

**Republic of Iraq**  
**Ministry of Higher Education**  
**and scientific research**  
**University of Babylon**  
**College of Engineering**  
**Chemical Engineering Department**



# **Optimization the Bipolar Electrocoagulation Parameter for Reactive Blue dye Removal from Textile Wastewater**

A Thesis

Submitted to the College of Engineering University of Babylon in a  
Partial Fulfillment of the Requirement for the Degree of Master of  
Science in Engineering / Chemical Engineering

By

**Doaa Riyadh Hadi Kathum**

Supervised

By

**Asst. Prof. Sata Kathum Ajjam**

**Asst. Prof. Dr. Forat Yasir Sharrad**

2022

بِسْمِ اللَّهِ الرَّحْمَنِ الرَّحِيمِ

(يَرْفَعِ اللَّهُ الَّذِينَ آمَنُوا مِنْكُمْ وَالَّذِينَ أُوتُوا الْعِلْمَ دَرَجَاتٍ وَاللَّهُ  
بِمَا تَعْمَلُونَ خَبِيرٌ)

صدق الله العلي العظيم.

[11] المجادلة:



# DEDICATION

*To my parents for their kindness ...*

*To My affectionate mother...*

*To my brothers ...*

*To my friends and colleagues...*

*To all knowledge seekers...*

*Doaa*

## **Acknowledgements**

First, thanks to Allah, the compassionate the merciful, for giving me patience and strength to accomplish this research.

I would like to express my deepest thanks to my *supervisors Asst.Prof. Sata Kathum Ajjam and Asst.Prof.Dr.Forat Yasir Sharrad* for their guidance and helpful suggestions during my work to complete this thesis. The combined experience and insight offer them have greatly influenced and impacted this study.

Also I would like to express my thanks to the head of the Department of chemical Engineering *Asst.Prof. Dr. Hameed Hussein Alwan* for the continuous support throughout the preparation of this work.

## **Abstract:**

A huge amount of textile wastewater is discharged every year from many industries that must be treated. This work aims to assess the ability of an electrocoagulation removal of Reactive Blue dye (RBD) from synthetic textile wastewater utilizing plane and perforated aluminum electrodes connected to DC power apply in a bipolar connection. The kinetics of reaction and thermodynamic parameters have been studied.

The impact of power supplied mode was investigated and compared utilizing two modes; Constant Current Mode (CCM) (at 3 A) and Constant Voltage Mode (CVM) (at 20 volt) along the period of experiments (the electrolysis time: 2-70 min), and the pH was 8. The findings gained demonstrate that the removal efficiency of RBD utilizing CCM was higher compared to that gained in the case of CVM. After 70 min, the former technique provided 98.82% of RBD removing while the latter mode provided 97.74% of removing efficiency at the same conditions of initial RBD amount (100 ppm) and pH 8.

Despite the slight difference between the two findings gained, this finding proved that the CCM was relatively more efficient and cost-effective than the CVM. The order of the CCM was a first-order while it was a second-order for the other mode. Moreover, both of these processes are endothermic, consequently an investigation have conducted under the impact of the reaction time, voltage applied, and pH according to the ranges (2-80 min), (15-25 volt), and (4-12), respectively.

The experimental design and the analysis of findings gained have done utilizing response surfaces methodology (RSM) kind central composites design (CCD) and Minitab-statistics programme. The core findings revealed the treatability of the present configuration of electrodes to obtain higher removal efficiency of RBD. The highest RBD-removal was attained at the optimum values of the

operating variables which were 45.33 min, 18.03 volt, and 4 of the reaction time, voltage applied, and pH, respectively.

The mathematical models were significant according to the ANOVA test ( $p < 0.001$ ). This study proved the ability of the electrocoagulation technology to remove RBD from textile wastewater utilizing the present configuration of electrodes .

## CONTINUED

<b>Subject</b>		<b>Page</b>
Abstract		I- II
Table of contents		III- VI
List of figures		VII-X
List of tables		XI
List of abbreviations		XII-XIV
<b>Content</b>		<b>Page</b>
<b>Chapter One:Introduction</b>		
1.1	General	1
1.2	1.2. Objectives	3
<b>Chapter Two :Concepts and Literature Review</b>		
2	Introduction	5
2.1	Overview of Wastewater	5
2.2	Dyes	6
2.3	Fundamentals	8
2.4	Operation and design of electrocoagulation reactor	13
2.5	Factors affecting electrocoagulation	14
2.6	Characterization of electrocoagulation process	18
2.7	Advantages and disadvantages of EC	21
2.8	Literature review	22-33

<b>Content</b>		<b>Page</b>
<b>Chapter Three :Experimental work</b>		
3.1	Introduction	34
3.2	The Construction of the electrocoagulation unit	34
3.3	Synthetic wastewater preparation	35
3.4	Apparatus	36
3.5	Chemicals material	36
3.6	Set up of experimental work	37
3.7	Experimental procedure	38
3.8	Design of the preliminary experiment	40
3.9	Set up of Box-Willson design	41
3.10	Analytical methods	44
3.11	Estimate of removing efficiency	46
<b>Chapter Four :Results and Discussion</b>		
4.1	Introduction	47
4.2	The Preliminary experiment	47
4.2.1	Removing efficiency removing	47
4.2.2	FT-IR Analysis	50
4.2.3	Thermodynamic study	51
4.2.4	Kinetics studies	52
4.3	Best design of electrodes	54

<b>Content</b>		<b>Page</b>
4.4	Main statistical analysis of electrocoagulation responses	55
4.4.1	The Statistical analysis of dye RBD removing efficiency response	56
4.4.1.1	Impact of reaction time	56
4.4.1.2	Impact of the applied voltage	57
4.4.1.3	Impact of initial pH on the RBD removing	58
4.4.2	The statistical analysis of energy consumption response	59
4.4.3	The statistical analysis of electrodes consumption response	61
4.4.4	The statistical analysis of final pH response	64
4.4.5	The statistical analysis of the final conductivity response	66
4.4.6	The statistical analysis of the final stable current applied response	69
4.4.7	The statistical analysis of the final TDS response	71
4.5	Mathematical models	74
4.6	Optimization of the functional parameters	76

<b>Content</b>		<b>Page</b>
<b>Chapter Five :Conclusions and Recommendations</b>		
5.1	Conclusions	78
5.2	Recommendations of future work	79
	References	80-89
	Appendix A	90-92
	Appendix B	93
	الملخص	94

## LIST OF FIGURES

<b>Figure No.</b>	<b>Titel of Figure</b>	<b>Page</b>
2.1	Chemical structure of Reactive Blue	7
2.2	Mechanism of electrocoagulation (EC)	9
2.3	(a) Schematic of the various regions of the electrical double layer based on the BDM model, and (b) variation of the potential versus distance from the surface	12
2.4	Arrangement of electrodes in electrocoagulation cells	15
2.5	Aluminum hydrolysis species distribution as a function of pH	16
3.1	Electrodes geometry	34
3.2	The electrocoagulation reactor: (1) Digital DC power supply; (2) Magnetic stirrer; (3) Cathode; (4) Anode (bipolar electrodes in parallel connection).	35
3.3	In this investigation, we employed an electrocoagulation system and other equipment.	38
3.4	calibration curve of RBD	45
3.5	1800 UV- Spectrophotometer	45
4.1	Amount decay of RB-dye with time. (a) CVM-mode. (b) CCM-mode	48
4.2	MB removing efficiency with time. (a) CVM-mode. (b) CCM-mode	49
4.3	FT-IR analysis result of the sludge	51
4.4	Estimation of the heat of enthalpy depending on the slope magnitude (a) CVM. (b) CCM	52
4.5	Results of kinetic study for CVM system. (a) First order reaction. (b) Second order reaction	53
4.6	Findings of kinetic study for CCM system. (a) First order reaction. (b) Second order reaction	54

## LIST OF FIGURES

Figure No.	Titel of Figure	Page
4.7	Impact of reaction time on RBD removing efficiency of 100 mg RBD/L simulated wastewater (Applied voltage= 20 volt, pH 8)	56
4.8	The impact of voltage applied on the RBD removing efficiency of 100 mg RBD/L of simulated wastewater (reaction time= 41 min, and pH 8)	57
4.9	The impact of pH on the RBD removing efficiency of 100 mg RBD/L of simulated wastewater (reaction time= 41 min, and voltage applied=20 volt)	58
4.10	The impact of reaction time applied on the energy consumption of simulated wastewater (ph=8, voltage=20 volt ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	60
4.11	The impact of Ph applied on the energy consumption of simulated wastewater (reaction time= 41 min, voltage=20 volt ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	60
4.12	The impact of voltage applied on the energy consumption of simulated wastewater (reaction time= 41 min, pH 8 ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	61
4.13	The impact of reaction time applied on the electrode consumption of simulated wastewater (ph=8, voltage=20 volt ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	62
4.14	The impact of Ph applied on the electrode consumption of simulated wastewater (reaction time= 41 min, voltage=20 volt ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	63

## LIST OF FIGURES

<b>Figure No.</b>	<b>Titel of Figure</b>	<b>Page</b>
4.15	The impact of voltage applied on the electrode consumption of simulated wastewater (reaction time= 41 min, pH 8 ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	63
4.16	The impact of reaction time on the final Ph of simulated wastewater (voltage =20 volt, pH 8 ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	64
4.17	The impact of Ph on the final Ph of simulated wastewater (reaction time=41 min ,voltage =20 volt,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	65
4.18	The impact of voltage applied on the final Ph of simulated wastewater (reaction time= 41 min, pH 8 ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	65
4.19	The impact of reaction time on the final conductivity of simulated wastewater (voltage =20 volt, pH 8 ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	67
4.20	The impact of Ph on the final conductivity of simulated wastewater (reaction time=41 min, voltage =20 volt,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	67
4.21	The impact of voltage on the final conductivity of simulated wastewater (reaction time=41 min, Ph =8 ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	68

<b>Figure No.</b>	<b>Titel of Figure</b>	<b>Page</b>
4.22	The impact of reaction time on the current applied of simulated wastewater (Ph =8 ,voltage =20 volt, Nacl=2g and 200 rpm) for 100 mg RBD/L removal	69
4.23	The impact of Ph on the current applied of simulated wastewater (reaction time =41 min ,voltage =20 volt, Nacl=2g and 200 rpm) for 100 mg RBD/L removal	70
4.24	The impact of voltage on the current applied of simulated wastewater (reaction time=41 min, Ph =8 , Nacl=2g and 200 rpm) for 100 mg RBD/L removal	70
4.25	The impact of reaction time on the final TDS of simulated wastewater (Ph =8 , voltage =20 volt, Nacl=2g and 200 rpm) for 100 mg RBD/L removal	72
4.26	The impact of Ph on the final TDS of simulated wastewater (reaction time=41 min, voltage=20 volt, Nacl=2g and 200 rpm) for 100 mg RBD/L removal	72
4.27	The impact of voltage on the final TDS of simulated wastewater (reaction time=41 min, Ph=8 ,Nacl=2g and 200 rpm) for 100 mg RBD/L removal	73
4.28	demonstrates the optimal settings of the functional parameters and responses for the remediation of simulated wastewater containing 100 mg RBD/L	75

## LIST OF TABLES

<b>Table No.</b>	<b>Title</b>	<b>Page</b>
2.1	Properties of Reactive Blue dye (RB)	8
3.1	Aluminum electrodes chemical.composition expressed.as.a percentage-of total weight	35
3.2	Functional.parameters	39
3.3	Functional parameters for Preliminary Experiment	41
3.4	The range of design for parameters	42
3.5	Set up the coded and the real parameters	42
3.6	Experimental design.condition.of mintab design with 3.factors	43
4.1	Findings of both experiments done for RBD removing efficiency	48
4.2	Findings of CVM and CCM modes for RBD removing	50
4.3	Best design of electrodes	55
4.4	Mathematical models	74-75

## List of Abbreviations

Symbol	Meaning	Unit
b	Energy of adsorptions	l/mg
Bi	Regression coefficient	----
Bij	Regression coefficient	----
Bo	Regression coefficient	----
C	Pollutant concentration	mol/m <sup>3</sup>
C <sub>e</sub>	Equilibrium concentration of adsorbate	mg/l
C <sub>i</sub>	Initial concentration of adsorbate	mg/l
C <sub>o</sub>	Initial concentration of dye	mol/m <sup>3</sup>
C <sub>t</sub>	concentration of dye at time (t)	mol/m <sup>3</sup>
CCM	Constant Current Mode	----
CVM	Constant Voltage Mode	----
EC	Electrocoagulation	----
EF	Electro-fenton	----
Eox	electrosorption, Electrooxidation	----
F	Faraday's constant	columb/mol
I	Current supplied	Ampere
k	Rate constant	----
K <sub>1</sub>	Reaction rate constant of first order	1/min
K <sub>2</sub>	Reaction rate constant of second order	[(mol/m <sup>3</sup> min)]-1
K <sub>d</sub>	Equilibrium constant	l <sup>2</sup> /g <sup>2</sup>
M	Molecular weight	g/mol
D	Distance between the electrode	cm
RTD	Resident time distribution	----
SSV	Settling sludge velocity	----
PC	Perox-coagulation	----

Symbol	Meaning	Unit
mA	Atomic weight of lead	g/mol
N	Order of reaction	----
n	Freundlich constant	----
ppm	Part per million	gm/m
$q_{max}$	Max. adsorptions capacity	mg/l
m	Mass of the adsorbent	g
R	Gas constant	J/K.mol
$R^2$	Regression coefficient	----
R	Correlation coefficient	----
$R_l$	Separation factor or equilibrium parameter	----
t	Electrolysis time	h or min.
U	Voltage applied	volt
V	Volume of the simulated wastewater	$m^3$ or liter
W	Weight of lead nitrate	g
$X_1$	Time	min.
$X_2$	Amount of lead	ppm
$X_3$	pH	----
Z	Number of electrons involved in the reaction	----
$\Delta G$	Gibbs energy	J/mol
$\Delta H$	Basic heat adsorptions	J/mol
$\Delta S$	Change in entropy	J/mol
Y	Removal efficiency response	
COD	Chemical oxygen demand	----
RSM	Response surface methodology	----
$q_a$	Absorption of light at aspecific wavelength	----
$q_e$	Quantity adsorbated	mg/g
Ao	Anodic oxidation	----

Symbol	Meaning	Unit
$\varepsilon$	Random error	----
RBD	Reactive blue dye	----
RB5	Black 5dye	----
BB3	Basic blue 3	----
BR46	Basic red 46	----
VBP	Banana peel	----
RG-19	Reactive green dye 19	----
MB	Methylene blue	----
MO	Methyl orange	----

## CHAPTER ONE

### INTRODUCTION

#### 1. General

Many sources of wastewater cause the pollution to the aquatic systems and soil which may have led to the crisis of clean water demand around the world (*Patel, et al, 2010*). Wastewater is discharged annually from domestic and many activities of factories such as textile plants, petroleum refineries, and petrochemical plants. Dyes are one of the components of paints, textile, printing inks, paper, and plastic industries (*Teng, et al, 2020*). The effluent from textile plants contains many pollutants such as dyes, organic compounds, and heavy metals (*AlJaberi (a), 2018*). Dyes are complicated pollutants presented in textile wastewater since a huge amount of dyes are produced and consumed every year (*Borhade, 2018*). More than one million commercial dyes exist, and 10,000 dyes with over  $7 \times 10^5$  metric tons produced annually and available for commercial use; 30% of these dyes' production exceeds 1000 tonnes/year, and 90 percent of textile goods have been utilized at the level of 100 tonnes/year. Colored and toxic wastewater discharged into the ecosystem goes through chemical and biological transformations, consuming dissolved oxygen (*Anantha Singh and Ramesh, 2013*).

The contaminants presented in the effluents of textile plants undergo chemical and biological changes which leads to consume dissolved oxygen and impacts aquatic life since some types of dyes when degraded produce carcinogens and toxic materials. Thereby, it is essential to eliminate dyes from wastewater before discharging to the aquatic system to meet ecological regulations (*Golder, et al, 2005*). The presence of dyes in water tends to prevent the light to passage through water, consequently, minimizing the efficiency of

photosynthesis in aquatic plants and impact their growth ( *Othmana,et al,2018*) It is important to remove dyes from water since they are a source of contamination. There are several methods for removing such as physical precipitation, chemical oxidation or reduction, filtration, biological and their combinations.

But these methods have disadvantages, including chemical methods in which a lot of chemicals are utilized that cause pollution as well as in terms of economic aspects (*Borhade,2018;Golder,2005*). As for biological methods in which living organisms are used, they cannot be applied to textile factories water since dyes are considered toxic to living organisms, which leads to an increase in the formation of thrombus and lack of efficiency of the process (*Othmana,et al,2018; AlJaberi (a) ,2020*).

Chemical coagulation is utilized to treat dye wastewater. However, pollution may be caused by the high amounts of chemicals used. Electrochemical wastewater remediation technologies received much attention in recent years. These technologies include electrodeposition, electro-Fenton , Electrooxidation (EOX) and electrocoagulation (EC) and their successive and concurrent combinations for the remediation of industrial wastewaters (*Özyurt and Camcioğlu ,2018*).

Excessive coagulant material can be avoided by EC utilizing electricity to treat wastewater. It was first proposed in the United Kingdom in 1889. Elmore as the first to suggest the application of electrolysis in mineral extraction in 1904. Dietrich first patented the principle of EC as a technique to treat bilge water from ships in 1906. The EC process is an ecofriendly, effective, speedy, and economic remediation for the removal of various toxicants, such as dissolved metals, tannin, phenol, and dyes, from water and wastewater (*Anantha Singh and Ramesh,2013*).

Electrocoagulation (EC) is an uncomplicated and effective method that treats many types of wastewater and has gained great success in removing most of the pollutants, (*AlJaberi (b) ,2018;Helder,2015*). The mechanism of EC is based on the release of numerous ions from anode and cathode by the process of dissolution. Then, by interacting to form electro-coagulants, i.e.  $\text{Al(OH)}_3$  in the case of aluminum electrodes, pollutants are removed by adsorption (*AlJaberi (b) ,2020*). Usually aluminum or iron anode and cathode are utilized and this method prevents the use of chemical additives (*Othmana,et al,2018; Changmai, et al ,2019*).

For the last three decades, EC was applied as a successful water-remediation technology to remove a wide range of pollutants. Renewed interest in EC as a produced water remediation technology is largely since the technological improvements which led to lower electricity use and cleaner output. (*Esmailirad et al., 2015*).

### **1.1. Objectives**

This work aims to remove Reactive Blue Dye (RBD) from simulated wastewater utilizing a batch electrocoagulation reactor involves new configuration of electrodes which were connected to a power source in a bipolar-parallel mode. The electrodes utilized in this work is made of aluminum and have been configured to be the interior two as perforated-plates while the outer two plates as not perforated. The experiments and findings were done utilizing response surfaces methodology (RSM) type central composites design (CCD).

The investigation also attempts at examining the impact of the operating variable Preliminary Experiment , electrolysis time, electric voltage applied, pH, salt amount, and stirring speed on the EC process. This work also aims to study the following responses: efficiency of RBD removal , consumption of energy

and electrodes, pH, electrical conductivity, and the applied steady current are all factors to be considered.

As for the initial experiment that was conducted before starting the design experiments, the goal is:

1. To investigate the treatability of an electrocoagulation reactor to remove (RBD) from simulated wastewater utilizing new configuration of electrodes containing plane and perforated aluminum electrodes connected in a bipolar-parallel connection, and compare the performance of this reactor between two modes which are the Constant Modes of Current and Voltage (CCM and CVM), respectively. The kinetics of reaction and thermodynamic parameters have been studied.
2. Find out how long the process will take to get the information needed to RSM design method.
3. To investigate the variation of the inside temperature of the simulated solution over time.

## Chapter two

### Concepts and Literature Review

#### 2.1. Overview of Wastewater

Wastewater is the water that has been polluted for various reasons by households, business enterprises, businesses, public institutions, textile plants, petroleum refineries and other similar entities. Although sewage is a more generic word that refers to any dirty water (including wastewater) that may contain organic and inorganic compounds, industrially wastes, groundwater that infiltrates and mixes with the contaminated water, storm runoff, and other similar liquids (*Arcadio and Gregoria,2002*).

Raw water in large volumes for a variety of applications is consumed by industrially operations. Wastewater is made up of a complicated mix of various substances. There are chemical and inorganic substances that are potentially hazardous and difficult to breakdown (*Özyurt and Camcioğlu, 2018*).

Dye Wastewater includes many unused dyes that are discharged by textile and paper factories and paper industries. The presence of these dyes in the water causes pollution to the water environment, colors water and increases the demand for chemical oxygen (COD) and this leads to toxicity and bad odor (*Islam, etal ,2011; AlJaberi (c),2018*).

These contaminants can be found in three various states: dissolved, colloidal, and suspended form (*Özyurt and Camcioğlu, 2018*). Conventional wastewater remediation techniques are utilized to treat the majority of wastewater (*Anantha Singh and Ramesh,2013*). There are several methods for removing pollutants such as physical precipitation, chemical oxidation or reduction, filtration, biological and their combinations. Physical processes, such as screening, flotation, filtration, and sedimentation (*Getaye,et al,2017*), chemical processes, such as coagulation/flocculation, chlorination, adsorption, and ion

exchange, and biological processes, such as activated sludge, aerated lagoons, and membrane bioreactors are all examples of conventional wastewater remediation technologies (*Özyurt and Camcioğlu, 2018*).

When metal hydroxides are being utilized as a coagulant, the adsorption of hydroxide on mineral surfaces is 100 times greater in 'situ' than on pre-precipitated hydroxides, making electrocoagulation (EC) as a simple and efficient method for the remediation of several kinds of wastewaters, including textile wastewater (*Mollah et al., 2004; Khandegar and Anil, 2013; Adhoum et al., 2004*).

## **2.2. Dyes**

The chemical structure of dyes can be utilized to classify them: application technique; a certain chemical class is frequently used; dyes from a variety of application types, as well as a specific type of dye. Dyes of many chemical kinds may fall under this category (*William, et al, 1971; Waring, and Hallas, 2013*).

1. Azoic dyes.
2. Cationic and related dyes.
3. Anthraquinone dyes.
4. Vat dyes.
5. Sulphur dyes.
6. Miscellaneous dyes.
7. Polycondensation and related dyes.
8. Phthalocyanine pigments and dyes.
9. Pigments other than phthalocyanines.
10. Solvent dyes.
11. Oxidation bases.

12. Retention of dyes in the fiber.
13. Fluorescent brighteners.
14. Color photography
15. Reactive dyes

One of the most significant breakthroughs in the field of synthetic coloring has been the development of reactive dyes for cellulosic fibers. With far-reaching implications since of the increased usage of these dyes, the demand for various types of dyes has dropped. To make these colors, cyanuric chloride is combined with two or three amino-substituted dye molecules. Chloro- and dichlorotriazinyl dyes for reactive ranges are made utilizing similar condensations. However, only two stages are required as one reactive chlorine atom must remain in the end product. An example of this type is the reactive blue dye utilized in this research (*Hunger, 2007*).

Reactive blue dye (RBD) is a water-soluble anionic dye and can cause very harmful impacts on humans and other negative influences on the environment. Therefore it is very essential to minimize its amount in wastewater before discharging to the aquatic systems and soil (*El Alouani, et al ,2018; Munagapati, et al,2010; Alardhi , et al ,2020 ;Al-Barakat, et al ,2020 ;Jafat,Ajam 2016*) . The chemical structure and properties of (RBD) are demonstrated in Fig. (2.1) and Table (2.1).

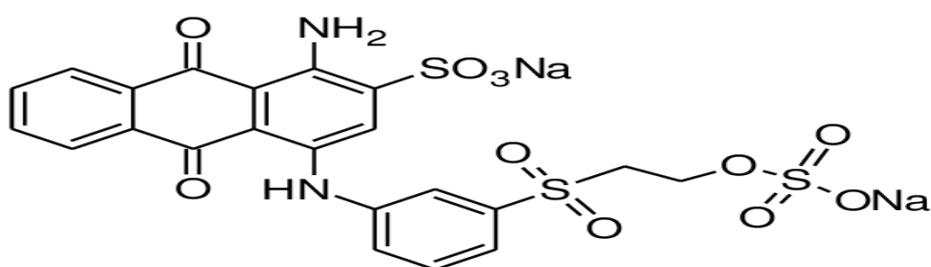


Figure (2.1). Chemical structure of Reactive Blue (*Hunger, 2007*).

Table (2.1). Properties of Reactive Blue dye (RB).

Chemical form	$\lambda$ max	Molar mass (g/mol)
$C_{22}H_{16}N_2Na_2O_{11}S_3$	585	626.50

### 2.3. Fundamentals:

#### 2.3.1. Principles of Electrocoagulation

EC theory synthesis of coagulant in a combination throughout the reactor distinguishes this approach. The chemically processing, which happen in the cell seem to be the reduction and oxidation processes that actually occur at the electrode/electrolytes contact. The anodes seems to be the electrode in which oxidation occurs, whereas the cathodes seem to be the electrode where reduction occurs. The anode, commonly called as the sacrifice electrodes, corrode to generate active coagulant cations, usually aluminum or iron, into the solutions. As a consequence, rather than environmental dosing, electro-coagulants inserts metallic ions in site (*Adelaide,2013*).

The EC process develops in three stages: (A) the oxidation process and the formation of coagulants, (B) fraction emulsions and instability of pollutants, and (C) combining pollutants to form masses that Khandegar float or precipitate according to the type of pollutants (*Khandegar and Anil,2013*) , as Fig (2.2) illustrates this .

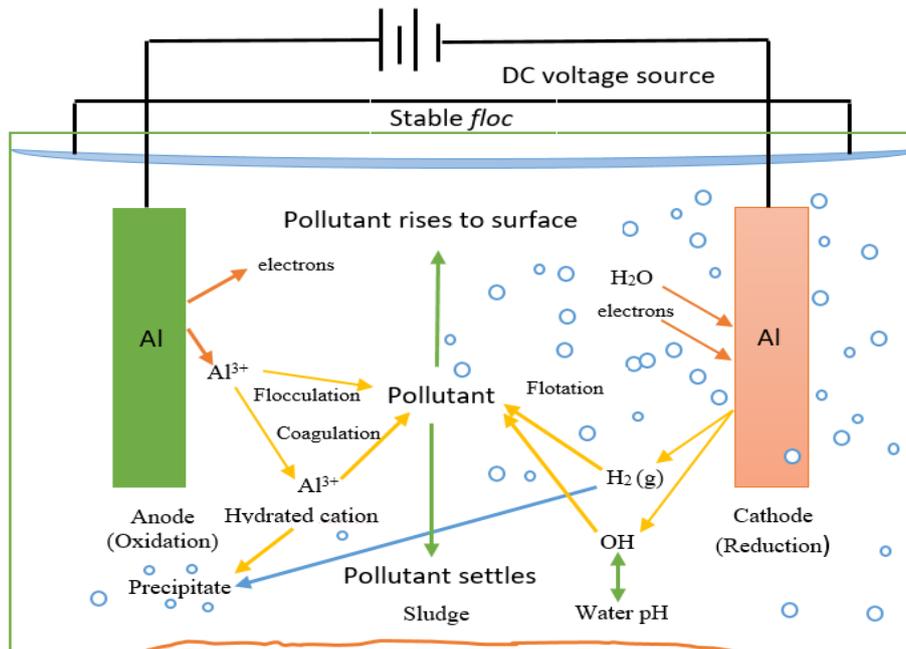


Figure (2.2): Mechanism of electrocoagulation (EC) (*Holt ,et al,2002*).

