochemical oxidation and EC for persistent organic consumption, specifically regeneration of urban wastewater. The study stated that energy consumption is capable of removal at values lower than  $4.5 \text{ KWh/m}^3$ . Bukhari (2008) investigated the EC treatment process for the removal of total suspended solids from Eastern Province of Saudi Arabia municipal wastewater. He found that the highest TSS removal efficiency of 95.4% occurred at a current of 0.8 A and contact time of 5 min using stainless steel electrode. Veli *et al.*

(2008) conclude that the treatment efficiency of 87% could be completed for removing COD of municipal leachate in 15 minutes operating time.

### **2.7.2 Industrial Wastewater Treatment**

Koby et al. (2006) investigated the treatment of wastewater from potato chips manufacturing by EC process by using iron and aluminum electrodes. They found that the removal efficiencies of COD and turbidity were high, being 60% and 98%, respectively, with retention time below than 40 min at 300 A/m<sup>2</sup>. Drogui et al. (2008) concluded that there was higher COD removal (82%) from EC process of pollutants from agro-industry wastewaters by using bipolar mild steel electrode in 90 minutes at  $19 \times 10^{-3}$  A/cm<sup>2</sup>. Khatibikamal et al. (2010) investigated EC process using aluminum electrodes for removing fluoride from treated industrial wastewater originated from steel industry. After treatment, the fluoride concentration was reduced from initial 4.0–6.0 mg/L to lower than 0.5 mg/L where about 93% removal was obtained during 5 minutes of treatment. Uğurlu et al. (2008) studied the removal of lignin and phenol from paper mill effluents by EC process and came out with 80% of lignin and 98% of phenol removal after 7.5 minutes by using Aluminum electrode. Zaied and Bellakhal (2009) studied the EC treatment of black liquor from paper industry. After 50 minutes by using of 14 mAcm<sup>-2</sup>, the treatment of black liquor has led to a removal capacity of 98% of COD, 92% of polyphenols and 99% of color intensity with a good repeatability relative standard deviation (R.S.D. < 3%) making it possible to highlight the industrial interest of this electrochemical process.

## **2.8 DESIGN OF EXPERIMENT**

Design of Experiment (DoE) is a structured, organized method that is used to determine the relationship between the different factors affecting a process and the output of that process. In addition, experimental design is a strategy to gather empirical knowledge which is based on the analysis of experimental data and not theoretical models. DoE



involves designing a set of ten to twenty experiments or more, in which all relevant factors are varied systematically. When the results of these experiments are analyzed, it helped to identify optimal conditions, the factors that most influence the results, and those do not, as well as details such as the existence of interactions and synergies between factors.

DoE worked as an efficient procedure in conducting experiment with analyzing resulted data significantly to give a valid an objective conclusion in the experiment (Montgomery, 2001). Hence the application of the experimental design (DoE) techniques early in process development results in improved process yields, reduced variability and closer conformance to target requirement, reduce development time and reduce overall costs. DoE can be divided into several groups based on the function and objective of statistical analysis. The most common design used in the experiment was factorial, response surface, mixture and crossed.

### **2.8.1 Response Surface Methodology**

Response surface methodology (RSM) is a useful methodology derived from the combination of both specific mathematics and statistic manner. In addition, Montgomery has defined that RSM is a collection of mathematical statistical techniques that are useful for the modeling and the analysis problems in which a response is influenced by several variables of factors, and the objective is to optimize this response. All the variables or factors are assumed continuous and controllable with negligible error whereas the response is assumed to be a random variable (Montgomery, 2001).

To assist the researcher in resolving problems related to optimum product design, procedure improvement or optimum system, RSM is the best way to provide a feasible analysis and resolution program as the characteristic of an industrial system has been significantly affected. In most RSM problems, the form of the relationship between the response and the factors is unknown. Thus, the first step in RSM is to find a suitable approximation for the true functional relationship between response and the set of factors.

If the response is well-modeled by linear function of the factors, then the approximating function is the first order model whereas if there is curvature in the optimum region, then polynomial of higher degree such as the second order model must be used (Montgomery, 2001).

RSM is a beneficial technique in obtaining the optimum experimental design and operational conditions for various industrial applications such as electronics, engineering, agriculture, chemical, biotechnology and food science (Chang and Shaw, 2009). There were several design classed in RSM including Central Composite design (CCD), Box-Behnken design, hybrid design and 3-Level Factorial. However, among these designs, CCD becomes most favorable design in all application primarily in membrane industry (Nosrati, 2011)

### 2.8.2 Central Composite Design

The central composite design (CCD) is the most frequently used under RSM design due to its flexibility and robustness. This type of design contains of fractional factorial design with center points that is augmented with a group of axial points (red points) that allow estimation of curvature as shown in Figure 2.11. The replicates at the center point have two main objectives: (i) provide a measure of pure error and (ii) stabilize the variance of the predicted response (Ferreira *et al*, 2007). To obtain more precise estimate error, the more replicates the better. Generally, CCD design consists of a  $2^k$  factorial (or fractional factorial) with  $n_f$  runs,  $2k$  axial or star runs and  $n_c$  center run, where  $k$  represents the number of factors or variables involved in the studies.

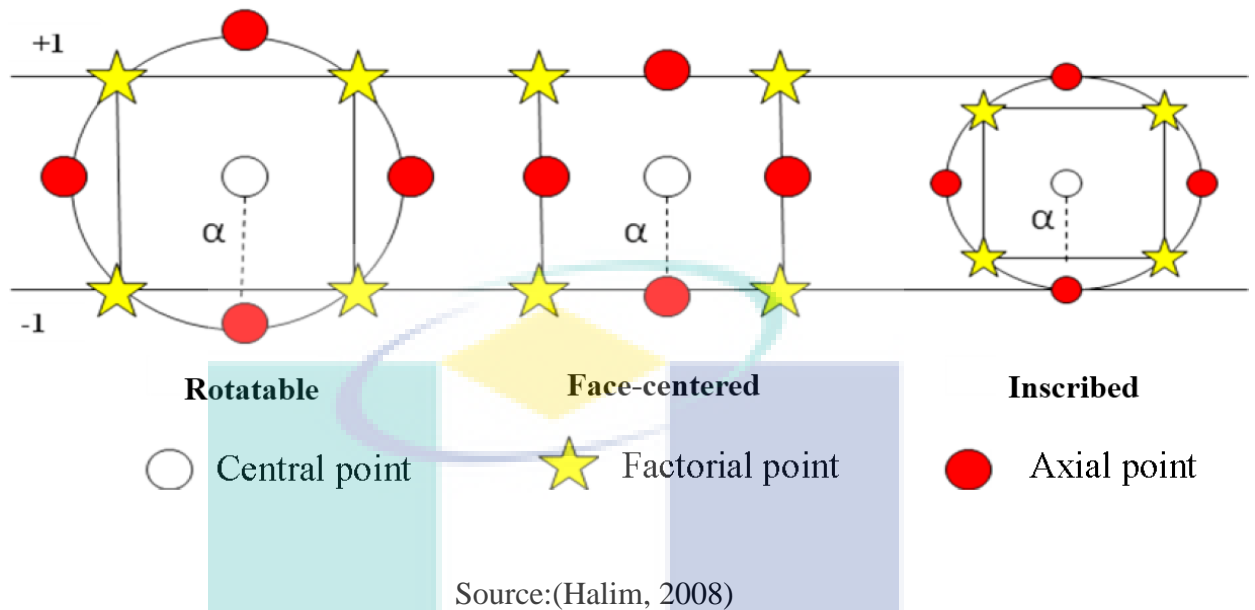
There are two parameters in the design that must be specified; for example distance  $\alpha$  of the axial points from the design center and the number of center points  $n_c$  (Myers and Montgomery, 2002). It is based on the number of factors,  $k$  in the design, where  $\alpha$  is equal to  $\sqrt{k}$ . These design required 5 levels for each factors. The choice of  $\alpha$  actually depends to a great extent on the region of operability and region of interest. The value of the axial

distance generally varies from 1.0 up to  $\alpha = \sqrt{k}$ , the former placing all axial points on the face of the cube or hypercube, the latter, resulting in all points being placed on a common sphere. If the distance from the center of the design space to a factorial point ( $2k$ ) is  $\pm 1$  unit for each factor, the distance from the center of the design space to a star point is  $\pm \alpha$  with  $[\alpha] > 1$ . Three main varieties of CCD available in most statistical software program by changing  $\alpha$  are rotatable, face-centered and inscribed. Table 2.2 summarizes the properties of the three varieties of CCD.

**Table 2.2: Central composite designs.**

Central Composite Design Type	Terminology	Comments
Rotatable	CCR	<ul style="list-style-type: none"> <li>• The original form of the central composite design.</li> <li>• The ● are at some distance <math>\alpha</math> from the center based on the properties desired for the design and the number of factors in the design.</li> <li>• The ● establish new extremes for the low and high settings for all factors.</li> <li>• The designs have circular, spherical or hyperspherical symmetry.</li> <li>• Require 5 levels for each factor.</li> <li>• Augmenting an existing fractional factorial design with ● can produces this design.</li> </ul>
Face centered	CCF	<ul style="list-style-type: none"> <li>• The ● are at the center of each face of the factorial space at <math>\alpha = \pm 1</math>.</li> <li>• Require 3 levels for each factor.</li> <li>• Augmenting an existing factorial design with appropriate ● can also produce this design.</li> </ul>
Inscribed	CCI	<ul style="list-style-type: none"> <li>• The CCI design is scaled down CCR design with each factor level for the CCR design divided by <math>\alpha</math>.</li> <li>• Require 5 levels for each factor.</li> </ul>

Source: Tobias and Trutna, (2006)



**Figure 2.2:** Three type of central composite design.

### 2.8.3 Analysis of the Data

A response surface model represents the functional form of a response surface. Response surface models can be based on either theoretical or empirical considerations. When a theoretical model cannot be specified in an experimental investigation (the usual case), polynomial models can often be used to approximate the response surface. A quadratic model can provide a useful approximation for a broad range of applications. In addition, a quadratic (second-order) model is the most widely used for design fitting in the CCD. A complete second-order model is of the form:

$$Y = b_0 + \sum_{i=1}^3 b_i x_i + \sum_{i=1}^3 b_{ii} x_i^2 + \sum_{i=1}^2 \sum_{j=i+1}^3 b_{ij} x_i x_j + \varepsilon \quad (2.5)$$

Where  $Y$  is the response (dependent variable),  $b_0$  is the constant coefficient,  $b_i$  is the coefficient for the linear effect,  $b_{ii}$  is the coefficient for the quadratic effect,  $b_{ij}$  is the coefficient for the interaction effect,  $x_i$  and  $x_j$  are the independent variables (factors).

The analysis of the data is started by fitting a response surface model using provided software inside DoE program. The stepwise regression procedure allowed the selection of probabilities ( $p$ -values) for adding or deleting model terms. Stepwise will generate a screen with recommended model terms checked and  $p$ -values shown (these are called “Prob>F” in the output). Sometimes, based on  $p$ -values, some term might be chosen to reduce or uncheck. However, hierarchy principle should be followed and keep all main effects that are part of significant higher-order terms or interactions, even if the main effect  $p$ -value is higher than 0.05 which is common statistical significance level chosen.

A next step was proceeding with the analysis of variance (ANOVA). The ANOVA statistics consists of value for  $R^2$ , adjusted  $R^2$ , predicted  $R^2$ , lack of fit test, standard deviation, etc. in order to confirm the adequacy of the regression model. Generally, the  $R^2$  value must be in the range of  $0 < R^2 < 1$ . However, a large value of  $R^2$  does not imply that the regression model is good (Myers and Montgomery, 2002).

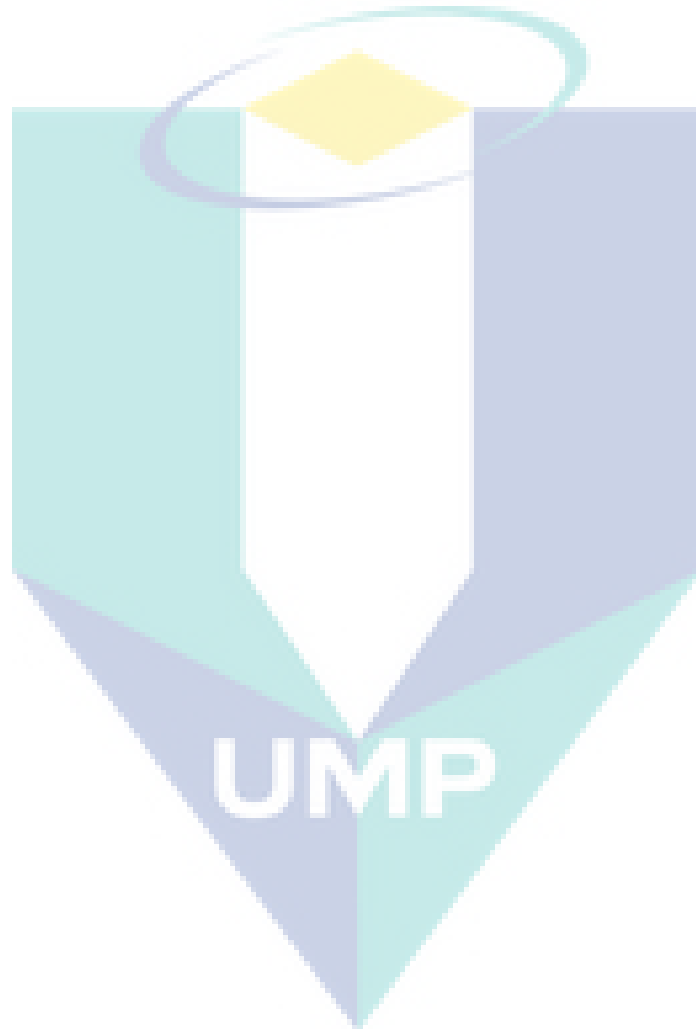
Then, the further work was done with diagnostic of the statistical properties of the regression model. Inspection of various diagnostic plots was carried out to statistically validate the model. The most important diagnostic will be the normal probability plot of the studentized residuals (normal probability distribution). The data points should be approximately linear. A non-linear pattern indicates non-normality in the error term, which may be corrected by a transformation. After each response get analyzed and the model fitting was satisfied, optimization can be done either by inspection of the interpretation plots or with the graphical and numerical tools provided in the software packages.

## 2.9 PRODUCTION OF HYDROGEN BY EC PROCESS

Besides treating sewage, EC process also produces hydrogen that can be used as energy. In a carbon constraint economy, renewable energy resources such as wind, solar and water are constantly replenished and claimed to be a non polluting alternative energy source. In conjunction hydrogen produced through non-fossil fuel sources such as water by using different forms of sustainable energy sources and process heats may then be considered to be a prime fuel in meeting energy supply, secured environmental and economical sustainability (Hotza and Diniz da Costa, 2008; Adnan Midilli and Ibrahim Dincer, 2008). In this regard, considering a long-term environmental damages created by consumption of fossil fuels, sustainable energy based hydrogen energy systems that enable us to store the sustainable energy sources in the form of hydrogen should be particularly put into practice. In fact as water is a resource generally available and abundant in many areas, the potential benefits of the hydrogen network would be the economically feasible production. In particular hydrogen based energy conversions or water fuel can be burned as a clean burning fuel, renewable, low cost and reliable energy solution.

The advantages of sustainable energy based hydrogen energy system can be enumerated as follows: It can provide environmental sustainability because hydrogen is high quality and environmental benign energy and a nontoxic clean energy carrier, and produces nontoxic exhaust emissions. It can also provide environmental stability because hydrogen can be safely transported in pipelines. It can be appropriate for the utilization of the sustainable energy sources and present long period-energy use, and thus ensure energy resource sustainability because hydrogen can be produced from non-fossil fuel sources by using different production techniques, and can be stored over relatively long periods of time, compared to electricity. It creates many additional industrial working fields, and thus ensures industrial sustainability because hydrogen is possible to advantageously use as a chemical feedstock in the petrochemical, food, microelectronics, ferrous and nonferrous metal, chemical and polymer synthesis, and metallurgical process industries, and as an energy carrier in clean sustainable energy systems (Soltermann and Silva, 1998; Rakopoulos *et al*, 2008).

The aspect of hydrogen recovery from the EC process has less been studied previously. Hydrogen is a main by-product of the EC process as it is generated at the cathodes by water electrolysis. With an effective gas–liquid–solid separation process, high quality hydrogen can be recovered from the EC process and used as an energy source or as a reactant for industrial processes.



## CHAPTER 3

### RESEARCH METHODOLOGY

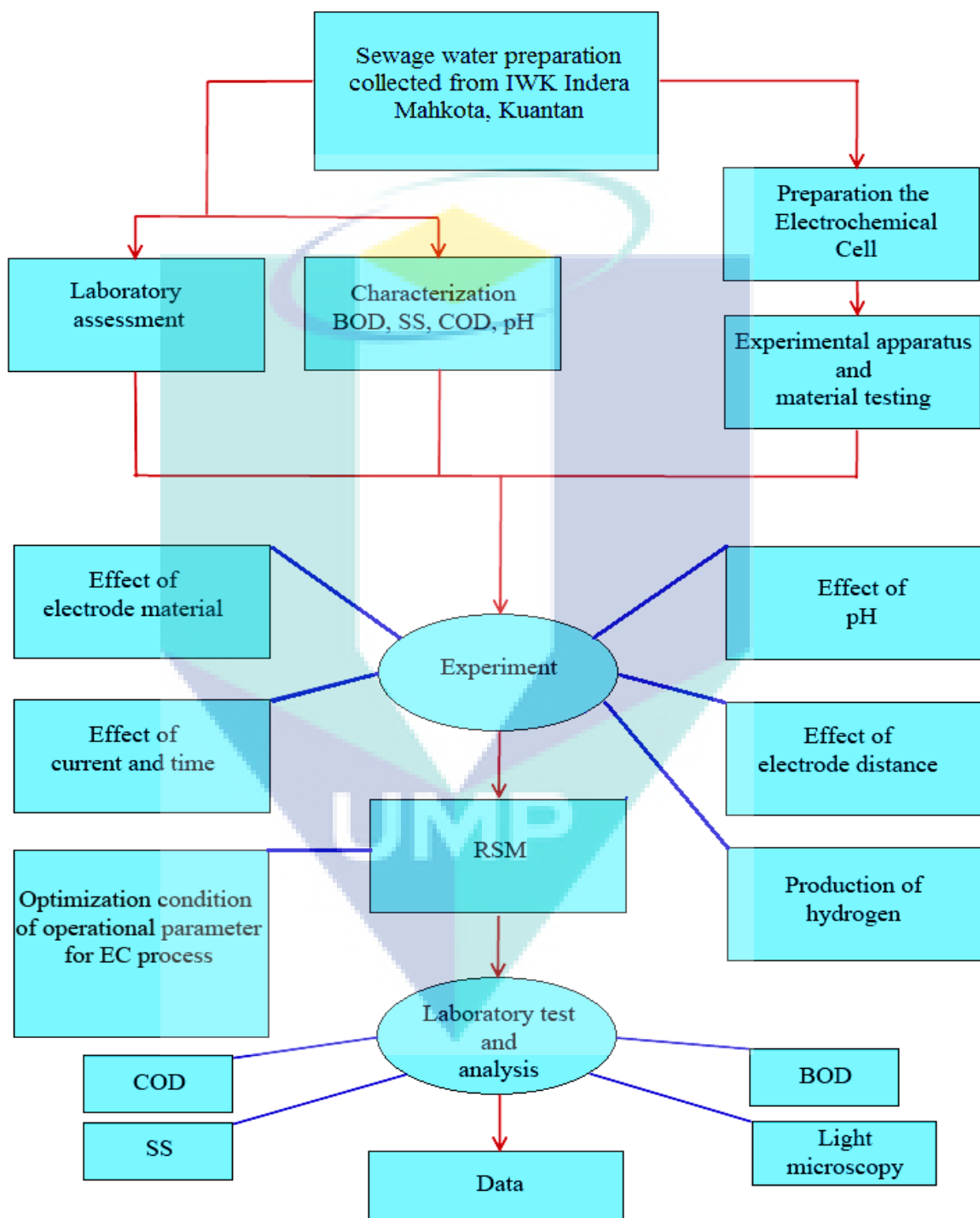
#### 3.1 INTRODUCTION

The aim of this project was to examine the efficiency of EC process in treating sewage water by using different type of electrode, current, time, pH and electrode distance, therefore to suggest how it can be improved. One of the basic stages was to do a number of experiments that would help examine how the efficiency is balancing and to validate a model that will be used to investigate the effects that influence the efficiency. Experimental data is quite useful in such projects as it usually varies with theory and hence proposals could be made in order to improve the operation modes. For this specific project, five types of experiments were chosen to help investigate the efficiency of the EC process, which were determining the best of electrode material, current, time, pH and electrode distance. Another experiment by using Response Surface Methodology (RSM) will be used as a supportive experiment. An additional experiment of determining the hydrogen production also will be carried out in order to test the effectiveness of EC process on hydrogen production.

This chapter presents the experimental set-up for this work. It will be started with the equipment such as EC reactor, materials and chemicals had been used in this study, sampling location for collecting the sewage sample and then, the preparation techniques for



the EC process. Operational framework of this research is conducted as according to the flowchart as shows in figure 3.1.



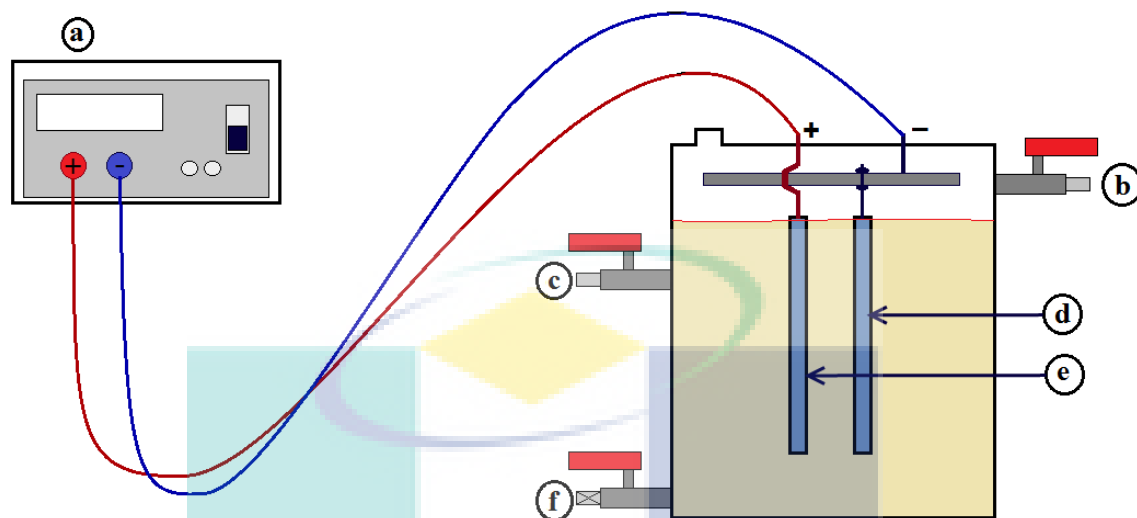
**Figure 3.1:** Flow chart of research framework.

### 3.1.1 Preparation of EC process

To run the experiment the important thing was to prepared the apparatus. The apparatus that involved in this EC process was an electrochemical cell, DC power supply, the amount of sewage water, tygon tube, electric wire, test tube, beaker, sodium hydroxide (NaOH) and hydrochloric acid (HCl) as pH controller.

### 3.1.2 Preparation of Electrochemical Cell

The lack of mechanistic understanding of EC is reflected in the design of electrochemical cell. No single empirical or systematic approach has emerged over the years in the design of electrochemical cell. It is therefore difficult to compare the performance of the cells. In this experiment, a stainless steel cuboids box with volume 2000 ml been used as the platform for EC process of sewage water. Monopolar metal plate electrodes (120 mm x 100 mm x 2 mm) were used with distance between each pair of electrodes is varied from 5 mm to 30 mm. Aluminum, iron and stainless as steel electrodes used for the experiment were cut mechanically according to the size and all the dirt and corrosion on the plates were removed using hydrochloric acid with concentration of 10 percent. All the plates were drown into the acid solution for about 5 minutes and wash with clean water before used. This method is applied in order to prevent the interruption of electron movement between the plates by the dirt and corrosion at the plate's surface that will affect the process performance. The plates are supported by a shoulder at both side of reactor to make sure both electrodes always kept in the right position and not moving during the experiment running. The total effective electrode was  $1.652 \times 10^{-2} \text{ m}^2$ . These electrodes are connected to terminals of a Regulated DC Power Supply (EDU-LABS TPR-3030D; 30V / 30 A). After all the apparatus is setup the electrolysis of sewage wastewater is run. The electric current can be controlled into desired value by DC power supply.



**Figure 3.2:** Electrochemical cell setup: a) Power Supply; b) Hydrogen collection valve; c) Sample collection valve; d) Cathode; e) Anode; f) Drain valve.

### 3.1.3 Preparation of Sewage Sample

Raw sewage was chosen in this study. For every trial, fresh sewage was collected on each of the experimental study. Sample was collected at Indah Water Konsortium (IWK) which located at Indera Mahkota, Kuantan, Pahang.

