BANANA BLOSSOMS PEELS ADSORBENT FOR THE REMOVAL OF MANGANESE IN WATER

NURUL NADIA BINTI RUDI

A thesis submitted in fulfillment of the requirement for the award of the Degree of Master of Engineering Technology

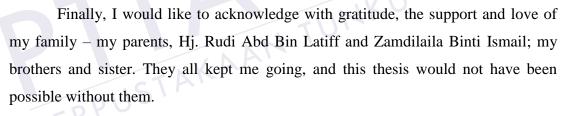
Faculty of Engineering Technology Universiti Tun Hussein Onn Malaysia

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ABSTRACT

Manganese is one of the persistent heavy metals detected in surface water lately that has become an environmental concern because manganese poses negative impacts to the ecosystem and water supply. This study investigates the effectiveness of banana blossom peels (BBP) as an adsorbent for the removal of manganese in water. The BBP adsorbent was activated via chemical treatment method. The Water Quality Index for Sungai Panchor revealed that the river is classified in Class II as clean river with the highest manganese concentration of 0.575 mg/L detected by using ICP-OES. FESEM-EDX analysis showed the morphology of BBP adsorbent was crimped with deeper dents, rough internals surface, and dense in nature, providing maximum surface area for the adsorption process. FTIR analysis confirmed the presence of carboxyl and hydroxyl groups in BBP adsorbent that contribute to the adsorption process. XRD analysis showed that the structure of the BBP adsorbent is amorphous. BET surface area of BBP adsorbent was 2.12 m^2/g with the total pore volume of 0.0139 cm³/g and average pore diameter of 64.35 nm. Batch adsorption study evaluated the parameters affecting the adsorption process, which include pH, adsorbent dosage, initial manganese concentration, and contact time. The optimum condition was attained at pH 7, adsorbent dosage of 0.5 g, 10 mg/L initial manganese concentration, and 150 minutes contact time. The adsorption isotherm study demonstrated that the Langmuir isotherm model best fit BBP adsorption toward manganese with $R^2 > 0.9963$. The adsorption of manganese followed the pseudo-second-order kinetics representing the chemisorption process. The maximum desorption rate of 92% was achieved in the first cycle, with a recovery rate of 94.18% within 30 minutes using 0.1M HCl. The BBP adsorbent able to remove 55% manganese in Sungai Panchor water sample with an initial manganese concentration of 0.095 mg/L. This study suggests that BBP adsorbent has the potential to be used as an adsorbent for manganese removal in surface water.

ABSTRAK

Mangan adalah antara logam berat yang dikesan di dalam air sungai sejak akhir-akhir ini dan menjadi permasalahan kepada alam sekitar kerana mangan akan menyebabkan kesan negatif kepada ekosistem dan juga bekalan air. Kajian ini menyelidiki keberkesanan kulit jantung pisang (BBP) sebagai penjerap untuk menyingkirkan mangan di dalam air. Penjerap BBP telah diaktifkan melalui kaedah kimia dan haba. Hasil Indeks Kualiti Air untuk Sungai Panchor menunjukkan bahawa sungai itu diklasifikasikan sebagai Kelas II, dikategorikan sebagai bersih dengan kepekatan mangan tertinggi sebanyak 0.575 mg/L dikesan menggunakan alat ICP-OES. Analisis FESEM-EDX menunjukkan penjerap BBP adalah berkerut dengan lekukan yang dalam, permukaan dalaman yang kasar dan tebal secara semulajadi di mana menyediakan permukaan yang maksimum untuk proses penjerapan. Analisis FTIR penjerap BBP mengesahkan penglibatan kumpulan karboksil and hidroksil yang menyumbang kepada proses penjerapan. Analisis XRD menunjukkan bahawa struktur penjerap BBP adalah amorfus. Luas permukaan penjerap BBP adalah 2.12 m²/g dengan jumlah pori $0.0139 \text{ cm}^3/\text{g}$ dan diameter pori 64.35 nm. Eksperimen penjerapan berkumpulan menilai parameter yang mempengaruhi proses penjerapan termasuklah pH, dos penjerap, kepekatan mangan awal dan masa sentuhan. Keadaan optimum dicapai pada pH 7, dos penjerap 0.5g, kepekatan awal mangan 10 mg/L dan 150 minit masa sentuhan penjerapan. Kajian isoterma penjerapan menunjukkan bahawa model Langmuir merupakan model yang paling sesuai untuk penjerapan BBP terhadap mangan dengan $R^2 > 0.9963$. Penjerapan mangan mengikuti kinetik pseudo-kedua yang mewakili proses kemisorpsi. Kadar penyahjerapan maksimum sebanyak 92% dicapai dalam kitaran pertama dengan kadar pemulihan sebanyak 94.18% dalam 30 minit menggunakan 0.1M HCl. Penjerap BBP mampu menyingkir 55% kandungan mangan di dalam air Sungai Panchor dengan kepekatan awalan mangan 0.095 mg/L. Kajian ini menunjukkan bahawa penjerap BBP berpotensi digunakan sebagai penjerap untuk menyingkirkan mangan dalam air sungai.

