TREATMENT OF ACETAMINOPHEN AND ACETYLSALICYLIC ACID BY CHICKEN BONE BASED ADSORBENT

NURSHUHADA AMIRA BINTI YUSOFF

A thesis submitted in fulfilment of the
requirements for the award of the degree of
Master of Philosophy

School of Chemical and Energy Engineering
Faculty of Engineering
Universiti Teknologi Malaysia

JULY 2019

DEDICATION

Specially for

My beloved father; Yusoff bin Ibrahim

My beloved mother; Rosnani binti Shaari

My dearest siblings,

And my forever husband; Zakaria bin Ismail

ACKNOWLEDGEMENT

I wish to express my deepest appreciation to all those who helped me, in one way or another, to complete this project. First and foremost I thank God Almighty who provided me with strength, direction and purpose throughout the project. Special thanks to my project supervisor Associate Professor Dr. Norzita binti Ngadi for all her patience, guidance and support during the execution of this project. Through her expert guidance, I was able to overcome all the obstacles that I encountered during my journey to complete this research. I could not have imagined having a better advisor and mentor who always gave me immense hope every time I consulted with her over problems relating to my project.

ABSTRACT

The presence of trace amounts of pharmaceutical waste in the environment has become a concern among the environmental community and scientist. Paracetamol (acetaminophen, ACT) and aspirin (acetylsalicylic acid, ASA) are the most common pharmaceutical waste that have been detected in wastewater and surface water throughout the world. In recent years, researchers have begun to explore the potential of organic waste as an adsorbent in adsorption process to remove the pollutant from wastewater. In this study, activated carbon derived from chicken bone waste was synthesized and characterized to compare with present organic waste adsorbent. The prepared adsorbent; bone char and activated carbon, were used to remove ACT and ASA from aqueous solution. Finally, the effectiveness of the adsorbent prepared to remove ACT and ASA was studied. The chicken bone was collected from a food stall, dried and heated up to 500 ºC in the furnace prior to being activated by phosphoric acid. The surface properties of adsorbents were characterized using Fourier transform infrared spectrophotometer, scanning electron microscopy and Brunauer-Emmett-Teller. The adsorption process was carried out in batch mode affected by different parameters; contact time (60-300 minutes), dosage (0.05-0.2 g), pH (2-11), initial concentration (1,000-5,000 mg/L) and temperature (25-70 ºC). The highest removal percentage was ACT (94.7 %) followed by ASA (92.78 %). Both pharmaceuticals waste were efficiently removed using activated carbon chicken bone compared to bone char. The experimental isotherm and kinetic data were evaluated using Langmuir and Freundlich isotherm models and pseudo-first-order, pseudo-second-order and intra-particle diffusion kinetics models. The experimental data obey the Freundlich isotherm model and pseudo-second-order kinetic model. In a thermodynamic study, the adsorption of ACT occurred spontaneously with the exothermic reaction while the adsorption of ASA occurred with the endothermic reaction. The high percentage removal of ACT (94.7 %) and ASA (92.78 %) using chicken bone based on activated carbon confirmed that chicken bone can be potentially used as an alternative adsorbent for pharmaceutical waste ACT and ASA.

ABSTRAK

Kehadiran sejumlah sisa farmaseutikal dalam persekitaran telah mendapat perhatian di kalangan ahli alam sekitar dan saintis. Parasetamol (asetaminophen, ACT) dan aspirin (asid asetailsalisailik, ASA) adalah sisa farmaseutikal yang biasa dikesan di dalam air kumbahan dan pada permukaan air di seluruh dunia. Kebelakangan ini, penyelidik telah mula untuk menerokai potensi sisa organik sebagai bahan penjerap dalam proses penjerapan untuk membuang bahan pencemaran tersebut daripada air kumbahan. Dalam kajian ini, karbon aktif dihasilkan daripada sisa tulang ayam telah disintesis dan dicirikan bagi membuat perbandingan dengan penjerapan sisa organik yang sedia ada. Penjerap yang disediakan iaitu arang tulang dan karbon aktif telah digunakan untuk penyingkiran ACT dan ASA daripada larautan akueus. Akhir sekali, keberkesanan penjerap yang disediakan untuk menyingkirkan ACT dan ASA telah dikaji. Tulang ayam dikumpul dari gerai makanan, dikeringkan dan dipanaskan sehingga suhu 500 °C dalam relau dan kemudian diaktifkan dengan menggunakan asid fosforik. Sifat-sifat permukaan bahan penjerap telah dicirikan dengan menggunakan spektroskopi transformasi inframerah Fourier, mikroskop imbasan elektron dan Brunauer-Emmett-Teller. Proses penjerapan telah dijalankan dalam mod kelompok yang dipengaruhi oleh beberapa pemboleh ubah yang berbeza seperti masa tindakbalas (60-300 minit), dos (0.05-0.2 g), pH (2-11), kepekatan awal (1,000-5,000 mg/L) dan suhu (25-70 °C). Peratus penyingkiran tertinggi ialah ACT (94.7%), diikuti oleh ASA (92.78%). Kedua-dua bahan farmaseutikal berkesan disingkirkan dengan menggunakan karbon teraktif tulang ayam berbanding arang tulang. Ujikaji isotem dan data kinetik telah dinilai menggunakan model isotem Langmuir dan model isotem Freundlich dan model kinetik pseudo-tertib-pertama, pseudo-tertib-kedua dan model kinetik penyebaran intra-zarah. Data ujikaji mematuhi kepada model isoterm Freundlich dan model kinetik pseudo-tertib-kedua. Dalam kajian termodinamik, penjerapan ACT berlaku secara spontan dengan tindak balas eksotermik manakala penjerapan ASA berlaku dengan tindak balas endotermik. Peratusan tinggi penyingkiran ACT (94.7%) dan ASA (92.78%) menggunakan karbon teraktif berasaskan tulang ayam mengesahkan bahawa tulang ayam berpotensi digunakan sebagai penjerap alternatif untuk menyingkirkan sisa farmaseutikal ACT dan ASA.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

