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The Potential of Nanocellulose Filter Paper (Neolamarckia Cadamba) for the Removal of Dyes from Textile Effluents

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Abstract: Cellulose Nanofibers (CNFs), in particular, has drawn considerable attention to the developments made in nanotechnology because of its unique properties. In this study, *Neolamarckia Cadamba* was used as a raw material in the manufacture of nanocellulose filter paper for the removal of dyes from textile wastewater. Therefore, the goal of this study is to investigate the effectiveness of the filter paper made from renewable forest resources through cross-flow filtration system in terms of the dosages of 60:40 and 70:30. Reactive dyes were thought to be the most problematic chemical in textile effluent due to their high-water solubility, stability, and persistence in nature. Normalized flux, colour removal, water quality parameters, and membrane surface properties were used to analyze membrane fouling behaviour. The results showed that colour removal was 91.21%, and the water quality parameters achieved were within the allowed wastewater discharge limit. The turbidity removal percentage is 99.66%. The goal of this project was to standardize wastewater treatment for the composite textile industry by using filter paper made of cellulose nanofibers (CNFs) as a membrane to remove dyes from wastewater. Thus, this study can verify the effectiveness of *Neolamarckia Cadamba* as raw source for filter paper.

Keywords: *Neolamarckia Cadamba*, Textile Wastewater, CNFs filter paper, Cross-Flow Filtration System

1. Introduction

Water is essential to human, plant, and animal survival. Today, a growing population, escalating agricultural and industrial demands, and the deteriorating effects of climate change all pose serious threats to the availability of water [1]. However, nowadays the elevation of industrial activities, uncontrolled usage of chemicals in domestic areas and urbanization process are keeps happening and it can lead to serious pollution because of that, it needs to minimize the safe water resources throughout the world. There are so many industrial activities that contribute to this pollution called dye chemicals

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**Corresponding author: norhazren@uthm.edu.my* such as food processing, textile, leather, dyeing and cosmetics [2]. This issue needs to be taken seriously as it will impact human health as well as the environment if the dye chemicals that have been discharged were not carefully treated especially when it discharged to the rivers or streams that happened to be Malaysian number one water resource [3].

Dyestuffs industries are the major contributors to the increment of environmental pollution which contained the largest group of organic compound contaminants [4]. One of the most significant sectors of manufacturing in Malaysia is the textile industry. The two main categories of textile fibres are natural and synthetic, which includes cotton, linen, wool, and silk as examples of man-made fibres. Natural polymers like viscose and cellulose acetate, as well as synthetic polymers like polyester, polyamide, polyacrylonitrile, poly urethane, and polypropylene, are the sources of man-made fibres [5]. Fibre is prepared in a variety of procedures in the textile industry. There are broad studies on textile wastewater solution treatment. Pollutants present in textile wastewater solution include inorganic substances, polymers, organic products, and colour. Chemical precipitation, adsorption, biological treatment methods, and membrane technologies can all be used to treat textile wastewater solutions. Due to their inexpensive cost, biological treatments are typically selected to remove the lingering colour from wastewater [6]. The most prevalent polymer found in nature is cellulose, which is produced by a range of organisms including bacteria, plants, and wood [7]. Cellulose is the most prevalent natural biopolymer on the planet in terms of biomass adsorbents. Nanocellulose, on the other hand, is a material that is smaller than 100 nm and can be divided into several formations, such as cellulose nanofibrils (CNFs), cellulose nanocrystals (CNCs), and bacterial nanocellulose (BNC). Nanocellulose can be produced from natural cellulose which consists of nanoscale fibrils and crystallites which permit the extraction of the nano constituents via mechanical and chemical methods, or through a combination of these techniques [8]. Forests and the by-products of wood and paper mills represent a wealth of renewable resources that can be used to generate everyday materials. Making and using these materials could reduce energy consumption, gas emissions and harmful waste disposal, and it could even lead to new employment opportunities. The starting material from Malaysian renewable resources which come from plantation forests will be used in this research in conjunction with Forest Research Institute Malaysia (FRIM). The type of renewable resource is *Neolamarckia Cadamba* which has been extracted into the CNFs form to produce filter paper. The main purpose of the CNFs filter paper is to be used in the elimination of dyes from untreated effluent.

