MASS TRANSFER KINETICS OF PHOSPHATE REMOVAL FROM SEWAGE TREATMENT PLANT EFFLUENT BY WASTE MUSSEL SHELL

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DEDICATION

This thesis is dedicated, in its entirety, to my parents, brother, and sister, who have supported me throughout the journey. I dedicate this humble monograph to lecturers at the Faculty of Chemical Engineering, Universiti Teknologi Mara, Shah Alam, Selangor, Malaysia.

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ABSTRACT

Excessive amount of phosphate (PO_4^{3-}) released from sewage treatment plant effluent (STPE) may trigger eutrophication of water causing degradation of aquatic ecosystem and human health. Even though the presence of PO_4^{3-} ions in aqueous solution can be removed using adsorption techniques, detailed description of adsorption kinetics is still not fully understood. In this study, the isotherm and kinetic adsorption of PO_4^{3-} from aqueous solution onto porous material were conducted in batch experiments. A typical design of the hybrid plug flow column reactor (HPFCR) was used to remove PO_4^{3-} from STPE. The kinetic models (i.e., pseudo-first-order (PFO) and pseudo-second-order (PSO)) and the isotherm models (i.e., Freundlich and Langmuir) were used to determine the adsorption kinetics and isotherms of PO_4^{3-} from STPE onto waste mussel shell (WMS) and iron-coated waste mussel shell (ICWMS) adsorbents. The empirical models of bed depth service time (BDST), Thomas, and modified mass transfer factor (MMTF) were used to describe the adsorption kinetic processes of PO_4^{3-} of WMS and ICWMS applied in the HPFCR. The experimental data for the adsorption of PO_4^{3-} onto both WMS and ICWMS adsorbents fitted very well with the PSO kinetic model and Freundlich isotherm model, respectively. The dynamic adsorption capacity of WMS and ICWMS described by the BDST model has shown to increase with increase in the plug flow column (PFC) depth. The hydrodynamic behavior of PO_4^{3-} global mass transfer can be described using the Thomas models for predicting the PFC performance. Employing the MMTF models enabled differentiation between the behavior of film mass transfer and porous diffusion. The resistance of PO_4^{3-} mass transfer is dependent on porous diffusion and this contributes to the development of advanced WMS and ICWMS adsorbents in enhancing the performance of the HPFCR system in the future.

ABSTRAK

Jumlah fosfat (PO_4^{3-}) berlebihan yang dilepaskan daripada efluen loji rawatan kumbahan (STPE) boleh menyebabkan eutrofikasi air lalu menyebabkan kemerosotan ekosistem air dan kesihatan manusia. Walaupun kehadiran ion PO_4^{3-} dalam larutan akueus boleh disingkirkan menggunakan teknik penjerapan, keterangan terperinci kinetik penjerapan masih tidak difahami sepenuhnya. Dalam kajian ini, isoterma dan kinetik penjerapan PO_4^{3-} daripada larutan akueus ke atas bahan berliang telah dijalankan dalam eksperimen kelompok. Reka bentuk tipikal reaktor turus aliran palam hibrid (HPFCR) digunakan untuk menyingkirkan PO_4^{3-} daripada STPE. Model-model kinetik (iaitu, tertib pertama pseudo (PFO) dan tertib kedua pseudo (PSO)) dan model-model isoterma (iaitu, Freundlich and Langmuir) digunakan untuk menentukan kinetik dan isoterma penjerapan PO_4^{3-} daripada STPE ke atas bahan penyerap daripada sisa cangkerang kupang (WMS) dan sisa cangkerang kupang bersalut ferum (ICWMS). Model-model empirik seperti masa penggunaan kedalaman lapisan (BDST), Thomas, dan faktor pindahan jisim terubahsuai (MMTF) digunakan untuk menggambarkan proses kinetik penjerapan dan penyingkiran PO_4^{3-} daripada STPE ke atas bahan penjerap daripada WMS dan ICWMS yang digunakan dalam HPFCR. Data eksperimen bagi penjerapan PO_4^{3-} ke atas bahan penjerap daripada WMS dan ICWMS masing-masing sangat sepadan dengan model kinetik PSO dan model isoterma Freundlich. Keupayaan penjerapan dinamik WMS dan ICWMS digambarkan oleh model BDST yang didapati meningkat dengan peningkatan turus aliran palam (PFC). Kelakuan hidrodinamik pindahan jisim global PO_4^{3-} boleh diterangkan dengan menggunakan model Thomas untuk meramalkan prestasi PFC. Penggunaan model MMTF dapat menerangkan perbezaan sebenar antara kelakuan pindahan jisim filem dan peresapan berliang. Kerintangan pindahan jisim PO_4^{3-} adalah bersandarkan pada peresapan berliang dan ini menyumbang kepada pembangunan bahan penjerapan termaju WMS dan ICWMS dalam meningkatkan prestasi sistem HPFCR pada masa akan datang.