ACMF	-	Australian Chicken Meat Federation
AC	-	Activated Carbon
ACT	-	Acetaminophen
ASA	-	Acetylsalicylic Acid
BC	-	Bone Char
BET	-	Brunauer Emmett Teller
BOD	-	Biological Oxygen Demand
CB	-	Chicken Bone
DoD	-	Department of Defenses
DBPs	-	Disinfection by-products
EPA	-	Environmental Protection Agency
EDCs	-	Endocrine disrupting chemical
FTIR	-	Fourier Transform Infrared
NSAIDs	-	Non-steroidal anti-inflammatory drugs
PAC	-	Powder activated carbon
PPCPs	-	Personal Care Products and Pharmaceuticals
POPs	-	Persistent organic pollutants
SEM	-	Scanning Electron Microscope
USGS	-	U. S. Geological Survey
UV	-	Ultra violet
WWTP	-	Wastewater treatment plant

LIST OF SYMBOLS

Δ	-	Delta
>	-	More than
<	-	Less than
%	-	Percent
β	-	Beta
T	-	Temperature
d	-	Diameter
t	-	Time
rpm	-	Revolution per minute
R^2	-	Coefficient correlation
R	-	Universal gas constant
min	-	Minutes
ΔG	-	Gibbs Free Energy
ΔH	-	Enthalpy
ΔS	-	Entropy

CHAPTER 1

INTRODUCTION

1.1 Background of Study

In a world with advanced technology, a very low concentration of new emerging pollutants has been detected by scientists around the world using new and advanced analytical instruments with several techniques. Emerging contaminant is any synthetic or naturally occurring chemical or any microorganisms that are not commonly monitored in the environment but they have the potential to enter the environment and cause known or suspected adverse ecological and human health (Fawell and Ong, 2012). There are numerous types of emerging contaminants including human and veterinary pharmaceutical, personal care products, pesticides, endocrine disruptive compound, and micro-constituents. These emerging contaminants may enter the water environment through various routes including the agricultural, industrial and urban channel. Various ranges of the emergent pollutants have been detected in the water environment at low concentration that is commonly in nanogram per liter range. This low concentration of emerging pollutants in natural water and drinking water may not cause an immediate fatal effect to humans but it may lead to a dangerous impact on human health in the long term.

Pharmaceuticals and Personal Care Products (PPCPs) is one of the most common emergent pollutants present in wastewater and drinking water. Several research studies showed that over 30 mg/L of pharmaceutical waste was discharged into rivers and lakes close to industrial outlets (Fawell and Ong, 2012). Paracetamol (acetaminophen) and aspirin (acetylsalicylic acid) are most widely used in our society as analgesic and anti-inflammatory drugs which are pain-controlling and fever-reducing. The presence of both drugs in the environment especially in wastewater and drinking water can cause ecological effects because their continuous introduction into the environment can lead to overwhelming transformation rates.

Several research studies showed that acetaminophen and acetylsalicylic acid were known to be present as high as nanogram per liter. It was found that acetaminophen had the highest concentration detected in Langat River, Malaysia, which was 350.3 ng/L in correspondence to the concentration of acetaminophen detected in Spain which was 250 ng/L and exceeded 1 µg/L concentration of acetaminophen detected in the UK (Al-Odaini *et a*l., 2013).

Removal of PPCPs from wastewater and drinking water has become a challenge among the community since there is no effective method to remove a variety of range of PPCPs from the water. It had been reported that conventional wastewater treatment plant was not adequate to eliminate PPCPs residues from the water. Several research studies showed that coagulation and filtration in conventional wastewater treatment processes were not the appropriate methods to eliminate PPCPs while disinfection process had been reported to only be able to remove some PPCPs (Shi *et al*., 2013). Typically, some treatments have been implemented to eliminate PPCPs including adsorption, advance oxidation process, nano-filtration and reverse osmosis membrane but some disadvantages have arisen from these methods including high investment and maintenance cost, the formation of secondary pollutant, and complex operation procedure. For example, the removal of PPCPs by advanced oxidation process can cause the formation of oxidation by-product (Grassi *et al*., 2012). However, the adsorption process has shown good potential in the removal of the emerging pollutants since its operation does not form any undesirable by-products.

