REMOVAL OF TRICLOSAN FROM AQUEOUS SOLUTION BY ACTIVATED CARBON AND NYLON 6,6 NANOFIBER

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DEDICATION

This thesis is specially dedicated to my dear husband,

Mohd Aliff Omar bin Seman

My beloved mother and father,

Nor Hamidah binti Arifin and Mohd Khori bin Mat Hassan

&

My beloved mother-in-law and father-in-law,

Noor Hashimah binti Arifin and Seman bin Bakar

Thanks for all your prayers, sacrifices, patience, support and understanding

throughout this journey

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ABSTRACT

Triclosan (TCS) is one of the biocide that functions as antibacterial and antifungal agent and has the ability to kill and hinder the growth of bacteria. Hence, it is used in many personal care and health care products such as in shampoo, detergent, first aid, deodorant and cosmetics. However, TCS can cause health and environmental problems such as environmental pollutions, carcinogenic impurities, acute toxicity and endocrine disruption. Due to rapid urbanizations and growth populations, TCS was detected in many sewage treatment plants, river, and soil. The conventional system that was used to remove TCS and other pollutants from water used a lot of chemicals, involved a time-consuming procedure and cannot effectively remove all the pollutants from water. Therefore, the effective method to remove TCS with high removal rate in shorter time was studied. The aim of this study was to investigate the removal of TCS from aqueous solution by combining the activated carbon (AC) with nylon 6,6 nanofiber. In this study, AC from coconut pulp waste (*Cocos nuciefera*) and nylon 6,6 nanofiber were used to remove TCS from aqueous solution. The effects of physico-chemical parameters and physical-chemical characteristics for both AC and nanofiber have been studied to determine the best possible conditions for maximum removal of TCS. The AC was prepared by using coconut pulp waste treated with Zinc Chloride and was carbonized under nitrogen flow at 300 ˚C for 1 hour. The nylon 6,6 nanofiber [14 wt.%] was prepared by using electrospinning machine with injection rate at 0.4 mL/h, tip-to-collector distance at 15 cm, rotation speed at 1000 rpm and high voltage at 26 kV. The parameters studied for AC and nylon 6,6 nanofiber during the adsorption test were contact time, adsorbent dosage, agitation speed, initial TCS concentration, pH and temperature of the TCS solution. Besides that, the filtration test was done by using flat sheet membrane test machine at pressure 1.0 bar. The characteristics of AC and nylon 6,6 nanofiber were analysed by using Field Emission Scanning Electron Microscopy (FESEM), Fourier Transform Infrared Spectroscopy (FTIR) and Brunauer–Emmett–Teller (BET). The experiments show that the adsorption method by using AC can remove 83.3 % of TCS within 20 minutes and the filtration method by using nylon 6,6 can remove 90.2 % TCS within 5 minutes. After combine the adsorption and filtration method of both AC and nylon 6,6 nanofiber, the removal of TCS increased to 100 % removal in less than 5 minutes. For isotherms study, the AC follow Langmuir isotherm and nylon 6,6 nanofiber follow Freundlich isotherm. While for kinetics study, both AC and nylon 6,6 follow pseudo-second-order model. This study proved that the combination of AC and nylon 6,6 nanofiber can increase the removal of TCS in water.

