NATURAL COMPOSITE ADSORBENT USING ALGINATE AS BINDER AID FOR LEACHATE TREATMENT

AWATIF BT MD ISA

UNIVERSITI SAINS MALAYSIA

2018

NATURAL COMPOSITE ADSORBENT USING ALGINATE AS BINDER AID FOR LEACHATE TREATMENT

by

AWATIF BT MD ISA

Thesis submitted in fulfilment of the requirements
for the degree of
Master of Science

August 2018

ACKNOWLEDGEMENT

ALHAMDULILLAH, and thank you ALLAH S.W.T for giving me the wisdom, strength, support and knowledge to explore things, for the guidance and help to surpass all the trials I have faced. This dissertation would not have been a success without the guidance and help of several individuals whom in one way or another contributed to this study. First and foremost, I would like to express my sincere gratitude to my supervisor, Prof Dr. Mohd Suffian Yusoff, who always gives me infinite guidance and advice throughout completing this project. I would like to extend my humble thanks to Dr. Mohamad Anuar Kamaruddin, for his advices and knowledge contribution throughout my research progress.

I would like to thank Universiti Sains Malaysia (USM) for funding my studies under Fellowship scheme and RUI USM grant (1001/PAWAM/814260). My sincere thanks also goes to Mr. Zaini, Mr. Mohad, Mr. Nizam and Mrs. Shamsiah for their technical guides during all experimental works. Apart from that, I would like to thank my dear parents, Md. Isa Ismail and Hasani Khalid from the core of my heart for their never ending love, and for giving me support throughout the journey of completing my studies. I would also like to express my deepest gratitude to my research colleagues and friends for their support and motivation. Finally, I would like to express my sincere thanks to my husband, Mohamad Hazlami and my daughter Maryam Humaira that have inspired and motivated for me to finish my research successfully.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF PLATES

LIST OF SYMBOLS

		Unit
C_e	Adsorbate concentration at the equilibrium	(mg/L)
C_o	Initial concentration of parameters	(mg/L)
K_F	Freundlich constant	$(mg/g)(L/mg)^{1/n}$
K_L	Langmuir adsorption constant	(L/mg)
K_1	Adsorption rate constant for pseudo-first-order	(min^{-1})
K_2	Adsorption rate constant for pseudo-second-order	(g/mg.h)
n	Constant for Freundlich isotherm	-
$\frac{1}{n}$	Freundlich heterogeneity factor	(mg/L)
Q_{max}	Maximum adsorption capacity for the solid phase loading (mg/g)	(mg/g)
q_e	The amount of adsorbate adsorbed per unit mass of adsorbent (mg/g)	(mg/g)
q_t	Amount of adsorbate adsorbed per unit mass of adsorbent at equilibrium at time, t	(mg/g)
q_{t}, cal	Calculated adsorption uptake at time, t	(mg/g)
q_{t}, exp	Experimental adsorption uptake at time, t	(mg/g)
R_L	Separation factor	-
R^2	Coefficient of determination	-
m	weight of composite adsorbent	(g)

V	volume of sample solution	(mL)
Y	Predicted responses	-
X	Composite adsorbent preparation variable	-

LIST OF ABBREVIATIONS

3D	3 dimension
2FI	Two Factor Interaction
AC	Activated carbon
AG	Alginate
ANOVA	Analysis of variance
APHA	American Public Health Association
ASBR	Anaerobic Sequencing Batch Reactor
ASTM	American Society for Testing and Materials
BET	Brunauer-Emmett-Teller
BOD_5	Biochemical Oxygen Demand
C	Carbon
CCD	Central composite design
CO_2	Carbon Dioxide
COD	Chemical Oxygen Demand
DOE	Department of Environment
EQA	Environment Quality Act
FTIR	Fourier Transform Infrared
H_2	Hydrogen gas
H_2O	Water
H_2S	Hydrogen sulphide
ICP-OES	Inductively-Coupled Plasma - Optimal Emission Spectroscopy