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LIST OF ABBREVIATIONS

APHA	-	American Public Health Association
BDST	-	Bed Depth Service Time
BET	-	Brunauer, Emmett, and Teller
BOD	-	Biochemical Oxygen Demand
COD	-	Chemical Oxygen Demand
DO	-	Dissolve Oxygen
EDXRF	-	Energy Dispersive X-Ray Fluorescence
EU	-	European Union
FMT	-	Film Mass Transfer
FTIR	-	Fourier Transform Infrared
GMT	-	Global Mass Transfer
HPFCR	-	Hybrid Plug Flow Column Reactor
ICWMS	-	Iron-coated Waste Mussel Shell
MMTF	-	Modified Mass Transfer Factor
MTF	-	Mass Transfer Factor
P	-	Phosphorus
PD	-	Porous Diffusion
PFC	-	Plug Flow Column
PFO	-	Pseudo-first Order
PSO	-	Pseudo-second Order
SEM	-	Scanning Electron Microscope
SF	-	Sand Filtration
SS	-	Suspended Solid
STP	-	Sewage Treatment Plant
STPE	-	Sewage Treatment Plant Effluent
USEPA	-	United States Environmental Protection Agency
WCS	-	Waste Cockle Shell
WES	-	Waste Egg Shell
WMS	-	Waste Mussel Shell
WOS	-	Waste Oyster Shell

XRD	-	X-Ray Diffraction

LIST OF SYMBOLS

$[k_\mathrm{L}a]_\mathrm{d}$	-	The internal mass transfer (porous diffusion) factor (h^{-1})
$[k_\mathrm{L}a]_\mathrm{f}$	-	The external (film) mass transfer factor (h^{-1})
$[k_\mathrm{L}a]_\mathrm{g}$	-	The global mass transfer factor (h^{-1})
C_e	-	The concentration of the adsorbate in the equilibrium solution (mg L^{-1})
C_f	-	The concentration of PO_4^{3-} solute in the aqueous solution (mg L^{-1})
C_i	-	The initial concentration of PO_4^{3-} solute in the aqueous solution (mg L^{-1})
C_o	-	The concentration of PO_4^{3-} solute entered the PFC (mg L^{-1})
C_s	-	The concentration of PO_4^{3-} solute passed through the PFC (mg L^{-1})
F_e	-	The error function (dimensionless)
K_a	-	The adsorption rate constant (L h^{-1} mg^{-1})
K_F	-	The Freundlich constant (mg g^{-1})
K_L	-	The adsorption energy coefficient (L mg^{-1})
K_T	-	The kinetic coefficient or the Thomas rate constant (L h^{-1} mg^{-1})
N_o	-	The dynamic adsorption capacity per unit volume of the plug flow column (mg L^{-1})
R^2	-	The correlation coefficient
k_1	-	Pseudo-first order constant (min^{-1})
k_2	-	Pseudo-second order constant (g mg^{-1} min^{-1})
$q_\mathrm{e(exp)}$	-	The equilibrium amount of adsorbate adsorbed by per unit mass of adsorbent obtained from the measurement during the experiments (mg g^{-1})
$q_\mathrm{e(theo)}$	-	The equilibrium amount of adsorbate adsorbed by per unit mass of adsorbent calculated by the model (mg g^{-1})
q_e	-	The equilibrium amount of adsorbate adsorbed by per unit mass of adsorbent (mg g^{-1})
q_max	-	The maximum adsorption capacity of the adsorbent (mg g^{-1})
q_o	-	The equilibrium PO_4^{3-} solute uptake per gram of the adsorbent (mg g^{-1})