This sample will be characterized to identify the COD, BOD, SS and pH first, and then should be stored in the freezer with temperature below than  $-4^{\circ}\text{C}$  to ensure that it would maintain the same condition as the same time as collected and to prevent any of bacteria activities. If the sample did not preserve properly, it should be taken back again at IWK.



**Figure 3.3:** Incoming flow of the raw sewage.

### 3.2 DETERMINE THE CHARACTERISTIC OF SEWAGE

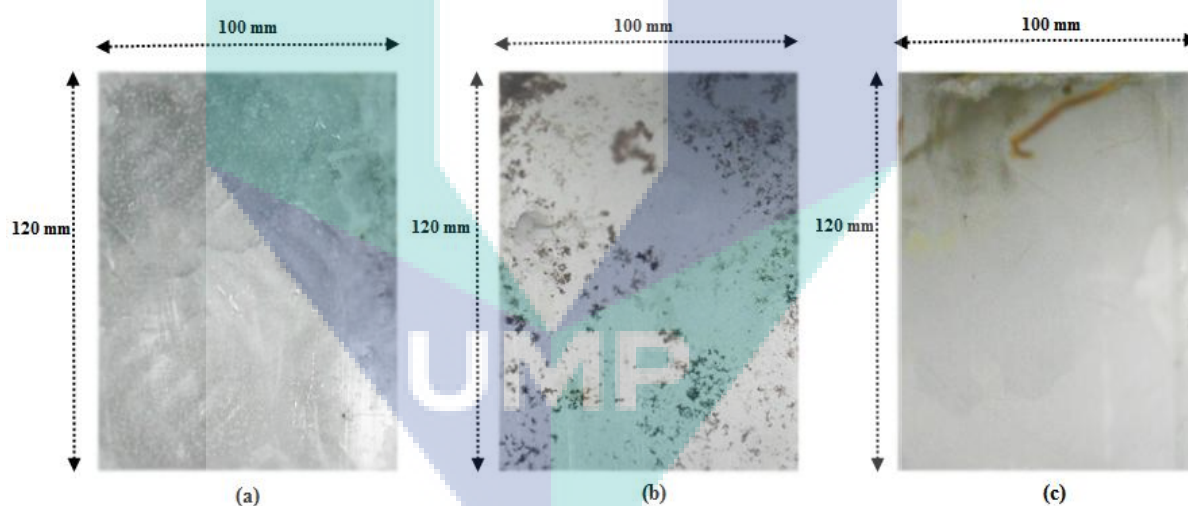
This experiment was carried out to determine the characteristic of sewage either it is weak, medium, strong or very strong strengths. The strength of sewage is usually judge by its COD and BOD<sub>5</sub>. Table 3.1 shows the strength of sewage water.

**Table 3.1:** Sewage strength in terms of COD and BOD<sub>5</sub> (Source: Mara, 1976)

Strength	COD (mg/L)	BOD <sub>5</sub> (mg/L)
Weak	<400	<200
Medium	700	350
Strong	1000	500
Very strong	>1500	>750

### 3.3 EFFECT OF ELECTRODE MATERIAL

This experiment was carried out to determine the appropriate electrode material between iron, stainless steel and aluminum in treating the raw sewage wastewater. The best performance of electrode material will be used in the next experiment. A couple of Iron, stainless steel and aluminum plates were fabricated with the same dimension of 120 mm x 100 mm x 2 mm. The electrodes were dipped in 2 L of sewage water under the operation of EC process with 30 A in 30 minutes to measure COD, BOD and SS. At the end of each run, the electrodes were rinsed in dilute HCl solution to remove any oxide film formed onto the surfaces during experiments. As a control, raw sewage was used by dipping each type of electrodes for 30 minutes without any current (electricity) applied. Figure 3.4 shows the electrode used in the experiment.



**Figure 3.4:** Type of electrodes material a) Aluminum; b) Iron; c) Stainless steel

### 3.4 EFFECT OF CURRENT AND CONTACT TIME

In this experiment, six different amounts of currents (three low currents and three high currents) were applied for 30 minutes and followed by measuring COD, BOD and SS in every 5 minutes. The value of low currents that have been used were controlled at 1 A, 2

A, and 3 A, while for high currents value were operated at 20 A, 25 A, and 30 A. In a control experiment the dipped electrode material was run without any current applied. The experiment is important to study the practical influence of EC, whether by its time or current. The result also shows the difference between low current and high current treatment. Electro-dissolution of the electrode material also been discussed. This experiment can determine the best current applied that should be used during the next experiment.

### **3.5 EFFECT OF INTER-ELECTRODE DISTANCE**

In this experiment, the distance between electrodes have been varied in order to determine the effect to the treatment at 30 minutes by 30 A of current applied. The inter-electrodes distances were studied at 5 mm, 10 mm, 20 mm, 30 mm and 40 mm respectively. In a control experiment, the EC process with each electrode distance mentioned were repeated without applying any current. The experiment is important to study the practical of inter-electrodes distance on EC process whether it efficient in low or in high distance.

### **3.6 EFFECT OF pH**

This experiment was performed to study the pH influence to the COD, BOD, SS and hydrogen production in EC process up to 30 minutes of treatment. It also necessary to determine the optimal pH values that could enhance the treatment of raw sewage water as well as hydrogen production. The initial pH value was run at 3, 4, 5, 6, 7, 8, 9, 10 and 11 at 30 A current applied. This experiment was repeated without applying any current as a control.

### 3.7 DESIGN OF EXPERIMENT: RESPOND SURFACE METHOD

A standard response surface methodology (RSM) with central composite design (CCD) was applied to develop a polynomial regression equation in order to analyze the correlation among the reaction time, current intensity, inter-electrode distance and pH to the responses. CCD consists of  $2n$  factorial runs,  $2(n)$  axial runs and six centre runs, where  $n$  is the number of factors. A complete design matrix of the experiments employed as shown in Table 3.2. Factorial CCD consists of 4 variables; 16 factorial points, 8 axial points and 6 replicates at the center points with  $\alpha$  value of 1.05 were employed. The total of 30 experiments for EC process of sewage was performed, as calculated from Equation 3.1:

$$N = 2^n + 2n + n_c = 2^4 + 2(4) + 6 = 30 \quad (3.1)$$



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**Table 3.2:** Experimental design matrix.

Run	Variables (coded level of variables)				pH
	Current	Reaction Time	Distance		
	(A)	(min)	(mm)		
	$x_1$	$x_2$	$x_3$	$x_4$	
1	30.00	30.00	5.00		3.0
2	30.00	30.00	5.00		11.0
3	30.00	5.00	5.00		11.0
4	1.00	30.00	5.00		3.0
5	1.00	30.00	5.00		11.0
6	15.50	17.50	17.50		7.0
7	30.00	30.00	30.00		3.0
8	1.00	5.00	30.00		3.0
9	1.00	5.00	5.00		11.0
10	15.50	17.50	17.50		7.0
11	30.00	5.00	5.00		3.0
12	1.00	5.00	30.00		11.0
13	1.00	30.00	30.00		11.0
14	15.50	17.50	17.50		7.0
15	15.50	17.50	17.50		7.0
16	1.00	5.00	5.00		3.0
17	30.00	5.00	30.00		11.0
18	30.00	30.00	30.00		11.0
19	1.00	30.00	30.00		3.0
20	30.00	5.00	30.00		3.0
21	15.50	30.63	17.50		7.0
22	15.50	17.50	17.50		7.0
23	15.50	17.50	30.63		7.0
24	15.50	17.50	4.38		7.0
25	15.50	4.38	17.50		7.0
26	15.50	17.50	17.50		11.2
27	30.73	17.50	17.50		7.0
28	0.27	17.50	17.50		7.0
29	15.50	17.50	17.50		7.0
30	15.50	17.50	17.50		2.8



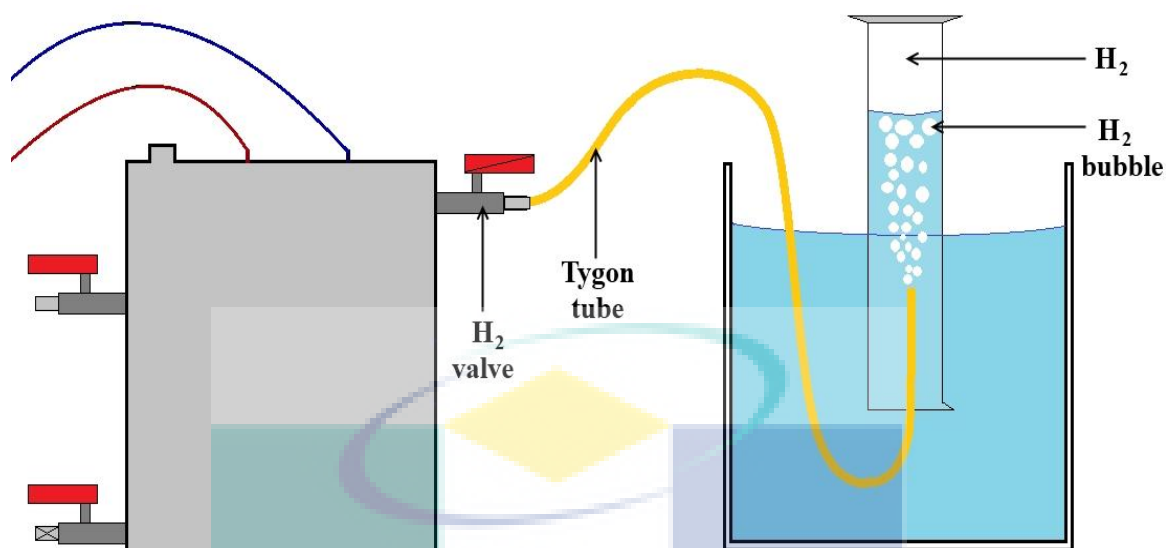
The experiments sequence was randomized in order to minimize the effects of the uncontrollable factors. The responses were COD ( $Y_1$ ), BOD ( $Y_2$ ) and SS ( $Y_3$ ). Each response was used to develop an empirical model which correlated the response to the two carbonization variables using a second-degree polynomial equation as given by Equation 3.2:

$$Y = \beta_0 + \beta_1x_1 + \beta_2x_2 + \beta_3x_3 + \beta_4x_4 + \beta_{11}x_1^2 + \beta_{22}x_2^2 + \beta_{33}x_3^2 + \beta_{44}x_4^2 + \beta_{12}x_1x_2 + \beta_{13}x_1x_3 + \beta_{14}x_1x_4 + \beta_{23}x_2x_3 + \beta_{24}x_2x_4 + \beta_{34}x_3x_4 \quad (3.2)$$

where  $Y$  is the predicted response,  $\beta_0$  the constant coefficient,  $\beta_1$ ,  $\beta_2$ ,  $\beta_3$  and  $\beta_4$  are the linear coefficients,  $\beta_{12}$ ,  $\beta_{13}$ ,  $\beta_{14}$ ,  $\beta_{23}$ ,  $\beta_{24}$  and  $\beta_{34}$  are the interaction coefficients, and  $\beta_{11}$ ,  $\beta_{22}$ ,  $\beta_{33}$  and  $\beta_{44}$  are the quadratic coefficients. The experimental data were analyzed using a statistical software Design Expert software version 6.0.8 (STAT-EASE Inc., Minneapolis, USA) for regression analysis to fit the second-degree polynomial equation and also for the evaluation of the statistical significance of the equations developed.

### 3.8 DETERMINE THE PRODUCTION OF HYDRGEN BY EC PROCESS

This experiment was performed to determine the hydrogen gas produced as by-product by EC process of raw sewage. Chemical reaction occurred at electrochemical cell and yet produces hydrogen gas and oxygen gas. The gases were collected in the test tube that was filled with the water by using water displacement method where the gases will displace the water in the test tube. The gases were collected in 60 seconds of EC process. The ampere of electrochemical process was set at 1 A, 2 A, 3 A, 20 A, 25 A and 30 A. While the inter-electrode distance pH and were set at the optimum condition of each parameters. Figure 3.5 shows the water displacement method.



**Figure 3.5:** Water displacement method.

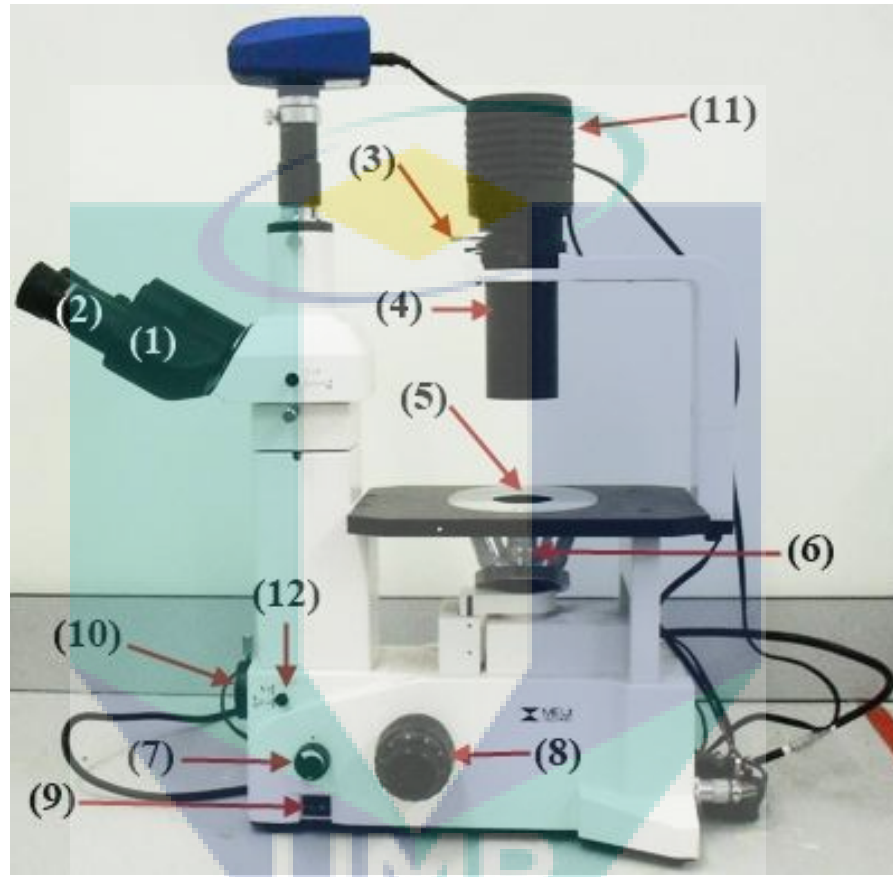
### 3.9 LABORATORY TEST AND ANALYSIS

Data processing and analysis involved data for before and after sample treated performance. All experiments involved such as Suspended solid, Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), pH and Gases were determined by standard method.

#### 3.9.1 Light Microscopy

In this study, the used of light microscope is compulsory to detect the aggregation from EC process. This instrument employs visible light to detect small objects where it can be observed and shape recognized at a mere 400X magnification. Magnification is the ability to view an object as larger. A good image is obtained when the amount of specimen detail is also increased. Magnification alone will not achieve this. Figure 3.6 shows the

Meiji Techno TC5200 Brightfield Inverted Microscope, Japan that will be used in this study.



**Figure 3.6:** Meiji Techno TC5200 Brightfield Inverted Microscope, Japan.

Where:

- |  |   |
|--|---|
| 1. Binocular head, 300 inclination               | 7. Low positioned variable brightness control |
| 2. Eyepieces SWF 10X                             | 8. Coaxial course and fine focusing controls  |
| 3. Aperture Diagram                              | 9. Power switch                               |
| 4. Extended working distance condenser           | 10. Front camera port                         |
| 5. Specimen stage                                | 11. Integrated illuminator 6V 30W             |
| 6. Quintuple nosepiece and phase objectives lens | 12. Beam splitter for camera port             |

In order to ascertain the total magnification when viewing an image with a compound light microscope, take the power of the objective lens which is at 10, 20x or 40x and multiply it by the power of the eyepiece which is typically 10x. Therefore, a 10x eyepiece used with a 20X objective lens, will produce a magnification of 200X. The naked eye can now view the specimen at a magnification 200 times greater and so microscopic details are revealed. In order to get a sharp and clear picture, the following steps should be noted:

- obtaining sufficient contrast
- finding the focal plane
- obtaining good resolution
- recognizing the subject when one sees it

The light microscope was used to observe the formation of lumps of particles that exist in sewage after EC process and to distinguish between the use of high current and low current to the agglomeration of particles that occurred.



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## CHAPTER 4

### RESULT AND DISCUSSION

#### 4.1 INTRODUCTION

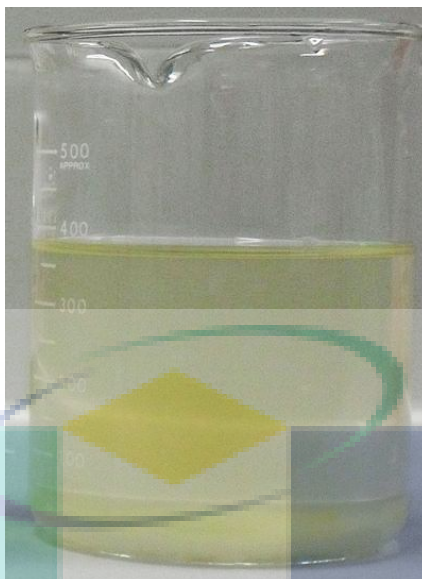
This research has been carried out to determine the effect of electrode material, current (high and low), inter-electrode distance and pH on sewage water. In this chapter, assessment of the efficiency of EC treatment in this phase was based on three main parameters: removal of COD, BOD and SS. The productions of hydrogen as by product at different operating conditions were also determined.

#### 4.2 CHARACTERISTIC OF SEWAGE WATER

Table 4.1 shows the characteristic of sewage water sample used before the treatment. As observed, the average COD, BOD and SS concentration is in the medium concentration.

**Table 4.1:** Characteristic of raw sewage water

COD (mg/L)	BOD (mg/L)	SS (mg/L)	pH
466	259	297	7.6



**Figure 4.1:** Sewage sample

### **4.3 EFFECT OF ELECTRODE MATERIAL**

Electrode assemblies as the heart of the EC process. Therefore, the appropriate selection of its materials is very concerned. In this experiment, the aluminum, iron and stainless steel electrode were used as these electrodes are known as cheap, readily available and proven effective to treat wastewater. Table 4.2 shows the treatment performance in every types of the electrode used. During EC process, aluminum, iron and stainless steel electrodes releases their own metal ion and form a range of coagulant species and metal hydroxides, which destabilize and aggregate the suspended particles or precipitates and absorb dissolve organic particles. The comparison of the treatment amongst electrodes used was recorded throughout the experimental condition. The experiment was initially operated in 30 minutes at 30 A at different types of electrode to obtain the favorable electrode in sewage water treatment. In control sample, the final concentration of COD, BOD and SS did not change much from the initial state where there was only a little bit drops has spotted. This drop was due to the particles in the sample settled at the bottom of EC cell

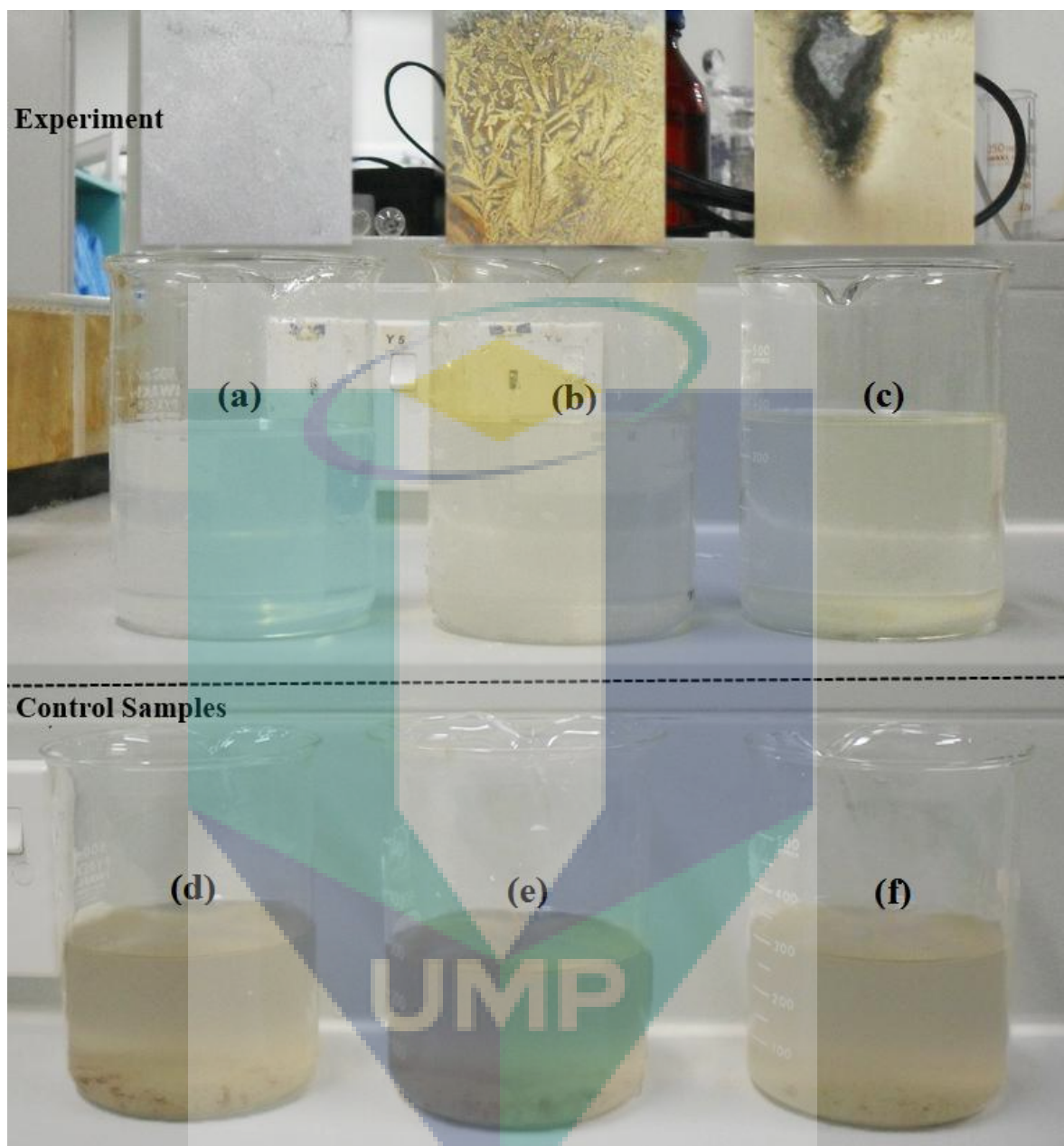
and meaning that the electrode materials have given no effect to the sewage water if there is no EC process occurred.

**Table 4.2:** The effect of different types of electrodes material on sewage's parameters

Electrode material	COD		BOD		SS	
	Treatment	Control	Treatment	Control	Treatment	Control
	sample	sample	sample	sample	sample	sample
<b>Aluminum</b>	9	462	5	254	7	293
<b>Iron</b>	11	463	7	255	13	293
<b>Stainless steel</b>	18	463	10	254	19	295

Note : current intensity = 30 A and reaction time = 30 minutes

Besides, by using iron electrodes, the treated solution begins to change into greenish color after 5 minutes and then switch into turbid and brownish color a few minutes later during treatment due to oxidation effect of iron electrode. The green color must be due to  $\text{Fe}^{2+}$  and the brown color is due to  $\text{Fe}^{3+}$ .  $\text{Fe}^{2+}$  can be easily oxidized into  $\text{Fe}^{3+}$  by dissolve oxygen in water.  $\text{Fe}^{3+}$  usually exist in the form of  $\text{Fe}(\text{OH})_3$  which is yellowish fine particles and hard to precipitate. For that reason, iron electrode is not suitable to be used in this process. As for the stainless steel, the treatment using this metal is not as good as expected as this metal also turns the solution into brownish color yet the treatment efficiency is below than the treatment by iron and aluminum. Figure 4.2 shows the comparison of the sample for sewage water after treatment among three types of electrode used. The removals by iron and stainless steel electrodes are below aluminum yet a little bit expensive compared to aluminum especially stainless steel. As a result, all following experiments were carried out with aluminum electrode.



**Figure 4.2:** Comparison of sample sewage water treated by different electrode; a) Aluminum electrode; b) Iron electrode; c) Stainless steel electrode; d) Control of aluminum electrode; e) Control of iron electrode, f) Control of stainless steel electrode.



#### 4.4 EFFECT OF CURRENT AND CONTACT TIME

In all the EC process, application of current intensity and operating time is the most important parameters in controlling the reaction rate. An increment of current and time resulted to an increase in the removal efficiency of COD, BOD and SS.

##### 4.4.1 Observation of Organic and Inorganic Removal in EC Process

Table 4.3 shows the value of COD every 5 minutes of treatment for 30 minutes in each current applied. As show in the result, all the application of low current in 30 minutes did not meet the regulation for the standard A as stated in table 2.1, except for 3 A current applied where the concentration of COD is 3mg/L below than standard A. However, all the application of low current intensity satisfied the regulation for standard B. While, in high current applied, all the final result met the regulation for standard A.

