

ADSORPTIVE COAGULATION-FLOCCULATION REMOVAL OF  
ANTIBIOTICS IN SEWAGE TREATMENT PLANTS

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ANTIBIOTICS IN SEWAGE TREATMENT PLANTS

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To my beloved family

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## ABSTRACT

The persistent existence of antibiotics in sewage wastewater treatment plants in recent years has emerged as a serious issue. This scenario has indicated that coagulation-flocculation process is ineffective in removing antibiotics from sewage. Therefore, adsorptive coagulation-flocculation process has been proposed as novel sewage treatment method to remove antibiotics. In this study, three types of natural zeolites (NZ01, NZ02 and NZ03) from different sources were employed as adsorbents in batch adsorption test to remove selected antibiotics which were tetracycline (TC) and oxytetracycline (OTC). The physical appearance and chemical properties of these natural zeolites were characterized using scanning electron microscopy (SEM), cation exchange capacity (CEC), Brunauer Emmett Teller (BET), Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD) and X-ray fluorescence (XRF) methods. Adsorption test was carried out in batch whereas adsorptive coagulation-flocculation was performed using jar test method. The adsorption data was evaluated in terms of equilibrium and kinetic adsorption mechanism. Important parameters including pH of solution, dosage of adsorbents, temperature, initial concentration of antibiotics and contact time were varied to study the effects of these parameters. It has been observed that at pH between 7 to 8, and temperature of 30 °C the adsorption of antibiotics was optimum with about 90% removal of TC and 70% removal of OTC at the dosage of 6 mg/ml of NZ02. In addition, the adsorption of TC and OTC has been proven to follow Elovich kinetic model and Langmuir model for TC and Temkin isotherm model for OTC. The optimum parameters were later applied in adsorptive coagulation-flocculation (ACF) process to remove antibiotics in synthetic wastewater. Langmuir model and pseudo-second order model turned out to be the most suitable isotherm model and kinetics model to represent the adsorption of antibiotics via ACF. The percentage of removal of antibiotics from synthetic wastewater using only alum was clearly proven to be ineffective (about 5%). However, by injecting NZ02 simultaneously, it increased dramatically (about 20-35%). Last but not least, the hybrid process managed to remove 60% of TC and 40% of OTC from sewage wastewater. In conclusion, adsorptive coagulation-flocculation has the potential to be one of the best alternative methods to isolate antibiotics in sewage treatment plants in the most environmentally and economically friendly way.