$q_{t(exp)}$	-	The experimental q value obtained from the measurement during the experiments (mg g^{-1})
$q_{t(theo)}$	-	The theoretical q value calculated by the model (mg g^{-1})
q_t	-	The amount of adsorbate adsorbed at adsorption time t (mg g^{-1})
h	-	The depth of PFC (cm)
B	-	The potential mass transfer index relating to driving force of the PO_4^{3-} mass transfer from bulk water to acceptor sites (mg g^{-1})
E	-	The removal efficiency (%)
Q	-	The volumetric flow rate (L h^{-1})
V	-	The solution volume (L)
a	-	The retention coefficient relying the speed of solute passed through the adsorbent (h cm^{-1})
b	-	The contact time constant relying to the availability of porous surface and acceptor sites (h)
c	-	Constant (h^{-1})
d	-	Constant (dimensionless)
m	-	The amount of adsorbent (g)
n	-	The heterogeneity factor (dimensionless)
q	-	The cumulative quantity of PO_4^{3-} deposited on the adsorbent (mg g^{-1})
t	-	The service time (h)
t	-	The interval time (min)
v	-	The linear flow rate (cm h^{-1})
β	-	The absorbate-adsorbent affinity parameter (g h mg^{-1})

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CHAPTER 1

INTRODUCTION

1.1 Background

Since the 20th century, the presence of phosphorus (P) in sewage treatment plant effluent (STPE) has received attention due to the realization of its negative impacts on receiving waters. In sewage treatment processing, P is a vital nutrient for bacteria needed to degrade and biologically stabilize the organic wastes (Nielsen *et al.*, 2019). P appears exclusively as condensed phosphates (polyphosphates), and organically bound phosphate. Condensed phosphates are used extensively as builders in detergents, and organic phosphates are constituents of body waste and food residue (Gomes *et al.*, 2016; Naden *et al.*, 2016). The discharge of excessive amount of phosphate (PO_4^{3-}) from STPE is of concern as it is one of the key nutrients that have the potential to contribute to eutrophication in surface water (Gu *et al.*, 2021).

Oversupply of PO_4^{3-} release from sewage into an aquatic ecosystem leading to the eutrophication, excessive growth of algae, and oxygen depletion may pose serious threat to the environment, aquatic life, and human health (Fink *et al.,* 2018). Briefly, the presence of phytoplankton in water surface may utilize the PO_4^{3-} compounds causing an excessive algae growth and giving rise to the appearance of cyanobacteria (Kakade *et al.,* 2021). Sufficient light and nutrients induced cyanobacteria to release the toxic secondary metabolites/cyanotoxins which are responsible for aquatic life illnesses and death, including humans via direct contact, inhalation, or ingestion (Dwivedi, 2018). The commonly found cyanotoxins are Microcystins, Cylindrospermopsin, Anatoxins, Saxitoxins, and Lyngbytoxins (Wang and Wang, 2018). These toxins are capable of accumulating in water, aquatic plants and the food web sequentially. Zooplanktons can accumulate cyanotoxins through phytoplankton grazing, which further bioaccumulates to higher trophic organisms such as fish and humans (Wang and Wang, 2018). For example, the fish population in Lake Victoria was decreased due to hypoxia and high cyanotoxin accumulation in them (Olokotum *et al.*, 2020). The accumulation of toxins in fish affects their growth, development, reproduction and causes severe histopathological damages, reducing their survival chances (Drobac *et al.*, 2016). Further, these algal mats on water surface decrease light penetration and cause hypoxia, which results in flora and fauna death. The severe anoxia at the bottom of deep lakes is because of the high respiratory activity of organic matter degrading bacteria produced by cyanobacteria. The toxins released from these blooms cause several hazards in humans such as headache, sore throat, and diarrhea (Szlag *et al.*, 2015). As a result, declined dissolve oxygen (DO) and the released toxins cause disturbance in the aquatic food web and harm humans by contact or consumption.