The adsorption process is the most favorable technique in the removal of pollutants in wastewater treatment compared to other techniques in terms of simplicity of design and operation; its operation does not produce undesired by-product. There are many adsorbents which can be used including soils, clays, silica, and hydrous oxides, but activated carbon has been chosen as an adsorbent to adsorb the low concentration of pollutants due to their unique characteristics which contain high surface area and highly-developed porous network making them able to react with other heteroatoms and creating a variety of surface functionalities on its surfaces. The greatest challenge in the implementation of activated carbon is their high production cost for a large industrial scale. Hence, several research studies were made to produce activated carbon by using waste materials as the precursor such as coffee residues (Laksaci *et al*., 2017), rice hulls (Mukoko *et a*l., 2015), fruit seeds and shells (Verla *et al*., 2012), wood (Chojnacka and Michalak, 2009), corn cob (Okeola and Odebunmi, 2010), bamboo (Liu *et al*., 2010) and animal bone (Djilani *et al*., 2016).

In 1990s, the global demand for meat reported by the Australian Chicken Meat Federation (ACMF) was 173 million tonnes, of which poultry made up 23%. However, the annual global demand for meat increased to 285 million tonnes in 2010, with poultry now comprising of 35% or 100 million tonnes of this (Mcdonald, 2013). In Poultry Trend 2018, world poultry production are nearly at 123 million tons, lead United State, China and Brazil (Poultry Trend, 2018). In Malaysia, the production and consumption of poultry showed an increasing pattern from 1960 to 2014 and it is expected to increase annually (Nor and Rosali, 2015). In a study conducted (Samsuddin *et al*., 2015), the total number of chicken meat production increase from 217, 227, 467 in 2010 to 268, 242 584 in 2012. The study showed that the demand for chicken was higher throughout the year. The increase of poultry production has contributed to the increase of poultry waste products such as chicken bone wastes. Chicken bone wastes are as abundant as food and organic wastes in municipal solid wastes which are fulfilling the landfill. The abundance of chicken bone wastes in the landfills are attributed by everyday consumption of chicken in human food. Since chicken bone is abundantly available and priceless, it is an advantage to use it as raw material to produce useful products.

1.2 Problem Statement

Pharmaceuticals are one of the emerging contaminants that have been widely used around the world with an important role in the treatment and prevention of diseases in both human and animal. Paracetamol (acetaminophen, ACT) and aspirin (acetylsalicylic acid, ASA) are known as the most widely used drugs around the world to reduce fever and pain. These drugs will be excreted by the body after the administration and absorption by the body to the sewage system via urine and feces and reach the wastewater treatment plant. The occurrence of these drugs in the environment has contributed to adverse effects, including acute and chronic damage, accumulation in tissues, reproductive damage, inhibition of cell proliferation, and behavioral changes although their occurrences have been detected in low concentration (Wu *et al*., 2012).

Questions have risen among the community concerning the effective treatment process to remove this low concentration of pharmaceuticals from drinking water and wastewater. As reported in the previous study, advanced oxidation process has been widely used in the industry to remove all these pollutants (Grassi *et al*., 2012). However, the drawbacks of this method, including the harsh reaction conditions, the formation of secondary pollutants and the high operational cost have made it not a favorable method to remove these pollutants. In the case of pharmaceuticals, conventional wastewater treatment has been demonstrated to not be effective to eliminate and/or degrade the majority of these compounds and they are only partially removed, therefore residual quantities remain in the treated water and have been found to accumulate in drinking water.

The adsorption process is the most preferable method by the industry being activated carbon as the adsorbent to remove the emerging pollutants. A review study has been made by a researcher regarding new approaches to remove pharmaceuticals from wastewater. The study states that the adsorption method is the most efficient method to remove pharmaceutical waste since it has a simple design, cost-effective and unaffected by toxicity (Kyzas *et al*., 2015). However, the utilization of activated carbon on large scale industrial processes in the adsorption process is limited because of the high cost in the manufacturing of activated carbon. Consequently, many industrials take other alternatives by replacing the activated carbons with cost-effective adsorbents such as sepiolites and diatomites even though their removal efficiency is much lower than activated carbon.

The economic cornerstone for the use of activated carbon in advanced water treatments based on adsorption process can be overcome by replacing the raw materials of the activated carbon with non-expensive materials to lower the production cost of activated carbon. In this sense, synthesis of solid waste materials as a precursor of adsorbent as an attractive approach in sustainable management and economic availability on the utilization of residues as precursors of carbon adsorbent is an interesting strategy that enables researcher to deal with problems regarding solid waste disposal and recycling since its production extremely increases each year due to the increase of the Malaysian population.

Food and organic waste is the main component of municipal solid waste in Malaysia comprising approximately half of the total municipal solid waste (Johari *et al.*, 2014). The utilization of chicken bones as the precursors of activated carbon preparation is able to overcome the cost of activated carbon manufacturing to reduce the everyday waste disposal since it comprises of food waste. In Australia, an average of 50 kg of chicken was consumed by each person per year (Znad and Frangeskides, 2014) while the trend of chicken consumption in Malaysia was seen to be increasing from 36 kg per capita in 2010 to 39 kg per capita in 2011 (Jayaraman *et al.*, 2013) and poultry production and consumption was estimated to increase by 24 % in 2013. The increase of poultry production and consumption has contributed to the abundance of chicken waste and as a result, chicken bones can be potentially used in this application. Apart from that, a previous study states that the abundance of animal wastes can cause environmental impacts on water and air pollution due to the methane emissions which contribute to global warming (Whitely *et al.*, 2006). The utilization of chicken bones is the best way to reduce the cost of activated carbon manufacturing which consequently reduces the municipal waste disposal issue. Apart from that, any study on the chicken bone as activated carbon has not been reported especially as an adsorbent in pharmaceutical removal.

