ACTIVATED CARBON FROM TEXTILE SLUDGE FOR THE ADSORPTION OF REACTIVE BLACK 5 AND METHYLENE BLUE

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Specially dedicated to my beloved parents.

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ABSTRACT

Adsorption is known to be one of the best techniques for treatment of dye in water. Many types of research had been done in preparing low-cost activated carbon from industrial wastes and agricultural by-products. In this study, activated carbon derived from textile sludge (TS) was synthesized and its performance was tested for removal of reactive black 5 (RB5) and methylene blue (MB) which represent anionic and cationic dyes, respectively. The activated carbon was synthesized through a chemical activation process by sulfuric acid followed with carbonization which was run at 650 ºC in a furnace under nitrogen flow. Textile sludge-activated carbon (TSAC) was characterized by Fourier transform infrared, field emission scanning electron microscopy, energy dispersive x-ray and nitrogen adsorption isotherm analysis. The characterization results revealed that the activated carbon from textile sludge had been successfully synthesized. The Brunauer, Emmett and Teller (BET) surface area of TSAC increased from 90.65 m^2/g (BET surface area for TS) to 221.52 m^2/g. The adsorption capacity was found to be dependent on adsorption time, pH, adsorbent dosage and initial dye concentration. The adsorption data for both dyes were well fitted to Freundlich isotherm model which explains the heterogeneous nature of the TSAC surface. The adsorption obeys the pseudo-second order kinetic model, which indicates that the adsorption was classified as chemisorption as the rate limiting step. The positive value of enthalpy change confirmed the endothermic nature of the overall process. The negative values of standard Gibbs free energy indicated the feasibility and spontaneity of the adsorption processes while the positive value of entropy change suggested the affinity of TSAC for both dyes and increased randomness at the solid/solution interface during the adsorption process. The highest adsorption capacity for the adsorption of RB5 and MB dyes obtained were 11.978 mg/g and 13.272 mg/g, respectively.

ABSTRAK

Penjerapan merupakan salah satu teknik yang paling berkesan dalam merawat masalah pewarna di dalam air. Pelbagai kajian telah dilakukan dalam menghasilkan karbon teraktif yang melibatkan kos yang rendah, iaitu daripada sisa-sisa industri dan bahan buangan produk pertanian. Dalam kajian ini, karbon teraktif daripada enapcemar tekstil (TS) telah dihasilkan dan kegunaannya diuji untuk menjerap dua jenis pewarna iaitu reaktif hitam 5 (RB5) yang mewakili pewarna anion dan metilena biru (MB) yang mewakili pewarna kation. Karbon teraktif disintesiskan melalui proses pengaktifan kimia oleh asid sulfurik diikuti pengkarbonan yang dijalankan pada suhu 650 ºC dibawah aliran gas nitrogen. Karbon teraktif daripada enapcemar tekstil (TSAC) dicirikan oleh infra-merah transformasi Fourier, mikroskopi pengimbas elektron pancaran medan, tenaga serakan sinar-x dan analisis penjerapan isoterma nitrogen. Hasil pencirian menunjukkan bahawa karbon teraktif daripada enapcemar tekstil ini telah berjaya disintesiskan. Luas permukaan bagi TSAC daripada keputusan Brunauer, Emmett dan Teller (BET) menunjukkan peningkatan daripada 90.65 m^2/g (luas permukaan BET bagi TS) kepada 221.52 m^2/g. Kapasiti penjerapan didapati bergantung kepada masa penjerapan, pH, dos penjerap dan kepekatan larutan pewarna. Data penjerapan telah dipadankan dengan model isoterma Freundlich yang menunjukkan ciri heterogen pada permukaan TSAC. Proses penjerapan ini mematuhi model kinetik pseudo-tertib-kedua bagi kedua-dua jenis pewarna yang mana menunjukkan penjerapan kimia sebagai langkah pengehadan kadar penjerapan. Nilai negatif perubahan entalpi mengesahkan sifat endotermik bagi keseluruhan proses penjerapan. Nilai negatif piawai tenaga bebas Gibbs menunjukkan kebolehlaksanaan dan proses penjerapan spontan dan nilai positif perubahan entropi mencadangkan pertalian TSAC dengan kedua-dua pewarna dan peningkatan rawak pada antaramuka penjerap/pewarna semasa proses penjerapan. Nilai penjerapan tertinggi yang diperoleh bagi penyingkiran pewarna RB5 dan MB masing-masing adalah 11.978 dan 13.272 mg/g.