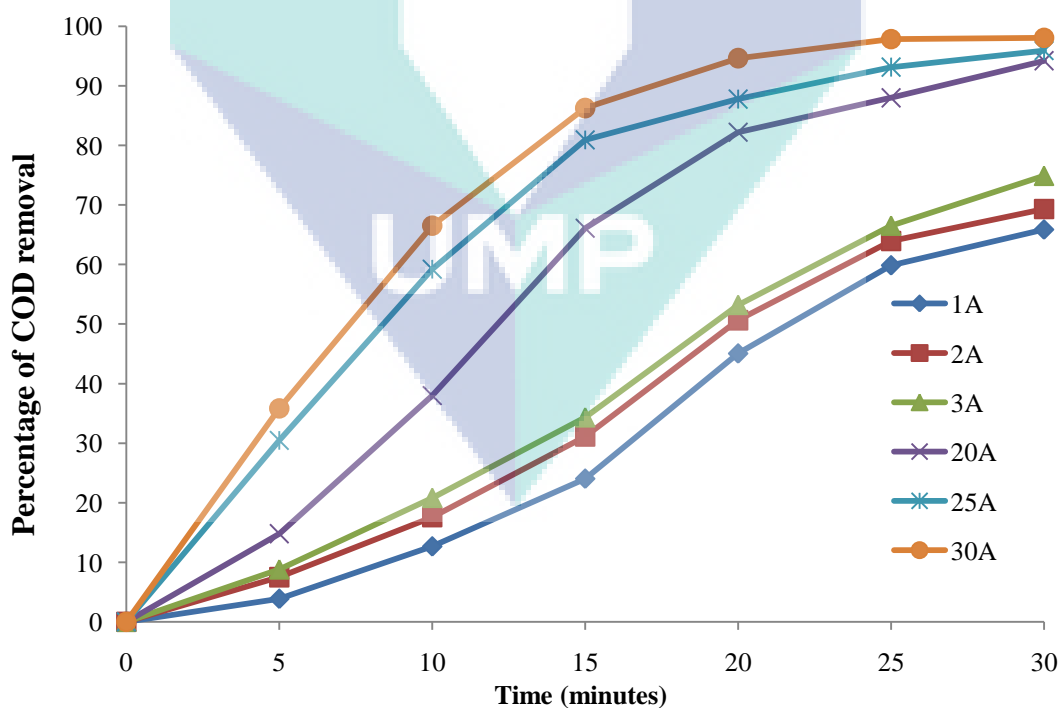
**Table 4.3:** Value of COD (mg/L)

	Current	Time (min)						
		0	5	10	15	20	25	30
Low current	1 A	466	448	407	354	256	187	159
	2 A	466	431	384	321	230	168	143
	3 A	466	425	369	306	218	156	117
High current	20 A	466	397	289	158	83	56	27
	25 A	466	324	190	89	57	32	19
	30 A	466	299	156	64	25	10	9
	Control	466	466	464	465	463	460	460

Note: Average COD value of control sample = 463 mg/L

Figure 4.3 shows the removal efficiency in each current applied for 30 minutes. High current application showed improvement removal efficiency in this experiment. As shown in 4.3, percentage of COD removal for 30 A current application was high which

reached over than 35% in only 5 minutes of treatment and it rapidly rise until it reach 86.26% at 15 minutes, then start slowing down. During 20 to 25 minutes of treatment process, there are only a little increment of removal efficiency and the final removal efficiency of 98.1% was observed at 30 minutes. By applying 25 A, the curve of the graph was almost the same as 3 A application which dramatically rise in early stage and slightly become slower when 15 minutes of treatment reached. The difference between 2 A and 3 A application was the value of removal by 25 A application a little bit lower than the application of 30 A. The final removal for 25 A was recorded up to 95.9%. In the treatment process of using 20 A, the removal efficiency starts to rapidly increase at 5 minutes, before slowing down at 20 minutes and reached 94.2% at 30 minutes. In low current applications, the curves of the graphs were analogous amongst 1 A, 2 A and 3 A. The treatment of low current application increasing gradually during first 15 minutes, then started to rise rapidly. However the treatment become slightly slowing down after 20 minutes. The final removal of 65.88%, 69.3% and 74.9% were obtained at the application of 1 A, 2 A and 3 A respectively.



**Figure 4.3:** Percentage of COD removal

Table 4.4 showed the value of BOD at interval 5 minutes of treatment for 30 minutes in each current applied. All BOD value of treated sample in high current application for 30 minutes were met standard A as stated in table 2.1 which below than 20 mg/L. While all BOD value of treated sample in low current application for 30 minutes were neither met standard A nor standard B.

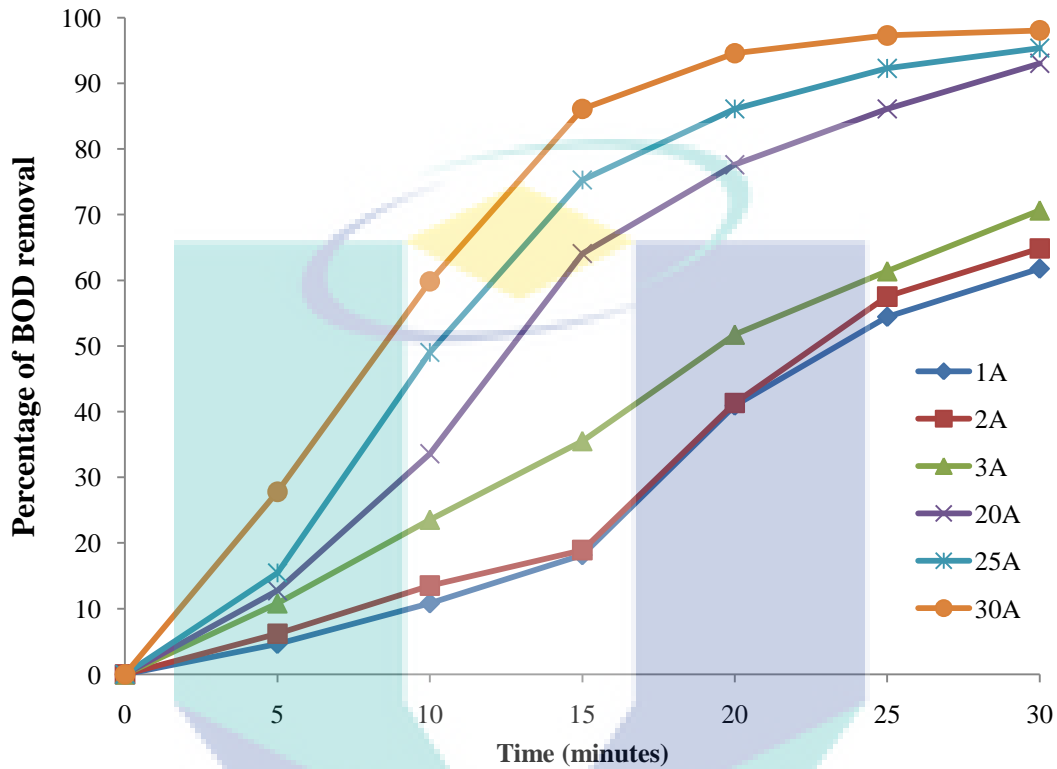
**Table 4.4:** Value of BOD (mg/L)

	Current	Time (min)						
		0	5	10	15	20	25	30
Low current	1 A	259	247	231	212	153	118	99
	2 A	259	243	224	210	152	110	91
	3 A	259	231	198	167	125	100	76
High current	20 A	259	226	172	93	58	36	18
	25 A	259	219	132	64	36	20	12
	30 A	259	187	104	36	14	7	5
	Control	259	257	257	257	255	254	254

Note: Average BOD value of control sample = 256 mg/L

Figure 4.4 shows the removal efficiency in every current applied for 30 minutes. A sharp rise of BOD removal in the early stage was observed at 30 A, unfortunately after 15 minutes, the removal steadily slowing down. Nevertheless, the removal efficiency in 15 minutes reached 86.1% and the final treatment of 98.1% was achieved. The same situation has been observed at 20 A and 25 A, where the removal start slowing down at 15 minutes and the final result of 93.1% and 95.4% respectively were achieved. While by applying 1 A and 2 A, the graphs start to rising dramatically after 15 minutes of treatment and the removal efficiency were 18.1% and 18.9% respectively which were about 5 times lower than the recorded treatment using 30 A at 15 minutes. The final removal efficiency for 1 A was 61.8% and 64.9% for 2 A. As for the treatment using 3 A showed almost linear graphs

where the removal efficiency was slightly better than the application of 1 A and 2 A and finally end up to 70.7% of removal.



**Figure 4.4:** Percentage of BOD removal

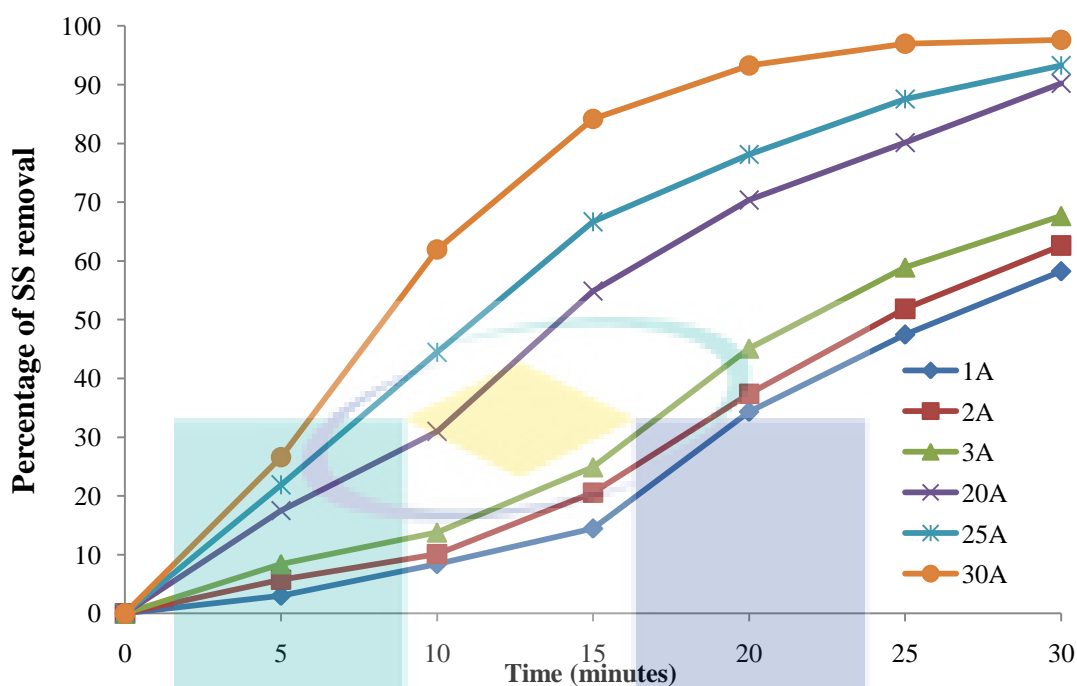
Table 4.5 show the value of SS every 5 minutes of treatment for 30 minutes in each current applied. High current application was important to reach the standard A of discharge value of 50 mg/L. However in low current application, the final results show that SS concentration did not meet the regulation for standard B as stated in table 2.1, except for the application of 3 A which the final value of 96 mg/L was obtained.

**Table 4.5:** Value of SS (mg/L)

	Current	Time (min)						
		0	5	10	15	20	25	30
Low current	1 A	297	288	272	254	195	156	124
	2 A	297	280	267	236	186	143	111
	3 A	297	272	256	223	163	122	96
High current	20 A	297	245	205	134	88	59	29
	25 A	297	232	165	99	65	37	20
	30 A	297	218	113	47	20	9	7
	<b>Control</b>	297	296	296	295	293	293	292

Note: Average SS value of control sample = 295 mg/L

Figure 4.5 shows the removal efficiency in low and high current application during 30 minutes of treatment. The highest treatment was obtained by the application of 30 A where the removal efficiency reached 97.6% at 30 minutes. From what have been shown in figure 4.3 and figure 4.4, similar trend happen to the SS removal at 30 A where the removal efficiency was rapidly increasing before slowing down in last 15 minutes. For both application of 20 A and 25 A, the final removal efficiency was 90.2% and 93.2% respectively. The graph's curve was not so apparent among three application of low current where at the beginning, the removals was slow then rapidly increasing after 15 minutes. The final removal efficiency of 1 A, 2 A and 3 A application was 58.2, 62.6 and 67.7% respectively.



**Figure 4.5:** Percentage of SS removal

#### 4.4.4 Explanation of Organic and Inorganic Removal in EC Process

As shown in figures 4.3, 4.4 and 4.5, the result indicates that current application was the most influence factor in controlling the reaction rate of EC process. When the current intensity increased, the efficiency of ion production at anode and cathode increased simultaneously. This is applicable to all removals of COD, BOD and SS.  $Al^{3+}$  is a main coagulant agent in this process.  $Al^{3+}$  produced by electrochemically sacrificing of aluminum anode. The dosage was determined by electrical current loading. This can be proved by the control samples where the concentration of COD, BOD and SS were not apparently change because no  $Al^{3+}$  produced if no electrical current loaded to the process. Reactive time is the additional influence treatment efficiency of EC process, as the reaction time increased, higher production rate of hydroxyl and metal ion produced on the electrodes. The coagulant  $Al^{3+}$  dosage is depending on the amount of aluminum electrode dissolves in the electrolyte sample.

The amount of aluminum losses was calculated by weighing the electrodes before and after each run. Table 4.6 shows the amount of Al released as a function of different current and time.

**Table 4.6:** Weight dissolution of aluminum by experiment

Current (A)	Time (min)/ dosage (mg)						
	0	5	10	15	20	25	30
<b>1</b>	0 mg	1.7 mg	2.8 mg	3.9 mg	4.8 mg	5.6 mg	6.5 mg
<b>2</b>	0 mg	2.8 mg	4.7 mg	6.9 mg	8.8 mg	11 mg	13 mg
<b>3</b>	0 mg	3.9 mg	7.1 mg	9.7 mg	13.1 mg	16.1 mg	19.4 mg
<b>20</b>	0 mg	21.1 mg	42 mg	63.1 mg	83.5 mg	104 mg	124.7 mg
<b>25</b>	0 mg	26.5 mg	52.1 mg	78.2 mg	104.1 mg	129.8 mg	155.3 mg
<b>30</b>	0 mg	32.6 mg	62.8 mg	93.8 mg	124.8 mg	155.3 mg	186 mg

Table 4.7 shows the duly loss of aluminum electrode which calculated by Faraday's Law. From the law, the quantity of electricity passed through the electrolytic solution and the process running will determine the amount of aluminum electrode dissolved.

**Table 4.7:** Weight dissolution of aluminum by theory

Current (A)	Time (min)/ dosage (mg)						
	0	5	10	15	20	25	30
<b>1</b>	0 mg	1.0 mg	2.1 mg	3.1 mg	4.1 mg	5.2 mg	6.2 mg
<b>2</b>	0 mg	2.1 mg	4.1 mg	6.2 mg	8.3 mg	10.4 mg	12.4 mg
<b>3</b>	0 mg	3.1 mg	6.2 mg	9.3 mg	12.4 mg	15.5 mg	18.7 mg
<b>20</b>	0 mg	20.7 mg	41.5 mg	62.2 mg	82.9 mg	103.6 mg	124.4 mg
<b>25</b>	0 mg	25.9 mg	51.8 mg	77.7 mg	103.6 mg	129.6 mg	155.5 mg
<b>30</b>	0 mg	31.1 mg	62.2 mg	93.3 mg	124.4 mg	155.5 mg	186.6 mg

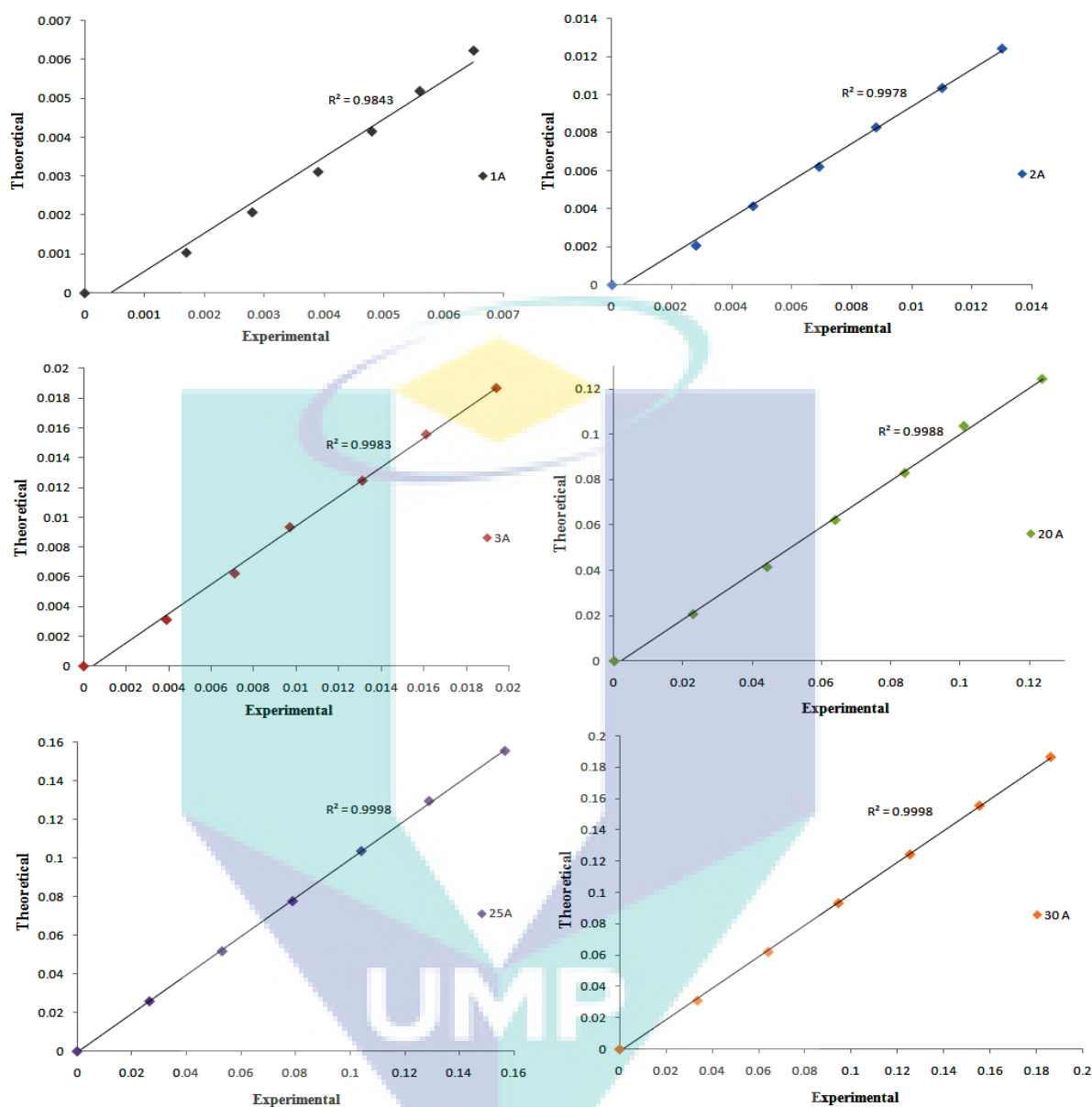
The results illustrated most of aluminum amount generated from anode always exceeded those calculated theoretically from Faraday's equation. This statement can be supported by Faradic efficiency. Table 4.8 shows the Faraday's efficiency of aluminum electrode loss. The reason may be due to the fact that Faraday's law does not take into account the impact of NaOH on metal dissolution while the NaOH has a strong effect on the rate of dissolution by increasing the conductivity of the solution. NaOH has been used as catalyst in order to enhance the process.

**Table 4.8:** Faradic efficiency of aluminum electrode dissolution

Current (A)	Time (sec)/ Faraday's efficiency (%)						
	0	5	10	15	20	25	30
<b>1</b>	NA	164.02%	135.08%	125.43%	115.78%	108.06%	104.52%
<b>2</b>	NA	135.08%	113.37%	110.96%	106.13%	106.13%	104.52%
<b>3</b>	NA	125.43%	114.17%	103.99%	105.33%	103.56%	103.99%
<b>20</b>	NA	101.79%	101.31%	101.47%	100.71%	100.34%	100.26%
<b>25</b>	NA	102.27%	100.54%	100.60%	100.44%	100.19%	99.89%
<b>30</b>	NA	104.85%	100.99%	100.56%	100.34%	99.89%	99.70%

Figure 4.6 shows the comparison between experimental and theoretical sacrificial aluminum anode consumption which has been achieved by calculating the theoretical aluminum loss (in gram) from the anode using Faradic equation and compared with those obtained experimentally.

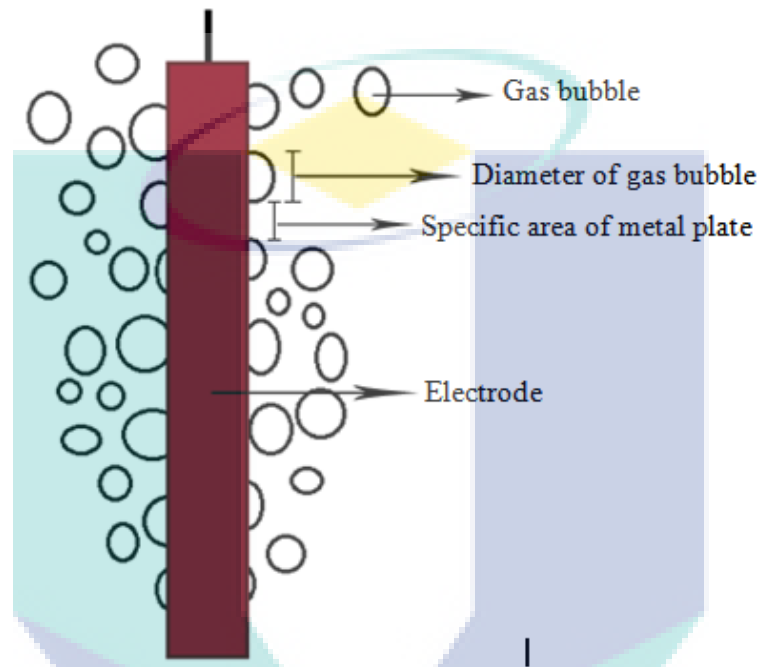




**Figure 4.6:** Experimental versus theoretical sacrificial electrode

Mass bubble also played a role in this process. As mentioned by Fukui and Yuu, (1985) the current directly influences the number and size of bubbles. Another previous study has reported that mass of bubbles produced is proportional to the current (Chen, 2004). This might explain the figures 4.3, 4.4 and 4.5 which the removal efficiency slowing down after certain time. The slowly increments of the removal efficiency while increasing

the current was mainly attributed to the number and size of gas bubbles whose specific area decreases with the diameter of gas bubbles and the influencing of bubbles production during electrolysis at one time. The figure 4.7 might showed the typical characteristic during the process.



**Figure 4.7:** Specific area decreases with the diameter of gas bubbles

#### 4.5 EFFECT OF INTER-ELECTRODE DISTANCE

The effect of inter electrode distance does not show much significant result in this experiment. However, when the distance between anode and cathode away too long, it will give the bad effect on the removal efficiency of COD, BOD and SS because the rate of electron transfer will become slow. Table 4.9 shows the value of COD, BOD and SS at 30 minutes of treatment.