IUPAC	International Union of Pure and Applied Chemistry
LS	Limestone
MB	Methylene Blue
MHLG	Ministry of Housing and Local Government
MSW	Municipal Solid Waste
N_2	Nitrogen
O_2	Oxygen
OVAT	One-Variable-At-a-Time
PAC	Powder activated carbon
PLS	Powder limestone
RSM	Response Surface Methodology
SBR	Sequencing batch reactor
SEM	Scanning Electron Microscopy
USGS	United States Geological Survey
VOAs	Volatile Organic Acid
w/w	Weight to weight
w/v	Weight to value

PENJERAP KOMPOSIT SEMULAJADI DENGAN MENGGUNAKAN ALGINAT SEBAGAI PEMBANTU PENGIKAT BAGI RAWATAN LARUT LESAPAN

ABSTRAK

Penjanaan larut lesapan merupakan satu isu yang sangat besar yang memberi kesan negatif kepada alam sekitar kerana mengandungi kepekatan bahan organik dan bukan organik yang tinggi. Penjerapan telah terbukti antara kaedah yang berkesan untuk merawat larut lesapan. Oleh itu, kajian ini mengkaji keberkesanan penjerap komposit yang dihasilkan daripada campuran serbuk karbon teraktif (PAC), serbuk batu kapur (PLS) dan alginat (AG) untuk menyingkirkan keperluan oksigen kimia (COD) dan Fe(II) di dalam larut lesapan. Hasil keputusan menunjukkan bahawa nisbah campuran (w/w) PAC-PLS dan kepekatan AG yang terbaik adalah 7:3 dan 2% w/v. Luas kawasan permukaan penjerap komposit pula adalah 555.2 m^2/g dengan saiz purata diameter liang iaitu 3.515 nm. Tambahan lagi, didapati bahawa kumpulan hidroksil, karboksil dan alkena yang ditemui dipermukaan penjerap komposit meningkatkan lagi proses penjerapan. Seterusnya, berdasarkan kajian kelompok, keadaan terbaik untuk rawatan larut lesapan adalah 16 g (dos penjerap komposit), pH 7 (pH awal), 200 rpm (kelajuan gegaran) dan 150 min (masa sentuhan). Berdasarkan kepada kajian isoterma, mekanisma penjerapan bagi COD boleh digambarkan melalui model Freundlich (penjerapan fizikal) dan Fe(II) pula melalui model Langmuir (penjerapan kimia). Dalam masa yang sama, melalui kajian kinetik mendapati bahawa penjerap komposit mematuhi model pseudo-tertib-kedua (penjerapan kimia) bagi kedua-dua parameter. Kesemua model memperoleh nilai $R^2>0.9$ menunjukkan bahawa proses penjerapan fizikal dan penjerapan kimia berlaku pada keseluruhan permukaan penjerap komposit semasa proses penjerapan.

NATURAL COMPOSITE ADSORBENT USING ALGINATE AS BINDER AID FOR LEACHATE TREATMENT

ABSTRACT

Generation of leachate is the biggest issue that caused negative impact to the environment due to high concentration of organic and inorganic materials. Adsorption is proven to be among the effective methods for leachate treatment. Therefore, this study examined the effectiveness of composite adsorbent producing from the mixture of powder activated carbon (PAC), powder limestone (PLS) and alginate (AG) to remove chemical oxygen demand (COD) and Fe(II) in leachate. The result indicated the best mix ratio (w/w) of the PAC-PLS and AG concentration were 7:3 and 2% w/v, respectively. Surface area of the composite adsorbent was 555.2 m^2/g with average pore diameter 3.5 nm. In fact, the hydroxyl, carboxyl and alkene groups discovered on the surface of the composite adsorbent that enhanced the adsorption process. Next, from batch study, the best condition for leachate treatment is 16 g (composite adsorbent), pH 7 (initial pH), 200 rpm (shaking speed) and 150 min (contact time). According to the isotherm study, adsorption mechanism for COD can be described by Freundlich model (physisorption) while Fe(II) is particularly represented by Langmuir model (chemisorption). Meanwhile, through the kinetic study, the composite adsorbent was also found to obey pseudo-second-order model (chemisorption) for both parameters. All the models obtained the value of R^2>0.9 and indicating that both physisorption and chemisorption processes occurred at the entire surface of composite adsorbent during adsorption process.