To achieve the aim of this study, three objectives are listed. The objectives are to study the ability of cellulose-based nanomaterials (*Neolamarckia Cadamba*) for the removal of synthetic dyes from textile industry wastewater, to investigate the effect of CNFs filter paper ratio (60:40 and 70:30) during the filtration process regarding normalized flux and to distinguish and investigate the functional group present in CNFs filter paper. Turbidity removal and colour removal from textile industry wastewater also been tested for study as a replacement for current filter paper material. Parameters such as pH and COD of the wastewater is going to be studied in order to provide better understanding of the impacts of wastewater.

2. Materials and Methods

2.1 Design Experimental Planning

There are a few procedures that should be followed to complete this study on the removal of dye wastewater from textile industrial. Firstly, the materials which is Cellulose Nano-Fibrils (CNFs) filter made from *Neolamarckia Cadamba* which is the tree are planted in Forest Research Institute Malaysia been tested in permeability test with pure water. After the permeability test, dye wastewater will go through filtration into the CNFs filter paper by using cross-flow filtration system. The characterization of the CNFs filter paper will be done by performing the analysis using the following instruments which are Fourier Transform Infrared Spectroscopy (FTIR).

2.2 Material

The materials which will be used in this study are CNFs filter paper from *Neolamarckia Cadamba* species which obtain from Forest Research Institute Malaysia (FRIM) Kepong, Malaysia. As previously indicated, Malaysian renewable forest resources will be used as raw materials in this research. *Neolamarckia Cadamba* is being used as starting materials in cooperation with the Forest Research Institute Malaysia (FRIM) which they extract the tree into cellulose nanofibrils (CNFs). The various chemical used in this study such as sodium hydroxide (NaOH), hydrochloric acid (HCl) for control and adjusted pH value.

2.3 Characterization methods of CNFs Filter Paper

To understand the mechanism and properties of materials, the characterizations and analysis steps will be done on the CNFs filter paper. The characterization of the CNFs filter paper will be done by performing the analysis using the following instruments which are Fourier Transform Infrared Spectroscopy (FTIR).

2.4 Analytical Method

The experimental set-up for this study is the cross-flow filtration system based on figure 1, permeability test for CNFs filter paper and set-up using wastewater effluent. The equilibrium the percentage of colour removal will be determined by using Equation 1.

$$Colour\ removal, R\ (\%) = \frac{C_0 - C_t}{C_0} \times 100\% \qquad \text{Eq.1}$$

where C_0 is the initial dye concentration in the solution (mg/L), C_t is the equilibrium dye concentration in the solution (mg/L), and V is the volume of the solution (L). Normalized flux values of the membranes analysis are function to investigate the CNFs filter paper fouling behaviour during the reaction in the cross-flow filtration system by using Equation 2 below. The values will be determined by plotting graph of normalized flux against operating time.

$$Normalized\ flux = \frac{J}{J_0} \qquad \text{Eq. 2}$$

where J is the instantaneous permeate flux (L/m^2h) and J_0 is the initial pure water permeate flux (L/m^2h).

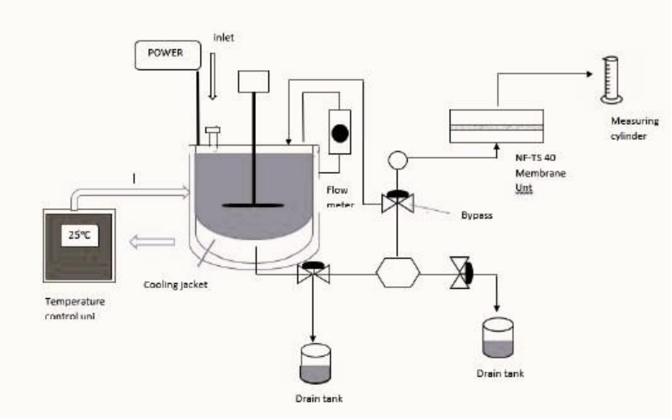

Figure 1: Cross-flow filtration system

3. Results and Discussion

3.1 Permeability Study

Figure 2 shows the water flux value of pure water using 70:30 and 60:40 filter papers under different operating pressure. The test was conducted to compare both filter papers on the resistance ability of absorbent again water with different hydrostatic pressure and also to compare on how efficiently effective the solvents were allowed to pass through. Increased operating pressure resulted in higher permeate flux in general [9]. Based on Figure 1, as operational pressures increased, the amount of pure water permeate flux increased linearly. The permeability test performance for the 70:30 filter was not as effective as the performance of the permeability test of 60:40.