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LIST OF SYMBOL AND ABBREVIATIONS

-	Percentage
-	Centimeter
-	Gram
-	Milligram
-	Milliliters
-	Liters
-	Volume
-	Microgram per liter
-	Milligram per liter
-	Milligram per liter Milligram per gram Gram per centimeter cube
-	Gram per centimeter cube
-	Gram per mole
-	Gram per liter
XA	Gram per milligram minute
-	moles per liter
-	Revolution per meter
-	Lambda
-	Degree Celsius
-	Second
-	Minute
-	Intelligence Quotient
-	Million tones
-	Ministry of Health
-	Banana Blossom Peels
-	Department of Environment
-	Field emission scanning electron microscopy
-	Energy-dispersive X-ray spectroscopy

FTIR	-	Fourier-transform infrared spectroscopy
XRD	-	X-ray Diffraction
BET	-	Brunauer-Emmett-Teller
ICP-OES	-	Inductively coupled plasma – optical emission
		spectrometry
MLD	-	Million per day
INWQS	-	Interim National Water Quality Standards
NAHRIM	-	National Hydraulic Research Institute of
		Malaysia
Mn	-	Manganese
WHO	-	World Health Organization
АРНА	-	American Public Health Association
EPA	-	United States Environmental Protection Agency
EC	-	Electrocoagulation
EO	-	Electrochemical oxidation
RO	-	Electrochemical oxidation Reverse osmosis Ultrafiltration
UF	-	Ultrafiltration
MF	-	Microfiltration
NF	-	Nanofiltration
PVA/CS	XA	Polyvinyl alcohol/chitosan
ΔG^0	-	Gibbs energy change
ΔH^0	-	Enthalpy change
ΔS^{0}	-	Entropy change
kJ/mol	-	Kilo joule per mol
Κ	-	Thermodynamic equilibrium constant
Т	-	Absolute temperature in Kelvin
R	-	Gas constant
NaOH	-	Sodium hydroxide
NaHCO ₃	-	Sodium bicarbonate
H_2SO_4	-	Sulfuric acid
HNO3	-	Nitric acid
H_3PO_4	-	Phosphoric acid
NaNO ₃	-	Sodium Nitrate
Na ₂ EDTA	-	Disodium salt dihydrate



HCl	-	Hydrochloric acid
DO	-	Dissolve Oxygen
COD	-	Chemical oxygen demand
BOD	-	Biochemical oxygen demand
TSS	-	Total suspended solid
WQI	-	Water quality index
R^2	-	Correlation coefficient
K_L	-	Langmuir constant
K_F	-	Freundlich constant
Ν	-	Intensity
q max	-	Adsorption capacity