1.3 Objectives of the Research

The objectives of this study are as follows:

(a) To synthesize and characterize activated carbon derived from chicken bone adsorbent to compare with present organic based adsorbent

(b) To evaluate the effectiveness of chicken bone charcoal and chicken bone-derived activated carbon on acetaminophen and acetylsalicylic acid removal

(c) To study the adsorption kinetics, adsorption isotherm and adsorption thermodynamic of chicken bone-derived activated carbon on the removal of acetaminophen and acetylsalicylic acid from an aqueous solution

1.4 Scope of Study

Activated carbon was prepared by two processes; (i) carbonization of raw materials and (ii) chemical activation by using phosphoric acid (H_3PO_4). Firstly, the raw material, chicken bones, were carbonized at 500 °C for one hour to produce bone charcoal. After that, the bone charcoal in the form of powder was impregnated by 0.5 M H_3PO_4. Finally, the activated carbon and the bone charcoal produced were characterized using Fourier Transform Infrared Spectrophotometer (FTIR) to identify the functional group present, scanning electron microscopy (SEM) analysis to identify the surface morphology, and Brunauer-Emmett-Teller (BET) theory to determine the specific surface area.

The effectiveness of chicken bone based activated carbon in removing acetaminophen and acetylsalicylic acid was conducted by batch adsorption process. The effect of several experimental parameters such as contact time (60-300 minutes), pH (2-11), activated carbon dosage (0.05-0.2 g), initial concentration (1000-5000 mg/L), and temperature (25-70 °C), was studied on the removal of acetaminophen and acetylsalicylic acid. The effectiveness of chicken bone based activated carbon was determined based on the percentage removal of pharmaceutical from the aqueous solution. The experiment was repeated with bone char using the same parameter to compare the effectiveness of bone char (BC) and chicken bone based activated carbon (AC).

Adsorption kinetic study of bone char and chicken bone-derived activated carbon was conducted by fitting the equilibrium data to the pseudo-first-order equation, pseudo-second-order-equation, and intra-particle diffusion equation. Two adsorption isotherm models were fitted to the equilibrium data which are Langmuir isotherm model and Freundlich isotherm model. Finally, the adsorption thermodynamic study was conducted by fitting the data of temperature effect to the thermodynamic equation to determine the Gibbs free energy (ΔG), enthalpy (ΔH) and entropy (ΔS) of the adsorption process.

1.5 Significant of Study

This study will be significant in wastewater treatment especially in adsorption process due to its high efficiency in the removal of all types of pollutants. This study is also beneficial to the industry in the manufacturing of large scale activated carbon with low manufacturing cost. The use of chicken bones as the raw material in the activated carbon manufacturing will also be significant to solid disposal management. It will also serve as a reference for new researchers to find other raw materials in the activated carbon manufacturing.

1.6 Thesis Organization

This thesis consists of 5 chapters and each chapter represents the sequence of this research as described below.

Chapter 1 describes the overview of pharmaceutical wastes in wastewater and common method used to remove the pharmaceutical wastes. This chapter consists of the problem statement, the objectives of research, the scope, and the significance of the research.

Chapter 2 covers the general and scientific knowledge on pharmaceutical wastes, acetaminophen, acetylsalicylic acid, activated carbon, chicken bones, and available treatment methods to remove the pharmaceutical wastes. This chapter also covers the previous study on the removal of pharmaceutical wastes using different types of activated carbon.

Chapter 3 consists of the methodology of research involving the experimental procedure and the characterization method. This chapter describes the experimental procedure to produce the adsorbent, evaluation of the adsorbent performance, and characterization of the adsorbent used.

Chapter 4 covers the results obtained from this study based on the experimental parameter tested. This chapter covers the results of adsorbent performance on the removal of the pharmaceutical wastes by adsorption process affected by several experimental parameters including contact time, pH of the solution, adsorbent dosage, initial concentration, and temperature.

Finally, chapter 5 covers the conclusion of this study. In this chapter, the conclusion on the performance of adsorbent and optimum condition for pharmaceutical removal were identified.

1.7 Summary

Pharmaceutical wastes have been detected in wastewater, groundwater, surface water, and drinking water at low concentration. The concentration has been detected as low as nano to microgram per liter which may not cause an immediate fatal effect to humans but it may lead to a dangerous impact on humans in the long term. Conventional wastewater treatment like sewage treatment which uses physical, chemical and biological treatment is not adequate to remove the pharmaceutical wastes. The adsorption process is the most effective treatment to eliminate pharmaceutical wastes from the environment. In the adsorption process, the pollutants will be adsorbed to the adsorbent called activated carbon. Activated carbon can be produced from several raw materials such as chicken bones. Therefore, in this study, chicken bones were used as the raw material to produce activated carbon. The performance of the activated carbon produced was evaluated in the adsorption process to remove acetaminophen and acetylsalicylic acid from aqueous solution.

REFERENCES

Abbaszadeh, S., Wan Alwi, S. R., Webb, C., Ghasemi, N., & Muhamad, I. I. (2016). Treatment of Lead-contaminated Water using Activated Carbon Adsorbent from Locally Available Papaya Peel Biowaste. Journal of Cleaner Production, 118, 210–222.

Abdel-raouf, N. (2012). Microalgae and Wastewater Treatment. Saudi Journal of Biological Sciences, 19(3), 257–275.

