# SYNTHESIS AND CHARACTERISATION OF MAGNETIC-POLYETHYLENEIMINE-CELLULOSE ADSORBENT FOR REACTIVE BLACK 5 DYE REMOVAL

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To my beloved father and mother,

Nordin Mohamadan
Jamaliah Maskan

To my supervisor,

Assoc. Prof. Dr. Norzita Ngadi

Also to all my friends,

Thank you for your love, support and guidance

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In the name of Allah, the Most Gracious and the Most Merciful

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# ABSTRACT

The application of adsorption method for removing dye from textile wastewater has been recognised and adsorption onto conventional activated carbon is known to be the best prospect in eliminating dye that exhibits colour in water. However, activated carbon is costly and inconvenient for regeneration process. Thus, alternative adsorbents have been investigated. It is well-known that the waste materials from agriculture can be obtained and employed as low-cost adsorbents. In this study, cellulose from oil palm empty fruit bunch was modified with polyethyleneimine (PEI) and magnetic nanoparticles via crosslinking method to remove reactive black 5 (RB5) dye. The best conditions to prepare the magnetic-PEI-cellulose adsorbent were investigated and the results obtained for cellulose to PEI ratio is 2:1, impregnation time is 6 hours, crosslinking contact time is 60 mins, volume of glutaraldehyde is 1 ml and cellulose-PEI to magnetic nanoparticles ratio is 2:1:0.25. Then, the synthesised magnetic-PEI-cellulose was characterised using Fourier transform infrared spectroscopy, Brunauer Emmett Teller surface area, vibrating sample magnetometer and point of zero charge. A batch adsorption experiment was conducted and the results obtained were excellent, with almost 100% RB5 removal under the following conditions: 180 min of contact time, 0.1 g of adsorbent, 0.1 g/L of initial RB5 concentration, pH 7 and at 27°C. Kinetics, isotherm and thermodynamics evaluation were also performed for the adsorption data. The adsorption data fitted well to the pseudo second order model with the influence of intraparticle diffusion. For isotherm study, the data best fitted to the Langmuir model ($\chi^2 = 3.478E\text{-}09$) with the maximum adsorption capacity of 330 mg/g. A thermodynamics analysis shows that the adsorption was endothermic, random and spontaneous. The magnetic-PEI-cellulose is able to be regenerated and reused for 4 times with RB5 percentage removal above 70%. In conclusion, magnetic-PEI-cellulose was successfully demonstrated and can be used as a new promising adsorbent for the removal of dye from textile wastewater.

# ABSTRAK

Penggunaan kaedah penjerapan bagi menyingkirkan pencelup daripada air sisa buangan tekstil telah diiktiraf dan penjerapan pada karbon teraktif telah dikenali sebagai prospek terbaik bagi menyingkirkan pencelup yang mempamerkan warna di dalam air. Namun begitu, karbon teraktif adalah mahal dan sukar untuk proses penggunaan semula. Oleh itu, penjerap alternatif lain telah dikaji. Jelas diketahui bahawa bahan-bahan sisa dari pertanian boleh diperoleh dan digunakan sebagai penjerap murah. Dalam kajian ini, selulosa daripada tandan kosong kelapa sawit telah diubahsuai dengan polietilenaimina (PEI) dan zarah halus magnet melalui kaedah sambung-silang bagi menyingkirkan pencelup reaktif hitam 5 (RB5). Keadaan yang terbaik bagi penyediaan penjerap zarah halus magnet-PEI-selulosa telah dikaji dan hasil dapatan yang diperoleh bagi nisbah antara selulosa dan PEI ialah 2:1, masa impregnasi ialah 6 jam, masa sentuh sambung-silang ialah 60 minit, isipadu glutaraldehida ialah 1 ml dan nisbah antara selulosa-PEI dan zarah halus magnet ialah 2:1:0.25. Kemudian, penjerap zarah halus magnet-PEI-selulosa yang dihasilkan telah dicirikan menggunakan spektroskopi inframerah jelmaan Fourier, luas permukaan Brunauer Emmett Teller, magnetometer sampel bergetar dan titik cas sifar. Kajian penjerapan kelompok telah dijalankan dan hasil yang diperolehi sangat cemerlang dengan hampir 100% penyingkiran RB5 pada keadaan berikut; 180 minit masa sentuh, 0.1 g penjerap, 0.1 g/L kepekatan awal RB5, pH 7 dan pada 27°C. Kajian kinetik, isoterma dan termodinamik juga telah dijalankan menggunakan data penjerapan. Model pseudo tertib kedua adalah yang paling sesuai dengan data penjerapan dengan pengaruh resapan antara zarah. Bagi kajian isoterma, model Langmuir adalah yang paling sesuai ($\chi^2$ = 3.478E-09) dengan kapasiti penjerapan maksimum ialah 330 mg/g. Analisis termodinamik menunjukkan bahawa penjerapan adalah endotermik, rawak dan spontan. Zarah halus magnet-PEI-selulosa berupaya digunapakai sebanyak 4 kali dengan peratusan penyingkiran RB5 melebihi 70%. Kesimpulannya, zarah halus magnet-PEI-selulosa telah berjaya dibuktikan dan boleh digunakan sebagai penjerap baharu yang berkeupayaan bagi menyingkirkan pencelup daripada air sisa buangan tekstil.

