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Eco-Friendly Nanocellulose Filter Paper: A Potential for Removal of Dye from Textile Wastewater

Noor Izzatul Husna Ishak[1], Hazren A. Hamid[1]*

[1]Department of Civil Engineering Technology, Faculty of Engineering Technology, Universiti Tun Hussein Onn Malaysia, 86400, Pagoh, Johor, MALAYSIA.

*Corresponding Author Designation

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Abstract: Textile wastewater has now become one of the largest producers of wastewater as the demand for that product increase. As a result, the high concentration of the dyes resulting from the process will cause harm towards the surrounding environment including humans, animals and the ecosystem. There are vary method of treatment such as filtration and adsorption in treating wastewater. The objectives of this study are to investigate the performance of the filter paper towards textile wastewater as well as to determine the optimum dosage of filter paper using filtration method. Different dosage and pH is used in order to find the best performance of filter paper through filtration. Then, it is significant in distinguishing and investigating the functional group present in CNFs filter paper via Fourier Transform Infrared Spectroscopy (FTIR). From the result obtained, the optimum pH and dosage removal of colour from textile wastewater samples are found to be at pH 9 and dosage of 60. The final parameter including turbidity recorded is 5NTU, the COD value is 52 mg/L while the colour is 64 ADMI after using the filter paper. If we compare the value of those parameters with the Acceptable conditions for discharge of Industrial Effluent or Mixed Effluent of Standard A and Standard B, they are all can be categories under Standard A. This show that the use of filter paper in removing the dye effluent in textile wastewater is applicable. However, some improvements need to be taken in order to create a better efficiency of filter paper performance.

Keywords: Filtration, Filter Paper, Dye Removal

1. Introduction

Water is an essential thing that has contributed continuously to the whole natural ecosystem. Water sources can be either rivers, lakes, reservoirs or groundwater. As the years passed, the population of humans increased which triggered water usage to increase as well. Based on a statement in the UN World Water Development Report 2019, there has been an increment of 1% per year in the use of freshwater worldwide resulting from population growth and socio-economic development [1]. The water demand is predicted to continue rising at a similar rate until 2050, accounting for an increase of

**Corresponding author: norhazren@uthm.edu.my*

20 to 30% exceeding the current level of water use [1]. However, water pollution has been described as a global problem that leads to a shortage of freshwater. Water pollution has become a major problem which causes the scarcity of useful water which causes the demand for developing eco-friendly wastewater treatment technology [2].

Textile industries are one of the fastest-growing industries that provide numerous employment for millions of people globally [3]. The global textile industries produce daily products such as textile yarn, fiber, fabric and finishing products including apparel [4]. The high demands that aim to fulfil the needs of the people increase the rate of product production and manufacturing process. Initially, most of the materials used is natural fibers such as cotton, wool and silk that met the requirements for human consumption [4]. Apart from that, humans also use synthetic dyes which have been manufactured from organic molecules [5]. Most of these synthetic dyes are widely used in the field of painting, textile and printing [5].

The 2019 Toxic Textiles report has stated that almost 43 million tons of chemicals are used to dye clothes each year with 8000 different chemicals used in manufacturing the clothes. In producing textile products, almost 60 – 80% of the chemical used for coloring is from azo dyes **[**6]. Chemicals tend to release easily as it is easy to come off fabrics leading to harmful diseases such as cancer or dermatitis [10]. The dye pollutants that are being discharged into the river also contribute to the degradation of the aesthetic quality of the environment. Moreover, sunlight cannot fully enter the water as the dye pollutants on the surface of the water will absorb the sunlight. It will disturb the growth of the bacteria as well as the photosynthesis rate of the aquatic plants [7]. Thus, to minimize the impact of textile wastewater, a sustainable alternative or procedure should be taken. The most favorable method is by adsorption with the presence of different cellulose nanofibrils (CNFs) dosages made up of burflower-tree (*neolamarckia cadamba*)[8]. The usage of these materials is advantageous as they are from renewable material sources, have high mechanical strength and a high aspect ratio [9].