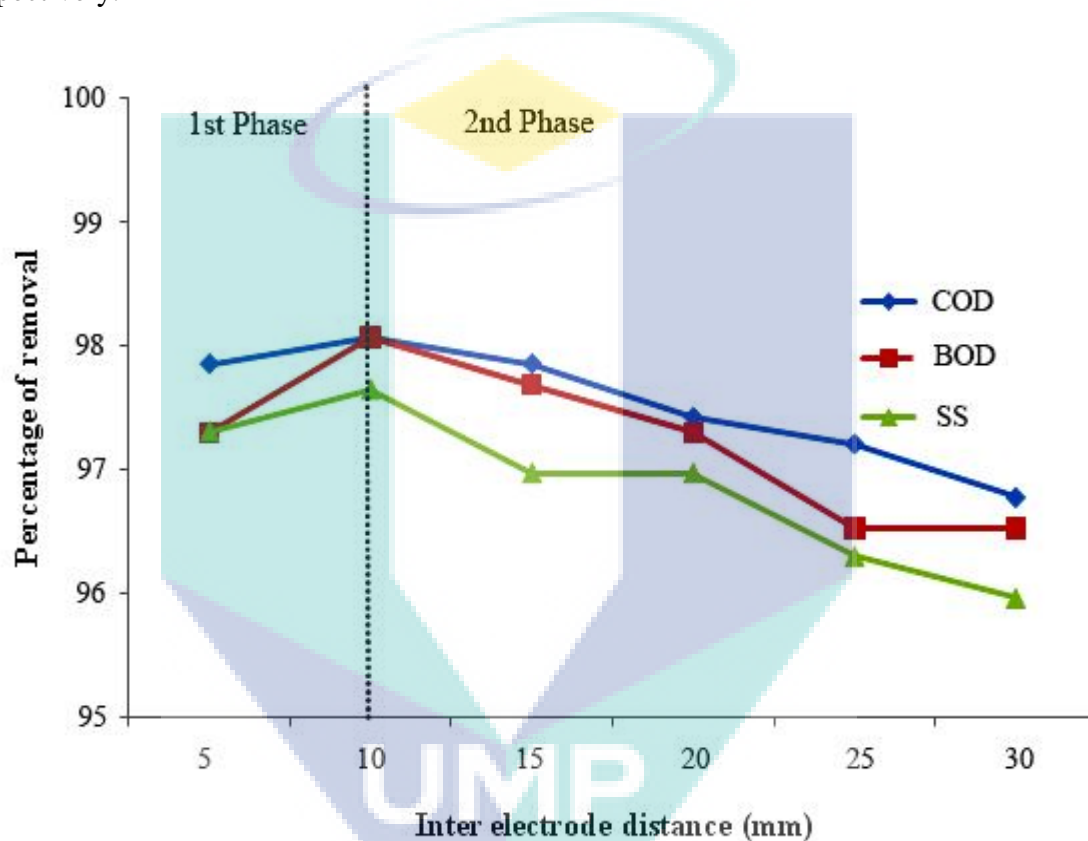
**Table 4.9:** The effect of inter-electrode distance on sewage's parameters

Inter-electrode distance (mm)	COD	BOD	SS
	Treatment sample	Treatment sample	Treatment sample
	5	10	7
10	9	5	7
15	10	6	9
20	12	7	9
25	13	9	11
30	15	9	12

Note: Average control sample for COD= 462.3 mg/L, BOD= 254.7 mg/L, SS= 293.8 mg/L

Variations of the percentage removal with inter electrode distance is shown in Figure 4.8. There were two phases involved in this experiment. The first phase was the state where the removal efficiency increasing and the second phase was the state where the removal efficiency decreasing. As shown in Figure 4.7, the increasing of inter-electrode distance from 5 to 10 mm leading to COD, BOD and SS removal efficiency increasing from 97.9%, 97.3% and 97.3% to 98.1%, 98.1% and 97.6% respectively. The increment probably occurred due to the electrostatic effect (Daneshvar *et al*, 2006). Electrostatic attraction is the attraction between atoms of opposite charge that holds the atoms together as ionic bonds. Any electric charge creates at the surrounding medium is called as electric field. Any electric field applies a force to any charged particle. Electrostatic force depends largely on the size of the structures and the distance between electrodes. In this case, the further the distance, the ion movements become slower and would be greater opportunity to generate aggregate flakes. Furthermore, these flakes were able to adsorb more molecules (Daneshvar *et al*, 2006). In the second phase, the removal efficiency of COD, BOD and SS decrease with increasing the inter electrode distance and this phenomena happens because of the increment of IR drop. This can be explained that decreasing the space between electrodes results in low resistance through the solution which in effect results in increasing

the rate of aluminum dissolution and  $Al^{3+}$  releases and consequently leads to more removal from the solution. On the other hand, decreasing the distance could enhance the flotation process by limiting the generated bubbles in a narrow space which results in higher removal efficiencies. For example, COD, BOD and SS removal efficiency decreases from 98.1%, 98.1% and 97.6% to 96.8%, 96.5% and 96% at 10 mm to 30 mm inter-electrode distances respectively.



**Figure 4.8:** Effect of inter electrode distance on removal efficiency

The electrostatic effect and IR drop play a big role in inter electrode distance in order to determine the removal efficiency of COD, BOD and SS. Therefore, the optimum distance between electrodes was at 10 mm.

#### 4.6 EFFECT OF PH

It has been established from the previous studies that pH is an important variable influencing the treatment performance of the EC process (Sengil and Ozacar, 2009; Kobya *et al*, 2006; Sengil *et al*, 2009). In order to examine its effect, the sewage water was adjusted to the desired pH by using potassium hydroxide (KOH) and hydrochloric acid (HCl). Table 4.10 shows the value of COD, BOD and SS where pH were varied from 3 to 11 at 30 A of current applied. In this experiment, the value of COD, BOD, and SS in neutral state (pH 7) are the lowest and the values increased in either acidic or alkaline sample solution.

**Table 4.10:** The effect of pH on sewage's parameters

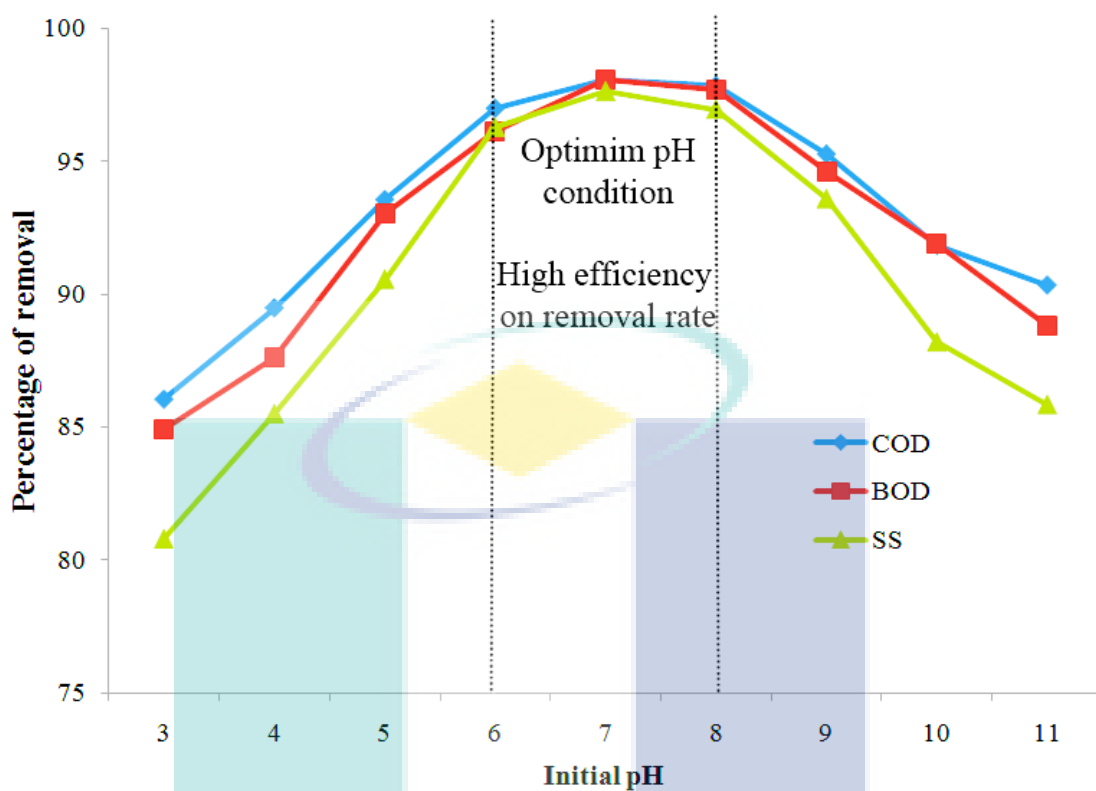
pH	COD		BOD		SS	
	Treatment sample	Control sample	Treatment sample	Control sample	Treatment sample	Control sample
3	65	463	39	255	57	294
4	49	462	32	256	43	295
5	30	462	18	255	28	294
6	14	463	10	253	11	293
7	9	462	5	255	7	295
8	10	464	6	255	9	294
9	22	463	14	254	19	294
10	38	462	21	253	35	293
11	45	462	29	255	42	293

Figure 4.9 showed the effect of pH on COD, BOD, and SS removal efficiency. From the figure, the removal efficiency were very low in pH 3 which can only remove 86%, 85% and 80.8% of COD, BOD and SS respectively. Almost similar with the sample

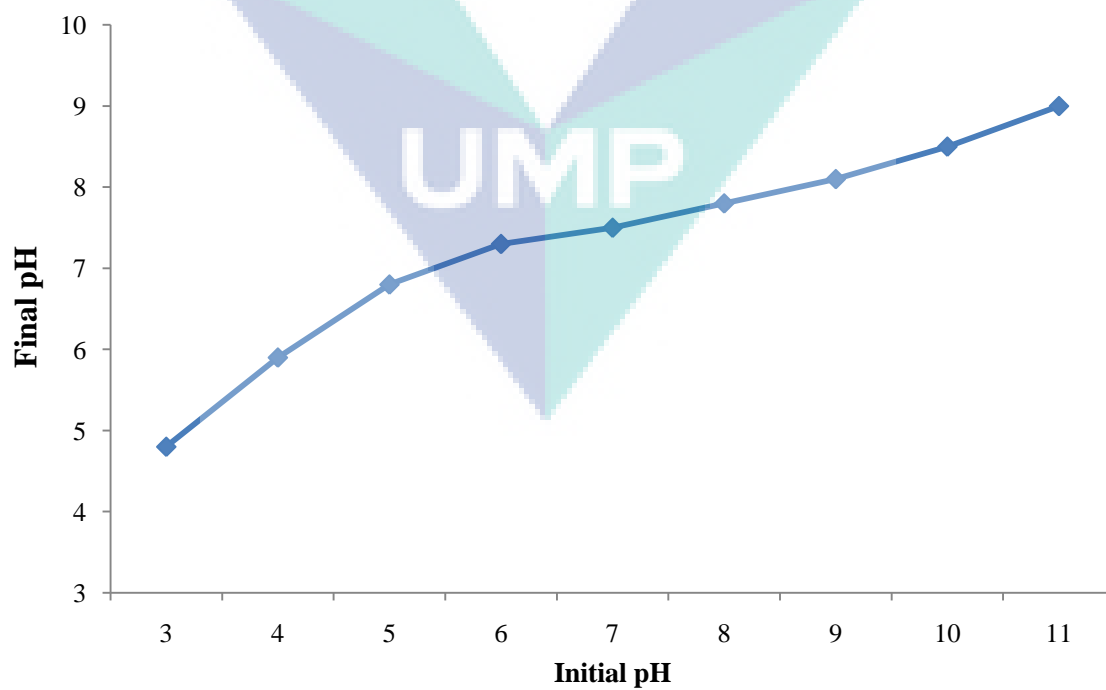
condition in pH 11 where the removal efficiency were also low which the removal efficiency for COD, BOD and SS were 90.3%, 88.8% and 85.9% respectively. However, in neutral electrolyte solution (initial pH 7), greater removal efficiency has been observed where for COD, BOD and SS were 98%, 98% and 97.7% respectively. Meanwhile in a little bit acidic and alkaline electrolyte solution, the removal efficiency for COD, BOD, and SS were close to the removal efficiency in neutral condition of electrolyte solution. For example in initial pH 8, the process can remove COD, BOD and SS of 97.8%, 97.6% and 97% respectively, as well as 97%, 96.1% and 96.3% respectively in initial pH 6.  $\text{Al(OH)}_2$  form in acidity condition but so in alkali condition,  $\text{Al(OH)}_3$  are produced.  $\text{Al(OH)}_3$  and  $\text{Al(OH)}_2$  settle while,  $\text{H}_2$  moves upward and causes flotation. Since  $\text{Al(OH)}_3$  has higher weight and density, it settles faster and has higher efficiency. However in natural condition, both of  $\text{Al(OH)}_3$  and  $\text{Al(OH)}_2$  are formed. Therefore, natural condition acts better in enmeshment of precipitate where the flocs of Aluminum Hydroxide acts as a blanket and brings all the stable suspended solid down while settling. Hence from the figure 4.9, the optimum range for pH values appeared between 6 and 8 which will provide an economical and effective treatment for the sewage water in practical.

The logo for UMP (Universiti Malaysia Perlis) is a large, downward-pointing arrow shape. It is composed of several overlapping triangles in shades of teal, light blue, and yellow. The letters 'UMP' are written in a bold, white, sans-serif font across the center of the arrow.

UMP

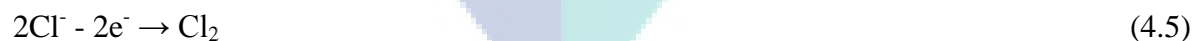


**Figure 4.9:** Effect of pH on removal efficiency, using 30 A current applied at 30 minutes



**Figure 4.10:** pH change during EC process

During EC process, initial pH of electrolyte will change due to the reaction happen in the sewage water. Figure 4.10 shows the pH of the sewage water after EC process. As being observed by other researcher (Vik *et al*, 1984), an increment of pH occurs when the initial pH is low. However, a pH drops occur when the initial pH is above 8. In other word this situation can give indirect benefit to the process, which the EC process can act as pH neutralization. This statement also supported based on the results of the present study and previous studies where EC process can act as a pH moderator (Bazrafshan *et al*, 2007; Kim *et. al.*, 2002). Then, there is no need to make further pH adjustment on sewage water after the treatment process and can be directly discharged into sewers because the range of initial sewage water's pH is normally from 6 to 9, so the final pH value will be over than 6.5 and less than 8.5. A complete examination of what happen in the EC process shows that there are the following electrochemical and chemical reactions occurs.





The reactions 4.1 to 4.8 were occurred during EC process. Reaction 4.2, 4.6 and 4.7 are quite complete and thus the total amount of  $H^+$  produced in reaction 4.2, 4.3, 4.6 and 4.7 was almost the same as that reduced in the hydrogen evolution. As shown in figure 4.10, when the initial pH was in the range of 3 to 7, the final pH was from 4.8 to 7.5 which could be explained by the excess of hydroxyl ion produced in cathode and by the liberation of  $OH^-$  due to the occurrence of a partial exchange of  $Cl^-$  with  $OH^-$  in  $Al(OH)_3$ . Actually there are a few mechanisms contributed to the pH change and one of them is the transfer of  $CO_2$  where at acidic condition,  $CO_2$  is oversaturated in sewage water that can be released. Releasing of this gas can make the  $H_2$  bubble disturbance which can cause pH increase. Other than that, some anions present in sewage water such as  $NO_3^-$ ,  $HCO_3^-$ ,  $SO_4^{2-}$ ,  $HSiO_4^-$  and many more can exchange partly with  $OH^-$  in  $Al(OH)_3$  to free  $OH^-$  which also causes a pH increase. When, the initial pH was in the range of 8 to 11, the final pH was from 9 to 7.8 indicate that the pH drop when it is in high pH condition. At high pH condition, the cation which present in the sewage water in the form of ionic metal such as  $Mg^+$ ,  $K^+$  and  $Ca^+$  can coprecipitate with  $Al(OH)_3$  in the form of metal hydroxide, leading to a pH decrease. Moreover, as  $Al(OH)_3$  is the typical amphoteric metal hydroxide, reaction 4.8 can take action at very high pH, leading to a pH decrease. Besides, the formation of  $Al(OH)_3$  near the anode would release hydrogen ion leading to decrease of pH. Furthermore, there is also oxygen evolution reaction leading to pH decrease.

#### 4.7 EXPERIMENTAL DESIGN FOR RSM

In this section, current ( $x_1$ ), treatment time ( $x_2$ ), electrode distance ( $x_3$ ) and pH ( $x_4$ ) were assigned to the statistical evaluation. A standard design in response surface methodology (RSM), central composite design (CCD) was applied to develop a polynomial regression equation in order to analyze the correlation between EC process preparation to the responses; percentage of COD removal ( $Y_1$ ), percentage of BOD removal ( $Y_2$ ) and percentage of SS removal ( $Y_3$ ). A complete design matrix EC process is given in Table 4.11.

**Table 4.11:** Complete design matrix of EC process

Run	Variables (coded level of variables)				Responses		
	Current	Time	Distance	pH	% COD	% BOD	% SS
	(A)	(min)	(mm)		removal	removal	removal
	$x_1$	$x_2$	$x_3$	$x_4$	$Y_1$	$Y_2$	$Y_3$
1	30.00	30.00	5.00	3.0	94.64	93.05	92.93
2	30.00	30.00	5.00	11.0	97.64	96.91	95.29
3	30.00	5.00	5.00	11.0	16.24	15.44	14.14
4	1.00	30.00	5.00	3.0	41.99	37.84	41.08
5	1.00	30.00	5.00	11.0	42.35	40.93	46.80
6	15.50	17.50	17.50	7.0	50.86	51.93	47.47
7	30.00	30.00	30.00	3.0	83.97	78.38	78.79
8	1.00	5.00	30.00	3.0	1.07	1.16	3.03
9	1.00	5.00	5.00	11.0	3.51	1.16	3.37
10	15.50	17.50	17.50	7.0	51.50	49.81	48.82
11	30.00	5.00	5.00	3.0	11.94	11.58	6.40
12	1.00	5.00	30.00	11.0	1.07	2.32	2.02
13	1.00	30.00	30.00	11.0	29.97	30.89	31.99
14	15.50	17.50	17.50	7.0	52.36	49.42	47.14
15	15.50	17.50	17.50	7.0	50.43	51.35	49.16
16	1.00	5.00	5.00	3.0	0.72	0.39	1.68
17	30.00	5.00	30.00	11.0	6.01	5.41	7.41
18	30.00	30.00	30.00	11.0	83.62	81.85	82.83
19	1.00	30.00	30.00	3.0	33.05	32.43	31.31
20	30.00	5.00	30.00	3.0	5.95	6.18	6.40
21	15.50	30.63	17.50	7.0	76.18	78.76	77.10
22	15.50	17.50	17.50	7.0	52.58	49.81	49.49
23	15.50	17.50	30.63	7.0	48.36	49.81	50.17
24	15.50	17.50	4.38	7.0	57.37	57.92	54.55
25	15.50	4.38	17.50	7.0	17.45	17.76	15.82
26	15.50	17.50	17.50	11.2	41.85	40.15	41.75
27	30.73	17.50	17.50	7.0	58.73	53.28	54.88
28	0.27	17.50	17.50	7.0	23.32	25.87	24.92
29	15.50	17.50	17.50	7.0	51.82	50.19	48.82
30	15.50	17.50	17.50	2.8	44.14	37.07	38.72

#### 4.7.1 Development of Empirical Model Equation for EC Process Preparation

Regression analysis is the general approach to fit the empirical model with the collected response variable data (Montgomery, 2001). For all responses, quadratic model were selected as suggested by the software. The coefficient of the full regression model equation and their statistical significance were determined and evaluated using Design-Expert software version 6.0.8. The final empirical formula models for responses in term of coded factors are presented by Equation 4.14 to 4.16, respectively.

% COD removal;

$$Y_1 = 51.19 + 15.57x_1 + 28.69x_2 - 4.05x_3 + 0.26x_4 - 8.67x_1^2 - 3.41x_2^2 + 2.07x_3^2 - 6.88x_4^2 + 11.17x_1x_2 - 1.09x_1x_3 + 0.43x_1x_4 - 1.73x_2x_3 - 0.45x_2x_4 - 0.86x_3x_4 \quad (4.9)$$

% BOD removal;

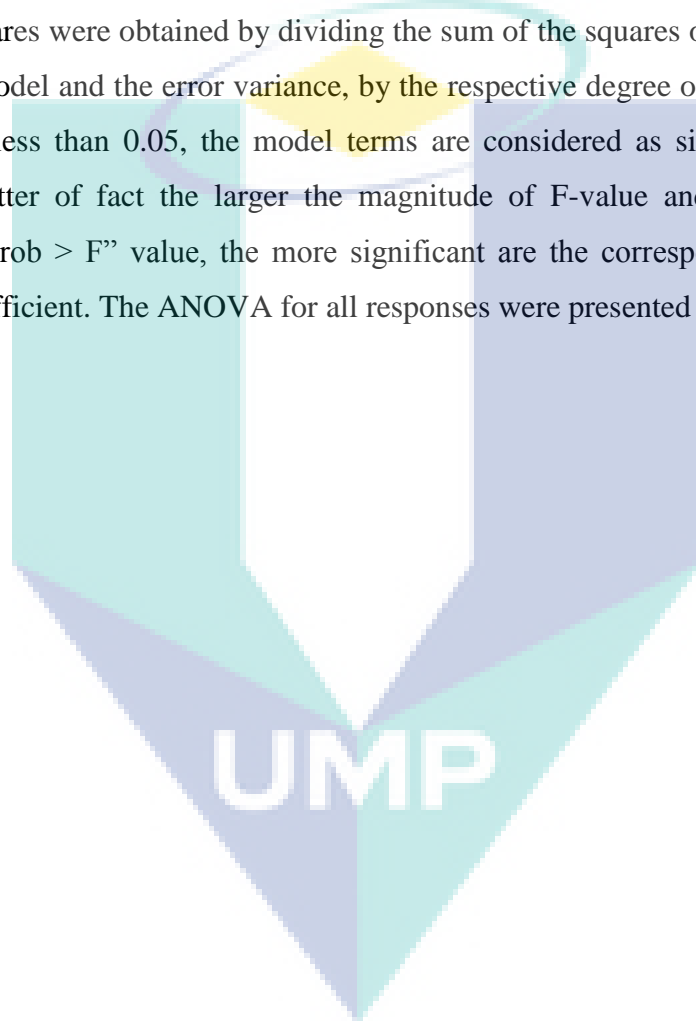
$$Y_2 = 50.25 + 14.86x_1 + 28.16x_2 - 3.69x_3 + 0.94x_4 - 9.28x_1^2 - 1.40x_2^2 + 3.68x_3^2 - 10.16x_4^2 + 10.91x_1x_2 - 1.98x_1x_3 + 0.43x_1x_4 - 1.98x_2x_3 + 0.24x_2x_4 - 0.58x_3x_4 \quad (4.10)$$

% SS removal;

$$Y_3 = 48.47 + 13.97x_1 + 28.61x_2 - 3.43x_3 + 1.40x_4 - 7.92x_1^2 - 1.97x_2^2 + 3.38x_3^2 - 7.62x_4^2 + 10.90x_1x_2 - 0.55x_1x_3 + 0.50x_1x_4 - 2.78x_2x_3 + 0.21x_2x_4 - 0.80x_3x_4 \quad (4.11)$$

For the responses, the model coefficient was estimated using multiple regression analysis technique which included in the RSM. The quality of the models developed was evaluated based on the standard deviation and correlation coefficient,  $R^2$  value. The capability of the model to predict the response is translated by closer of  $R^2$  value to unity. In this study, standard deviation of equations (4.9), (4.10) and (4.11) were recorded as 1.34, 1.27 and 1.33, respectively, while the  $R^2$  were closed to 0.9989, 0.9990 and 0.9989, respectively. These indicate that 99.89, 99.9 and 99.89% of the total variation in the responses, were attributed to the experimental variables studied. These values were considered high as the values were close to unity and lead to small variation in response predicted from these models.

The significance and adequacy of the models were further justified through analysis of variance (ANOVA). For each response, the statistical significance of quadratic model was determined by  $F$ -value and  $\text{Prob.} > F$ .  $F$ -value is a measurement of variance of data about the mean, based on the ratio of mean square of group variance due to error. The mean squares were obtained by dividing the sum of the squares of each of the variation sources, the model and the error variance, by the respective degree of freedom (Sahu et al., 2010). The mean squares were obtained by dividing the sum of the squares of each of the variation sources, the model and the error variance, by the respective degree of freedom. If the value of  $\text{Prob.} > F$  less than 0.05, the model terms are considered as significant (Sahu et al., 2010). As matter of fact the larger the magnitude of  $F$ -value and correspondingly the smaller the “ $\text{Prob} > F$ ” value, the more significant are the corresponding model and the individual coefficient. The ANOVA for all responses were presented in Tables 4.12 to 4.14.