ABSTRAK

Triklosan (TCS) adalah salah satu daripada biosid yang berfungsi sebagai agen antibakteria dan antikulat dan mempunyai keupayaan untuk membunuh dan menghalang pertumbuhan bakteria. Oleh itu, ia digunakan dalam banyak produk penjagaan diri dan penjagaan kesihatan seperti syampu, detergen, alat pertolongan cemas, pewangi ketiak dan kosmetik. Bagaimanapun, TCS boleh menyebabkan masalah kesihatan dan persekitaran seperti pencemaran alam sekitar, kekotoran karsinogen, ketoksikan akut, gangguan endokrin dan lain-lain. Oleh sebab perkembangan proses pembandaran dan populasi yang semakin pesat, TCS telah banyak dikesan di dalam loji rawatan air kumbahan, sungai, dan dalam tanah. Sistem konvensional yang digunakan untuk membuang TCS dan bahan pencemar lain dari air menggunakan banyak bahan kimia, melibatkan kaedah dan proses yang memakan masa dan tidak dapat menghapuskan semua pencemar dari air dengan berkesan. Oleh itu, kaedah berkesan untuk membuang TCS dengan kadar penyingkiran yang tinggi dalam masa yang lebih singkat dikaji. Tujuan kajian ini adalah untuk mengkaji penyingkiran TCS dari larutan akues dengan menggabungkan karbon diaktifkan (AC) dengan nilon 6,6 nanogentian. Dalam kajian ini, AC dari sisa hampas kelapa (*Cocos nuciefera*) dan nilon 6,6 nanogentian digunakan untuk membuang TCS daripada larutan akues. Kesan parameter dan ciri-ciri fizik-kimia untuk kedua-dua AC dan nanogentian telah dikaji untuk menentukan keadaan terbaik bagi menyingkirkan TCS secara maksima. AC disediakan dengan menggunakan sisa hampas kelapa yang dirawat dengan Zink Klorida dan dikarbonkan di bawah aliran nitrogen pada 300 ˚C selama 1 jam. Nilon 6,6 nanogentian [14 wt. %] pula disediakan dengan menggunakan mesin elektroputaran dengan kadar suntikan pada 0.4 mL/h, jarak antara jarum ke pengumpul pada 15 cm, kelajuan putaran pada 1000 rpm dan voltan tinggi pada 26 kV. Parameter yang dikaji untuk AC dan nilon 6,6 nanogentian semasa ujian penjerapan adalah masa persentuhan, dos penjerap, kelajuan goncangan, kepekatan awal larutan TCS, pH dan juga suhu. Selain itu, ujian penapisan dilakukan dengan menggunakan mesin uji nanogentian lembaran rata pada tekanan 1.0 bar. Ciri-ciri AC dan nilon 6,6 nanogentian dianalisa dengan menggunakan Mikroskop Elektron Pengimbas Pelepasan Medan (FESEM), Spektroskopi Inframerah Transformasi Fourier (FTIR) dan Brunauer-Emmett-Teller (BET). Eksperimen menunjukkan bahawa kaedah penjerapan menggunakan AC boleh mengalih keluar 83.3% TCS dalam masa 20 minit dan kaedah penapisan dengan menggunakan nilon 6,6 nanogentian boleh mengeluarkan 90.2% TCS dalam masa 5 minit. Selepas menggabungkan kaedah penjerapan dan penapisan kedua-dua AC dan nanogentian, penyingkiran TCS meningkat kepada 100% penyingkiran dalam tempoh kurang daripada 5 minit.Untuk kajian isoterma, AC mengikut isoterma Langmuir dan nilon 6,6 mengikut isoterma Freundlich. Manakala untuk kajian kinetik, kedua-dua AC dan nilon 6,6 nanogentian mengikut model pseudo-tertib-kedua. Kajian ini membuktikan bahawa gabungan AC dan nilon 6,6 nanogentian boleh meningkatkan penyingkiran TCS dalam air.

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LIST OF ABBREVIATIONS

AC	-	Activated Carbon
ATR	-	Attenuated Total Reflectance
ATTM	-	Ammonium Tetrathiomolybdate
BADGE	-	Bisphenol A Diglycidyl Ether
BET	-	Brunauer–Emmett–Teller
BP3	-	Benzophenone-3
BPA	-	Bisphenol A
BPS	-	4,4′-Sulfonyldiphenol
CECs	-	Chemical of Emerging Concerns
Cr(VI)	-	Hexavalent Chromium
CTAB	-	Cetyltrimethylammonium Bromide
DDBS	-	Sodium Dodecylbenzenesulfonate
DEHP	-	Di-(2-ethylhexyl) phthalate
DMOB	-	3, 3′-dimethoxybenzidine
DMP	-	Dimethyl phthalate
DOP	-	Dioctyl phthalate
EDC	-	Endocrine Disrupting Chemicals
EtP	-	Ethyl Paraben
FDA	-	Food And Drug Administration
FESEM	-	Field Emission Scanning Electron Microscopy
FTIR	-	Fourier Transform Infrared Spectroscopy
H_2SO_4	-	Sulfuric Acid
H_3PO_4	-	Phosphoric Acid
HB	-	Hydroxybenzoic Acid
HCl	-	Hydrochloric Acid
KOH	-	Potassium Hydroxide
MeP	-	Methyl Paraben
MF	-	Microfiltration
$MgCl_2$	-	Magnesium Chloride