Abdullah, A. H., Kassim, A., Zainal, Z., Hussien, M. Z., Kuang, D., Ahmad, F., & Sim Wooi, O. (2001). Preparation and Characterization of Activated Carbon from Gelam Wood Bark (Melaleuca cajuputi). Malaysian Journal of Analytical Sciences, Vol. 7(No. 1), 65–68.

Abechi, S. E., Gimba, C. E., Uzairu, a, & Dallatu, Y. a. (2013). Preparation and Characterization of Activated Carbon from Palm Kernel Shell by Chemical Activation. Research Journal of Chemical Sciences, 3(7), 54–61.

Al-Khateeb, L. A., Almotiry, S., & Salam, M. A. (2014). Adsorption of Pharmaceutical Pollutants onto Graphene Nanoplatelets. Chemical Engineering Journal, 248, 191–199.

Ali, I., Asim, M., & Khan, T. A. (2012). Low Cost Adsorbents for the Removal of Organic Pollutants from Wastewater. Journal of Environmental Management, 113, 170–183.

Ansari, R. (2009). Activated Charcoal :Preparation, Characterization and Applications : A Review Article. International Journal of ChemTech Research, 1(4), 859–864.

Baccar, R., Sarrà, M., Bouzid, J., Feki, M., & Blánquez, P. (2012). Removal of pharmaceutical com- pounds by activated carbon prepared from agricultural by-product. Chem. Eng. J., 211-212(2012), 310–317.

Ben-Ali, S., Jaouali, I., Souissi-Najar, S., & Ouederni, A. (2016). Characterization and Adsorption Capacity of Raw Pomegranate Peel Biosorbent for Copper Removal. Journal of Cleaner Production, 142, 3809–3821.

Beninati, S., Semeraro, D., & Mastragostino, M. (2008). Adsorption of Paracetamol and Acetylsalicylic Acid onto Commercial Activated Carbons. Adsorption Science & Technology, 26(9), 721–734.

Bolong, N., Ismail, A. F., Salim, M. R. and Matsuura, T. (2009) 'A review of the effects of emerging contaminants in wastewater and options for their removal', Desalination. Elsevier B.V., 239(1–3), pp. 229–246. Boxall, A. B. a. (2004). The environmental side effects of medication. EMBO reports (Vol. 5). UK.

Bui, T. X., & Choi, H. (2009). Adsorptive Removal of Selected Pharmaceuticals by Mesoporous Silica SBA-15. Journal of Hazardous Materials, 168(2–3), 602–608.

Cantuaria, M. L., de Almeida Neto, A. F., Nascimento, E. S., & Vieira, M. G. A. (2016). Adsorption of Silver from Aqueous Solution onto Pre-treated Bentonite Clay: Complete Batch System Evaluation. Journal of Cleaner Production, 112, 1112–1121.

Cecen, F. (2014). Activated Carbon. Kirk-Othmer Encyclopedia of Chemical Technology. John Wiley & Sons, Inc.

Cheung, W. H., Szeto, Y. S., & McKay, G. (2007). Intraparticle Diffusion Processes During Acid Dye Adsorption onto Chitosan. Bioresource Technology, 98(15), 2897–2904.

Chojnacka, K. and Michalak, I. (2009) 'Using Wood and Bone Ash to Remove Metal Ions from Solutions', Global Nest Journal, 11(2), pp. 205–217

Chowdhury, Z. Z., Hamid, S. B. A., Das, R., Hasan, M. R., Zain, S. M., Khalid, K., & Uddin, M. N. (2013). Preparation of Carbonaceous Adsorbents from Lignocellulosic Biomass and Their use in Removal of Contaminants from Aqueous Solution. BioResources, 8(4), 6523–6555.

Dada, O, A., Olalekan, P, A., Olatunya, M, A.O. (2012). Langmuir, Freundlich, Temkin and Dubinin–Radushkevich Isotherms Studies of Equilibrium Sorption of Zn 2+ Unto Phosphoric Acid Modified Rice Husk. IOSR Journal of Applied Chemistry, 3(1), 38–45.

Demiral, H., & Güngör, C. (2015). Adsorption of Copper(II) from Aqueous Solutions on Activated Carbon Prepared from Grape Bagasse. Journal of Cleaner Production, 124, 103–113.

Deng, H., Yang, L., Tao, G., & Dai, J. (2009). Preparation and Characterization of Activated Carbon from Cotton Stalk by Microwave Assisted Chemical Activation — Application in Methylene Blue Adsorption from Aqueous Solution, 166, 1514–1521.

Desta, M. B. (2013). Batch Sorption Experiments: Langmuir and Freundlich Isotherm Studies for the Adsorption of Textile Metal Ions onto Teff Straw (Eragrostis tef) Agricultural Waste. Journal of Thermodynamics, 1(1).

Djilani, C., Zaghdoudi, R., Djazi, F., Bouchekima, B., Lallam, A., & Magri, P. (2016). Preparation and Characterisation of Activated Carbon from Animal Bones and its Application for Removal of Organic Micropollutants from Aqueous Solution. Desalination and Water Treatment, 57(52), 25070–25079.

Dutta, M., Das, U., Mondal, S., Bhattachriya, S., Khatun, R., & Bagal, R. (2015). Adsorption of Acetaminophen by using Tea Waste Derived Activated Carbon. International Journal of Environmental Sciences, 6(2), 270–281.