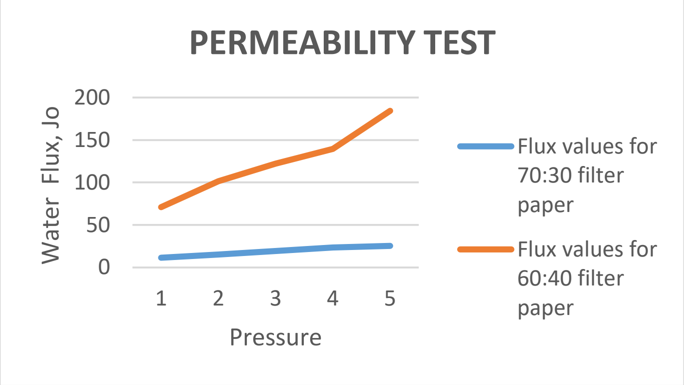

Figure 2: Permeability test for 70:30 and 60:40 filter paper

3.2 Effect of dosage

There are two dosages that were used which are 60:40 and 70:30. For the optimum dosage to be choose, the behaviours of membrane (filter paper) fouling need to be analyzed. A comparative analysis of fouling behaviours was conducted by graphing the graph of normalized flux against the operating period. Figure 3 shows the normalized flux of dye wastewater effluent with both dosages 60:40 and 70:30 filter paper. For these cases, the 60:40 filter shows higher normalized flux than the 70:30 filter paper. Filter paper dosage 60:40 need to be chosen as the optimum dosage as gives way less cost than filter paper 70:30. 70:30 filter paper cost is higher because it needs more CNFs when in the making of the filter paper. Every industry sector will prefer to choose a low cost when it comes to this matter as long as it is effective to use.

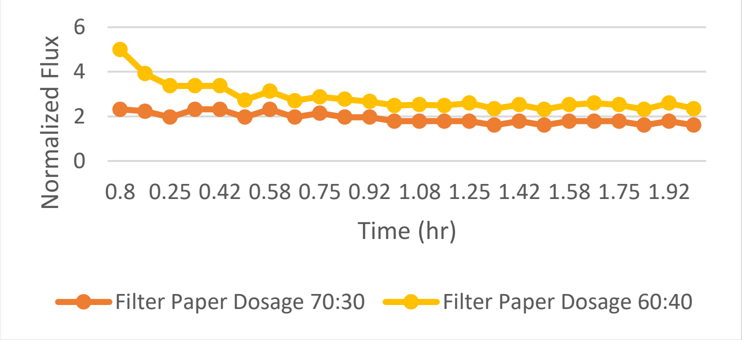

Figure 3: Comparison performance between the dosage 70:30 with dosage 60:40

3.3 Effect of pH

The value of the pH of wastewater needs to be compared to choose the best pH to be the optimum for wastewater effluent to get through the cross-flow filtration system. For the adjusted pH of wastewater to go through the cross-flow filtration system, there are four different values to be considered which are 3, 6.5, 7 and 10. Figure 4 shows the pH relationship with turbidity removal. Their concentration was fixed and only pH is different. The final turbidity of for experiment with pH 3 was 97.72 % and 99.66 % for the experiment with pH 6.5. For pH 7, the final turbidity removal was recorded at 98.95% and the pH of 10 is 98.60%. According to these tests, wastewater with a pH of 6 removed the most turbidity, followed by wastewater with a pH of 7, and pH 3 removed the least. The association between pH and the removal of turbidity from wastewater is not strongly supported by the data obtained, thus it is possible that it has an impact only when combined with other parameters that it would not have had on its own.

