

PHYTOREMEDIATION SYSTEM FOR TREATING METALS IN CERAMIC  
INDUSTRY WASTEWATER

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*Dear Allah, the most gracious and the most merciful*  
*Dedicated to who have been asking Allah for His blessing and guidance for me*  
*To my husband and my parents*

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## ABSTRACT

Phytoremediation system using aquatic plants is increasingly being applied by researchers due to its capability in metal removal. The study is aimed at evaluating the potential of selected aquatic plants by phytoremediation system to remove selected elements in ceramic industrial wastewater namely boron, copper, cadmium, chromium, ferum, manganese, and zinc. The system was tested with different types of plant, namely water hyacinth, water lettuce, and water spinach. Based on different plants, control and free flow method with hydroponic tank were applied in order to compare the effectiveness of circulation method by phytoremediation system. Phytoremediation system with applied volume of wastewater at  $0.024\text{m}^3$  using three columns of phyto-rig was prepared to treat contaminants in ceramic wastewater using different plants within 11 days of treatment. Metal concentration analysis is determined by Inductively Coupled Plasma Mass Spectrometer (ICP-MS). The accumulation of contaminants in the plants was identified using Scanning Electron Microscope (SEM). Phytoremediation, which is a natural method, reduced the contaminants up to 90% by translocating metals in roots, leaves and shoots of the selected plants. Water hyacinth shows a ratio of removal percentage where  $\text{Cr}=\text{Zn}=\text{Fe}>\text{Cd}>\text{Mn}>\text{Cu}>\text{B}$ . Meanwhile, water lettuce shows removal percentage in the following order  $\text{Fe}=\text{Zn}>\text{Cr}=\text{Cd}>\text{Mn}>\text{B}>\text{Cu}$ . The ratio result of water spinach shows the removal where  $\text{Zn}>\text{Cd}>\text{Fe}>\text{Mn}>\text{Cr}>\text{B}>\text{Cu}$ . Based on the application of three types of plants, water hyacinth is the best phytoremediator as it can absorb high concentrations of contaminants. The concentration of metals in roots is much higher than in leaves and stems. This study successfully proved that these three plants are a good phytoremediator with high potential to remove contaminants in ceramic wastewater and will provide a useful guideline for on-site treatment.

## ABSTRAK

Sistem fitoremediasi menggunakan tumbuhan akuatik yang semakin banyak digunakan kerana keupayaannya dalam penyingkiran logam. Kajian ini bertujuan untuk menilai potensi tumbuhan akuatik terpilih melalui sistem fitoremediasi untuk mengurangkan elemen terpilih dalam air sisa industri seramik iaitu boron, kuprum, kadmium, kromium, ferum, mangan, dan zink. Sistem ini telah diuji dengan pelbagai jenis tumbuhan iaitu keladi bunting, salad air, dan kangkung. Berdasarkan tumbuhan yang berbeza, kaedah kawalan dan kaedah aliran bebas dengan tangki hidroponik telah digunakan untuk membandingkan keberkesanan kaedah kitaran oleh sistem fitoremediasi. Sistem fitoremediasi yang menggunakan isipadu air sisa sebanyak  $0.024\text{m}^3$ , dengan 3 takungan pelantar-fito yang digunakan untuk merawat bahan cemar air sisa seramik menggunakan tumbuh-tumbuhan yang berbeza dalam tempoh 11 hari rawatan. Analisis kepekatan logam ditentukan menggunakan Spektrofotometer Plasma Berganding Secara Aruhan-Spektrofotometer Berjisim (ICP-MS). Pengumpulan bahan cemar dalam tumbuh-tumbuhan telah dikenal pasti menggunakan Mikroskop Imbasan Elektron (SEM). Fitoremediasi adalah kaedah semula jadi yang mengurangkan bahan cemar sehingga 90% melalui pemindahan logam ke akar, daun dan batang tumbuh-tumbuhan yang terpilih. Keladi bunting berjaya membuat penyingkiran mengikut nisbah  $\text{Cr}=\text{Zn}=\text{Fe}>\text{Cd}>\text{Mn}>\text{Cu}>\text{B}$ . Salad air menunjukkan peratus pengurangan mengikut nisbah  $\text{Fe}=\text{Zn}>\text{Cr}=\text{Cd}>\text{Mn}>\text{B}>\text{Cu}$ . Keputusan nisbah rawatan menggunakan kangkung adalah  $\text{Zn}>\text{Cd}>\text{Fe}>\text{Mn}>\text{Cr}>\text{B}>\text{Cu}$ . Berdasarkan penggunaan 3 jenis tumbuhan, keladi bunting adalah fitoremediasi terbaik kerana ia boleh menyerap bahan cemar pada kepekatan yang tinggi. Kepekatan logam dalam akar adalah lebih tinggi daripada daun dan batang. Kajian ini berjaya membuktikan ketiga-tiga tumbuh-tumbuhan adalah agen fitoremediasi yang baik yang berpotensi tinggi untuk merawat logam dalam air sisa seramik dan menyediakan satu garis panduan yang berguna untuk rawatan di kawasan tapak.