Eutrophication threatens human health and aquatic life. For example, eutrophication in Lake Taihu, China has endangered the drinking water supply of millions of people in Shanghai (Fink *et al.*, 2018). The discharge of a high amount of PO_4^{3-} from domestic wastewater effluent into Lake Izabal, Guatemala stimulated algal blooms and killed fish (Obrist-Farner *et al.*, 2019). The city of Toledo, Ohio, United States had to shut down the drinking water supply of half a million people due to the presence of harmful algal blooms near the intake pipes of the water treatment plant on Lake Erie, in North America (Isaac, 2020). The increased amount of PO_4^{3-} inputs from residential areas triggered eutrophication in Slim River Lake, Perak, and consequently degraded the lake water quality (Aeriyanie *et al.*, 2021). The most severe fish kill event occurred in Tebrau River, Johor, due to the presence of harmful algal blooms (Yñiguez *et al.*, 2021).

In order to alleviate eutrophication, the acceptable limits of PO_4^{3-} discharged into surface waters have been regulated by the Malaysia Environmental Quality (Sewage) Regulations 2009, the United States Environmental Protection Agency (USEPA), the European Union (EU) Wastewater Directive, the Swiss Federal Water Protection Law (SFWPL) and the China's Water Quality Regulations (CWQR). The acceptable limit for PO_4^{3-} concentration according to the Malaysian effluent standards is 5 mg L^{-1}. The USEPA permits the PO_4^{3-} concentration of less than 0.8 mg L^{-1} in the effluent and the effluent limit of PO_4^{3-} regulated by the EU Wastewater Directive is 2 mg L^{-1} for every agglomeration with population equivalent of 10,000-100,000 (Fulazzaky *et al.,* 2014; Kumar *et al.,* 2019). Swiss and China effluent standards allow the PO_4^{3-}concentration is set at the level of 0.8 and 0.5 mg L^{-1}, respectively to prevent eutrophication (Preisner *et al.,* 2020). It seems that the Malaysian effluent standards can tolerate the amount of PO_4^{3-} in effluent up to 5 mg L^{-1} higher than EU Wastewater Directive, USEPA, Swiss and China effluent standards (Osman *et al.,* 2020; Sabeen *et al.,* 2018). This argues that the higher concentration of PO_4^{3-} permitted by the Malaysian effluent standards make the operator of sewage treatment plant (STP) less sensitive to the eutrophication of aquatic environment. The limit of environmentally realistic PO_4^{3-} concentration regulated by the Malaysian Environmental Quality Act is required to avoid the eutrophication of water body in Malaysia.

