CATALYTIC CHELATING TECHNIQUE ON LEAD, NICKEL AND CADMIUM REMOVAL FROM *FENNEROPENAEUS MERGUIENSIS*

ISMAIRUL IZAD BIN ISMAIL

UNIVERSITI TEKNOLOGI MALAYSIA

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ISMAIRUL IZAD BIN ISMAIL

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For the love one and his prophet.

Erni Johan, Aleeya Hanna and Aleesya Hanna.

My mother and family.

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ABSTRACT

Fenneropenaeus merguiensis is one of the main sources of protein in Malaysian dietary. The heavy metals content (0.8495, 0.4923 and 0.0854 μg/g for lead, nickel and cadmium respectively) in *F. merguiensis* were below permissible limit of Malaysia Food Regulations (1985) and EU Food Regulations. However, as the pollution increased at tremendous rate, continuing uptake of food containing heavy metals may affect human health. In this study, chelation and catalytic chelation techniques were applied to remove Pb, Ni and Cd in *F. merguiensis*. The heavy metals analyses were performed using flame atomic absorption spectroscopy (FAAS). The chelation technique was carried out at three different parameters; chelating agent dosage, treatment time and treatment temperature. Three chelating agents used were trisodium citrate, disodium oxalate and sodium acetate. The study revealed that the optimum conditions of chelation technique were at 29.5°C, 2 hours treatment time and 600 mg/L of trisodium citrate. Percentage of removal of Pb, Ni and Cd were 71.51%, 42.82% and 44.85% respectively. The introduction of CaO/Al_2O_3 and MgO/Al_2O_3 catalysts had enhanced the removal capacity of the heavy metals in *F. merguiensis.* The catalytic chelation technique was conducted at optimum condition of chelation technique but the treatment time was changed to 1 hour, to maintain the freshness of *F. merguiensis.* The percentage of removal of Pb and Cd was higher when CaO/Al_2O_3 was used with percentage removal were 90.23% and 94.95% respectively, while the percentage of removal of Ni was 59.09% when MgO/Al_2O_3 was used as catalyst. Percentage of Pb removal obtained from Central Composite Design (CCD) is in good agreement with experimental result, which *p*-value (<0.0001) indicated that, the percentage of Pb removal is statistically significant.

ABSTRAK

Fenneropenaeus merguiensis merupakan salah satu sumber protein dalam diet seharian rakyat Malaysia. Kandungan logam berat (0.8495, 0.4923 dan 0.0854 μg/g masing-masingnya untuk plumbum, nikel dan kadmium) di dalam *F. merguiensis* adalah lebih rendah daripada had yang telah ditetapkan oleh Peraturan Makanan Malaysia (1985) dan Peraturan Makanan EU. Namun begitu, tahap pencemaran yang sentiasa meningkat dan pengambilan berterusan makanan yang mengandungi logam berat boleh menjejaskan kesihatan pengguna. Di dalam kajian ini, teknik pengkelatan dan pengkelatan bermangkin digunakan untuk menyingkirkan kandungan Pb, Ni dan Cd di dalam *F. merguiensis*. Analisa kandungan logam berat dilakukan menggunakan teknik spektroskopi serapan nyalaan atom (FAAS). Tiga parameter kajian bagi kaedah pengkelatan yang digunakan iaitu kepekatan egen pengkelat, masa rawatan dan suhu rawatan. Tiga ejen pengkelat yang digunakan adalah trinatrium sitrat, dinatrium oksalat dan natrium asetat. Kajian mendapati, keadaan optimum bagi kaedah pengkelatan adalah pada suhu 29.5°C, 2 jam masa rawatan dan menggunakan 600 mg/L trinatrium sitrat. Peratus penyingkiran Pb, Ni dan Cd adalah masing-masing 71.51%, 42.82% dan 44.85%. Kehadiran mangkin CaO/Al_2O_3 dan MgO/Al_2O_3 telah meningkatkan peratus penyingkiran logam berat. Kaedah pengkelatan bermangkin dijalankan pada keadaan optimum kaedah pengkelatan dengan pengubahsuaian masa rawatan dari 2 jam kepada 1 jam bagi menjamin kesegaran *F. merguiensis* yang digunakan. Penggunaan mangkin CaO/Al_2O_3 memberikan penyingkiran tertinggi bagi logam Pb dan logam Cd dengan masing-masing 90.23% dan 94.95% penyingkiran, manakala penggunaan mangkin MgO/Al_2O_3 memberikan peratus tertinggi penyingkiran logam Ni dengan nilai 59.09%. Peratus penyingkiran menggunakan model Central Composite Design (CCD) menunjukkan keputusan yang sama seperti yang diperolehi melalui kaedah ujikaji, dengan nilai *p* (<0.0001), yang menunjukkan kajian ini diterima secara statistik.

