

AMINE-FUNCTIONALIZED SILVER-EXCHANGED ZEOLITE NaY AS
ANTIBACTERIAL AGENT

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A thesis submitted in fulfilment of the
requirements for the award of the degree of
Doctor of Philosophy (Biosciences)

Faculty of Biosciences & Medical Engineering
Universiti Teknologi Malaysia

MARCH 2017

I would like to dedicate my thesis to:

My loving parents

Mohd Hanim bin Osman

Azlana bte Amat

My beloved husband

Mohd Firdaus bin Mohd Jaafar

whose affection, love, encouragement and prays of day and night enabled me to get
such success and honor

ACKNOWLEDGEMENT

First and foremost, I want to send my deepest gratitude to Allah the Almighty for always giving me the strength to complete my study.

I sincerely want to thank my supervisor, Dr. Nik Ahmad Nizam bin Nik Malek, for his advice, his support, his patience and for giving me the great opportunity to work with him. I am deeply grateful for his understanding at all times and his continuous encouragement even for small achievements.

I also want to thank Prof. Dr. Zaharah binti Ibrahim for her supervision during my study. The help of my lab mates, for always giving me motivation and strength during lab works have been invaluable and are deeply appreciated.

Many thanks to our lab staff, En. Hasrul bin Ishak, for his assistance on every technical problem in the development of my experiments. Not forgetting, Cik Zaleha, the officer in charge during my whole period of study for always reminding and managing the process involved in finishing my study.

Special thanks to the Ministry of Higher Education Malaysia on MyPhD 15 for their financial support in my studies and research.

Finally, thank you so much to my parents Mohd Hanim bin Osman and Azlena bte Amat, my mother in law, Siti Jumiah binti Saripin and very specially my lovely husband Mohd Firdaus bin Jaafar, for understanding, listening, supporting and encouraging me at all times.

ABSTRACT

Bacterial resistance to antibacterial agents has becoming a serious concern worldwide. Current single-approach antibacterial agent is no longer effective towards these resistant bacteria. Hence, the aim of this research was to produce a newly modified material with multi-approach antibacterial agent with enhanced performance. The material comprised of zeolite NaY (CBV 100) as the carrier for two antibacterial compounds; silver (Ag^+) and 3-aminopropyltriethoxysilane (APTES), a type of silane coupling agent was studied. The preparation of amine-functionalized silver-exchanged zeolite (ZSA) began with the ion exchange at different Ag concentrations (25, 50, 100, and 200%) based on the zeolite cation exchange capacity (CEC) (CEC: 255 meq/100 g) producing silver-exchanged zeolites (ZS), which were then functionalized with different APTES concentrations (0.01, 0.2 and 0.4 M). All prepared materials were characterized according to their structural, morphological, elemental analysis and physicochemical properties related to their usage as a carrier and antibacterial agent. Characterization results of ZSA showed that the zeolite framework was not distorted after the modifications while the Ag-exchanged zeolite was successfully functionalized with APTES. The antibacterial activity of ZSA was investigated by using several antibacterial assays including the minimum inhibitory concentration (MIC) test, disc diffusion test (DDT) and inhibition growth study (IGS) against four types of bacteria, *Escherichia coli* (ATCC 11229), *Pseudomonas aeruginosa* (ATCC 15422); *Staphylococcus aureus* (ATCC 6538) and *Enterococcus faecalis* (ATCC 29212). All antibacterial assays showed that ZSA samples have higher antibacterial activity compared to ZS samples. Results also showed that ZSA samples were more effective towards the Gram negative bacteria compared to the Gram positive bacteria. This is possibly due to the thin peptidoglycan layer of Gram negative cell wall. The ZSA-50-0.2 (zeolite with 50% CEC Ag^+ and 0.2 M APTES) had highest antibacterial activity compared to others. The Ag^+ release study was carried out for ZSA-50-0.2 and ZS50 in order to study the mechanism of the antibacterial activity of ZSA and ZS. Different parameters were studied including incubation period, sodium chloride solution concentrations and type of bacteria. Results showed that Ag^+ released from ZSA was very low compared to ZS at 30 minutes and 24 hours of incubation. However, there was no significant effect from the type of bacteria and sodium chloride solution concentrations towards the Ag^+ released for both ZSA and ZS. These results proved that amine-functionalized silver-exchanged zeolite (ZSA) which is a multi-approach antibacterial agent have higher antibacterial activity than ZS, a single-approach antibacterial agent and hence, ZSA could possibly be used as an alternative antibacterial agent.