Electrocoagulation process does not need the addition reagents or chemicals. So, it is very economical, does not form secondary pollutants, and its efficiency is greater than other conventional methods. It is also easy to designing, can be tuned for any capacity of processing plant, easy to operate and requires the formed which is less and easy to be removed (*Getaye, et al ,2017; Al-Barakat,et al, 2020*).

The electrochemical cell is made up of a reactor with two electrodes, the anodes electrodes and the cathode electrodes, both of which contain the electrolyte solution to CD power supply ( Voltage and current) (*Abd UL-Shaheed,2018*). The mechanism of EC is based on the dissolution process releasing a large number of ions from the anodes and cathode, which then interact to produce electro-coagulants, such as  $Al(OH)_3$  in the case of aluminum electrodes, which subsequently adsorb pollutants ( *Anantha Singh and Ramesh,2013*). Usually aluminum or iron as electrodes are used. This method prevents the use of chemical additives (*Getaye,et al,2017; Bazrafshan, et al, 2012*).

Coagulation is caused by metallic cations produced by anodes hydrolysis, which create compounds of polyhydroxymetallic, polyhydroxides, and hydroxides with a significant attraction for ions counter and dispersed grains.

### 2.3.2 Main reactions throughout the EC

The electrodes were usually in the form of plates which are either of iron or aluminum. The type of metal utilized in the electrodes affects the type of coagulant that directly affects the removal efficiency. When starting the electrode coagulation process and applying the required voltage or current to a cell, a certain process of the anode and the reduction process of the cathode are occurred. The pH increases when hydrogen gas is released during the reduction process Eq. (2.1) and Eq. (2.2) (*AlJaberi (d), 2018; Essadki,2012*).



An investigation of the chemical reactions occurring in the electro-coagulation process demonstrates that the main reactions occurring at the aluminum electrode are (*Bazrafshan,2012*):



Monomeric kinds, including  $Al(OH)_4^{+2}$ ,  $Al_2(OH)_2^{+4}$ ,  $Al(OH)_2^{+}$ , and  $Al(OH)^{+2}$ , as well as polymeric kinds, including  $Al_{13}(OH)_{34}^{+5}$ ,  $Al_{13}O_4(OH)_{24}^{+7}$ ,  $Al_8(OH)_{20}^{+4}$ ,  $Al_7(OH)_{17}^{+4}$ , and  $Al_6(OH)_{15}^{+3}$  have been created throughout the electrocoagulation process (*Canizares,et al, 2005 ; Can ,et al,2003*). The main function of Aluminum hydroxide is to act as a coagulant, to coagulate contaminants present in the liquid surrounding media, thus removing them (*Bazrafshan, et al ,2012*).

The amount of dissolved aluminum ions from the anodes is theoretically estimated by Faraday's law of electrolysis:

$$W=I t M/(ZF) \quad (2.6)$$

where  $w$  is the dissolved anodes mass ( $\text{g}/\text{cm}^2$ ),  $I$  is the current ( $\text{A}/\text{cm}^2$ ),  $t$  is time (s),  $M$  is the anodes material's molecular weight,  $Z$  is the valence of dissolution in the oxidation/reduction reaction, and  $F$  is Faraday's constant ( $96,487 \text{ C}/\text{eq}$ ). According to Eq. (2.6), the anodic dissolving rate increases as the current raises, and the resultant ion hydroxides form more flocs, improving the coagulation process. In addition, the rate of bubble production raises, separating the contaminants by float (*Pajootan, et al,2012; Anantha Singh and Ramesh,2013*).

### **2.3.3. Coagulation**

It is the most important step in the process of destabilizing the pollutant by destabilizing the pollutant charge, and this leads to clumping of suspended grains forming (*Benefield, et al,1982*). Reducing the energy barrier leads to the accumulation of pollutant grains, determining the repulsive forces of the double layer and the gravitational attraction of the vander Waals. Surface charge is the advantage of grains in water resulting from chemical reactions or the ions' adsorptions on the grain's surface, symmetric substitution, disliquid surrounding media of ions. A double electrical layer is formed resulting from the charged grain's surface and the opposite charge from the anticron ions which were first described by Helmholtz in 1879 (*Eilbeck and Mattock ,1987*). The generally recognized model, which is depending on the model's Bockris, Devanathan, and Muller (BDM), is depicted in Fig (2.3) (a) and (b), which demonstrates the corresponding fluctuation of the distances versus potential from the surfaces.

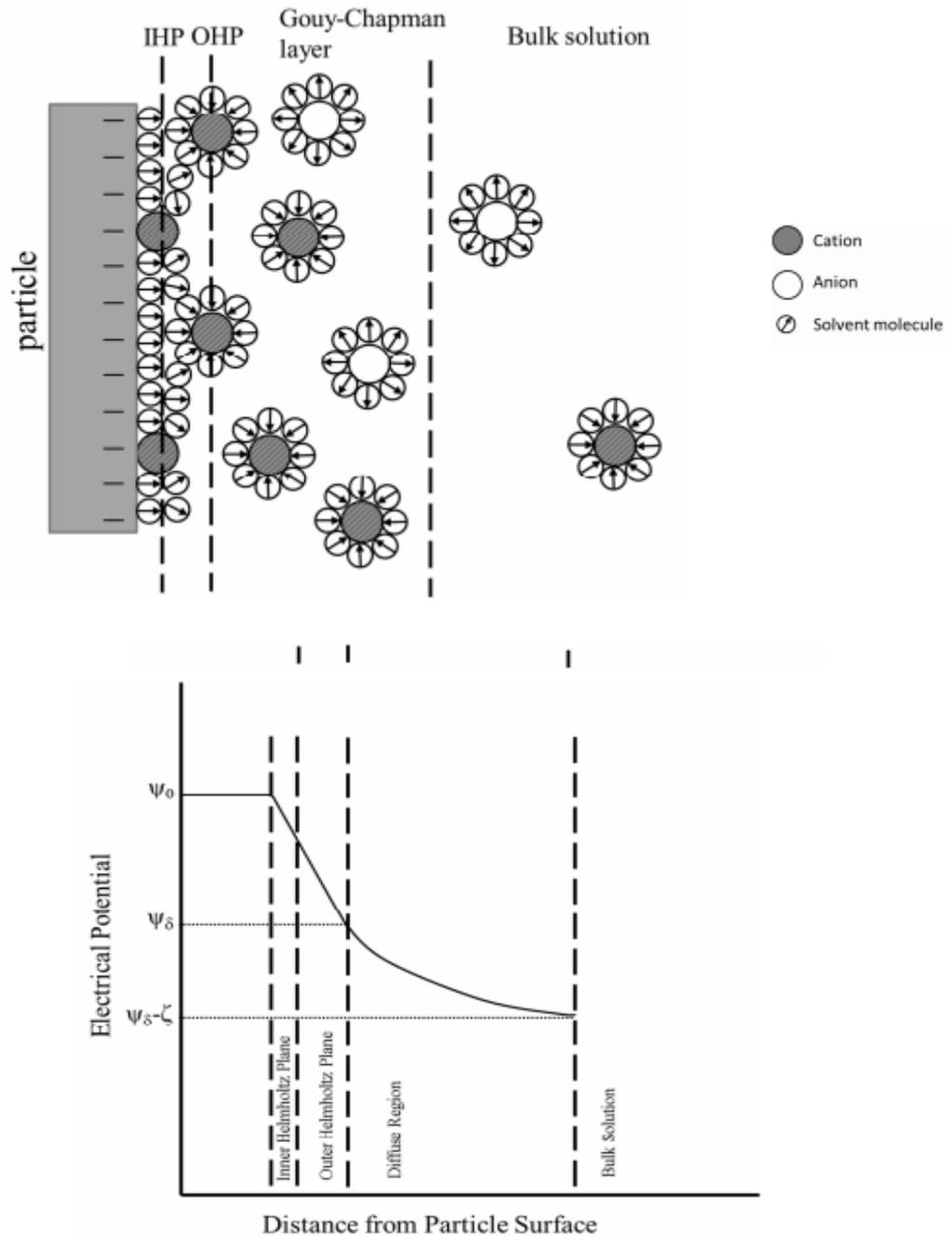


Figure (2.3): (a) Schematic of the various regions of the electrical double layer based on the BDM model, and (b) variation of the potential versus distance from the surface (Adelaide, 2013).

The first section is called the stern layer and consists of adsorbing ions. This layer is divided into two parts, the outer OHP which contains moist counter ions, and the inner which contains IHP adsorbing ions in the form of It is special and is located directly after the interface. As noted in the Fig. (2.3), the potential

decreases from the magnitude of  $(\Psi_0)$  to  $(\Psi_\delta)$  in this layer (*Adelaide,2013*). The potential decreases dramatically as the distances from the interfaces increases. This layer has been referred to as the scattering layer, its limits however are referred to as the shear plane. A model's Buchris, Devanathan, and Muller design that studies the impact of the solvent adjacent to the interfaces and has proven the presence of a layer of water inside its inner plane. As for the adsorbents ions, they displace part of the water molecules, as seen in Fig. (2.3) (a). The first layer is followed by the other layers of water except for the binaries. The electrodes are not stable as in the first layer (*Abd Ul-Shaheed,2018*).

#### **2.3.4. Flocculation**

The stage of agglomeration and the collection of pollutant grains is in to two stages. Flocculation refers to the physical process of bringing particles together, once they have been destabilised by the coagulation process. In the flocculation process, the collisions of the microflocs formed during the coagulation step cause them to bond to produce larger, visible flocs. The size of the flocs continues to increase because of the collisions and interactions with the coagulant as shown in Eqs (2.3),(2.4),(2.5) (*AlJaberi (a),2018*).

The second stage is the release of hydrogen ions at the cathode and the formation of adsorbents for metal hydroxides when a potential difference is applied at the beginning of the electrocoagulation process (*Adelaide,2013*).

#### **2.4. Operation and *Designing* of Electrocoagulation Reactor**

The batch and continuous modes of operation are the two types of electrocoagulation method. In addition to the other factors of electrocoagulation reactor designing, both types of operating modes have an impact on the magnitude of pollutant removal efficiency. An experimental design is a method for determining a relation between the operating variables and observable

responses, or a set of variables that will result in some practical benefit. It has been one of the most successful strategies to enhance the in performance.

## **2.5. Main factors affecting EC treatment**

There are several factors affecting the electrocoagulation process that must be studied along with their impacts on the quality and performance of the process such as electrodes materials ,electrodes arrangements, pH of solution ,electrolyte type and amount , impact of applied voltage.

### **2.5.1. Electrodes Materials**

In every electrochemical process, the electrode material has a big influence on how the effluents are treated. It must be nontoxic, affordable, and widely available for the remediation of potable water. The electrodes of iron and aluminum are the most popular electrode materials used in electrocoagulation systems. The electrodes in the most of wastewater remediation settings are made of the same material, mostly due to the fact that they have the same resistant and potential. The type of the contaminants, as well as the chemical parameters of waste water or electrolyte, influence the electrode material selection. In most kinds of contaminants, aluminum is more effective than iron in terms of removing efficiency (*Abd Ul-Shaheed ,2018; Bertsch and Parker ,2020*).

### **2.5.2. Electrodes Arrangements**

There are several ways to arrange the electrodes. They can be arranged according to a monopolar system so that all anodes and cathodes are connected in a series with each other making the same current flow through each pole . Alternatively, they can be arranged according to a bipolar system so that the external electrodes are connected in parallel with a source equipped with voltage and current in which the electric current is divided between the electrodes, as in the Fig (2.4) ( *Kabdaşlı, et al ,2012*). The positive side opposite the other electrodes side is polarized with a negative charge and vice versa. The

arrangement of the electrodes is an important factor in the coagulation process that affects the removal efficiency. It has been proven by some studies of the removal efficiency that the bipolar system is the most efficient (*Adelaide,2013*).

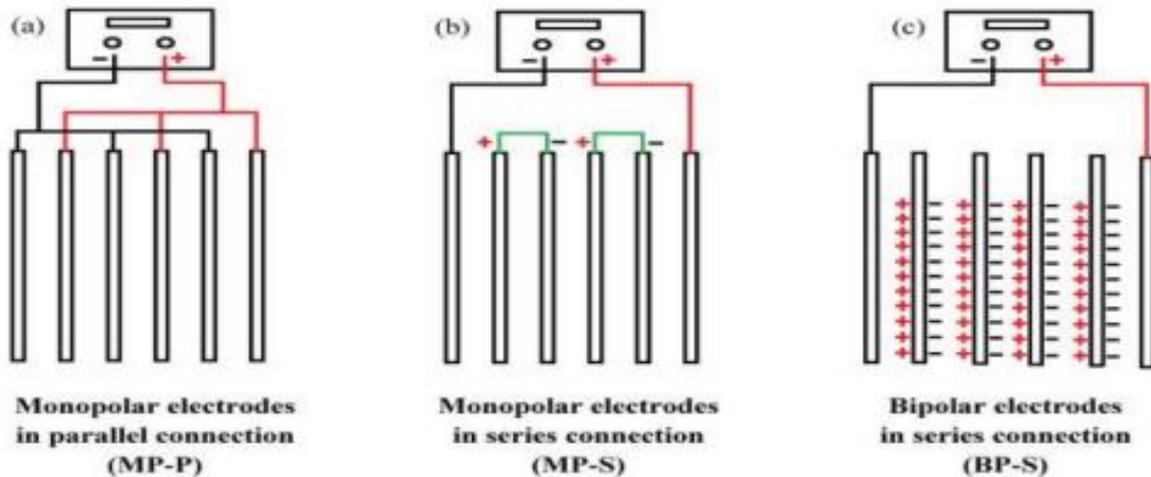


Figure (2.4): Arrangement of electrodes in electrocoagulation cells (*AlJaberi (a),2018*).

### 2.5.3. pH of solution

The coagulation process is influenced by the ultimate pH of the solution. Therefore, the pH fluctuates during the electrotreatment process. The acidity of the solution increases with an increase in the initial pH when it is within 4 or less and decreases when the initial pH is within 8 or less. However, in the case of the initial pH within 6 -8, a slight change in pH will occur.

The process of production and consumption of metal hydroxide ions and charge neutralization during the electrolysis process is called the buffer pH capacity ( *Kabdashli,et al ,2012*). The pH decreases when metal hydroxides are formed at the sites near the anodes and the final pH increases since the production of hydroxyl ions at the cathode (*Akbartabar,et al,2017*). Fig (2.5) demonstrates the percentage of the metal hydroxyl with the pH value.

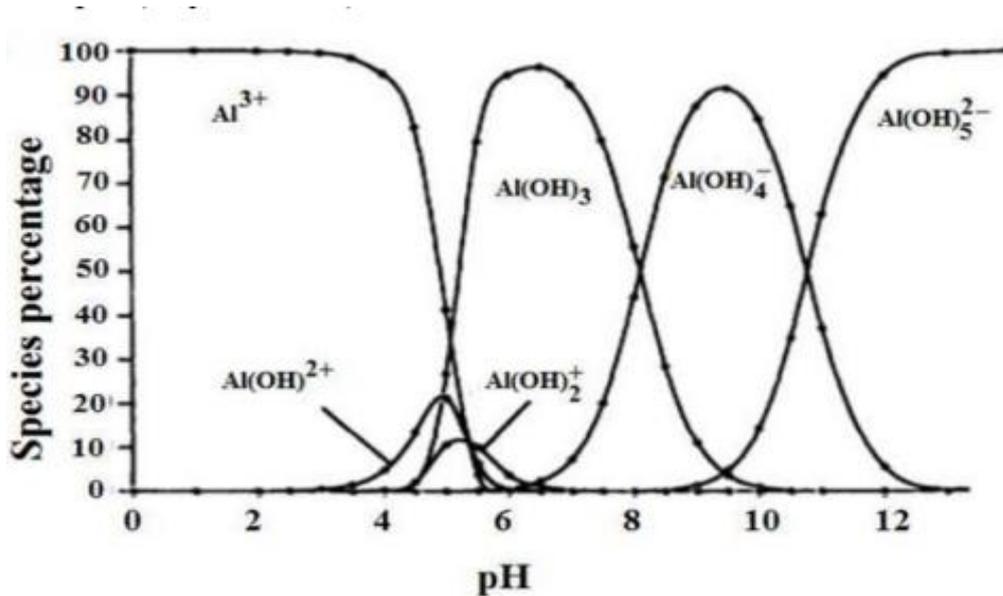


Figure (2.5): Aluminum hydrolysis species distribution as a function of pH (AlJaberi (d),2018)

The best hydrogen and oxygen bubbles that provide a sufficient interfacial space for unstable pollutants to collect molecules and form flocs. It can be said that pH is the main controller for the coagulation and adsorptions process since it controls the metal of the electrodes dissolved in the liquid surrounding media (Chen,2004).

#### 2.5.4. Electrolyte Type and Amount

In the process of electrocoagulation, the conductivity of the solution must be enhanced. The solution is utilized for this purpose as metal chlorides, and this leads to an increase in the efficiency of the remediation process (Abd Ul-Shaheed,2018) . In addition, the electrochemical process is heavily reliant on conductivity since it boosts removal efficiency and decreases the development of an oxide layer on the cathode electrodes by introducing metal salts like chlorides or sulfates metals  $\text{Na}_2\text{SO}_4$  (s),  $\text{NH}_4\text{Cl}$  and  $(\text{NH}_4)_4\text{SO}_4$  and reduce the applied effort to complete the experiment .

Also, this factor is involved in reducing operating costs, since the addition of metal chloride leads to a decrease in energy consumption, as it increases the

conductivity of the solution. This can be observed in practice during experiments. It must also be mentioned that the amount of metal chlorides should be improved and in known and specific quantities since excessive amount leads to the dissolution of the anodes electrodes significantly and the corrosion of the electrodes may be unregulated (*Kabdaşlı, et al, 2012*).

The presence of harmful ions such as  $\text{HCO}^{-3}$  and  $\text{SO}^{-2}$  which limit the dissociation process of the metal anodes is another reason for the addition of metal chlorides to form chloride ions which prevent its occurrence in addition to oxidizing agents ( $\text{OCl}^-$ ) and ( $\text{HOCl}$ ) resulting from the (EOX) process that occurs as a result of direct or indirect oxidation. Direct oxidation of the pollutant occurs as a result of the formation of hydroxyl radicals on the surface of the electrodes. (*AlJaberi (b), et al, 2020*).



At the anodic electrodes, a side reaction occurs, resulting in the creation of oxygen, which is an undesired formation since it lowers the electrochemical process efficiency and raises the operation's economic expenses (*Chen, 2003*).

### 2.5.6. Impact of Applied Voltage

The voltage supplied at the electrodes is one of the most critical factors affecting the performance and efficiency of the electrocoagulation process. The number of  $\text{Al}^{3+}$  ions released from the various electrodes and the amount of coagulant produced are determined by the current supplied to the electrocoagulation system. such as, more ions dissolve in the solution, increasing the rate of  $\text{Al}(\text{OH})^3$  production (*Bazrafshan, et al, 2012; Mollah, et al, 2001*).

## **2.6. Characterization of Electrocoagulation Process**

### **2.6.1. Removal efficiency**

It is one of the important parameters on which the electrocoagulation process depends. It gives an indication of the performance of the cell and the designing of the electrodes through which the efficiency of removing a contaminant and the required rate of the coagulation process gained depends on it. It is calculated through the mathematical relation : removal efficiency (Y (percent)) has been calculated utilizing Eq. (2.11):

$$Y = \frac{C_o - C_t}{C_o} \times 100 \text{ percent} \quad (2.11)$$

where Y is the removal efficiency response; Co and Ct are the initial and final amount of pollutant (ppm) (*Adelaide ,2013; Khandegar and Anil ,2013*).

### **2.6.2. Energy Consumption**

It is important for the electrocoagulation process through which the economical issue of the process is studied under the effect of several factors, namely the time of electrolysis, the amount of current, the applied voltage, and the volume of the electrolyte solution. All these variables related into a mathematical correlation through which the energy consumption. Eq. (2.12) as follows:

$$\text{Energy Consumption} = (U.I. t)/(1000 .V) \quad (2.12)$$

Whereas: U denotes the applied voltage in volts, I denotes the applied current in amps, t denotes the electrolysis time in hours, and the contaminated water volume ( $m^3$ ) is V (*AlJaberi (d) ,2018*).

There are also some factors that affect energy consumption is related to the:

- Designing of the reactor, the composition and arrangement of the electrodes, and the distance between them.

- The type of metal the electrodes are made of, since it is responsible for the formation of adsorbents materials. The floc component the faster it is, the lower the energy consumption.
- The nature of the electrolyte solution and the amount of the pollutant, which strongly affects the energy consumption.
- pH which is related to the formation of metal hydroxide (*Andrade, et al, 2007*).

### **2.6.3. Electrodes Consumption**

The amount of consumed electrodes decreases when choosing the best design and arrangement of electrodes since the time to complete the electrocoagulation process is less. At the anodes, the consumption is large since it produces metal hydroxides, and the theoretical consumption of electrodes is calculated from faraday's law (2.6).

Through the faraday's law, the amount of consumption of the electrodes depends on the time of electrolysis, the molecular weight of the metal and the amount of electric current passing through electrodes these factors are directly proportional to the amount of consumption, while the number of free electrons is inversely proportional to the consumption. The removal efficiency of the electrocoagulation process is dependent on the amount of coagulants formed to complete the remediation process (*Daneshvar, et al, 2003*).

### **2.6.4. Applied Current**

Adjusting the supplied current can govern the generated adsorbents by electrochemical effect. Current densities are extremely impacting color removal. The relation between current density and the quantity of adsorbents formed in the solution is described by Faraday's law Eq. (2.6).

In addition, it has an influence on the electrode potential, which determines the reactions taking place on the electrode surface. Indeed, it has been reported

that the dissolution rate of the anode can be lower than the theoretical value computed by Faraday's law, The concentration of  $Al^{+3}$  and  $Fe^{+2}$  ions produced from the anodes is usually calculated according to Faraday's law, provided that the current and electrocoagulation times are known (*Adelaide,2013*).

### 2.6.5. Electrocoagulation Reactor Kinetic Approach

Several kinetic models are proposed to describe the behavior of the reaction occurring in the reactor, such as the first-order and second-order formulas depending on the general formula of designing Eq. (2.13) to refer the kinetic of pollutant degradation by electrocoagulation technology.

$$\frac{dC}{dt} = -kC_t^n \quad (2.13)$$

where C, n, k, and t refer to the RBD concentration, order of reaction, the rate constant, and the electrolysis time (*Othmana,et al,2018*).

### 2.6.6. Thermodynamics Study

Thermodynamic parameters could be measured depending on the variation of solution temperature as the remediation process proceeds as well as on the equilibrium constant ( $K_d$ ) at each temperature that could be obtain from Eq. (2.14) as follows:

$$K_d = (q_e / C_e) (V/W) \quad (2.14)$$

where  $q_e$  is the quantity adsorbated in (mg/g),  $C_e$  is the RBD equilibrium concentration (mg/l), V is the solution theoretical volume (0.75 liter), and W is the electro-coagulant weight (g) (*Khan and Singh,1987*).

The entropy ( $\Delta S$ ), Gibbs energy ( $\Delta G$ ), and the heat of adsorption ( $\Delta H$ ) of the remediation process for both modes of CVM and CCM could be evaluated utilizing Eqs. (2.15 to 2.17):

$$\text{Log } K_d = -(\Delta H / 2.303 RT) + \text{constant} \quad (2.15)$$

$$\Delta G = -R T \ln K_d \quad (2.16)$$

$$\Delta S = (\Delta H - \Delta G)/T \quad (2.17)$$

The positive sign of the heat of adsorption ( $\Delta H$ ) and the entropy ( $\Delta S$ ) indicates that the operation is endothermic and ascribed to a random irregularity at the solid-liquid, whereas the negative magnitude of the Gibbs energy ( $\Delta G$ ) refers to the spontaneous nature and more favorable adsorptions of the remediation process (*Harrache, et al, 2019*).

### **2.7. Advantages and Disadvantages of EC**

Electrocoagulation has characteristics that distinguish it from other methods:

- simplicity of tools and ease of operation;
- low cost;
- the remediation system is compact and does not contain many moving parts, which leads to a decrease in maintenance;
- can be run on solar energy;
- no need to add chemical coagulants since these materials lead to secondary pollution;
- this method makes the sludge layer less acidic and stable, containing a small percentage of water, making it resistant and easy to filter and separate water from it;
- water can be recovered since the resulting sludge does not contain chemical coagulants and the percentage of dissolved solids is less. This is considered an economical characteristic of this method; and,
- the release of hydrogen gas during the remediation process from the cathode electrodes. These bubbles help carry pollutants to the surface of the treated water, which facilitates their collection and removing.

As for the disadvantages of this method, they can be summarized as follows:

- the use of electrical energy, which restricts its use and increases operating costs;
- during the oxidation process of the anodes, oxides form on the surfaces of the electrodes, which reduces the removal efficiency;
- the great conductivity of the solutions leads to a rapid increase in the consumption of the anodes electrodes, which requires replacement; and,
- the findings are acquired in a controlled environment in laboratories, so it is not always possible to apply conditions to the industrially sector (*Anantha Singh and Ramesh,2013*).

## **2.8. Literature Review**

Several researchers have been employed the electrocoagulation method to treat wastewater containing dyes.

*Daneshvar, et al. (2003)* conducted a batch EC reactor to remove Orange II dye under the impacts of water temperature, current density, electrode spacing, dye concentration, stirring speed, initial pH, and other variables. When iron anode was utilized and 200 mg orange II per liter, the removal efficiency was (98 percent) and COD was minimized by (84%) according to the experimental findings. The efficient removal of orange II was obtained at the optimal current density of 34.62 A/m<sup>2</sup>.

**Golder, et al. (2005)** investigated the electrocoagulation (EC) efficiency of aqueous dye solution of two distinct commercial colors in a batch stirred cell in their work. They have carried out the experiments utilizing 200 ppm for each of eosin yellowish and methylene blue. The impact of operation duration and current densities of current on dye and COD removal, and electrical conductivity and pH change during remediation have been investigated using

NaCl electrolyte. Starting from a 200 mg/l dye amount, 1.5 KWh of electric power lowers 0.21 and 0.11 kg COD for MB and EY, respectively, from 0.24 and 0.14 kg of baseline COD .

**Kobyá, et al. (2006)** have utilized electrocoagulation with aluminum sacrificial anodes to decolorize the levafix orange textile dye. As a consequence of electrolysis duration, initial dye amount, current densities, conductivity, and initial pH, the performance of processing is evaluated depending on the removal efficiency and key factors that are related, including electrodes and energy usages. The efficiency of electrochemical remediation for textile dye wastewater has been shown in this research. At appropriate operating conditions, including the current density of 100 A/m<sup>2</sup>, reaction time of 12 min, and initial solution pH 6.4, a 95 percent decolorization efficiency was achieved. Where a 1.8 kg Al/kg dye and 35 kWh/kg dye of electrodes and energy consumption, respectively .