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LIST OF ABBREVIATIONS

Al	-	Aluminium
Al_2O_3	-	Aluminium Oxide
ANOVA	-	Analysis of Variance
As	-	Arsenic
ATSDR	-	Agency for Toxic Substances and Disease Registry
BBD	-	Box-Behnken Design
BET	-	Brunauer-Emmett-Teller
Br	-	Bromine
bw	-	Body weight
CaO	-	Calcium Oxide
CCD	-	Central Composite Design
Cd	-	Cadmium
CHC	-	Cysteamine Hydrochloride
Cl	-	Chlorine
Cr	-	Chromium
Cu	-	Copper
$CuSO_4$	-	Copper(II) Sulfate
DFO	-	Deferroxamine
DFP	-	Deferiprone
DMPS	-	2, 3-dimercaptopropane-1-sulfonate
DOE	-	Department of Environmental
DTC	-	Sodium Dimethyldithiocarbamate
DTPA	-	Diethylenetriaminepentaacetate
EDDS	-	Ethylenediaminedisuccinic
EDTA	-	Ethylenediaminetetraacetic acid
EDX	-	Energy Dispersive X-ray
EU	-	European Union

FAAS	-	Flame Absorption Atomic Spectroscopy
FAO	-	Food and Agriculture Organization
Fe	-	Iron
FESEM	-	Field Emission Scanning Electron Microscopy
FTIR	-	Fourier Transform Infrared Spectroscopy
HD	-	Historical Design
Hg	-	Mercury
ICP-AES	-	Inductively Coupled Plasma-Atomic Emission Spectroscopy
ICP-MS	-	Inductively Coupled Plasma-Mass Spectroscopy
JECFA	-	Joint Food and Agriculture Organization and World Health Organization Expert Committee on Food Additives
MFR	-	Malaysia Food Regulation s
MgO	-	Magnesium Oxide
Mn	-	Manganese
MRT	-	Multiple Range Test
MSWI	-	Municipal Solid Waste Incineration
MTs	-	Metallothioneins
NaOH	-	Sodium Hydroxide
Ni	-	Nickel
$NiSO_4$	-	Nickel(II) Sulfate
NTA	-	Nitrilotriacetate
OFD	-	One Factor Design
PBD	-	Potassium Butyl Dithiophosphate
PMDA	-	Pyromellitic Dianhydride
PTWI	-	Provisional Tolerable Weekly Intake
Rb	-	Rubidium
RSM	-	Research Surface Methodology
SEM	-	Scanning Electron Microscope
Sr	-	Strontium
TBA	-	Tetrathio Bicarbamic Acid
TMBTCA	-	Benzene-1,2,4,5-tetracarboxamide
TMT	-	Tetramethylthiuram

UNEP	-	United Nations Environment Programme
USD	-	US Dollar
WB	-	Weight Base
XRD	-	X-Ray Diffraction Spectroscopy
Zn	-	Zinc

LIST OF SYMBOLS

°C	-	Degree celcius
cm	-	Centimetre
g	-	Gram
g/kg	-	Gram per kilogram
L^{n-}	-	Ligand with n charge
M^{m+}	-	Metal ion with m charge
$[M]_{initial}$	-	Initial concentration
$[M]_{after\ removal}$	-	Concentration after treatment
mg/kg	-	Miligram per kilogram
mg/L	-	Miligram per liter
$ML^{(m-n)}$	-	Chelated complex
mL	-	Mililiter
mm	-	Milimeter
m^2/g	-	Meter square per gram
ppm	-	Part per million
r/min	-	Revolutions per minute
USD	-	US dollar
v/v	-	Volume per volume
WB	-	Weight base
µg/g	-	Micro gram per gram
%	-	Percentage

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CHAPTER 1

INTRODUCTION

1.1 Introduction

Seafood is an important sources of protein in Malaysians diet. Fish, crab, cockles, squid and prawn are a few examples of normal seafood in Malaysian dietary. It is popular among Malaysians because it is considered as a healthy food due to their high protein and omega fatty acid with low saturated fat contents (Gu *et al*., 2015). It can easily found in the markets with various ranges of prices depends on the quality and type of seafood. There are two sources of this seafood, either it is from aquaculture or harvested from the sea or river. Naturally, aquaculture seafood is cheaper than capture fisheries products.