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Water plays crucial role in human survivability and national development. Therefore, it has to be safe, clean and palatable for consumption (Alam *et al.*, 2018). Water supply in Malaysia is mainly extracted from surface water sources, particularly from a river and in some areas from groundwater sources (Moni *et al.*, 2017). In general, surface water with minimum treatment is potable if it is not affected by anthropogenic activities. However, rivers nowadays have been used as disposal routes for liquid and solid waste, mainly from residential, agricultural and industrial activities (Afroz *et al.*, 2014). The river water quality in Malaysia is worsening due to the rapid development in various sectors and population growth, contributing to the increase in water pollution. The pollution has made the water quality issue becomes a great concern, and access to clean water sources has become challenging over the years (Ashraf and Hanfiah, 2017; Ghani *et al.*, 2017).

Hanfiah, 2017; Ghani et al., 2017).
Heavy metal pollution has gained public attention in recent decades due to its abundance, persistence, and toxicity in water that cause many environmental problems and pose a threat to human health when present in drinking water (Fallahzadeh et al., 2017; Salam et al., 2019). The situation worsens as heavy metals are not degraded naturally and persist in the sediment of water bodies (Paul, 2017). Industries that caused an abundance of production of heavy metal, such as the chemical manufacturing industry, mining, municipal effluents, and other anthropogenic activities, has contributed to heavy metal pollution (Salam et al., 2019). Non-industrialised areas were also severely contaminated due to the widespread of heavy metals along the river (Wu et al., 2016). Heavy metals such as manganese, copper,



zinc, and iron are needed as nutrients in small quantities. However, these metals can cause deterioration to the human body at higher concentrations or excess consumption in the long term (Paul, 2017). Some heavy metals may have acute or chronic impacts when present in aquatic habitats, drinking water sources, and recreational water bodies (Huber *et al.*, 2016).

Manganese (Mn) is a trace metal that is found mainly as oxides, carbonates, and silicates in many different minerals, with pyrolusite (manganese dioxide) as the most common naturally-occurring form (Milatovic and Gupta, 2018). Manganese can be found abundantly in the earth's crust and water sources, which exist in a broad range of oxidation states and species in the water (Tobiason *et al.*, 2016). Manganese has been commonly detected in surface water which resulted in major water quality issues over the years. Manganese has been found in various types of agricultural, municipal, domestic and industrial wastewaters effluent (Marsidi *et al.*, 2018). Industries dealing with metals such as steel alloy production, battery manufacturing, pesticides, mine quarry operations and pharmaceuticals contributes to manganese pollution and its release to the environment, especially through wastewater discharges (Mthombeni *et al.*, 2016). Therefore, it is necessary to remove manganese from surface water.



The World Health Organization (2011) has set the standard of 0.05 mg/L as the maximum manganese concentration level in domestic water supplies. A high level of manganese in drinking water that exceeds the permissible value is considered unacceptable because it affects not only the aesthetic water quality, but also operational problems in the distribution systems, such as iron pipe corrosion. Manganese causes turbidity and a black-brown colour to drinking water (Tobiason *et al.*, 2016; Rose *et al.*, 2017). Excessive manganese accumulation in specific brain areas produces neurotoxicity, leading to degenerative brain disorder (Milatovic and Gupta, 2018). The initial toxic symptom associated with manganese is a psychiatric nature disorder known as *locura manganica* resembling schizophrenia, followed by a permanently crippling neurological (extrapyramidal) disorder that is clinically similar to Parkinson's disease (Bjørklund *et al.*, 2017).

High manganese concentrations have been found in some Malaysian rivers. The highest manganese concentration of 4.7 mg/L was detected at Kepayang River, Perak (Affandi and Ishak, 2018). Apart from that, high manganese concentrations that exceed the standard for raw and drinking water were also found in the Kelantan River Basin, Benut river, Sembilang River, Langat River, Kerian River, Semenyih River and Sedili Kecil River (M. *et al.*, 2012; Al-Badaii *et al.*, 2016; Ismail *et al.*, 2017; O. Basheer *et al.*, 2017; Ibrahim *et al.*, 2020; Mohamed *et al.*, 2020b; Abdul Maulud *et al.*, 2021). The high manganese concentrations reported in these rivers could be attributed to mining activities, industrial sewage, agricultural activities, and also industrial effluents. Since water used mainly derived from surface water, the water source should be safe and palatable for consumption and drinking, without harmful contamination of manganese.