ABSTRAK

Kerintangan bakteria terhadap agen antibakteria adalah satu perkara yang membimbangkan kepada seluruh dunia. Pendekatan agen antibakteria pada masa kini tidak lagi berkesan terhadap bakteria ini. Oleh itu, tujuan kajian ini adalah untuk menghasilkan bahan baru yang diubahsuai dengan agen antibakteria yang mempunyai pelbagai pendekatan dengan prestasi yang dipertingkatkan. Bahan baru ini terdiri daripada zeolit NaY (CBV 100) sebagai pengangkut bagi dua sebatian antibakteria; perak (Ag^+) dan 3-aminopropyltriethoxysilane (APTES), sejenis agen gandingan silana telah dikaji. Penyediaan zeolit perak difungsikan dengan amina (ZSA) bermula dengan pertukaran ion pada kepekatan Ag yang berbeza (25, 50, 100 dan 200%) berdasarkan kapasiti pertukaran kation (CEC) (CEC: 255 meq/100 g) menghasilkan zeolit mengandungi perak (ZS), yang kemudiannya difungsikan pada kepekatan APTES yang berbeza (0.01, 0.2 dan 0.4 M). Semua bahan yang disediakan dicirikan mengikut struktur, morfologi, analisis unsur dan ciri-ciri fizikokimia yang berkaitan dengan penggunaannya sebagai pembawa dan agen antibakteria. Keputusan pencirian ZSA menunjukkan bahawa kerangka zeolit itu tidak berubah selepas pengubahsuaian manakala zeolit yang mengandungi perak telah berjaya difungsikan dengan APTES. Aktiviti antibakteria ZSA telah dikaji dengan menggunakan beberapa ujian antibakteria termasuk ujian kepekatan perencatan minima (MIC), ujian cakera penyebaran (DDT) dan kajian perencatan pertumbuhan (IGS) terhadap empat jenis bakteria, iaitu *Escherichia coli* (ATCC 11229), *Pseudomonas aeruginosa* (ATCC 15422), *Staphylococcus aureus* (ATCC 6538) dan *Enterococcus faecalis* (ATCC 29212). Kesemua ujian antibakteria menunjukkan bahawa sampel ZSA mempunyai aktiviti antibakteria yang lebih tinggi berbanding sampel ZS. Hasil kajian juga menunjukkan bahawa sampel ZSA lebih efektif terhadap bakteria Gram negatif berbanding Gram positif. Ini berkemungkinan kerana lapisan peptidoglikan nipis dalam dinding sel bakteria Gram negatif. ZSA-50-0.2 (zeolit dengan 50% CEC Ag dan 0.2 M APTES) mempunyai ciri-ciri aktiviti antibakteria yang paling optimum berbanding sampel lain. Kajian pelepasan ion perak telah dijalankan untuk ZSA-50-0.2 dan ZS50 untuk mengkaji mekanisme aktiviti antibakteria ZSA dan ZS. Parameter yang berbeza telah dikaji iaitu tempoh pengeraman, kepekatan natrium klorida dan jenis bakteria. Hasil kajian menunjukkan bahawa ion perak yang dikeluarkan oleh ZSA adalah lebih rendah berbanding ZS pada masa pengeraman 30 minit dan 24 jam. Walau bagaimanapun, tidak ada kesan yang ketara daripada jenis bakteria dan kepekatan natrium klorida ke arah ion perak yang dikeluarkan oleh ZSA dan ZS. Keputusan ini membuktikan bahawa zeolit perak yang difungsikan dengan amina (ZSA) dengan pelbagai pendekatan agen antibakteria mempunyai aktiviti antibakteria yang lebih tinggi daripada ZS, iaitu agen antibakteria dengan satu pendekatan dan dengan itu, ZSA mungkin boleh digunakan sebagai agen antibakteria yang alternatif.