**Daneshvar, et al. (2006)** have employed EC to remove dye from samples having Basic Blue 3 (BB3) and Basic Red 46 (BR46). The influence of operational variables of current density, initial solution pH, electrolysis time, initial dye concentration, and electrical conductivity were investigated. The results revealed that increasing the current density to 60–80 A/m<sup>2</sup> improved color removal efficiency, the electrolysis time was 5 min, and the pH range was 5.5–8.5 for the two dye solutions studied. In dye samples having BB3 and BR46, COD reduced by more than 75 percent and 99 percent, respectively, throughout the EC processing under the optimum conditions .

**Song, et al. (2007)** studied what factors impact the efficiency of the ozonation process and electrocoagulation using iron electrodes to remove dye from wastewater containing Reactive Black-5dye (RB5). Several operating

variables have been investigated, including initial pH, initial dye amount, current density, salt ( $K_2SO_4$ ) amount, temperature, ozone flow rate, and spacing between the electrodes. The results of the experiments show that the color of RB5 in the aqueous phase has been successfully eliminated. The color removal efficiency was 94 percent under the following factors: initial dye concentration =100 mg/l, initial pH of 5.5, current density of 10 mA/cm<sup>2</sup>, salt amount of 5000 ppm, temperature of 20 degree centigrade, ozone flow rate of 20 ml/min (ozone dosage of 0.20 g/h), and inter-electrode spacing of 10 mm. The process consumed around 33 kWh of energy every kilogram of COD removed.

**Şengil and Özacar (2009)** performed an EC reactor containing iron electrodes to remove Reactive Black 5 dye from synthesized wastewater under the effect of the initial concentration of dye, electrolysis time, current density, initial pH, and solution conductivity. The dye removal efficiency of 98.8 percent was achieved when the initial dye amount was 100 ppm, the initial pH was 5, the current density was 4.575 mA/cm<sup>2</sup>, the salt amount was 3000 ppm, the temperature was 20 degree centigrade, and the inter-electrodes spacing was 2.5 cm. The consumption of energy per kilogram was 4.96 kWh/kg .

**Patel, et al. (2010)** conducted an EC process for removing Direct Black 22 and Acid Red 97 colors, COD, and organic compounds from wastewater using iron and aluminum. The effect of operating variables including densities of current, initial solution pH, duration of electrolysis, and electrodes materials on the batch process was investigated to achieve maximum removal of contaminants.

**Islam, et al. (2011)** employed the EC method to remove different concentrations of Orange II dye from simulated wastewater using an Iron

sacrificial anode and 30V for 30 minutes. They attained 65 to 92% of removal efficiency for 10 to 60 ppm of dye concentration with a considerably low total operating cost. They documented that the EC is a potentially viable and inexpensive process for dye removal from wastewater.

**Gomes, et al. (2011)** proved the usefulness of the EC approach for removing dye, including direct red dye utilizing iron electrodes as well as the optimal dye concentration, current density, and pH conditions. SEM/EDS, XRD, and FTIR were used to analyze the EC-floc.

**Patel, et al. (2011)** investigated the ability of a batch EC reactor to remove 25 ppm of reactive black dye (RB5) from synthesis floor-wash wastewater. More than 90 percent of removal efficiency was achieved, in less than one hour, at a current density ranging from 4.5 to 7.5 mA/cm<sup>2</sup> in the presence of the supporting electrolytes NaCl and Na<sub>2</sub>SO<sub>4</sub>. The energy consumption was 29 kWh/kg RB5 in the presence of NaCl that was almost half that of Na<sub>2</sub>SO<sub>4</sub>. They revealed that EC can be efficiently used as a primary treatment for dye removal from floor-wash in the presence of NaCl .

**Pajootan, et al. (2012)** In a batch electrochemical reactor, investigated binary (Acid Yellow 220 and Acid Black 52) systems color removal by EC utilizing aluminum electrodes. The operational factors were solution pH, initial concentration of dye, conductivity, and current density. The dye removal efficiency was improved when the current density has been raised to 40 A/m<sup>2</sup>, and the optimal pH of 5. The color removal was unaffected by the increase of the electrolyte content from 0 to 8 g/l, although the energy consumption was reduced. They conducted that the electrocoagulation process is an effective method to remove dyes from textile wastewaters.

**Bayar, et al. (2012)** conducted EC to remove Direct Red 23 from wastewater using aluminum electrodes with constant spacing among them. This investigation was done under the influence of the initial pH, stirring speed, and supporting electrolyte type while the current density and temperature were fixed at 0.1 mA/cm and 20 degree centigrade, respectively. The highest removal efficiency of 98 percent was achieved at the optimal conditions of pH 5, 150 rpm stirring speed, and 5 mmol NaCl.

**Pi, et al. (2014)** investigated the use of electrocoagulation with periodic electrodes reversal to treat synthetic wastewater containing Methyl Orange (MO) and azo dye. The impact of experimental settings on color removal, energy and electrodes consumption, and sludge generation per kg has been optimized using (RSM). pH 7.4, solution conductivity 9.4 mS/m, cell voltage 4.4 V, current density 185 mA/cm<sup>2</sup>, electrocoagulation duration 14 min, a cycle of periodic reversal of electrodes 15 s, inter-electrodes distance 3.5 cm, and initial MO concentration of 125 ppm were determined to be optimum conditions. A 97.2 percent of the color has been eliminated under these conditions, and the energy and electrodes consumption, and sludge generation values have been 44.3 kWh/ kg(MORs), 4.1 kg(Al)/kg(MORs), and 17.2 kg(sludges)/kg(MOr), respectively.

**Chigozie and Joseph (2014)** tested the combination of Nanofiltration and electrocoagulation in removing dye from synthetic wastewater. Vat dyes (Vat yellow), synthetic dyes (Erythrosine), and Azo dye have been chosen as typical dyes molecules for the synthetic dyes wastewater (Orange-G dye) using an iron electrode as a sacrificial anode. The operating variables were the temperature, initial concentration of dye, initial solution pH, current density, and electrolysis time. With an initial concentration of dye of 100 mg/l, a density of current of 1559 A/m<sup>2</sup>, and reaction time of 25 min, and initial

solution pH of 10, the findings revealed that 99.958 percent of Orange-G, 99.854 percent of Erythrosine, and 85.956 percent of Vat yellow were decolorized .

**Alizadeh, et al. (2015)** investigated the effectiveness of the 2 Liter batch EC method using electrodes made of aluminum in eliminating the reactive green dye (RG-19) from wastewater using four aluminum electrodes. The effect of the operational variables on the removal effectiveness was examined, including voltage, time of reaction, initial concentration of dye, energy consumption, KCl amount, pH, and inter-electrodes distance. At optimal conditions (KCL quantity of 0.005 M, pH 11, and distance of 1 cm), the maximum removing effectiveness of RG dye was determined to be 33.49%, 60.32%, 72.43%, 93.63%, and 94.91% for applied voltages of 10 V, 20 V, 30 V, 40 V, and 50 V, respectively. They found that the removal efficiency attained 99.88% with the increase of dye from 25 to 150 mg/l .

**Carvalho, et al. (2015)** investigated the use of an electrocoagulation (EC)/banana peel (BP) adsorption coupling technique to enhance methylene blue (MB) adsorption from aqueous solutions. The effect of current density on the EC/BP combination process, removal efficiency, and energy consumption were investigated. They found that when the combination of electrocoagulation and BP adsorption led to an increase in dye removal. Particularly at lower current densities, and a significant reduction in contacting time when compared to the traditional simple EC and simple adsorption processes, with removal efficiency rates of approximately 99 percent short contacting time and low energy consumption.

**Mahmad, et al. (2016)** performed Electrocoagulation for the removal of total chromium, color, and turbidity contamination in landfill leachate using

stainless steel and aluminum electrodes. The pH range was (3, 4, 5, 6, and 7) while the voltages were 1.5, 2.0, and 2.5V. They proved that the efficiency of eliminating total chromium, color, and turbidity was depending on the metal of the electrodes (stainless steel or aluminum). According to the core findings, aluminum electrodes are the best for removing turbidity and color .

**Ghalwa, et al. (2016)** investigated the efficiency of an EC reactor employing aluminum and iron electrodes to remediate synthetic wastewater including Reactive Red 24 under the effects of current density, kind of electrolyte, pH, initial concentration of dye, electrolyte amount, inter-electrode distance, and temperature. The core findings revealed that iron electrodes removed 99.6 percent and 91.5 percent of dye and COD, respectively, whereas aluminum electrodes removed 97.9percent and 83.8 percent. The dye removal was pseudo-first-order, with high  $R_2$  (0.99 and 0.955 for Al and Fe electrodes, respectively).

**Rekha and Murthy (2016)** investigated the ability of EC technology to reactive dye from wastewater under the impact of the electrolysis time at various applied currents, and pH values. They found that at a neutral pH, 90 percent of color removal was attained with little sludge generation and anode consumption.

**Shah, et al. (2017)** proved the beneficial use of EC treatments of the binary dye systems containing Reactive red (R223) dye and Coomassie brilliant blue (CBBR250) dye using aluminum and iron for the anode and the cathode, respectively. The electrolysis process was preceded under the effects of the operational variables of pH, NaCl, a voltage applied, and reaction time using the RSM statistical method. As response variables, the percent color and COD reductions have been investigated. At the optimal values, the removal

efficiency of RR223, CBBR250 dyes, and COD was 89 percent, 94 percent, and 100 percent.

**López, et al. (2017)** used a novel electrocoagulation reactor with a cylindrical shape employing a 3D steel wool anode based on a filter cartridge that has been modified to hold the electrodes. The resident time distribution (RTD) was utilized to investigate the electrolyte flow behavior inside the reactor. The novel reactor was utilized to effectively remove Remazol Red RB 133 dye in continuous operation mode, with a dye removal of 99 percent

**Getaye, et al. (2017)** used an EC method to remove malachite green, an antibacterial color used in aquaculture. Utilizing Fe and Al as scarifying anodes, the effect of initial dye amount, current density, pH, inter-electrodes spacing, and electrolyte amount on dye removal were studied. Furthermore, optimal current density ( $76.5\text{A/m}^2$ ) and solution pH (8.0) are evaluated to attain maximum removal of malachite green dye from wastewater. Using an electrolyte (NaCl) concentration of 0.4 g/l, an inter-electrodes spacing of 1 cm, and optimal pH and current density, complete color removal of a 100 mg/l malachite green wastewater was obtained within 30 minutes. The color removal efficiency using Al and Fe as sacrificial electrodes was nearly the same, with the exception of their reaction to inter-electrodes spacing, which has been varied, with the color removal rate of Fe as anodes declining more quickly.

**Mook, et al. (2017)** suggested regressions model for the electrocoagulation treating of contaminated water containing reactive black dye (RB5) using response surface methodology (RSM). The operating variables were treatment time, applied current, and initial solution pH. The highest removal efficiency of 83.3 percent was obtained at pH 6.63, the applied current of 0.075 A, the electrolyte dosage of 0.11 g/l, and the electrolysis time of 50.3 minutes .

**Khorram and Fallah (2018)** investigated the ability of the EC method for the remediation of industrial dye wastewater under the impact of the operating factors including initial pH ranging from 4 to 9, a current density of current of 15 to 35 mA/cm<sup>2</sup>, and EC time from 20 to 60 min). It has been evaluated utilizing a short settling period of 30 minutes, as opposed to previous studies that utilized a settling time of 12 hours or more. Under the following optimal conditions, decolorization has been achieved to be 98 percent: EC time of 23 minutes, the current density of 15 mA/cm<sup>2</sup>, and initial pH of 5.5. Under ideal conditions, COD removal efficiency, the velocity of settling sludge (SSV), electrodes, and energy consumption all have been assessed. A 40 percent, 0.004 cm/min, 1.3 kg/ m<sup>3</sup>, and 7.64 kWh/m<sup>3</sup> were the values, respectively. Moreover, 40 percent of settled trash may be recovered under ideal conditions; otherwise, additional settling time is needed.

**Duan, et al. (2018)** used Electrocoagulation technology with periodical electrodes reversal (PEREV-EC) to remediate synthetic wastewater containing methylene blue (MB). An applied voltage of 3.1 V, electrodes reversal duration of 2.5 s, the current density of 347 mA/cm<sup>2</sup>, and inter-electrodes gaps of 2 cm have been shown to be the ideal condition for decolorization, electrode consumption, and sludge generation using the Surface Responses Methodology (RSM) approach. The electrodes consumption and sludge generation have been  $0.25 \pm 0.02$  kg(Al)/kg(MBR-removed) and  $1.9 \pm 0.9$  kg(sludge)/kg(MBR), correspondingly, under these conditions  $97 \pm 2$  % of the color has been removed.

**Naraghi, et al. (2018)** achieved 96 percent of Azo Reactive Black 5 (RB5) from textile wastewater at pH 6 and 80 min of reaction time. They concluded that the consumption of electrodes and energy, pH, the ultimate temperature of effluent, and removal efficiency was increased as reaction time increases .

**Zazou, et al. (2019)** examined the use of EC in combination with electrochemical advanced oxidation processes (EAOPs) including electro-Fenton (EF), peroxi-coagulation (PC), and anodic oxidation (AO) to treat real effluents containing a mixture of reactive dyes (primarily methylene blue). Sequential EC-EF treatment was more efficient compared to other EAOPs investigated because it delivered electro-generated hydroxyl radical OH, particularly when employing boron-doped diamonds. Utilizing the EC-EF combined process, TOC, turbidity, and color removal have been reduced by 97 percent, 100 percent, and 100 percent, respectively .

**Nippatla and Philip (2019)** They have assessed the individual and combined performance of Pulse Powers Plasma Treatments (PPT) and Electro-coagulations-Flotation (EC-F) methods in the remediation of textile wastewater involving Congo Red dye (CR) diazo dye, and Methylene Blue (MB). The operating variables were the influence of the initial dye amounts, current density, electrical conductivity, and reaction time. At a current density of 14.2 mA/cm<sup>2</sup>, the electrical conductivity of 8 mS/cm, the concentration of dye was reduced from 50 ppm to below detection level and 50 ppm to 3.1 ppm for CR and MB dyes, respectively, using EC-F alone. Electrolysis times of 2 and 14 minutes were required for removing CR and MB, respectively .

**Abdulhadi, et al. (2019)** studied the effects of electrode gap (EG: 0.5, 1, and 1.5 cm) and current density (CD: 2, 4, and 6 mA/cm<sup>2</sup>) on the EC process for removing reactive red 120 dye from potable water. The results indicated that the increment of CD increased the dye removal rate. Nevertheless, increasing the EG reduced the dye removal efficiency. Dye elimination increases from 87 to 98 percent when current density increased from 2 to 6 mA/cm<sup>2</sup>, according to the core results. While the dye removal reduced from 96 percent to 80 percent when the EG was increased from 0.5 to 1.5 cm .

**Hashim, et al. (2019)** used an electrocoagulation reactor involving aluminum electrodes to evaluate the impact of the initial solution pH on the removal efficiency of reactive black (RB5) dye from wastewater. Several groups of continuous mode studies were conducted at five various values of initial pH ranging from 4 to 8. The variables of the current density applied, inter-electrode distance, and RB5 concentrations were fixed at 2 mA/cm<sup>2</sup>, 4 mm, and 25 ppm, respectively. The dye removal efficiency increased gradually as the initial pH increased from 4 to 6, reaching 96 percent at the neutral solution pH, then decreasing to 74 percent when the initial value of pH increased to 8.

**Tlaiaa, et al. (2020)** compared the chemical coagulation including alum, polyaluminum chloride, ferric chloride, ferrous sulfate, and ferric sulfate, and the electrocoagulation involving aluminum electrodes. The highest removal efficiencies for dye and COD using chemical coagulation were 98% and 90%, respectively, using ferric sulfate at optimal pH and dose of 4,200 ppm. While the highest removal efficiencies of these pollutants were achieved at the optimal values of pH, NaCl amount, initial dye concentration, and applied voltage .

**Teng, et al. (2020)** have compared the performance among the electro-Fenton (EF), electrocoagulation (EC), and electro-oxidation (EO) processes to remove methylene blue from simulated wastewater. The EO employs graphite as a cathode, iron rods as the anode, and fly ashes–red mud particles as electrodes particle. The best performance of EO was attained in the pH ranging from 3 to 11 compared to EC and EF integrated system. The energy consumption of these technologies was 21.35, 36.55, and 81.51 kWh/m<sup>3</sup> for EO, EC, and EF, respectively.

**Alwash, et al. (2021)** used the technique to enhance the electrocoagulation (EC) removal of 28 ppm of reactive black (RB5) dye from wastewater. Simulated wastewater was treated initially using the EC reactor for 1 hour at 4, 5, 6, 7, and 8 of initial pH, and current density ranging from 1 to 3 mA/cm<sup>2</sup>. Then, the wastewater was treated again but the sonication effect was done before for 10 min. The finding showed that 82.14 percent of dye removal was attained at pH 4 and 2 mA/cm<sup>2</sup> of current density while the complete removal was attained when the sonication process was done before the EC process.

**Abd Ul-Shaheed ,Sh .H., Ajjam,S,K. (2018)**, A laboratory batch electrocoagulation reactor of four monopolar electrodes is designed to investigate the effects of various parameters on the nitrate removal efficiency, such as initial pH (7,9,11) , distance between electrodes (1,2,3)cm , applied voltage (5,10,15)V and the electrolysis time (30,60,90)min . From the experimental results , the most parameters effect on the removal of nitrate are initial pH and voltage applied to the treatment reactor . It is found that the nitrate removal is increased as the initial pH increases from (7-9) and gives the best removal at pH = 9, also when the voltage increases from (5-10)V , and for the distance effect the removal decreases as the distance increases from (1-3) cm .While the time effecton the nitrate removal increases at time increases from(30-75)min . The maximum nitrate removal efficiency is 93.3% at (pH=9 ,10v ,Ci = 50 ppm , d=1cm , 120min , 100rpm , 0.5g/l NaCl )

## Chapter Three

## Experimental Work and procedure

## 3.1. Introduction

This chapter aims to detail the features of the electrocoagulation system and its accessories, in addition to the method of the electrocoagulation experiment consistent with the suggested design.

## 3.2. The Construction of the Electrocoagulation Unit

A one-liter batch electrocoagulation reactor demonstrated in Fig. (3.1) consists of a PVC cover and four plate-electrodes manufactured of aluminum in a bipolar-parallel mode.

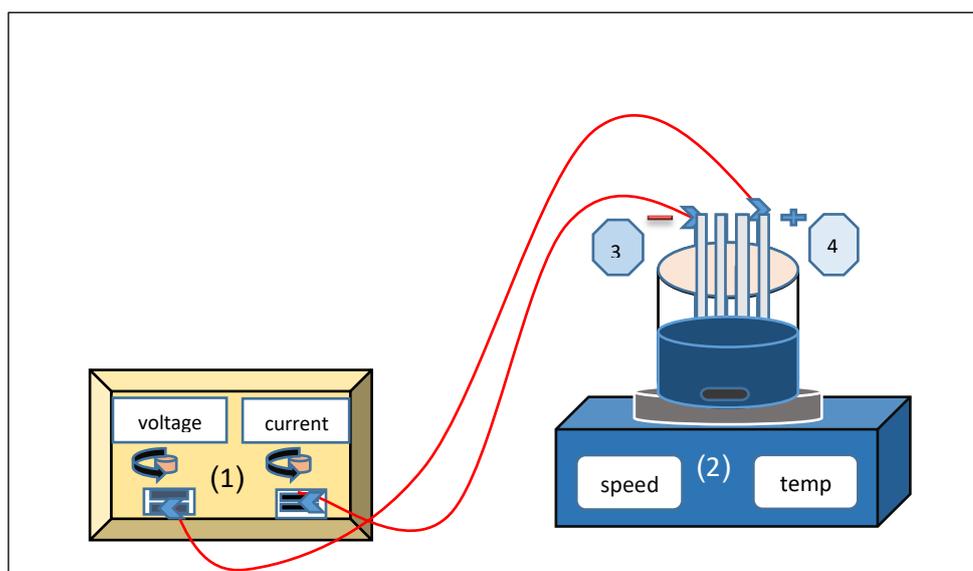


Figure (3.1) The electrocoagulation reactor: (1) Digital DC power supply; (2) Magnetic stirrer; (3) Cathode; (4) Anode (bipolar electrodes in parallel connection).

These plates are plane designed to be and perforated, as demonstrated their configuration in Fig. (3.2). Each of perforated plates contains 49 holes with a diameter of 0.5 cm and a distance of 0.5 cm from other holes where the total effective area is  $255 \text{ cm}^2$ . The composition of the aluminum electrodes was

examined by (EDS) the use of energy dispersive spectroscopy testing (Oxford instruments-X-act) and X-rays testing (X-Rays Diffractometers XRD 6000/Shimadzu), as demonstrated in Table (3.1).



Figure (3.2): Electrodes geometry

Table (3.1): Aluminum electrodes chemical analysis expressed as a percentages of total weight

<b>Standard ident</b>	<b>Al</b>	<b>Si</b>	<b>Mg</b>	<b>Ti</b>	<b>Mn</b>	<b>Zn</b>	<b>Cu</b>	<b>Fe</b>	<b>Cr</b>	<b>Sr</b>
<b>%Weight</b>	<b>98.137</b>	<b>0.6</b>	<b>0.45</b>	<b>0.2</b>	<b>0.2</b>	<b>0.15</b>	<b>0.1</b>	<b>0.1</b>	<b>0.05</b>	<b>0.013</b>

**3.3. Synthetic Wastewater Preparation**

The synthetic wastewater (1000 PPM) reactive blue dye was prepared by dissolving one gram of (RBD) in 1000 ml of distillate water. Each run containing 750 ml of RBD-wastewater was treated under the impact of the operating variables listed in Table (3.2). A certain amount of the stock solution was mixed

with distilled water to prepare the (750 ml) of (100 ppm RBD) synthetic wastewater solution for each experiment.

### **3.4. Apparatus**

1. (QJ3005X II) digital DC regulated power supply; (0–30) volt and (0–5) A.
2. Digital balance (500 g × 0.001 g) (TH-3068 Professional Digital jewelry scale company, China).
3. Magnetic stirrer (HP-3000); 60–1,500 rpm.
4. pH meter (ATC company, China).
5. Digital timer.
6. Three sets of alluminum plates (plane and perforated).
7. TDS -conductivity meter.
8. UV-1800 spectrophotometer (Shimadzu Inc., Japan).
9. Thermometer for measuring temperature.
10. Glass wares including pipettes, beakers, volumetric flasks and others.

### **3.5. Chemicals**

The chemicals materials that were utilized in the experiments:

1. Reactive blue dye (RBD) from M/s Loba Chemie, (India) .
2. Sodium chloride (NaCl), 99.9 % wt purity, produced by HiMedia Laboratories Pvt. Ltd., India.
3. Sodium Hydroxide, (NaOH) 96.0 % wt purity, produced by BDH Limited Poole England.
4. Hydrochloric acid (HCl), 35-38% wt , produced by BHARAT MAHAL, Pvt. Ltd., India.
5. pH- Buffer liquid surrounding medias 4, 7, and 10.

6. Distillated water .

### **3.6. Set up of Experimental Work**

1. A batch electrocoagulation reactor of 1 L of volume is consisting of four plain and perforated plate-electrodes which are manufactured of aluminum. They are connected to a DC-power supply in a bipolar-parallel mode. The outer plates are not perforated while the entire two plates are perforated (49 holes with a hole diameter of 0.5 cm).

2. The mixing process of the reactor components was carried out utilizing a magnetic stirrer (Model: HP-3000; 60–1500 rpm) to provide 200 rpm of stirring speed. A high 1 cm from the base of the reactor, the electrodes were placed in order to obtain the best mixing efficiency.

3. To determine the necessary weight of (RBD) in accordance with an electronic magnitude for the initial amount (RBD) of that was planned A capacity of digital balance was utilized (500g) with a margin of error of (0.001g).

4. Conductivity measuring meter. The conductivity and TDS (ppm) are measured.

5. The starting pH and conductivity (NaCl amount) of the reaction solution were adjusted with (0.1M) of (NaOH, HCl) and (2 g/L NaCl), respectively.

6. A thermometer used to measure the change in temperature during the preliminary experiment.

In addition, Figure (3.3) clearly depicts the electrocoagulation cell and other equipment.



Figure (3.3): In this investigation, we employed an electrocoagulation system and other equipment.

### 3.7. Experimental Procedure

#### 3.7.1. Simulated Wastewater Samples

1. After the reactor was completely set up, 750 mL of wastewater, of 100 ppm RBD .
2. To improve conductivity, 2 g/l NaCl salt was utilized.
3. Add 0.1M NaOH or HCl to get the appropriate pH.

#### 3.7.2. The Functional Parameters

The type of metal electrodes used, as well as their design, arrangements, whether bipolar, in parallel, batch mode , voltage applied at constant current, electrolysis duration, solution pH, current applied at constant voltage , and other factors, were investigated in this study.

The objective of this study was to overcome the complexity of these issues by utilizing a design of four plates , perforated and non- perforated to avoid some drawbacks and to obtain the following benefits:

1. A larger cathodic active area aids the electroflotation process by releasing a greater amount of hydrogen bubbles, allowing it for having more duration to adsorb the fine contaminant and allow to rise solution surfaces.
2. Reducing the ohmic drop between electrodes and reducing the energy consumption throughout the process of electrocoagulation.

Table (3.2) demonstrates functional parameters studied in the experiment.

Table. (3.2):-Functional Parameters

Parameters	Ranges or constant values
Reaction time (min)	2-80
Applied voltage (volt)	15-25
pH	4-12
Initial RBD concentration (ppm)	100
Electric Current (Amp.)	5
Stirring speed (rpm)	200

### **3.7.3. The General Procedure of Each Experiment**

1. Before the experiment, the DC power supply was initialized in terms of the applied voltage and current according to each experiment. The DC-device provided the electrocoagulation cell with the required voltage while simultaneously turning on the stirrer when the electrodes were immersed in the simulated wastewater.

2. On a regular basis, samples are gained from the treated wastewater. As a consequence, an UV-1800 spectrophotometer equipment was utilized to quantify the quantity of (RBD) that remained in each sample.
3. Before and after each experiment, the pH of the solution was measured.
4. Measurements of the solution conductivity before and after each experiment were measured.
5. Calculate the weight of the electrodes before and after each experiment in order to calculate the actual consumption of the electrodes during the experiment.
6. Measure the final current applied before the end of each experiment.
7. After each experiment, the electrodes are washed utilizing diluted hydrochloric acid and washed several times utilizing distilled water.

### **3.8. Design of The Preliminary Experiment**

In order to verify the impact of the change in voltage and current on the electrocoagulation process in the reactor and the dye removal in particular. Two parts were done in this study, the first one employed at a Constant Current Mode (CCM) of 3 Amp, while the second part was done at Constant Voltage Mode (CVM) of 20 volts. Each run containing 750 ml of 100 ppm RBD-wastewater was treated under the impact of the operating variables listed in Table (3.3). Periodic sampling was conducted according to the interval of (2, 5, 10, 20, 30, 40, 50, 60, and 70 min), consequently, measuring the concentration of RBD utilizing UV-spectrophotometer. Table (3.3) demonstrates the findings. Except for the contacting duration, which was (2-70) minutes, the operating parameters were gained at their mean values.