Various treatment technologies have been developed to treat surface water that contains manganese. The adsorption process is one of the potential alternatives for low-cost and efficient methods to improve surface water quality (Burakov *et al.*, 2018). This process has been broadly studied to develop high selectivity and efficient adsorbent materials (Liu *et al.*, 2017b; Sajid *et al.*, 2018). Adsorption is a simple process that can be synthesised from waste materials (Jain *et al.*, 2016). The materials for adsorbents are relatively cheap, less toxic, easy to functionalise over other substrates, and highly efficient. Many factors affect the efficiency of adsorbents, including initial concentration, temperature, adsorbent dose, pH, contact time, and stirring speed (Cermakova *et al.*, 2017).



Banana blossom peels (BBP) are considered one of the new and potential materials for the adsorption of heavy metals (Padmaja, 2017). To date, investigations on banana blossom peel as adsorbent had only been studied on lake water sample. The results showed that BBP adsorbent was able to reduce 79.72% turbidity, 88.24% total solids, 90.15% electrical conductivity, and 61.01% chloride in the lake water sample (Gopakumar *et al.*, 2018). However, there is insufficient technical information regarding the synthesis process of the adsorbent, mechanism of the adsorption process and effects of parameters that govern the adsorption process. In order to produce a safe and cost-effective method in treating surface water containing manganese, this study investigates the synthesis process of BBP as an adsorbent.

1.2 Problem Statement

Sungai Panchor has been used as the water source for the people in Pekan Panchor, Muar, Johor since 2017 according to data by Department of Environment (DOE, 2018). In 2018, the Department of Environment Malaysia reported that Sungai Panchor was slightly polluted (Class III) with a water quality index (WQI) of 61 and needed extensive water treatment. The need for treatment is due to the anthropogenic activities resulted from agricultural runoff from palm oil plantations nearby, construction, domestic waste, residential, food stalls, and natural erosion (Rosman, 2016). The demand for clean water is increasing as the population of Pekan Panchor increases (DOSM, 2020). Furthermore, the anthropogenic activities had caused issues of nutrients and minerals contamination in the river, especially manganese ions. Hence, it is important to remove manganese from the river water by employing an effective treatment process.

Several techniques can be applied to remove manganese from water, including precipitation, coagulation/flocculation, ion exchange, membrane filtration, and oxidation (Patil *et al.*, 2016). All these methods have drawbacks of the expensive and complex process except for adsorption. Adsorption is preferable owing to cost-effectiveness and high efficiency in removing contaminants.

In the adsorption process, the adsorbents from agricultural waste must be pretreated by chemicals to prevent secondary pollution and increase the adsorption capacity in terms of the existence of hydroxyl and carbonyl groups in their molecular structure. The use of natural agricultural wastes usually contributes to problems such as high concentration of chemical oxygen demand, biological oxygen demand, total organic carbon, and low adsorption capacity value. The problems are due to the released of soluble organic compounds in the plant resulting in secondary pollution (Acharya *et al.*, 2018).

This study intends to investigate the use of low-cost BBP adsorbent to remove manganese from water. The BBP is considered waste because only a few types of banana blossom are suitable to be consumed. The banana blossom has to be harvest to enhance the quality of banana fruits. The harvesting generates a significant amount of waste (Palomo, 2018). Moreover, the usage of banana wastes such as its peels and blossom peels are increasing. Hence, the utilisation of BBP would reduce the overall banana waste disposal problems.

Recent research on the utilisation of BBP as an adsorbent is very limited, and the only reported research was successfully conducted by Padmaja (2017) for dye removal. Understanding the capability of BBP as a new potential adsorbent to adsorb manganese can help in future studies. Furthermore, the major characteristics of banana blossom peel that contributes to the manganese removal are its chemical composition consisting of cellulose and lignin, which contain a variety of functional groups.