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LIST OF SYMBOLS

°C	-	Degree Celsius
cm	-	Centimeter
g	-	Gram
L	-	Liter
M	-	Molar
m	-	Meter
mg	-	Miligram
min	-	Minute
h	-	Hour
mL	-	Mililiter
mm	-	Milimeter
nm	-	Nanometer
mV	-	Milivolt
kV	-	Kilo volt
rpm	-	Rotation per minute
v	-	Volume
v/v	-	Volume per volume
Å	-	Angstrom
μL	-	Microliter
λ	-	Lambda
Θ	-	Theta
°	-	Degree

LIST OF ABBREVIATIONS

APTES	-	3-aminopropyltriethoxysilane
BET	-	Brunauer, Emmett & Teller
CEC	-	Cation exchange capacity
CFU	-	Colony forming unit
DDT	-	Disc diffusion test
EDX	-	Energy dispersive x-ray
FTIR	-	Fourier transform infrared
GFAAS	-	Graphite furnace atomic absorption spectrophotometer
IGS	-	Inhibition growth study
IR	-	Infrared
LB	-	Luria-Bertani
MHA	-	Mueller Hinton agar
MIC	-	Minimum inhibition concentration
MRSA	-	Methicillin resistant <i>Staphylococcus aureus</i>
NA	-	Nutrient agar
ND	-	Not detected
OD	-	Optical density
TGA	-	Thermogravimetric analysis
XRD	-	X-ray diffraction
Z	-	Zeolite
ZP	-	Zeta potential
ZS	-	Silver-exchanged zeolite
ZSA	-	Amine-functionalized silver-exchanged zeolite

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CHAPTER 1

INTRODUCTION

1.1 Introduction

In the present time, technologies in biomedical and health sciences area are developing rapidly following the development of technologies in many areas. As a developing country, Malaysia is not an exception from the development achieved by other developed countries. Biomedical areas have experienced much progress in the development from the slightest to the very life changing development. For example, with the presence of technologies such as artificial body part replacements and implants such as artificial pacemaker, artificial heart valve, arm prostheses and leg prostheses have saved many lives and also reduced the death rate apart from giving a better life towards patients. The use of these devices however requires operation procedures to enable the devices to be placed into the patient's body. The operation scale and complexity depend on the type of implant that will be implanted. This operation however comes with a risk. Attention needs to be given in every operation due to the risk of infection by microorganism. Every operation being carried out must be ensured in a sterile surrounding with sterile equipment and apparatus. All surfaces including operation table, walls, ceilings and everything in the operation room must be sterilised and free of microorganisms such as bacteria and fungi. This is to avoid the occurrence of microorganism infection towards the patient especially by resistant microorganism which can endanger the patient's life or prolong their hospital stay and hence increase their healthcare cost. Hence, the control of microorganism must be carried out not only during operation procedures, but also around the clock in healthcare institutions. New and improved ways to combat bacterial resistance must be studied and discovered to get ahead of the bacteria which is capable of evolving

and acquiring resistant. Knowledge sharing between various fields can benefit all of us. Knowledge in materials science can be applied in biomedical field as well as medical microbiology. Materials science is an interdisciplinary field concerning the study and design of new materials especially solids while biomedical field is a field that combines medicine and biology for healthcare purposes. Materials science knowledge can complement the biomedical field in a way that the implants and medical devices can be made from materials with antibacterial properties. In this way, early prevention of bacterial infection can be made. These antibacterial materials have been introduced specifically to prevent the related diseases. New antibacterial agents will continuously emerge as long as infections and diseases exist within the communities because people are always looking for a better cure. Therefore, this study is an effort that can be done to improve the quality of human health and life as this study aims to develop a new and perhaps a better antibacterial agent that employs an improved mechanism in its antibacterial action.

1.2 Problem Statement

The treatment of bacterial infections is becoming more complicated due to the ability of bacteria to develop resistance towards antimicrobial agent. Antimicrobial resistance is a natural biological phenomenon of response from microbes including bacteria, parasites, fungi and viruses towards antimicrobial agent (Sharma *et al.*, 2005). For instance, there is a report by Tenover (2006) that common bacteria with antibacterial resistance in healthcare institutions are *Staphylococcus aureus*, *Escherichia coli* and *Pseudomonas aeruginosa*. Antimicrobial resistance is a concern because it often causes treatment failures in healthcare institutions (Tenover, 2006). Resistance increases morbidity, mortality and cost, which can cause serious economic, social, and political implications. Current antimicrobial agents used to kill bacteria such as ethanol, silver nitrate and surfactant always have drawbacks. For example, ethanol can cause skin irritation, volatile and inflammable. Besides that, to be effective, the contact time on the surface must be at least 20 minutes, which is difficult because it can easily evaporate even at room temperature (Tilton and Kauffman, 2004). According to Livermore (2003), there is a need for the

development of new antibacterial agents to keep ahead of the bacteria. With the relative absence of new antimicrobials available to the market and the increasing frequency of antimicrobial resistance, efforts must be increased to intensify the search for new therapeutics (Levy and Marshall, 2004). Thus, research for a better antibacterial agent should be performed nowadays.