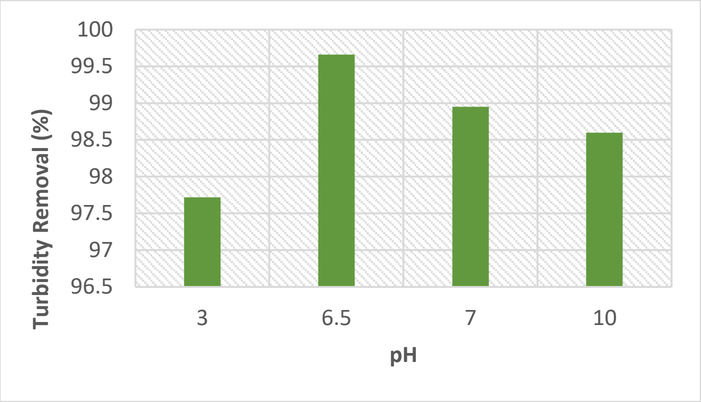

Figure 4: Graph Turbidity Removal (%) against pH

3.4 Effect of Dilution

The concentrations that were used in this experiment are 75% and 100%. 100% are equal to a 500 mL wastewater sample. For 75% concentration, 75% which equals to 375 mL of wastewater and the remaining 25% (125 mL) is dilution pure water. By plotting the graph of normalized flux against the operation period, a comparative investigation of fouling behaviors was carried out. Figure 5 shows the normalized flux of wastewater effluent concentrations of 100% and 75%. It could be observed that the 75% concentration showed a higher normalized flux than the concentration of 100%. Permeate flux increases and membrane fouling is reduced due to superior performance in the cross-flow filtration system can be linked to an improvement in dye wastewater effluent normalized flux. In comparison to the 100% concentration of dye wastewater effluent sample, this case clearly indicates that concentrations of 75% significantly improve and eliminate fouling propensity in cross-flow filtration systems. Since membrane fouling gets more severe as feed concentration rises, the drop in permeate flux was expected to be greater at higher concentrations. This could be attributed to a rise in dye wastewater adsorption on the membrane surface, as well as concentration polarisation [10]. Since it has a cheap initial cost and uses little energy, industry sectors prefer a low pressure driven membrane with the lowest operating pressure and the least amount of dilution required (at least 80% of dilution). Therefore, the performance of dye wastewater removal treatment using a cross-flow filtration system was positively influenced by the concentration. It was observed that a lower percentage of the concentration of dye wastewater permeate better off the membrane as the cake layer formation on the membrane surface was subsequently reduced.

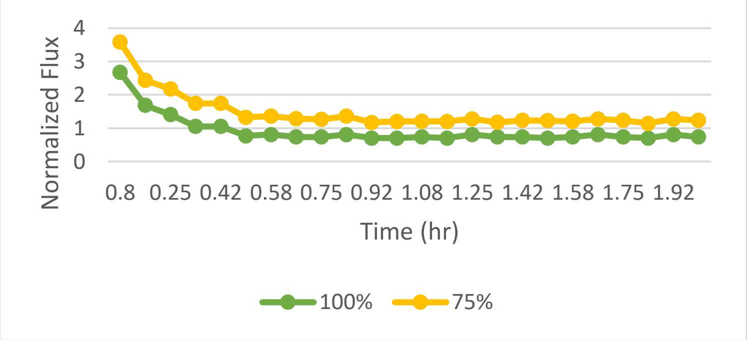

Figure 5: Comparison performance between the concentration 75% with 100%

3.5 Wastewater characterization

The result for wastewater characterization on parameters is shown in Table 1. For Chemical Oxygen Demand (COD), the initial COD reading is 379 mg/L which means it indicates that there are more oxidizable materials in the dye wastewater. If so, the water's dissolved oxygen content will have decreased. It needs proper treatment before discharge into the water body to minimise pollution potential since the value is higher. For the final result of COD which is after the filtration process, the value is 23 mg/L. The final COD show a low COD level compared to the initial. This means that the filtration process was managed to reduce the COD level.

For turbidity removal, the initial turbidity is 594 NTU while the final is 2 NTU. This show that before the filtration, the turbidity is high compared to after the filtration. The turbidity removal for this is 99.66%. This proved that the efficiency of CNFs filter paper produced from *Neolamarckia Cadamba* on turbidity removal was very excellent. Based on the legislation in Malaysia, the turbidity required was less than 10 NTU (Malaysia Wastewater Effluent Discharge Standards 1979). The turbidity of wastewater may be efficiently filtered using the CNFs filter paper and ensured to be within the range permitted by the government.