1.3 **Objectives**

This study aims to synthesise and evaluate the performance of the BBP adsorbent to remove manganese in water. It embarks on the following objectives:

(i) To synthesise BBP as an adsorbent using a chemical treatment method and characterise its physiochemical properties.

(ii) To investigate the effects of adsorbent dosage, pH, manganese initial concentration, and contact time toward manganese removal using the BBP adsorbent. (iii) To elucidate the adsorption mechanism of BBP adsorbent by isotherm and kinetic study.

(iv) To determine the reusability of the manganese-loaded BBP adsorbent.

1.4 Scope of the Study

MINA The scope of this study includes the collection of water samples conducted at Sungai Panchor, Muar, Johor, and the evaluation of water quality of Sungai Panchor. Furthermore, manganese concentrations in Sungai Panchor were detected for several weeks under different weather conditions.

BBP was synthesised as an adsorbent via a chemical treatment method using hydrochloric acid (HCl) and sodium hydroxide (NaOH). The BBP adsorbent was characterised before and after the adsorption process by using field emission scanning electron microscopy (FESEM), energy-dispersive X-ray spectroscopy (EDX) analysis, Fourier-transform infrared spectroscopy (FTIR), X-ray diffraction (XRD) analysis, and Brunauer-Emmett-Teller (BET) analysis.

The performance of BBP adsorbent in removing manganese was evaluated using batch adsorption experiments. The investigated parameters include the effects of adsorbent dosage (0.1g, 0.3g, 0.5g), pH (4, 5.5, 7), and manganese initial concentration (10 mg/L, 30 mg/L, 50 mg/L) toward contact time (5, 10, 30, 60, 90, 120, 150 min). The concentrations of manganese ions before and after adsorption were evaluated using inductively coupled plasma optical emission spectrometry (ICP-OES).

The adsorption isotherm data obtained at different manganese concentrations were analysed by the Langmuir and Freundlich isotherm models. The experimental



data were further analysed using pseudo-first-order and pseudo-second-order models to elucidate the adsorption kinetic of manganese onto BBP adsorbent.

The reusability of BBP adsorbent was investigated using HCl as desorbing agent up to three desorption cycles. The potential application of the BBP adsorbent was evaluated in a batch adsorption study using surface water from Sungai Panchor to test manganese removal efficiency.

1.5 Significance of Study

The significance of this study to the field of water treatment technology is the new knowledge on the synthesis and application of BBP adsorbent for the removal of manganese. There have been many previous studies on the performances of various adsorbents for removing manganese. However, none have reported on the removal of manganese using BBP as a low-cost and environmentally friendly adsorbent.

Hence, this study intends to establish new knowledge on the effects of pH, adsorbent dosage, and manganese concentration against contact time using BBP adsorbent. This study also analysed in detail the manganese removal capability, characteristics, and mechanism involved in the adsorption process.

The knowledge on the synthesis of BBP adsorbent towards future research is important because it can be applied by the waterworks industry in Malaysia that demands economic and effective processes.

In addition, the BBP adsorbent has the potential to improve the quality of surface water by reducing manganese concentration. The porous surface structure and functional groups generated in the synthesis process can promote adsorption, thereby enhancing manganese removal by the adsorbent.

The toxicity caused by the excessive exposure and long-term effects of manganese in surface water on human health can be reduced. The BBP adsorbent is a new alternative towards an effective water treatment process in removing manganese from surface water.



CHAPTER 2

LITERATURE REVIEW

This chapter provides an overview of surface water contamination and information on heavy metal contamination caused by industrial activities. The occurrences of manganese, the negative impacts on living organisms, and the application of the adsorption process for its removal in water treatment are discussed in this chapter. After the adsorption, this chapter also explains the reusability process that helps maintain adsorbent performances. The adsorption mechanism of manganese ions and the adsorbent is further elucidated in isotherm and kinetic study.