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and (B) after rhizofiltration

**LIST OF ABBREVIATIONS**

APHA	-	American Public health Association
B	-	Boron
BOD	-	Biochemical Oxygen Demand
Cd	-	Cadmium
CoF	-	Control test of free flow system
CoC	-	Control test of circulation flow system
COD	-	Chemical Oxygen Demand
Cr	-	Chromium
Cu	-	Copper
DO	-	Dissolved Oxygen
DOE	-	Department of Environment
EDTA	-	Ethylenediaminetetraacetic acid
EHS	-	Environmental Health and Safety
Fe	-	Iron
FWS	-	Free Water Surface
HF	-	Horizontal Flow
HNO <sub>3</sub>	-	Acid Nitric
HRT	-	Hydraulic Retention Time
ICP-MS	-	Inductively Coupled Plasma-Mass Spectrophotometer
IFC	-	International Finance Corporation
Mn	-	Manganese
MSIG	-	Malaysian Sewerage Industry Guideline
NIAST	-	National Institute of Agricultural Science and Technology
Pb	-	Plumbum

SEM	-	Scanning Electron microscope
SSF	-	Subsurface Flow
TOC	-	Total Organic Carbon
TSS	-	Total Suspended Solid
UN	-	United Nation
UTHM		Universiti Tun Hussein Onn Malaysia
UTM	-	Universiti Teknologi Malaysia
VF	-	Vertical Flow
WhF		Water hyacinth by free flow treatment
WhC		Water hyacinth by circulation flow treatment
WHO	-	World Health Organization
WlF		Water lettuceby free flow treatment
WlC		Water lettuce by circulation flow treatment
WsF		Water spinach by free flow treatment
WsC		Water spinach by circulation flow treatment
Zn	-	Zinc

**LIST OF SYMBOLS**

cm	-	Centimeter
g	-	Gram
h	-	Hour
L	-	Liter
m	-	Meter
metal <sub>L</sub>	-	Metal in leave
metal <sub>R</sub>	-	Metal in root
NH <sup>3</sup> -N	-	Ammonia nitrogen
ppb	-	Part per billion
TP	-	Total Phosphorus
μ	-	micro



## CHAPTER 1

### INTRODUCTION

#### 1.1 Background of Study

Ceramic industry is requiring the consumption of sand, clay, and bricks for its production. It is an important industry due to its vast application ability. One of its applications is that it can be used to filter contaminants from waste as materials in wastewater treatment (Garcia *et al.*, 2011). The production of ceramic consist of on several activities such as milling, charging, spraying, pressing, glazing, and sorting (Jing *et al.*, 2010). These activities will produce wastewater through the preparation and casting process with different process of glazing, decorating, polishing, and wet grinding. This wastewater contained suspended solids such as clays and insoluble silicates, suspended and dissolved heavy metals such as lead and zinc, sulfates, boron, and traces of organic matter.

Phytoremediation, using plants to remove contaminants, is a method on developing in situ strategies for remediation of environmental contaminants. Phytoremediation provides low cost, low technology treatment process which apply selected plants and microorganisms work together to metabolize, absorb, and accumulate, harmless multiple environmental contaminants (Prasad, 2004). A specific type of phytoremediation, namely rhizoremediation that involves rhizosphere microbes, can occur naturally. Root are played important role in this stage of phytoremediation (Lee and Yang, 2010) . The application of phytoremediation system are successfully executed when there are a study in Nigeria were using water lettuce to accumulates metals more than 300 times the concentration accumulated by conventional plants (Abubakar *et al.*, 2014). While in Slovakia, the removal of zinc and cadmium were studied to look into the effectiveness applied from contaminated wastewater (Zuzana *et al.*, 2014).

In Malaysia, the ceramic wastewater is recycled and reuse as a part of cleaning and mixing process at most of the factory. The wastewater going to selected points of the process with a specified route to ensure the part of body will not use that particular water. Even it was a recycle part, the standard of effluent discharge are specified in order to maintain the sustainability and the quality of product and as a mitigation way for body contact (Bovea *et al.*, 2010). The concentration of heavy metal is actually low depends on the level of the processes and treatments itself. Effluents which come up as a result of compounds processing and equipment cleaning usually contain the same raw materials and support materials and this makes compounds are water insoluble.

This research aims to investigate the ability of selective plants to improve the performances of concentration especially metals from solution in ceramic wastewater. The elemental of each plant is always collaborating with the concentration of metals. In order to make it worthy and support the growth, rhizofiltration is a best way to remove the metals from wastewater using roots of plants. The selection of plants must be

synchronizing to the nutrient needs which can be given from wastewater itself. Based on the characterization, the absorption potential, and the growth, three of plants, namely water hyacinth, water lettuce and water spinach was the best to remediate ceramic wastewater.