## ABSTRAK

Kewujudan berterusan antibiotik di kumbahan loji rawatan air sisa pada tahun-tahun kebelakangan ini telah muncul sebagai satu isu yang serius. Senario ini telah menunjukkan bahawa proses pembekuan-pemberbukuan tidak berkesan dalam menyingkirkan antibiotik daripada kumbahan. Oleh itu, proses jerapan pembekuan-pemberbukuan telah dicadangkan sebagai kaedah baru rawatan kumbahan untuk menyingkirkan antibiotik. Dalam kajian ini, tiga jenis zeolit semulajadi (NZ01, NZ02 dan NZ03) daripada sumber-sumber yang berbeza telah digunakan sebagai penjerap dalam kumpulan ujian penjerapan untuk meresap antibiotik yang terpilih iaitu tetrasiklin (TC) dan oksitetrasiklin (OTC). Sifat fizikal dan kimia zeolit semulajadi telah dikaji dengan menggunakan mikroskopi elektron imbasan (SEM), kapasiti penukaran kation (CEC), *Brunauer Emmett Teller* (BET), spektroskopi inframerah transformasi Fourier (FTIR), pembelauan sinar X (XRD) dan pendarfluor sinar X (XRF). Ujian penjerapan telah dijalankan secara kelompok manakala process hibrid penjerapan dan penggumpalan-pengelompokan telah dilakukan dengan menggunakan kaedah ujian balang. Data penjerapan telah dinilai dari segi kajian isoterma dan mekanisme kinetik penjerapan. Parameter penting termasuk pH larutan, dos penjerap, suhu, kepekatan awal antibiotik dan masa proses telah diubah untuk mengkaji kesan parameter-parameter tersebut. Pada pH di antara 7 hingga 8 dan suhu 30 °C, penjerapan antibiotik adalah optimum dengan penyingkiran kira-kira 90% TC dan 70% OTC dicapai pada dos 6 mg/ml NZ02. Di samping itu, penjerapan TC dan OTC dipercayai mematuhi model kinetik *Elovich*, model isoterma *Langmuir* untuk TC dan model isoterma *Temkin* untuk OTC. Parameter optimum kemudiannya digunakan dalam proses hibrid penjerapan dan pembekuan-pemberbukuan (ACF) untuk menyingkirkan antibiotik daripada air sisa sintetik. Model *Langmuir* dan model pseudo-tertib kedua ternyata menjadi model isoterma dan model kinetik yang paling sesuai untuk penjerapan antibiotik melalui ACF. Peratusan penyingkiran daripada air sisa antibiotik sintetik dengan menggunakan alum sahaja ternyata tidak berkesan (kira-kira 5%). Walau bagaimanapun, dengan menggunakan NZ02 pada masa yang sama, penyingkiran meningkat secara mendadak (lebih kurang 20-35%). Akhir sekali, proses hibrid berjaya menyingkirkan 60% TC dan 40% OTC daripada air sisa kumbahan. Kesimpulannya, proses hibrid penjerapan pembekuan-pemberbukuan mempunyai potensi untuk menjadi salah satu daripada kaedah alternatif terbaik untuk menyingkirkan antibiotik dalam loji rawatan kumbahan dengan cara yang paling mesra alam dan ekonomi.

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## LIST OF SYMBOLS

$a$	-	Initial adsorption rate ( $\mu\text{mol/g}\cdot\text{min}$ )
$\beta$	-	Sorption energy ( $\text{mol}^2/\text{kJ}^2$ )
$b$	-	Desorption constant ( $\text{g}/\mu\text{mol}$ )
$B$	-	Heat of adsorption ( $\text{kJ/mol}$ )
$b_T$	-	Temkin model constant
$c_o$	-	Initial concentration ( $\mu\text{M}$ )
$c_e$	-	Equilibrium concentration ( $\mu\text{M}$ )
$c_1$	-	Remaining magnesium concentration ( $\text{mmol/L}$ )
$c_2$	-	Corrected magnesium concentration ( $\text{mmol/L}$ )
$c_{b1}$	-	Magnesium concentration in blank ( $\text{mmol/L}$ )
CEC	-	Cation exchange capacity ( $\text{cmol/kg}$ )
$\Delta G^\circ$	-	Change of standard Gibbs free energy ( $\text{kJ/mol}$ )
$\Delta H^\circ$	-	Change of standard enthalpy ( $\text{kJ/mol}$ )
$\Delta S^\circ$	-	Change of standard entropy ( $\text{kJ/mol}\cdot\text{K}$ )
$\varepsilon$	-	Dubinin-Radushkevich constant
$E$	-	Mean free energy ( $\text{kJ/mol}$ )
$k_1$	-	Lagergren rate constant ( $\text{min}^{-1}$ )
$k_2$	-	Rate constant of pseudo-second-order model ( $\mu\text{mol}/\text{min}$ )
$k_d$	-	Intraparticle diffusion rate constant ( $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{min}^{-0.5}$ )
$K_d$	-	Equilibrium constant ( $\text{L/g}$ )
$K_F$	-	Freundlich sorption capacity constant ( $\text{L}^{1/n}\mu\text{mol}^{1-1/n}/\text{g}$ )
$K_L$	-	Langmuir equilibrium constant ( $\text{L}/\mu\text{mol}$ )
$K_T$	-	Equilibrium binding constant ( $\text{L}/\mu\text{mol}$ )
$K_{OW}$	-	Octanol water partition coefficient
$m$	-	Mass of dried natural zeolite ( $\text{g}$ )