MPEG-PLGA	-	Methoxy Polyethylene Glycol-Poly(Lactide-Co-Glycolide)
MTCS	-	Methyltriclosan
MWCO	-	Molecular Weight Cut-Off
NaOH	-	Sodium Hydroxide
Na_2CO_3	-	Sodium Carbonate
Na_2SO_4	-	Sodium Sulfate
NEP	-	New Emerging Pollutants
NF	-	Nanofiltration
NOM	-	Natural Organic Matter
PAN	-	Polyacrylonitrile
PC	-	Polycarbonates
PCL	-	Polycaprolactone
PDLLA	-	Poly(D,L-Lactide)
PES	-	Polyethersulfone
PESH	-	Polyethylenesulfonic Acid
PEtOx	-	Poly(2-Ethyl-2-Oxazoline)
PI	-	Polyamide, Polyimide
PLA	-	Polylactide
PLGA	-	Poly(D,L-Lactide-Co-Glycolide)
PrP	-	Propyl Paraben
PSF	-	Polysulfone
PTFE	-	Polytetrafluoroethylene
PVA	-	Polyvinyl Alcohol
PVDF	-	Polyvinylidene Fluoride
PVP	-	Polyvinyl Pyrrolidone
SiO_2	-	Silicon Dioxide
TCS	-	Triclosan
TiO_2	-	Titanium Dioxide
TTIP	-	Titanium Tetraisopropoxide
TX-100	-	Triton X-100
UF	-	Ultrafiltration
U.S	-	United States

UV-Vis	-	Ultraviolet-Visible
$ZnCl_2$	-	Zinc Chloride

LIST OF SYMBOLS

%	-	Percent
˚C	-	Celcius
˚K	-	Kelvin
µ	-	Micro
$>$	-	Greater than
$<$	-	Smaller than
-	-	Until
~	-	Approximate
A	-	Temkin equilibrium binding constant
B	-	Temkin constant related to heat of sorption
C	-	Boundry layer thickness
C_0	-	Concentration at initial
C_e	-	Concentration at equilibrium
C_x	-	Concentration after adsorption
cm	-	Centimetre
Da	-	Dalton
g	-	Gram
h	-	Hour
J	-	Flux
k	-	Kilo
k_1	-	Pseudo-first-order rate constant
k_2	-	Pseudo-second-order rate constant
k_{dif}	-	Intraparticle diffusion rate constant
K_F	-	Freundlich constant
K_L	-	Langmuir constant
L	-	Litre
m	-	Metre
M	-	Mass
n	-	Freundlich constant
Pa	-	Pascal

pk_a	-	Pollutant dissociation constant
q_e	-	Adsorption Capacity at equilibrium
Q_m	-	Maximum adsorption capacity
R^2	-	Correlation factor
R_L	-	Separation factor of Langmuir
rpm	-	Revolutions per minute
t	-	Time
V	-	Voltage
V	-	Volume
wt %	-	Percent weight

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Rivers and lakes are important sources of water that need to be protected from any pollutants. Nowadays, there are new emerging pollutants (NEP) detected in water sources especially in the rivers and lakes such as dyes, pesticides, pharmaceutical and personal care products (PPCPs), herbicides and fertilizers. PPCPs are the products used for individual health and cosmetics reasons such as medicine, deodorant, shower and so on. Ebele et al (2017) stated that, PCPPs is one of the Endocrine Disrupting Chemicals (EDC) contaminates that has ability to induce physiological effects in human and aquatic life even at low doses. EDC are chemicals that may happen naturally or from industrial and plasticizers substances that consist of hundreds or more exogenous chemicals, or mixtures of chemicals that disturb any aspect of hormone action (Zoeller et al, 2012; Ribeiro et al, 2016).

Triclosan (TCS) is a chlorinated aromatic compound that has functional groups of both ethers and phenols. TCS is one of the antibacterial and antifungal agents that are normally used in medical and consumer products, such as surgical scrubs, toothpastes, hand wash soaps, mouthwash, shampoos, plastics, toys, textiles and deodorants (Yueh et al, 2014; Teitelbaum et al, 2015). It has the ability to hinder the growth of microorganisms and due to its presence in many consumer products, it has been detected in most of the sediments, biosolids, surface water, soil, and aquatic species (Montaseri and Forbes, 2016). Wang et al (2017) reported that, the concentrations of TCS detected from conventional wastewater treatment plants at their influent, effluent and slurries produced were approximately 26.8 μgL^{-1}, 2.7 μgL^{-1} and 373.4 μgL^{-1}, respectively, and more than 90 % of the TCS was removed from water and wastewater treatment plants. But, TCS has a high log K_{ow} at 4.76 that shows its high sorption potentials to adsorb onto the sewage sludge, normally used as fertilizers (Dhillon et al, 2015).