CHAPTER ONE

INTRODUCTION

1.1 Overview

Municipal solid waste (MSW) is defined as any scrap materials that are broken, contaminated, and unable to be used which requires disposal by the authority (UNDP, 2008). Furthermore, by referring to Malaysia's Solid Waste and Public Cleansing Management Act of 2007, the source of MSW mainly comes from commercial, household, institutional and public solid wastes (Act 672) (Tan et al., 2014). United States Environmental Protection Agency (USEPA) also defined that MSW is unwanted materials (product packaging, grass clippings, furniture, clothing, bottles, food scraps, newspapers, appliances, paint, and batteries) of daily items originating from homes, schools, hospitals, and commercial areas (USEPA, 2017).

A study conducted by Tan et al. (2014) reported that the increasing trend in MSW generation is directly proportional to the population of Malaysia. The amount of waste generation has increased by 31.62%, following the expansion of Malaysian population from 21.13 million in 1997 to 28.60 million in 2010. Moreover, further increment of MSW generation beyond 2010 is also predicted as the population continues to grow over the years. For the years between 2020 to 2030, the population is projected to grow from 32.40 million (with MSW of 9.82 Metric ton) to 36.09 million (with MSW of 13.38 Metric ton), respectively. Moreover, the increasing rate of MSW production is also directly influenced by rapid economic growth, industrialization and urbanization process (Ismail and Manaf, 2013).

Landfill has been the most common practice for MSW disposal as compared to the others. For a more advanced practice, such as incineration, requires intensive knowledge because the sophisticated technology releases gaseous pollutants that needs ultimate disposal (Fazeli et al., 2016). Meanwhile, landfill is preferred in tackling the overwhelming MSW conundrums due to lower cost implication in preparing for the site and simplicity in terms of technical and operational systems. In Malaysia, 95% to 97% of the MSW was disposed in landfills. However, this percentage will be reduced by year 2020 as 44% of the MSW will be disposed in sanitary landfills (Periathamby et al. 2009).

In landfill disposal sites, degradation of the organic fraction of wastes, together with percolating rainwater, can lead to production of leachate (Abouri et al., 2016). Percolation occurs when the magnitude of gravitational forces exceeds the holding forces. Leachate is a dark coloured liquid with strong smell, produced by the organic and inorganic matters leaching out from the landfill wastes (Fauziah et al., 2013; Peng, 2013). Moreover, leachate is a high strength wastewater with extreme level of pH, chemical oxygen demand (COD), biochemical oxygen demand (BOD), ammonia nitrogen (NH_3-N), inorganic salts and heavy metals (Kamaruddin et al., 2013). Based on the data recorded in 2010, about 26,000 tons of MSW was generated each day that resulted in the production of leachate is 3.9 million per day in Malaysia (Kamaruddin et al., 2017).

1.2 Problem Statement

Based on the current practice of MSW management, Malaysia is highly dependent on landfill to treat MSW, with an estimated amount of 93.5% MSW is disposed at the landfill site (Pariatamby et al., 2009). This can create problems and issues if a proper collection system is not provided at the landfill site. In environmental aspect, raw leachate from landfills laterally seeps into soil compartments and causes soil contamination (Emenike et al., 2016). Consequently, it can lead to higher possibilities of groundwater contamination. On top of that, the existing contents of iron (Fe(II)) and COD in leachate can potentially contribute risks to human and environment (Mojiri et al., 2015).

