# Study on Colour Removal of Organic Dyes (Methylene Blue and Congo Red ) by Using Coconut Husk

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#### Abstract

In the present research, the plant-base waste material coconut husk was prepared to be used as adsorbent for the removal of organic dyes (methylene blue and congo red) from aqueous solution. The coconut husk powder (CHP) was used as an adsorbent in this research. The physicochemical properties of collected coconut sample such as pH (7.23), moisture content (18.50%), bulk density (0.40) and ash content (8.00%) were observed. The coconut husk powder was characterized by modern techniques such as SEM and FT IR. Surface morphology of coconut husk samples were observed. The adsorption capacity of prepared samples were determined with the different parameters such as initial concentration, contact time and dosage. The dye removal of coconut husk samples were studied by using model dyes (methylene blue and congo red ). The maximum percent removal of dye was found by using initial concentration of 10 mgL<sup>-1</sup>, 0.1 g of dosage and 1 h contact time. The removal percent of methylene blue (MB) and congo red (CR) by coconut husk were found to be (97.65%) and (71.87%), respectively. It was found that the coconut husk were cheap and low cost effective biosorbent for the removal of organic dyes from aqueous solution.

Keywords: Coconut Husk, Organic dyes, Sorption, SEM, FT-IR, Methylene Blue, Congo Red

#### Introduction

The coconut tree (*Cocosnucifera*) is a member of the family *Arecaceae* (palm family) and the only species of the genus *Cocos*. The term coconut can refer to the entire coconut palm or the seed, or the fruit, which, botanically, is a drupe, not a nut. Coconuts are known for their great versatility, as evidenced by many traditional uses, ranging from food to cosmetics. They form a regular part of the diets of many people in the tropics and subtropic. Coconuts are distinct from other fruits for their large quantity of "water", and when immature, they are known as tender-nuts or jelly-nuts and may be harvested for their potable coconut water. When dried, the coconut flesh is called copra. The oil and milk derived from it are commonly used in cooking and frying, as well as in soaps and cosmetics. The husks and leaves can be used as materials to make a variety of products for furnishing and decorating. Figure (1) shows the photograph of coconut fruits and coconut husk samples (Pearsall, 1999).





Figure (1) Coconut fruits and coconut husk samples.

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### Adsorption

Adsorption is a process in which the molecules or atoms of one phase interpenetrate nearly uniformly among those of another phase to form a solution with the second phase. The term "sorption", which includes both adsorption and absorption, is a general expression for a process in which the second phase in solid (Win Yin Cho Nwe *et al.*, 2010).

# **Types of Adsorption**

### Physisorption (or) Physical Adsorption

This is the type of adsorption in which the adsorbate adhere to the surface only through van der Waals (weak intermolecular) interactions. The forces responsible for the non-ideal behaviour of real gas.

#### **Chemisorption (or) Chemical Adsorption**

This is the type of adsorption whereby a molecule adheres to a surface through the formation of a chemical bond. The forces opposed to the van der Waals force (Malik, 2003).

### Adsorbent

Adsorbent is a substance, such as activated charcoal, that takes up another by the process of adsorption, as by the attachment of one substance to the surface of the other.

#### **Methylene Blue**



Figure (2) Structure of Methylene blue.

Methylene blue (3,7bisDimethylamino-phenazathionium chloride)

Molecular formula -  $C_{16}H_{18}N_3SCL$ 

Molecular mass -  $319.85 \text{gmol}^{-1}$ 

Methylene blue, also known as methylthioniumchloride, is a medication and dye. The structure of Methylene blue is shown in figure (2). Methylene blue is highly stable in the human body, and if ingested, it resists the acidic environment of the stomach as well as many hydrolytic enzymes present. It is not significantly metabolized by the liver, and is instead quickly filtered out by the kidneys. Therefore, it is necessary to make sure the effluent contained methylene blue was treated first before being released to the environment (Sharifah, 2006).

## **Congo Red**

Congo red is the sodium salt of  $3,3'-([1,1'-biphenyl]-4,4'-diyl)bis(4-aminonaphthalene-1-sulfonic acid) (formula: <math>C_{32}H_{22}N_6Na_2O_6S_2$ ; molecular weight: 696.66 g/mol). It is a secondary diazo dye. Congo red is water-soluble, yielding a red colloidal solution; its solubility is better in organic solvents such as ethanol. The structure of Congo Red is shown in figure (3).