**Table 4.12:** ANOVA for response surface quadratic model for COD removal

Source	Sum of Squares	DF	Mean Square	F Value	Prob > F	
<b>Block</b>	568.65	1	568.65			
<b>Model</b>	22896.82	14	1635.49	904.21	< 0.0001	significant
<b>x<sub>1</sub></b>	4413.42	1	4413.42	2440.03	< 0.0001	
<b>x<sub>2</sub></b>	14989.39	1	14989.39	8287.13	< 0.0001	
<b>x<sub>3</sub></b>	298.83	1	298.83	165.21	< 0.0001	
<b>x<sub>4</sub></b>	1.20	1	1.20	0.66	0.4286	
<b>x<sub>1</sub><sup>2</sup></b>	229.72	1	229.72	127.01	< 0.0001	
<b>x<sub>2</sub><sup>2</sup></b>	35.61	1	35.61	19.69	0.0006	
<b>x<sub>3</sub><sup>2</sup></b>	13.05	1	13.05	7.22	0.0177	
<b>x<sub>4</sub><sup>2</sup></b>	144.72	1	144.72	80.01	< 0.0001	
<b>x<sub>1</sub>x<sub>2</sub></b>	1996.77	1	1996.77	1103.95	< 0.0001	
<b>x<sub>1</sub>x<sub>3</sub></b>	19.17	1	19.17	10.60	0.0057	
<b>x<sub>1</sub>x<sub>4</sub></b>	2.99	1	2.99	1.66	0.2191	
<b>x<sub>2</sub>x<sub>3</sub></b>	47.95	1	47.95	26.51	0.0001	
<b>x<sub>2</sub>x<sub>4</sub></b>	3.24	1	3.24	1.79	0.2018	
<b>x<sub>3</sub>x<sub>4</sub></b>	11.93	1	11.93	6.60	0.0223	
<b>Residual</b>	25.32	14	1.81			
<b>Lack of Fit</b>	22.92	10	2.29	3.81	0.1045	not significant
<b>Pure Error</b>	2.40	4	0.60			
<b>Cor Total</b>	23490.79	29				

Table 4.12 shows ANOVA for response surface quadratic model for COD removal. The model  $F$ -value of 904.21 and  $\text{Prob}>F$  was less than 0.0001 implied that the model was significant. In this case  $x_1$ ,  $x_2$ ,  $x_3$ ,  $x_1^2$ ,  $x_2^2$ ,  $x_3^2$ ,  $x_4^2$ ,  $x_1x_2$ ,  $x_1x_3$ ,  $x_2x_3$  and  $x_3x_4$  were significant model terms. Other model terms especially the main effect of pH ( $x_4$ ) was relatively less

significant in influencing COD removal as their confidence level were less than 95% ( $P > 0.05$ )

**Table 4.13:** ANOVA for response surface quadratic model for BOD removal

Source	Sum of Squares	DF	Mean Square	F Value	Prob > F	
<b>Block</b>	556.99	1	556.99			
<b>Model</b>	22055.67	14	1575.41	969.92	< 0.0001	significant
<b>x<sub>1</sub></b>	4018.07	1	4018.07	2473.77	< 0.0001	
<b>x<sub>2</sub></b>	14438.40	1	14438.40	8889.17	< 0.0001	
<b>x<sub>3</sub></b>	248.02	1	248.02	152.70	< 0.0001	
<b>x<sub>4</sub></b>	16.13	1	16.13	9.93	0.0071	
<b>x<sub>1</sub><sup>2</sup></b>	263.13	1	263.13	162.00	< 0.0001	
<b>x<sub>2</sub><sup>2</sup></b>	6.01	1	6.01	3.70	0.0749	
<b>x<sub>3</sub><sup>2</sup></b>	41.39	1	41.39	25.48	0.0002	
<b>x<sub>4</sub><sup>2</sup></b>	315.11	1	315.11	194.00	< 0.0001	
<b>x<sub>1</sub>x<sub>2</sub></b>	1903.58	1	1903.58	1171.96	< 0.0001	
<b>x<sub>1</sub>x<sub>3</sub></b>	62.57	1	62.57	38.52	< 0.0001	
<b>x<sub>1</sub>x<sub>4</sub></b>	3.01	1	3.01	1.85	0.1949	
<b>x<sub>2</sub>x<sub>3</sub></b>	62.73	1	62.73	38.62	< 0.0001	
<b>x<sub>2</sub>x<sub>4</sub></b>	0.93	1	0.93	0.57	0.4615	
<b>x<sub>3</sub>x<sub>4</sub></b>	5.36	1	5.36	3.30	0.0908	
<b>Residual</b>	22.74	14	1.62			
<b>Lack of Fit</b>	18.32	10	1.83	1.66	0.3305	not significant
<b>Pure Error</b>	4.42	4	1.10			
<b>Cor Total</b>	22635.40	29				

Table 4.13 shows ANOVA for response surface quadratic model for BOD removal. The quadratic model was significant due the model  $F$ -value of 969.92 and  $\text{Prob}>F$  was less

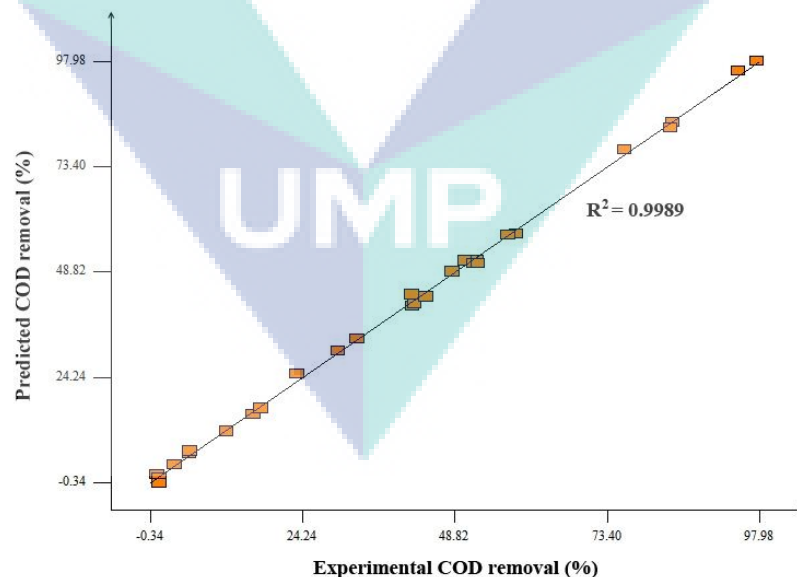
than 0.0001. From the table,  $x_1, x_2, x_3, x_4, x_1^2, x_3^2, x_4^2, x_1x_2, x_1x_3, x_2x_3$  were significant model term while  $x_2^2, x_1x_4, x_2x_4, x_3x_4$  were insignificant model term to the response as their confidence level were less than 95% ( $P > 0.05$ ).

**Table 4.14:** ANOVA for response surface quadratic model for SS removal

Source	Sum of Squares	DF	Mean Square	F Value	Prob > F	
<b>Block</b>	506.81	1	506.81			
<b>Model</b>	21626.45	14	1544.75	877.28	< 0.0001	significant
<b>x<sub>1</sub></b>	3554.14	1	3554.14	2018.45	< 0.0001	
<b>x<sub>2</sub></b>	14905.32	1	14905.32	8464.95	< 0.0001	
<b>x<sub>3</sub></b>	214.63	1	214.63	121.89	< 0.0001	
<b>x<sub>4</sub></b>	35.47	1	35.47	20.14	0.0005	
<b>x<sub>1</sub><sup>2</sup></b>	191.67	1	191.67	108.85	< 0.0001	
<b>x<sub>2</sub><sup>2</sup></b>	11.86	1	11.86	6.74	0.0212	
<b>x<sub>3</sub><sup>2</sup></b>	34.92	1	34.92	19.83	0.0005	
<b>x<sub>4</sub><sup>2</sup></b>	177.25	1	177.25	100.66	< 0.0001	
<b>x<sub>1</sub>x<sub>2</sub></b>	1901.18	1	1901.18	1079.71	< 0.0001	
<b>x<sub>1</sub>x<sub>3</sub></b>	4.79	1	4.79	2.72	0.1215	
<b>x<sub>1</sub>x<sub>4</sub></b>	4.07	1	4.07	2.31	0.1507	
<b>x<sub>2</sub>x<sub>3</sub></b>	123.49	1	123.49	70.13	< 0.0001	
<b>x<sub>2</sub>x<sub>4</sub></b>	0.71	1	0.71	0.40	0.5357	
<b>x<sub>3</sub>x<sub>4</sub></b>	10.22	1	10.22	5.81	0.0303	
<b>Residual</b>	24.65	14	1.76			
<b>Lack of Fit</b>	21.48	10	2.15	2.70	0.1750	not significant
<b>Pure Error</b>	3.18	4	0.79			
<b>Cor Total</b>	22157.91	29				

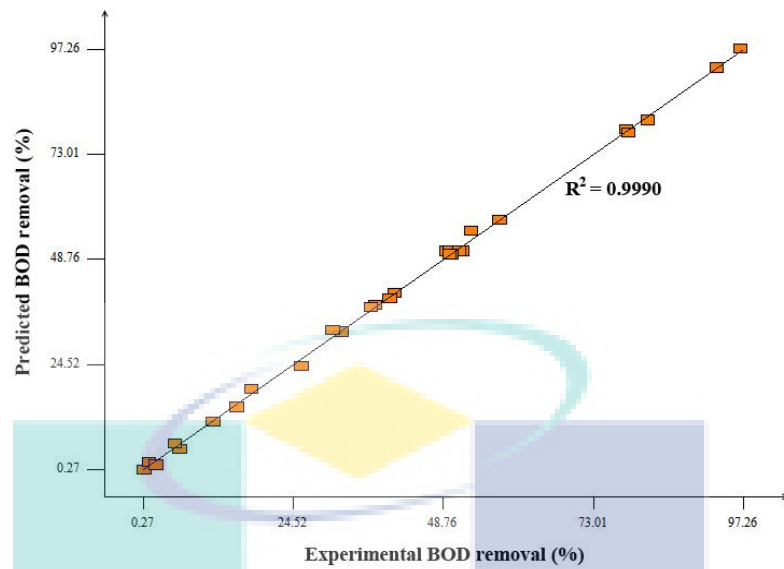
Table 4.14 shows ANOVA for response surface quadratic model for SS removal. The model  $F$ -value of 877.28 and  $\text{Prob}>F$  of less than 0.0001 implied that the model was significant. In this case  $x_1, x_2, x_3, x_4, x_1^2, x_2^2, x_3^2, x_4^2, x_1x_2, x_2x_3, x_3x_4$  were significant model term while  $x_1x_3, x_1x_4, x_2x_4$  were less significant model term to the response.

Based on the statistical result obtained, the models were adequate to predict the removal efficiency within the range of variable studies.  $\text{Prob}>F$  is the probability that the variation in the results are due to random error and thus the low values obtained for all models reflect the result are not random and the terms model have a significant effect in the response. In addition, Figures 4.11 to 4.13 show the predicted values versus the experimental values for removal efficiency of COD, BOD and SS respectively. Experimental values are the measured response data for a particular run, and the predicted values evaluated from the model and generated by using the approximating functions. It can be seen that the response models show good fits to the experimental data that reflected to good predictions of the models.

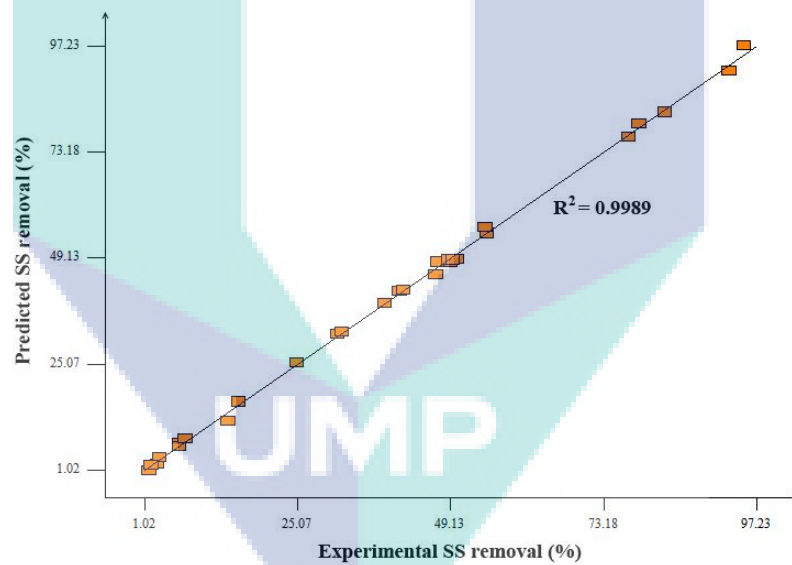


**Figure 4.11:** Predicted versus experimental percentage of COD removal





**Figure 4.12:** Predicted versus experimental percentage of BOD removal



**Figure 4.13:** Predicted versus experimental percentage of SS removal

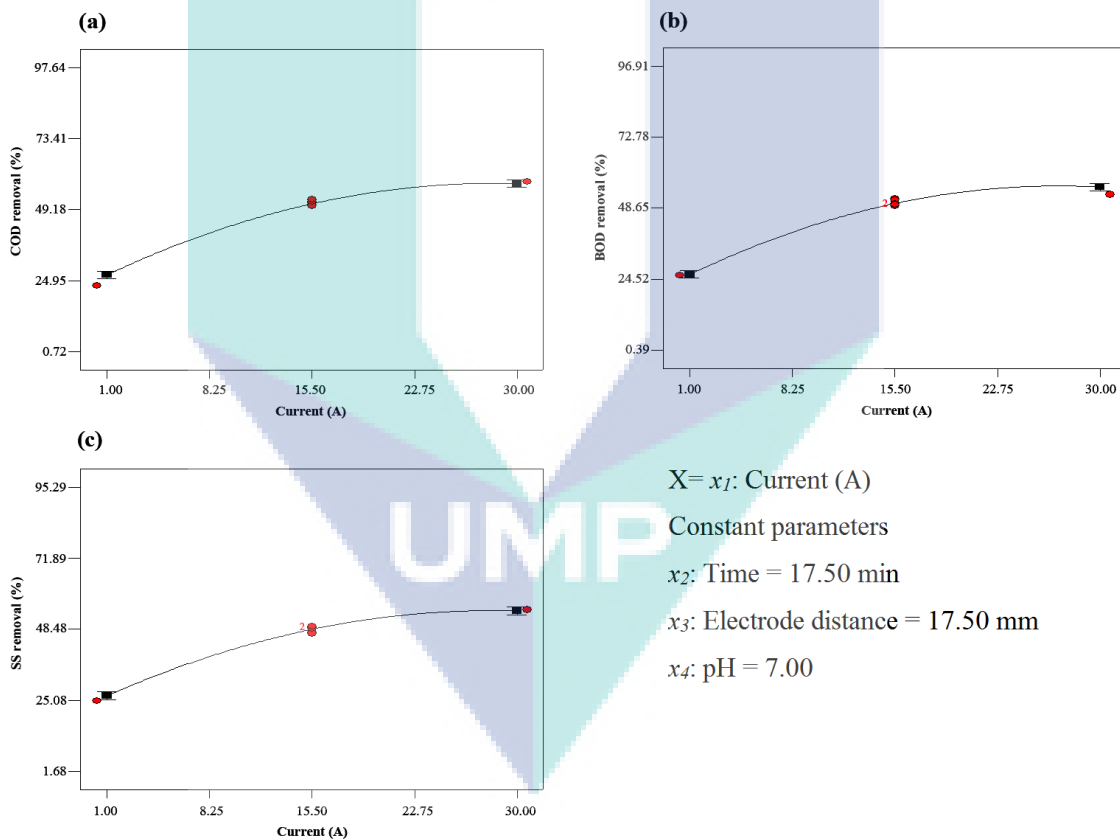
#### 4.7.2 Effect of Single Process Variables

Tables 4.12 to 4.14 indicate that the individual parameters studies; current applied ( $x_1$ ), operating time ( $x_2$ ), electrode distance ( $x_3$ ) and pH ( $x_4$ ) demonstrated significant to the developed models. Among of them, the operating time ( $x_2$ ) has the most significant effect

by the highest F-values which 8287.13, 8889.17 and 8464.95 were obtained from removal of COD, BOD and SS respectively. While pH ( $x_4$ ) has the less significant effect by the lowest F-values, where 0.66, 9.93 and 20.14 were obtained from the removal of COD, BOD and SS respectively.

#### 4.7.2.1 Current Application ( $x_1$ )

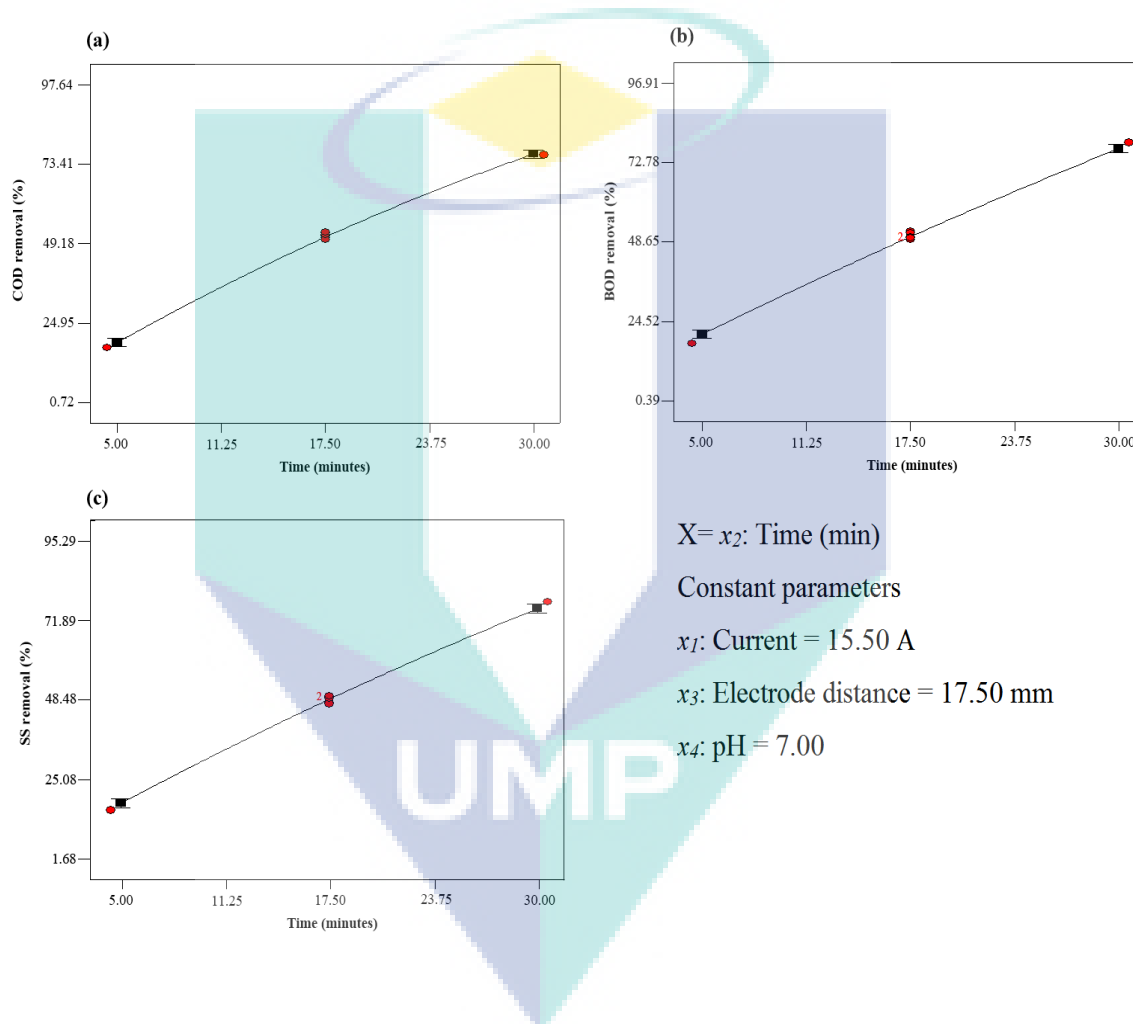
Based on Figure 4.14, the removal of COD, BOD and SS were increased as the current increase from 1 A to 30 A.



**Figure 4.14:** Individual effect of current applied: (a) COD removal, (b) BOD removal, (c) SS removal

#### 4.7.2.2 Operating Time ( $x_2$ )

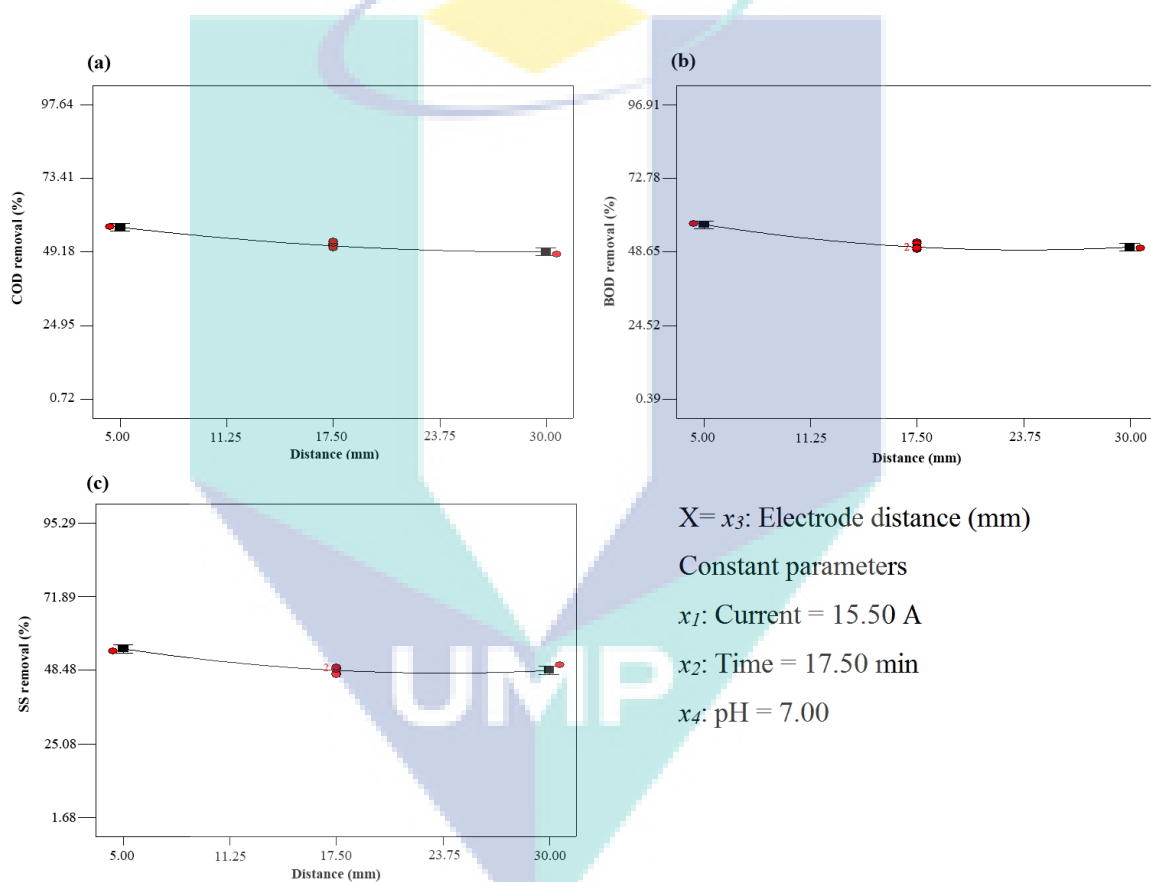
The operating time was found to induce the increase of COD, BOD and SS removal, as shown in Figure 4.15. This parameter actually gives the most effect toward the responses as describe by the F-value in table 4.12 to 4.14.



**Figure 4.15:** Individual effect of operating time: (a) COD removal, (b) BOD removal, (c) SS removal

### 4.7.2.3 Inter-electrode Distance ( $x_3$ )

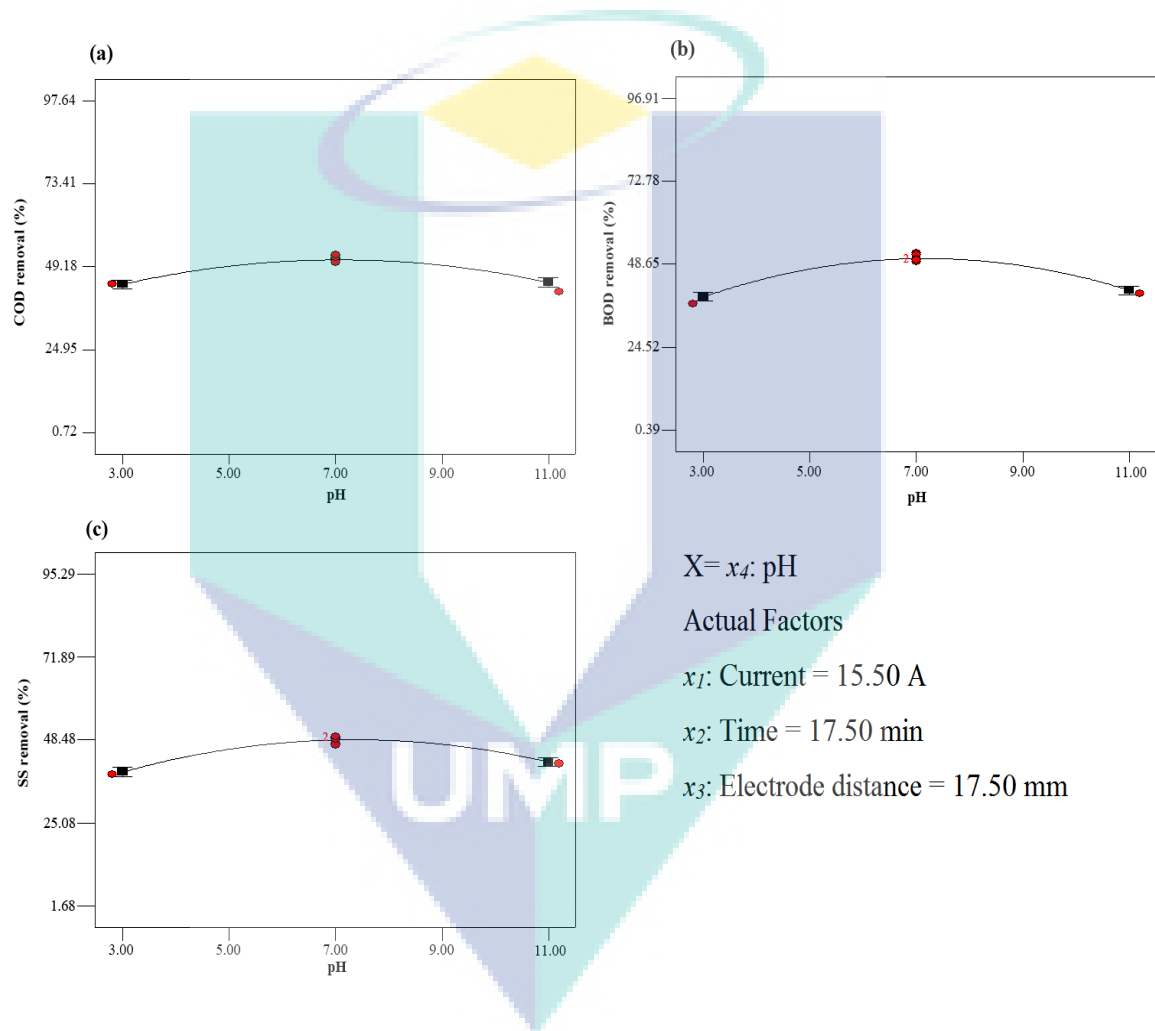
Figure 4.16 shows that there were slightly decreased in COD, BOD and SS removal, when the gap between anode and cathode increased. However this parameter gives only less effect toward the responses as describe by the F-value in table 4.9 to 4.11. Thus electrode distance can be use as a tool to fine tune for the effect of other parameter since it exhibit small effect to the treatment process.