This study is conducted to investigate the performance of the filter paper towards the textile wastewater as well as to determine the optimal dosage of filter paper. Different dosage and pH is used in order to find the best performance or optimal dosage and pH for filter paper through filtration. Then, it is significant in distinguishing and investigating the functional group present in CNFs filter paper via Fourier Transform Infrared Spectroscopy (FTIR).

There have been numerous studies focused on the removal of dye effluent. However, previous studies show minimal work on the manufacturing of CNFs into filter paper from Malaysian renewable forest resources to be used as filter paper to eliminate dye pollutants in wastewater treatment methods and this will be the research gap in this study.

2. Materials and Methods

There are a few procedures that should be considered in completing this research on the filtration of wastewater by using cellulose nanofibrils (CNFs). Firstly, the materials of adsorbent and filter paper are selected. After that, the CNFs adsorbents are prepared before collecting and interpreting data for textile dye effluent. The characterization of the filter paper is analyzed to obtain the functional group present on it. The characterization for both before and after the filtration of the textile wastewater sample process was conducted before being compared. Additionally, some analysis of the parameters was conducted which includes pH, colour removal, total suspended solids (TSS), and Chemical Oxygen Demand (COD). The next step is conducting a filtration experiment using a real wastewater sample. The whole procedure is presented in **Figure 1**.

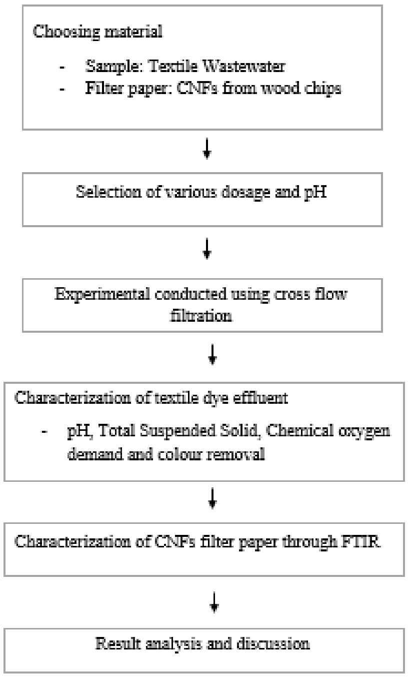

Figure 1: The research methodology carried out in this study

2.1 Design Experimental Planning

There are a few procedures that should be followed to complete this study. The sample of wastewater was taken at Textile Industry, located in Senai, Johor. Then, the filter paper made up of cellulose from *Neolamarckia cadamba* was taken at Forest Research Institute Malaysia (FRIM).

2.2 Selection of various variables and ranges

The selection of the independent variables and ranges were made to obtain the optimal dosage and pH in filtrating the wastewater sample. The selection of the variables is presented as shown in the **Table 1** below.

Table 1: The selection of the independent variables and ranges

Dosage of filter paper (gsm)	pH
50	4
	7
	9
60	4
	7
	9

2.3 Experimental Set Up

Identifying key parameters that influence the filtration of CNFs filter paper is the most critical phase in the design of the experiment (DoE) technique. The permeability test was conducted on CNFs filter paper using distilled water at various pressure.

2.4 Analytical Method

After undergo the cross flow filtration, the sample will experience the analytical method involving normalized flux analysis and colouring removal. The formula used for calculating the filter paper fouling behavior and colour removal of the wastewater sample is as follows:

$$Normalized\ flux = \frac{J}{J_0} \qquad Eq.1$$

$$Colour\ Removal\ (\%) = \frac{C_0 - C_1}{C_0} x\ 100\% \qquad Eq.2$$

Where C_0 is the initial dye concentration while the C_1 is the final dye concentration.

3. Results and Discussion

This section explained about the results and discussion are made after the experiments and analysis conducted.