Table (3.3 ): Operating parameters

parameters	Range or constant values
Initial dye amount (ppm)	100
pH	8
Voltage (volt)	20
Electric Current (Amp.)	3
Stirring speed (rpm)	200

### 3.9. Set up of Box-Willson Design

The parameters of operation are: time of electrolysis (X1), pH of solution (X2) and voltage (X3). where (Y) refers to the studied response .For three parameters the quadratic polynomial formula can be referred as follows:

$$Y = B_0 + \sum_{i=1}^k B_i X_i + \sum_{i=1}^k B_i X_i^2 + \sum_j \sum_j^k B X_i X_j + \varepsilon \quad (3.1)$$

$$Y = ( B_0 + B_1 X_1 + B_2 X_2 + B_3 X_3 + B_4 X_1^2 + B_5 X_2^2 + B_6 X_3^2 + B_7 X_1 X_2 + B_8 X_1 X_3 + B_9 X_2 X_3 ) \quad (3.2)$$

Y is the predicated magnitude,

B0-is the .constant, B1, B2, B3 are the linear coefficients.

Quadratic coefficients are B4, B5, and B6.

Cross-production coefficients are B7, B8, and B9, and k is the number of components; in this example, k equals three .

The number of experimental runs required to cover range the three parameters is:

$$N = 2^K + 2K + 5 \quad (3.3)$$

$$N = 2^3 + 2 * 3 + 5 = 19$$

The coded parameters ( $\alpha$ ) were between (-1.732) and (1.732), the range of real parameters for the system can be referred in Tables (3.4), (3.5) and (3.6), which refer the experimental Box-Willson design (*shahad,2018*).

Table (3.4): The range of design for parameters

Time (min)	PH	Voltage(volt)
2-80	4-12	15-25

Table (3.5): Set up the coded and the real parameters

Parameters	levels				
	-1.732	-1	0	1	1.732
X1= time (min)	2	18	41	64	80
X2= pH	4	6	8	10	12
X3= voltage (v)	15	17.1	20	22.9	25

Table (3.6):- Results of the studied responses

Coded parameters				Real parameters		
Run	A	B	C	Time (min)	Applied Voltage (Volt.)	pH
1	-1	-1	-1	18	17.1	6
2	1	-1	-1	64	17.1	6
3	-1	1	-1	18	22.9	6
4	1	1	-1	64	22.9	6
5	-1	-1	1	18	17.1	10
6	1	-1	1	64	17.1	10
7	-1	1	1	18	22.9	10
8	1	1	1	64	22.9	10
9	-1.732	0	0	2	20.0	8
10	1.732	0	0	80	20.0	8
11	0	-1.732	0	41	15.0	8
12	0	1.732	0	41	25.0	8
13	0	0	-1.732	41	20.0	4
14	0	0	1.732	41	20.0	12
15	0	0	0	41	20.0	8
16	0	0	0	41	20.0	8
17	0	0	0	41	20.0	8
18	0	0	0	41	20.0	8
19	0	0	0	41	20.0	8
20	0	0	0	41	20.0	8

For regression and graphical analysis of the findings, Minitab-17 was used. Based on the prior findings, the following objects were calculated:

1. Time of the electrolysis.
2. Reaction order study.
3. Removal efficiency.
4. Energy consumption.
5. Electrode consumption.
6. Final. pH.
7. final conductivity.
8. final applied current stable.
9. Optimization .

### **3.10. Analytical methods**

A number of analytical methods can be utilized for the analysis of dyes presented solution, such as UV-visible demonstrated in Fig. (3.5), absorption spectrophotometer method. The principle of spectrophotometry involves radiation being selectivity absorbed when passed through a specific wavelength. Beer–law Lambert's eq. (3.4) which may be utilized to determine an unknown amount of an analyte by measuring the quantity of light that a sample absorbs. If the absorbency coefficient is unknown, a working curve of absorbance  $A$  against concentration  $C$  (mg/l) produced from standards can be utilized to calculate the unknown amount (3.4).

$$q_A = K.C. + E. \quad (3.4)$$

Whereas:

$q_A$ : absorption of light at a specific wavelength,

$K$ : the coefficient of absorbance (linear relation's slope),

C.: dye concentration in liquid surrounding media.(mg/l),

And E: the linear relationship's intercept.

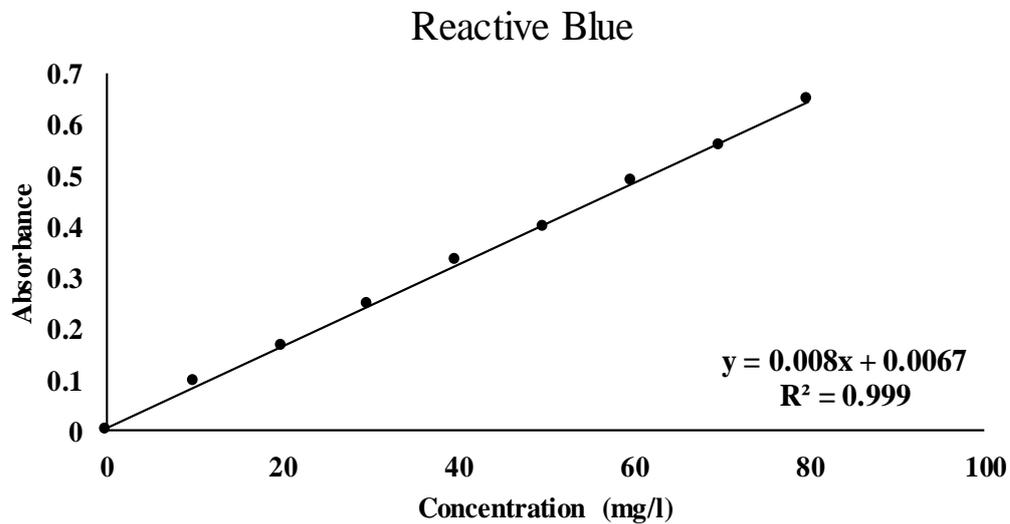


Figure (3.4): calibration curve of RBD



Figure (3.5): 1800 UV- Spectrophotometer

**3.11. Estimate of Removal efficiency**

Each electrocoagulation cell sample was utilized using a 1800 UV-Spectrophotometer, the data was analyzed. The removal efficiency of RBD from wastewater could be estimated using the following equation (3.5).

$$R\% = \frac{C_0 - C}{C_0} * 100 \quad (3.5)$$

R% = RBD removal efficiency,

$C_0$  = initial concentration of RBD.

C = concentration of RBD at any time.

## **Chapter Four**

### **Results and Discussions**

#### **4.1. Introduction**

This chapter includes two parts, the first part, the preliminary experiment to estimate the reaction time to remove the dye reactive blue (RBD). The effect of power supplied mode was investigated and compared using two modes; Constant Current Mode (CCM) (at 3 A) and Constant Voltage Mode (CVM) (at 20 volt) along the period of experiments, and design of the electrodes and the selection of the best arrangement. The second part, the impact of three parameters (reaction time, pH, and applied voltage on the removal efficiency) are studied.

#### **4.2. The Preliminary Experiment**

So as to identify the reaction time for the electrochemical process, the variable will take be at their mean values, meaning that the pH of 8, the current is equal to 3 amperes and the voltage is 20 volt at 200 rpm of stirrer speed and then the effect of power supplied mode was investigated and compared using two modes; Constant Current Mode (CCM) and Constant Voltage Mode (CVM) under the impact of the operating variables to investigate which of these modes possess higher performance to remove RBD from simulated wastewater.

##### **4.2.1. Removal efficiency:**

Table (4.1) explains the findings of the comparison of RBD amount and removal efficiency for both modes along the period of experiments with the conditions of other variables.

Table (4.1). Findings of both experiments done for RBD removal efficiency.

Run	Electrolysis time (min)	CCM	Experiment	CVM	Experiment
		Dye Conc. (ppm)	Dye removal efficiency %	Dye Conc. (ppm)	Dye removal efficiency %
0	0	100	0.00	100	0.00
1	2	73.411	26.59	53.2	46.80
2	5	61.529	38.47	35.553	64.45
3	10	55.411	44.59	30.965	69.04
4	20	29.059	70.94	20.612	79.39
5	30	18.235	81.77	10.494	89.51
6	40	5.529	94.47	3.61	96.39
7	50	1.65	98.35	2.965	97.04
8	60	1.411	98.59	2.564	97.44
9	70	1.1765	98.82	2.259	97.74

The minimizing of RBD concentration with time for both cases is demonstrated clearly in Fig (4.1) which reveals the effective configuration of the present design of electrodes for both cases that provided higher elimination of RBD-pollutant regardless of the operation mode whether it is CVM or CCM.

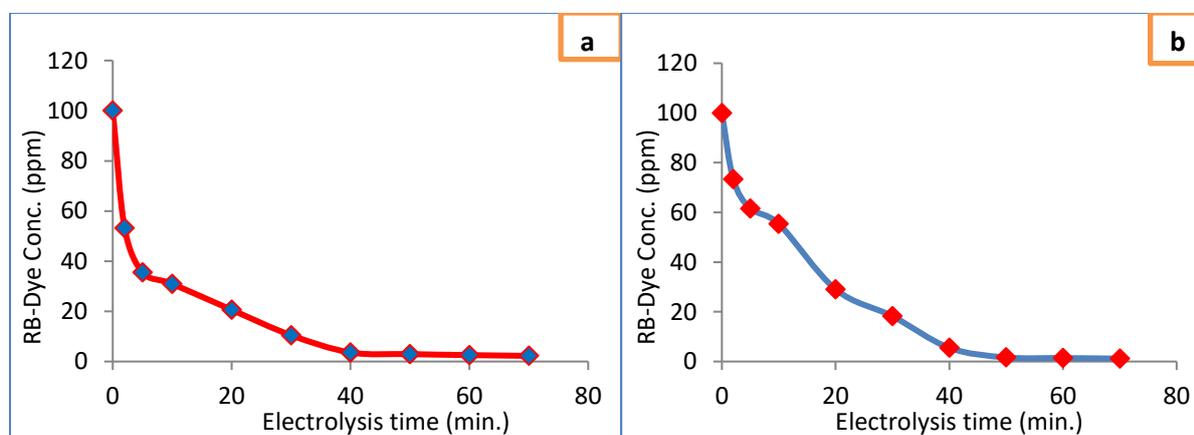


Figure (4.1): Amount decay of RBD with time. (a) CVM-mode. (b) CCM-mode at mean values of variables (100 ppm, pH=8, 20 volt, 200rpm).

Figure (4.2) explain the behavior of RBD removal efficiency for both cases of CVM and CCM which demonstrate that the removal efficiency of RBD improved with increasing electrolysis time until it reached the saturation status. The increment of removing response with time occurred since the adsorption process

between the RBD-pollutant and the electro-coagulant adsorbents that formed since the redox reactions. The continuous supplying of current at constant voltage or voltage at constant current will cause to release aluminum ions from anode and hydroxide ions at the cathode that will assist to generate adsorbents, consequently, the removing process will proceed. Moreover, the perforating action of entire electrodes will increase the releasing of hydrogen and oxygen soft bubbles that will assist to rise pollutant to the solution surface then removed by screaming process. As observed, the CCM process was more effective than CVM process which means that the significant elimination of RBD could be attained in the case of a steady current of 3 Amps throughout the experiment while the applied voltage remained constant will alter according to the observation of electrodes consumption and owing energy consumption. The mathematical correlation of RBD removal efficiency for both modes are explained in Eqs. (4.1) and (4.2) The highest removal efficiency of RBD and the real consumption of electrodes for both cases are explained in Table (4.2).

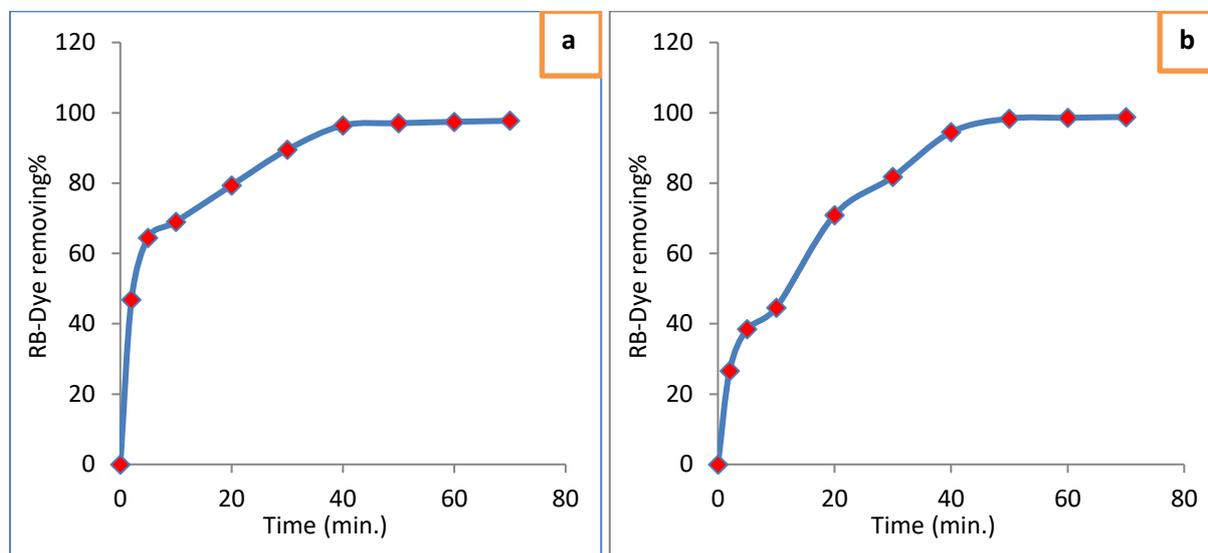


Figure (4.2 ) RBD removing efficiency with time. (a) CVM-mode. (b) CCM-mode at mean values of variables (100 ppm, pH=8, 20 volt, 200rpm).

$$\text{CVM: } -0.0289 (\text{time})_2 - 2.8694 (\text{time}) + 68.052 \quad R^2: 0.81 \quad (4.1)$$

$$\text{CCM: } -0.0291(\text{time})_2 - 3.1891(\text{time}) + 85.419 \quad R^2:0.97 \quad (4.2)$$

Table (4.2): Findings of CVM and CCM modes for RBD removing and the consumption electrodes and energy

Operation modes	Highest removing efficiency %	Electrodes consumption . (g)	Energy cons.(kW/m3)
CVM	97.74	1.4	10.99
CCM	98.82	0.32	2.8

As seen from data presented in Table (4.2) that the value of removal efficiency approximately are closed together, but the difference is observed clearly in the values of the energy and electrodes consumption where the constant current mode (CCM) was the lowest. This finding indicates that the CCM operating mode was more cost-effective than the CVM operating mode.

#### 4.2.2. FT-IR Analysis:

The characterization of the dried sludge was done utilizing FT-IR test where the spectra in the ranging of  $4000\text{--}500 \text{ cm}^{-1}$  on behalf of the rare materials, as demonstrated in Fig .(4.3). FTIR spectral study established the damagingly charged efficient sets (amine, carboxyl, and hydroxyl) being found on the surface of the magnetite. The comprehensive band at  $3454.62\text{--}3097.78 \text{ cm}^{-1}$  in unadulterated bark powder is recognized since hydroxyl (O-H), (C-H), and (N-H) stretching of amines and amides. The (C-N) and silicone compounds gave the symmetrical bending in the area  $2472.21\text{--}1754.15 \text{ cm}^{-1}$ . The conjugation of (C=O) with two aromatic rings and (C-O) are presented at the peaks of  $1641.68$

and  $1398.44\text{ cm}^{-1}$ . The peak of  $1519.95\text{ cm}^{-1}$  is mentioning in the aromatic vibration (C-C) of lignin and the peak of  $1330.93\text{ cm}^{-1}$  indicates the aromatic rings, while  $1072.48\text{ cm}^{-1}$  is related to the alkyl amine. The  $(\text{CH}_3)$  angular compound demonstrated in the band current underneath  $771.55\text{ cm}^{-1}$ . The rest of peaks refers to other compounds such  $\nu$  and beta-rings. This observation of FT-IR analysis is since the chemical compositions of RBD and electrodes utilized in the present study.

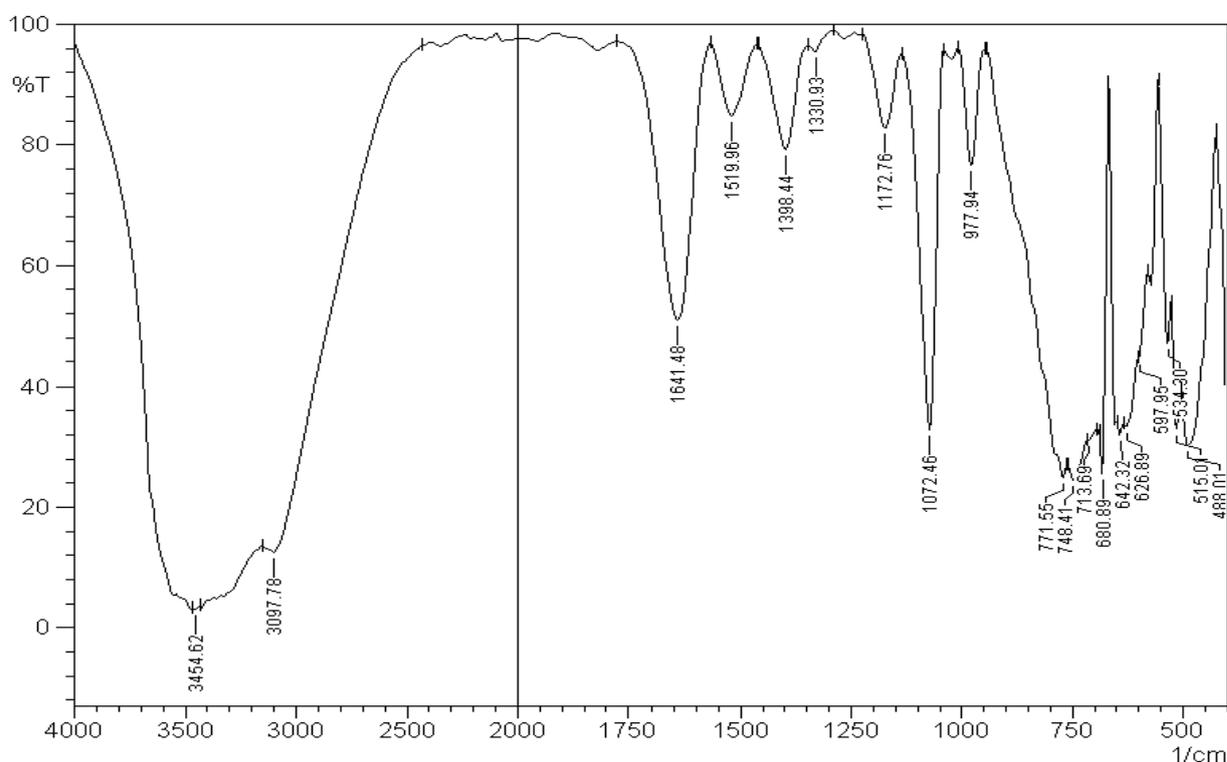


Figure (4.3) FT-IR analysis result of the sludge

### 4.2.3. Thermodynamic Study

For both CVM and CCM, the thermodynamic parameters of entropy (S), Gibbs energy (G), and heat of reaction (H) of the remediation process were estimated as demonstrated in Tables (A1 to A4:Appandix A) depending on the variation of solution temperature observed along the periods of both experiments and findings gained from Fig (4.4) depending on Eqs. (2.14) and (2.15), plotting on the values

of (log Kd) against (1/T), and then evaluating the value of the H from the Slope ( $-\Delta H/ 2.303 R$ ), as seen in Figure (4.4). The values of entropy ( $\Delta S$ ) and Gibbs energy (G) are estimated in accordance with Eqs. (2.16) and (2.17).

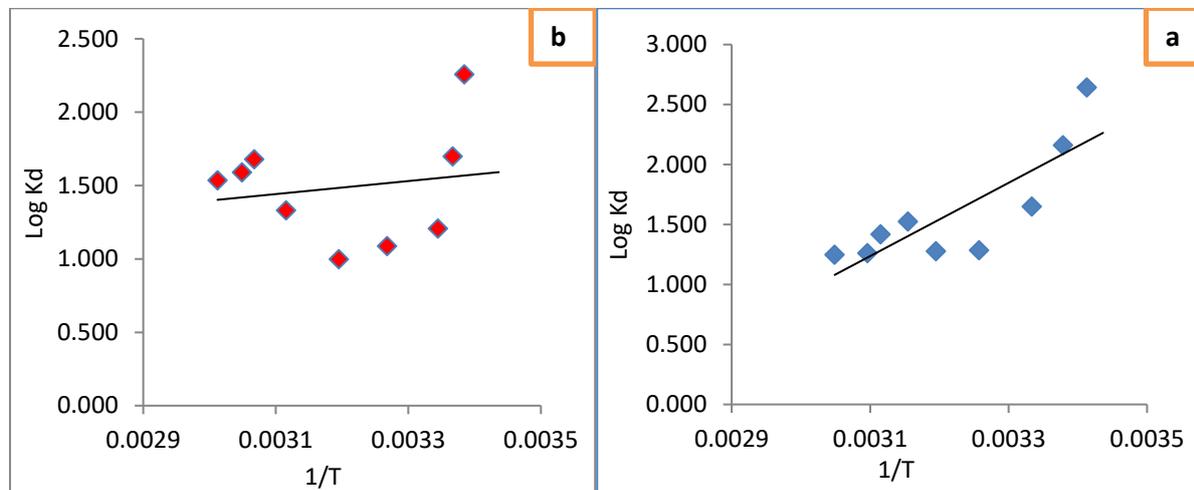


Figure (4.4) Estimation of the heat of enthalpy depending on the slope magnitude  
(a) CVM. (b) CCM at mean values of variables (100 ppm, pH=8, 20 volt, 200rpm)

As observed, the positive sign of the heat of adsorptions ( $\Delta H$ ) and the entropy ( $\Delta S$ ) indicates that the operation is endothermic and ascribed to a random irregularity at the solid-liquid. The heat required for the endothermic process could be provided by the electrical current that applied on the electrodes. Whereas the negative value of the Gibbs energy ( $\Delta G$ ) refers to the spontaneous nature and more favorable adsorption of the remediation process.

#### 4.2.4. Kinetics Studies

Several kinetic models were proposed to describe the behavior of the reaction occurring in the reactor, including the first-order and second-order formula depending on the general formula of design Eq.(4.3) to refer the kinetic of RBD degradation by electrocoagulation technology.

$$\frac{dC}{dt} = -kC_t^n \quad (4.3)$$

$$\ln \frac{C_t}{C^0} = K_1 t \quad (\text{first order}) \quad (4.4)$$

$$\frac{1}{C_t} - \frac{1}{C^0} = K_2 t \quad (\text{second order}) \quad (4.5)$$

where t, k, n, and C referring to the electrolysis time, the rate constant, reaction's order, and the RBD concentration. Fig (4.5) , (4.6) and Tables (A5 to A7:Appandix A) explain the kinetic investigation of dye removal from simulated wastewater for both cases of CVM and CCM. The constant voltage mode (CVM) obeys the second order kinetic of reaction while the operation mode of constant current (CCM) obeys the first order kinetic reaction compared to the CVM system which means that the CCM system is more reliable and effective than CVM system.

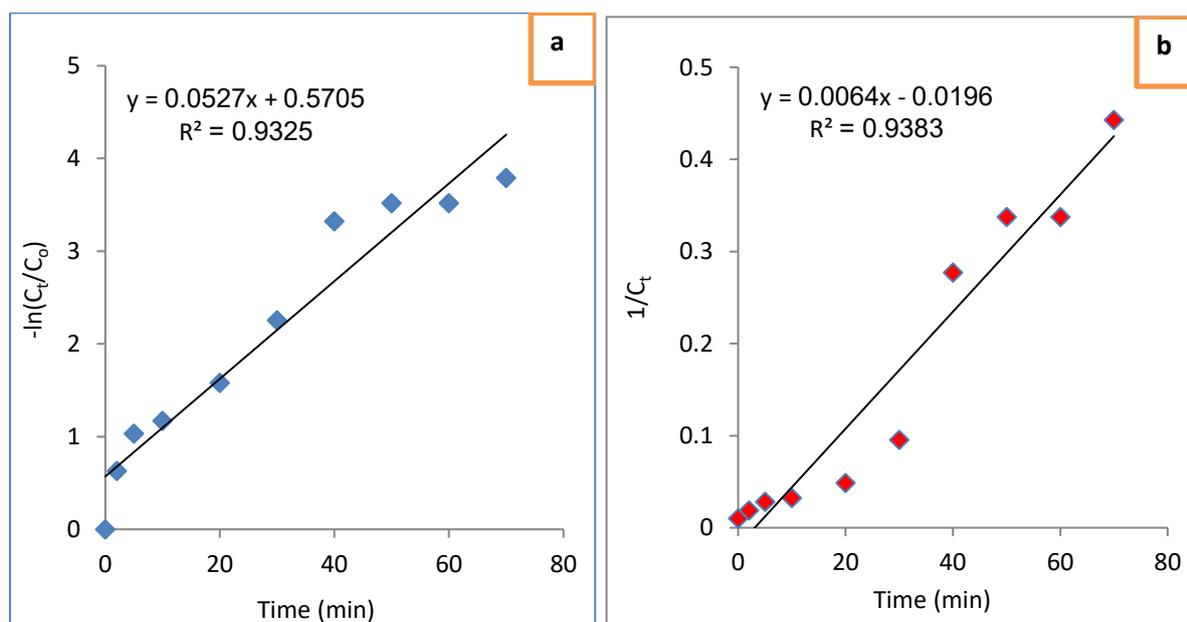


Figure (4.5). Findings of kinetic study for CVM system. (a) First order reaction (b) Second order reaction at mean values of variables (100 ppm, pH=8, 20 volt, 200rpm)

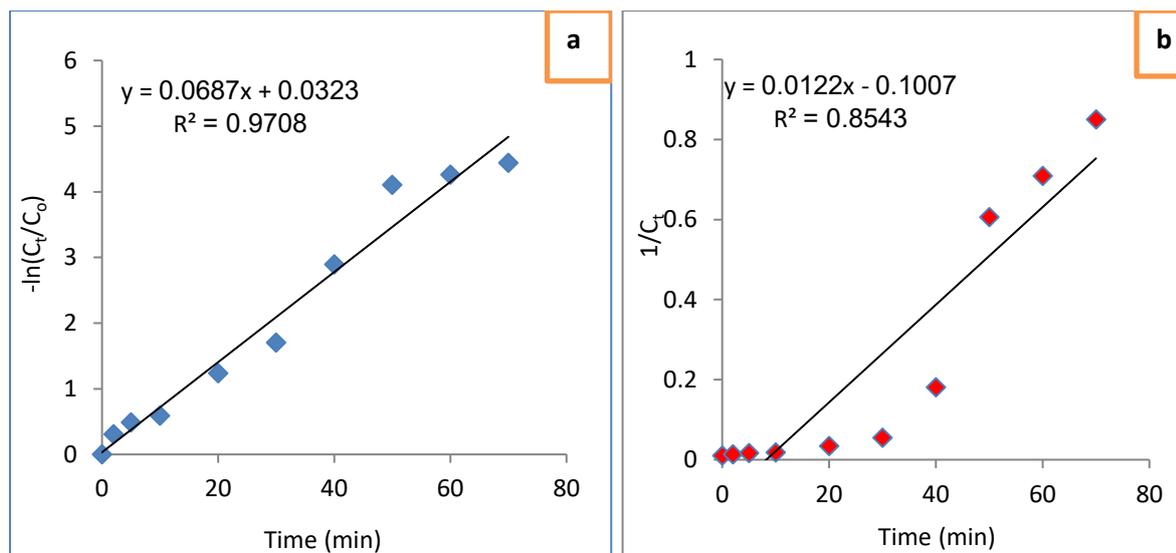


Figure (4.6) Findings of kinetic study for CCM system. (a) First order reaction  
(b) Second order reaction at mean values of variables (100 ppm, pH=8, 20 volt, 200rpm)

### 4.3. Best design of electrodes

To choose the best design of the electrodes, three experiments were conducted for each particular design experiment at the mean values of the three parameters, then the removal efficiency was measured to choose the best design of electrodes for remediation. The first experiment (A) was to design three non-perforated electrodes and one perforated electrode located toward the anode electrode, the second experiment (B) was three non-perforated electrodes and one perforated electrode located toward the cathode electrode, which compared with in the third experiment (C), the outer plates are not perforated while the entire two plates are perforated, as demonstrated in Table (4.3).