It has a strong, though apparently noncovalent, affinity to cellulose fibers. However, the use of Congo red in the cellulose industries (cotton textile, wood pulp, and paper) has long been abandoned, primarily because of its toxicity and tendency to run and change color when touched by sweaty fingers (Tezcanli, 2003).

Properties

Chemical formula -  $C_{32}H_{22}N_6Na_2O_6S_2$ 

Molar mass-696.665



Figure (3) Structure of Congo Red.

# **Removal of Organic Dyes by Adsorption Process**

Dyes are widely used in textile, paper, plastic, food and cosmetic industries. The wastes coming from these industries can effect on our atmosphere causing pollution. The level of the pollutants even in very low concentration is highly visible and will affect aquatic life as well as food web. Many dyes are difficult to degrade. They are generally stable to light, oxidizing agents and are resistant to aerobic digestion. Contaminations due to dyes pose not only a severe public health concern, but also many serious environmental problems because of their persistence in nature and non-biodehradable characteristics. The number of conventional methods are available for colour removal from industrial effluents including ion exchange, adsorption, membrane technology and coagulation (Tahir *et al.*, 2008).

Effectiveness of physicochemical methods varies and depends on the type of dyes being eliminated. Each dye is, in most cases, a complex organic compound. It has an electronegective nature depending on pH of the solution. Total elimination of dyes can be achieved by the application of coagulation together with sorption. The coagulation should be performed through the addition of two reagents in a specifically determined proportioning sequence. The dyes from the solution are precipitated together with a sediment which is a product of a reaction between the reagents (coagulants) supplied to the solution (Anielak, 1996).

Synthetic dyes present in watercourses is aesthetically unacceptable and may be visible at the concentration as low as 1ppm. Moreover, they may also affect photosynthetic activity in aquatic systems by reducing light penetration. Due to low biodegradability of dyes, a conventional biological treatment process is not very effective in treating a dye wastewater. The adsorption of the molecules onto various adsorbents is an ideal option for decolourization, which is evidenced by the effectiveness of adsorption for various dye types (Benaissa, 2005).

### **Materials and Methods**

Coconut husks were collected from Hinthada Township, Ayeyawady Region. After the collection, they were cut into small pieces. The air-dried coconut husks were washed thoroughly with tap water, sun dried at ambient temperature and ground into fine powder. Dried coconut husk powder was sieved into the size of  $100\mu m$ . The powder was then soaked and washed severally with cold tap water until clean. Clean coconut husks powder was soaked for 48 hours in distilled water, and then the samples was filtered and rinsed with distilled water, then dried under the air. After all, the samples were stored in plastic containers (Nabilah *et al.*, 2011).

The prepared samples of coconut husk powder were used to determine the physicochemical properties such as moisture content, pH, bulk density and ash content. These samples were characterized by using SEM, FT IR analysis.

A stock solution of 100 mgL<sup>-1</sup> was prepared by dissolving 0.1 g of dye powder in 1000 mL distilled water. Dye solutions of different concentrations were then prepared by dilution on the stock solution with distilled water. The dye solution, Methylene Blue (MB) and congo red were studied. Analyses were carried out by colorimetric method using 2550, U.S.A Spectrophotometer and calibration curve of dye stuff solution was plotted. Based on the calibration curve or by using absorption coefficient ( $\epsilon$ ), concentration of dye solution was calculated. Using the equilibrium contact time the nature sorption properties of various dyes were evaluated. Exactly 25 mL of dye solution of known initial concentration was mixed with a required dose of coconut husk powder sample in the flask. The flask was placed on a magnetic stirrer at room temperature and was stirred for 1 hour. After one hour the samples were filtered and the absorbance measured. The percentage of colour removal of coconut husk powder was also calculated. Figure (4) shows coconut husk samples before and after sun dried (Ming, 2011).



Figure (4) Coconut husk samples (before and after sun dried).

### **Results and Discussion**

The physicochemical properties of coconut husk powder, the moisture content, pH, bulk density and ash content of samples were determined. The bulk density was 0.40gm<sup>-3</sup>, the material is apparently porous and has a higher surface area of particle. The moisture content was observed 18.50%. In general, the ash content is attributed to show non-volatile inorganic minerals. The ash content 8.00% and the pH value 7.23 were observed.