El Haddad, M., Mamouni, R., Saffaj, N., & Lazar, S. (2012). Removal of a cationic dye - Basic Red 12 - from aqueous solution by adsorption onto animal bone meal. Journal of the Association of Arab Universities for Basic and Applied Sciences, 12(1), 48–54.

El Haddad, M., Slimani, R., Mamouni, R., ElAntri, S., & Lazar, S. (2013). Removal of Two Textile Dyes from Aqueous Solutions onto Calcined Bones. Journal of the Association of Arab Universities for Basic and Applied Sciences, 14(1), 51–59.

ElShafei, G. M. S., ElSherbiny, I. M. A., Darwish, A. S., & Philip, C. A. (2014). Silkworms' feces-based activated carbons as cheap adsorbents for removal of cadmium and methylene blue from aqueous solutions. Chemical Engineering Research and Design, 92(3), 461–470.

Fawell, J., & Ong, C. N. (2012). Emerging Contaminants and the Implications for Drinking Water. International Journal of Water Resources Development, 28(March 2013), 247–263.

Ferreira, R. C., Junior, O. M. C., Carvalho, K. Q., Arroyo, P. A., & Barros, M. A. S. D. (2015). Effect of Solution pH on the Removal of Paracetamol by Activated Carbon of Dende Coconut Mesocarp. Chemical and Biochemical Engineering Quarterly Journal, 29(1), 47–53.

Ferreira, R. C., Lima, H. H. C. De, Cândido, A. A., Junior, O. M. C., Arroyo, P. A., & Carvalho, K. Q. De. (2015). Adsorption of Paracetamol Using Activated Carbon of Dende and Babassu Coconut Mesocarp. International Journal of Biological, Biomolecular, Agricultural, Food and Biotechnological Engineering, 9(7), 674–679.

Galhetas, M., Andrade, M. A., Mestre, A. S., Kangni-foli, E., Villa de Brito, M. J., Pinto, M. L., Carvalho, A. P. (2015). The Influence of the Textural Properties of Activated Carbons on Acetaminophen Adsorption at Different Temperatures. Physical Chemistry Chemical Physics : PCCP, 17(18), 12340–12349.

Ghaemi, A., & Tavakkoli, H. (2013). Full Length Research Paper Adsorption Behavior Investigation of Malachite Green Dye onto Animal Bone Meal ; Kinetic and Isotherm Studies. International Journal of Scientific Research in Knowledge, 1(11), 484–492.

Ghiaci, M., Dorostkar, N., & Gil, A. (2014). Chicken Bone Ash as an Efficient Metal Biosorbent for Cadmium, Lead, Nickel, and Zinc from Aqueous Solutions. Desalination and Water Treatment, 52(16–18), 3115–3121.

Grassi, M., Kaykioglu, G., Belgiorno, V., & Lofrano, G. (2012). Removal of Emerging Contaminants from Water and Wastewater by Adsorption Process. In G. Lofrano (Ed.), Emerging Compounds Removal from Wastewater (pp. 15–37). Springer Netherlands.

Gros, M., Petrović, M., Ginebreda, A., & Barceló, D. (2010). Removal of Pharmaceuticals During Wastewater Treatment and Environmental Risk Assessment Using Hazard Indexes. Environment International.

Gunasekara, S. N. (2011). Improvements on Municipal Wastewater Treatment by : Chemical Pre-Precipitation with Ca2+ & Mg2+ and Acid Hydrolysis of Toilet Paper. Industrial Ecology, Royal Institute of Technology.

Gupta, V. K., Ali, I., Saleh, T. A., & Agarwal, S. (2012). Chemical Treatment Technologies for Wastewater Recycling: An Overview, 6380–6388.

Hesas, R. H., Arami-Niya, A., Wan Daud, W. M. A., & Sahu, J. N. (2013). Preparation and Characterization of Activated Carbon from Apple Waste by Microwave-assisted Phosphoric Acid Activation: Application in Methylene Blue Adsorption. BioResources, 8(2), 2950–2966.

Ho, Y. S. (2006). Review of Second-Order Models for Adsorption Systems. Journal of Hazardous Materials, 136(3), 681–689.

Hu, Z., Srinivasan, M. P., & Ni, Y. (2000). Preparation of Mesoporous High-Surface-Area Activated Carbon. Adv Mater, 12(1), 62–65.

Hu, Z., Srinivasan, M. P., & Ni, Y. (2001). Novel Activation Process for Preparing Highly Microporous and Mesoporous Activated Carbons. Carbon, 39(6), 877–886.

Iriarte-Velasco, U., Sierra, I., Zudaire, L., & Ayastuy, J. L. (2015). Conversion of Waste Animal Bones into Porous Hydroxyapatite by Alkaline Treatment: Effect of the Impregnation Ratio and Investigation of the Activation Mechanism. Journal of Materials Science, 50(23), 7568–7582.

Ismail, Z. Z., & AbdelKareem, H. N. (2015). Sustainable Approach for Recycling Waste Lamb and Chicken Bones for Fluoride Removal from Water Followed by Reusing Fluoride-bearing Waste in Concrete. Waste Management, 45, 66–75.