Table (4.3) Best design of electrodes

Electrode modes	Dye Amount (ppm)	Removing efficiency %
A	8.35	91.65
B	1.88	98.12
(C) Best design	1.18	98.82

Table (4.3) proved that the highest removal efficiency was achieved when design (C) consisting of four plates the outer plates are not perforated while the entire two plates are perforated.

#### 4.4. Main Statistical Analysis of Electrocoagulation Responses

The purpose of this study is to investigate into the parameters that have an impact on the responses . A nonlinear regression analysis was utilized to relate the experimental data, and a second order polynomial was predicted that properly correlates the response in terms of the operating variables. The model and experimental data were then statistically evaluated. The model would eventually provide optimal operating conditions that yielded the highest removal efficiency (*Abd Ul-Shaheed,2018*).

The empirical correlations between the particular responses and the operating factors provided in Table (3.2) that is a form of response surfaces methodology (RSM), which has a significant relevance in studying systems where several operating variables impact responses also the effective response optimization, were calculated utilizing four plate-electrodes made of aluminum. The mathematical correlations of responses could be gained utilizing Eq(3.1) .

Tables (3.5) and (3.6) list the specifics of the planned experiments utilized to calculate the empirical correlations of the specific parameters. For regression and A graphical representation of the findings Minitab-17 was employed. ( *AlJaberi (d),2018*).

#### 4.4.1. The Statistical Analysis of RBD Removal efficiency Response

##### 4.4.1.1. Impact of Reaction Time

As observed in Fig. (4.7), the RBD removal efficiency improved with increasing the reaction time at the mean values of the voltage applied solution pH where the reaction time is a very essential parameter in any electrochemical remediation technique. According to the range chosen in this study (2-80 min), the removal efficiency of RBD had clearly increased to reach more than 85% within 60 min. Then, it tends to get slightly minimized due to the a decrease in the formation of the absorbents material. Thus, the sites on the surface of the absorbent material become insufficient to adsorb the RBD (. So, the greater the reaction time, the more absorbent materials, that is, the absorbents material that adsorbs the dye pollutant, which leads to a decrease in the amount of the dye in the solution and a higher removal efficiency obtained (*AlJaberi (a), et al,2020*).

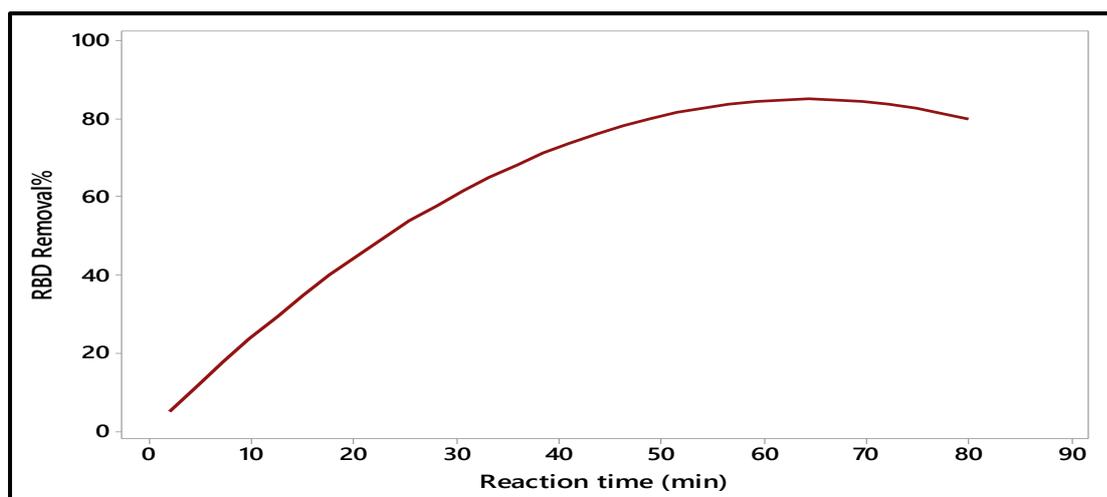


Figure (4.7): Impact of reaction time on 100 ppm RBD/l removal efficiency of from simulated wastewater (Applied voltage= 20 volt, pH 8,Nacl=2g,200 rpm)

The following formula refer to the impact of the time on the removal efficiency in the case mean values of other variables:

$$Y = 2.642 x_1 - 0.02055 x_1^2 + C \quad R^2 = 94.7\% \quad (4.6)$$

#### 4.4.1.2. Impact of Initial pH on the RBD Removal

The value of solution pH affects directly the removal efficiency of pollutants from wastewater; therefore, this study investigated the impact of solution pH value on the ability of the present reactor to remove RBD from the simulated wastewater. The range of pH selected covered the acidity and base nature of wastewater. As noted in Fig. (4.8), the lower removal efficiency the more basic solution while the highest removal was obtained at value less than the neutral medium. So, this type of dyes is removed efficiently at the acidic medium (*AlJaberi, Mohammed (c) ,2018; Andrade ,2007*).

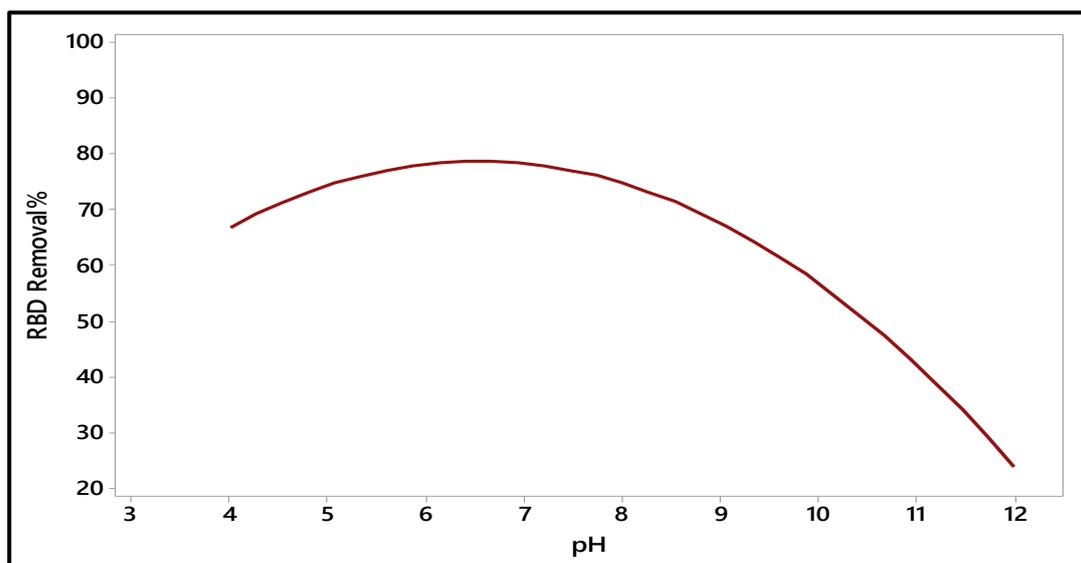


Figure (4.8 ):The impact of pH on the RBD removing efficiency of 100 mg RBD/l of simulated wastewater (reaction time= 41 min, and voltage applied=20 volt ,Nacl=2g ,200rpm)

The following formula refer to the impact of the pH on the removal efficiency in the case mean values of other variables :

$$Y = 24.05 x_2 - 1.837 x_2^2 + C \quad R^2 = 94.1\% \quad (4.7)$$

#### 4.4.1.3. Impact of the Applied Voltage

Electrocoagulation reactors are extremely dependent in their operation on the voltage or current applied (*AlJaberi (a), et al, 2020; AlJaberi (d), 2018*).

The present work investigates the impact of varying the value of voltage applied on the RBD removal efficiency.

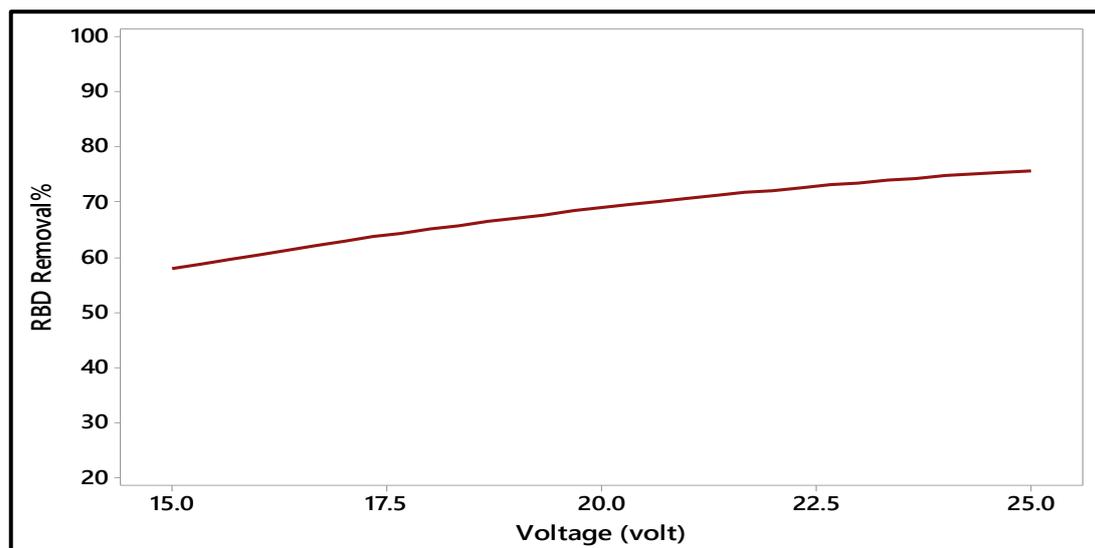


Figure. (4.9): The impact of voltage applied on the RBD removing efficiency of 100 mg RBD/l of simulated wastewater (reaction time= 41 min, and pH 8 ,Nacl=2g, 200 rpm)

The following formula refer to the impact of the voltage on the removal efficiency in the case mean values of other variables :

$$Y = 5.121x_3 - 0.08379x_3^2 + C \quad R^2 = 92.1\% \quad (4.8)$$

Figure (4.9) revealed that the treatability of the present design of electrocoagulation reactor affected by the value of the voltage applied. As seen, the removal efficiency improved with increasing the voltage applied at the mean magnitudes of reaction time and solution pH. The RBD removal increased from 58% to about 80% when the applied voltage raises from 15 to 25 volt. This could be interpreted that the continuous raising of voltage applied will increase the formation of various ions required to form adsorbents. But the optimum value

should be taken into consideration to prevent the excess consumption of electrodes (*AlJaberi (a), et al, 2020; AlJaberi (c), et al, 2020; AlJaberi (d), 2018*).

#### **4.4.2. The Statistical Analysis of Energy Consumption Response**

Figures (4.10) ,(4.11) and (4.12) depicts the direct relationship between energy consumption responses and the operating variables that could contribute to the enhancement of electrically conductivity and therefore the energy consumed to some extent depending on the electrolyte concentration, solution. The impact of contacting time on the amount of energy consumption is comparable to the change in the value of the voltage applied. In the event of high levels of the provided voltage of the pollutant electrochemical cell, it is obvious that the quantity of energy consumed grows with the growing contacting time.

Because since existence of electrical conductivity, that substantially helps to reducing the energy amount used, the basic solution has continued to utilize the least energy amount. This responses, on the other hand, depends upon the voltage amount that has been applied on the systems, as the energy amount consumed is responsible for the increase in voltage, that seems appropriate given the numerical law relating to the computation of energy consumed in the electrocoagulation reactors depending on the experimental period.

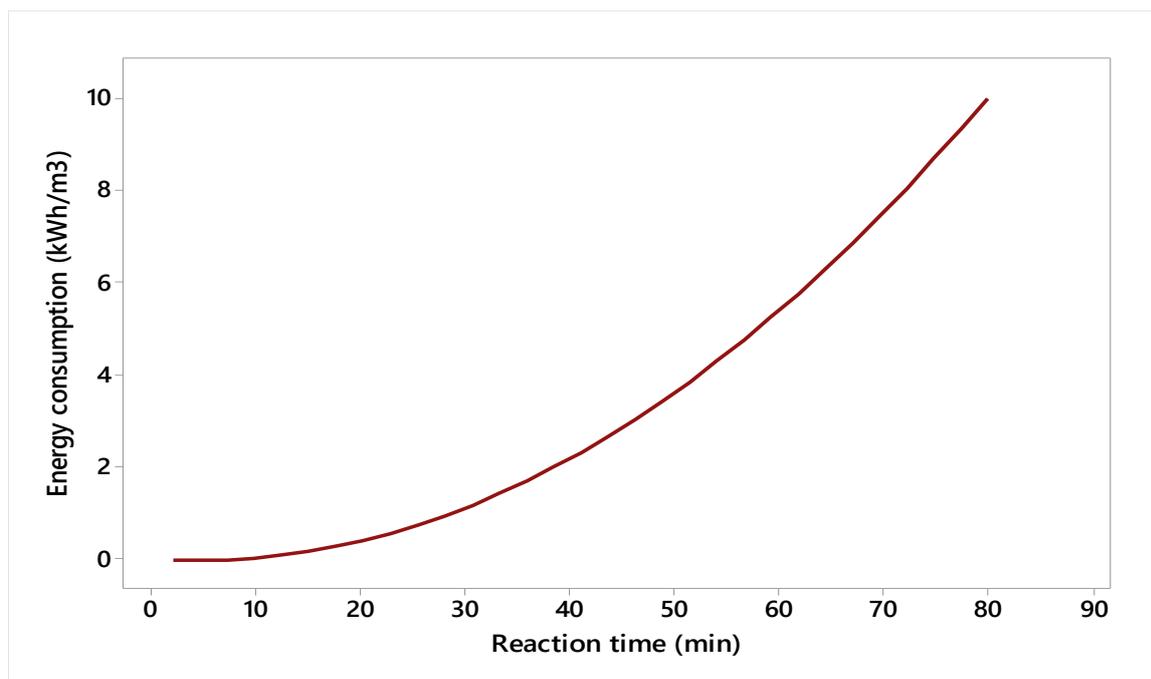


Figure. (4.10): The impact of reaction time applied on the energy consumption of simulated wastewater (ph=8, voltage=20 volt ,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

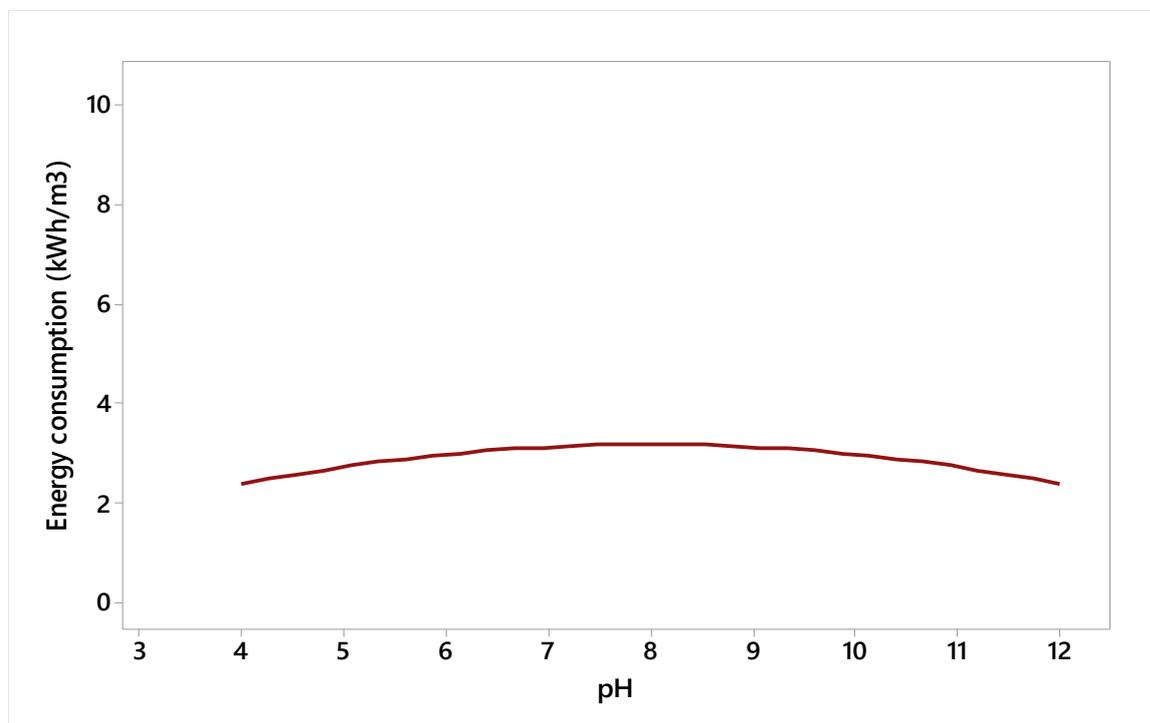


Figure. (4.11): The impact of Ph applied on the energy consumption of simulated wastewater (reaction time= 41 min, voltage=20 volt ,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

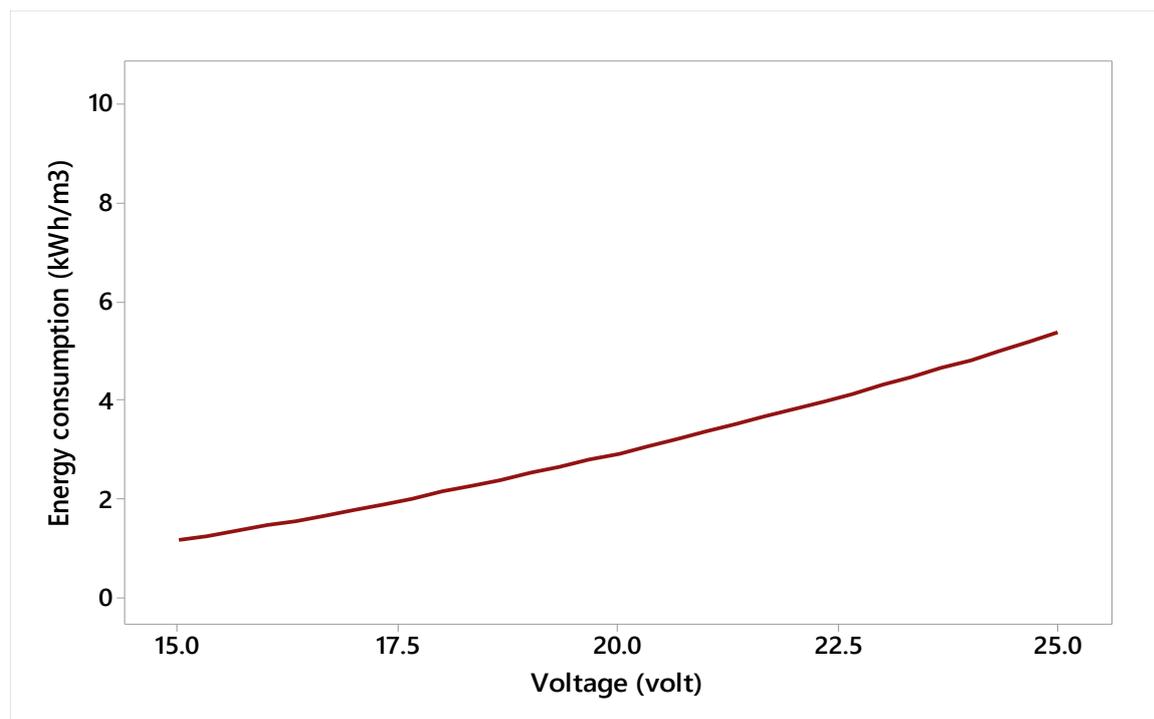


Figure. (4.12): The impact of voltage applied on the energy consumption of simulated wastewater (reaction time= 41 min, pH 8 ,NaCl=2g and 200 rpm) for 100 mg RBD/l removal

The following correlations refer the to impacts of the operating variables on the consumption value of energy:

$$Y = -0.01577x_1 + 0.0017x_1^2 + C \quad R^2 = 87.8\% \quad (4.9)$$

$$Y = 0.797 x_2 - 0.049 x_2^2 + C \quad R^2 = 52.5\% \quad (4.10)$$

$$Y = -0.1254 x_3 + 0.0136 x_3^2 + C \quad R^2 = 58.6\% \quad (4.11)$$

#### 4.4.3. The Statistical Analysis of Electrodes Consumption Response

The relation between real consumption of electrodes and operating variables is clearly demonstrated in Figs. (4.13) ,(4.14) and (4.15). Since the anode's consumption is high and is closely related to the amount of RBD in the solution when the electrolysis was started.

While the acidic solution requires less consumption of the anode than the basic solutions, the aluminum consumption lasts longer. The duration of the experiment is much greater than that of the hydroxyl ion in the high basic solutions. This response appears to be more consistent with its different behavior when the current values vary over the duration of the experiment and the consumption of aluminum remains the more as supplied current increased. This is a natural result due to continuous ion releasing and dissolution of electrodes and their consumption accordingly.

The pH influences the electrodes' consumption response, with consumption being lower when the pH is acidic, and the behavior of this response varies during the the experiment with changing values of the parameters. But the persistence of the electrode's consumption is associated with an increase in the applied voltage that leads to the release of ions and dissolution of the electrodes, thus increasing their consumption. The consumption of the electrodes is directly related to the increase of the voltage applied and pH.

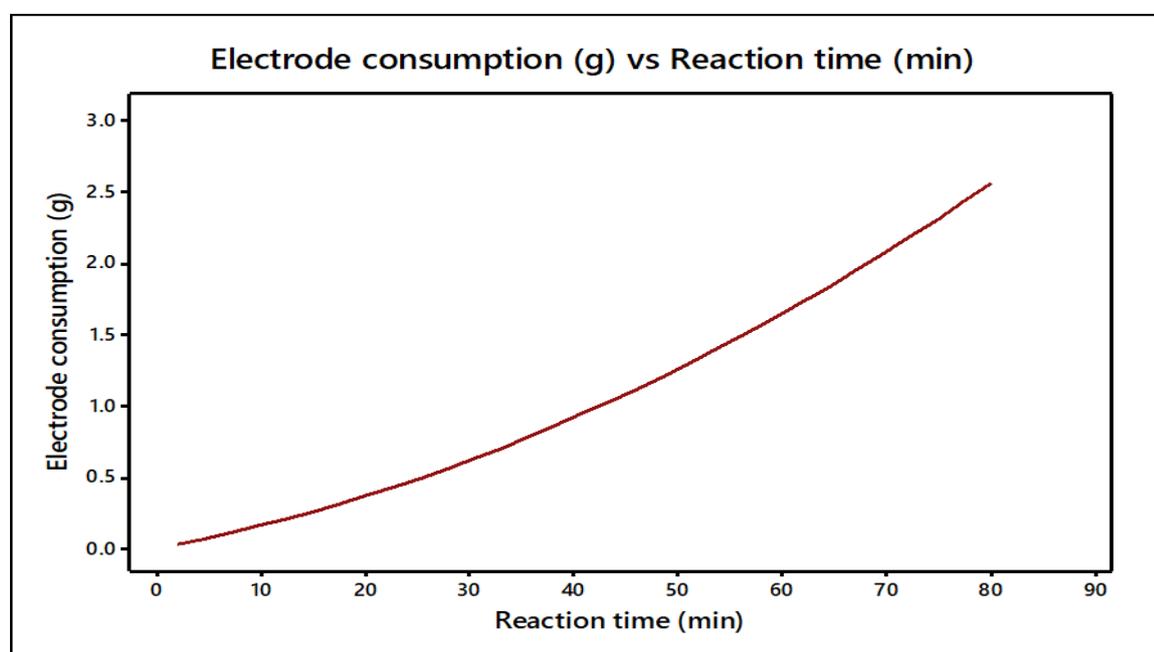


Figure. (4.13): The impact of reaction time applied on the electrode consumption of simulated wastewater (ph=8, voltage=20 volt ,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

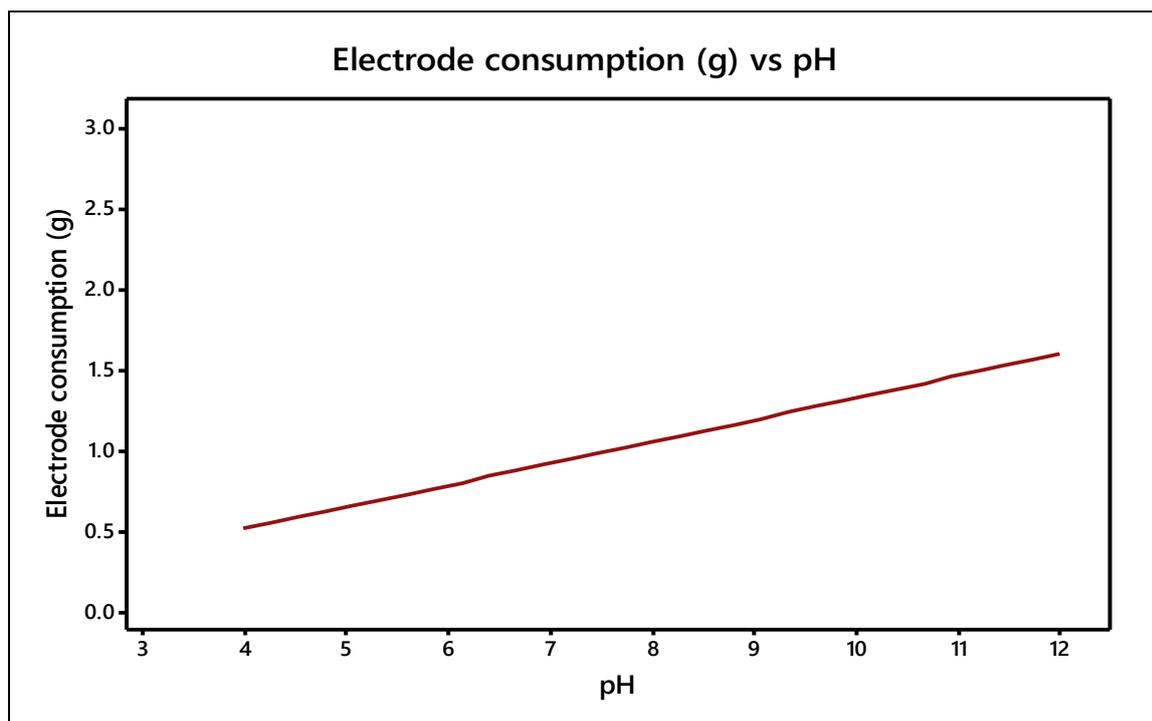


Figure. (4.14): The impact of Ph applied on the electrode consumption of simulated wastewater (reaction time= 41 min, voltage=20 volt ,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

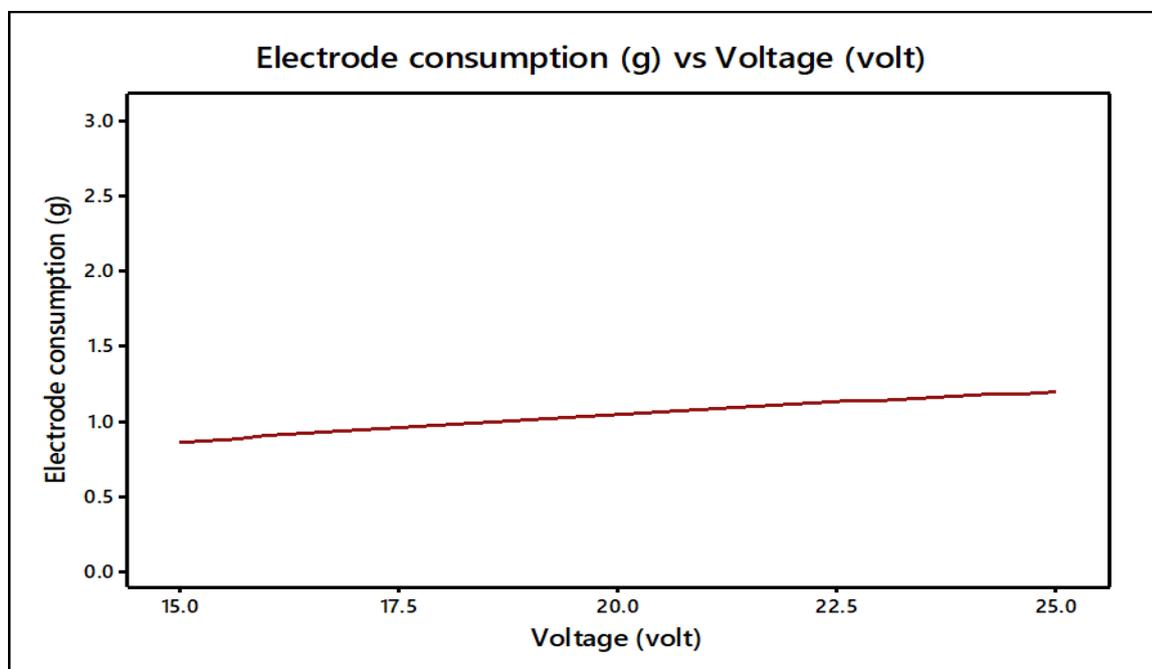


Figure. (4.15): The impact of voltage applied on the electrode consumption of simulated wastewater (reaction time= 41 min, pH 8 ,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

The following correlation refer to the impacts of the operating variables on the consumption value of electrodes:

$$Y = 0.01385 x_1 + 0.000227 x_1^2 + C \quad R^2 = 81.9\% \quad (4.12)$$

$$Y = 0.1248 x_2 + 0.00044 x_2^2 + C \quad R^2 = 67.2\% \quad (4.13)$$

$$Y = 0.07104 x_3 - 0.000930 x_3^2 + C \quad R^2 = 62.0\% \quad (4.14)$$

#### 4.4.4. The Statistical Analysis of Final pH Response

The impact of increasing the electrolysis time on the behavior of the contaminated solution ultimate pH in the electrolysis reactor is demonstrated in Figs. (4.16), (4.17) and (4.18). The quantity of free hydroxyl ion and its influence on pH, as well as the usage of the same ion to create components of various aluminum hydroxides, induce a rise in the amount of free hydroxyl ion.

