

**EXTRACTION, CHARACTERIZATION AND APPLICATION OF
CELLULOSE FROM PANDAN LEAVES AS GRAFTED FLOCCULANT
FOR DYES TREATMENT**

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EXTRACTION, CHARACTERIZATION AND APPLICATION OF CELLULOSE
FROM PANDAN LEAVES AS GRAFTED FLOCCULANT FOR DYES
TREATMENT

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*This project I dedicated to my beloved mother, siblings, lecturers and fellow friends.
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this project.*

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ABSTRACT

A new polyacrylamide grafted cellulose (PAM-g-cellulose) was successfully synthesised by using microwave assisted synthesis method. Cellulose was extracted from Pandan leaves by carrying out alkali and bleaching treatments. The determination of chemical composition was done which covers the percentage of lignin, hemicellulose and cellulose. Analysis of Scanning Electron Microscopy (SEM), Fourier Transform Infrared Spectroscopy (FTIR), X-Ray Diffraction (XRD) and Thermogravimetric (TGA) were conducted to the raw Pandan leaves and extracted cellulose. For grafting synthesis study, the highest grafting percentage was achieved at 170%. The best grafted sample (i.e. highest percentage) then was characterized using SEM and FTIR. The PAM was successfully grafted onto the backbone of cellulose as confirmed by SEM and FTIR results. The grafting mechanism involved a synergism of microwave radiation and ceric ammonium nitrate (CAN) as chemical free radical initiator to initiate the free radical grafting reaction. The flocculation studies of the developed grafted copolymers have been evaluated in synthetic dye (Reactive Black 5 (RB5)) suspension. Response surface methodology (RSM) and Box-Behnken design (BBD) were employed to explore the relationship of process variables (i.e. initial dye concentration, flocculant dosage and pH) on percentage reduction of initial dye concentration and COD level of RB5 dye as well as to find the optimum process condition. The flocculant dosage and pH were the most significant factors in affecting the degree of flocculation efficiency. The optimum condition for reduction of initial dye concentration and COD were achieved at initial dye concentration of 0.03 g/l, flocculant dosage of 0.06 g and pH 11.72. Under this condition, the reduction of initial dye concentration and COD were 23.51% and 54.24%, respectively.

ABSTRAK

Gabungan antara poliakrilamida dan selulosa (PAM-g-selulosa) telah berjaya disintesis dengan menggunakan kaedah yang dibantu oleh gelombang mikro. Selulosa telah diekstrak dari daun Pandan melalui rawatan alkali dan pelunturan. Komposisi kimia telah ditentukan yang merangkumi peratusan lignin, hemiselulosa dan selulosa. Analisis Pengimbas Mikroskop Elektron (SEM), Jelmaan Fourier Spektroskopi Inframerah (FTIR), Pembelauan X-Ray (XRD) dan Termogravimetri (TGA) telah dijalankan terhadap daun Pandan dan selulosa yang telah diekstrak. Di dalam kajian mengenai sintesis gabungan, peratusan gabungan tertinggi dicapai adalah sebanyak 170%. Sampel gabungan yang terbaik (i.e. peratusan tertinggi) kemudian telah dicirikan menggunakan SEM dan FTIR. PAM telah berjaya digabungkan dengan selulosa seperti yang telah dibuktikan oleh keputusan SEM dan FTIR. Mekanisma gabungan melibatkan sinaran gelombang mikro dan serium ammonium nitrat (CAN) sebagai bahan kimia radikal bebas untuk memulakan tindakbalas gabungan. Gabungan yang terhasil ditunjukkan oleh SEM dan FTIR di mana kedua-duanya menunjukkan perubahan pada mikrograf dan spektra. Kajian pengelompokan telah dijalankan terhadap pewarna sintetik (Reactive Black 5 (RB5)). Kaedah respon permukaan (RSM) dan reka bentuk Box-Behnken (BBD) telah digunakan untuk mengkaji hubungan antara pembolehubah (kepekatan awal pewarna, dos flokulan dan pH) dengan peratusan penurunan kepekatan awal pewarna dan tahap COD pewarna RB5 serta untuk mencari keadaan optimum proses. Dos flokulan dan pH adalah faktor yang paling penting dalam mempengaruhi tahap kecekapan pengelompokan. Keadaan optimum untuk mengurangkan kepekatan awal pewarna dan COD dicapai apabila kepekatan awal pewarna adalah sebanyak 0.03 g/l, dos flokulan sebanyak 0.06 g dan pH 11.72. Dalam keadaan ini, pengurangan kepekatan awal pewarna dan COD adalah masing-masing 23.51% dan 54.24%.