## **1.2 Problem Statement**

Research and publication of remediation process on organic chemicals and metals had been developed as a remedial strategy to improve environmental solution. There are many published result regarding to phytoremediation. It is well-suited for use at large site area where other methods of remediation are not cost-effective at sites over long period (Schnoor, 1997). However, the fundamental solution in field regarding to uptake in ceramic industrial wastewater is limited. Ceramic wastewater that accumulated from the production process, which is contains small quantities of numerous organic materials as well as some heavy metal, are easily trap inside the body skin to become harmful. The manufacturing of ceramic also produces the insoluble particulate matter, organic, and inorganic materials in wastewater. This may produce large quantity of contaminants in wastewater. Currently, the treatment system applied mechanical and chemical application may damage and the concentration of organic and inorganic matter will be increased. In term of that, phytoremediation system with phytoremediator plant had been studied to improve the current system. This system proposed the simple and natural method without any harmful mechanisms and contributes to an uptake of ceramic wastewater solution.

### **1.3 Objectives of Study**

This study aimed to investigate the potential of aquatic plants namely water hyacinth, water lettuce, and water spinach to be used for phytoremediation in wastewater from ceramic industry. Specific objectives of this study are as follows:

- (i). To determine the effectiveness of free flow system in reducing pollution potential of ceramic wastewater by determination of organic and heavy metals uptake and absorption rate
- (ii). To determine the effectiveness of circulation system in reducing pollution potential of ceramic wastewater by determination of organic and heavy metals uptake and absorption rate.
- (iii). To evaluate the ability of plants to absorb contaminants by determination of physical and morphological adaptive capacity.

### **1.4 Scope of Study**

This research conducted a cleanup system by lab-scale phytoremediation system with installation of phyto-rig (0.28 x 0.19 x 0.46) m<sup>3</sup>. It was applied with 3 types of phytoremediator plants namely water hyacinth, water lettuce, and water spinach. This study involved field and laboratory activities. The determinations of physical characteristics were determined at site area in ceramic industrial area in Kluang for the real result. All laboratory works involved determination of chemical properties had been conducted in Environmental Laboratory in Faculty of Civil Engineering and Chemical

Engineering, UTM Johor. The samples collected had been treated in 11 days. The water quality parameter tested on the organic and inorganic constituents includes of Total Organic Carbon (TOC), Chemical Oxidation Demand (COD), Dissolved Oxygen (DO), pH, Total Suspended Solid (TSS), and heavy metals analysis. The heavy metals includes of Boron (B), Manganese (Mn), Copper (Cu), Cadmium (Cd), Chromium (Cr), Iron (Fe), and Zinc (Zn). The selection parameters of physical, chemical, and heavy metals constituents was basically chosen based on the highest reading of influent produced from the site area in factory. The result had been compared to previous study which related to ceramic industry wastewater. The selection of parameters also based on the uptake organic contaminants from ceramic wastewater consistent with nutrients needs by plants as stated more in chapter 4.

## **1.5 Significance of Study**

This research is necessary in solving the problem in remediation and treatment of the industrial wastewater. By using natural system such as phytoremediation in wastewater treatment will enhance sustainable development with environmental friendly condition. This will eventually help to provide balance to the ecosystem through its dynamic vegetation process which is using phytoremediation green technology supported by plant as the main treatment component. Furthermore, treatment of the industrial wastewater by using rhizofiltration technology seems to be promising and can offer reliable and feasible alternative method.

Specific findings of this study as follow:

- (i). This study provides comparison of phytoremediation by free flow system and circulation system for ceramic industry wastewater treatment.
- (ii). New insight in identify the best plants selection to reduce pollution in ceramic wastewater.
- (iii). New theoretical and practical knowledge in determination of organic and heavy metals uptake and adsorption rate.
- (iv). Exploration of physical and morphological adaptive capacity by selected plants

## **1.6 Organization of Thesis**

This study explained on the effectiveness of selected plants to react with rhizofiltration system in removal physical, chemical, and metal constituents. The scope was identified as major exploration of this study by looking into the physical and morphological of sample specified which is aquatic plants and ceramic wastewater.

Chapter 2 introduced the definition and concept of this study based on review of previous researches. The introduction was started by looking into the characteristics of ceramic wastewater and the relationship to the contaminants absorption by selected plants. The reviews on the plants were also focused on the aquatic habitat with the growth development and nutrients absorption.

Chapter 3 provided a brief description on the work flow that involved in this study. This was an important chapter in this study because of the relations on the data profile and as a root to the conclusion remarks. All the measurement and operation was based on the objectives proposed in this study. This chapter also introduced the mechanisms related with data provided in the next chapter explanation.

Chapter 4 was all about the study on the performances of specified plants namely water hyacinth, water lettuce and water spinach in removal the contaminants in ceramic wastewater. The analysis determination on the significance of the variables also been explained in this chapter. The end of this chapter concluded the relationship for both of plants and ceramic wastewater in adsorption capability.

Chapter 5 highlighted a statement which explained the best plants and the most contaminants absorption rate that successfully performed. The rhizofiltration method between stagnant and circulation was proposed and recommended to further study. All the limitation on the operational process and discussion from the previous study was summarized in this chapter.

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