The initial pH value influences the ultimate pH value, thus when the pH value is acidic, the pH level tends to rise, and when the pH value is basic, the pH level tends to fall. Throughout the experiment, the voltage supplied had a noticeable influence on increasing the pH value until about an hour later, as the value began to drop as the amount of hydrogen bubbles and oxide layer generated on the cathode surface increased.

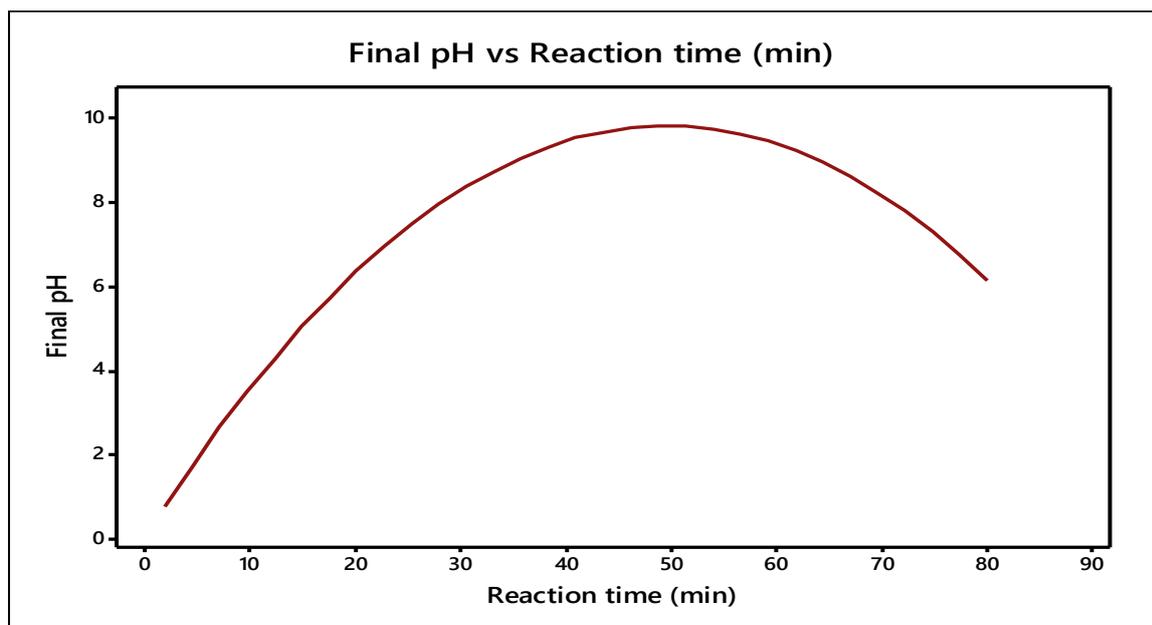


Figure. (4.16): The impact of reaction time on the final Ph of simulated wastewater (voltage =20 volt, pH 8 ,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

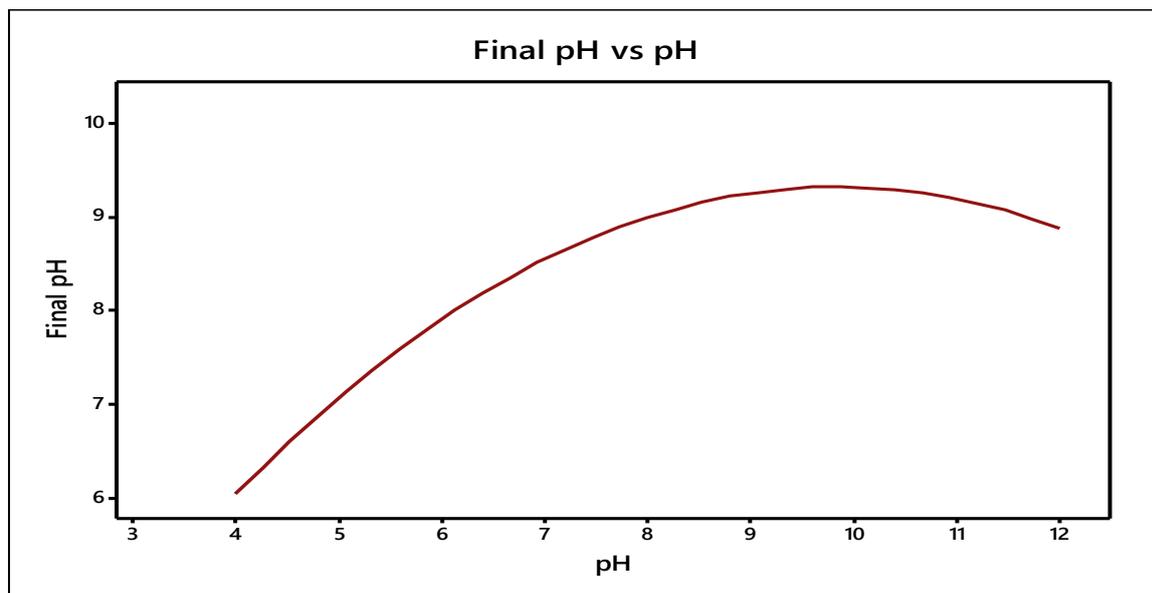


Figure. (4.17): The impact of Ph on the final Ph of simulated wastewater (reaction time=41 min ,voltage =20 volt,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

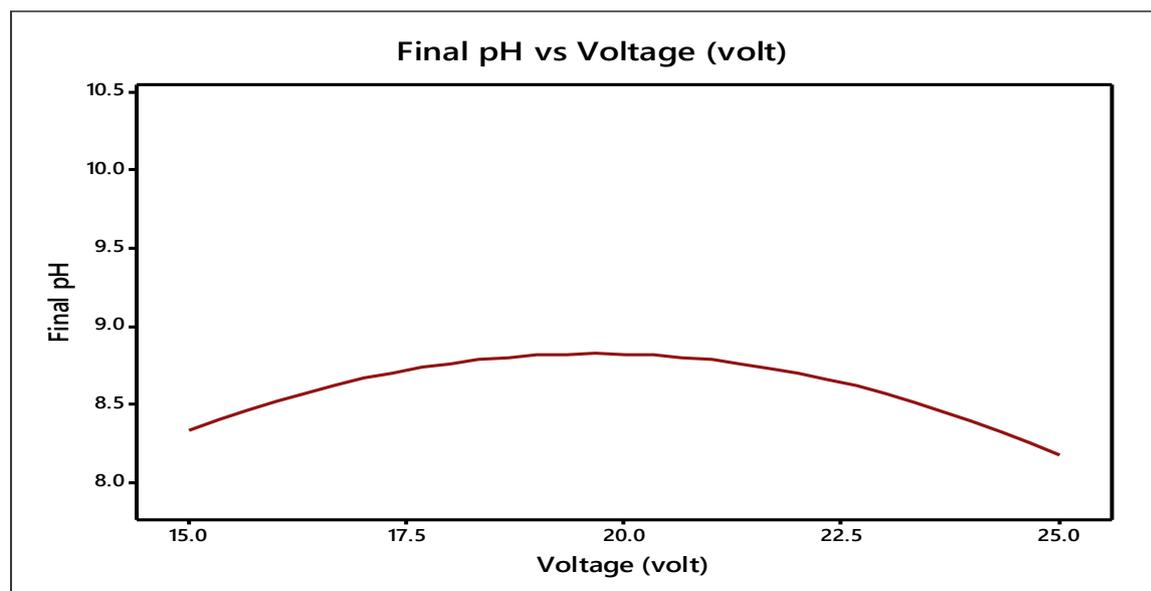


Figure. (4.18): The impact of voltage applied on the final Ph of simulated wastewater (reaction time= 41 min, pH 8 ,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

The following correlations refer the to impacts of the operating variables on the final Ph:

$$Y = 0.3957 x_1 + 0.003985 x_1^2 + C \quad R^2 = 93.3\% \quad (4.15)$$

$$Y = 1.894 x_2 - 0.09618 x_2^2 + C \quad R^2 = 99.1\% \quad (4.16)$$

$$Y = 0.8982 x_3 - 0.02286 x_3^2 + C \quad R^2 = 99.5\% \quad (4.17)$$

#### 4.4.5. The Statistical Analysis of the Final Conductivity Response

Figures. (4.19) ,(4.20) and (4.21) shown the impact of operating variable on the behavior of the final electrical conductivity response during the dye RBD-contaminated water remediation process. The conductivity increased with the increase of electrolysis time due to the continuous releasing of different ions from the electrodes ,then it decreased due to the formation of oxide layer.

The impact of pH on conductivity may also be demonstrated. As the pH is acidic, the conductivity increases, taking into account the impact of voltage and

time for electrolysis. This is owing to the fact that hydroxyl ions were readily available during the experiment.

There seems to be a small variation in the value of the voltage source, where the greater the voltage, the greater the conductivity, and conversely at low voltage values, due to the abundant supply of various ions produced once the current value is continued to increase, and the conductivity slightly increasing as a result.

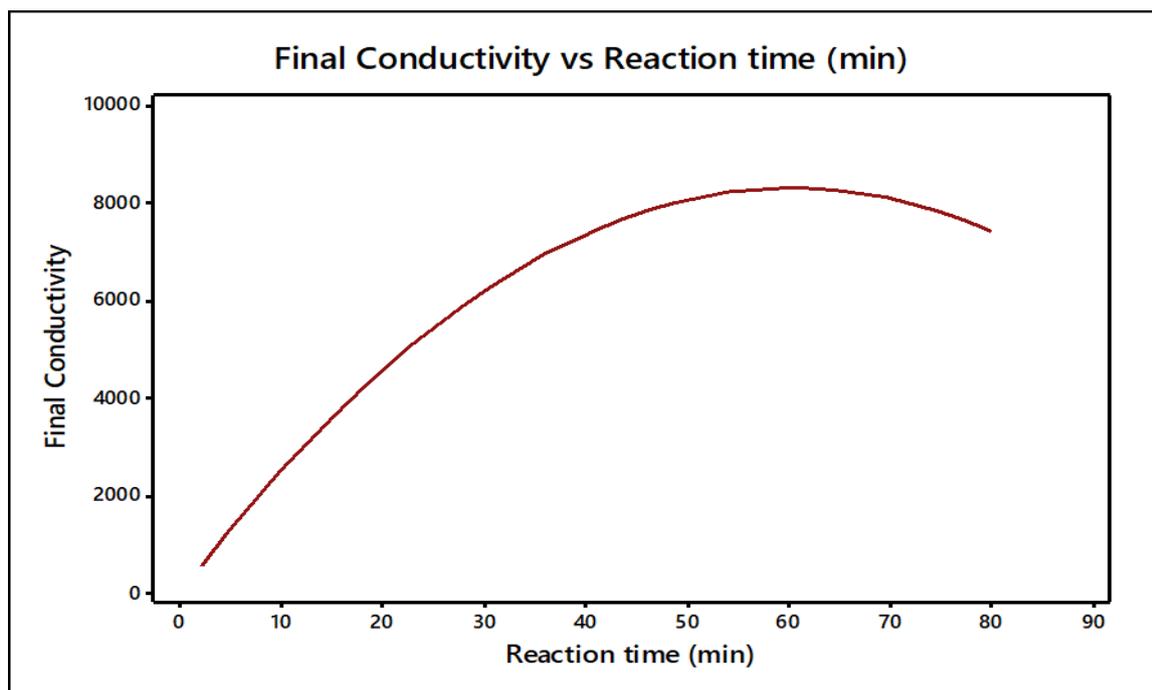


Figure. (4.19): The impact of reaction time on the final conductivity of simulated wastewater (voltage =20 volt, pH 8 ,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

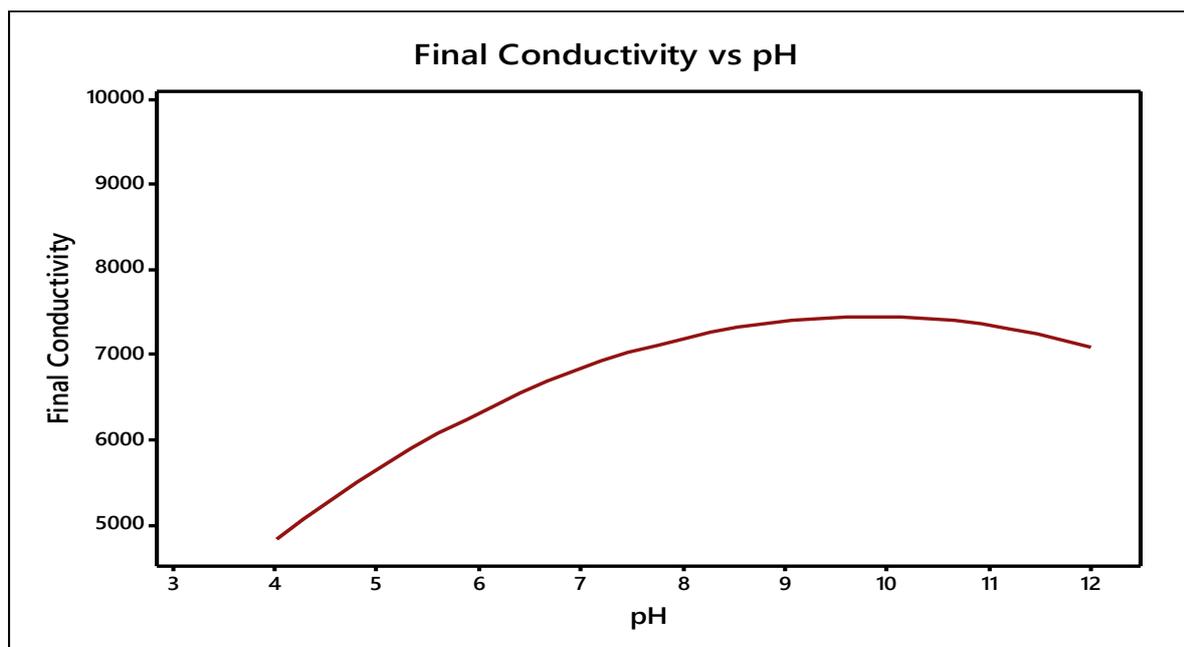


Figure. (4.20): The impact of Ph on the final conductivity of simulated wastewater (reaction time=41 min, voltage =20 volt,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

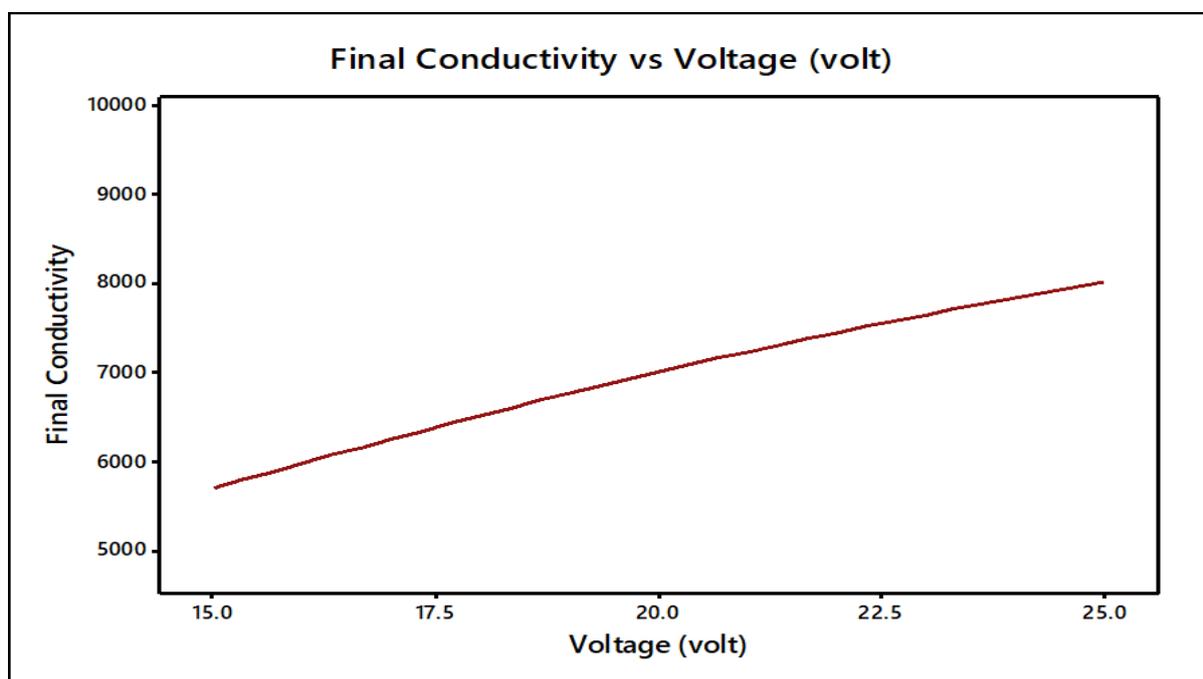


Figure. (4.21): The impact of voltage on the final conductivity of simulated wastewater (reaction time=41 min, Ph =8 ,Nacl=2g and 200 rpm) for 100 mg RBD/l removal

The following correlations refer the to impacts of the operating variables on the Final Conductivity:

$$Y = 274.9 x_1 - 2.278 x_1^2 + C \quad R^2 = 95.3\% \quad (4.18)$$

$$Y = 1516 x_2 - 77.19 x_2^2 + C \quad R^2 = 95.7\% \quad (4.19)$$

$$Y = 467.8 x_3 - 5.893 x_3^2 + C \quad R^2 = 97.25 \quad (4.20)$$

#### 4.4.6. The Statistical Analysis of the Final Stable Current Applied Response

As the contaminated solution is electrolyzed in the electrolysis reactor, the value of the final stable current response increased sharply with the reaction time, as illustrated in Figures. (4.22) ,(4.23) and (4.24).

Since of the number of ions necessary to create the adsorbent materials and the voltage's stability differential between the two selected electrodes, basic solution consume less current as opposed to acidic solution, which have a higher magnitude of this response. Since of the quantity of ions produced by the solution , the relation between voltages and pH is straightforward.

It was discovered that there was a clear relation between the response value and the amount of voltage applied during the remediation period; the greater the voltage, the higher the electric current's ultimate value.

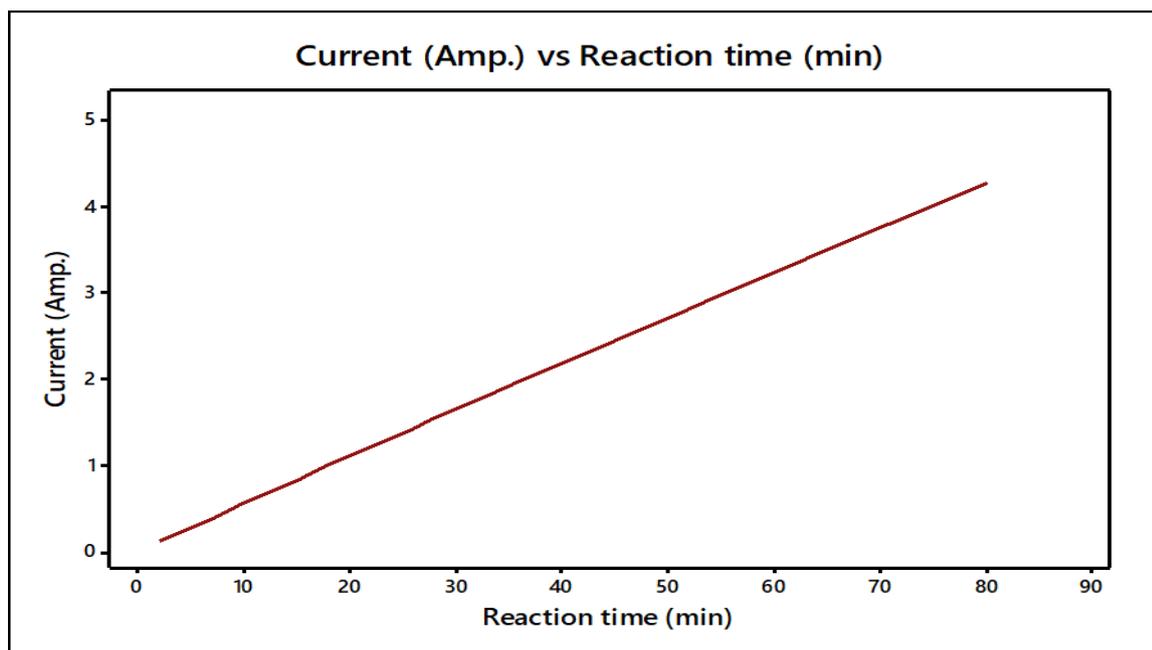


Figure. (4.22): The impact of reaction time on the current applied of simulated wastewater (Ph =8 ,voltage =20 volt, Nacl=2g and 200 rpm) for 100 mg RBD/l removal

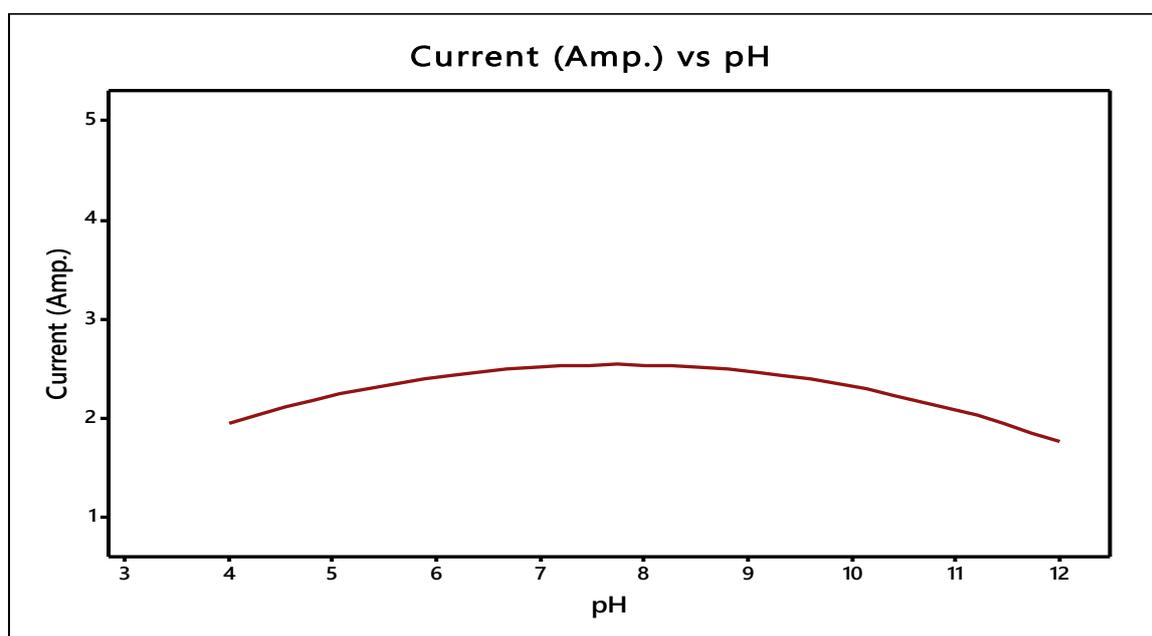


Figure. (4.23): The impact of Ph on the current applied of simulated wastewater (reaction time =41 min ,voltage =20 volt, Nacl=2g and 200 rpm) for 100 mg RBD/l removal

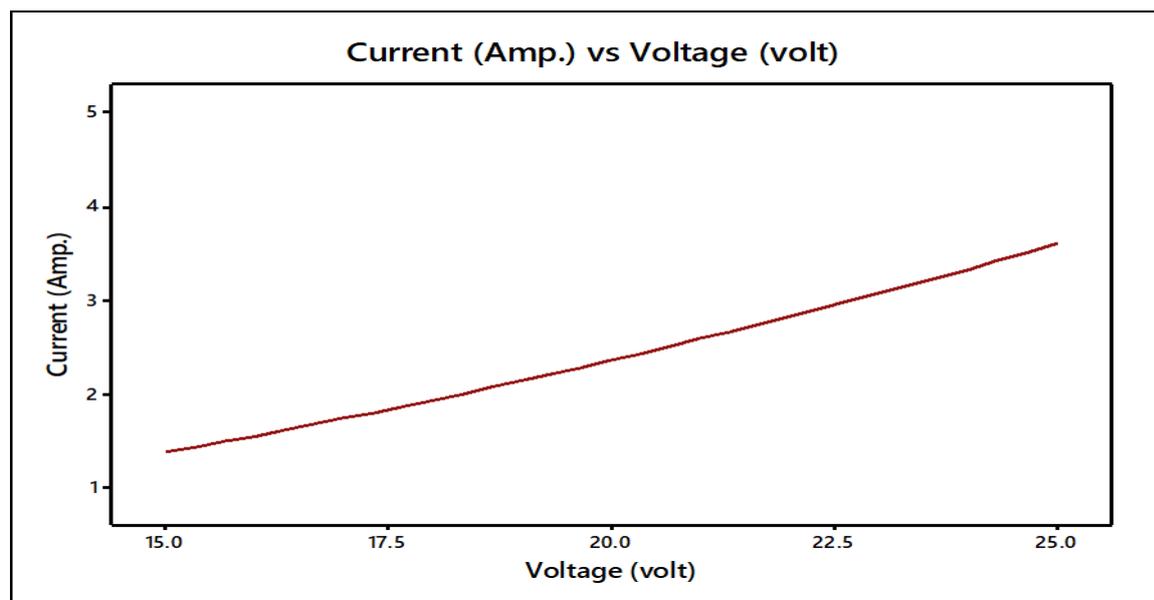


Figure. (4.24): The impact of voltage on the current applied of simulated wastewater (reaction time=41 min, Ph =8 , Nacl=2g and 200 rpm) for 100 mg RBD/l removal

The following correlations refer the to impacts of the operating variables on the Final Stable current Applied:

$$Y = 0.05583 x_1 - 0.000031 x_1^2 + C \quad R^2 = 85.7\% \quad (4.21)$$

$$Y = 0.6573 x_2 - 0.04255 x_2^2 + C \quad R^2 = 81.9\% \quad (4.22)$$

$$Y = 0.01348 x_3 + 0.005226 x_3^2 + C \quad R^2 = 86.5\% \quad (4.23)$$

#### 4.4.7. The Statistical Analysis of The Final TDS Response

The impact of pH on TDS may also be demonstrated in Figs(4.25) ,(4.26) and (4.27) .At acidic pH, the value of (TDS) decreases, and at base pH, the value of (TDS) increases, taking into account the impact of voltage and time of electrolysis. The impact of the applied voltage is directly proportional to this response.