Although TCS is an antibacterial agent, it also gives a potential risk to the human health and the environment. Zhu et al (2016) reported that, TCS was detected in 97 % of urine samples of 471 men with concentrations from 0.41 to 2.95 ng (mg creatinine)$^{-1}$ and it caused some adverse effects to the semen quality, such as, low sperm production and poor forward mobility. Besides, TCS has a high bioaccumulation potential and it can enter the food web system and cause toxicity to some aquatic life species (Dhillon et al, 2015).

The presence of TCS in the aquatic environment is well known to be a source of pollutants. Hence, several treatment methods have been implemented to remove TCS from the water such as using cellulose acetate (CA) membrane (Zhang et al, 2015), ammonia amendment and bioaugmentation in nitrifying activated sludge (Lee et al, 2015), dielectric barrier discharge plasma combined with activated carbon fibers (Xin et al, 2016) and ozonation (Orhon et al, 2017). However, some of these treatments involved complex procedures, high costs of treatments and maintenance, large volumes of chemicals and long processing times (Wang et al, 2013a).

In recent years, the adsorption process is one of the famous methods applied to remove chemicals and dyes in water and wastewater treatments, as compared to the other methods, due to its wide range of applications and less sludge being produced. Several adsorption studies to remove TCS were done by using rice straw-derived activated carbon (Liu et al, 2014), charcoal-based activated carbon (Behera at al., 2010), conventional activated carbon (Weinwr et al, 2017), civilian protective gas mask activated carbon (Sharipova et al, 2016), magnetic carbon composites from hydrochar (Zhu et al, 2014) and wastewater biosolids-derived biochar (Tong et al, 2016). High surface areas, micro-porous structures and high degrees of surface reactivity make activated carbons as versatile adsorbents, particularly effective for the adsorption of organic and inorganic pollutants from aqueous solutions (Pezoti et al, 2016; Sayğılı and Güzel, 2016).

But, the preparation for commercial activated carbons caused increased costs and this has encouraged researchers to search for low cost materials as alternatives (Wirasnita et al, 2014). The agricultural wastes are one of the promising sources as they are inexpensive, easy to collect and environmental friendly (Rahmat et al, 2016). Besides, they also have a high efficiency in trap and remove chemicals and dyes in water due to the availability of many functional groups, such as alcohols, phenolic, amido, amino, carboxyl, carbonyl and ester (Lazim et al, 2015a). The agricultural wastes from coconut trees are one of the promising materials to be used as adsorbents due to their abundance in nature, high porous structures and high adsorption capability. The coconut tree parts commonly used as adsorbents were the frond (Njoku et al, 2014), pulp waste (Kamari et al, 2014), husk (Dabwan et al, 2015), coir (Hettiarachchi et al, 2016), bunch (Rahmat et al, 2016), leaves (Jawad et al, 2016) and shell (Kaman et al, 2017).

Other than that, membrane is one of the technologies used for treatments of various chemicals and pollutants in water. It is an advanced treatment technology that is well known and it has become one of the preferred options for water and wastewater treatments, food industries, petrochemical industries, chemical industries and pharmaceutical industries (Salehi, 2014; Padaki et al, 2015; Zheng et al, 2015; Jafarinejad, 2017; Piacentini and Giorno, 2017; Mistry and Maubois, 2017). Membrane treatment is one of the most potential and favourable methods because it has many advantages, such as no addition of chemicals required, no secondary pollutants produced, low energy consumption, easy to handle, low operating and maintenance costs, easy to scale-up, high porous structure and high recovery and reusability (Xu et al, 2015; Conidi et al, 2016; Castro-Muñoz et al, 2016; Jasni et al, 2017b).

