# Electrocoagulation and Microfiltration Hybrid System for Water Treatment

By

#### **GANESH SHARMA**



University of Technology, Sydney

Submitted in fulfilment for the degree of

**Master of Engineering** 

Faculty of Engineering and Information Technology

University of Technology, Sydney (UTS)

Australia

July, 2011

#### **CERTIFICATE OF AUTHORSHIP**

I certify that the work in this thesis has not previously been submitted for any degree nor has it been submitted as part of requirements for a degree except as fully acknowledge within the text.

I also certify that the thesis has been written by me. And help that I have received in my research work and the preparation of the thesis itself has been acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

#### Signature of Candidature

Production Note: Signature removed prior to publication.

(Ganesh Sharma)

Sydney, July 2011

#### ACKNOWLEDGEMENT

First and foremost, I offer my sincerest gratitude to my supervisor, Dr HK Shon, who has supported me throughout my thesis with his patience, knowledge, motivation and guidance. One could not simply wish for a better and friendlier supervisor. I would also like to thank him for his financial support during my study.

I am heartily thankful to Dr. Rupak Aryal for his encouragement, guidance and assistance throughout this study. I wish to acknowledge Sherub Phuntsho for always providing generous help in the initial phase of the experimental works and assistance in writing thesis and journal papers. I would like to thank Jason Choi from University of Sydney for working with us on solar powered electrocoagulation.

In my daily work I have been blessed with a friendly and cheerful group of fellow students. My appreciation goes to Thanh, Ibrahim, Yousef, Wen Xing, Chinu, Johir and Rana for their generous help in the experimental phase of this research and staffs in the research office for their cooperation. I would also like to thank Dr. Hao for his support working in the Environmental laboratory.

I wish to thank my mother, sisters and brothers for their love and support. I could not have completed my study without their support and encouragement. Thanks are also to my room and house mates in Sydney. I offer my regards and blessings to all of those who supported me in any respect during the completion of this study.

### **TABLE OF CONTENTS**

Title Page	i
Certificate	ii
Acknowledgement	iii
Table of contents	iv
Nomenclature	X
List of the tables	xi
List of the figures	xii
Abstract	xvi

#### **CHAPTER 1**

INT	TRODUCTION		1-1
1 1	Introduction		1 2

1.1	Introduction	1-2
1.2	Objectives of the study	1-3

#### CHAPTER 2

2-1

2.1	Memb	Membrane Filtration	
	2.1.1	Historical Development of Membranes	2-2
	2.1.2	Overview of Membrane Technology	2-3
	2.1.3	Membrane Filtration Mechanisms	2-4
	2.1.4	Membrane Filtration Modes	2-5
	2.1.5	Submerged Membrane System	2-6
2.2	Memb	rane Fouling	2-7
	2.2.1	Definitions and Causes of Membrane Fouling	2-7

#### **TABLE OF CONTENTS**

Title Page	i
Certificate	ii
Acknowledgement	iii
Table of contents	iv
Nomenclature	Х
List of the tables	xi
List of the figures	xii
Abstract	xvi

#### **CHAPTER 1**

INTRODUCTION		1-1

1.1	Introduction	1-	-2
1.2	Objectives of the study	1-	-3

#### CHAPTER 2

LITE	RATU	RE REVIEW	2-1
2.1	Memb	rane Filtration	2-2
	2.1.1	Historical Development of Membranes	2-2

	2.1.2	Overview of Membrane Technology	2-3
	2.1.3	Membrane Filtration Mechanisms	2-4
	2.1.4	Membrane Filtration Modes	2-5
	2.1.5	Submerged Membrane System	2-6
2.2	Memb	rane Fouling	2-7