As shown in Figs. (4.25) ,(4.26) and (4.27). the conct of TDS increased with the reaction time due to the continous relaing of different ions from electrodes .

then it tends to minimize because of the formation of oxide layer and gas bubbles that may decrease the relaing of ions from electrodes that may decrease the TDS value .

While the behavior of TDS response is increased with voltage till the and of the experiment at mean value of time and pH variables . ( *AlJaberi (d),2020*; *AlJaberi (e),2020*).

The impact of applied on the voltage investigated reactions is very important since the pace at which electro-coagulants and gas bubbles are released has a significant impact on the rate at which flocs develop, which is dependent on the provided voltages. Because it regulates the quantity of ions' aluminum released from electrodes, the bubbles of gas emission, and, as a result, the formation of flocs, it must be recognized in each treatment technique. The concentration of final TDS is signiticanthy increased with the increase of Ph value. At acidic conditions ; Then it tends to slightly decreased. this behavior is dependent on the preesent and consumption of hyrodxil ions .

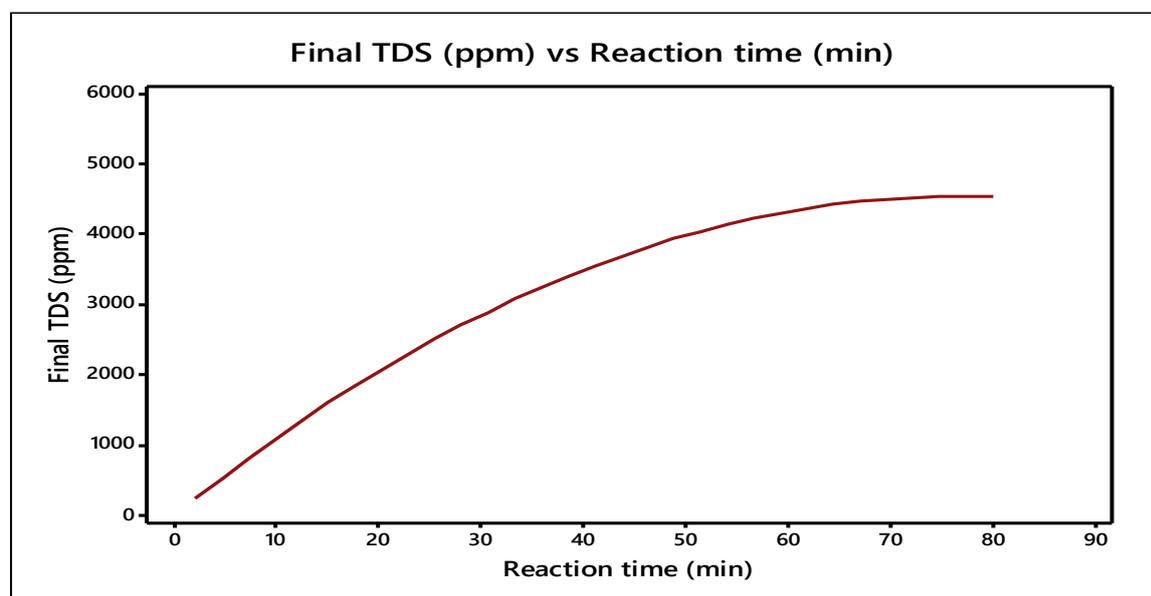


Figure. (4.25): The impact of reaction time on the final TDS of simulated wastewater (Ph =8 , voltage =20 volt, Nacl=2g and 200 rpm) for 100 mg RBD/l removal

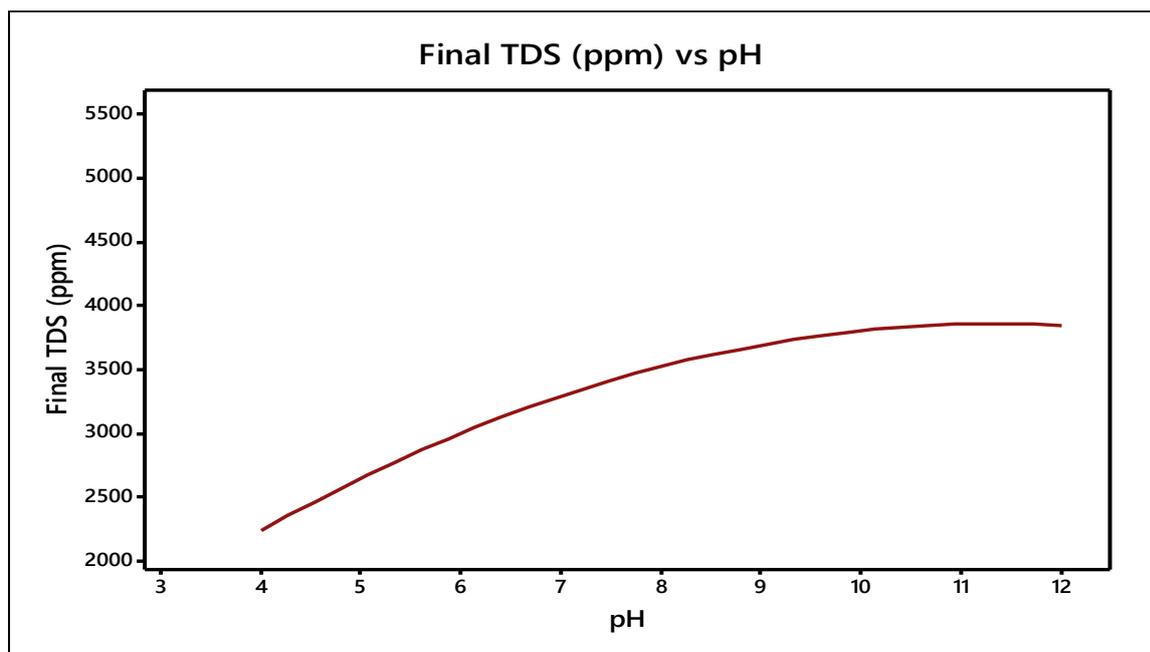


Figure. (4.26): The impact of Ph on the final TDS of simulated wastewater (reaction time=41 min, voltage=20 volt, NaCl=2g and 200 rpm) for 100 mg RBD/l removal

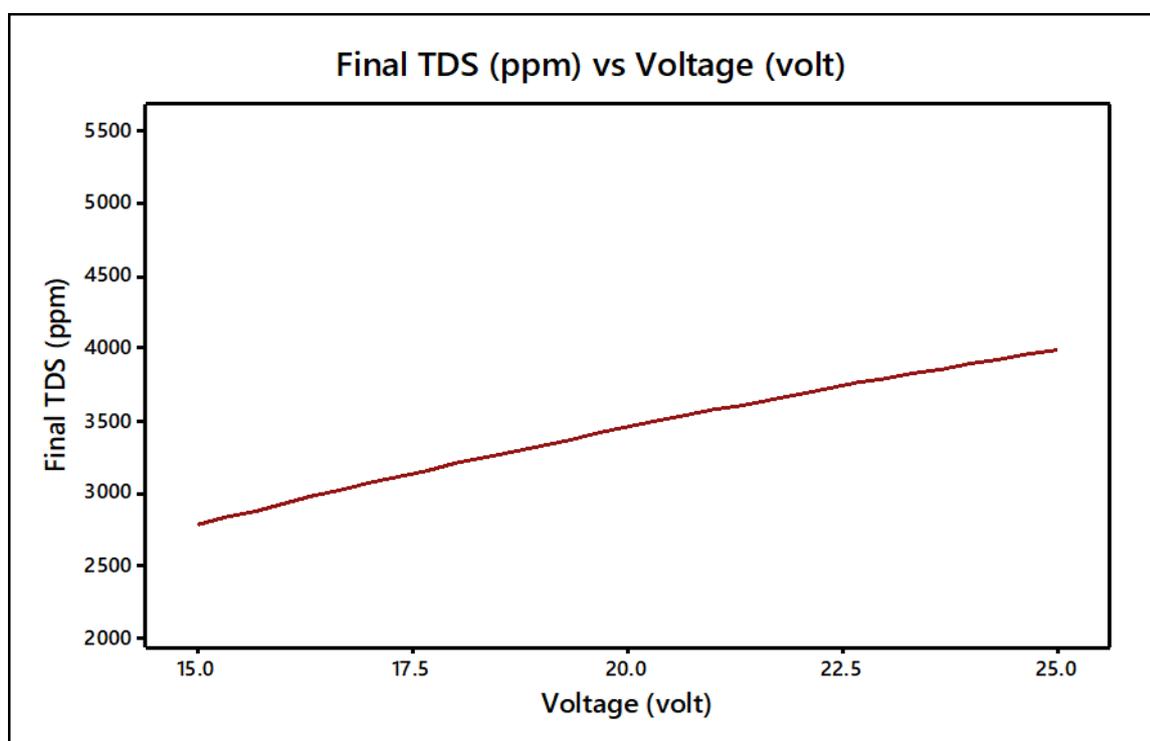


Figure. (4.27): The impact of voltage on the final TDS of simulated wastewater (reaction time=41 min, Ph=8 ,NaCl=2g and 200 rpm) for 100 mg RBD/l removal

The following correlations refer to the impacts of the operating variables on the final TDS:

$$Y = 117.9 x_1 - 0.7669 x_1^2 \quad R^2 = 94.6\% \quad (4.24)$$

$$Y = 679 x_2 - 29.91 x_2^2 \quad R^2 = 93.6\% \quad (4.25)$$

$$Y = 223.9 x_3 - 2.573 x_3^2 \quad R^2 = 94.5\% \quad (4.26)$$

#### 4.5. Mathematical Models

Table 2 explained the mathematical models of the studied responses of RBD removal efficiency, electrodes and energy usage where the ANOVA test proved the significance of the models estimated ( $p < 0.001$ ).

Table 4.4. Mathematical models

Response	Mathematical model	R2	ANOVA Test
RBD removal efficiency %	$Y = 204 + 1.07 x_1 - 10.38 x_2 - 13.55 x_3 - 0.0084 x_1^2 + 0.117 x_2^2 - 0.524 x_3^2 + 0.023 x_1 x_2 - 0.0132 x_1 x_3 + 0.800 x_2 x_3$	94.94%	$p < 0.001$ $F = 20.85$
Energy Consumption	$Y = 25.0 - 0.474 x_1 - 1.67 x_2 - 0.77 x_3 + 0.00231 x_1^2 + 0.0317 x_2^2 + 0.0137 x_3^2 + 0.0176 x_1 x_2 + 0.0065 x_1 x_3 + 0.0129 x_2 x_3$	95.52%	$p < 0.001$ $F = 23.70$

Electrodes Consumption	$Y = 14.74 - 0.085 x_1 - 1.214 x_2 - 0.382 x_3 + 0.00044 x_1^2 + 0.029 x_2^2 - 0.0074 x_3^2 - 0.0002 x_1 x_2 + 0.0102 x_1 x_3 + 0.0112 x_2 x_3$	95.33%	$p < 0.001$ $F = 22.68$
Final pH	$Y = 11.11 - 0.0279 x_1 + 0.100 x_3 - 0.911 x_2 - 0.000264 x_1^2 - 0.0040 x_3^2 + 0.0467 x_2^2 - 0.00019 x_1 x_3 + 0.00697 x_1 x_2 + 0.0056 x_2 x_3$	83.45 %	$p < 0.001$ $F = 26.46$
Final Conductivity	$Y = 5805 - 7.1 x_1 - 106 x_3 - 480 x_2 - 0.014 x_1^2 + 16.9 x_3^2 + 100.0 x_2^2 + 2.02 x_1 x_3 + 2.20 x_1 x_2 - 53.6 x_2 x_3$	91.32 %	$p < 0.001$ $F = 11.69$
Final stable current Applied	$Y = 15.0 - 0.1716 x_1 - 1.004 x_3 - 0.559 x_2 + 0.001100 x_1^2 + 0.0258 x_3^2 + 0.0161 x_2^2 + 0.00398 x_1 x_3 + 0.00447 x_1 x_2 + 0.0030 x_2 x_3$	82.27 %	$p < 0.009$ $F = 5.16$
Final TDS	$Y = 7819 - 84.1 x_1 - 291 x_3 - 650 x_2 + 0.3620 x_1^2 + 11.59 x_3^2 + 52.63 x_2^2 + 1.85 x_1 x_3 + 6.68 x_1 x_2 - 6.4 x_2 x_3$	97.44 %	$p < 0.001$ $F = 42.37$

### 4.6 Optimization of the operating Parameters

The optimum values of the operating parameters, reaction time, applied voltage and pH, were estimated as 45.33 min, 18.03 volt, and 4, respectively, utilizing Minitab statistical software where the composite desirability (D-indicator) of these findings equals 0.9284 Fig. (4.28).

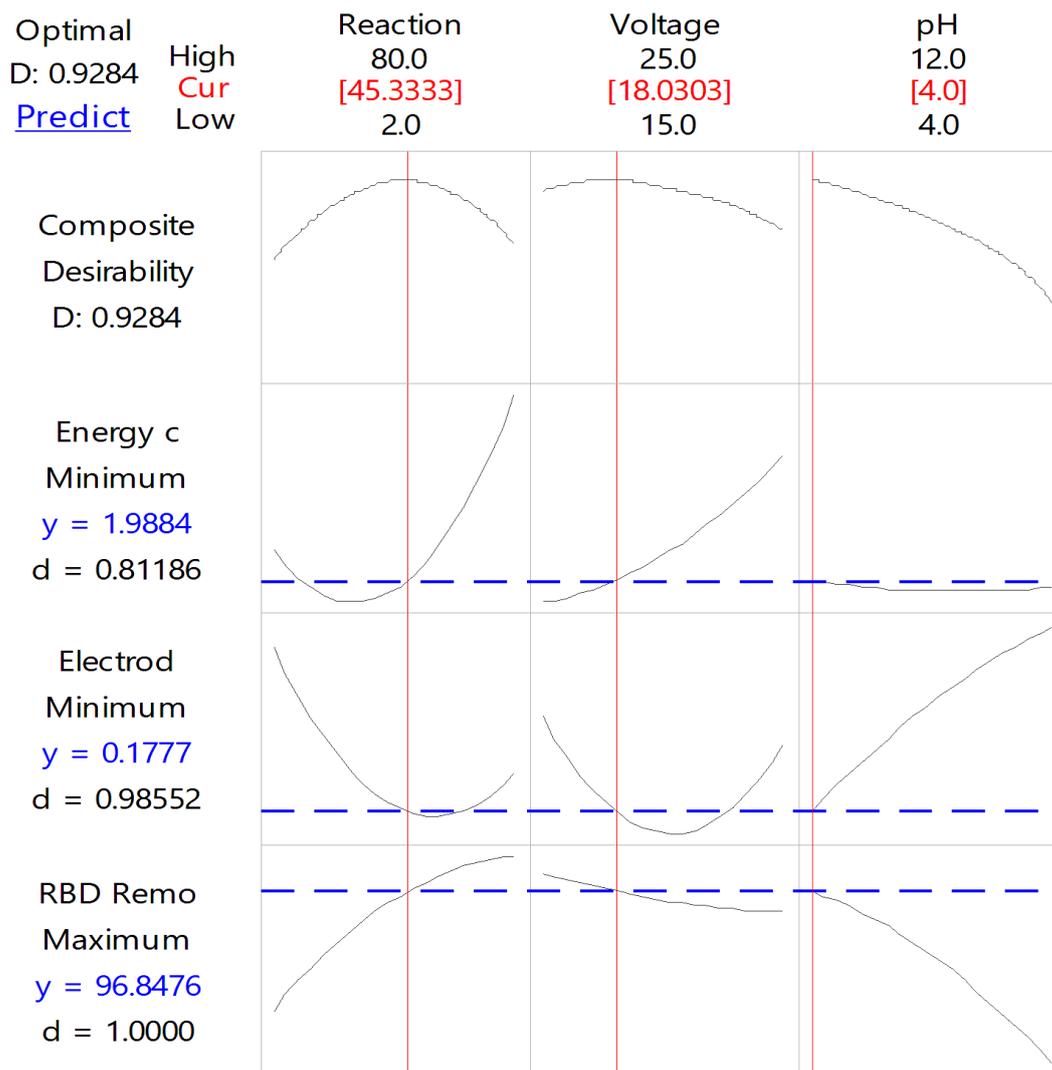


Figure (4.28) demonstrates the optimal settings of the functional parameters and responses for the remediation of simulated wastewater having 100 mg RBD/L.

Which proved the ability of the present design of the EC reactor to remove dyes from wastewater.

Table (4.5) : lists a compararison between the present study and other previous studies for dye wastewater treatment .

References	Pollutants	Electrodes metals		Operational variables	Removal efficiency %
		Anode	Cathode		
<b>Nazrul Islam et al. (2011)</b>	Orange II	Fe	Fe	Voltage ,time ,dye concentration	92%
<b>Rekha et al.(2016)</b>	remazol Red Rb 133	Fe	Fe	electrolyte concentration, current , pH, time	90%
<b>Nizam Mahmud et al. (2016)</b>	Chromium ,Colour	SS Al	SS Al	Types of Electrodes Material, pH and voltage	94.76%
<b>Patel et al.(2017)</b>	RB5	Fe	Al	current density , supporting electrolyte , initial pH	90%
<b>Mook et al.(2017)</b>	RB5	Fe	Fe	Initial pH, current, electrolyte dose EC time	80.9%
<b>Shah et al.(2017)</b>	R223 and CBBR250	Al	Fe	including pH, NaCl, voltage and electrolysis time	89% 94%
<b>Duan et al. (2018)</b>	MB	Al	Al	current density, voltage, distance between electrodes and reversal time	91%
<b>Assémian et al.(2018)</b>	MB	Fe	Fe	initial solution pH, current density, initial concentration of dye and energy consumption	93.2%
<b>Madhuri Damaraju , et al. (2018)</b>	Organic Carbon (TOC)	Fe	Fe	Initial pH , reaction time , and current density	85%
<b>Teng et al.(2020)</b>	MB	Fe graphite	Graphite Fe	Initial concentration ,ph, Voltage	86.46%

## **Chapter Five**

### **Conclusions and Recommendations**

#### **5.1. Conclusions**

The current project employed a batch electrocoagulation reactor consists of new configuration of electrodes. Four aluminum plates have utilized as electrodes in a bipolar-parallel connection mode . The following are the key conclusions:

- 1- The investigation of MB-dye removal has conducted and compared between two operating systems which are the constant modes of voltage and current (CVM and CCM), respectively.
- 2- Thermodynamic study proved that this reaction is endothermic, a random irregularity at the solid-liquid, and favorable. Both systems gave higher removal efficacies but the CCM was the highest.
- 3- The highest removing efficiency when design type (C) consisting of four plates the outer plates are not perforated while the entire two plates are perforated.
- 4- The RBD removal efficiency increased with increasing of the reaction time and voltage apply ,this type of dyes is remove efficiently at the acidic medium.
- 5- The period of electrolysis and the electric voltage provided have a significant influence on the energy consumption response, but the impact of other functional factors is minimal .
- 6- The operating variable, particularly electrolysis time and electric voltage, has a significant impact on the consumption of both electrodes.
- 7- When compared to its original value, the final pH varies with time, with a greater value when the solution is acidic and a lower value when the solution is alkaline.

8- In the current study, the impact of operating factors on the final value of electrical conductivity is clearly visible. When the pH of the contaminated solution varied, the behavior of conductivity at its ultimate value appeared to be more erratic.

A- The operating factors, particularly the voltage supplied to the electrocoagulation reactor, influence the final stable current applied response.

B- The impact of the three parameters, reaction time, voltage and pH , is directly proportional to the removal response.

9- The core results proved the efficient ability of the present configuration of electrodes which provide 96.85 %, 1.98 kWh/m<sup>3</sup>, and 0.178 g of the RBD removal efficiency ,the consumption of energy and electrodes respectively.

The main conclusion that the present configuration of electrodes containing perforated electrodes assisted the remediation process in the electrocoagulation reactor and detected findings at the optimum values proved that the current reactor of electrocoagulation is economic and cost-effective.

## **5.2. Recommendations**

- 1- Using various electrode designs to explain how they affect removal efficiency and other responses values.
- 2- Extend the functional parameter ranges to explain how effect the innovative design can be in these new ranges.
- 3- Investigating the remediation of real wastewater with the current electrode design.
- 4- Using continous mode to investigate the ability of the present design.

### References:

1. Abd Ul-Shaheed ,Sh .H., Ajjam,S,K., 2018, " Electrocoagulation – flotation for Nitrate Removal from Industrial Wastewater by mono-polar electrodes" ,University of Babylon.
2. Abdulhadi, B.A., Kot, P., Hashim, K.S., Shaw, A. and Khaddar, R.A. , 2019,"Influence of current density and electrodes spacing on reactive red 120 dye removal from dyed water using electrocoagulation/electroflotation (EC/EF) process", (Materials Science and Engineering )584: 012035 .
3. Adelaide D., M.Sc., 2013, "Electrocoagulation for Water Treatment: the Removal of Pollutants utilizing Aluminium Alloys, Stainless Steels and Iron Anodes", (National University of Ireland Maynooth, August).
4. Adhoum, N., and Monser, L. ,2004," Decolourization and removal of phenolic compounds from olive mill wastewater by electrocoagulation", (Chemical Engineering and Processing: Process Intensification), 43(10), 1281-1287.
5. Akbal, F., & Kuleyin, A. ,2011," Decolorization of levafix brilliant blue E-B by electrocoagulation method", (Environmental Progress & Sustainable Energy), 30(1), 29-36.
6. Akbartabar ,I. , Yazdanshenas, M. E. Y., Tayebi ,H.T. , Nasirizadeh ,N., 2017," Investigation of Acid Blue 62 dye adsorptions utilizing SBA-15/Polyaniline mesoporous nanocomposite: Kinetic and Thermodynamic study", (Iranian Journal of Health Sciences) ; 5(3): 17-34.
7. Alardhi ,S.M., AlJaberi, F. Y. and AlSaedi , L.M. , 2020, " Studying the treatability of various types of nanoparticles for oil content removal from oily wastewater produced from refinery process", (Egyptian Journal of Chemistry), 63, 4963-4973 .

## References

---

8. Al-Barakat ,H.S. , Matloub ,F. K., Ajjam, S. K. and Al-Hattab, T.A.,2020, "Modeling and Simulation of Wastewater Electrocoagulation Reactor", (IOP Conf. Series: Materials Science and Engineering) 871.
9. Alizadeh, M., Ghahramani, E., & Sadeghi, S. ,2015,"Removal of Reactive Green 19 dye from synthetic wastewater using electrocoagulation and aluminum electrodes", (Journal of advances in environmental health research), 3(1), 42-48.
- 10.AlJaberi, F. Y, 2018, "Studies of autocatalytic electrocoagulation reactor for lead removal from simulated wastewater", (J Environ Chem Eng. 6), 6069-6078 (d).
- 11.AlJaberi, F. Y. , Ahmed ,S.A. . and Makki ,H. F., 2020," Electrocoagulation treatment of high saline oily wastewater: evaluation and optimization", (Heliyon) , 6(6), e03988(a).
- 12.AlJaberi, F. Y. ,2018," Studies of autocatalytic electrocoagulation reactor for lead removal from simulated wastewater", (Journal of environmental chemical engineering), 6(5), 6069-6078 (a).
- 13.AlJaberi, F. Y., 2018,"Investigation of electrocoagulation reactor design effect on the value of total dissolved solids via the treatment of simulated wastewater ", (Desalination Water Treat),120, 141-149 (b) .
- 14.AlJaberi, F. Y., Abdul-Majeed, B. A., Hassan, A. H., Ghadban, M. L., 2020,"Assessment of an electrocoagulation reactor for the removal of oil content and turbidity from real oily wastewater utilizing response surface method", (Recent innovations in Chemical Engineering), 13 (1) 55-71 (c).
- 15.AlJaberi, F. Y., Mohammed, W. T., 2018,"Effecting of pH parameter on simulated wastewater treatment utilizing electrocoagulation method", ( Journal of Engineering), 24 , 73-88 (c).