Zeolites have been widely used in many applications. Although natural zeolites are abundant and cheap, they hold many drawbacks as compared to synthetic zeolites. Natural zeolites and synthetic zeolites resemble one another and are similar in applications. However, the natural zeolites have variable phases of purity and contaminated by the chemical impurities from other minerals, which are costly to be removed and this make synthetic zeolites to be more attractive for specific applications where uniformity and purity are very important. Natural zeolites also have lower surface area (Payra and Dutta, 2004). On the other hand, synthetic zeolites are high in purity, with larger internal pore volumes, molecular-size pores, regular crystal structures, diverse framework chemical compositions and have large cation exchange capacity (CEC) depending on the types of zeolites due to a lower Si/Al ratio (Sherman, 1999; Yusof and Malek, 2009). This make synthetic zeolites with low Si/Al ratio such as zeolites A, X or Y interesting to be used as an adsorbent for the modification of zeolite in this research as it has a higher adsorption capacity for polar molecules and provide more exchange sites. However, zeolite Y shows to be a more promising type in this research as zeolite Y has large pore, high cation exchange capacity (CEC) value, low Si/Al ratio and have high crystallinity. Zeolite NaY was chosen because the high CEC of zeolite enables large amount of silver ion to be exchanged into the framework structure of the zeolite, which is important in this research to produce an antibacterial agent with high and improved antibacterial activity.

Silver is the most powerful metal ion with antibacterial properties as compared to the other heavy metal-ions and it shows an oligodynamic effect with a minimum development of bacterial resistance and low toxicity (Bastan and Ozbek, 2013). Silver has been used in medical applications especially in the treatment of burn wounds in the form of silver nitrate. However, simple silver (I) salts can

precipitate in the solution as silver chloride, causing irritation to the wound area and also reduces the antibacterial activity of the silver (Clement and Jarrett, 1994). It is also often ineffective when bacterial infection has established. Moreover, the use of silver itself can cause argyria, a type of skin condition where the colour turns grey caused by the accumulation of silver on the skin (Baker *et al.*, 2011). Increasing the concentration of silver incorporated in materials such as catheters, prostheses and tubes, increases the antimicrobial effect, but its cytotoxicity effects will also increase. Hence, to overcome the potential negative effects of silver, the incorporation of a secondary chemical or support system is needed and this is where zeolite plays this role (Bastan and Ozbek, 2013). Silver-loaded zeolites act as an inorganic reservoir and release silver ions in a controlled way in exchange for other cations (Kwakye-Awuah *et al.*, 2007).

There have been numerous studies on silver-loaded zeolites. Silver loaded-zeolites have been used mainly for the purpose of killing or inhibiting bacterial growth. One such study is the study of antibacterial activity of silver-zeolite against oral microorganisms such as *Streptococcus mutans*, *Lactobacillus casei*, *Candida albicans* and *Staphylococcus aureus* using the disc diffusion assay, minimum inhibition concentration and minimum lethal concentration. This study concluded that silver-zeolite had antibacterial effects towards the oral microorganism and that it may be useful to be applied to oral hygiene products for protection against oral infection (Saengmee-Anupharb *et al.* 2013). Although zeolite acts as a reservoir for the silver ions, silver ions tend to leach out from the zeolite over time and also precipitate out in the presence of chloride ions (Marambio-Jones and Hoek 2010). Hence, to overcome this problem, surface functionalization of zeolite might be the solution which is a technique in modifying the surface of materials by adding extra functionalities to overcome the material shortcomings in order to be used for a particular application (Treccani *et al.* 2013). The most popular technique is by silanization by silane coupling agent such as 3-aminopropyltriethoxysilane (APTES). The adsorption of silane coupling agents on the surface of zeolite might reduce the leaching of silver ions into the solution because the narrowing of pores by the silane molecules situated in the zeolite surface (Nik *et al.* 2012). This technique could also help in reducing the precipitation of silver ions by chloride ions. Silane molecules

could bind to the anions in the solution especially chloride ions and therefore preventing the binding of chloride ions with silver ions (Kang *et al.* 2009; Emami Khansari *et al.* 2015).