**Figure 4.16:** Individual effect of electrode distances: (a) COD removal, (b) BOD removal, (c) SS removal

#### 4.7.2.4 pH ( $x_4$ )

The value of pH was found to have a significant effect on the removal of COD, BOD and SS, as shown in Figure 4.17. The best removal was appeared when the value of pH was in the range of 6 to 8.



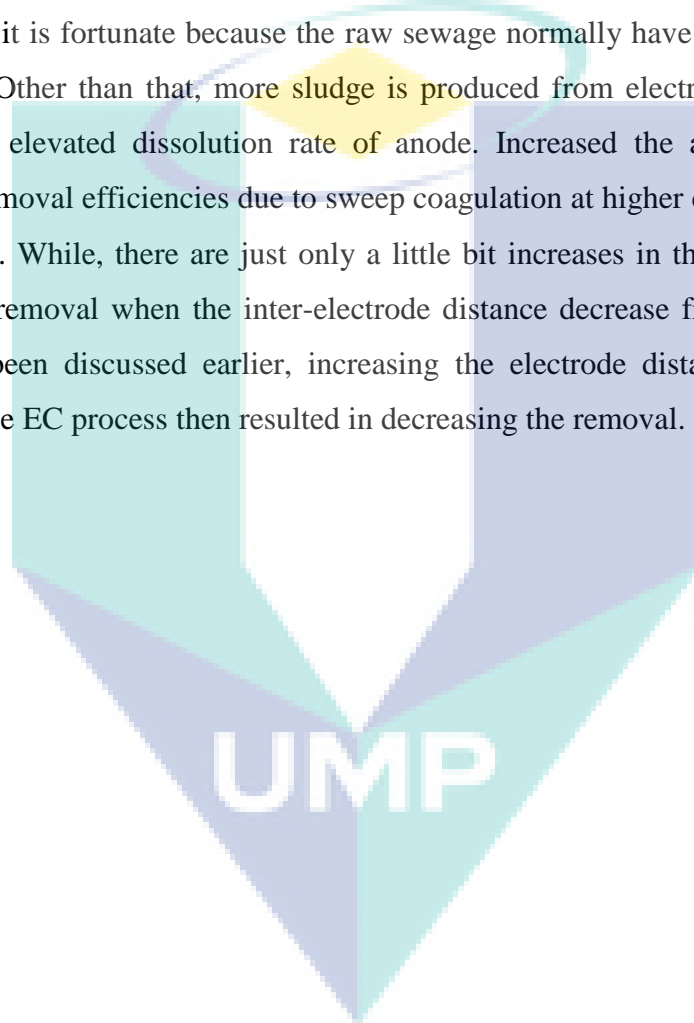
**Figure 4.17:** Individual effect of pH: (a) COD removal, (b) BOD removal, (c) SS removal

### 4.7.3 Combine Effect on COD, BOD and SS

The combine parameters of current applied with operating time ( $x_1x_2$ ), current applied with electrode distance ( $x_1x_3$ ), current applied with pH ( $x_1x_4$ ), operating time with electrode distance ( $x_2x_3$ ), operating time with pH ( $x_2x_4$ ) and electrode distance with pH ( $x_3x_4$ ) were studied. Tables 4.12 to 4.14 indicate that all the combine parameters studies were significant to the developed models except the combination of  $x_1x_4$  and  $x_2x_4$  which F-value is 1.66 and 1.74 respectively for COD removal model;  $x_1x_4$ ,  $x_2x_4$  and  $x_3x_4$  which F-value is 1.85, 0.57 and 3.30 respectively for BOD removal model; and  $x_1x_3$ ,  $x_1x_4$  and  $x_2x_4$  which F-value is 2.72, 2.31 and 0.40 respectively for SS removal model. Among the combine parameters,  $x_1x_2$  had the most significant effect by the highest F-value which 1103.95, 1171.96 and 1079.71 obtained from COD, BOD and SS removal respectively. The significant of current application with operating time ( $x_1x_2$ ) were shown in Figure 4.18, 4.19 and 4.20 where the highest removal efficiency for COD, BOD and SS reach up to 94.5%, 93.5% and 92.1% respectively.

Equations 4.14, 4.15 and 4.16 in EC process are used to visualize the effects of experimental factors on responses in 3D graphs. 3D surface plots in Figure 4.18 to 4.20 were shown to visualize the more than one factor and responses. As shown in Figure 4.18 to 4.20 and Table 4.11, all COD, BOD and SS removals showed a similar removal efficiency trends. The percentage of COD, BOD and SS removal are found to increase with an increase in operating time at any value of current application, electrode distance and pH. Similar pattern was observed for current application. Generally, it is known that COD, BOD, and SS removals increase with increasing of coagulant dosages in EC. It may be inferred from Figure 4.18 to 4.20, that higher COD, BOD and SS removals can be achieved at higher current and operating time. At higher current, higher dissolution of electrode material (Faraday's law) with higher rate of formation of aluminum hydroxides and some polymeric metal complexes may result in higher removal due to their co-precipitation. The monomeric species such as  $\text{Al}(\text{OH})_2^+$ ,  $\text{Al}(\text{OH})_3^+$ ,  $\text{Al}_2(\text{OH})_2^{4+}$ , and  $\text{Al}(\text{OH})_4$  and polymeric species such as  $\text{Al}_6(\text{OH})_{15}^{3+}$ ,  $\text{Al}_7(\text{OH})_{17}^{4+}$ ,  $\text{Al}_8(\text{OH})_{20}^{4+}$ ,  $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$ , and  $\text{Al}_{13}(\text{OH})_{34}^{5+}$  are formed during the EC process (Can *et al*, 2003). Formation rates of these different

species depend on pH of the medium and the types of ions present which play an important role in the EC process. A decrease of the removal efficiency at pH less than 5 and higher than 8 is observed. The occurrence of this behavior is attributed to an amphoteric behavior of  $\text{Al(OH)}_3$  which led to the soluble of  $\text{Al}^{3+}$  cations when the initial pH was low and led to the monomeric anions,  $\text{Al(OH)}_4^-$  when the initial pH was high. These soluble species are useless for water treatment. Therefore, initial pH in the EC process is kept in the range of 5–8. Actually, it is fortunate because the raw sewage normally have the value of pH in the range of 5-8. Other than that, more sludge is produced from electrodes at higher current values due to elevated dissolution rate of anode. Increased the amount of sludge can enhance the removal efficiencies due to sweep coagulation at higher current application and operating time. While, there are just only a little bit increases in the percentage of COD, BOD and SS removal when the inter-electrode distance decrease from 30 mm to shorter distance. As been discussed earlier, increasing the electrode distance will increase the resistance to the EC process then resulted in decreasing the removal.



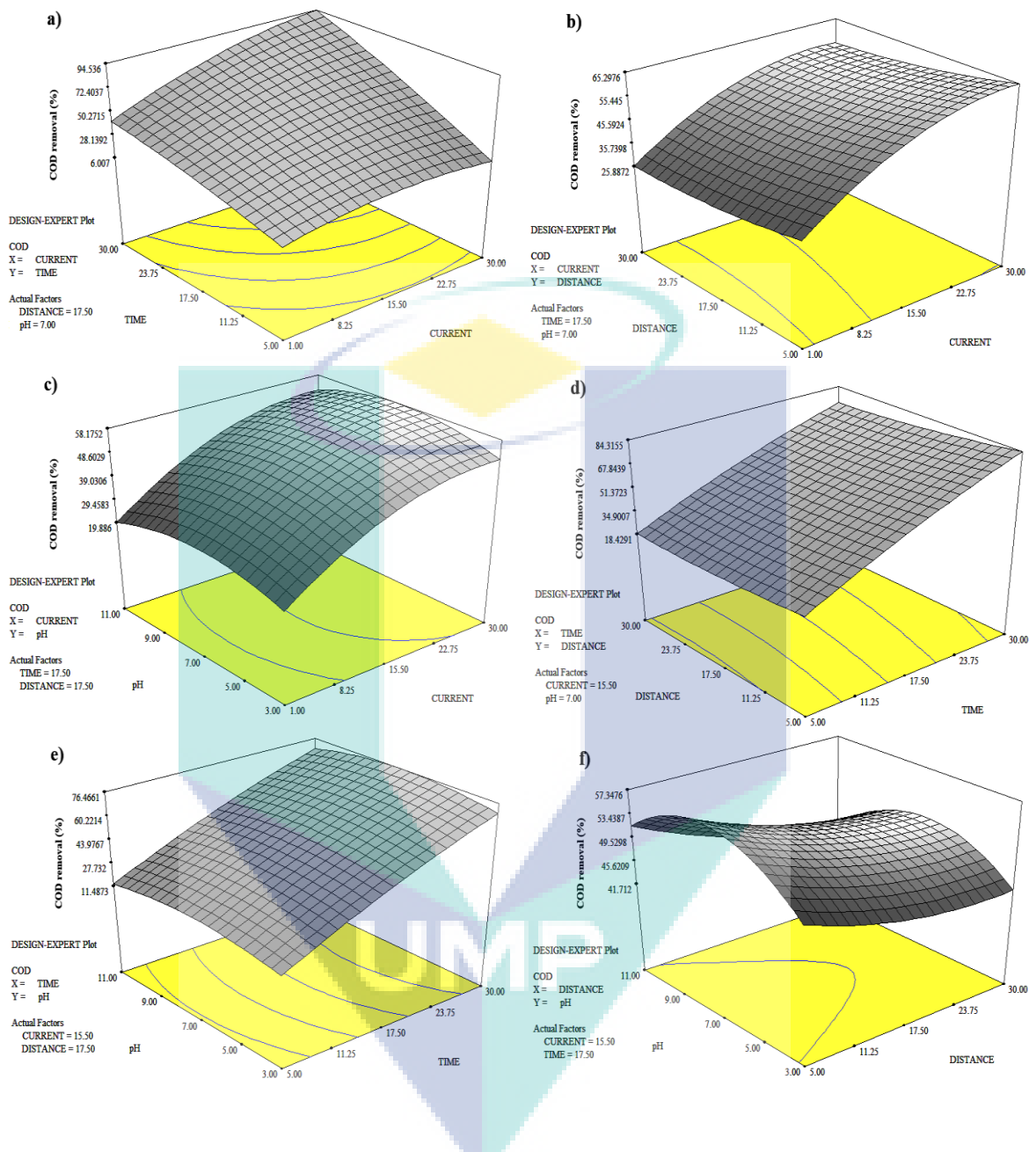


Figure 4.18: 3D surface plot for COD removal efficiency



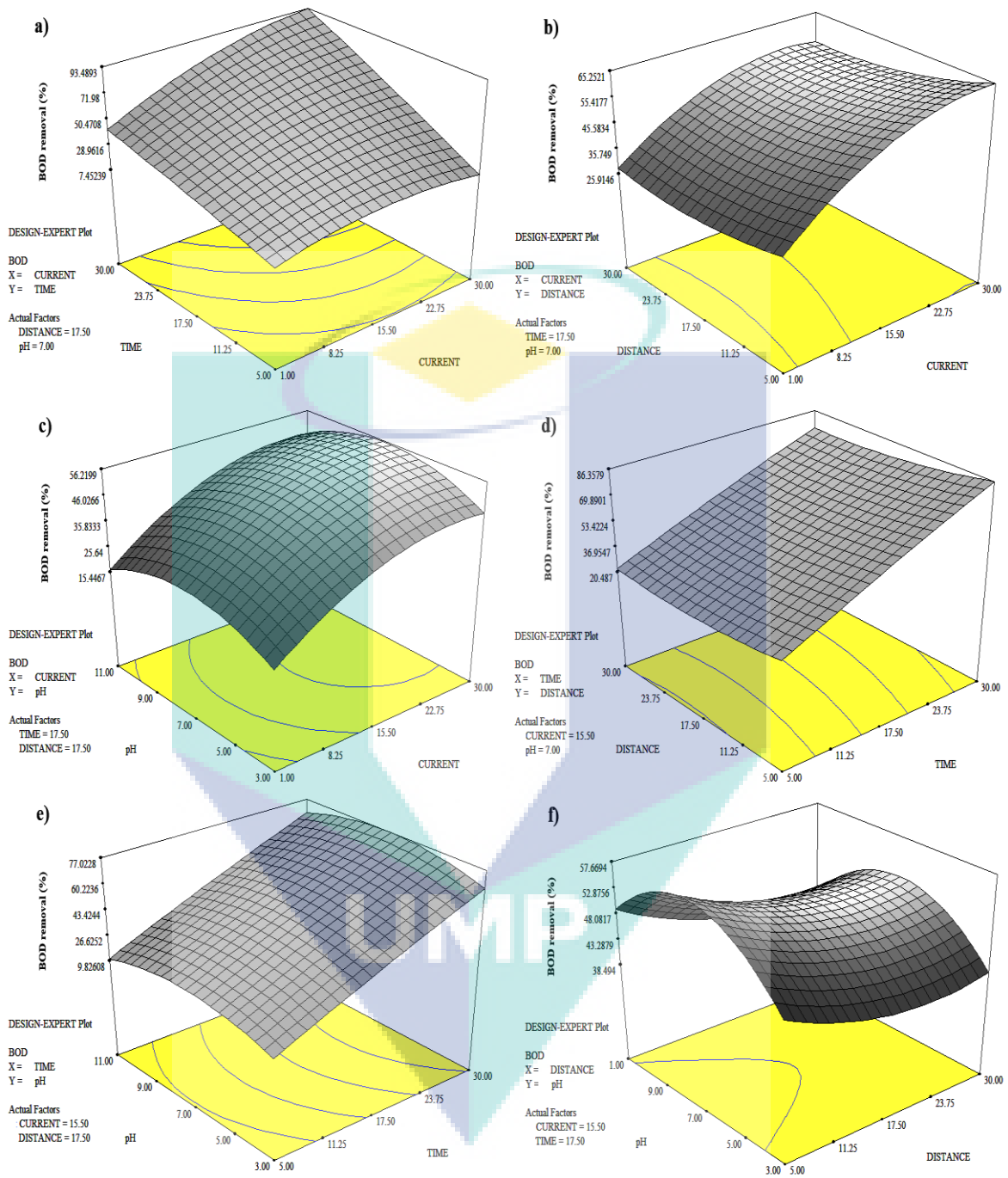


Figure 4.19: 3D surface plot for BOD removal efficiency

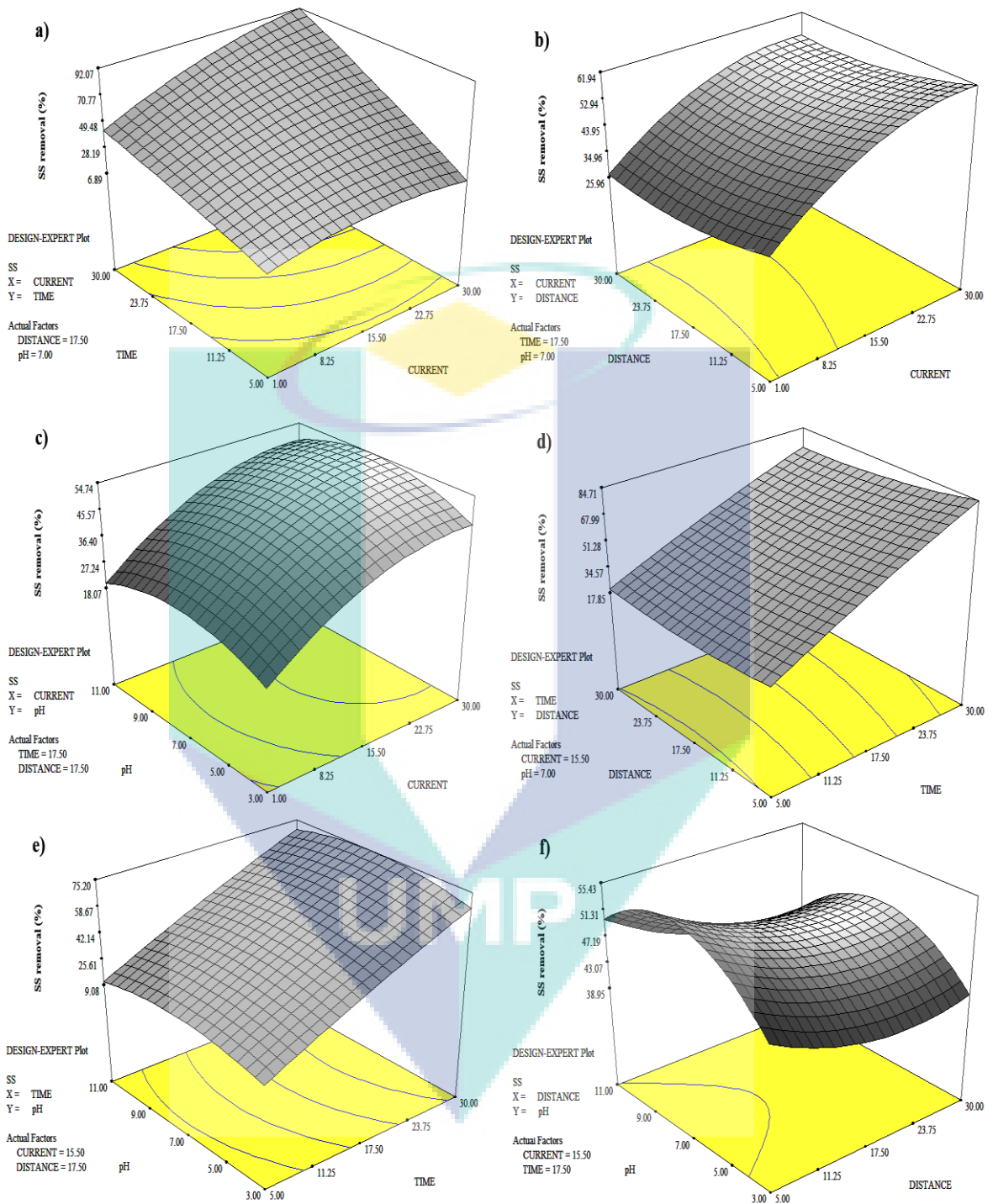


Figure 4.20: 3D surface plot for SS removal efficiency

#### 4.7.4 Optimization of Experimental Parameters

It is necessary to find out the optimal point as a compromise among current applied, operating time, inter-electrode distance and pH values. The desirability function approach is one of the most widely used methods for optimization of multiple response processes. In this case, Design Expert software with desirability function was applied to compromise the responses. The experimental conditions with the highest desirability are shown in Table 4.15. When operating variables were in range, the removal efficiencies of COD, BOD and SS was maximized in the RSM model with desirability of 1.000. Optimization results for the maximum removal efficiencies of COD, BOD and SS as 97.84%, 97.77% and 95.72% respectively at 29.09 A of current, 29.86 minutes of operating time, 10.71 mm of inter-electrode distance and pH value of 7.04 in the EC process were obtained for the system. It is clear that all 6 mg/L of COD, 3mg/L for BOD and 10mg/L for SS satisfied the effluent standard A for STP. Based on the above results, the EC process may be now to suggest in engineering application especially to treat sewage.

**Table 4.15:** Optimization result of variables and responses

Name	Goal	Lower Limit	Upper Limit	Optimization Result
<b>Current, <math>x_1</math></b>	is in range	20	30	29.09
<b>Time, <math>x_2</math></b>	is in range	15	30	29.86
<b>Distance, <math>x_3</math></b>	is in range	5	15	10.71
<b>pH, <math>x_4</math></b>	is in range	5	8	7.04
<b>% COD Removal</b>	maximize	0.72	97.64	97.84
<b>% BOD Removal</b>	maximize	0.39	96.91	97.77
<b>% SS Removal</b>	maximize	1.68	95.29	95.72
<b>Desirability</b>	maximize	0	1	1

3D surface plots in Figure 4.20 to 4.22 were shown to visualize the effects of experimental factors on percentage of COD, BOD and SS removal responses under

optimized conditions. As seen from Figure 4.21 to 4.23, COD, BOD and SS removal efficiencies showed the similar removal trends with experiments results.