Most of the membranes are made with polymeric materials and they are available in various types of structures and properties. Some of the organic polymers used are polysulfone (PSF), polycarbonates (PC), polyethersulfone (PES), polyamide and polyimide (PI) (Topaloğlu, 2015). Meanwhile, nylon 6,6 is a polyamide polymer that is excellent in mechanical strength, toughness, rigidity and stability with self-lubricating properties and cost effective in nature (An et al, 2017; Jasni et al, 2017a). It is also hydrophilic, thinner, highly porous, highly permeant, better in fouling resistant and less complicated in structures (Huang and McCutcheon, 2014; Bilad et al, 2018). These advantages have promoted nylon as a functional polymer for many biomedical and environmental applications (Jasni et al, 2017a).

However, finding the best and the most affordable treatments for TCS, so that its long term effects to the aquatic life, wild life and human health can be prevented remains a concern for the researchers. Therefore, this research studied the efficiency of two methods, adsorption and filtration by using selected activated carbon and nylon 6,6 membrane respectively and the suitability of both adsorption and filtration methods combined together to remove TCS in water.

1.2 Problem Statement

In recent years, most of the people are getting more concerned about their health issues. This concern has encouraged them to use antibacterial-based products in their daily life, such as hand wash, shower cream, deodorant, toys, cosmetics, detergent and others. In urban areas, antimicrobial agents were detected in most of their wastewater treatment plants effluents, rivers and lakes (Elmekki, 2014; Halden, 2014). In Malaysia, TCS was detected in Lui River, Selangor with concentration at 20.80 ng/L (Praveena et al, 2018). Other than that, China also detected with TCS in their rivers with concentration 105.0 ng/L (Yang et al, 2018). The present of this pharmaceutcal substance is most likely due to the large pharmaceuticals production including over-the counter medications, prescription drug products, nutraceuticals, traditional medicines, and health supplements in all dosage forms (Hassali et al., 2009).Singer et al (2002) reported that, 79% of TCS was biologically degraded, 15% was adsorbed to the sludge and 6% escaped the wastewater treatment plant and flew out in the final effluent to the water sources. Although only a small number of TCS leaves the water and wastewater treatment plants, it can cause bioaccumulation and negatively affect human health and animals (Dhillon et al, 2015). Besides that, photodegradation and biodegradation processes take place to TCS in water and then produced chlorophenols, dioxins (such as 2,8-DCDD) and methyltriclosan (MTCS) that are very toxic and can kill aquatic life (Tohidi and Cai, 2017). In addition, over 75% of the US populations have been detected with TCS in their blood and urine (James et al, 2015).

There were many treatments that had been conducted to remove and reduce TCS from water sources, either by physical, chemical or biological methods. However, some of the treatments involve high maintenance costs, complex operation procedures and are time consuming, besides producing secondary chemicals (Wang et al, 2013a). The adsorption treatment is one of the methods that can remove TCS from water. The previous researches reported that, granular-activated carbon can remove 87 % of TCS (Katsigiannis et al, 2015), wastewater biosolids-derived biochar can remove 75 % of TCS (Tong et al, 2016) and gas mask activated carbon can remove 88 % of TCS (Sharipova et al, 2016) from the water. Even so, the conventional activated carbon productions are expensive. The efforts to reduce water treatment cost had encouraged the researchers to find cheaper materials to replace the conventional activated carbon.

The usage of agricultural wastes can help to reduce costs of adsorbent materials due to their low price and abundance in nature. However, only a few studies were done using agricultural wastes to remove TCS in water such as ricestraw (Liu et al, 2014) and stevia plant residue (Yokoyama et al, 2019). Agricultural waste consist of some basic elements such as lipids, lignin, cellulose, hemicellulose, hydrocarbons, extracts, proteins, starch, simple carbohydrates and water and also containing a variety of functional groups that present in the binding process and can sequestering of pollutant (Lazim et al, 2015a). Cellulose elements are significance in adsorption process since it has potential sorption capacity for various pollutants. Besides that, agricultural waste has many functional groups such as alcohols, phenolic, carboxyl, carbonyl, amino and esters that can help to form chemical bonding with pollutants (Lazim et al, 2015b). Besides that, in Malaysia, there are a lot agricultural wastes that did not fully utilized and disposed by the user and industries such as coconut waste, sugarcane waste, paddy field waste and so on. Some of the agricultural waste are abundance in nature and idle. Hence, some potential agricultural wastes were selected and studied in order to find new material that can remove TCS in water.