With the knowledge that aminosilane and silver possess the antibacterial activities, the material that will be developed in this study is a combination of aminosilane, silver and zeolite because aminosilane and silver need zeolite as a support system. Hence, this research has been conducted by expecting that aminosilane can effectively enhance the antibacterial activity of silver-zeolite.

1.3 Objectives of Research

There are four main objectives in this study:

- 1) To prepare and characterize 3-aminopropyltriethoxysilane (APTES) functionalized NaY zeolite with different APTES concentrations.
- 2) To study the antibacterial activity of APTES functionalized-zeolite.
- 3) To prepare and characterize APTES-functionalized silver-exchanged NaY zeolite at different concentrations of APTES and silver.
- 4) To study the antibacterial activity and mechanism of APTES-functionalized silver-exchanged NaY zeolite.

1.4 Scope of Research

This research focuses on the application of modified zeolite NaY as a new antibacterial agent. The antibacterial properties of APTES-functionalized zeolite (ZA) were firstly determined by disc diffusion method (DDT) after its preparation and characterization. The zeolite was firstly modified with silver and then, the silver-zeolite was functionalized with APTES. The silver-zeolite was prepared by adding NaY zeolite with four known concentrations of silver nitrate according to the zeolite cation exchange capacity (CEC) which were 25%, 50%, 100% and 200% for ion

exchange process of silver ions with the cations in the zeolite to occur. Next, the silver-zeolite was functionalized using different concentrations of 3-aminopropyltrimethoxysilane (APTES) which were 0.01 M, 0.2 M and 0.4 M. The APTES-functionalized silver-exchanged zeolite NaY (ZSA) was then characterized using Fourier transform infrared spectroscopy (FTIR), field emission scanning electron microscopy (FESEM), energy dispersive X-ray (EDX), X-ray Diffraction (XRD), Brunauer, Emmett and Teller (BET), and thermogravimetric analysis (TGA) to determine its elemental, structural and morphology characteristics. The material was then tested for its antibacterial activity towards Gram-positive and Gram-negative bacteria including *E. coli* ATCC 11229, *P. aeruginosa* ATCC 15442, *S. aureus* ATCC 6538 and *E. faecalis* ATCC 29212 by disk diffusion test (DDT), minimum inhibitory concentration (MIC) test and growth inhibition study (GIS). The antibacterial activities of all prepared materials were compared with each other to determine the most effective antibacterial agent. Finally, the antibacterial and silver release properties of ZSA were studied intensively by conducting MIC at different concentrations of saline solution and silver release study against several parameters such as incubation time, type of bacteria and concentration of sodium chloride solution.

1.5 Outline of Research

This overall research methodology was designed so that each objective could be achieved in the expected time frame. Research design was divided into eight stages in order to achieve each objective. Stages 1 to 3 were discussed in chapter 3 and stages 4 to 8 were discussed in chapter 4 in this thesis. The flow diagram for the outline of research can be seen in Figure 1.1.