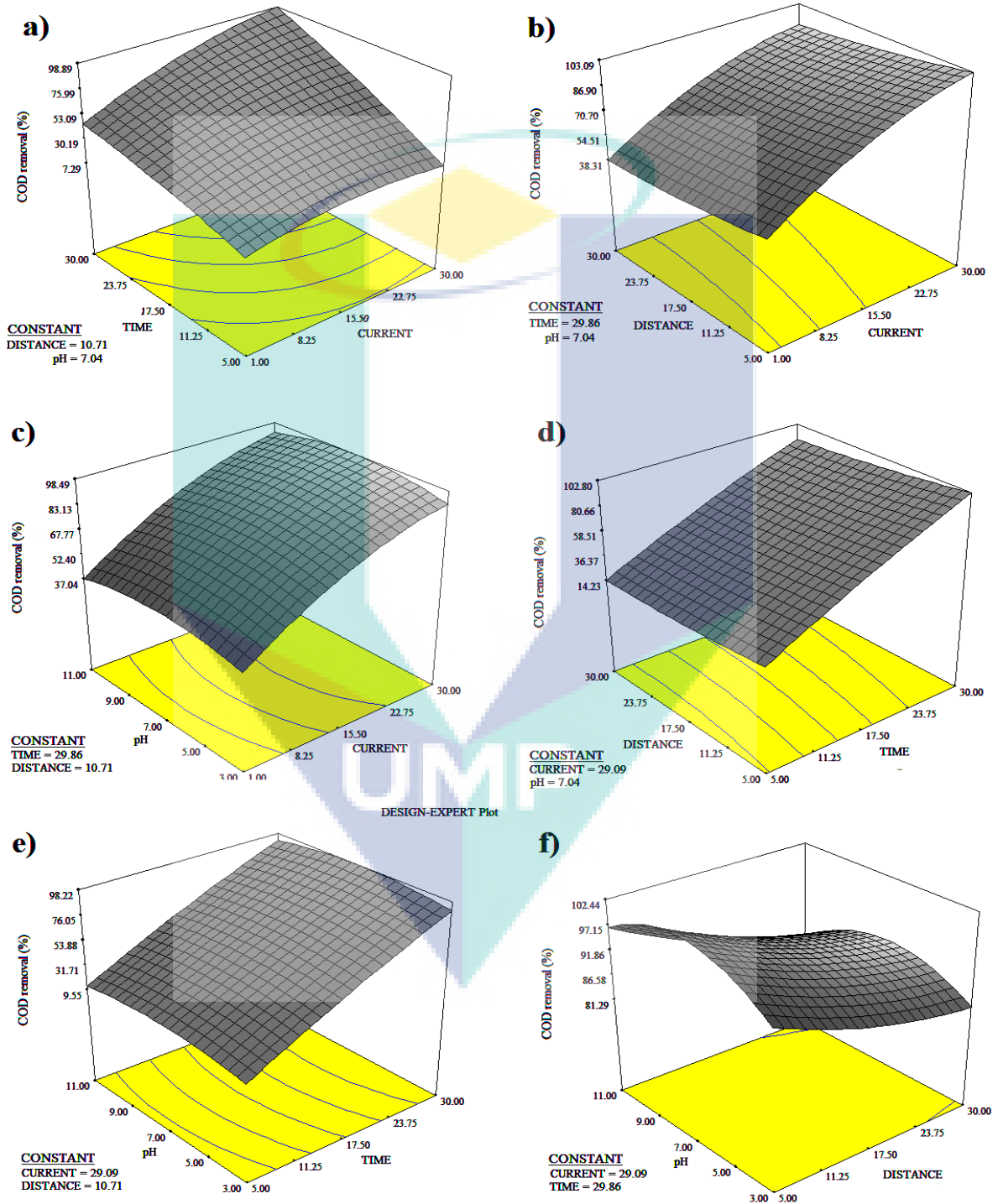


Figure 4.21: Optimization in 3D plot of COD removal efficiency

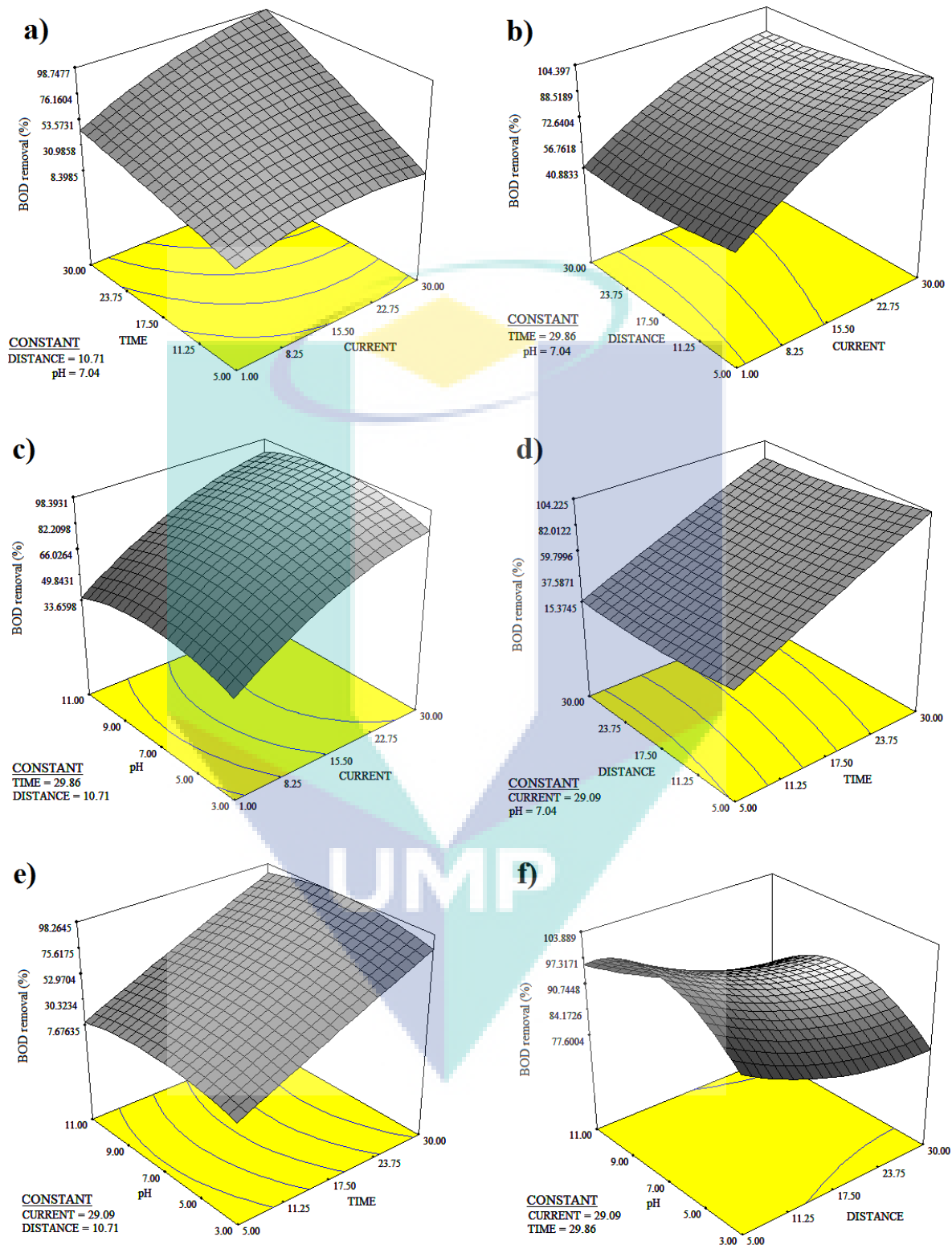


Figure 4.22: Optimization in 3D plot of BOD removal efficiency

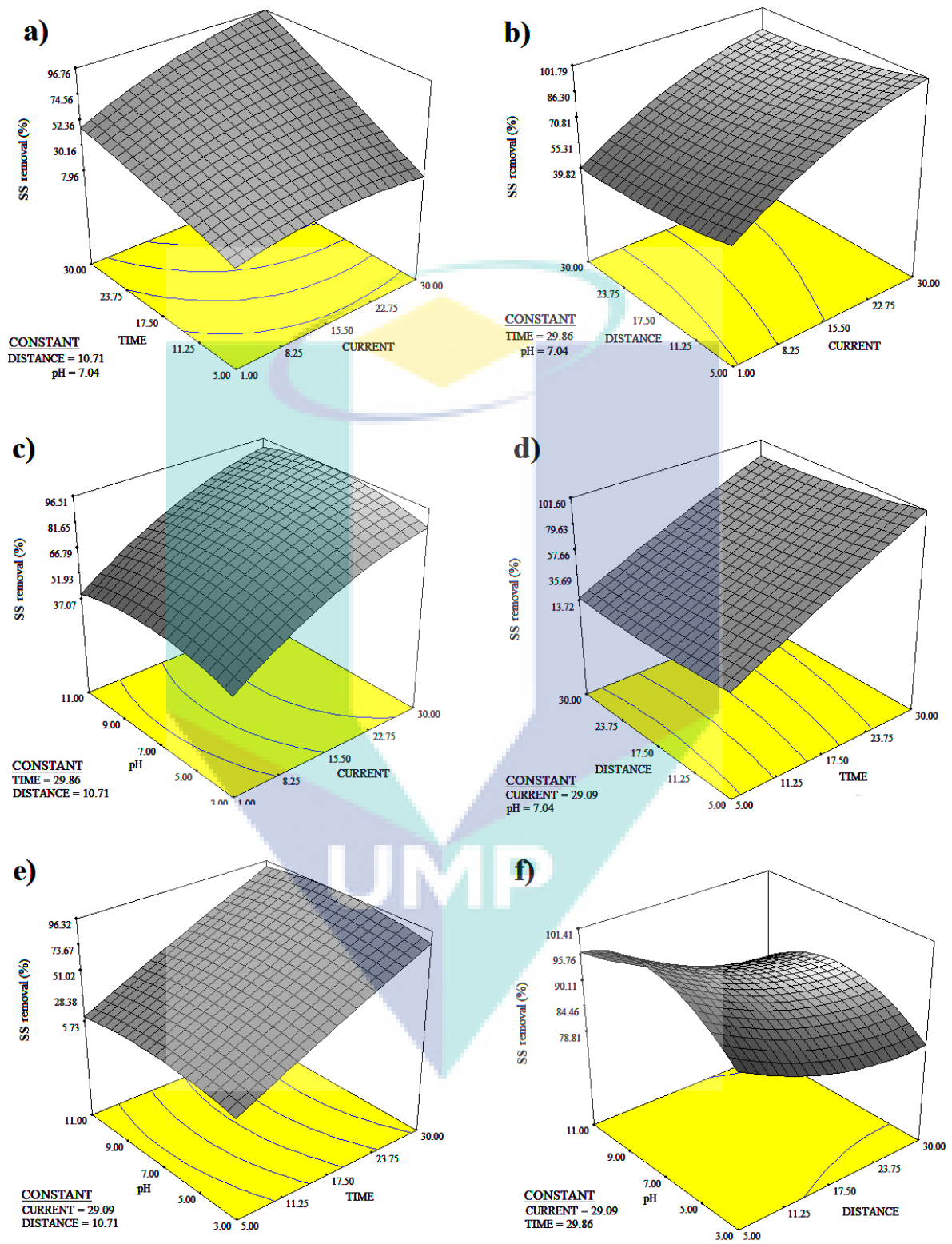


Figure 4.23: Optimization in 3D plot of SS removal efficiency

#### 4.8 HYDROGEN GAS PRODUCTION BY EC PROCESS

In this experiment, pH value and inter-electrode distance have been kept constant at the optimum treatment condition while the current was varied. The time given for each measurement was also kept constant at 60 sec. The table shows the volume of hydrogen gas produced in low and high current intensity application.

**Table 4.16:** Hydrogen gas production by different current applied.

Current (A)	H <sub>2</sub> production (ml)
1	6.40
2	14.00
3	21.80
20	144.10
25	181.40
30	210.70

Note: pH = 7.04, inter-electrode distance = 10.71 mm

By keep increasing the current, it leads to continuous hydrogen gas development. The theoretical volume of hydrogen can be calculated using Faraday's equation.

$$V_{\text{H}_2\text{theoretical}} = \frac{ItV_m}{nF} \quad (4.12)$$

The term I is the current used in mA, t is the time each measurement was taken and V<sub>m</sub> is the molar volume of hydrogen and considered to be equal to 24 L/mol at room temperature and normal pressure. The constant n is the number of electrons being exchanged in order to release one particle at the electrode and in this case is equal to 2. The Faraday's constant is equal to 96484 C/mol.

From the experimental result, it is possible to calculate the Faraday's efficiency of hydrogen production in every current application of the process. The Faradic efficiency can be calculated using equation (4.13).

$$\eta_{Faraday} = \frac{V_{H_2experimental}}{V_{H_2theoretical}} \times 100\% \quad (4.13)$$

Where:

$V_{H_2experimental}$  = volume of H<sub>2</sub> obtained from experiment

$V_{H_2theoretical}$  = volume of H<sub>2</sub> calculated using Faraday's equation

Both experimental and theoretical results of this thesis were interesting about the EC process. The numbers on the experimental sessions show that there is a difference with theoretical predictions. Table 4.17 shows the hydrogen production rate and Faradic efficiency of this experiment.

**Table 4.17:** Hydrogen production rate and Faradic efficiency

<b>Current (A)</b>	<b>Experimental H<sub>2</sub> production (ml)</b>	<b>H<sub>2</sub> rate (ml/min)</b>	<b>Theoretical H<sub>2</sub> (ml)</b>	<b><math>\eta_{Faraday}</math> (%)</b>
<b>1</b>	6.40	6.40	7.46	85.79
<b>2</b>	14.00	14.00	14.92	93.83
<b>3</b>	21.80	21.80	22.39	97.36
<b>20</b>	144.10	144.10	149.25	96.55
<b>25</b>	181.40	181.40	186.56	97.23
<b>30</b>	210.70	210.70	223.87	94.12

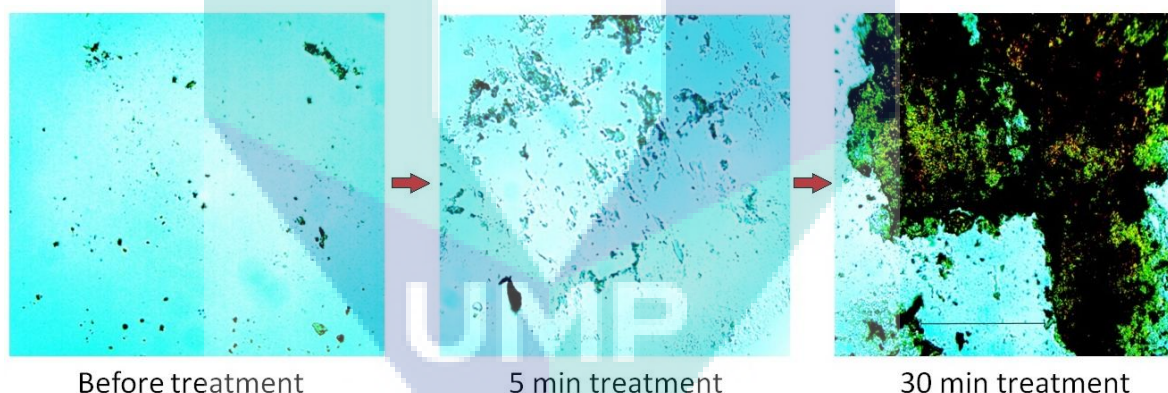
The Faradic efficiency of the EC cell/reactor expresses how much of the current is converted in the desired reaction. In order the EC cell/reactor to be efficient, Faradic efficiency must be close to 100 %. In case it is much smaller than one, it would mean that



there were several reason and reactions that were affecting the system such as corrosion. This would be a huge disadvantage, since not only it would shorten the service of the EC cell/reactor but also it would result in high-energy input, which is economically inefficient.

#### 4.9 LIGHT MICROSCOPY RESULTS

Particles in sewage water are expected to agglomerate and aggregate during EC process. The volume of coagulant added into the sewage water by EC process determines the agglomeration and aggregation to happen. Hence, the more time consumed and the more current applied will lead to the more pollutant removal. In order to provide image of sewage's particle, the light microscopy has been used.

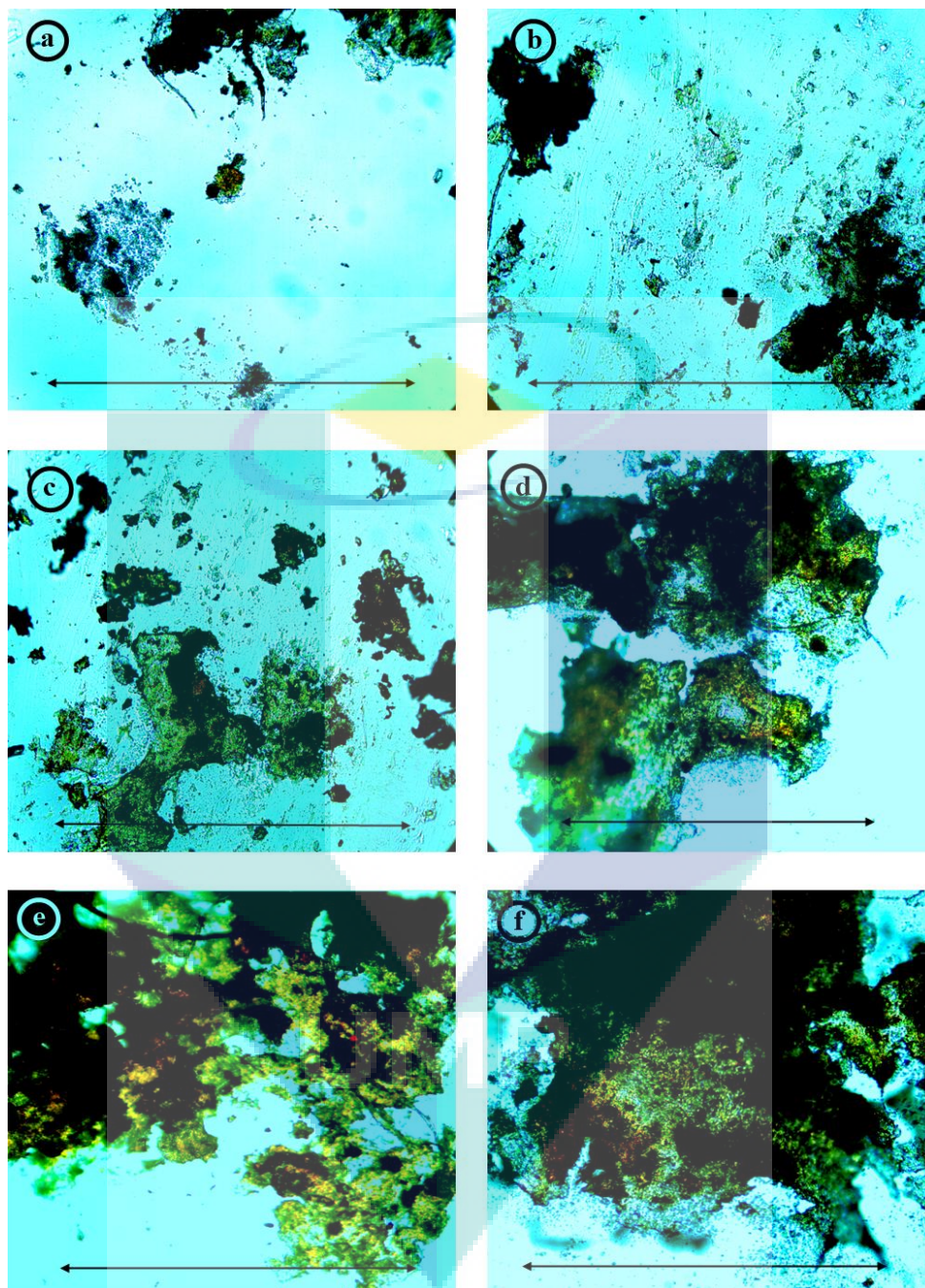


**Figure 4.24:** Microscopic images (20X) of sewage particles before treatment, 5 minutes and 30 minutes of treatment at 30 A.

A typical magnified photograph of the sewage particles is shown in figure 4.24. The sewage's particles were exists in various shapes and sizes. Before treatment, the numerous dots were observed in the background. That are the smaller pollutant particles and they were difficult to observe at this magnification. Overall the dots are majority and were disparate implying a stable pollutant at very lower sizes. The aggregate formed after 5

minutes of EC. It clearly can be seen that coagulant addition aggregated the sewage particles to form larger clusters. In fact, more dots appeared indicate that there were so many particles which were smaller than the dots that informed earlier. These smaller particles can be seen bonded together forming a dot and then the dot combine with another dot to form larger particles. The particles formed possessed an irregular shape and varied in sizes. Normally, the aggregates formed were elongated with irregular branching. The aggregates increased in size over time because by increase the reaction time, it also increased the coagulant addition. Larger aggregates were duly settled, as they were assuming to have a big size and also heavy. Besides, the large weightless particles can have interaction with the rising bubbles and thus will be removed by flotation. Hence either flotation or sedimentation, both process are the mechanism in pollutant removal. Larger aggregates are shown at 30 minutes of reaction time where the particle length is already more than 200 $\mu\text{m}$ .

The current applications also give the impact on particle's aggregates. Figure 4.25 shows the effect of current applied on the aggregates at 30 minutes of treatment. As can be seen, there were huge different between low current and high current applied. The size of aggregates formed in high current are approximately twice compared to low current applied. Larger aggregates would be expected to settle more readily, assuming shape and structure do not alter the aggregates' drag coefficient significantly. Alternatively, the aggregates' irregular shape and larger size (relative to primary clay particle) increased the probability of interaction with rising bubbles and thus removal by flotation. Hence aggregation resulted in pollutant removal by either sedimentation or flotation.



**Figure 4.25:** Microscopic images (20X) of sewage particles at 30 minutes at variety of current applied. (a) using 1 A, (b) using 2 A, (c) using 3 A, (d) using 20 A, (e) using 25A, (f) using 30 A.

#### 4.10 EC KINETIC

The pollutant is generally adsorbed at the surface of the flocs generated electrochemically. Critical analysis of the EC process of organic pollutants reveals that there are two separate processes taking place:

- Electrochemical process through which the metal flocs are generated
- Physio-chemical process through which the effluents are adsorbed on the surface of the flocs.

The removal of pollutant is similar to conventional adsorption except the generation of coagulants. During EC process the insoluble metal hydroxides removes pollutants by surface adsorption. In the adsorption it is assumed that the pollutant can act as a ligand to bind a hydrous Al in situ formed gelatinous precipitate.

$$\frac{-dC}{dt} = kCC_a \quad (4.14)$$

where,  $C_a$  refers adsorbent concentration in the system. Since the generation of aluminum hydroxide can be assumed constant for a given current density, the above equation can be simplified to pseudo first-order kinetics as stated by Balasubramanian et al. (2009):

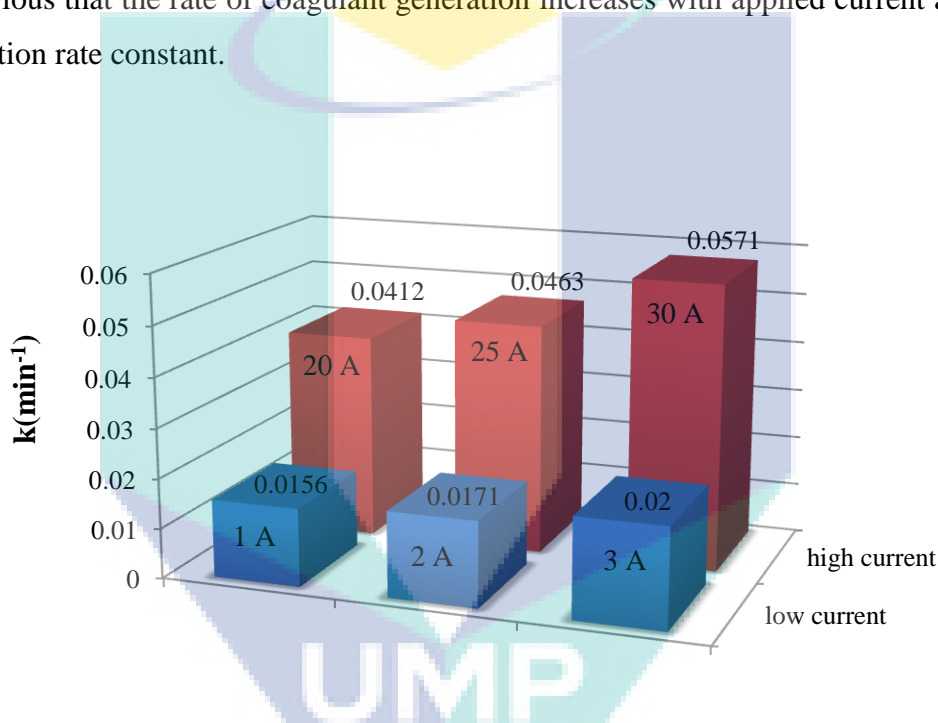
$$\frac{-dC}{dt} = kC \quad (4.15)$$

The integration of equation. (20) yields (Yadav, 2010):

$$\frac{C}{C_i} = \exp^{-k_1 t} \quad (4.16)$$

The reaction rate constant  $k_1$ , can be estimated from the plot  $\ln[C/C_i]$  versus electrolysis time. It has been observed from the present experimentation that the operating parameters have significant influence on the reaction rate constant.

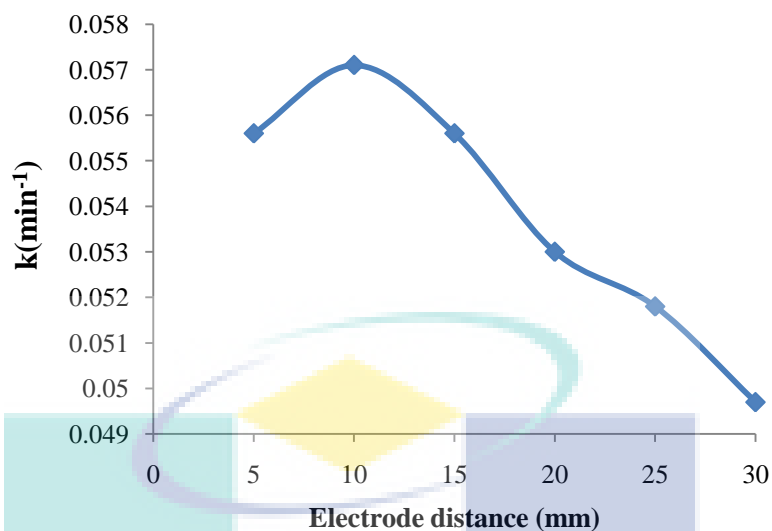
Figure 4.26 shows the influence of applied current density on reaction rate constant. It can be noticed that the reaction rate constant increases with an increase in the applied current density. There are much difference between low current and high current applied as it is obvious that the rate of coagulant generation increases with applied current and in turn the reaction rate constant.



Note: 30 minutes of treatment at pH 7

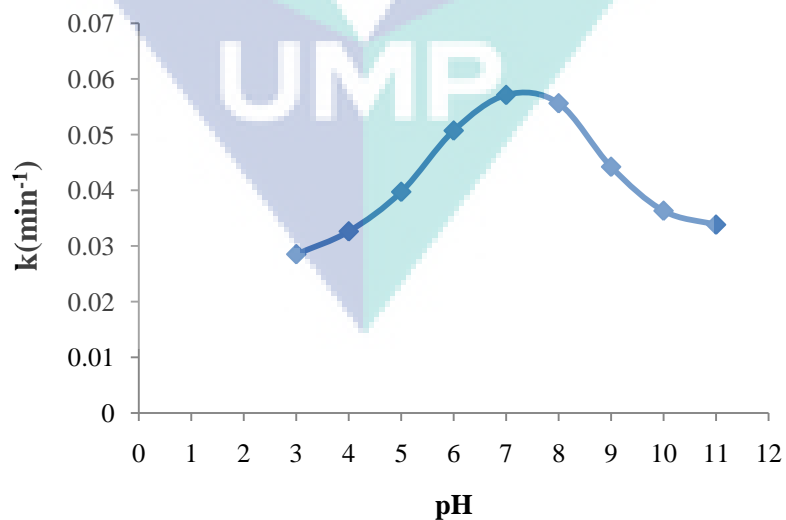
**Figure 4.26:** Effect of current on rate constant,  $k$

Figure 4.27 shows the influence of electrode distance on reaction rate constant. It can be assured that the rate constant decreases with an increase in the electrode distance. This could be due to the rising of Ohmic loss (IR drop) which eventually inhibits the rate of COD removal and in turn the reaction rate constant.



**Figure 4.27:** Effect of electrode distance on rate constant,  $k$

Figure 4.28 shows the influence of electrolyte pH on rate constant for aluminum anode. It can be ascertained that the rate constant is maximum at a pH value of 7. This is similar to the pH effect on percentage COD removal, the EC process is optimum at a pH value of 7, in turn maximum rate constant at a pH value of 7.