The various methods of the physical, chemical, and biological processes have been proposed for the removal of PO_4^{3-} from an effluent. Using the biological method can achieve a high efficiency of PO_4^{3-} removal but it has several types of variables to be controlled to ensure optimum performance of the treatment process (Bali and Gueddari, 2019; Nagoya *et al.,* 2019). The application of chemical method requires the cost of chemical usage in a sewage treatment system (Lalley *et al.,* 2016). The combination of biological and chemical processes can allow to protect the eutrophication of receiving water from PO_4^{3-} pollution while saving on PO_4^{3-}-sludge disposal costs (Tomei *et al.,* 2020) but it is still complicated and expensive. Among all these purification methods, adsorption is a promising and attractive method of PO_4^{3-} removal from effluent due to the adsorption process may gain popularity for the reasons of simplicity of operation and cost effectiveness while the possibility of producing secondary pollutant is minimal (De Gisi *et al.,* 2016; Zhang *et al.,* 2019). Different types of materials such as activated carbon, laterites, metal oxides, and metal sulfate can be used as adsorbents for the removal of inorganic and organic pollutants from waters (Fulazzaky *et al.,* 2014; Huang *et al.,* 2017). The promising potential of waste mussel shell (WMS) can be proposed for the removal of PO_4^{3-} from wastewater since the interaction of nanocalcium hydroxide with PO_4^{3-} ions allowing a precipitation in the form of value-added hydroxyapatite on the WMS surface may increase the adsorption capacity of PO_4^{3-} compared to activated carbon, laterites, metal oxides and metal sulfate (Khan *et al.,* 2020). In addition, the calcium oxide content of the WMS adsorbent has exhibited promising performance for the adsorption of PO_4^{3-} ions from water (Barbachi *et al.,* 2019; Romar-Gasalla *et al.,* 2016).

In Malaysia, about 1,800 metric tonnes per year of WMS are produced from food industry and dumped into landfills; approximately 500 metric tonnes of WMS in Malaysia are used for fertilizers and handicrafts while the remainder (1,300 metric tonnes) constitutes a serious solid waste problem (Sainudin *et al*., 2019). Scientific optimism and societal concerns of using the WMS as an alternative low-cost adsorbent for the removal of PO_4^{3-} needs to be scrutinized due to it is abundantly produced in Malaysia with no economical value. The modification of calcined mussel shell powder decorated with surfactants has been proposed to improve the oil-cleaning and antistatic properties that offer a green solution to reduce the environmental pressures (Wei *et al.,* 2018). The addition of powdered WMS into an activated sludge process can increase the removal efficiency of heavy metals enabling the formation of heavier activated sludge flocs to enhance the settling properties of activated sludge (Papadimitriou *et al.,* 2017).The practical application of the WMS adsorbent has been investigated by using the calcined mussel shells for the removal of basic fuchsin dye (El Haddad, 2016) and by using the mussel-inspired Fe_3O_4@Polydopamine(PDA)-MoS_2 core-shell nanosphere for the removal of Pb^{2+} from aqueous solutions (Wang *et al.,* 2019). The adsorption isotherm of PO_4^{3-} onto WMS has been investigated following the Langmuir model for a short contact time and the Freundlich model for the longer batch and column contact times (Paradelo *et al.,* 2016).

Many recent studies have been focused on the interpretation of adsorption kinetic and isotherm models to understand the mechanism of PO_4^{3-} adsorption onto the various types of adsorbents, but these models are limited to the batch experiments (Fulazzaky *et al.,* 2019; Khan *et al.,* 2020; Paradelo *et al.,* 2016). Two types of empirical adsorption models of such as the bed depth service time (BDST) and Thomas have been broadly used for predicting the breakthrough curves of adsorption process in continuous mode of operation (Charola *et al.,* 2018; Rout *et al.,* 2017; Wang *et al.,* 2016). The BDST models can be used to determine the effect of various operating variables on the performance of adsorbent to remove the PO_4^{3-} ions from aqueous solution. However, the dispersion of PO_4^{3-} caused by mass transfer resistance is negligible during the adsorption process (Pember *et al.*, 2016). The Thomas models assume the kinetic process of PO_4^{3-} adsorption onto adsorbent from aqueous solution needs to follow the adsorption-desorption kinetics of the Langmuir model with no any axial dispersion and neglect any mass transfer resistances (Ahmed and Hameed, 2018). Even though several empirical adsorption models have been proposed to improve the adsorption performance of removing a solute from aqueous solution, the application and validity of the empirical adsorption models to describe the mass transfer factors of PO_4^{3-} adsorption onto the WMS and iron-coated waste mussel shell (ICWMS) from aqueous solution is still not fully understood. Therefore, a mechanistic understanding of the global, external, and internal mass transfer of the adsorption kinetics and mechanisms revealed by the modified mass transfer factor (MMTF) models needs to be understood.

