# UNIVERSITI TEKNOLOGI MARA

# SURFACE PROPERTIES AND ANTIMICROBIAL PERFORMANCE OF COPPER COATINGS ELECTRODEPOSITED ON 304 STAINLESS STEEL

## NIK NORZIEHANA BINTI CHE ISA

Thesis submitted in fulfillment of the requirements for the degree of **Doctor of Philosophy** 

**Faculty of Applied Sciences** 

December 2017

### ABSTRACT

Bacterial contamination resulted from frequently-touched surface materials is responsible in the acquisition of infection especially in hospital. The utilization of copper as antimicrobial touch surfaces offers a solution that can serve as an additional line of defense against pathogens. Copper is the first and currently the only one surface metal that has received registration from Environmental Protection Agency (EPA) as antimicrobial material which is capable to inhibit biofilms, retains the antimicrobial activity under typical indoor conditions. In this study, electrodeposition technique was used to coat copper on 304 stainless steel using 0.01 M Cu<sup>2+</sup> solutions containing either uncomplexed CuSO4 or complexed Cu-EDTA based systems. Effects of applied potential (based on CV analysis) and deposition time were investigated to study the formation, nucleation and growth of copper on the 304 stainless steel. Copper coatings Cu/SS(-0.25 V, CuSO4 pH 1, 900 s) and Cu/SS(-1.1 V, Cu-EDTA pH 8, 900 s) prepared at low overpotential from both electrolyte solutions possess good physicochemical and surface properties. In addition, they also have high contact angle, high surface roughness and well-adhered to the substrate. Both copper coatings showed excellent antimicrobial activity towards E. coli (100 % reduction within 5min of exposure) and S. aureus (100 % reduction within 10 min of exposure), compared to solid copper (100 % of reduction for both bacteria required within 10 min of exposure) whereas stainless steel surface showed no antibacterial activity even after 30 min of exposure. A significant observation is on the reduction of S. aureus after 5 min of exposure with Cu/SS(-1.1 V, Cu-EDTA pH 8, 900 s) showed outstanding antibacterial activity compared to Cu/SS(-0.25 V, CuSO4 pH 1, 900 s) and solid copper. Even though solid copper has higher dissolution rate (0.1292 mm/yr) than the copper coatings (i.e.: Cu/SS(-0.25 V, CuSO4 pH 1, 900 s) = 0.0531 mm/yr and Cu/SS(-1.1 V, Cu-EDTA pH 8, 900 s) = 0.08048 mm/yr) but both copper coatings have faster killing rate of bacteria than solid copper. This indicates that the killing rate does not only depend on dissolution rate but also surface properties. Cu/SS(-1.1 V, Cu-EDTA pH 8, 900 s) sample with high surface roughness, hydrophobic, high compositions of copper with uniform distribution of nano-sized grains structure resulted in better and more efficient contact killing of bacteria by allowing copper structure to interact closely with bacterial membranes.

## ACKNOWLEDGEMENT

#### Bismillahirrahmanirrahim,

Alhamdulillah. Thanks to Allah SWT, whom with His willing giving me the opportunity and strength to complete this journey. Thank you Allah because had blessed me with more than I ever asked for. I appreciate all the good things in my life.

The success of this thesis can been attributed to the extensive support and assistance from my supervisor, Associate Professor Dr Yusairie Mohd, who always gives valuable advice, guidance, comments, especially kindness, suggestions and also sacrifice his time to me. I would like to express my gratitude to him for supervising me on how to do this study systematically and consistently and how to write the story beautifully.

Many thanks go to my co-supervisor, Associate Professor Dr Sharifah Aminah Syed Mohamad and all members of the "Micres group", especially Dr Suhaidi Ariffin and Fadhil, who guide me in terms of antimicrobial part and teach me how to handle antibacterial testing from zero until I master it. Special thanks to Mrs Shadatul Hanum Rashid, who support me a lot in terms of financial through her grant, Mr Hayub Ta and Mrs Nida for their assistance on the FESEM, Mr Abul Khamis for his assistance on the XRD, Mrs NurulWahida for her assistance on the AFM, Mrs Azizan for her assistance on the contact angle, all lab staffs and administration staffs. This research would not have been possible without the advice and support from a lot of people.