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LIST OF ABBREVIATIONS

AM	-	Acrylamide
Ag	-	Agar
ANOVA	-	Analysis of variance
AGU	-	Anhydroglucose
AOX	-	Alternative oxidase
BBD	-	Box-behnken design
BOD	-	Biological oxygen demand
CAN	-	Ceric ammonium nitrate
CCD	-	Central composite design
CMG	-	Carboxymethyl guar gum
CMS	-	Carboxymethylstarch
COD	-	Chemical oxygen demand
C.I.	-	Colour index
DF	-	Degree of freedom
EFB	-	Empty fruit bunch
EDA	-	Ethylene diamine
FTIR	-	Fourier transform infrared
g	-	Graft
GGI	-	Gum ghatti
H ₂ O ₂	-	Hydrogen peroxide
Hyd.	-	Hydrolyzed
O ₃	-	Ozone
MnO ₄	-	Permanganate

PAM	-	Polyacrylamide
PAC	-	Poly (aluminium chloride)
Psy	-	Psyllium
RB5	-	Reactive black 5
RBF	-	Round bottom flask
RSM	-	Response surface methodology
SEM	-	Scanning electron microscopy
SAG	-	Sodium alginate
NaClO ₂	-	Sodium chlorite
NaOH	-	Sodium hydroxide
NaOCl	-	Sodium hypochloride
St	-	Starch
TAM	-	Tamarindus indica mucilage
TAPPI	-	Technical Association of Pulp and Paper Industry
TDS	-	Total Dissolve solid
TGA	-	Thermogravimetric analysis
TSS	-	Total Suspended solid
TKP	-	Tamarind kernel polysaccharide
UV/Vis		Ultraviolet-visible
XRD		X-ray diffraction

LIST OF SYMBOLS

θ	-	Angle
%G	-	Grafting percentage
$-\text{SO}_2-\text{CH}_2\text{CH}_2-\text{SO}_3-$	-	Sulphatoethylsulphone
$-\text{N}=\text{N}-$	-	Two nitrogen atoms
λ	-	Wavelength

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CHAPTER 1

INTRODUCTION

1.1 Research Background

Textile industry is one of the largest water and chemicals consumer industries in Malaysia (Kumar *et al.*, 2011; Subki and Hashim, 2011). Textile industry in Malaysia is concentrated mainly in the states of Johor, Penang and Selangor (Muyibi *et al.*, 2008). Generally, textile wastewater is characterized with high in colour, biological oxygen demand (BOD), chemical oxygen demand (COD), total dissolve solid (TDS), total suspended solid (TSS), pH and temperature (Oke *et al.*, 2006). Although most of the textile factories are equipped with treatment facilities, the problems still persist since all the available treatment methods have own positive and negative aspects. Reife and Freeman (1996) stated that, the details of the wastewater characterization are important step in selecting wastewater treatment methodologies.

The discharge of textile wastewater contains various pollutants such as sulphur, salts, toxicants, degradable organics and colour. These pollutants are generated during various stages of textile manufacturing process. The processes include, desizing, scouring, bleaching, mercerizing, dyeing, printing and finishing (Verma *et al.*, 2012). These processes led to the alteration of chemical structure in the discharge effluents which then result in the toxicity of the wastewater (Koyuncu, 2003; Ahmad and Puasa, 2007).

The important material used in textile industry is dyes. Dyes are materials that impart appearance of the fabrics. Dyes give a good attraction to the fabrics through the colours that being created. At the same time, dyes are the biggest contributor of the highly toxic discharge of textile wastewater. This important material further is in charge of all the characteristics of textile wastewater. Dyes can give a high negative impact towards textile wastewater characteristics such as chemical oxygen demand (COD), suspended solids, colour and pH. Those characteristics are significant in evaluating textile wastewater quality (Chung *et al.*, 1992; Tufekcu *et al.*, 2007; Saratale *et al.*, 2011). Dyes are considered as xenobiotic compounds that are very recalcitrant to biodegradation. Besides that, the absorption of light due to textile dyes creates problem to photosynthetic aquatic plants and algae. The reduction of photosynthesis deteriorates the water quality and lowers the gas solubility mainly in aqueous ecosystem (Anjaneyulu *et al.*, 2005; Saeed *et al.*, 2010).