Figure (5) shows the effect of dosage on removal of methylene blue and congo red by coconut husk powder samples. It was observed that the removal efficiency (0.02 to 0.12) increases with the little decrease of adsorbent dose in methylene blue and congo red. The lower percent removal may be attributed to the normal quantity of sorbent dose being used. This is due to the increase in adsorbent dose attributed to increase in surface area and availability of adsorption site. From those data, MB was more effective than CR by using coconut powder. The optimum sorbent dosage was found to be 0.1 g for both dyes.

Figure (6) shows the effect of contact time on the removal of methylene blue. The initial concentration of 10 mgL<sup>-1</sup> dye solutions of congo red and methylene blue, 0.12 g of coconut husk powder (CHP) was used and then the solutions were equilibrated. The removal percent, a little increases as the contact time increases. After the equilibrium contact time (60 min), the percent removal is nearly constant. The percent removal of CR (78.73%) is less than that of MB (98.27%).

Figure (7) shows the effect of initial concentration of dyes (MB) and (CR) on the coconut husk powder samples. The initial concentrations were varied from 10 mgL<sup>-1</sup> to 50 mgL<sup>-1</sup>. According to the experimental data, the removal percent decreases as the initial concentration increases. It was observed that the removal percent of MB was greater than that of CR.



Figure (5) The effect of dosage on removal of MB and CR by coconut husk powder samples.



Figure (6) The effect of contact time on the removal of MB and CR coconut husk powder samples.



Figure (7) The effect of initial concentration of dyes MB and CR coconut husk powder samples.

Silicon grease was coated on the brass stub at first. Then the sample was poured (spread) onto the grease and the stub was inserted into the ion sputter for gold coating on the sample. The stub with gold-coated specimen was placed in the sample holder and put into the Scanning Electron Microscope. The permanent records were obtained by the photographing of the sample crystals. The SEM micrograph is shown in Figures (8, 9 & 10).



Figure (8) SEM micrograph of coconut husk powder samples.



Figure (9) Methylene blue with coconut husk samples.



Figure (10) Congored with coconut husk samples.

FTIR spectra of coconut husk samples were recorded on 1% KBr pellet using fourier transform infrared spectrometer. The FT-IR spectra of coconut husk powder were shown in Figure (11) and the assignments of FT IR spectra for coconut husk powder was recorded in Table (1).



Figure (11) FT IR spectra of coconut husk powder sample.

<b>Observed Frequency of CHP</b>	Literature Frequency Range	Band Assignment
( <i>cm</i> <sup>-1</sup> )	( <i>cm</i> <sup>-1</sup> )	
3412.19	3500-3200	UOH
		Stretching
2922.25	3000-2850	υ <sub>OH</sub>
		Saturated
		/unsaturated
1730.21	1738-1700	C=O
		Stretching
1620.26	1650-1580	N-H
		Bending
1375.29	1440-1320	$\delta_{CH}$
		Benching
1255.70-825.56	1300-800	U <sub>C</sub> -0
		stretching alcohol

Table (1) FT IR Spectral Assignments of CHP.

CHP = Coconut Husk Powder

# Conclusion

The coconut husk powder sample in this investigation showed that it can be used as an effective biosorbent for the colour removal of dye from aqueous solution.

The determination of physicochemical properties such as the bulk density of coconut husk powder was found to be  $(0.40 \text{ gcm}^{-3})$ , the moisture content (18.50%) and the ash content (8.00%) were observed. The observed pH value was 7.23 for coconut husk powder.

The characterization of prepared sample by SEM and FT IR were able to reveal the surface morphology and spectral data of coconut husk powder.

In the effect of initial concentration, it was observed that the higher the concentration becomes, the fewer the available sites of adsorption and hence adsorption capacity decrease.

In the effect of contact time, the adsorption capacity increases with the increasing contact time and the equilibrium contact time reach about 60 min.

In the effect of adsorbents dosage, the adsorption capacity increase with the increasing dosage of sorbent.

From these results, the advantages of this project are being free from the difficulties of plucking and cleaning the coconut fruit, and then peeling the husk. In contract to, the color removal of methylene blue was more excellent than congo red by the coconut husk powder samples. It can be concluded that coconut husk powder can be served as an eco-friendly sorbent material in the color removal of dye from industrial waste water.

#### Acknowledgements

We would like to express our deep appreciation to Dr Tin Htwe, Rector of Hinthada University and Dr Ma Lar, Pro-Rector of Hinthada University, for their kind permission to carry of this research. We also wish to express our profound gratitude to Dr Cho Cho Than, Professor and Head, Departement of Chemistry, Hinthada University, for her numerous invaluable suggestions, kind encouragement and comments without which this work would not have been completed.

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