1.2 Problem Statement

In 2019, there were 7,115 public STP operated in Malaysia with a total capacity of 27.06 million population equivalent (SPAN, 2019). Of these, old treatment systems such as oxidation ponds are still being widely used until today. These old plants are not effectively designed to remove nutrients, particularly PO_4^{3-}. The concentration of PO_4^{3-} released into water bodies contravenes with the Environmental Quality (Sewage) Regulations 2009 that would only allow 5 mg L^{-1}. Moreover, the standards set by EU Wastewater Directive, USEPA, Swiss, and Russia are stricter than Malaysia. An excessive amount of PO_4^{3-} in the aquatic environment can cause an increased growth of algae and other aquatic plants, which can result in a decreased level of DO in water leading to depopulate the aquatic life and deteriorate the quality of water. Considering a huge loading of PO_4^{3-} into water bodies and the nature of PO_4^{3-} as a rate limiting nutrient in the freshwater environment, action must be taken to equip the STP with PO_4^{3-} removal technology. Thus, this study proposed the hybrid plug flow column reactor (HPFCR) system of combining sand filtration (SF) and plug flow column (PFC) as the laboratory-scale device to remove PO_4^{3-} from STPE collected from the Taman Sri Pulai of Johor Bahru in Malaysia. An understanding of the removal efficiency and the adsorption behavior in the HPFCR system is important for the control of full-scale operating systems, and for more widespread use in real wastewater treatment. However, factors that affect the behavior of mass transfer between adsorbate/solute and adsorbent need to be considered, taking into account that in order to understand the applicability of HPFCR system in the removal of PO_4^{3-} from STPE. In spite of reducing the concentration of PO_4^{3-} can be accomplished through an adsorption technique, the resistance of mass transfer of the adsorption in a hydrodynamic column needs to be verified. Therefore, the use of MMTF models is crucial for describing the removal of one or more solutes from waters to provide better understanding of the movement of solutes from the bulk water to active sites within the pores of a porous material. The use of the MMTF models could be useful to describe the real difference between the behaviors of film mass transfer (FMT) and porous diffusion (PD).

1.3 Objectives

This study embarks on the following objectives:

1. To verify the applicability of the adsorption kinetic models of pseudo-first-order (PFO) and pseudo-second-order (PSO) kinetic models and the adsorption isotherm models of Freundlich and Langmuir observed from the batch experiments,

2. To analyze the reliability of the BDST and Thomas models for predicting the kinetic behaviors of PO_4^{3-} adsorption onto the WMS and ICWMS adsorbents from STPE processed in a hydrodynamic column,

3. To verify the applicability of the MMTF models to predict the mechanisms and kinetics of mass transfer for the adsorption of PO_4^{3-} deposited onto the WMS and ICWMS adsorbents from STPE applied in a hydrodynamic column, and

4. To determine if the resistance of mass transfer is located at either FMT or PD during the adsorption process of PO_4^{3-} deposited onto the WMS and ICWMS adsorbents from STPE.

1.4 Scope of The Study

The scope of this study is as follows:

1. Characterization of STPE

   The characteristics of STPE were determined by monitoring several parameters of such as chemical oxygen demand (COD), suspended solid (SS), ammonium (NH_4^+), iron (Fe^{3+}), PO_4^{3-}, nitrate (NO_3^-), sulfate (SO_4^{2-}), chloride (Cl^-), ph, and DO. This study used the STPE collected from the Taman Sri Pulai of Johor Bahru in Malaysia (latitude 1°33'59.1"N and longitude 103°36'56.7"E).

2. Characterization of adsorbents

   Two types of adsorbents were used in this study i.e., WMS and ICWMS. The physical and chemical properties of adsorbents, such as chemical composition, types of mineral phases, surface morphology, surface functional group, and specific surface area were determined.