There are several technologies that have been developed in solving textile wastewater problems. The technologies include physical, chemical and biological treatment methods. However, the requirements for every treatment process are for it to be efficient, simple, low cost and environmentally friendly. Therefore, every limitation of the readily available processes is extensively worked on in order to provide some improvement. There are various continuing researches carried out to develop new technologies of textile wastewater.

1.2 Problem Statement

Textile wastewater contains a wide range of dyes which give rise to serious problems to the environment and human life. Dyes contribute to the negative characteristics of textile wastewater such as high in BOD, COD, colour, TSS and etc. Among all dyes treatment in textile wastewater such as adsorption, oxidation, coagulation and/or flocculation, biosorption and membrane technologies, flocculation process is in demand because of its effectiveness and economical characteristics.

In flocculation process, flocculant is the important chemical used to increase the flocculation efficiency. There are two categories of flocculants which are inorganic and polymeric. Inorganic flocculants are needed in large dosage and thus the production of sludge is high. In comparison, polymeric flocculants are extensively used because the dosage and the production of sludge are less than inorganic flocculant.

Apart from that, polymeric flocculants can be further classified into natural and synthetic. Natural flocculants are low in cost and have good biodegradability. However, the biodegradability of these flocculants limits their storage life and flocculation performance. Synthetic flocculants on the other hand, has high efficiency in flocculation process. But still, the biggest limitations of these flocculants are on their shear degradability that can be considered to be hazardous to the environment and human health.

Another class of polymeric flocculants which are grafted copolymers flocculant have been synthesized by a number of researchers, which combine the advantages between natural and synthetic flocculants. Besides that, the disadvantages of both types can be diminished or fully terminated. Grafted

copolymers flocculant has been claimed that can increase the flocculation efficiency besides being professed as eco-friendly flocculant technology.

This flocculant has been studied by many researchers by means of finding the best grafted flocculant in order to solve wastewater problems. The findings were led to various treatment performances. The performance was based on the type of the wastewater and also the materials used (natural and synthetic) to synthesize the grafted flocculant. Therefore, it is necessary to choose the suitable materials to achieve a good flocculation performance.

This study was aimed to synthesize a new grafted copolymers flocculant with high flocculation efficiency in treating textile wastewater. The flocculant was synthesized by grafting cellulose derived from Pandan leaves and polyacrylamide (PAM). Cellulose has been extracted from several other sources. Extracted cellulose from Pandan leaves was targeted as excellent natural polymer to be grafted with PAM. The effectiveness of this synthesized flocculant was investigated for the first time in treating Reactive Black 5 (RB5) dye. The flocculation efficiency was defined through percentage reduction of initial dye concentration and COD level.

1.3 Hypothesis

The hypothesis for this research is that new grafted copolymers flocculant is more superior compared to inorganic flocculants and solely natural and synthetic polymeric flocculants in treating textile wastewater. This grafted copolymers flocculant has the advantages from both natural and synthetic polymer flocculants because of the extent of the attachment of both materials onto their backbone. Moreover, the findings will provide an alternative for a low cost, environmentally friendly textile wastewater treatment.

1.4 Objectives

This study aims to achieve the following objectives:

- i. To extract cellulose from Pandan leaves (*Pandanus amaryllifolius* Roxb.)
- ii. To synthesize and characterize the polyacrylamide grafted cellulose (PAM-g-cellulose)
- iii. To investigate the effectiveness of synthesized polyacrylamide grafted cellulose (PAM-g-cellulose) as a flocculant for Reactive Black 5 (RB5) dye in textile wastewater

1.5 Scope

In order to achieve the objectives, specific scopes have been set which limit the range of the study. This study was firstly, emphasized on extraction of cellulose from Pandan leaves (*Pandanus amaryllifolius* Roxb.). The cellulose was extracted using chemical treatment method which was alkali treatment and bleaching. After that, the extracted Pandan leaves were characterized by means of chemical composition (using TAPPI method), morphology (using SEM analysis), functional group (using FTIR spectroscopy analysis), crystallinity (using XRD analysis) and thermal stability (using TGA).