**Figure 4.28:** Effect of pH on rate constant,  $k$

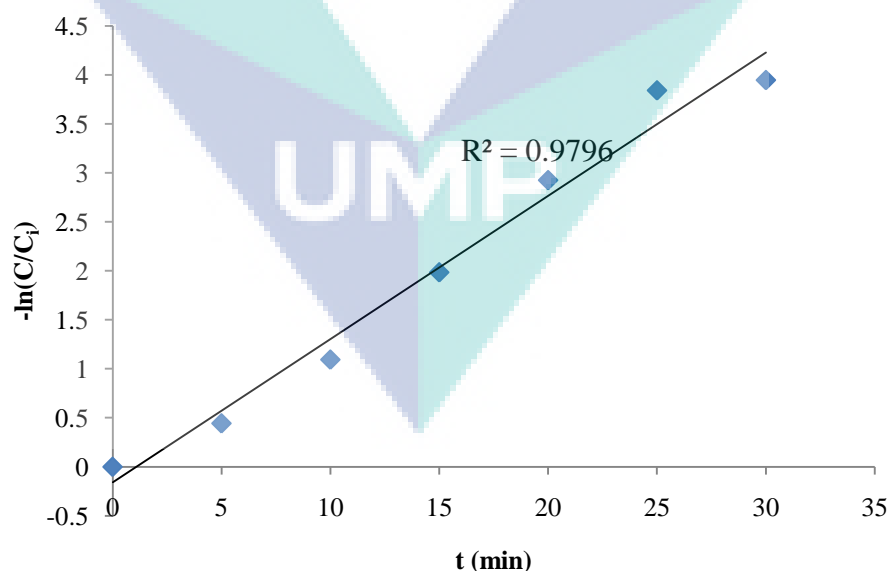
The pseudo second order kinetic equation can be given as

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \quad (4.17)$$

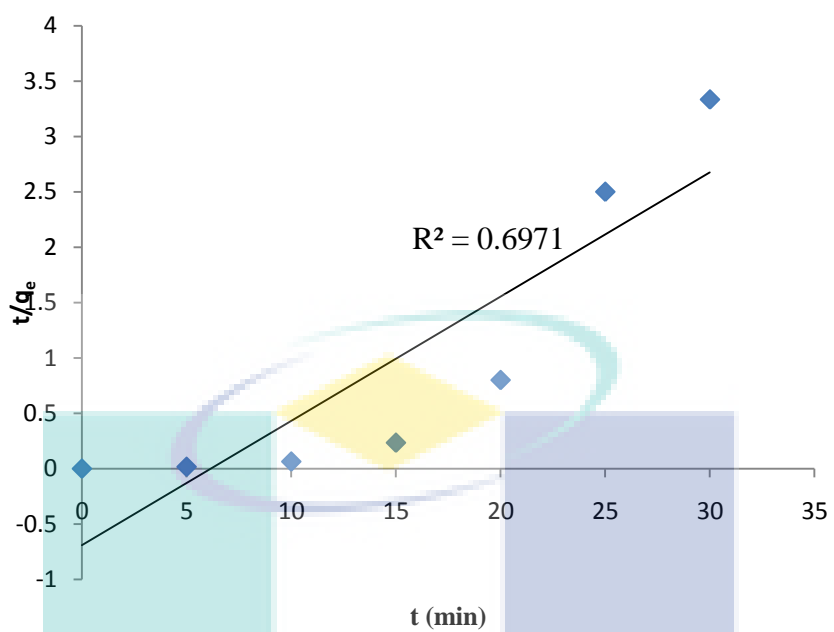
where  $k_2$  is the equilibrium rate constant of pseudo-second order adsorption, Where  $q_e$  and  $q_t$  refer to the amount of dye adsorbed at equilibrium and at any time,  $t$ . Integrating the equation (4.17) for the boundary condition  $t=0$  to  $t$  and  $q_t=0$  to  $q$ , gives,

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (4.18)$$

The intercept of plot  $t/q_t$  versus  $t$  was used to calculate the second-order rate constant  $k_2$ . The present experimental data have been verified with the above pseudo-first and pseudo second order and observations are presented in figure 4.29 and 4.30. It was found that the pseudo-first order fit with this experiment.



**Figure 4.29:** Pseudo-first order kinetic of EC, current 30 A; pH 7; Electrode distance 10 mm.



**Figure 4.30:** Pseudo-second order kinetic of EC, current 30 A; pH 7;  
Electrode distance 10 mm.

UMP



## CHAPTER 5

### CONCLUSION AND RECOMMENDATION

#### 5.1 INTRODUCTION

This chapter concludes the findings of all relevant sources in EC process to make sure that all the objectives were achieved.

#### 5.2 GENERAL CONCLUSION

Wastewater treatment by EC process is an environment-friendly process that requires no addition of chemicals, yields high quality effluent, requires short treatment times and simple operation. This study was made to treat sewage water by EC process using various operating parameters that are types of electrode, current, time, electrode distance and pH. Sewage sample classification is in medium strength as COD, BOD and SS is recorded as 466 mg/L, 259 mg/L and 297 mg/L respectively.

Electrochemical cell has successfully been made with stainless steel cuboids box of 2L. Monopolar metal plate electrodes (120 mm x 100 mm x 2 mm) of aluminum, iron and stainless steel were studied with distance between each pair of electrodes varied from 5 mm to 30 mm. The efficiency of electrode material was measured by the effectiveness of electrode in treating sewage water and the most effective was aluminum where it can

remove COD, BOD, and SS higher than 97% in 30 minutes of treatment. Removal of pollutant from sewage water is depending on the coagulant produced in-situ in EC process by the electrode. Increasing current and operating time resulted in increasing of coagulant and this fact was ascertained by Faraday's law. There are two groups of current applied in this experiment in order to determine their effect. The first was using low current (1 A, 2 A and 3 A) and the second was using high current (20 A, 25 A and 30 A). All treatments using high current in 30 minutes were satisfied the standard A of effluent discharge specified by The Environment Quality Act 1974 (Amendment 2009), in term of COD, BOD and SS. While, all the treatment using low current did not meet the regulation even for standard B except for SS removal which the value was 96 mg/L by using 3 A. So, the treatment using high current is more effective in treating sewage water for 30 minutes. As for the operating time parameter, the more time given, the more development in COD, BOD and SS removals were obtained and it applied on both types of current applied. There were not much changes on the removal was shown when the electrode was varied from 5 to 30 mm at 30 A and 30 minutes of treatment. The decreases on the COD, BOD and SS removal were occurred when inter-electrode distance increases from 10 to 30 mm and this must be due to the accretion of resistance and IR-drop effect. The decrease was also detected when electrode distance decrease from 10 to 5 mm and it must be due to electrostatic force effect. Although there were fewer changes in removal, the distance of electrode should be in the range of 5 to 15 mm to make sure that the EC process operates in optimum condition. pH is an important parameter in this study where the change of pH condition resulted in change of removal efficiency. The pH of sample was altered by using KOH and HCl into desired pH condition. In 30 minutes of operating time by using 30 A current applied, the best pH condition to treat sewage water was at neutral condition (pH 7). The first objective was accomplished where the effect of EC parameters on sewage characteristic have been determined.

The study using RSM modeling has been done to compare with the experimental without using RSM. There are 4 parameters assigned as variables to the statistical evaluation that are current, time, electrode distance and pH in order to analyze the correlation with the responses that are COD, BOD and SS. ANNOVA indicated the

response surface quadratic models for COD, BOD and SS removal to be satisfactory fitted with  $R^2$  of 0.9989, 0.9990, and 0.9989 respectively. The second objective was achieved where the optimization results for the maximum removal efficiencies of COD, BOD and SS as 97.84%, 97.77% and 95.72% respectively at 29.09 A of current, 29.86 minutes of operating time, 10.71 mm of inter-electrode distance and pH value of 7.04. This result fit with the experiment without using RSM where the optimum condition was obtained at 30 A, 30 minutes, 10 mm of electrode distance and pH value of 7.

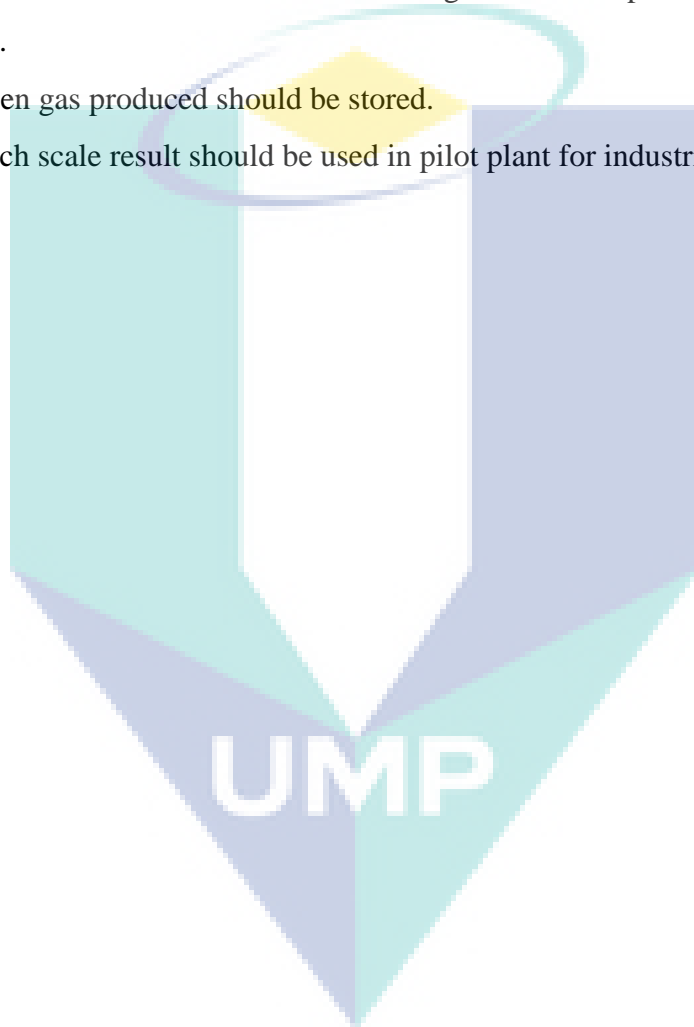
Production of hydrogen by EC process has been studied. In this paper, a technique of hydrogen recovery from an EC treatment of sewage waste is presented. The experimental hydrogen yields are compared with those calculated from theory. The yield of hydrogen increasing in increasing of applied current. Production of hydrogen gas obtained from experiment in 60 seconds at 1 A, 2 A, 3 A, 20 A, 25 A and 30 A were 6.40 ml, 14.00 ml, 21.80 ml, 144.10 ml, 181.40 ml and 210.70 ml respectively while the Faradic efficiency were 85.79%, 93.83%, 97.36%, 96.55%, 97.23% and 94.12% respectively. Therefore, the third objective in this study was achieved.

Light microscopy was used to provide images of sewage particles and aggregates. Clearly, coagulant addition aggregated the clay particles to form larger clusters. The result showed more clots occur if current and time increased because of more coagulant generated. The greater amount of coagulant (and its hydrolysed species) resulted in a greater number of reactive and binding sites in solution. The size of aggregates formed in high current are approximately twice compared to low current applied. The pollutant is generally adsorbed at the surface of the flocs generated electrochemically. In the adsorption it is assumed that the pollutant can act as a ligand to bind a hydrous Al in situ formed gelatinous precipitate. The experiment satisfied pseudo first order kinetic where the  $R^2$  was 0.9796.

### 5.3 RECOMMENDATION

EC process was successfully used as a treatment of sewage water. However, further work should take the following directions;

- Solar panel should be used as a power source to generate electrochemical cell.
- The cost effectiveness should be investigate and compared with other treatment method.
- Hydrogen gas produced should be stored.
- The batch scale result should be used in pilot plant for industrial application.



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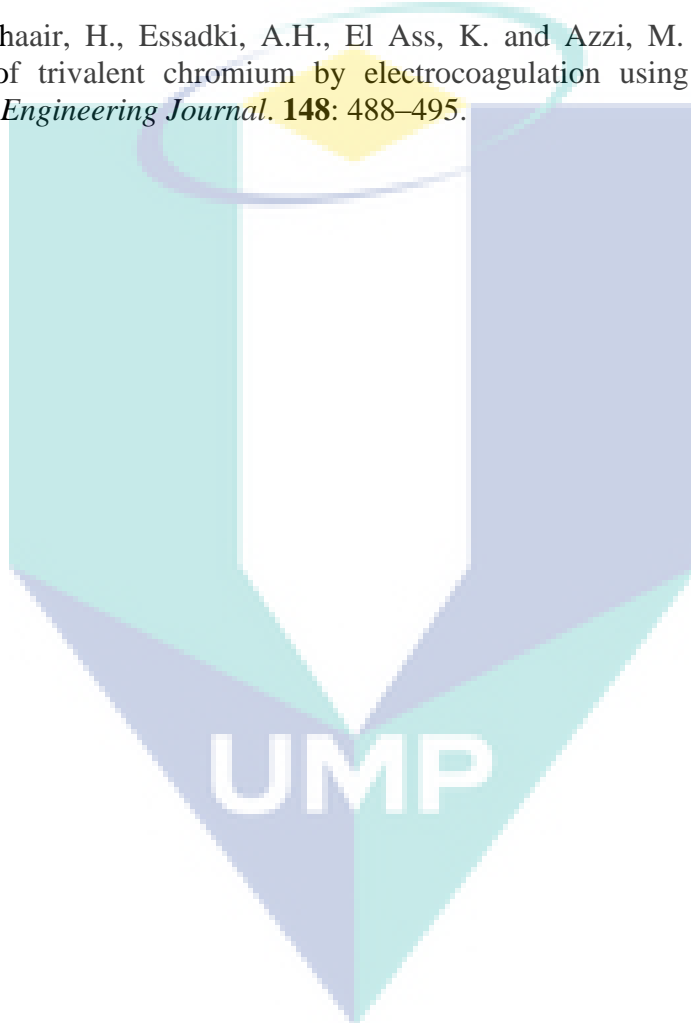
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## APPENDICES

## APPENDIX A1

**REMOVAL PERCENTAGE OF COD, BOD AND SS AT 1 A, 10 MM INTER-ELECTRODE DISTANCE, PH 7 BY USING ALUMINUM ELECTRODE**

Time (min)	COD			BOD			SS		
	Av.	±SD	±SE	Av.	±SD	±SE	Av.	±SD	±SE
0	0	0	0	0	0	0	0	0	0
5	3.86	0.45	0.26	4.63	0.51	0.29	3.03	0.39	0.23
10	12.66	0.39	0.23	10.81	0.52	0.30	8.42	0.88	0.51
15	24.03	0.49	0.28	18.15	0.31	0.18	14.48	0.46	0.27
20	45.06	0.57	0.33	40.93	0.57	0.33	34.34	0.48	0.28
25	59.87	0.36	0.21	54.44	0.43	0.25	47.47	0.53	0.31
30	65.88	0.66	0.38	61.78	0.62	0.36	58.25	0.72	0.42

(values in mean ± SD, n= 3)

## APPENDIX A2

**REMOVAL PERCENTAGE OF COD, BOD AND SS AT 2 A, 10 MM INTER-ELECTRODE DISTANCE, PH 7 BY USING ALUMINUM ELECTRODE**

Time (min)	COD			BOD			SS		
	Av.	±SD	±SE	Av.	±SD	±SE	Av.	±SD	±SE
0	0	0	0	0	0	0	0	0	0
5	7.51	0.44	0.25	6.18	0.56	0.32	5.72	0.45	0.26
10	17.60	0.63	0.36	13.51	0.71	0.41	10.10	0.51	0.29
15	31.12	0.54	0.31	18.92	0.36	0.21	20.54	0.46	0.27
20	50.64	0.59	0.34	41.31	0.93	0.54	37.37	0.44	0.25
25	63.95	0.79	0.46	57.53	0.46	0.27	51.85	0.58	0.33
30	69.31	0.55	0.32	64.86	0.51	0.29	62.63	0.61	0.35

(values in mean ± SD, n= 3)

## APPENDIX A3

**REMOVAL PERCENTAGE OF COD, BOD AND SS AT 3 A, 10 MM INTER-ELECTRODE DISTANCE, PH 7 BY USING ALUMINUM ELECTRODE**

Time (min)	COD			BOD			SS		
	Av.	±SD	±SE	Av.	±SD	±SE	Av.	±SD	±SE
0	0	0	0	0	0	0	0	0	0
5	8.80	0.45	0.26	10.81	0.31	0.18	8.42	0.49	0.28
10	20.82	0.34	0.20	23.55	0.56	0.32	13.80	0.78	0.45
15	34.33	0.51	0.29	35.52	0.71	0.41	24.92	0.59	0.34
20	53.22	0.53	0.31	51.74	0.33	0.19	45.12	0.62	0.36
25	66.52	0.98	0.57	61.39	0.52	0.30	58.92	0.42	0.24
30	74.89	0.44	0.25	70.66	0.48	0.28	67.68	0.35	0.20

(values in mean ± SD, n= 3)

UMP

## APPENDIX A4

**REMOVAL PERCENTAGE OF COD, BOD AND SS AT 20 A, 10 MM INTER-ELECTRODE DISTANCE, PH 7 BY USING ALUMINUM ELECTRODE**

Time (min)	COD			BOD			SS		
	Av.	±SD	±SE	Av.	±SD	±SE	Av.	±SD	±SE
0	0	0	0	0	0	0	0	0	0
5	14.81	0.44	0.25	12.74	0.35	0.20	17.51	0.39	0.23
10	37.98	0.37	0.21	33.59	0.42	0.24	30.98	0.47	0.27
15	66.09	0.61	0.35	64.09	0.46	0.27	54.88	0.38	0.22
20	82.19	0.55	0.32	77.61	0.69	0.40	70.37	0.54	0.31
25	87.98	0.65	0.38	86.10	0.48	0.28	80.13	0.56	0.32
30	94.21	0.57	0.33	93.05	0.53	0.31	90.24	0.56	0.32

(values in mean ± SD, n= 3)

UMP

## APPENDIX A5

**REMOVAL PERCENTAGE OF COD, BOD AND SS AT 25 A, 10 MM INTER-ELECTRODE DISTANCE, PH 7 BY USING ALUMINUM ELECTRODE**

Time (min)	COD			BOD			SS		
	Av.	±SD	±SE	Av.	±SD	±SE	Av.	±SD	±SE
0	0	0	0	0	0	0	0	0	0
5	30.47	0.46	0.27	15.44	0.39	0.23	21.89	0.32	0.18
10	59.23	0.61	0.35	49.03	0.45	0.26	44.44	0.43	0.25
15	80.90	0.54	0.31	75.29	0.64	0.37	66.67	0.41	0.24
20	87.77	0.63	0.36	86.10	0.61	0.35	78.11	0.57	0.33
25	93.13	0.33	0.19	92.28	0.55	0.32	87.54	0.45	0.26
30	95.92	0.41	0.24	95.37	0.65	0.38	93.27	0.52	0.30

(values in mean ± SD, n= 3)

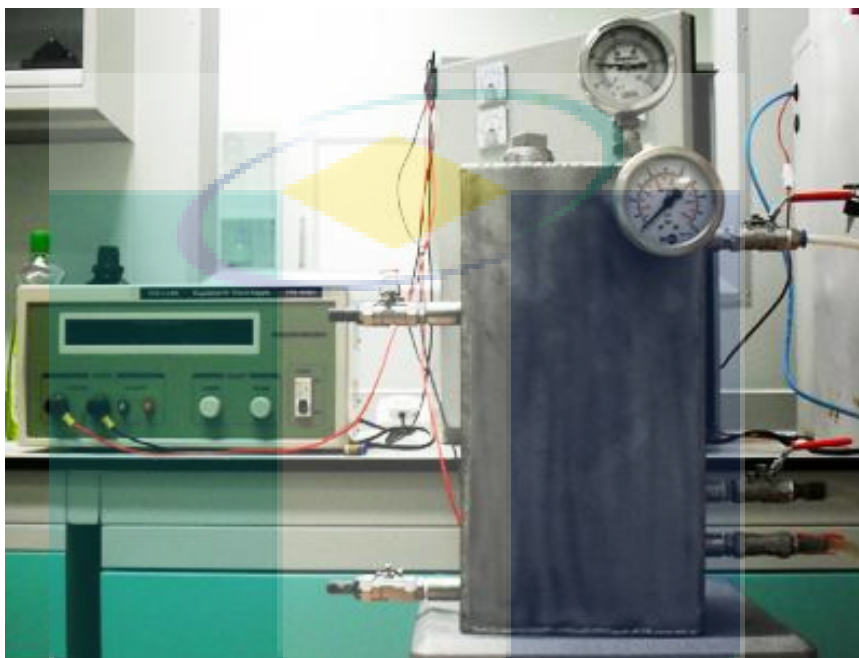


## APPENDIX A6

**REMOVAL PERCENTAGE OF COD, BOD AND SS AT 30 A, 10 MM INTER-ELECTRODE DISTANCE, PH 7 BY USING ALUMINUM ELECTRODE**

Time (min)	COD			BOD			SS		
	Av.	±SD	±SE	Av.	±SD	±SE	Av.	±SD	±SE
0	0	0	0	0	0	0	0	0	0
5	35.84	0.36	0.21	27.80	0.46	0.27	26.60	0.24	0.14
10	66.52	0.41	0.24	59.85	0.57	0.33	61.95	0.31	0.18
15	86.27	0.29	0.17	86.10	0.39	0.23	84.18	0.51	0.29
20	94.64	0.48	0.28	94.59	0.48	0.28	93.27	0.38	0.22
25	97.85	0.75	0.43	97.30	0.45	0.26	96.97	0.54	0.31
30	98.07	0.59	0.34	98.07	0.47	0.27	97.64	0.68	0.39

(values in mean ± SD, n= 3)

**APPENDIX B1****ELECTROCHEMICAL CELL MODEL****UMP**

**APPENDIX B2**

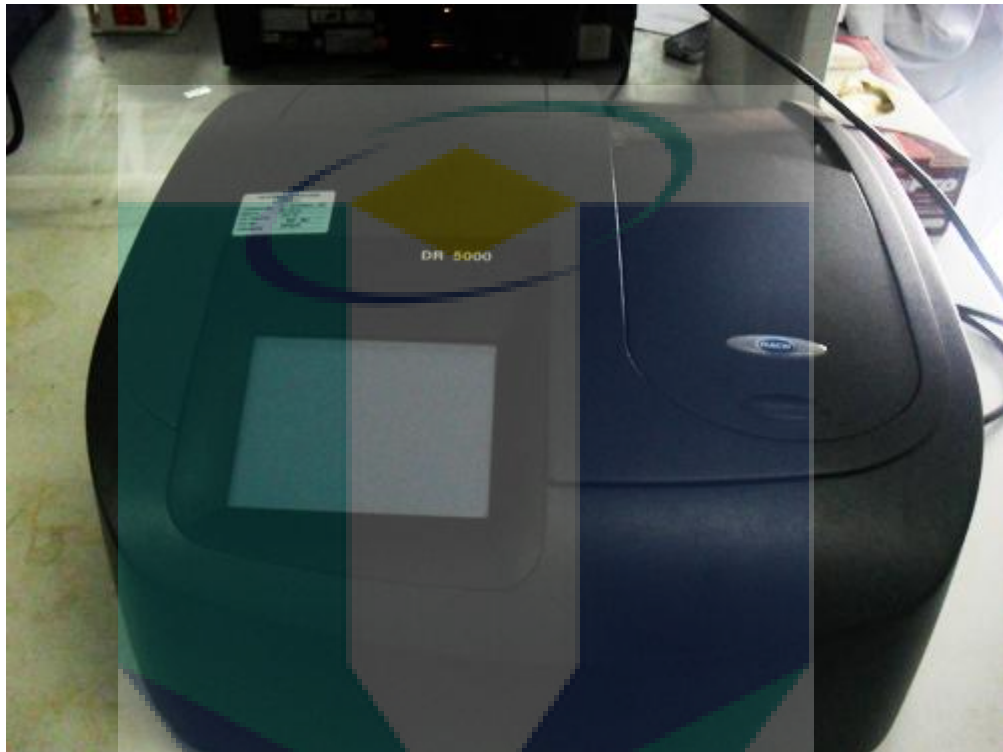
**INCUBATOR**



**UMP**

**APPENDIX B3**

**SPECTROPHOTOMETER DR 5000**



UMP

**APPENDIX C3**  
**LIST OF PUBLICATIONS**

Nasrullah Mohd, Mimi Sakinah, Abd. Syukor Abd Razak, Anwar Ahmad, Zahrizzan Zakaria, Zularisam Ab. Wahid\*, Ideris Zakaria. 2011. Hydrogen as the future sustainable Energy – A review. *International Journal of Civil Engineering and Geo-Environment*. **2**:47-57. ISSN: 21802742. Accepted.

Mohd Nasrullah, Lakverr Singh, Zularisam Ab. Wahid\*. 2012. Treatment of Sewage by Electrocoagulation and the Effect of High Current Density. *Energy and Environmental Journal*. **1**: 27-31. ISSN: 2227-3344. Accepted.

Mohd Nasrullah, Zularisam Ab. Wahid\*. 2012. Sewage Water Treatment by Electrocoagulation Process. *Energy and Environmental Journal*. Volume 2. Accepted.

Mohd Nasrullah, Lakveer Singh, Zularisam Ab. Wahid\*. 2012. Treatment Sewage Water using Electrocoagulation Process. *Journal of Industrial and Engineering Chemistry*. manuscript number JIEC-D-12-00845. ELSEVIER. Under Review.

Mohd Nasrullah, Lakveer Singh, Zularisam Ab. Wahid\*. 2012. Treatment Sewage Water by Electrocoagulation Process by Low current. *Asian Journal of Chemistry*. Under Review.