In addition, the membrane technology is used in many treatment processes especially for water and wastewater treatment. Membrane has a lot of functional group that can help to form bonding with the pollutants such as carbonyl groups, ether, ester, hydroxyl groups and so on. The hydrogen bonding between pollutants and carbonyl groups of membrane can help to increase pollutants removal in water. But, some of the pollutants are not completely removed by membrane technology due to some problems such as the tendency for fouling to occur during the treatment process. Xu et al (2014) reported that, the mesoporous nanofibers could remove 60 % to 72 % of TCS from the water. Hence, the combination of both activated carbon and membrane treatment to remove TCS in water was investigated in order to maximize the removal of TCS in water. Furthermore, there is a lack of studies that analyzed the combination of two treatment methods that apply both filtration and adsorption concept to treat TCS from the water. The lacking of previous study might be due to the function of the treatment as both of the treatments apply filtration concept but with different media

1.3 Research Objectives

The aim of this study was to investigate the adsorption of TCS from aqueous solution by combining the coconut pulp waste activated carbon with nylon 6,6 nanofiber. The objectives of this study are as follows:

1) To determine the potential agricultural wastes as adsorbent to remove TCS from water.
2) To investigate the effects of physico-chemical parameters for the TCS removal by using coconut pulp waste activated carbon and nylon 6,6 nanofiber.
3) To evaluate the adsorption kinetics and isotherms of TCS on coconut pulp waste activated carbon and nylon 6,6 nanofiber.
4) To determine the performance of combination method using coconut pulp waste activated carbon and nylon 6,6 nanofiber in removing TCS.
5) To study the physical and chemical characteristics of both activated carbon and nylon 6,6 nanofiber.

1.4 Scope of Study

This study was conducted by using activated carbon from coconut pulp waste and nylon 6,6 membrane. A total of eight types of agricultural wastes, namely, coconut frond, coconut husk, coconut pulp waste, dates seed, galangal stem, rambutan peel, orange peel and sugarcane bagasse were used for the screening process. The coconut pulp waste was selected as the best adsorbent to remove TCS in water due to its highest TCS removal within 24 hours. Subsequently, the selected material was prepared as activated carbon by using a horizontal furnace. For nylon 6,6 membrane, the membrane sheet was fabricated using an electrospinning machine.

The batch studies were done to investigate the effects of various parameters on the TCS removal for both activated carbon and membrane during the adsorption study. The parameters tested were adsorbent dosage, pH, temperature, contact time, agitation speed and initial TCS concentration. Besides, the filtration study was done for nylon 6,6 membrane by using a flat sheet membrane test machine. The compaction test, water flux and TCS flux were analyzed during this study. The TCS residues after being filtered through the permeation cell were collected and measured. The TCS residues from all of the experiments were read using UV-Vis spectrophotometer at wavelength (λ_{max}) 279 nm.

In this research, the TCS removal by the combination method using both filtration and adsorption processes was conducted under the conditions of the best parameters obtained from activated carbon and membrane batch studies. The experiments were carried out by using a flat sheet membrane test machine. The percentages of TCS removal using a single method from adsorption and filtration were compared with those obtained using the combination of both methods.

The surface morphology and functional groups of activated carbon and membrane before and after TCS removal were investigated using Fourier Transform Infrared Spectroscopy (FTIR) and Field Emission Scanning Electron Microscope (FESEM). For Brunauer-Emmett-Teller (BET) test, only activated carbon was analyzed. The surface areas of the raw coconut pulp waste, coconut pulp waste activated carbon and the activated carbon after TCS adsorption were studied. The BET test was not applied for membrane.

1.5 Significance of the Study

The significance of this research was to carry out the best treatment to remove TCS in water by a combination of adsorption and filtration methods using coconut pulp waste activated carbon and nylon 6,6 membrane, respectively. Even though there were many treatment methods studied to remove TCS, this combined method has not been studied yet by any researchers. Besides, this method can achieve a maximum TCS removal in high percentages, in less than 24 hours.

Malaysia is a country that produces a lot of agricultural products. The agricultural wastes produced are not fully utilized and they are often thrown to the landfill or left to decompose by natural processes. Hence, this study can help to give ideas for further usages of agricultural wastes. The coconut pulp waste is an abundant agricultural waste and it is normally thrown or used as animal feed. Moreover, the coconut pulp waste is not widely used as activated carbon. This research can help coconut milk industries to reduce their waste and encourage economic activities for agricultural industries. Therefore, this research can give benefits, both environmentally and economically to people in Malaysia.