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# LIST OF ABBREVIATIONS

| | | |
|---|---|---|
| RB5 | - | Reactive Black 5 |
| FTIR | - | Fourier Transform Infrared Spectroscopy |
| BET | - | Brunauer-Emmett-Teller |
| VSM | - | Vibrating Sample Magnetometer |
| BJH | - | Barret-Joyner-Halenda |
| PZC | - | Point of Zero Charge |
| UV-Vis | - | Ultraviolet-visible |
| n.a | - | not available |
| MNPs | - | Magnetic Nanoparticles |
| AC | - | Activated Carbon |
| GLA | - | Glutaraldehyde |
| HCl | - | Hydrochloric Acid |
| NaOH | - | Sodium Hydroxide |
| PEI | - | Polyethyleneimine |
| NaCl | - | Sodium Chloride |

# LIST OF SYMBOLS

% - Percentage
°C - Degree Celcius
$\Delta G$ - Gibbs Free Energy
$\Delta H$ - Enthalpy
$\Delta S$ - Entropy
$cm^{-1}$ - Reciprocal Centimeter
$cm^3$ - Centimeter Cubed
$\lambda max$ - Maximum wavelength
HCl - Hydrochloric Acid
NaOH - Sodium Hydroxide
$N_2$ - Nitrogen Gas
rpm - Revolution per minute
v/v - Volume per volume
g/L - Gram per Liter
$cm^3/g$ - Centimeter cubed per gram
min - Minute
g - Gram
$mol\ L^{-1}$ - Mol per liter
L - Litre
ml - Milliliter
nm - Nanometer
M - Molarity
mg/g - - Milligram per gram
mg/L - Milligram per liter
K - Kelvin

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# CHAPTER 1

# INTRODUCTION

## 1.1 Research Background

The development of industrialisation and rapid urbanisation has significantly contributed to the increasing of water pollution and has made this a serious restraint for a reasonable standard of urban living [1]. Textile industry is one of the most chemically severe industries which generate dye wastewater during the processes of dyeing and finishing. The dyes contained in the wastewater discharged from the textile industry are highly observable and potentially harmful for the environment and human life, even at low concentration (1.0 mg/L) [2].

There are several types of dyes including anionic (direct, acid, and reactive dyes), cationic (basic dyes), and nonionic (disperse dyes). Among them, anionic dyes are found to be the brightest class of the soluble dyes, which highly cause serious environmental and health problems [3, 4]. According to Gottlieb *et al.* [5], for humans, the maximal concentration of anionic RB5 allowed is 27.5 mg/L, which indicates that even a small exposure to these dyes can lead to undesirable consequences to human being. The consequences include direct and indirect toxic effects on humans as dyes are associated with cancer, jaundice, tumours, skin irritation, allergies, heart defects, and mutations. The adversity of the effects therefore strengthens the need of the removal of dye pollutants from textile industry wastewater [6, 7].

Some of the treatment methods include reverse osmosis, filtration, adsorption, chemical precipitation, coagulation, electroplating, evaporation, ion exchange, activated sludge, aerobic and anaerobic treatment are available to remove the pollutants [8-10]. Among them, adsorption technologies have proven to be an excellent and versatile method to remove dyes from textile wastewater due to its simplicity and feasibility.

In order to enhance the effectiveness of the adsorption processes, it is vital to develop cheaper adsorbents with higher adsorption capacity. Many efforts have focused on developing inexpensive adsorbent from natural or waste materials as it is abundantly available and easy to obtain. One of potential natural waste to be used as adsorbent is from agricultural waste. Agricultural waste is mainly made up of cellulose, hemicellulose and lignin. Cellulose has received high consideration for its properties such as possess high contents of hydroxyl groups, non-toxicity and safe disposability after us [11]. However, crude natural adsorbent such as cellulose has poor adsorption capacity, which limit their uses [12]. Therefore, modification of cellulose on its hydroxyl functional groups with some other functional groups such as carbonyl group [13], sulfonate group [14], phosphate group [15], and amino group [16] is necessary to increase its adsorption capacity.

Polyethyleneimine is water-soluble which composes plenty of amino groups on the polyamine chains. A large number of primary and secondary amino groups in a molecule show good ability of sorption when they are adsorbed on the adsorbent surface [17]. In addition, the existence of nitrogen groups on its molecular chains makes it easy to be positively charged, thus good for adsorption of pollutants that carry negative charge [18]. But, PEI cannot be directly used as adsorbent to treat wastewater because of the difficulty to achieve liquid or solid phase separation due to its strong water-solubility property. Therefore, to overcome the water-solubility property, PEI molecules are commonly be hybridised or coated on an insoluble solid support to act as an effective adsorbent.