2.2.1 Definitions and Causes of Membrane Fouling 2-7

	2.2.2	Types of Membrane Fouling	2-7
		2.2.2.1 Membrane foulants	2-7
		2.2.2.2 Particulate/Colloidal Fouling	2-8
		2.2.2.3 Organic Fouling	2-9
		2.2.2.4 Inorganic Fouling	2-10
		2.2.2.5 Biofouling	2-10
2.3	Foulin	g Prevention	2-10
2.4	Pretre	atment 2.4.1 Adsorption	2-11 2-12
		2.4.2 Media Filter	2-12
		2.4.3 Cartridge Filter	2-12
		2.4.4 UV irradiation	2-13
		2.4.5 Flocculation/Coagulation	2-13
		2.4.6 pH Adjustment	2-13
		2.4.7 Ion Exchange	2-14
		2.4.8 Lime (Ca(OH) <sub>2</sub> ) Softening	2-14
		2.4.9 Chlorination	2-14
		2.4.10 Dechlorination	2-14
		2.4.11 Biofiltration	2-15
		2.4.12 Electrical pretreatment	2-15
2.5	Micro	filtration Hybrid System	2-15
	2.5.1	Coagulation-Microfiltration Hybrid System	2-16
	2.5.2	Adsorption-Microfiltration Hybrid System	2-17
2.6	Electro	ocoagulation (EC)-Membrane Hybrid System	2-19
2.7	Electro	ocoagulation	2-21
	2.7.1	Historical Background	2-21

	2.7.2	Electrocoagulation Theory and Fundamentals	2-22
		2.7.2.1 Reactions at the electrodes	2-24
		2.7.2.2 Advantages of EC over conventional	
		chemical coagulation	2-25
2.7.3	Applic	cations of Electrocoagulation	2-26
2.8	Solar	Powered Electrocoagulation	2-27
СНА	PTER	83	
EXPI	ERIMI	ENTAL	3-1
3.1	Introd	uction	3-2
3.2	Exper	imental Materials	3-2
	3.2.1	Wastewater	3-2
		3.2.1.1 Synthetic Surface Water	3-2
		3.2.1.2 Synthetic Wastewater	3-2
	3.2.2 1	Membrane	3-4
3.3	Exper	imental Methods	3-5
	3.3.1	Chemical Coagulation	3-5
	3.3.2	Electrocoagulation	3-5
	3.3.3	Solar Powered Electrocoagulation	3-7
	3.3.4	Crossflow Microfiltration Setup	3-8
3.4	Analy	tical Methods	3-9
	3.4.1	Turbidity and pH	3-9
	3.4.2	Dissolved Organic Carbon (DOC)	
		and UV Absorbance	3-9
	3.4.3	Zeta Potential	3-10

#### **CHAPTER 4**

RESU	JLTS	AND DISCUSSION	4-1		
4.1	Electro	ocoagulation with sacrificial iron electrodes for water treatment	4-2		
	4.1.1	Turbidity removal with electrocoagulation using			
		iron electrodes	4-2		
	4.1.2	Organic removal with electrocoagulation using iron electrodes	4-3		
	4.1.3	Performance of microfiltration with electrocoagulation using			
		iron electrodes as pretreatment	4-4		
	4.1.4	Comparison of electrocoagulation with iron electrodes with			
		chemical coagulation	4-6		
		4.1.4.1 Turbidity removal with chemical coagulation using			
		ferric chloride	4-6		
		4.1.4.2 Organic removal with chemical coagulation using			
		ferric chloride 4.1.4.3 Performance of microfiltration flux with chemical	4-8		
		coagulation using ferric chloride as pretreatment	4-9		
4.2	Electrocoagulation with sacrificial aluminium electrodes				
	for wa	ater treatment	4-11		
	4.2.1	Effect of pH on turbidity and zeta potential			
		with electrocoagulation	4-12		
	4.2.2	Effect of pH on organic matter removal			
		with electrocoagulation	4-13		
	4.2.3	Effect of pH on turbidity and zeta potential with chemical			
		coagulation using alum	4-14		
	4.2.4	Effect of pH on organic matter removal with chemical			
		coagulation using alum	4-15		

		without charge controller	4-36
	4.5.5	Performance of microfiltration with solar power	
		electrocoagulation pretreatment using battery	
		and charge controller	4-37
4.6	Study	of fouling mechanism in electrocoagulation-crossflow	
	micro	filtration system	4-38
	4.6.1	Fouling mechanisms	4-40
	4.6.2	Fouling mechanism with electrocoagulation	4-43
CHA	PTEF	R 5	
CONCLUSION 5			5-1
5.1	Electr	ocoagulation with iron electrodes as pretreatment	
	to mi	crofiltration	5-2
5.2	Electr	ocoagulation with aluminiumn electrodes as pretreatment to	
	micro	ofiltration	5-3
5.3	Solar	powered electrocoagulation	5-4
5.4	Foulir	ng mechanism in electrocoagulation-crossflow	
	micro	filtration system	5-5
<b>REFERENCES</b> R			R-1
APP	ENDE	X	
List of publications based on this research			A-1