The *Fenneropenaeus merguiensis* (Pe´rez-Farfante and Kansley, 1997) which previously known as Penaeus *merguiensis De man* (Holthuis, 1980) is one of the species that belong to the Penaeidae family of prawn that commonly referred as penaeid shrimp. One hundred twelve species of penaeid shrimps can be found in the Western Central Pacific. *F. merguiensis* and *F. monodon* are two species in Penaeidae family is most commercially and economically important (Carpenter and Niem, 1998). In South East Asia, Philipines and Thailand are two countries that provide main fishing ground for Penaeidae family (Naylor *et al*., 2000). *F. merguiensis* species are found in different parts of the world, from the Persian Gulf to Thailand, found in Indonesia, Philippines, New Guinea, North Australia, Hong Kong and New Caledonia (Grey *et al*., 1983) is shown in Figure 1.1.

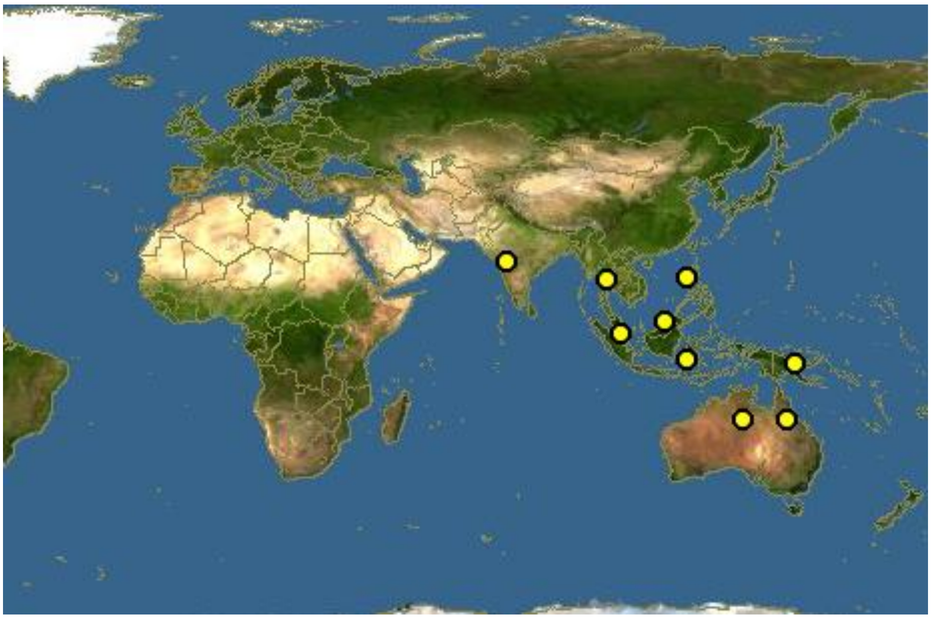

Figure 1.1 Geographical distribution of *F. merguiensis*

In Malaysia, *F. merguiensis* species (Figure 1.2), are not only found along the Malacca Straits and by the west and east coasts of the Malaysian peninsular but also in Sarawak and Sabah. It is locally known as udang kaki merah. In Indonesia, *F. merguiensis* is known as udang putih or udang jerbung, Thailand (Kung chaebauy), Hong Kong (Pak ha) and in Australia as banana prawn (Holthuis, 1980).

Figure 1.2 The *Fenneropenaeus merguiensis*

F. merguiensis life cycle almost similar to the other family members of Penaeidae (Figure 1.3). It can be found both onshore and offshore. Adults prawn had higher tendency to do their spawning activities in open sea. Each individual *F. merguiensis* with carapace length between 28 cm to 45 cm can produce approximately 100000 to 450000 eggs and their peak spawning seasons are in August, January and April (Crocos and Kerr, 1983). The pelagic larvae will then migrate inshore. The growth process continues where the juveniles inhabit in mangrove-lined with muddy banks for several months before emigrating to offshore (Dall *et al*., 1990, Kenyon *et al*., 2004) and the abundance of juveniles *F. merguiensis* increase in the upper reaches small creek (Vance *et al*.,1998). For offshore, *F. merguiensis* can be found in a sea with depth range 10 to 45 metres. In India, it is found up to 150 metres depth in east and west coast of India but normally inhabit in less than 30 metres depth (Jose, 2013).