$m_1$	-	Mass of dried zeolite and test tube (g)
$m_2$	-	Mass of wet zeolite and test tube (g)
$M$	-	Mass of natural zeolites (g)
$pK_a$	-	Acidic equilibrium constant
$q_e$	-	Adsorption capacity ( $\mu\text{mol/g}$ )
$q_m$	-	Maximum adsorption capacity ( $\mu\text{mol/g}$ )
$q_t$	-	Adsorption capacity at time $t$ ( $\mu\text{mol/g}$ )
$q_{\text{cal}}$	-	Calculated adsorption capacity ( $\mu\text{mol/g}$ )
$q_{\text{ex}}$	-	Experimental adsorption capacity ( $\mu\text{mol/g}$ )
$R$	-	Gas constant (8.31 kJ/mol.K)
$R^2$	-	Correlation of determination
$RE$	-	Removal efficiency (%)
$t$	-	Time (min)
$T$	-	Temperature (K)
$V$	-	Volume of solution (L)
$\chi^2$	-	Chi square analysis ( $\mu\text{mol/g}$ )

**LIST OF ABBREVIATIONS**

AAS	-	Atomic absorption spectrophotometer
ACF	-	Adsorptive coagulation-flocculation
BET	-	Brunauer-Emmett-Teller
BJH	-	Barrett-Joyner-Halenda
BOD	-	Biological oxygen demand
CEC	-	Cation exchange capacity
COD	-	Chemical oxygen demand
DR	-	Dubinin-Radushkevich
EDC	-	Endocrine damaging chemical
EDL	-	Electrodes discharge lamp
FAU	-	Faujasite
FTIR	-	Fourier transform infrared spectroscopy
MOR	-	Mordenite
MP	-	Micropollutant
NZ	-	Natural zeolite
OTC	-	Oxytetracycline
PFO	-	Pseudo-first order
PSO	-	Pseudo-second order
SEM	-	Scanning electron microscopy
STP	-	Sewage treatment plant
TC	-	Tetracycline
TSS	-	Total suspended solid
XRD	-	X-ray diffraction
XRF	-	X-ray fluorescene

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## **CHAPTER 1**

### **INTRODUCTION**

#### **1.1 Research Background**

Water is one of the most common compounds in this world. It covers 70% of the surface of planet Earth. However, only 3% of water is freshwater and only freshwater is consumable for human for interior purpose and exterior usage. Without clean freshwater, the existence of human and most organism is impossible. Therefore, it is utmost important to make sure the quality of freshwater is well preserved.

Due to the growth of global population and rapid development, the demand of water has been increasing for the last few decades. In addition, active industrial, domestic and agro-industrial activities have increased the disposal of wastewater into natural water body. As a matter of fact, wastewater consists of organic and inorganic pollutants which are originated from chemicals, pharmaceutical, electrochemical, electronics, petrochemical and food processing industries. Besides, heavy metals such as arsenic, mercury and lead which are toxic to almost all organisms have been found in sewage. Recently, traces of pharmaceutical have also been discovered in wastewater (Zupanc *et al.*, 2014).

Without proper treatment of sewage before disposal, the environment and human health will be endangered. There have been severe cases reported worldwide which are related to inefficient sewage treatment process. For instance, in the mid-1950, the residents near the Agano River and Yatsushiro Sea in Japan suffered from the Minamata disease caused by methyl mercury in industrial wastewater. The chemical in the wastewater attacked the central nervous system and caused the consumers to suffer from brain damage. Besides tragic case in Minamata, heavy metals were detected in Chao Phraya River in Thailand (Wijaya *et al.*, 2013), *Cryptosporidium* oocysts and *Giardia* cysts which are dangerous parasites were found in the Sagami River, Japan (Hashimoto *et al.*, 2002), and organic pollutants and pesticides had been detected in Yangtze River in China (Qi *et al.*, 2014). From the fact above, conclusion might be drawn that Asian countries did not have efficient technology in treating wastewater or they did not have the awareness about water hygiene.