## *References*

---

16. AlJaberi, F. Y., 2018, "Investigation of electrocoagulation reactor design effect on the value of total dissolved solids via the treatment of simulated wastewater", (Desalination Water Treat.120) , 141-149 (e) .
17. AlJaberi, F. Y., 2020, "Removal of TOC from oily wastewater by electrocoagulation technology", (In IOP Conference Series: Materials Science and Engineering), 928 (2), 022024. IOP Publishing (b) .
18. AlJaberi, F.Y., 2020, "Removal of TOC from oily wastewater by electrocoagulation technology", (In IOP Conference Series: Materials Science and Engineering), 928 (2), 022024. IOP Publishing (d).
19. Alwash, R. S., Safaa, K. H., & Al-Janabi, A. , 2021, "Sono-assisted treatment of textile wastewater: reactive black 5 dye a case study", ( In IOP Conference Series: Materials Science and Engineering), (Vol. 1184, No. 1, p. 012021), IOP Publishing .
20. Anantha Singh, T. S., and Ramesh, S. T. (2013). New trends in electrocoagulation for the removal of dyes from wastewater: a review. *Environmental Engineering Science*, 30(7), 333-349.
21. Andrade, L. S., Ruotolo , L. M., Rocha-Filho, R. C., Bocchi, N., Biaggio , S. R., Iniesta , J., Garcí'a-Garcia, V. and Montiel, V., 2007, "On the performance of Fe and Fe,F doped Ti–Pt/PbO<sub>2</sub> electrodes in the electrooxidation of the Blue Reactive 19 dye in simulated textile wastewater", (*Chemosphere*), 2035–2043.
22. Arcadio ,P. S., Gregoria, A. S., 2003, "Physical–Chemical treatment of water and wastewater", (International Standard Book Number) 1-58716-124-9.
23. Bazrafshan ,E. , KordMostafapoor, F. , Soori, M.M. and Mahv , A.H., 2012, " Application Of Combined Chemical Coagulation And Electro-Coagulation Process For Carwash Wastewater Treatment" ,(Fresenius Environmental Bulletin) , Volume 21 – No 9a\_
24. Benefield, L. D., Judkins, J. F., & Weand, B. L. , 1982, " Process chemistry for water and wastewater treatment".

## *References*

---

25. Bertsch, P. M., & Parker, D. R., 2020, "Aqueous polynuclear aluminum species", (In *The environmental chemistry of aluminum*) (pp. 117-168). CRC Press.
26. Borhade, A. V., Kankrej, S. R. and Kulkarni M. S., 2018, "An Efficient Removal of Erioglaucine Dye utilizing MCM-41 Synthesized from Waste Coal Fly Ash" (International Journal of Chemical and Physical Sciences), IJCPS Vol. 7, Special Issue ICAFM.
27. Can, O.T., Bayramoglu, M. and Kobya, M., 2003, "Decolorization of reactive dye liquid surrounding medias by electrocoagulation utilizing aluminum electrodes", (Ind. Eng. Chem. Res. 42), 3391-3396.
28. Canizares, P., Carmona, M., Lobato, J., Martinez, F. and Rodrigo, M. A., 2005, "Electrodisliquid surrounding media of aluminum electrodes in electrocoagulation processes", (Ind. Eng. Chem. Res. 44), 4178-4185.
29. Changmai, M., Pasawan, M., Purkait, M.K. 2019, "Treatment of oily wastewater from drilling site utilizing electrocoagulation followed by microfiltration", (Separation and Purification Technology), 210, 463-472.
30. Chen, G., 2004, "Electrochemical technologies in wastewater treatment", (Separation and Purification Technology), 38(1), 11-41.
31. Chigozie, U. F., & Joseph, N. T., 2014, "Removal of Orange-G, Vat Yellow, Erythrosine dyes from synthetic wastewater by electrocoagulation and nanofiltration", (J Adv Chem Eng), 4(112), 2.
32. Daneshvar, N., Ashassi-Sorkhabi, H. and Tizpar, A., 2003, "Decolorization of orange II by electrocoagulation method", (Separation and Purification Technology), 31, 153/162.
33. de Carvalho, H. P., Huang, J., Zhao, M., Liu, G., Dong, L., & Liu, X., 2015, "Improvement of Methylene Blue removal by electrocoagulation/banana

## References

---

- peel adsorption coupling in a batch system", (Alexandria Engineering Journal), 54(3), 777-786 .
- 34.Duan, X., Wu, P., Pi, K., Zhang, H., Liu, D., & Gerson, A. R. , 2018,"Application of modified electrocoagulation for efficient color removal from synthetic methylene blue wastewater", (Int J Electrochem Sci), 13(6), 5575-88.
- 35.Eilbeck, W. J., and Mattock, G. (1987). Chemical processes in waste water treatment. Ellis Horwood Ltd. Chichester, Sussex, 331.
- 36.El Alouani, M., Alehyen, S., El Achouri, M., & Taibi, M. H, 2018," Removal of Cationic Dye – Methylene Blue- from Aqueous Liquid surrounding media by Adsorptions on Fly Ash-based Geopolymer", ( J. Mater. Environ. ),Sci., 9 (1) 32-46.
- 37.Esmaeilirad ,N., Terry ,C. , Kennedy ,H., Li ,G. and Carlson ,K., 2015,"Optimizing Metal-Removal Processes for Produced Water With Electrocoagulation", ( Oil and Gas Facilities) , 4(02), 087-096.
- 38.Essadki, A. H. ,2012, " Innovative Electrochemical Reactors for Electrocoagulation/Electroflotation" , (Electrochemical Cells – New Advances in Fundamental Researches and Applications),107,pp. 17- 56.
- 39.Getaye, M., Hagos, S., Alemu, Y., Tamene, Z., & Yadav, O. P. ,2017," Removal of malachite green from contaminated water using electro-coagulation technique", ( J Anal Pharm Res), 6(4), 00184.
- 40.Ghalwa, N. M. A., Saqer, A. M., & Farhat, N. B. ,2016,"Removal of Reactive Red 24 dye by clean electrocoagulation process using iron and aluminum electrodes", ( Journal of Chemical Engineering & Process Technology), 7(1), 269.

## References

---

41. Golder, A.K., Hridaya, N., Samanta, A.N. and Ray, S., 2005, "Electrocoagulation of methylene blue and eosin yellowish utilizing mild steel electrodes", (Journal of Hazardous Materials) 127(1-3), 134–140.
42. Gomes, J. A., Jame, S. A., Chen, D., Palla, V., Bernazzani, P., & Cocke, D., 2011, "Removal of Textile Dye Using Electrocoagulation", (In EPD Congress 2011 (pp. 835-844)). Hoboken, NJ, USA: John Wiley & Sons, Inc, February.
43. Harrache, Z., Abbas, M., Aksil, T. and Trari, M., 2019, "Thermodynamic and kinetics studies on adsorption of Indigo Carmine from aqueous solution by activated carbon", (Microchemical Journal), 144, 180-189.
44. Hashim, K. S., Hussein, A. H., Zubaidi, S. L., Kot, P., Kraidi, L., Alkhaddar, R., ... & Alwash, R., 2019, "Effect of initial pH value on the removal of reactive black dye from water by electrocoagulation (EC) method", (In Journal of Physics: Conference Series) (Vol. 1294, No. 7, p. 072017), IOP Publishing, September.
45. Helder, P. de C., Jiguo, H., Meixia, Z., Gang, L., Lili, D. and Xingjuan, L., 2015, "Improvement of Methylene Blue removal by electrocoagulation/banana peel adsorptions coupling in a batch system", (Alexandria Engineering Journal) Vol.54, Issue 3, 777-786.
46. Holt, P.K., Barton, G.W., Wark, M., Mitchell, C.A., 2002 "A quantitative comparison between chemical dosing and electrocoagulation", (Colloids and Surfaces A: Physicochemical and Engineering Aspects) Volume 211, Issues 2–3, Pages 233-248.
47. Hunger, K. (Ed.), 2007, "Industrial dyes: chemistry, properties, applications".
48. Islam, S. M. N., Rahman, S. H., Adyel, T. M., Ahmed, Md. S., R. A. Yesmin, Rahman, Md. M. and Kaiser, N., 2011, "Electrocoagulation (EC) Technique

## References

---

- for Color Removal from Orange II Dye", (Journal of Environmental Research), 9, 45-52.
49. Jafat, R. Z. and Ajjam, S.K., 2016, "Removal of lead ions ( $Pb^{+2}$ ) from a synthetic wastewater by electrocoagulation utilizing aluminum (Al) as a rotating electrode", (International Journal of ChemTech Research), Vol.9, No.10, pp 166-176.
50. Jarusiripota, C., 2014, "Removal of Reactive Dye by Adsorptions over Chemical Pretreatment Coal Based Bottom Ash", (Procedia Chemistry), 121 – 130.
51. Kabdaşlı, I., Arslan-Alaton, I., Ölmez-Hancı, T., & Tünay, O., 2012, "Electrocoagulation applications for industrial wastewaters: a critical review", (Environmental Technology Reviews, 1(1)), 2-45.
52. Khan, A.A. and Singh, R.P., 1987, "Adsorptions Thermodynamics of Carbofuran on Sn (IV) Arsenosilicate in  $H^+$ ,  $Na^+$  and  $Ca^{2+}$  Forms", (Colloids and Surfaces), V24, 33-42.
53. Khandegar, V. and Anil, K. S., 2013, "Electrocoagulation for the treatment of textile industry effluente A review", (Journal of Environmental Management), Volume 128, 15 October, Pages 949-963.
54. Khorram, A. G., & Fallah, N., 2018, "Treatment of textile dyeing factory wastewater by electrocoagulation with low sludge settling time: optimization of operating parameters by RSM", (Journal of environmental chemical engineering), 6(1), 635-642.
55. Kobya, M., Demirbas, E., Can, O. T., & Bayramoglu, M., 2006, "Treatment of levafix orange textile dye solution by electrocoagulation", (Journal of hazardous materials), 132(2-3), 183-188.
56. López, A., Valero, D., García-Cruz, L., Sáez, A., García-García, V., Expósito, E., & Montiel, V., 2017, "Characterization of a new cartridge type

## References

---

- electrocoagulation reactor (CTECCR) using a three-dimensional steel wool anode", (Journal of Electroanalytical Chemistry), 793, 93-98.
57. Mahmad, M. K. N., Rozainy, M. M. R., Abustan, I., & Baharun, N. , 2016, "Electrocoagulation process by using aluminium and stainless steel electrodes to treat total chromium, colour and turbidity", (Procedia Chemistry), 19, 681-686 .
58. Mollah ,M.Y.A., Paul ,Morkovsky, Jewel,A.G.G., Mehmet,K., Jose,P. and David .L.C ,2004," Fundamentals, present and future perspectives of electrocoagulation", (Journal of Hazardous Materials), Volume 114, Issues 1–3, 18 October, Pages 199-210.
59. Mollah, M., Schennach, R., Parga, J.R. and Cocke, D.L. ,2001, "Electrocoagulation. (EC)-science and applications", ( J. Hazard. Mater), B84, 29.
60. Mook, W. T., Aroua, M. K., Szlachta, M., & Lee, C. S. ,2017," Optimisation of Reactive Black 5 dye removal by electrocoagulation process using response surface methodology", (Water Science and Technology), 75(4), 952-962.
61. Munagapati,V. S ., Yarramuthi,V., Nadavala, S.K. , Reddy Alla ,S., and Krishnaiah, A. ,2010, "Biosorption of Cu (II), Cd (II) and Pb (II) by *Acacia leucocephala* bark powder: Kinetics, equilibrium and thermodynamics," (Chem. Eng. J.), vol. 157, no. 2–3, pp. 357–365.
62. Naraghi, B., Baneshi, M. M., Amiri, R., Dorost, A., & Biglari, H. , 2018,"Removal of Reactive Black 5 dye from aqueous solutions by coupled electrocoagulation and bio-adsorbent process", ( Electronic physician), 10(7), 7086.
63. Nippatla, N., & Philip, L. ,2019," Electrocoagulation-floatation assisted pulsed power plasma technology for the complete mineralization of potentially toxic dyes and real textile wastewater", (Process Safety and Environmental Protection), 125, 143-156 .

## References

---

64. Othmana, N. H. , Alias ,N.H. , Shahrudin ,Z.S ., Abu Bakar ,N. F.A, Him,N. N.R. and Lau ,W.J. , 2018,"Adsorptions kinetics of methylene blue dyes onto magnetic graphene oxide", (Journal of Environmental Chemical Engineering. 6 (2)),2803-2811.
65. Özyurt, B., and Camcıoğlu, Ş. ,2018," Applications of combined electrocoagulation and electrooxidation treatment to industrial wastewaters" (In Wastewater and water quality (pp. 71-89). BoD–Books on Demand.
66. Pajootan, E., Arami, M., & Mahmoodi, N. M. ,2012," Binary system dye removal by electrocoagulation from synthetic and real colored wastewaters", (Journal of the Taiwan Institute of Chemical Engineers), 43(2), 282-290.
67. Patel ,N. B. , Soni, B. D. and Ruparelia, J. P., 2010,"Studies on Removal of Dyes from wastewater utilizing Electro-coagulation Process", ( Nirma university journal of engineering and technology), 1 (1), 20-25 .
68. Patel, U. D., Ruparelia, J. P., & Patel, M. U. ,2011," Electrocoagulation treatment of simulated floor-wash containing Reactive Black 5 using iron sacrificial anode", (Journal of hazardous materials), 197, 128-136.
69. Pi, K. W., Xiao, Q., Zhang, H. Q., Xia, M., & Gerson, A. R., 2014,"Decolorization of synthetic methyl orange wastewater by electrocoagulation with periodic reversal of electrodes and optimization by RSM", (Process safety and environmental protection), 92(6), 796-806,.
70. Şengil, İ. A., & Özacar, M. , 2009,"The decolorization of CI Reactive Black 5 in aqueous solution by electrocoagulation using sacrificial iron electrodes", (Journal of hazardous materials), 161(2-3), 1369-1376.
71. Shim, H. Y., Lee, K. S., Lee, D. S., Jeon, D. S., Park, M. S., Shin, J. S. and Chung, D. Y. ,2014," Application of electrocoagulation and electrolysis on the precipitation of heavy metals and particulate solids in washwater from the soil washing", ( Journal of Agricultural Chemistry and Environment, 3)(04), 130.

## *References*

---

72. Song, S., He, Z., Qiu, J., Xu, L., & Chen, J. ,2007," Ozone assisted electrocoagulation for decolorization of CI Reactive Black 5 in aqueous solution: An investigation of the effect of operational parameters", (Separation and purification technology), 55(2), 238-245 .
73. Teng, X., Li, J., Wang, Z., Wei, Z., Chen, C., Du, K., ... & Li, Y. , 2020,"Performance and mechanism of methylene blue degradation by an electrochemical process", (RSC Advances), 10(41), 24712-24720.
74. Tlaiaa, Y. S., Naser, Z. A. R., & Ali, A. H. ,2020," Comparison between coagulation and electrocoagulation processes for the removal of reactive black dye RB-5 and COD reduction", (DESALINATION AND WATER TREATMENT), 195, 154-161.
75. Waring, D. R., and Hallas, G. (Eds.). ,2013," The chemistry and application of dyes", ( Springer Science & Business Media).
76. William C. Agosta, R. S. Nyholm, FRS,1971, "Meredith Corporation" ,440 Park Avenue South New York, N.Y. 10016 Copyright R. L. M. Allen.
77. Zazou, H., Afanga, H., Akhouairi, S., Ouchtak, H., Addi, A. A., Akbour, R. A., ... & Hamdani, M. ,2019," Treatment of textile industry wastewater by electrocoagulation coupled with electrochemical advanced oxidation process", (Journal of Water Process Engineering), 28, 214-221.
78. Rekha, H. B., & Murthy, U. N. ,2016," Decolorization of reactive dye solutions by electrocoagulation using iron electrodes", ( Nature Environment and Pollution Technology), 15(1), 87.
79. Shah, A. R., Tahir, H., Ullah, H. M. K., & Adnan, A. ,2017,"Optimization of electrocoagulation process for the removal of binary dye mixtures using response surface methodology and estimation of operating cost", (Open Journal of Applied Sciences), 7(09), 458.

## APPENDIX- A

### Results of The Preliminary Experiment

**Table 1: Calculations of Van't Hoff formula to estimate the value of  $\Delta H$  of CCM experiment**

time	Temp. (°C)	Temp. (K)	(TEC)(g)	$q_e/C_e$	1/T(K)	$K_d$	Log $K_d$
0	18	291	0		0.00344		
2	22.5	295.5	0.03	19.66	0.00338	439.489	2.643
5	24	297	0.08	16.21	0.00337	144.897	2.161
10	26	299	0.17	9.97	0.00334	44.552	1.649
20	33	306	0.34	8.61	0.00327	19.242	1.284
30	40	313	0.50	12.71	0.00319	18.938	1.277
40	48	321	0.67	29.84	0.00312	33.349	1.523
50	53	326	0.84	29.26	0.00307	26.160	1.418
60	55	328	1.01	24.39	0.00305	18.173	1.259
70	59	332	1.17	27.63	0.00301	17.646	1.247

**Table 2: values of thermodynamics parameters of CCM experiment**

Temp. (°C)	Temp. (K)	$\Delta H$ (J/mol)	$\Delta G = -RT \ln K_d$	$\Delta S = (\Delta H - \Delta G)/T$
18	291	58475.372		
22.5	295.5		-14951.054	248.482
24	297		-12287.089	238.258
26	299		-9438.084	227.135
33	306		-7523.089	215.681
40	313		-7653.804	211.275
48	321		-9359.508	211.324
53	326		-8847.254	206.511
55	328		-7908.101	202.389
59	332		-7923.266	199.996

**Table 3: Calculations of Van't Hoff formula to estimate the value of  $\Delta H$  of CVM experiment**

time	Temp. (°C)	Temp. (K)	(TEC)(g)	$q_e/C_e$	1/T(K)	$K_d$	Log $K_d$
0	18	291	0	----	0.00344	----	----
2	20	293	0.03	19.66	0.00341	439.489	2.643
5	23	296	0.08	16.21	0.00338	144.897	2.161
10	27	300	0.17	9.97	0.00333	44.552	1.649
20	34	307	0.34	8.61	0.00326	19.242	1.284
30	40	313	0.50	12.71	0.00319	18.938	1.277
40	44	317	0.67	29.84	0.00315	33.349	1.523
50	48	321	0.84	29.26	0.00312	26.160	1.418
60	50	323	1.01	24.39	0.00310	18.173	1.259
70	55	328	1.17	27.63	0.00305	17.646	1.247

**Table 4: Values of thermodynamics parameters of CVM experiment**

Temp. (°C)	Temp. (K)	$\Delta H$ (J/mol)	$\Delta G = -RT \ln K_d$	$\Delta S = (\Delta H - \Delta G)/T$
18	291	58475.372	-----	----
20	293		-14824.564	250.171
23	296		-12245.719	238.923
27	300		-9469.649	226.483
34	307		-7547.674	215.059
40	313		-7653.804	211.275
44	317		-9242.879	213.622
48	321		-8711.560	209.305
50	323		-7787.550	205.148
55	328		-7827.805	202.144

**Table 5: Summary of calculations for the kinetic rate formula estimation of CVM experiment**

time	Dye Conc. (ppm)	Dye removal %	$C_t/C_0$	$-\ln(C_t/C_0)$	$C_t$	1/ $C_t$
0	100	0.00	1.00	0	100	0.01
2	53.2	0.47	0.53	0.63	53.20	0.02
5	35.553	0.64	0.36	1.03	35.55	0.03
10	30.965	0.69	0.31	1.17	30.97	0.03
20	20.612	0.79	0.21	1.58	20.61	0.05
30	10.494	0.90	0.10	2.25	10.49	0.10
40	3.61	0.96	0.04	3.32	3.61	0.28
50	2.965	0.97	0.03	3.52	2.97	0.34
60	2.964	0.97	0.03	3.52	2.96	0.34
70	2.259	0.98	0.02	3.79	2.26	0.44

**Table 6: Summary of calculations for the kinetic rate formula estimation of  
CCM experiment**

<b>time</b>	<b>Dye Conc. (ppm)</b>	<b>Dye removal %</b>	<b>C<sub>t</sub>/C<sub>0</sub></b>	<b>-.ln(C<sub>t</sub>/C<sub>0</sub>)</b>	<b>C<sub>t</sub></b>	<b>1/C<sub>t</sub></b>
0	100	0.00	1.00	0	100	0.01
2	73.411	0.27	0.73	0.31	73.41	0.01
5	61.529	0.38	0.62	0.49	61.53	0.02
10	55.411	0.45	0.55	0.59	55.41	0.02
20	29.059	0.71	0.29	1.24	29.06	0.03
30	18.235	0.82	0.18	1.70	18.24	0.05
40	5.529	0.94	0.06	2.90	5.53	0.18
50	1.65	0.98	0.02	4.10	1.65	0.61
60	1.411	0.99	0.01	4.26	1.41	0.71
70	1.1765	0.99	0.01	4.44	1.18	0.85

**Table 7: Reaction Orders Correlations and Constants Preliminary  
Experiment**

<b>Preliminary Experiment</b>	<b>First-Order</b>		<b>Second-Order</b>	
	<b>R<sup>2</sup></b>	<b>Correlation(y)</b>	<b>R<sup>2</sup></b>	<b>Correlation(y)</b>
<b>CCM</b>	<b>0.9708</b>	<b>0.0323x+0.0687y</b>	<b>0.8543</b>	<b>0.0122x+0.1007y</b>
<b>CVM</b>	<b>0.9325</b>	<b>0.5705x+0.0527y</b>	<b>0.9385</b>	<b>0.0064x+0.0196y</b>

## APPENDIX -B

### Results of The Studied Responses

Tabel .B1 : Results of The Studied Responses

Functional parameters				Final Results								
Run	Time (min)	Applied Voltage (Volt.)	pH	BD (ppm)	BD removal%	pH	TDS (ppm)	Conductivity	Current (Amp)	Temp. (C )	Energy cons. kWh/m3	Electrode cons.(g)
1	18	17.1	6	31.883	68.117	8.5	2541	5406	1.79	20	0.754	0.074
2	64	17.1	6	4.118	95.882	8.2	3173	6751	2.1	45	3.041	0.008
3	18	22.9	6	40.353	59.647	8.5	3070	6531	3.03	39.5	1.71	0.638
4	64	22.9	6	5.88	94.12	7.9	4546	9672	4.22	85	8.184	0.98
5	18	17.1	10	50.71	49.29	8.9	2541	5406	1.62	30	0.683	0.136
6	64	17.1	10	38.471	61.529	9.8	4925	8478	2.7	50	3.91	2.637
7	18	22.9	10	50.53	49.47	8.8	2992	6365	2.78	39	1.569	0.7
8	64	22.9	10	19.55	80.45	9.9	5497	8695	5.06	96	9.813	3.013
9	2	20.0	8	74.14	25.86	8.2	2262	4812	1.43	22	0.076	0.35
10	80	20.0	8	10	90	8	4775	8159	4.8	90	10.24	2.5
11	41	15.0	8	38.941	61.059	8.6	2732	5812	1.54	46	1.263	3
12	41	25.0	8	3.788	96.212	8.2	3782	8046	2.63	52	3.594	1.41
13	41	20.0	4	3.294	96.706	8.3	3283	6985	2.54	25	2.777	0.36
14	41	20.0	12	72.024	27.976	10.2	4337	9227	0.86	40	0.94	0.909
15	41	20.0	8	24.847	75.153	9.3	2865	6095	1.57	40	1.717	0.748
16	41	20.0	8	28.847	71.153	8.2	2936	6246	2	40	2.187	0.689
17	41	20.0	8	8.376	91.624	8.2	3283	6985	2.05	44	2.241	0.797
18	41	20.0	8	26.611	73.389	8.6	2973	6325	1.6	41	1.749	0.523
19	41	20.0	8	29.2	70.8	8.5	2918	6365	1.54	40	1.684	0.995

## الملخص:

يتم تصريف كمية كبيرة من مياه الصرف الصحي للنسيج كل عام من العديد من الصناعات التي يجب معالجتها. يهدف هذا العمل إلى تقييم القدرة على إزالة التبخير الكهربائي للصبغة الزرقاء التفاعلية (RBD) من مياه الصرف الصحي للنسيج الصناعي باستخدام أقطاب الألومنيوم المثقبة ويكون نوع الربط ثنائي القطب (bipolar). تمت دراسة حركية التفاعل والمعلمات الديناميكية الحرارية. تم دراسة تأثير وضع الإمداد بالطاقة ومقارنته باستخدام وضعين ؛ وضع التيار الثابت (CCM) (عند 3 أمبير) ووضع الجهد الثابت (CVM) (عند 20 فولت) طوال فترة التجارب (وقت التحليل الكهربائي: 2-70 دقيقة) ، وكان الرقم الهيدروجيني 8. النتائج التي تم الحصول عليها أظهرت أن كانت كفاءة إزالة RBD باستخدام CCM أعلى مقارنة بتلك التي تم الحصول عليها في حالة CVM. بعد 70 دقيقة ، قدمت التقنية السابقة 98.82% من إزالة RBD بينما قدمت الطريقة الأخيرة 97.74% من كفاءة الإزالة في نفس الحالة لتركيز RBD الأولي (100 جزء في المليون) ودرجة الحموضة 8. على الرغم من الاختلاف الطفيف بين النتيجتين المتحصل عليهما ، فإن هذا أثبتت النتائج أن آلية التنسيق بالدولة كانت نسبياً أكثر كفاءة وفعالية من حيث التكلفة من آلية التقييم الطارئ. كان ترتيب CCM من الدرجة الأولى بينما كان ترتيباً ثانياً للوضع الآخر. علاوة على ذلك ، كلتا العمليتين ماصتان للحرارة .

لقد تم إجراء هذا البحث تحت تأثير زمن التفاعل والجهد المطبق ودرجة الحموضة وفقاً للنطاقات (2-80 دقيقة) و (15-25 فولت) و (4-12) ، على التوالي. تم إجراء التصميم التجريبي وتحليل النتائج التي تم الحصول عليها باستخدام منهجية سطح الاستجابة (RSM) من نوع التصميم المركب المركزي (CCD) وبرنامج Minitab الإحصائي. كشفت النتائج الأساسية عن قابلية معالجة التكوين الحالي للأقطاب الكهربائية لتحقيق كفاءة إزالة أعلى لـ RBD. تم تحقيق إزالة RBD الكاملة عند القيم المثلى لمتغيرات التشغيل والتي كانت 45.33 دقيقة و 18.03 فولت و 4 من وقت التفاعل والجهد المطبق والأس الهيدروجيني ، على التوالي. كانت النماذج الرياضية مهمة وفقاً لاختبار ANOVA ( $P < 0.001$ ).

أثبتت هذه الدراسة قدرة تقنية التبخير الكهربائي على إزالة RBD من مياه الصرف باستخدام التكوين الحالي للأقطاب الكهربائية.



جمهورية العراق  
وزارة التعليم العالي والبحث العلمي  
جامعة بابل / كلية الهندسة  
قسم الهندسة الكيماوية

## الاختيار الامثل للتخثير الكهربائي ثنائي القطب لازالة الصبغة الزرقاء الفعالة من مياه الصناعات النسيجية

الرسالة

مقدمة إلى كلية الهندسة - جامعة بابل كجزء من متطلبات نيل  
الماجستير في الهندسة/الهندسة الكيماوية

من قبل  
دعاء رياض هادي كاظم

أ.م.د. فرات ياسر شراد

أ.م. ساطع كاظم اعجام

2022