3.1 Permeability study

The performance of the CNFs filter paper is measured by letting the distilled water to run through it at different pressure range from 1-5 bar which is known as water flux. The analysis is conducted using the cross-flow filtration system. The dilution used in this study is fixed at 100% of wastewater sample. The data trend in **Figure 2** shows that one variable increases as the other increases. The water flux is increasing moderately as the pressure is added.

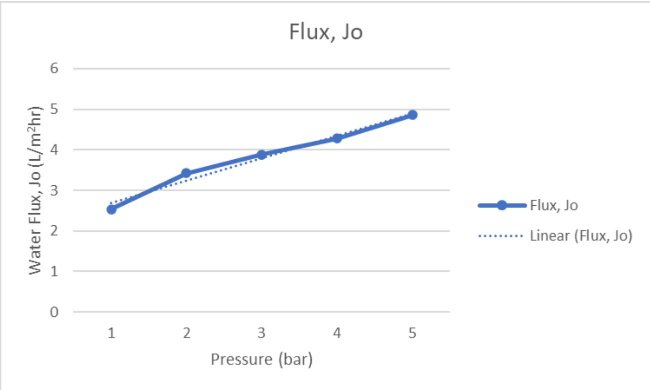

Figure 2: Graph of the water flux against the pressure

Meanwhile, the data presented in **Table 2** shows the data from the water permeability study of the optimum condition using a dosage of 60. As the pressure increased, the shortage time took the water flux to reach 2mL. Generally, the higher water pressure will trigger greater water flow. In addition, filter paper is a specialty paper with a high level of added value that has extremely high permeability, tiny pores, and high porosity. The result of the water flux for dosage of 60 gsm and pH 9 are as below in **Table 2**:

Table 2: Water flux at dosage of 60 gsm and pH 9

No	Pressure	Volume	Time (sec)	Time (h)	Flux, Jo
1	1	0.002	1380	0.383333	2.5327
2	2	0.002	1020	0.283333	3.4266
3	3	0.002	900	0.25	3.8835
4	4	0.002	816	0.226667	4.2833
5	5	0.002	720	0.20	4.8544

3.2 Results of the effect of dosage

The effect of dosage was investigated on the filtration of the CNFs filter paper by using 500 mL of textile wastewater for each sample with different pH and constant dilution of 100. From the data, the optimum dosage of this study is 60. This is because both the colour removal and turbidity removal percentage are high. It cannot be denied that the removal in dosage 80 is more effective. However, using this dosage, it is highly cost which will hardly attract industries to apply this filtration concept. The differences between initial and final filter paper can be observed in **Figure 3** after undergo the filtration process.

Table 3: The initial and final colour removal of dosage

Quantity of dosage, gsm	Initial colour concentration, ADMI	Final colour, ADMI	Colour removal (%)
50	1706	153	91
60	1706	134	96
80	1706	17	99

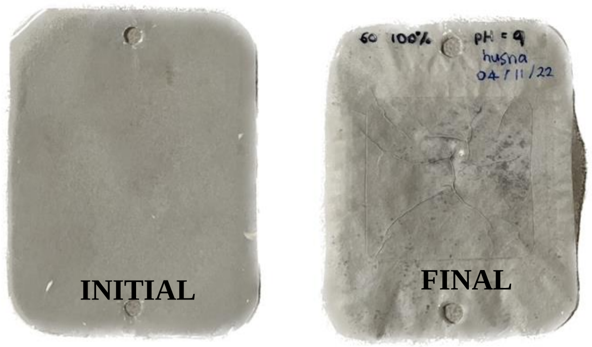

Figure 3: The differences between initial and final filter paper after filtration

3.3 Results of the effect of pH

The effect of dosage was investigated on the filtration of the CNFs filter paper by using 500 mL of textile wastewater for each sample with different pH and constant dilution of 100. One of the parameters that can be related to pH is turbidity. Based on the graph in **Figure 4**, the turbidity removal for pH 4 and 7 has the lowest percentage recorded compared to pH 9. According to Yatim et al., 2021, increasing the pH value will decrease the turbidity value.The turbidity is measured by using the DR6000. Based on the data obtained from the experiment conducted, the value of turbidity is at 5 NTU. The initial turbidity recorded is 1706 NTU. Therefore, the turbidity removal obtained is 99% which is better using the filter paper. Additionally, research conducted by Mostafa in 2020 found that most of the effluent from textile wastewater is between 7.65 to 9.10 which is alkaline.