1.6 Thesis Organization

This thesis consists of five main parts, beginning with chapter one and ending with chapter five, all being interconnected with each other. In chapter one, the discussions are focusing on the background of the study, problem statement, research objectives, scope of the study and the significance of this study. Meanwhile, in chapter two, all issues, facts and knowledge related to TCS, activated carbons and membranes are elaborated. Previous researches, articles, books and other resources were used as references. Following this, in chapter three, the focus is on the ways

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APPENDIX A

Adsorption Isotherms Calculations for both Activated Carbon and Membrane

ADSORPTION BY COCONUT PULP WASTE:

1. Langmuir Isotherm

From Figure 4.12, all the Langmuir constants were calculated as below;

y = mx + C

$\frac{C_e}{q_e} = \left(\frac{1}{Q_m}\right) C_e + \frac{1}{K_L Q_m}$ (a) Langmuir equation

Y = 0.0307 x + 0.1509 (b) Straight line equation from Langmuir graph

R^2 = **0.9714**

By comparison: (a) = (b)

$1 / Q_m = 0.0307$

$Q_m = 1/ 0.0307$

Q_m = **32.5731 mg/g**

$1 / (K_L Q_m) = 0.1509$

$1 = (0.1509) (K_L \times 32.5731)$

K_L = **0.2034 L/mg**

$R_L = 1 / (1 + (K_L C_o))$

R_L = 1 / (1 + (0.2034 L/mg x 90 mg/L))

R_L = **0.0518**

2. Freundlich Isotherm

From Figure 4.13, all the Feundlich constants were calculated as below;

$y = mx + C$

$\log q_e = \left(\frac{1}{n}\right) \log C_e + \log K_F$ (a) Freundlich equation

$Y = 0.5149\, x + 0.778$ (b) Straight line equation from Freundlich graph

$R^2 =$ **0.7726**

By comparison: (a) = (b)

$1 / n = 0.5149$

$n =$ **1.9421**

$\log K_F = 0.7780$

$K_F =$ **5.9980 (mg/g)(L/mg)$^{1/n}$**

3. Temkin Isotherm

From Figure 4.14, all the Temkin constants were calculated as below;

$y = mx + C$

$q_e = B(\ln C_e) + B(\ln A)$ (a) Temkin equation

$y = 7.1441x + 5.5208$ (b) Straight line equation from Temkin graph

$R^2 =$ **0.9124**

By comparison: (a) = (b)

B = **<u>7.1441 J/mol</u>**

B (ln A) = 5.5208

A = **<u>2.1658 L/g</u>**

ADSORPTION BY NYLON 6,6 MEMBRANE:

1. Langmuir Isotherm

From Figure 4.24, all the Langmuir constants were calculated as below;

y = mx + C

$$\frac{C_e}{q_e} = \left(\frac{1}{Q_m}\right) C_e + \frac{1}{K_L Q_m}$$ (a) Langmuir equation

Y = - 0.0350 x + 0.3061 (b) Straight line equation from Langmuir graph

R^2 = **<u>0.6605</u>**

By comparison: (a) = (b)

$1 / Q_m$ = - 0.0350

$- Q_m$ = 1/ (- 0.0350)

Q_m = **<u>28.5714 mg/g</u>**

$1 / (K_L Q_m)$ = 0.3061

1 = (0.3061) (K_L x 28.5714)

K_L = **<u>0.1143 L/mg</u>**

$R_L = 1 / (1 + (K_L C_o))$

$R_L = 1 / (1 + (0.1143 \text{ L/mg} \times 90 \text{ mg/L}))$

$R_L =$ **0.0886**

2. Freundlich Isotherm

From Figure 4.25, all the Feundlich constants were calculated as below;

y = mx + C

$\log q_e = \left(\frac{1}{n}\right) \log C_e + \log K_F$ (a) Freundlich equation

Y = 1.4528 x + 0.5569 (b) Straight line equation from Freundlich graph

$R^2 =$ **0.9821**

By comparison: (a) = (b)