## NOMENCLATURE

DOC		dissolved organic carbon
EC	=	electrocoagulation
F	=	faraday's constant (mol <sup>-1</sup> )
i		current density (A cm <sup>-2</sup> )
J	=	filtrate flux at a given time (1 m <sup>-2</sup> hr)
$\mathbf{J}_0$		pure water flux (1 m <sup>-2</sup> hr)
$K_1$	=	cake filtration constant $(l^2 min^{-1})$
$k_{l}$		filtration constant
М		relative molar mass of the electrode
MF	=	microfiltration
PV	-	photovoltaic
$Q_0$	=	initial flux (ml min <sup>-1</sup> )
SPEC		solar powered electrocoagulation
Т	=	electrocoagulation time (s)
t	=	filtration time (min)
V	=	permeate volume (ml)
$V_{\mathrm{f}}$	=	volume of permeate producing hydraulic resistance equal to
		membrane (l)
W	=	quantity of electrode material dissolved (g of M $cm^{-2}$ )
Z	=	number of electrons transferred in the reaction

Х

### LIST OF TABLES

Table 2.1	Characteristics of membranes (Adapted from Stephenson et al., 2000)
Table 2.2	Membrane foulant types
Table 2.3	Methods to reduce membrane fouling (Adapted from Mulder, 1996)
Table 3.1	Properties of synthetic water
Table 3.2	Constituents and characteristics of the synthetic wastewater
Table 3.3	Properties of flat sheet microfiltration membrane module
Table 4.1	Optimum parameters of electrocoagulation operation in terms of removal of humic acid (HA) and turbidity
Table 4.2	Results for the fouling mechanisms
Table 4.3	Results of the fouling mechanism for EC-MF according to standard law of filtration
Table 4.4	Results of the fouling mechanism for EC-MF according to classical cake filtration model

#### LIST OF FIGURES

- Figure 2.1 Membrane filtration modes
- Figure 2.2 Experimental set-up of submerged membrane adsorption hybrid system (Adapted from Guo et al., 2006)
- Figure 2.3 Schematic diagram of a bench-scale two-electrode electrocoagulation cell (Holt et al.2002)
- Figure 2.4 Schematic flow-diagram of a typical wastewater treatment plant and complex processes that can be replaced by electrocoagulation (Adapted from Mohammad et al., 2004)
- Figure 2.5 Solar radiation resource possessed by Australia (Adapted from Richards and Schafer, 2002)
- Figure 3.1 Scanning electron microscope image of microfiltration membrane used in this study
- Figure 3.2 Schematic diagram for the experimental setup of chemical coagulation
- Figure 3.3 Schematic diagram of a bench-scale two-electrode electrocoagulation cell
- Figure 3.4 Experimental setup for solar powered electrocoagulation
- Figure 3.5 Schematic diagram of the cross flow microfiltration unit
- Figure 3.6 Zetasizer Nano Series-Zs (Malvern, UK)
- Figure 4.1 Turbidity removal efficiency at different EC generation time (current density: 12 Am<sup>-2</sup>, pH 8, iron electrodes)
- Figure 4.2 DOC (left) and UV Abs (right) removal at different EC generation time (current density: 12 Am<sup>-2</sup>, pH 8, iron electrodes)