3. Batch experiment

   The batch experiments were conducted to determine the kinetics and isotherm of adsorption using a synthetic solution and STPE. The PFO and PSO kinetic models were used to evaluate adsorption kinetic. The Freundlich and Langmuir isotherm models were used to assess the adsorption isotherm.

4. HPFCR performance

   The laboratory-scale HPFCR system was performed to determine the removal efficiency of PO_4^{3-} from STPE. The concentration of PO_4^{3-} solute is the independent parameters while the flow rate, height, and mass of adsorbent in the PFC are dependent parameters. The concentrations of PO_4^{3-} were regularly monitored at inlet in the storage tank, after the SF and outlet of the HPFCR system using UV Spectrophotometer HACH DR 5000. The parameters of such as COD, SS, NH_4^+, SO_4^{2-}, Cl^-, pH, and DO were measured at inlet and outlet of HPFCR. The BDST and Thomas models were used to determine the design parameter and to predict the breakthrough curve behavior in HPFCR.

1.5 Significance of The Study

The significances of this study are as follows:

1. The removal of excessive amount of PO_4^{3-} from STPE is crucial to prevent the eutrophication of surface water and to meet the stringent effluent standards.

2. The analysis of experimental data obtained from a laboratory-scale device is important to gain better understanding of the further practical and operational information in designing the industrial scaling up purposes.

3. The findings of this study revealed an understanding of the mechanisms and kinetics of adsorption that are important for further improvement of the adsorption capacity, adsorbate–adsorbent affinity, and adsorbent surface properties for wide applications.

4. The determination of mass transfer resistance controlled by either FMT or PD for the adsorption of PO_4^{3-} onto WMS and ICWMS adsorbents can make an important contribution to the development of advanced WMS and ICWMS adsorbents for enhancing the HPFCR performance in the future.

1.6 Thesis Organization

This thesis is organized into six chapters. After briefly introducing the background in Chapter 1, this study reviews the literatures in Chapter 2 for concern with PO_4^{3-} in waters and its removal processes. The materials and methods in Chapter 3 include the batch experiments, configuration of HPFCR, HPFCR operating procedure and analytical methods. Discussion of the results can be found in Chapter 4 and Chapter 5. Chapter 4 describes the physical and chemical properties of the adsorbents and discusses the experimental result obtained from batch studies that include analysis adsorption kinetics and isotherm in synthetic solution and STPE. Chapter 5 presents the application of the empirical model i.e., BDST, Thomas, and MMTF models and discusses the adsorption kinetics of PO_4^{3-} deposited on the surface of WMS and ICWMS observed from a HPFCR. The last chapter presents the conclusions of this study and the recommendation for future works.

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APPENDIX

Appendix A Characterization of STPE

Because of the content of PO_4^{3-} (see Table 1) is higher than the limit value regulated by the law (see Table 2), the tertiary treatment of STPE is required before discharging into a river. This study used the STPE collected from the Taman Sri Pulai of Johor Bahru in Malaysia (latitude 1°33’59.1”N and longitude 103°36’56.7”E) as the effluent samples to feed the HPFCR treatment system. The characteristics of STPE quality can be classified as a moderate concentration based on the average value of the parameters of PO_4^{3-}, NH_4^{+}, SO_4^{2-}, Cl^{-}, COD, SS, pH, DO, and conductivity monitored during six months (see Table 1). The adsorption of PO_4^{3-} onto WMS and ICWMS adsorbents from STPE can be proposed to allow the design characterization of HPFCR system and to describe the kinetic behaviors of PO_4^{3-} removal