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LIST OF SYMBOLS

Å	-	Angstrom
C_e	-	Equilibrium concentration
C_o	-	Initial concentration
k_1	-	Adsorption rate constant of first order adsorption
k_2	-	Adsorption rate constant of second-order adsorption
K_F	-	Freundlich constant
K_L	-	Langmuir constants related to the rate of adsorption
k_p	-	Intraparticle diffusion rate constant
n	-	Freundlich constant
q_e	-	Amount of adsorbent at equilibrium
Q_o	-	Langmuir constants related to adsorption capacity
q_t	-	Equilibrium rate constant
R^2	-	Correlation coefficient
R_L	-	Separation factor
t	-	Time
$t^{1/2}$	-	Half-life time
V	-	Volume
m	-	Mass of adsorbent
ΔG	-	Gibbs Free Energy
ΔH	-	Enthalpy
ΔS	-	Entropy
λ_{max}	-	Maximum wavelength

LIST OF ABBREVIATIONS

BET	-	Brunauer, Emmett and Teller
CAC	-	Commercial Activated Carbon
DR	-	Dubinin-Radushkevich
EDX	-	Energy Dispersive X-Ray
EPA	-	Environmental Protection Agency
FESEM	-	Field Emission Scanning Electron Microscopy
FTIR	-	Fourier Transform Infrared Spectroscopy
IUPAC	-	International of Pure and Applied Chemistry
MATRADE	-	Malaysia External Trade Development Corporation
MB	-	Methylene Blue
N	-	Normality
NAD	-	Nitrogen Adsorption-Desorption Isotherm Analysis
RB5	-	Reactive Black 5
TGA	-	Thermogravimetric Analyzer
TS	-	Textile Sludge
TSAC	-	Textile Sludge Activated Carbon
UV-Vis	-	Ultra Violet Viscometer

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CHAPTER 1

INTRODUCTION

1.1 Research Background

Dye is one of the major pollutants in water ecosystem. It is highly noticeable because of the visibility of its colour. Dyes in wastewater consist of several contaminants such as acids, bases, dissolved solids, toxic compounds and colour (Chu, 2001). It is estimated about 10-15% of dyes were released into the environment through the dyeing process in the industry (Padhi, 2012). The textile industry is the largest consumer of dye stuff besides dyeing industries, paper and pulp industries, and paint industries. Dyes and their degradation products have been related to toxicity. Thus, the treatment of wastewater containing dyes has been taken seriously before the it is discharged to the environment.

Dyes can be classified into cationic, anionic and nonionic dyes. Anionic dyes comprise of acid, direct and reactive dyes, whereas cationic dyes comprise of basic dyes and nonionic dyes comprise of disperse dyes (Eren, 2009). Acid dyes from the anionic dyes group have a harmful effect on human beings since they are organic sulphonic acids (Attia *et al.*, 2006). Anionic dyes have a low degree of fixation due to the hydrolysis of reactive groups in the water phase. Therefore, the discharging of this type of reactive dyes into the environment is undesirable (Tabak *et al.*, 2009). On the other hand, cationic dyes are also considered as harmful and toxic colourants which can cause allergic dermatitis, skin irritation, mutations and cancer (Eren, 2009). Cationic dyes carry a positive charge in their molecule. It is water soluble and yield coloured cations in solution (Salleh *et al.*, 2011). Methylene blue is an

important basic dye and widely used in the textile industry. It is not strongly hazardous. However, acute exposure to methylene blue might cause increasing of heart rate, shock, vomiting, cyanosis, jaundice, quadriplegia, heinz body formation and tissue necrosis in humans (Vadivelan and Kumar, 2005).

Various techniques and technologies have been developed in solving dye-containing wastewater problem. It consists of chemical, biological and physical treatment (Robinson *et al*,. 2001; Salleh *et al.*, 2011). Some of the techniques are seen to have weaknesses in their application. For instance, flocculation technique may result in the significant of sludge production, whereas the membrane filtration may result to membrane fouling. Likewise, the practice of membrane filtration is also high in costing. Among these techniques, the adsorption by activated carbon had been found to be the best technique compared to all other techniques for treating dyes in wastewater (Tan *et al*., 2007). This is due to its efficiency, the simplicity of design and applicability of adsorbent on large scale application.

Activated carbon is a high-value carbonaceous adsorbent. It has high surface area and porosity from the highly developed pore structures (Ioannidou and Zabaniotou, 2007). The porosity structure of activated carbon enhances high adsorption capacity. Beside dyes, the application of activated carbon has been widely used in treating variety of wastewaters. However, the used of activated carbon was restricted due to its high associated cost. This is due to the high cost of the raw materials used in the production of activated carbon (Chakraborty *et al.*, 2005). Commercial activated carbon (CAC) is typically prepared from woods or coals precursor. These materials are non-renewable materials, which were found to be expensive and also limited in its applicability (Li *et al.*, 2011c). Therefore, lots of research have been done in preparing a low-cost activated carbon using waste, mainly industrial or agricultural by-products (Rio *et al*., 2006; Rozada *et al*., 2005). Any resources with high carbon contents with low inorganics have potentials to be used as the precursor for the activated carbon production (Tsai *et al.*, 1997). One of the interests was towards sludge waste. Some of sludge wastes involved were sewage sludge (Chen *et al*., 2002; Rio *et al*., 2006; Rio *et al*., 2005; Rozada *et al*., 2005), paper mill sewage sludge (Khalili *et al*., 2000; Li *et al.*, 2011a) and sludge

from food processing industry (Mahapatra *et al*., 2012). Sludge productions generally are resulted from the wastewater treatment plants in the industries. Thus, this undesirable sludge in the industry was used and transformed into more added value product.