For colour removal, the initial ADMI of textile dye wastewater with optimum parameter tested was 1706 while the final is 150. The colour removal percentage obtained for the optimum was 91.21%. Based on the observation, the color of filtered effluent is essentially the same as the color of pure water, if not exactly the same. Figure 6 shows the colour of textile dye wastewater before and after the filtration. The use of CNFs filter paper was thought to have excellent colour removal performance.

Table 1: Characteristics of textile wastewater after filtration process

	Final pH	**Final Colour (ADMI)**	**Final COD (mg/L)**	**Final Turbidity (NTU)**
Dye Sample	8.04	150	23	2

Figure 6: The colour of textile dye wastewater before and after the filtration

3.6 Characterization of CNFs filter paper

Fourier Transform Infrared Spectroscopic (FTIR) provides structural and compositional information on the functional groups present in a sample. By using FTIR, the functional groups found in the immediate composition of filter paper were examined. Figure 7 show the FTIR spectrum of filter paper before and after the filtration process. Table 2 shows the summary peaks of CNFs filter paper before and after the filtration process. There are 13 peaks that can be observed on the filter paper before filtration process. However, after the filtration process, the number of peak shows is increasing with additional of 1 functional group present on the filter paper. The result presented shows that the peak of the filter paper characterization before were located at 3324.79, 2896.14, 1051.11, 1107.02, 1159.20 and 1427.57 cm^{-1}. Some of the functional group appeared were hydroxide ($\mathrm{OH^-}$), Carboxyl, Alkane, Carboxylate and secondary cyclic alcohol. The broad peak observed at the graph for before filtration is at 3324.79cm^{-1} which represent the hydroxyl group ($\mathrm{OH^-}$). However, with the mixture of dye wastewater effluent has resulting to the additional functional group on the filter paper. The additional functional group that been found after filtration process is C=C stretching bond of alkynes molecule. The peak located were at 2109.67 cm^{-1}.

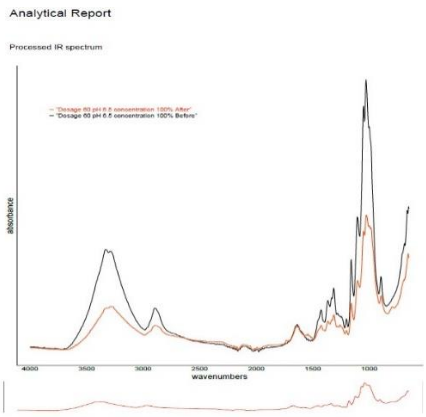

Figure 7: The FTIR spectrum of filter paper before and after

Table 2: Summary of peaks of CNF Filter paper after filtration process

WAVENUMBER AFTER FILTRATION (cm^{-1})	**FUNCTIONAL GROUP**
3287.51	OH^-
2896.14	C-H
1051.11	Secondary Cyclic Alcohol
1103.29	Carboxyl
1159.20	Alkane
1427.57	Carboxylate
2109.67	C=C stretching bond of alkynes molecule

4. Conclusion

In conclusion, this study was successfully conducted as the dye wastewater removal process through filtration process using *Neolamarckia Cadamba* via cross-flow filtration system show positive performances. The characterization of the membrane before and after the filtration process supports the ability of the membrane to remove dye wastewater and increase the water quality. The cross-flow filtration system helps to enhance CNF filter paper, helps to keep longer membrane life by preventing irreversible membrane fouling and maintains a uniform flow rate of permeate. Among of two ratio filter papers, 60:40 filter paper was the best choice because of its best performance and also cheap cost. 100% of the initial concentration of dye wastewater effluent was the ideal concentration to implement in the industry because it saves cost and does not need to dilute with pure water. This study also manages to prove that *Neolamarckia Cadamba* has a high percentage of colour removal which is 91.21% and also turbidity removal with percentage of 99.66%. The final Chemical Oxygen Demand (COD) reading also shows a good result. Overall, it can be concluded that this study was able to contribute to the treatment of the textile industry for wastewater effluent.

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