- Figure 4.3 Normalised permeate flux at different generation time of EC followed by fast and slow mixing using flocculator (current density: 12 Am<sup>-2</sup>, transmembrane pressure 10 kPa, cross flow velocity 0.5 1 min<sup>-1</sup>, pH 8)
- Figure 4.4 Turbidity removal at different dosing at neutral condition (left) and at different pH with optimum dosing (right)
- Figure 4.5 DOC removal at different dosing at neutral condition (left) and at different pH with optimum dosing (right)
- Figure 4.6 UV removal at different dosing at neutral condition (left) and at different pH with optimum dosing (right)
- Figure 4.7 Normalised permeate flux at different dosing of ferric chloride (transmembrane pressure 10 kPa, cross flow velocity 0.5 1 min<sup>-1</sup>, pH 6.5)
- Figure 4.8 Turbidity removal efficiency and zeta potential after EC treatment at different pH (current density: 12 Am<sup>-2</sup>; EC time: 30 min)
- Figure 4.9 DOC removal efficiency and normalised UV absorbance after EC treatment at different pH (current density: 12 Am<sup>-2</sup>; EC time: 30 min)
- Figure 4.10 Turbidity removal efficiency and zeta potential after chemical coagulation experiments at different pH
- Figure 4.11 DOC removal efficiency and normalised UV absorbance after chemical coagulation experiments at different pH
- Figure 4.12 Normalised permeate flux at different generation time of EC (current density: 12 Am<sup>-2</sup>, transmembrane pressure 10 kPa, cross flow velocity 0.5 1 min<sup>-1</sup>, pH 8)
- Figure 4.13 Normalised permeate flux at different generation time at different dosing of chemical coagulation (transmembrane pressure 10 kPa, cross flow velocity 0.5 l min<sup>-1</sup>, pH 6.5)
- Figure 4.14 Normalised permeate flux through MF for EC using aluminium plates (transmembrane pressure 10 kPa, cross flow velocity 0.5 l min<sup>-1</sup>)

- Figure 4.15 Normalised permeate flux through MF for chemical coagulation (transmembrane pressure 10 kPa, cross flow velocity 0.5 l min<sup>-1</sup>)
- Figure 4.16 Variation of turbidity (left) and UV removal (right) under different current intensity (Initial turbidity = 80 NTU, initial UV =  $0.150 \text{ cm}^{-1}$ , pH = 8.0 and electrodes gap = 2 cm)
- Figure 4.17 Variation of turbidity (left) and UV removal (right) at different pH (Current density= 11.5 mA cm<sup>-2</sup>, initial turbidity=80 NTU, initial UV= 0.150 cm<sup>-1</sup> and electrodes gap= 2 cm)
- Figure 4.18 Variation of turbidity (left) and UV removal (right) at different gaps of electrodes (Current density =  $11.5 \text{ mA cm}^{-2}$ , initial turbidity = 80 NTU, initial UV=  $0.150 \text{ cm}^{-1}$  and pH= 8.0)
- Figure 4.19 Variation of turbidity (left) and UV removal (right) at different concentration of humic acid (Current density = 11.5 mA cm<sup>-2</sup>, initial turbidity = 80 NTU, initial UV = 0.150 cm<sup>-1</sup>, electrodes gap = 2 cm and pH = 8.0)
- Figure 4.20 Variation of turbidity at five different times in a day (Initial turbidity = 80 NTU, initial UV= 0.150 cm<sup>-1</sup> electrodes gap = 2 cm and pH = 8.0. Experiment conducted on 4th April 2010)
- Figure 4.21 Variation of UV absorbance (254 nm) at five different times in a day (Initial turbidity = 80 NTU, initial UV= 0.150 cm<sup>-1</sup>, electrodes gap= 2 cm and pH= 8.0, experiment conducted = 4th April 2010, weather condition = fine)
- Figure 4.22 Variation of turbidity at three different times in a day (Initial turbidity = 80 NTU, initial UV =  $0.150 \text{ cm}^{-1}$ , electrodes gap = 2 cm, pH = 8.0 and current density= 2.11 mA cm<sup>-2</sup>)
- Figure 4.23 Variation of UV at three different times in a day. Initial turbidity = 80 NTU, initial UV =  $0.150 \text{ cm}^{-1}$ , electrodes gap = 2 cm, pH= 8.0 and current density= 2.11 mA cm<sup>-2</sup>)

- Figure 4.24 Variation of DOC at five different times in a day (Initial turbidity = 80 NTU, initial DOC = 5.5 mg/l, initial UV= 0.150 cm<sup>-1</sup>, electrodes gap = 2 cm and pH = 8.0. Experiment conducted = 4th April 2010, weather condition = fine)
- Figure 4.25 Variation of DOC at three different times in a day (Initial turbidity = 80 NTU, initial UV = 0.150, electrodes gap = 2 cm and pH = 8.0, current density =  $2.11 \text{ mA cm}^{-2}$ )
- Figure 4.26 Normalised permeate flux after SPEC pretreatment at five different times in a day (SPEC operation: 35 min, transmembrane pressure: 10 kPa, cross flow velocity: 0.5 1 min<sup>-1</sup>, pH 8)
- Figure 4.27 Normalised permeate flux after SPEC pretreatment at three different times in a day (SPEC operation: 35 min, transmembrane pressure: 10 kPa, cross flow velocity: 0.5 1 min<sup>-1</sup>, pH 8)
- Figure 4.28 Relationship between t/V and t for kaolin concentration of 100 mg/l at three different CFV
- Figure 4.29 Relationship between t/V and t for kaolin concentration of 400 mg/l at three different CFV
- Figure 4.30 Relationship between t/V and t for kaolin concentration of 800 mg/l at three different CFV
- Figure 4.31 Permeate flux with respect to time at various EC time