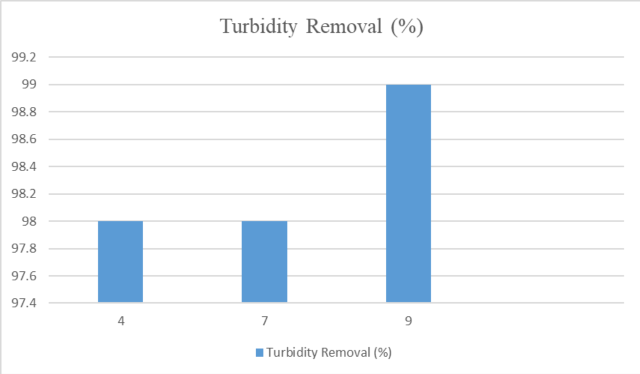

Figure 4: Turbidity of each sample according to pH

3.4 Results of Chemical Oxygen Demand (COD)

The experiment for Chemical Oxygen Demand was conducted using the optimum samples for the high range (200 – 15000 mg/L). Based on **Table 4**, the COD values drop drastically after undergo the filtration process which minimizes the pollution potential. The initial textile wastewater indicates the high value of Chemical Oxygen Demand value. It was influence with the present of high concentration of organic and inorganic compounds. Plus, the high COD value also indicates the high level of industrial effluent [9].

Table 4: Result of Chemical Oxygen Demand (COD)

Sample	Reading (mg/L)
Blank	0
Initial textile wastewater	378
Final textile wastewater	52

3.5 Result of Colour removal percentage

Using the DR6000 and the ADMI 1 inch programme, a decolourization test was conducted to evaluate how well textile wastewater removed colour. 1706 ADMI is the initial colour of textile effluent with the best parameter tested at 100% concentration. The final colour was then tested once more at 64 ADMI after the textile wastewater had been filtered using CNFs filter paper. The formula in Equation 3.4 was then used to calculate the percentage of colour removal. The percentage of colour removed was almost 96%. According to the observations, the water changes are easily distinguishable because it is now more colourless than it was before filtering. It means that the performance of the filter paper can be considered good to filtrate the colour from the wastewater effluent. The values obtained in this study are compliance with the Industrial Effluent or Mixed Effluent of Standard A which is 100 ADMI. This indicates that the filter paper's ability to remove colour from wastewater effluent can be regarded as effective. The results of this study are in accordance with Standard A's Industrial Effluent or Mixed Effluent, which is 100 ADMI. Table 5 shows the summary of colour removal for each sample. The results of this study are in accordance with Standard A's Industrial Effluent or Mixed Effluent, which is 100 ADMI.

Table 5: Summary of colour removal of each sample

Dosage	pH	Colour removal (%)
50	4	84
	7	87
	9	91
60	4	87
	7	92
	9	99
80	4	92
	7	99
	9	99

3.4 Result of FTIR Graph

FTIR spectroscopy allows the reading of the structural and compositional information on the functional groups present in the sample. The existence of the functional group in the optimum filter paper for both before and after were compared and presented in **Figure 5**.