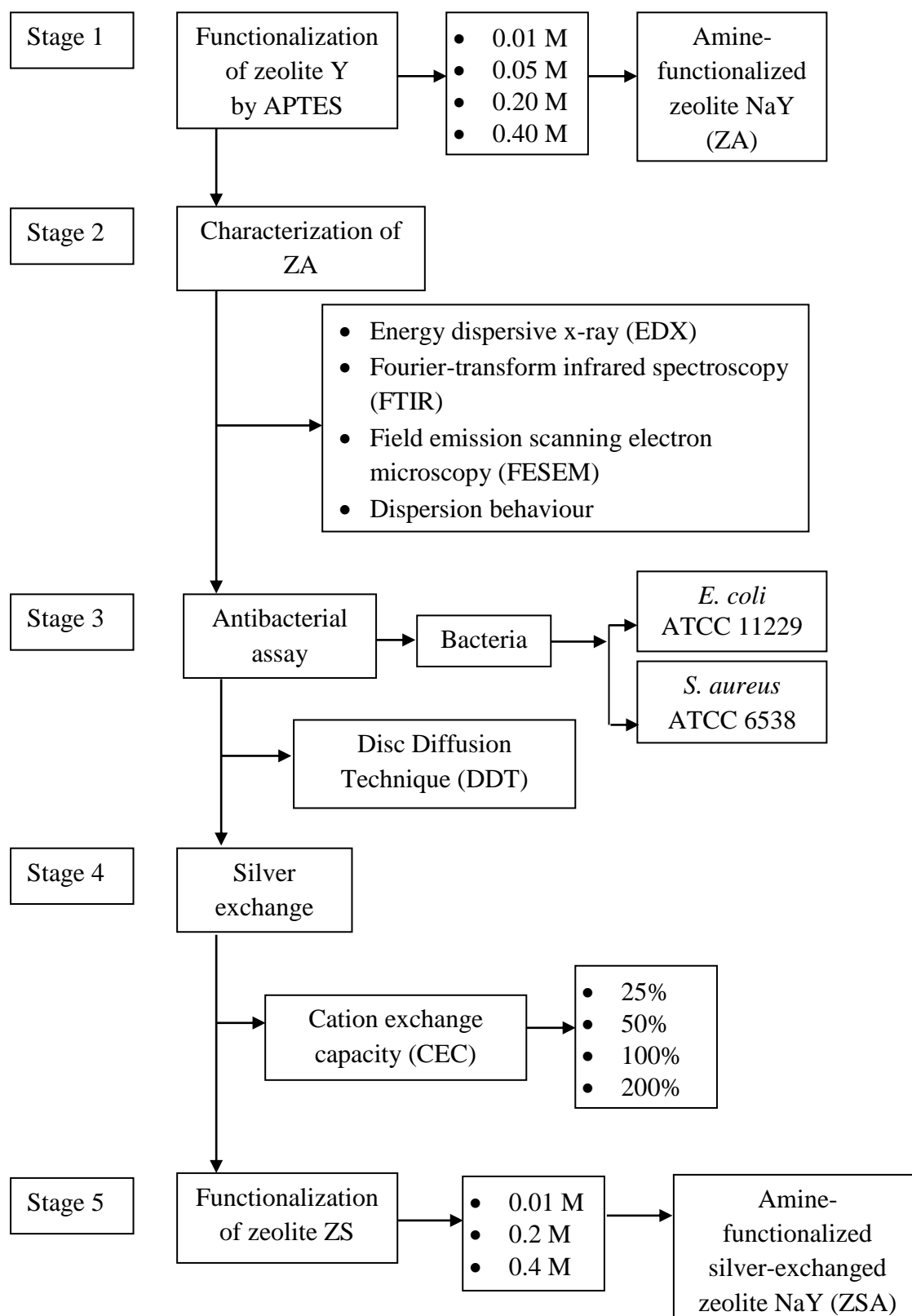


Figure 1.1 Flow diagram of the research outline

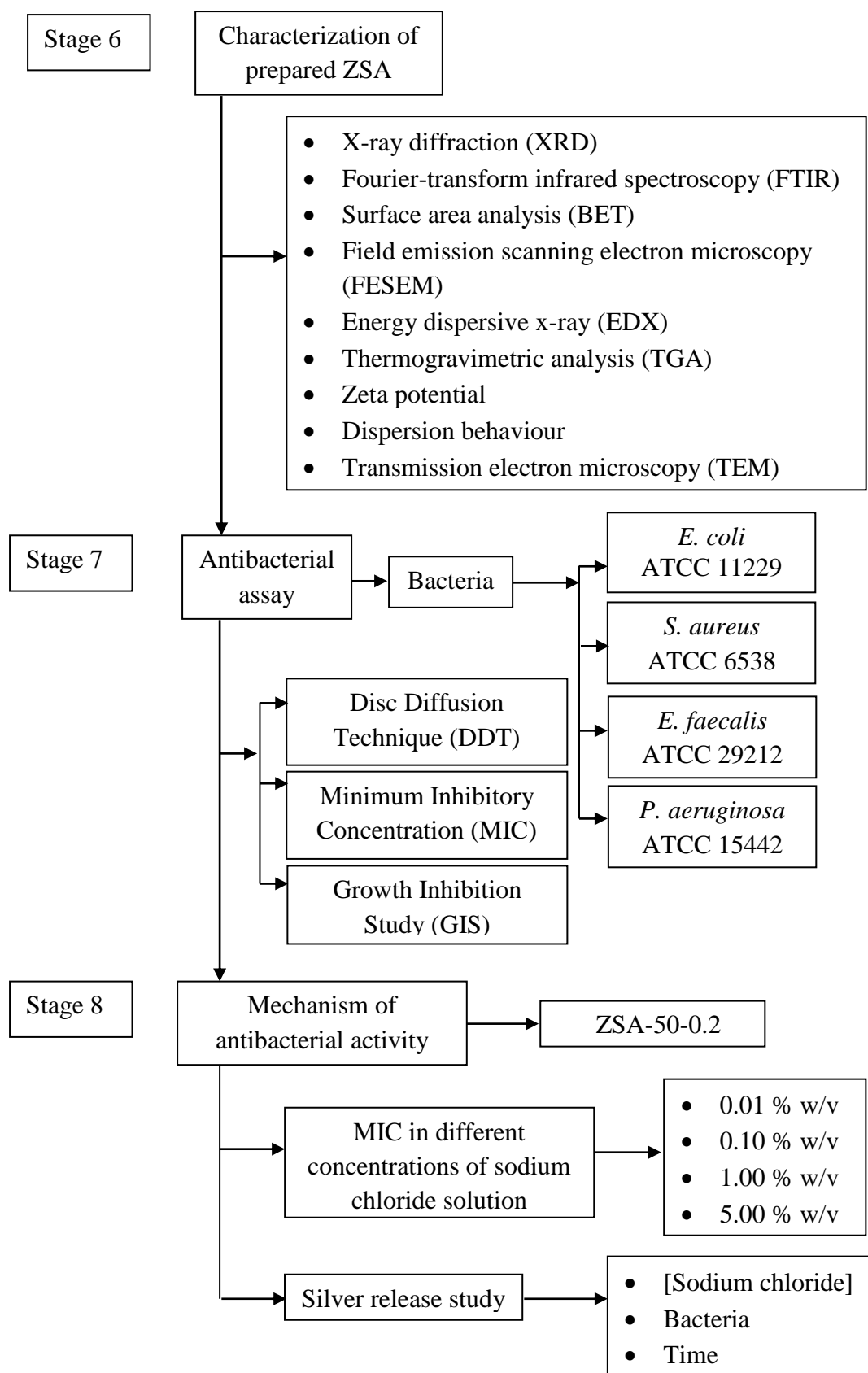


Figure 1.1 (cont) Flow diagram of the research outline

1.6 Research Significance

Development of a new antibacterial agent is needed to combat the antimicrobial resistance and keep ahead of the bacteria. The antibacterial agent must be efficient in its antibacterial activity to reduce the occurrence of antimicrobial resistance. From this research, an antibacterial agent with an improved antibacterial activity was developed. This antibacterial agent has a broad-spectrum in which it is able to kill both Gram-positive and Gram-negative bacteria. Two antibacterial compounds, aminosilane and silver ion were combined in one carrier system and these compounds work synergistically in its bactericidal activity resulting in a higher bacterial death. Besides that, this material has a multi-approach antibacterial activity, by contact-killing and release-based approach, hence having a higher efficiency as an antibacterial agent. The high effectiveness and efficiency of an antibacterial especially those that have more than one type of bacteria-killing mechanisms could avoid the occurrence of antibacterial resistance because during the course of its application as an antibacterial agent, very low amount to no bacteria will be able to survive the antibacterial effect and hence, mutation or resistance will be less likely acquired by the bacteria strain. Antibacterial resistance is a very serious problem that brings many negative implications to the community by posing clinical problems and life-threatening infections. This problem of antibacterial resistance can only be addressed by development of new antibacterial agents that are more powerful and effective. Hence, the wide usage of ZSA as a new alternative for antibacterial agent can very much reduce the occurrence of bacterial resistance towards the antibacterial agents. Finally, this may also start a new demand for the research, development and production of multi-approach antibacterial rather than single-approach antibacterial agent in which its ultimate drawbacks is the higher possibility of occurrence of antibacterial resistance.

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