#### ABSTRACT

Membrane technology for water and wastewater treatment offers many advantages over other conventional treatment systems. However, membrane process is usually hampered by the problem of membrane fouling which restricts its widespread application. Membrane fouling decreases permeate flux and plant productivity, increases hydraulic resistances thereby increasing energy consumption and increases the operational and maintenance costs ultimately affecting the overall plant economy. Pretreatment of feed water is considered one of the most effective means to reduce membrane fouling. Pretreatment increases the membrane lifetime and reduces membrane deterioration. Although several pretreatment options are available, only few studies have been reported so far for electrocoagulation (EC) as an attractive pretreatment method for membrane filtration.

The main objectives of this study are i) to evaluate water treatment by EC using aluminium and iron electrodes, ii) to evaluate the performance of microfiltration (MF) with EC as pretreatment, iii) to determine the EC operating conditions favouring removal of organic matter and turbidity, iv) to optimise EC-MF hybrid system for water treatment, v) to investigate the feasibility of solar powered electrocoagulation (SPEC) for applications in remote communities of Australia, vi) to access the feasibility of SPEC as a sustainable pretreament option for MF and finally vii) to identify the fouling mechanisms involved in the crossflow MF system when EC is used as pretreatment for the feed water.

EC pretreatment of synthetic water using iron electrodes did not reduce MF fouling due to the release of soluble ferrous ions ( $Fe^{2+}$ ) as it was not capable of colloidal destabilisation and  $Fe^{2+}$ -organic matter complexation prevents  $Fe(OH)_3$  precipitation and floc formation. However, EC pretreatment with aluminium electrodes significantly improved the performance of MF. The permeate flux for pretreated feed water was more than 55% higher than the feed water without pretreatment under optimum EC operating conditions. The isoelectric point for EC with aluminium electrodes occurred at pH 8. The highest removal efficiency (dissolved organic carbon (DOC) by 78%, UV abs by 85% and turbidity by 88%) occurred at the isoelectric point, where charge neutralisation occurred. Similarly, the highest organics and turbidity removal by chemical coagulation using aluminium sulphate also occurred at the isoelectric point (pH 6.5).

The potential for using solar powered electrocoagulation (SPEC) as an attractive technology for small and decentralised water purification system was explored. SPEC offered a suitable candidate for applications in the remote communities where renewable solar energy such as solar power is abundant. SPEC reactor was designed by connecting to photovoltaic panel (PV) either directly or through a set of batteries and charge control system. SPEC process system was observed sensitive to variation of solar irradiation when connected directly with PV panels and without any charge control system. SPEC reactor operated for five different times in a day (4 April 2010), yielded the highest organics removal at around midday i.e. between 10:00 AM-2:00 PM (DOC by 75%, UV abs by 85% and turbidity by 87%) under optimum EC operating conditions. However, when SPEC process was supported by batteries and charge control system, the process removal efficiency improved and also became more consistent. The variation in organic and turbidity removal was within the range of 10% for experiments conducted on three different times in a day (9 April 2010) with the highest removals at 10:30 AM in the morning.

The feasibility for SPEC as a sustainable pretreatment option, SPEC-MF hybrid system was evaluated. SPEC pretreatment using PV panel only without the charge control system improved the flux however the flux performance fluctuated due to the variation in the solar irradiation. The connection to batteries and charge control system improved the performance of MF permeate flux and also became more stable.

The fouling mechanism of crossflow MF was studied comparatively with feed water containing kaolin suspension with and without EC pretreatment. When the feed water was pretreated by EC, the fouling was found to follow both standard law of filtration and classical cake filtration model.