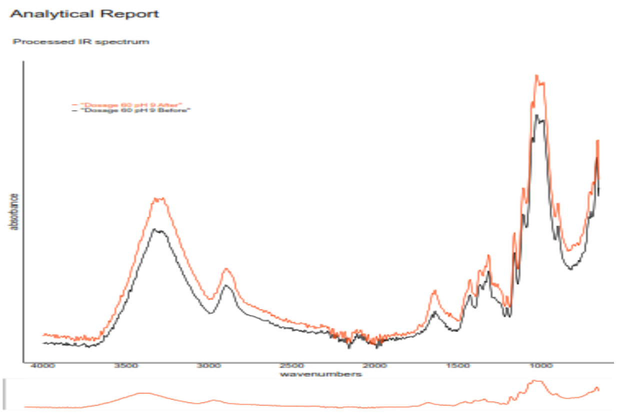

Figure 5: Overlay result of characterization of the before and after filter paper

The wave numbers on the infrared spectrum were plotted between. There are 16 peaks that can be observed on the filter paper before going through the filtration process. However, after the filtration process, the number of peak shows is increasing with an additional 4 functional groups present on filter paper. As shown in the Figure 4.4, the graph for optimum dosage in after is higher than the dosage in before. The result presented shows that the peaks of the filter paper characterization before were located at 3328.51, 1107.02, 1159.20, 2896.14, 1427.57 and 1051.11 cm-1. Some of the functional groups that appeared were hydroxide (OH-), Carboxyl and Alkane. The broad peak observed at the graph before is at 3328.51 cm-1 which represents the hydroxyl group (OH-). Moreover, the band points at 2896.14 cm-1 which represent the C-H bond.

However, the mixture of the dye pollutant from the wastewater sample has resulted in the additional functional group on the filter paper. The additional data includes the C-O Bond, C=C Stretching bond molecule and Silicon compound. The peaks located were at 697.01, 2255.04, 2202.86 and 2038.85 cm-1. From the line graph after the filtration, it shows that the broad peak was still at hydroxyl group (OH-) while the highest peak is at 1025.02 cm-1 which represents the secondary cyclic alcohol. The summary of the data obtained for the characterization of filter paper using FTIR are presented in Table 6 below.

Table 6: The summarization of the wavenumber after filtration

Wavenumber after filtration (cm^{-1})	Functional group
3276.33	Hydroxyl group
2896.14	C-H Bond
2255.04	C-O Bond
2202.86	C=C Stretching bond molecule
2038.85	Silicon compound
1427.57	Carboxylate
1051.11	Secondary cyclic alcohol

3.4 Summarization

According to the result and discussion the normalized water flux declines as time increases. It might be influenced by the quantity of effluent trapped on the filter paper which affects the water flow. The optimal pH and dosage obtained in this study are 9 and 60. The colour removal can cover about 99% which means the filter paper can be used. Moreover, the other characterization of wastewater parameters includes the COD, pH and turbidity. The COD Value recorded after the filtration process is low which is important as it reflect the level of pollution and contamination in the wastewater sample. If the COD vale remain high, the dissolved oxygen level will be reduced which is harmful for aquatic life forms. If we compare the value of those parameters with the Acceptable conditions for the discharge of Industrial Effluent or Mixed Effluent of Standard A and Standard B, they all can be categorised under Standard A. However, further improvement on filter paper should be taken as the volume of water flux was not high enough.

4. Conclusion

Based on the studies conducted, the dye wastewater treatment process using *Neolamarckia Cadamba* via cross-flow filtering system performing well, this study was successful in its objectives. The capacity of the membrane to remove dye wastewater and improve water quality is supported by the characterisation of the membrane before and after the filtration process. The cross-flow filtration method preserves a regular permeate flow rate, prolongs the life of the membrane by preventing irreversible membrane fouling, and improves CNF filter paper. In light of the findings and subsequent debate, 100% of the initial concentration of dye wastewater effluent was the best concentration to utilise in the industry because it was less expensive and did not require pure water dilution.Additionally, this study is able to demonstrate that *Neolamarckia Cadamba* has a significant percentage of colour and turbidity reduction. A positive outcome is also indicated by the final Chemical Oxygen Demand (COD) value. Overall, it can be said that this study was able to help remediate wastewater produced by the textile sector.

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