However, Asian countries are not left behind in overcoming water related problem. Japan and China have shown promising commitment in overcoming water issues locally and globally by applying innovative advanced wastewater treatment technologies after being the victims of water pollution (Kato *et al.*, 2013; Yuan *et al.*, 2010). Unfortunately, there are emerging pollutants in water due to increased complexity of the chemical process in industry whereby their presence has raised up significant concerns among scientists. This is because the existing sewage treatment plants do not have the capability to remove those pollutants in water. Pharmaceutical compounds especially antibiotics are one of them which are very hard to be removed.

Antibiotics were detected in nature water body for almost 30 years ago. However, due to its' usage in large amount for many years as medicine, people began to aware of its' increased presence in municipal wastewater, groundwater, and surface water in the mid-1990s. Even though they only exist in a very minute concentrations in sewage (normally in the unit of ng/L), it has the possibility to endanger human and environment health in the long term. It could alter aquatic ecology by promoting the mutation of antibiotic-resistance micro bacterial, creating



diseases which could not be treated effectively in the years to come (Tong *et al.*, 2009). As mentioned above, the existing sewage treatment plants are ineffective in treating wastewaters which have highly polar pollutants such as antibiotics. Thus, this proposed research is to develop alternative cost saving and effective ways in removing antibiotics in sewage treatment plants.

## 1.2 Problems Statements

More and more regions in the world are recycling wastewater into drinking water by a series of wastewater treatment process which remove undesired macro and micro pollutant either it is organic or inorganic so that the standard of quality water is reached and safe to be consumed (Al-Rifai *et al.*, 2007). Generally, pollutants found in wastewater consist of heavy metals, nitrogen, pathogen, pesticides, magnesium, calcium and suspended solids. In recent years, there are other emerging contaminants in wastewater such as endocrine-disruptive compound and pharmaceutical compound. The fact is that the rise of antibiotics in wastewater due to its' extensive use in veterinary and healthcare medicine has stirred up the awareness starting from scientists and researchers to public (Batt *et al.*, 2006). Traces of antibiotics had been detected at several regions at all parts of the world. Even though only a minute amount of antibiotics present in wastewater, negative impacts brought by antibiotics could be felt and clearly proved by analytical method in long term. Firstly, the reproductive system of aquatic organism such as freshwater flea and crustacean is adversely affected by continual exposure to antibiotics. Furthermore, the population of antibiotic resistance genes and bacteria has been spreading as a result of the consistence presence of antibiotics in wastewater. In conclusion, antibiotics should be removed from wastewater as it poses harmful consequences to the biodiversity and human health.

However, current designs of conventional wastewater treatment plants which include coagulation and flocculation could not effectively remove antibiotics from wastewater (Rizzo *et al.*, 2013). Adsorptive removal of antibiotics has been studied and the results is promising (Chao *et al.*, 2014). Adsorption is defined as the process of attachment of liquid or gas particles on the surface of a solid surface. It is a feasible method in wastewater treatment as it has attractive initial cost, simplicity of design, ease of operation and insensitivity to toxic substances. In fact, more than 90% of antibiotics (ciprofloxacin and norfloxacin) in wastewater could be removed by using adsorption on activated carbon (Ahmed and Theydan, 2014).

Nevertheless, the shortcomings of activated carbon which are costly and difficulty in regeneration have become the driving force in seeking unto other adsorbents. Currently, natural zeolite had found various applications worldwide such as for catalytic purpose, sorption and building materials. According to research, natural zeolite could be used in wastewater treatment plant such as to remove ammonium and heavy metals (Widiastuti *et al.*, 2011; Egashira *et al.*, 2012). In addition, it could also remove pharmaceutical products such as antibiotics (Wang and Peng, 2010). Moreover, natural zeolite could be modified to enhance the performance of adsorption (Guo *et al.*, 2013).

Adsorptive coagulation and flocculation is a new process in wastewater treatment which is the combination of adsorption and coagulation-flocculation. Both processes take place at the same time. It is an economical method as adsorbents with target pollutants attached on will settle down at sediment tank via coagulation-flocculation. Without coagulation-flocculation, more expensive adsorption column such as fixed bed adsorption column must be used so that the adsorbents and adsorbed pollutants will not be discharge out from wastewater treatment units. In short, novel economical attractive removal method which is adsorptive coagulation-flocculation using natural zeolite is proposed to assist current sewage treatment plants to remove antibiotics.