Joshi, S., & Pokharel, B. P. (2013). Preparation and Characterization of Activated Carbon from Lapsi (Choerospondias axillaris) Seed Stone by Chemical Activation with Potassium Hydroxide Preparation of Adsorbent Characterization of Activated Carbons Thermal Behavior of Lapsi seed stone par. Journal of the Institute of Engineering, 9(1), 79–88.

Kaouah, F., Boumaza, S., Berrama, T., Trari, M., & Bendjama, Z. (2013). Preparation and Characterization of Activated Carbon from Wild Olive Cores (oleaster) by H3PO4 for the Removal of Basic Red 46. Journal of Cleaner Production, 54, 296–306.

Karthikeyan, G., & Ilango, S. S. (2008). Adsorption of Cr (VI) onto Activated Carbons Prepared from Indigenous Materials. E-Journal of Chemistry, 5(4), 666–678.

Kyzas, G. Z., Fu, J., Lazaridis, N. K., Bikiaris, D. N., & Matis, K. A. (2015). New Approaches on the Removal of Pharmaceuticals from Wastewaters with Adsorbent Materials. Journal of Molecular Liquids, 209, 87–93.

Lawrence, M., & Jiang, Y. (2017). Porosity , Pore Size Distribution ,. S. Amzinane and F. Collet (eds.). Bio-aggregates Based Building Materials (39-71). UK: RILEM State of the Art Reports.

Liu, H., Ning, W., Cheng, P., Zhang, J., Wang, Y., & Zhang, C. (2013). Evaluation of Animal Hairs-based Activated Carbon for Sorption of Norfloxacin and Acetaminophen by Comparing with Cattail Fiber-based Activated Carbon. Journal of Analytical and Applied Pyrolysis, 101, 156–165.

Marsh, H., & Rodríguez-Reinoso, F. (2006). Activation Processes (Chemical). In Activated Carbon (pp. 322–365). Amsterdam, the Netherlands: Elsevier Science.

Mashayekh-Salehi, A., & Moussavi, G. (2016). Removal of Acetaminophen from The Contaminated Water using Adsorption onto Carbon Activated with NH4Cl. Desalination and Water Treatment, 57, 12861–12873.

Mestre, A. S., Pinto, M. L., Pires, J., Nogueira, J. M. F., & Carvalho, A. P. (2010). Effect of Solution pH on the Removal of Clofibric Acid by Cork-based Activated Carbons. Carbon, 48(4), 972–980.

Mestre, A. S., Pires, R. A., Aroso, I., Fernandes, E. M., Pinto, M. L., Reis, R. L., … Carvalho, A. P. (2014). Activated Carbons Prepared from industrial Pre-treated Cork: Sustainable Adsorbents for Pharmaceutical Compounds Removal. Chemical Engineering Journal, 253(106637), 408–417.

Mittal, A., Kurup, L., & Mittal, J. (2007). Freundlich and Langmuir Adsorption Isotherms and Kinetics for the Removal of Tartrazine from Aqueous Solutions using Hen Feathers. Journal of Hazardous Materials, 146(1–2), 243–248.

Mphahlele, K., Onyango, M. S., & Mhlanga, S. D. (2015). Adsorption of Aspirin and Paracetamol from Aqueous Solution using Fe/N-CNT/??-cyclodextrin Nanocomopsites Synthesized via a Benign Microwave Assisted Method. Journal of Environmental Chemical Engineering, 3(4), 2619–2630.

Mukoko, T., Mupa, M., Guyo, U., & Dziike, F. (2015). Preparation of Rice Hull Activated Carbon for the Removal of Selected Pharmaceutical Waste Compounds in Hospital Effluent. Journal of Environmental & Analytical Toxicology, s7.

Murdoch, K. (2015). Pharmaceutical Pollution in the Environment : Issues for Australia , New Zealand and Pacific Island countries. Australia. Retrieved from http://www.ntn.org.au/wp/wp-content/uploads/2015/05/NTN-Pharmaceutical-Pollution-in-the-Environment-2015-05.pdf

Naderi, M. (2014). Surface Area: Brunauer-Emmett-Teller (BET). In S. Tarleton (Ed.), Progress in Filtration and Separation (pp. 585–608). UK: Elsevier Ltd.

Nartey, O. D., & Zhao, B. (2014). Biochar preparation, characterization, and adsorptive capacity and its effect on bioavailability of contaminants: An overview. Advances in Materials Science and Engineering, 2014.

Norbayu, S., Subari, M., Osman, R. and Saim, N. (2017) 'SCIENCE & TECHNOLOGY Occurrence , Source Apportionment and Environmental Risk Assessment of Pharmaceuticals in Klang River , Malaysia', 25, pp. 119–128.

Pakhre, V., & Srivastava, V. C. (2012). Adsorption of Benzoic Acid and Salicylic Acid onto Granular Activated Carbon. Environment, 261–265.

Papita, S., & Chowdhury, S. (2011). Insight into Adsorption Thermodynamics. In M. Tadashi (Ed.), Thermodynamics (pp. 349–364). InTech.

Park, J. H., Cho, J. S., Ok, Y. S., Kim, S. H., Kang, S. W., Choi, I. W., Seo, D. C. (2015). Competitive Adsorption and Selectivity Sequence of Heavy Metals by Chicken Bone-derived Biochar: Batch and Column Experiment. Journal of Environmental Science and Health, Part A, 50(11), 1194–1204.