I am also very grateful to my sponsors: The Ministry of Higher Education (Malaysia) for the financial support through myPhD under Mybrain15 scholar and Fundamental Research Grant Scheme (FRGS) 600-RMI/FRGS 5/3 (139/2015).

I would like to acknowledge all my friends who have participated in my life during this journey, but space does not permit me to name all of you. However, I would be remiss if I did not name just a few: Dr Noorazmi, Dr Nor Zakiah, Dr Nurain, Dr Sukri Hadi, Mohammad Hafizudden, Siti Sarah, Muhammad Aliff, Mohd Hafiz, Mohd Muizz Fahimi, Zahidah, Monica, Mohamad Aslam, Wan Saidatul Syida, Norismawatiaizan, Akmar, Fazeela, Nazmi Zakaria, Fauzi Aryaan, Aidil Anuar and Mohamad Faridz Osman. You all are true friends during this wonderful journey, who make me laugh, who make me happy, who encourage me, who push me, who challenge me, who are celebrating happy time together and be my side in sad times.

I would like to thank my deepest appreciation to my family for their continuing and loving support, especially my parents (Che Isa and Nik Hakipah). This thesis specially dedicated to my late mother who could not see this thesis completed.

Finally, once again thanks to everyone who involved scientifically and as friends during this journey. I appreciate immensely your time, effort, support, friendship and kindness. I think without all of you and my family, this journey for sure not being that nice.

## **TABLE OF CONTENTS**

CONFIRMATION BY PANEL OF EXAMINERS	ii
AUTHOR'S DECLARATION	iii
ABSTRACT	iv
ACKNOWLEDGEMENT	v
TABLE OF CONTENTS	vi
LIST OF TABLES	ix
LIST OF FIGURES	x
LIST OF ABBREVIATIONS	xvii

## CHAPTER ONE: INTRODUCTION

1.1	Background of Study	1
1.2	Problem Statements	5
1.3	Objectives of Study	6
1.4	Significance of Study	7
1.5	Scope and Limitations of Study	7
1.6	Thesis Organization	8
1.7	Conceptual Framework	10

### CHAPTER TWO: LITERATURE REVIEW

2.1	Introduction		
2.2	Hospital-acquired Infections (HAIs)		
	2.2.1 Bacterial Infections	15	
2.3	Conventional Infection Controls	18	
2.4	Copper and Copper Alloys	19	
2.5	Antimicrobial Copper	21	
2.6	Copper as Antimicrobial Touch Surfaces	24	
2.7	The Science behind Antimicrobial Copper	34	
2.8	Copper Coatings	38	

Page

2.9	Electrodeposition of Copper	
	2.9.1 Electrodeposition Techniques	51
	2.9.1.1 Cyclic Voltammetry (CV)	51
	2.9.1.2 Choronoamperometry (CA)	54
	2.9.2 Electrodeposition of Copper from Aqueous System	57
2.10	Electrochemical Studies Related to the Copper Electrodepositi	on 62
	2.10.1 Effect of Adding Additives	64
	2.10.2 Metal-Chelate Complexes	69
	2.10.3 EDTA as Metal Chelate Complexes	70

## **CHAPTER THREE: METHODOLOGY**

3.1	Introduction	74
3.2	Materials and Chemicals	74
3.3	Instruments and Apparatus	75
3.4	Preparation of Stainless Steel Substrates	75
3.5	Preparation of Electrolyte Solutions	75
3.6	Electrochemical Studies of Copper on Stainless Steel Substrate	76
3.7	Characterization of Copper Coatings	77
3.8	Dissolution Rate Measurement	79
3.9	Antimicrobial Testing	80
3.10	Theoretical Framework	82

## CHAPTER FOUR: RESULTS AND DISCUSSION

4.1	Introc	83	
4.2	Stainl	83	
4.3	Stabil	87	
4.4	Electr	rodeposition using Uncomplexed CuSO <sub>4</sub>	92
	4.4.1	Cyclic Voltammetry Analysis	92
		4.4.1.1 Effect of Adding H <sub>2</sub> SO <sub>4</sub>	94
		4.4.1.2 Effect of pH	97
		4.4.1.3 Chronology Events during CV Scan	98
	4.4.2	Chronoamperometry Study	100
		4.4.2.1 Effect of Applied Potential	101