Sludge from the industry had been found to be a good precursor for activated carbon production. In this study, sludge from textile industry was selected. The use of textile sludge shows a high potential as the precursor for the activated carbon production. The used of textile sludge in the production of activated carbon has been studied by Kaçan and Kütahyali (2012). However, the activated carbon produced was only focused on the adsorption of strontium, Sr^{2+} from the aqueous solution. Therefore, the application of activated carbon from textile sludge was extended to removal of two different type of dyes which are anionic dyes, Reactive Black 5 (RB5) and cationic dyes, Methylene Blue (MB) from aqueous solutions.

1.2 Research Statement

Wastewater containing dyes may cause harmful effect to human over exposure due to the toxicity of dyes compound. However, this colour contamination issue from the dyes containing wastewater is not a new arising issue. Many types of research had been done in finding the best solution for this dyes problems. One of the interests is the adsorption method by activated carbon.

The adsorption by activated carbon was found to be a superior technique in dyes treatment compared to all other chemical, biological and physical techniques (Robinson *et al.*, 2001; Salleh *et al.*, 2011). It is good removal of wide variety of dyes promising the best solution for solving this wastewater containing dyes problem. In the past, the used of activated carbon was restricted due to its high associated cost cause by the expensive or non-renewable raw materials (Li *et al.*, 2011c). However, many types of research had been done and the used of low-cost raw materials for the production of activated carbon shows promising results. Industrial waste or agricultural by-product were some of the low-cost raw materials

for activated carbon production that are widely used. Besides, interest had been shown in the used of sludge from industries as the activated carbon precursor.

Textile sludge is produced from wastewater treatment activity and their production is expected to gradually increase along with the increasing of textile industry in Malaysia. However, uncontrolled disposal of this sludge may resulted in contamination of land and water resources. Thus, these might relate to other various environmental issues. Research has found that the textile sludge showed high nutrient composition and has the potential to turn into high value-added products (Rosa *et al.*, 2007). Besides, textile sludge also contains a high value of organic carbon, which is suitable to be used for activated carbon production. The use of sludge wastes, which are difficult to manage for the production of activated carbon can turn a waste to an economically valuable product.

Research on the use of textile sludge as the precursor for the production of activated carbon had been studied by Kaçan and Kütahyali (2012). However, that study only focused on the adsorption of strontium, Sr^{2+} in aqueous solutions. Therefore, in this study, the used of activated carbon derived from textile sludge will be tested to adsorb two different groups of dyes which are anionic dyes, Reactive Black 5 (RB5) and cationic dyes, Methylene Blue (MB). Moreover, the different method was used in the activated carbon production to enhance the dyes adsorption capacity. The performance and characteristic of the synthesized textile sludge activated carbon were studied and analysed to compete with the conventional activated carbon.

This study is about the use of activated carbon adsorbent produced from textile sludge for the treatment of anionic and cationic dyes. In this way, a double benefit would be obtained, as it would improve sludge management by turning the sludge waste into a low-cost raw material for activated carbon production and its ability to adsorb two different groups of dyes. It is expected that the synthesized activated carbon showing a good performance for the dyes adsorption process.

1.3 Research Objectives

This research embarks on the following objectives:

i. To study the chemical and physical properties of textile sludge (TS) as the raw material for activated carbon preparation.
ii. To synthesize and characterize the activated carbon derived from textile sludge (TSAC).
iii. To investigate the effectiveness of synthesized textile sludge activated carbon (TSAC) as an adsorbent for removal of anionic dyes, Reactive Black 5 (RB5) and cationic dyes, Methylene Blue (MB).

1.4 Scopes of Research

The scope of this research will be limited to the following;

i. The starting material of textile sludge was obtained from Anfi Industries Sdn. Bhd., Batu Pahat. The collected sludge was sieved to average size of 0.5 mm.
ii. The chemical and physical properties of TS were studied through the characterization of TS. The characterizations involved are Fourier Transform Infrared (FTIR) Spectroscopy, Thermogravimetric Analyser (TGA), Field Emission Scanning Electron Microscopy (FESEM) and Energy Dispersive X-ray (EDX) and Nitrogen Adsorption Isotherm Analysis (NAD).
iii. The synthesis of TSAC was formed by chemical activation method and carbonization at 650 ºC. Initially, screening was done by three different chemical activating agents ($ZnCl_2$, KCl and H_2SO_4) with the same concentration of 1 M. The best chemical activating agent (H_2SO_4) was selected to be used in the synthesis of TSAC for the subsequent dyes adsorption studies.

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