### 1.3 Research Objectives

This research was conducted based on the following objectives in accordance to the problem statements: (a) to characterize natural zeolites as adsorbents; (b) to evaluate the isotherms and kinetics of antibiotics removal process and (c) to evaluate the antibiotics removal performance.

### 1.4 Scopes

The scopes of the research were presented in order to achieve the three outlined objectives:

- (a) Natural zeolites (NZ01, NZ02 and NZ03) from different sources were characterized to determine the ideal structure for the adsorption of organic micropollutants. Characterization of natural zeolites was carried out by several methods including X-ray Diffraction (XRD), X-ray Fluorescence (XRF), Scanning Electron Microscopy (SEM), Cation Exchange Capacity (CEC), Brunauer-Emmett-Teller (BET) method and Fourier Transform Infrared Spectroscopy (FTIR).
- (b) Tetracycline (TC) and oxytetracycline (OTC) were chosen as model antibiotics in this study. The isotherms and kinetics of antibiotics adsorption will be carried out using batch adsorption procedure. Several adsorption parameters such as initial concentration of antibiotics, pH, adsorbent dosage, temperature and time were studied. Isotherms data of antibiotics adsorption was analyzed using the existing equilibrium models such as Langmuir, Freundlich, Temkin and Dubinin-Radushkevich. On the other hand, kinetics data will be analyzed using pseudo-first order model, pseudo-second order model, Elovich model and intra-particle diffusion model.

- (c) The removal performance of antibiotics adsorption was evaluated using jar test study by adding adsorbents and coagulants. The natural zeolite was used as a adsorbent and alum as a coagulant. Tetracycline and oxytetracycline were selected as model antibiotics in the adsorptive coagulation-flocculation process. Removal performance was evaluated at various conditions.

## **1.5 Thesis Outline**

There are 5 chapters all together in this research proposal. Chapter 1 included research background, problem statement, objectives and scope of study, research proposal outline, and summary. Chapter 2 consisted of the review of previous studies in recent years including conventional sewage treatment, antibiotics in wastewater and technologies involved in the removal of antibiotics, adsorptive coagulation and flocculation and its' application in separating antibiotics. The last part of this chapter reviewed the use of natural zeolite and modified zeolite in wastewater treatment whereby the wastewater contains antibiotics. In Chapter 3, research methodology was discussed. The chemical and physical properties of research materials which were general chemicals, antibiotics and natural zeolites were listed out. Besides, experimental procedures were established. In addition, characterization procedures and analytical procedures were also described in detail. Characterization procedures covered the characterization of zeolite and sewage using various methods whereas analytical procedures evaluated the adsorption performance based on the final concentration of antibiotics in sample solution. Chapter 4 exhibited the experimental results obtained from characterization of adsorbents, batch adsorption and adsorptive coagulation and flocculation. Lastly, Chapter 5 structured the final conclusions about the findings of the entire research and recommendations for future research were suggested for the aim of improvement.

## 1.6 Summary

The continuous presence of antibiotics in wastewater has raised concerns that it will disturb the balance of aquatic biodiversity as well as bringing harmful effect to human health. The conventional sewage treatment plants were not designed to remove polar contaminants such as antibiotics. The adsorptive coagulation-flocculation (ACF) was proposed as the alternative advanced wastewater treatment in coping with the global mutual problem in recent decades. Natural zeolites were selected as the adsorbents due to their excellent cation exchange capability. Batch experiments were conducted to evaluate the thermodynamics and kinetics of the adsorption of antibiotics on the adsorbents. In addition, the effect of several parameters such as initial concentration of antibiotics, pH, temperature, dosage of adsorbent and contact time of mixing was investigated. Finally, jar test was carried out to evaluate the percentage of removal of antibiotics from the synthetic wastewater and sewage wastewater via adsorptive coagulation-flocculation (ACF) using natural zeolite was evaluated.

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