Mangrove-lined estuaries provide natural nursery habitat for juveniles *F. merguiensis* (Staples *et al*.,1991). During flood tide, this juveniles moved further up the mangrove forests (Robertson, 1988). The matured *F. merguiensis* will later emigrate offshore but adjacent to mangrove-lined forests where they were juveniles (Rao *et al*., 1993).

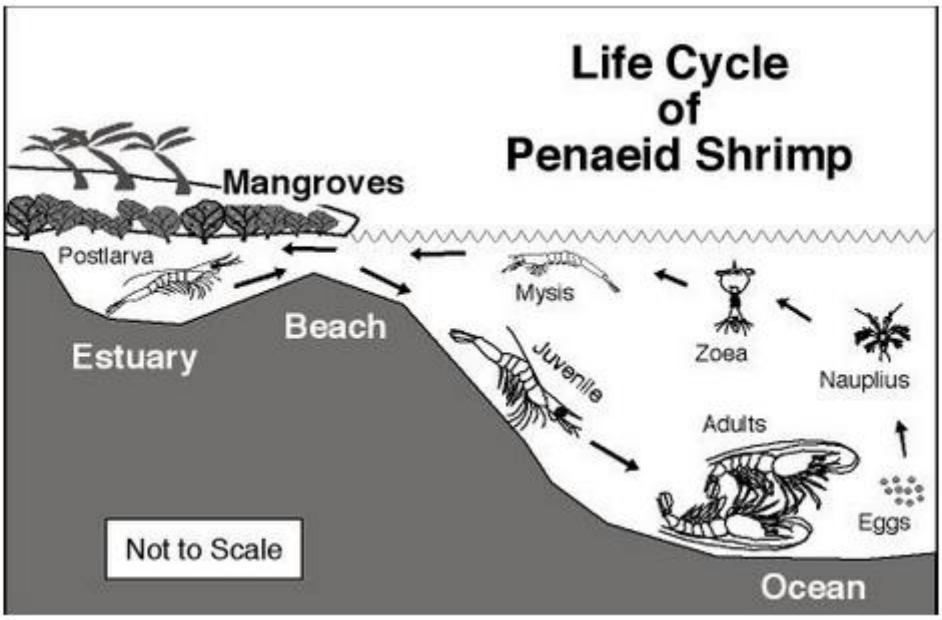

Figure 1.3 The life cycle of *Penaeid Shrimp* (Rosenberry, 2009)

Other than *F. merguiensis*, there was Indian white prawns, *F. indicus* that occupy the same area (Kenyon *et al*., 2004) and shared the same life cycle, therefore it is difficult to distinguish between these two species using their morphological characters (Pendrey *et al*., 1999). Taxonomic tree of *F. merguiensis* is shown in Table 1.1.

Table 1.1: The taxonomic tree of *Fenneropenaeus merguiensis*

Kingdom	Animalia
Phylum	Arthropoda
Subphylum	Crustacea
Class	Malacostraca
Subclass	Eumalacostraca
Order	Decapoda
Family	Penaeidae
Genus	Fenneropenaeus
Species	*Fenneropenaeus merguiensis*

1.2 Background of Problem

Malacca Straits are one of the most busy straits in the world, where number of ships using this lane increases from time to time. It is unfortunate that this strait becomes contaminated and polluted with Ni and Cu from petroleum spills and tanker wreckages that led to the accumulation of heavy metal in aquatic system (Santos-Echeandia *et al*., 2009). Urbanization also contributed to the contamination of the Malacca Straits. Pollutants discharged points along the straits increases through rapid urbanization where it received waste from both sea-based and land-bases sources (Abdullah *et al*., 1999) and also natural and anthropogenic (socio economic activities) sources (Yap *et al*., 2003).

Economic developments often contribute to marine pollutions. Industrialization and uncontrolled land use cause in increase of heavy metal (Cd, Cu, Pb and Hg) pollutants, especially in the littoral states of Malacca Straits like Pulau

Pinang, Selangor, Malacca and Perak (DOE, 2008). West Coast of Peninsular Malaysia also received heavy metal pollutant from manufacturing industry (Thia-Eng *et al*., 2000) and agricultural activities such as fertilizer and pesticides.