1 / n = 1.4528

n = **0.6883**

$\log K_F = 0.5569$

$K_F =$ **3.6050 (mg/g)(L/mg)$^{1/n}$**

3. Temkin Isotherm

From Figure 4.26, all the Temkin constants were calculated as below;

$y = mx + C$

$q_e = B(\ln C_e) + B(\ln A)$ (a) Temkin equation

$y = 18.8060\,x + 3.4592$ (b) Straight line equation from Temkin graph

R^2 = **0.9159**

By comparison: (a) = (b)

B = **18.8060 J/mol**

B (ln A) = 3.4592

A = **1.2019 L/g**

APPENDIX B

Adsorption Kinetics Calculations for both Activated Carbon and Membrane

ADSORPTION BY COCONUT PULP WASTE:

1. Pseudo-First-Order

From Figure 4.15, all the pseudo-first-order constants were calculated as below;

$y = mx + C$

$\ln(q_e - q_t) = -k_1 t + \ln q_e$ (a) Pseudo-first-order equation

$y = -0.1416x + 0.1357$ (b) Straight line equation from pseudo-first-order graph

$R^2 =$ **0.7224**

By comparison: (a) = (b)

$k_1 =$ **0.1416 mg/g min**

$\ln q_e = 0.1357$

$q_e =$ **1.1453 mg/g**

2. Pseudo-Second-Order

From Figure 4.16, all the pseudo-second-order constants were calculated as below;

$y = mx + C$

$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}$ (a) Pseudo-second-order equation

$y = 0.4528x + 0.8045$ (b) Straight line equation from pseudo-second-order graph

R^2 = **0.9880**

By comparison: (a) = (b)

$1 / q_e = 0.4528$

q_e = **2.2085 mg/g**

$1 / (k_2 q_e^2) = 0.1357$

k_2 = **0.02549 mg/g min**

3. Intraparticle Diffusion

From Figure 4.17, all the intra-particle diffusion constants were calculated as below;

$y = mx + C$

$q_t = k_{dif} t^{½} + C$ (a) Intra-particle diffusion equation

$y = 0.4201x + 0.2277$ (b) Straight line equation from intra-particle diffusion graph

R^2 = **0.8562**

By comparison: (a) = (b)

k_{dif} = **0.4201 mg/g min$^{1/2}$**

C = **0.2277 mg/g**

ADSORPTION BY NYLON 6,6 MEMBRANE:

1. Pseudo-First-Order

From Figure 4.27, all the pseudo-first-order constants were calculated as below;

$y = mx + C$

$\ln(q_e - q_t) = -k_1 t + \ln q_e$ (a) Pseudo-first-order equation

$y = -0.5775x + 0.0872$ (b) Straight line equation from pseudo-first-order graph

$R^2 =$ **0.5158**

By comparison: (a) = (b)

$k_1 =$ **0.5775 mg/g min**

$\ln q_e = 0.0872$

$q_e =$ **1.0911 mg/g**

2. Pseudo-Second-Order

From Figure 4.28, all the pseudo-second-order constants were calculated as below;

$y = mx + C$

$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}$ (a) Pseudo-second-order equation

$y = 0.4531x + 0.0763$ (b) Straight line equation from pseudo-second-order graph

$R^2 =$ **0.9981**

By comparison: (a) = (b)

$1 / q_e = 0.4531$

$q_e =$ **2.2070 mg/g**

$1 / (k_2 q_e^2) = 0.0763$

$k_2 =$ **2.6907 mg/g min**

3. Intraparticle Diffusion

From Figure 4.29, all the intra-particle diffusion constants were calculated as below;

$y = mx + C$	
$q_t = k_{dif} t^{½} + C$	(a) Intra-particle diffusion equation
$y = 0.8597 x + 0.5466$	(b) Straight line equation from intra-particle diffusion graph
R^2 = **0.8236**	

By comparison: (a) = (b)

k_{dif} = **0.8597 mg/g $min^{1/2}$**

C = **0.5466 mg/g**

LIST OF PUBLICATIONS

Journal with Impact Factor

1) Mohd Khori, N. K. E., Hadibarata, T., Elshikh, M. S., Al-Ghamdi, A. A., Salmiati, S & Yusop, Z. (2018). Triclosan removal by adsorption using activated carbon derived from waste biomass: Isotherms and kinetic studies. *Journal of the Chinese Chemical Society*, *65*(8), 951-959.