The second part of the study covers the synthesis of grafted copolymers flocculant. This part was carried out by grafting the polyacrylamide (PAM) with the extracted cellulose using microwave assisted method. The best grade of grafted copolymers (higher percentage grafting (%G)) then was characterized using SEM analysis and FTIR spectroscopy. Further, the mechanism of the developed flocculant was also investigated in this study.

The performance of the grafted copolymers (PAM-g-cellulose) as a flocculant was tested in flocculation process using a standard jar test. For jar test conditions, the mixing rate was fixed at 200 rpm for rapid mixing (3 minutes) and 30 rpm for slow mixing (30 minutes). The supernatant after the flocculation process was allowed to settle for 30 minutes. Three independent variables including initial dye concentration (g/l), flocculant dosage (g) and pH of the wastewater were used. The numerical values for all three variables have been specified through response surface methodology (RSM) method.

In order to produce the experimental design specifying all conditions, STATISTICA software version 8.0 was used. The range for the initial dye concentration is 0.025 to 0.1 g/l, 0.02 to 0.1 g for dosage of flocculant and 3 to 11 for pH. The fixed conditions and the ranges were selected according to the preliminary studies done and from several literature reviews from previous works. The treated wastewater was analyzed for percentage reduction of initial dye concentration and chemical oxygen demand (COD). To distinguish the superiority of the grafted copolymers flocculant, the test was also performed using solely Polyacrylamide (PAM) and cellulose extract using the obtained optimum conditions from RSM.

1.6 Significance of Study

There are several reasons for the execution of this research. Firstly, the selection for Pandan leaves (*Pandanus amaryllifolius* Roxb.) is due to its availability (especially in Malaysia), low cost of materials and simplicity in handling. In Malaysia, the interests in Pandan leaves are only focused to food additives and food colorant. The potential of Pandan leaves as a flocculant has never been explored by any researcher. Besides that, the study of cellulose extracted from Pandan leaves also has not been conducted yet.

In recent years, considerable attention has been paid on the synthesis of grafted copolymers that combines the advantages of both natural and synthetic polymers. Thus, a great number of graft copolymers has been synthesized by grafting synthetic polymers, mainly, flexible chain of polyacrylamide (PAM) on some natural polymers backbone, such as gum guar (Wang *et al.*, 2007), starch (Chang *et al.*, 2008), konjac glucomannan (Tian and Xie, 2008), chitosan (Zhang *et al.*, 2010), carboxymethylstarch (Sen, *et al.*, 2009) and tamarind kernel (Ghosh *et al.*, 2010).

However, there is no work been reported on the grafting of synthetic polymers (i.e. polyacrylamide) onto cellulose extract from Pandan leaves (*Pandanus amaryllifolius* Roxb.) to date. Moreover, cellulose extract from Pandan leaves grafted with synthetic polymers has never been used as a flocculant to treat any type of wastewater. In addition, a new alternative of grafted copolymers flocculant to treat wastewater was developed through this study.

1.7 Thesis Outline

There are 5 chapters in this thesis and each chapter describes the sequence of this research.

Chapter 1 presents brief overview of wastewater from textile industry and common method used to treat wastewater from textile industry. This chapter also presents the problem statement, hypothesis, research objectives, scopes of the study and the significance of the study.

Chapter 2 covers the deep view of related knowledge about textile industry wastewater, dyes and the available treatment methods to treat dyes in textile wastewater. After that, this chapter covers the previous studies of grafted copolymers as flocculant with the available grafting methods. Finally, the chapter was end up with the literature of cellulose characteristics and method uses to extract cellulose from natural sources.

Chapter 3 refers to the materials and methods used in the study. The chapter described the experimental procedure that being used in this research for the extraction of cellulose from Pandan leaves, preparation of the grafted copolymers flocculant (PAM-g-cellulose) and the flocculation preparation and process via RSM to treat Reactive Black 5 (RB5) dye.

Chapter 4 presents all of the results obtained in the study which cover the extraction study of cellulose from Pandan leaves, physical study on the synthesized grafted copolymers flocculant (PAM-g-cellulose) and also the performance of the developed flocculant to treat Reactive Black 5 (RB5) dye using RSM.

Chapter 5 refers to overall conclusions that are based on the findings obtained in the results and discussion which explained in Chapter 4. Besides that, the recommendations for future research were also given in this chapter to improve the structure of the study and also the findings.

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