Therefore, a study of COD concentration is important in order to measure the amount of organic matter present in leachate. The COD concentration in leachate is predicted to reduce over time due to the reduction of organic pollutants, which has undergone leaching in the landfill (Lee and Nikhraj, 2014). However, it is still essential to remove organic materials to ensure its concentration is below allowable threshold as following standard of discharge limit by Environment Quality Act (EQA) which is 400 mg/L (DOE, 2010). The most significant impact of biodegradable organic material is it can cause a reduction of oxygen concentration in water. Hence, this will affect the aquatic communities, such as species of plants and animals. The species will then migrate to an area that is absence of organic pollutants. In addition, the metabolism of these organic materials by anaerobic bacteria produces methane gas (CH_4), hydrogen sulfide (H_2S) and NH_3-N, which consequently pollutes the water and causes adverse effects to human and other living things if the water is consumed (Jumaah et al., 2016).

Heavy metal like Fe(II) is needed for living organisms. However, when the concentration exceeds the allowable effluent as stated in EQA (5 mg/L) (DOE, 2010) it becomes toxic and gives adverse effects to the discharge area. As a result, Fe(II) element will then enter into human body through food and water consumption that comes from this contaminated source which cannot be broken down and remain either in environment or human body for a long time. Hence, this causes reduction of growth and development, cancer, damage to nervous system, human physiology and other biological systems such as irreversible brain damage (Pariatamby et al., 2015; Jayanthi et al., 2017; Karnib et al., 2014).

There are several complaints received from the public regarding this leachate issue. As reported by Berita Harian on 18 December 2016, fishermen committee in Changkat, Pulau Pinang have reported that untreated leachate was directly discharged into the sea from Pulau Burung Landfill Site (PBLS). As the distance from PBLS is located just within 200 meters from the sea, the untreated leachate has created pollution to marine ecosystem and affected the food chain of aquatic life, which eventually gives bad impacts to human health.

Adsorption is one of the physical-chemical treatment methods that has several advantages in terms of its operational cost and method. The studies on leachate treatment for stabilization landfill have been conducted widely, especially on the adsorption of an individual precursor (AC, LS, zeolite, silica and polymeric adsorbent) and composite adsorbent to eliminate the pollutants (Ghani et al., 2017; Luukkonen et al., 2015; Othman et al., 2010). Above all, AC and LS are the most preferable materials to be used in leachate and wastewater treatment (Mojiri et al., 2015; Ghaly et al., 2007; Kamaruddin et al., 2014a). This is because AC is a non-polar compound containing small particles in size and has the affinity to adsorb organic substances, whereas LS is commonly applied in removing of heavy metals (Ali et al., 2012). However, application of AC alone is not economical since high energy is required during the activation process (Shehzad et al., 2015). Furthermore, the application of AC as a main ingredient in a large scale will only raise up the operational cost of a treatment plant due to high cost of AC over the years (Ali, 2010). Furthermore, the utilization of LS also give disadvantages which is the efficacy adsorption of organic matter is low (Suhara, 2010). Thus, mixing of LS and AC will reduce the usage of AC in composite adsorbent and more economical since LS is a low cost adsorbent. Furthermore, the idea of combining hydrophobic (AC) and hydrophilic (LS) compounds in order to make an effective composite adsorbent for leachate treatment is highly recommended.

In a previous research, AG acted as a binder for AC and LS was introduced by Kamaruddin (2015a) in removing both organic and inorganic substances in textile wastewater. Moreover, the application of AG is widely used for dye treatment (Thomason, 2011; Hassan et al., 2014; Ai et al., 2009; Benhouria et al., 2015). Nevertheless, there are no studies conducted by previous researchers regarding the application of AG as a binder for AC and LS, especially on the removal of COD and Fe(II) in landfill leachate. The success of using AG as a binder for AC and LS will give benefits to the leachate treatment field because AG does not only act as a binder, but also contribute to the uptake of heavy metal ions and COD. Therefore, this research is expected to demonstrate the effectiveness and performance of the composite adsorbent for landfill leachate treatment.