Two main activities that led to the contamination of Pb into Malacca straits are from waste-water treatment plants where waste-water is discharged based in Juru, Pulau Pinang (Shazili *et al*., 2006) and as a result from human activities (Yap *et al.*, 2003). Pulau Pinang is the national largest hub for electronic and semiconductor industries (UNEP, 2002). Production of batteries (nickel-cadmium) as part of electronic industries contributed to Cd pollutions along with mining and shipping activities (Nazli and Hashim, 2010).

1.3 Statement of Problem

F. merguiensis or banana prawn is one of popular seafood in Malaysia and very important sources of protein. Globally, prawn is one of the most traded fishery products that represent 15% of the total value of internationally traded fish product (FAO, 2010). More than 200000 tonnes of prawn are caught or produce from farms in Malaysia in 2010. The worldwide value of prawn that produced both from wild capture fisheries and aquaculture is almost 40000 million USD (FAO, 2014). Thus, shows that prawn has high demand, not only in Malaysia but across the world. The quality of the prawn had been affected by increasing concentration of heavy metal in its flesh due to contamination of seawater.

Various studies have been conducted by researchers on *F. merguiensis,* but mostly focused on the accumulation patterns of heavy metals on the species, however, no commercially technology was ever developed in terms of simplicity, practicality, quick, efficient and household safe and housewives-friendly; in the removal of heavy metal from contaminated *F. merguiensis*. This study is very important to develop new technology for removing heavy metal to sustain the quality of *F. merguiensis* and acquire the safe permissible limit stated by the Malaysia Food

Regulations (MFR, 1985) and Joint Food and Agriculture Organization and World Health Organization Expert Committee on Food Additives (FAO/WHO 1983, 1984) standards. In addition, the studied of catalyst to enhance removal of heavy metal from *F. merguiensis* in term of rates and chemical reactions are also necessary.

1.4 Significance of the Study

Human activities such as industrial and agricultural wastes are the main reason of the contaminating presence in aquatic life including *F. merguiensis* and consequently transferred the pollutants to human body through food chain. A few methods were used in removing heavy metal from the flesh tissue of aquatic life in the past such as depuration and cooking methods. The problem with this previous methods are poor removal of heavy metals, time consuming is not practical and unable to maintain the freshness of flesh tissue and probably loss on some importance mineral during the removal process.

This study is very important to reduce heavy metal toxicant in human bodies below the permissible amount of heavy metals such as lead, nickel and cadmium. Edible chelating agent extracted heavy metal from flesh tissue of *F. merguiensis* and form soluble chelate complex and capable to be removed from the flesh tissue without damaging the protein. In addition, the food grade catalyst enhanced capability of removal heavy metal and enable the process to accomplish in hours rather than days. Moreover, both chelation and catalytic chelation treatment are practicable process and can be done at convenient.

As a result, thus decreasing the amount of heavy metals in *F. merguiensis* and make it safe for humans and increase quality and economical value of *F. merguiensis* as it is one of the most important fisheries product of Malaysia. Furthermore, the catalytic chelation technique has huge potential of removal heavy metal in large scale contamination of aquatic life and becomes a standardized procedure in fisheries and food industries and normal practise in the household.

1.5 Objectives of the Study

The objectives of this study are to:

i. optimize the catalytic chelation treatment conditions for the removing of heavy metals from *F. merguiensis* while maintaining their quality.
ii. determine heavy metals (Pb, Ni and Cd) in *F. merguiensis* after treat *F. merguiensis* with edible chelating agents and food grade catalysts.

1.6 Scope of the Study

This study is to investigate the effectiveness of the catalytic chelation technique for removal of lead, cadmium and nickel from *F. merguiensis*. Samples were collected from Pantai Remis, Perak (Figure 1.4). Chelating agents used are sodium acetate, sodium oxalate and trisodium citrate. Two types of catalyst are CaO/Al_2O_3 and MgO/Al_2O_3.

For chelation treatment conditions, dosages of chelating agent, treatment time and treatment temperature are three parameters which were optimized. The optimal conditions of this three parameter, then monitored by Response Surface Methodology (RSM) for statistical analysis using Central Composite Desigin as a design of experiment. Lastly, for catalytic chelation treatment conditions, another two parameters which are types of catalyst and treatment time are optimized.

Figure 1.4 Sampling sites for *F. merguiensis* in Pantai Remis, Perak, Malaysia. (Azimah Ismail *et al.*, 2016)

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