Patel, S., Han, J., & Gao, W. (2015). Sorption of 17β-estradiol from Aqueous Solutions on to Bone Char Derived from Waste Cattle Bones: Kinetics and Isotherms. Journal of Environmental Chemical Engineering, 3(3), 1562–1569.

Pathak, P. D., Mandavgane, S. A., & Kulkarni, B. D. (2016). Utilization of Banana Peel for the Removal of Benzoic and Salicylic Acid from Aqueous Solutions and its Potential Reuse. Desalination and Water Treatment, 57(27), 12717–12729.

Plazinski, W., & Rudzinski, W. (2009). Kinetics of Adsorption at Solid/solution Interfaces Controlled by Intraparticle Diffusion: A Theoretical Analysis. Journal of Physical Chemistry C, 113(28), 12495–12501.

Reck, I. M., Paixão, R. M., Bergamasco, R., Vieira, M. F., & Vieira, A. M. S. (2018). Removal of Tartrazine from Aqueous Solutions using Adsorbents Based on Activated Carbon and Moringa Oleifera Seeds. Journal of Cleaner Production, 171, 85–97.

Ribeiro, A. V. F. N., Silva, A. R. da, Cunha, T. P. da, Santos, R. T. L. dos, Oliveira, J. P. de, Pereira, E. V., … Ribeiro, J. N. (2016). Banana Peel for

Acetylsalicylic Acid Retention. Journal of Environmental Protection, 07(12), 1850–1859.

Rivera-Utrilla, J., Sánchez-Polo, M., Ferro-García, M. Á., Prados-Joya, G., & Ocampo-Pérez, R. (2013). Pharmaceuticals as Emerging Contaminants and Their Removal from Water. A Review. Chemosphere, 93(7), 1268–1287.

Samsuddin, S., Sharaai, A.H. & Ismail, M.M., 2015. Sustainability of Chicken Meat Production in achieving Food Security in Malaysia. , 9(October), pp.1–6.

Shi, H., Cheng, X., Wu, Q., Mu, R., & Ma, and Y. (2012). Assessment and Removal of Emerging Water Contaminants. Journal of Environmental & Analytical Toxicology, s2, 1–14.

Sing, K. S. W. (2013). Adsorption by Active Carbons. Adsorption by Powders and Porous Solids: Principles, Methodology and Applications: Second Edition (2nd ed.). Elsevier Ltd.

Soleimani, M., & Kaghazchi, T. (2007). Agricultural Waste Conversion to Activated Carbon by Chemical Activation with Phosphoric Acid. Chemical Engineering and Technology, 30(5), 649–654.

Teo, H. T., Siah, W. R., & Yuliati, L. (2016). Enhanced adsorption of acetylsalicylic acid over hydrothermally synthesized iron oxide-mesoporous silica MCM-41 composites. Journal of the Taiwan Institute of Chemical Engineers, 65, 591–598.

Thommes, M., Kaneko, K., Neimark, A. V, Olivier, J. P., Rodriguez-reinoso, F., Rouquerol, J., & Sing, K. S. W. (2015). Physisorption of Gases , with Special Reference to the Evaluation of Surface Area and Pore Size Distribution (IUPAC Technical Report).

Tsibranska, I., & Hristova, E. (2011). Comparison of Different Kinetic Models for Adsorption of heavy Metals onto Activated Carbon from Apricot Stones. Bulgarian Chemical Communications, 43(3), 370–377.

Vesna, R., Vladislav, R., Marija, K., Otman, O., & Aline, A. (2015). The Adsorption of Pharmaceutically Active Compounds from Aqueous Solutions onto Activated Carbons. Journal of Hazardous Materials, 282, 141–149.

Vining, W., Young, Day, R., & Botch, B. (2014). Thermodynamics: Entropy and Free Energy. In General Chemistry (pp. 617–650). Boston, USA: Cengage Learning.

Wei, S., Li, D., Huang, Z., Huang, Y., & Wang, F. (2013). High-capacity Adsorption of Cr(VI) from Aqueous Solution using a Hierarchical Porous Carbon Obtained from Pig Bone. Bioresource Technology, 134, 407–411.

Wu, S., Zhang, L., & Chen, J. (2012). Paracetamol in The Environment and Its Degradation by Microorganisms. Applied Microbiology and Biotechnology, 96(4), 875–884.

Y. Bell, K., J. M. Wells, M., A. Traexler, K., Pellegrin, M.-L., Morse, A., & Bandy, J. (2009). Emerging Pollutants. Water Environment Research, 81(10), 2211–2254.

Yahya, M. A., Al-Qodah, Z., & Ngah, C. W. Z. (2015). Agricultural Bio-waste Materials as Potential Sustainable Precursors Used for Activated Carbon Production: A review. Renewable and Sustainable Energy Reviews, 46, 218–235.

Yakout, S. M., & Sharaf El-Deen, G. (2012). Characterization of Activated Carbon Prepared by Phosphoric Acid Activation of Olive Stones. Arabian Journal of Chemistry.

Zhao, Y., Yang, S., Wang, G., & Han, M. (2016). Adsorption Behaviors of Acetaminophen onto the Colloid in Sediment. Polish Journal of Environmental Studies, 24(2), 853–861.

Znad, H., & Frangeskides, Z. (2014). Chicken Drumstick Bones as an Efficient Biosorbent for Copper (II) Removal from Aqueous Solution. Desalination and Water Treatment, 52(7–9), 1560–1570.