Table 1 Main characteristics of STPE

Parameter	**Unit**	**Concentrations**		
		Minimum	**Maximum**	**Average**
DO	mg L^{-1}	5.1	5.6	5.43
COD	mg L^{-1}	170	195	180
SS	mg L^{-1}	12	84	31
pH	-	6.6	7.4	7.1
NH_4^{+}	mg L^{-1}	7.4	11.8	10.2
PO_4^{3-}	mg L^{-1}	5.6	8.4	7
Cl^{-}	mg L^{-1}	10	37.5	18
SO_4^{2-}	mg L^{-1}	8	15	13
Conductivity	μS cm^{-1}	369	414	384

LIST OF PUBLICATIONS

Journal with Impact Factor

1. Fulazzaky, M. A., **Salim, N. A. A**., Khamidun, M. H., Puteh, M. H., Mohd Yusoff, A. R., Abdullah, N. H. Syafiuddin, A. and Ahmad Zaini, M. A. (2021). 'The mechanisms and kinetics of phosphate adsorption onto iron-coated waste mussel shell observed from hydrodynamic column', *International Journal of Environmental Science and Technology,* (Q3, IF: 2.540) **Status: Published.**

2. **Salim, N. A. A.,** Zaini, M. A. A., Fulazzaky, M. A., Puteh, M. H., Khamidun, M. H. and Abdullah, N. H. (2021). 'A two–stage batch system for phosphate removal from wastewater by iron–coated waste mussel shell to assess the optimum adsorbent dosage', *Journal of Water Chemistry and Technology*, (Q4, IF: 0.504) **Status: Accepted.**

Indexed Journal

1. **Salim, N. A. A**., Puteh, M. H., Yusoff, A. R. M., Abdullah, N. H., Fulazzaky, M. A., Rudie Arman, M. A. Z., Khamidun, M. H., Zaini, M. A. A., Syafiuddin, A., Ahmad, N., Lazim, Z. M., Nuid, M. and Zainuddin, N. A. (2020). 'Adsorption isotherms and kinetics of phosphate on waste mussel shell', *Malaysian Journal of Fundamental and Apllied Sciences*, 16(3), 393–399. **(Indexed by Scopus)**

2. **Salim, N. A. A**., Fulazzaky, M. A., Ahmad Zaini, M. A., Puteh, M. H., Khamidun, M. H., Mohd Yusoff, A. R., Abdullah, N. H., Ahmad, N., Lazim, Z. M. and Nuid, M. (2021). 'Phosphate removal from wastewater in batch system using waste mussel shell', *Biointerface Research in Applied Chemistry*, 11(4), 11473–11486. **(Indexed by Scopus)**

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Non-Indexed Conference Proceedings

1. **Salim, N. A. A.**, Puteh, M. H., Abdullah, N. H., Khamidun, M. H., Fulazzaky, M. A., Yusoff, A. R. M. and Zaini, M. A. A. (2019) Kinetics Study of Phosphate Adsorption onto Waste Mussel Shell. *International Conference on Environmental Sustainability and Resource Security (IC-ENSURES)*. 5-6 November. Malaysia: Centre for Environmental Sustainability and Water Security (IPASA), 201-205.

Book Chapter

1. **Salim, N. A. A**., Abdullah, N. H., Ahmad, N. and Puteh, M. H. Phosphorus and Eutrophication in Water. In: M. H. Puteh and N. H. Abdullah. eds. *Issues and Technology in Water Contaminants*. Malaysia: UTM Press. 121–137; 2020.

2. Abdullah, N. H. and **Salim, N. A. A.** Introduction to Water Pollutants: Adsorptions and Solutions. In: N. H. Abdullah and N. A. A. Salim. eds. *Water Pollutants: Adsorptions and Solutions*. Malaysia: UTM Press. 2021. 1-6: 2021. **(Editor)**

3. **Salim, N. A. A**., Abdullah, N. H., Zaini, M. A. A. and Khamidun, M. H. Tackling Phosphorus Eutrophication with Mussel Shell. In: N. H. Abdullah and **N. A. A. Salim**. eds. *Water Pollutants: Adsorptions and Solutions*. Malaysia: UTM Press. 2021. 121-139: 2021. **(Editor)**