Magnetic nanoparticle, $Fe_3O_4$ material has good magnetic separation character such as its easy phase separation with aqueous solutions. In past studies [18, 19], a magnetic based adsorbent was successfully developed via a simple modification method of chemical reaction using a crosslinker. The magnetic based adsorbent was used on removal of pollutants in wastewater and has showed excellent performance where the pollutants could be easily separated from aqueous solution due to the presence of superparamagnetic $Fe_3O_4$ carrier. Therefore, it is highly expected that the hybridization of $Fe_3O_4$ and PEI onto cellulose could act as good adsorbents for removal of pollutants from dyeing wastewater such as anionic dyes.

## 1.2 Problem Statement

There are several wastewater treatments such as adsorption, advanced oxidation, membrane separation, coagulation, and flocculation. Amongst all, the adsorption process is the most preferred treatment due to its low-cost, easy operation, flexibility, and simplicity [13] to remove synthetic dyes from wastewater [20]. Among all the adsorbent used in adsorption, activated carbon has been proven to be the most promising adsorbent due to its high adsorption efficiency compared to many other available adsorbents such as zeolites [6], silica gel [21], bacterial biomass and polymeric materials [22]. Consequently, a commercial AC has been found most widely used in textile wastewater for the treatment of dye waste [23].

Although commercial AC is highly efficient, it is not practicable to be used in treating dye-contaminated wastewater, especially in large scale waters [24]. The synthesis of activated carbon is relatively expensive where high temperature and energy are required. Besides that, the regeneration of exhausted AC is not easily possible on a commercial scale. Tedious procedures are required to separate the AC from the effluent since the AC is mostly used as fine powder and result in the loss of the adsorbent [25]. Consequently, various low-cost adsorbents derived from natural and waste materials were investigated in order to provide a competitive substitute for commercial AC in treating the coloured wastewater [26-28]. However, these low-cost adsorbents were either inefficient in adsorption capacity [29] or may cause more serious damage to the environment such as producing harmful by-products after adsorption process [30, 31].

More recently, with the development of nanotechnology, magnetic nanoparticles are being increasingly used in adsorption treatment due to its magnetic separation character of $Fe_3O_4$ which could be conveniently separated from aqueous solution by applying external magnetic field. Nonetheless, studies that have used MNPs to remove dye pollutants are still scarce and the studies mainly focused on the removal of heavy metal ions from wastewater. Besides that, the study on synthesis conditions of the adsorbent is also limited because most of them have focused on changing the type of material use only to increase adsorption efficiency. But, it is necessary to study the best conditions for synthesis the adsorbent because the reactivity of the materials is depend on the preparation process which will influence the adsorbent characteristics.

Thus, in order to fulfil the gap on the aforementioned drawbacks, this study was carried out to develop an effective magnetic-PEI-cellulose adsorbent at the best synthesis conditions via crosslinking method with characters of high adsorption capacity, fast kinetics, and easy separation. The performance of magnetic-PEI-cellulose adsorbent towards removal of synthetic dye, RB5 was also studied.

## 1.3 Objectives of Research

The objectives of the research are as follows:

1. To identify the best synthesis conditions of modified magnetic-Polyethyleneimine-cellulose adsorbent and its characterization.

2. To investigate adsorption performance of synthesized adsorbent (magnetic-PEI-cellulose) towards the removal of reactive black 5.

3. To identify the adsorption of kinetic, isotherm and thermodynamic of adsorbent.

4. To study the regeneration and reusability of the adsorbent.

## 1.4 Scopes of Research

The modified adsorbent of magnetic-PEI-cellulose was synthesized via crosslinking method and glutaraldehyde was used as a crosslinking agent. Several parameters were varied during synthesis process in order to find the best conditions of adsorbent preparation. The parameters involved were the effect of solid to liquid ratio between cellulose and PEI (1:1, 1:2, 2:1, 3:1), effect of impregnation time (30min-24hour), effect of crosslinking time (10-80min), volume of glutaraldehyde (0.5-10ml) and effect magnetic nanoparticle (1, 0.5, 0.25, 0.125, 0.0625) to cellulose:PEI ratio. Then, the adsorbent was characterized based on properties such as functional group and surface area using the FTIR, BET, VSM and PZC. The amino content on the adsorbent was determined using titration method with hydrochloric acid.

The adsorption performance was evaluated by varying of several parameters such as contact time (0 to equilibrium), initial dye concentration (0.025-0.3g/L), adsorbent dosage (0.05-2.0g), temperature (27-70°C) and pH solution (2-9). Then, the percentage RB5 removal was analysed using UV-Vis spectrophotometer.

The kinetic process was determined by fitting the adsorption data to pseudo first order, pseudo second order and intraparticle diffusion models. The adsorption isotherm was identified using Langmuir and Freundlich models. The thermodynamic was evaluated by identifying the enthalphy, entropy and Gibbs energy.

The regeneration of the adsorbent was investigated by identifying the best regenerants between HCl and NaOH at different concentration (0.1 M, 1.0 M and 5.0 M). The best regenerant was chosen based on the adsorption performance of the adsorbent for four cycles.

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