Previous studies revealed the development of various agricultural waste as low-cost adsorbents to remove manganese, which provide insight for this study to improve the adsorbent properties. The utilisation of banana blossom peels (BBP) portrays a potential new adsorbent as described by its characteristics and efficiency. The application of BBP adsorbent is also possible for a future low-cost adsorbent in the water treatment process.

2.1 Surface Water Quality

Water is an essential component in life; for social and economic development (Ahmed *et al.*, 2014). It is needed for cell growth, body coolant, tissue protection, digestion, adsorption, excretory system, and maintaining a good weight (Marsidi *et al.*, 2018). In Malaysia, 98% of the total water used is mainly derived from the river, whereas the remaining 1% originates from groundwater (Lee Goi, 2020). In 2020, the population in Malaysia was approximately 32.7 million and projected to increase yearly (DOSM, 2021). Water usage in Malaysia was estimated to rise to approximately 25,455 million litter per day (MLD) in 2050. Nevertheless, the common issue of surface water is the

sources that are prone to pollution and water level declined during the dry season (Kasim *et al.*, 2016). Most rivers in Malaysia have been used as dumping sites for toxic wastes, especially heavy metals discharged from industry (Daniel and Kawasaki, 2016). In regard to this, the Ministry of Health (MOH), Malaysia, has set guidelines for Malaysia standard drinking water quality and raw water quality, as shown in Table 2.1.

Parameter	Group	Recommended Raw Water Quality Acceptable Value (mg/litre (unless otherwise stated))	Drinking Water Quality Standards Maximum Acceptable Value (mg/litre (unless otherwise stated))
рН	1	5.5 - 9.0	6.5 - 9.0
Turbidity	1	1000 NTU	5 NTU
Temperature	1	.vV	101
Ammonia Nitrogen	2	1.5	1.5
Chemical Oxygen Demand (COD)	K2A	10	-
Biochemical Oxygen Demand (BOD)	2	6	-
Manganese	2	0.2	0.1

Table 2.1: Drinking and raw water standard in Malaysia (Ministry of Health
Malaysia, 2012)

Table 2.2 depicts the National Water Quality Standards for Malaysia according to its classes. Surface water quality could be gradually improved or upgraded to be a better water class based on the standard values of 72 parameters in 6 water use classes. The Interim National Water Quality Standards (INWQS) has defined six different river classes (I, IIA, IIB, III, IV, and V) based on their classification, descending water quality and uses. Class I is referred to as the best, while water quality for class IV is very poor.

Parameter	Unit	Class					
	Unit	I	IIA	IIB	III	IV	V
Ammoniacal Nitrogen	mg/l	0.1	0.3	0.3	0.9	2.7	>2.7
BOD	mg/l	1	3	3	6	12	>12
COD	mg/l	10	25	25	50	100	>100
DO	mg/l	7	5-7	5-7	3-5	<3	<1
pH	-	6.5-8.5	6-9	6-9	5-9	5-9	-
TSS	mg/l	25	50	50	150	300	300
Temperature	°C	-	Normal	-	Normal	-	-
Turbidity	NTU	5	50	50	-	-	-
Manganese	mg/l	Absent	0.1	0.1	0.1	0.2	Above
Water Quality Index	-	<92.7	76.5-92.7	76.5-92.7	51.9-76.5	31.0-51.9	>31.0
emarks: Class I	Conserva	tion of natu	ral environme	nt.			AM
Water Supply I - Practically no treatment necessary							

Table 2.2: National water quality standards in Malaysia (INWQS, 2016)

Class I	Conservation of natural environment.			
	Water Supply I - Practically no treatment necessary.			
	Fishery I - Very sensitive aquatic species.			
Class IIA	Water Supply II - Conventional treatment required.			
	Fishery II - Sensitive aquatic species.			
Class IIB	Recreational use with body contact.			
Class III	Water Supply III - Extensive treatment required.			
	Fishery III - Common, of economic value and tolerant species; livestock drinking.			
Class IV	Irrigation.			
Class V	None of the above.			



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