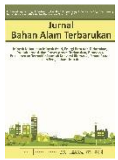
JBAT 13(1) (2024) 82 – 89

Jurnal Bahan Alam Terbarukan

Accredited SINTA 2 by Ministry of Research and Technology/National Research and Innovation Agency
SK No. 200/M/KPT/2020
http://journal.unnes.ac.id/nju/index.php/jbat

p-ISSN 2303 0623
e-ISSN 2407 2370



Application of Cellulose Acetate Propionate Biopolymer Membrane in The Treatment of Textile Wastewater Containing Remazol Dye

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DOI: https://doi.org/10.15294/jbat.v13i1.50295

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Article Info

Article history:
Received
14 March 2024
Revised
21 May 2024
Accepted
5 June 2024
Online
June 2024

Keywords:
Cellulose acetate propionate;
Membrane;
Phase inversion;
Remazol dye;
Rejection

Abstract

Textile liquid waste is pollution resulted from textile industry activities that often produce hazardous and toxic materials. Therefore, appropriate processing techniques are needed. There are many processing methods that can be used in treating this textile liquid waste, one of which is using membrane technology with phase inversion techniques. Membrane technology has advantages over other processing methods such as, being biopolymeric, relatively lower energy consumption, does not use chemicals in the processing process, and does not cause new waste in the processing process. In this study, 3 variations of cellulose acetate propionate (CAP) concentration were carried out in the manufacture of membranes including, 13% CAP; 14% CAP; and 15% CAP. Membranes that have been made are then carried out several analyses, namely porosity analysis; flux analysis; rejection analysis; and color concentration reduction analysis. The results in this study found that the highest porosity value, rejection value and flux value were found in the CAP membrane with a concentration of 13%. The CAP membrane has the best rejection value on reducing the remazol concentration of 43% with a membrane of 13% CAP.

INTRODUCTION

Liquid waste from the textile industry is pollution resulting from textile industry activities that often produce hazardous and toxic materials (B3) (Kishor et al., 2021; Rizkiah & Luciana, 2022; Yuniarti & Widayatno, 2022) and these wastes are non-biodegradable and resistant to good physical-chemical treatment methods (Enrico, 2019; Rizkiah & Luciana, 2022; Sausan et al., 2021; Sunoko, 2018). In the production of the textile industry, out of 100% of the dyes used in fact only 5% of dyes are effectively used. One type of dye that is often used in the textile industry is Remazol type dye (Amelia et al., 2023; Subagyo, 2021; Rahmiati et al., 2018) (Ismail et al., 2019).

Remazol is a textile dye with the molecular formula C18H14N2Na2O10S3 with a molecular weight of 560.5 g/mol (Yuniarizky & Nazriati, 2021). The impact of remazol dyes on humans includes eye irritation, skin irritation, cancer, carcinogenic, mutagenic, and has a high synthetic color content (Ayni & Ningsih, 2021; Kishor et al., 2021; Sari et al., 2018; Setiawan et al., 2023; Sunoko, 2018). Therefore, there is a need for processing techniques or methods in this case. One of them uses membrane technology (Kiswanto & Rahayu, 2019).

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Membrane technology is the treatment of liquid waste, one of which is the treatment of textile wastewater (Faizal et al., 2022; Jiang & To, 2022; Korkut et al., 2024; Maulana & Marsono, 2021; Mu'aliyah et al., 2022). Compared to other separation processes, this treatment process has several advantages, including the use of membranes that do not change the structure or arrangement of the separated substances, are biopolymers, can be operated at room temperature, relatively low energy consumption, non-toxic, no chemical additives in the process, and is a clean technology (Aburideh et al., 2021; Bousbih et al., 2021; Harianingsih & Maharani, 2018; Masri et al., 2020; Sianipar et al., 2023; Wang et al., 2023), lower initial investment cost than conventional systems, can remove harmful substances from textile wastewater (Chen et al., 2022; Hartini et al., 2018; Jin et al., 2018; Khan et al., 2020; Maulana & Marsono, 2021; Osman et al., 2024; Wen et al., 2021). One of the techniques commonly used in this process is phase inversion.

Phase inversion itself is a method in membrane manufacturing where the polymer is converted from the liquid phase to the solid phase with a certain control mechanism (Khan et al., 2020; Sofyana et al., 2020). The phase inversion process occurs by evaporating the precipitating solvent. The phase separation mechanism that occurs can be explained by a three-phase diagram with three main components, namely polymer, solvent and non-solvent (Lusiana & Prasetya, 2020), as shown in Figure 1.

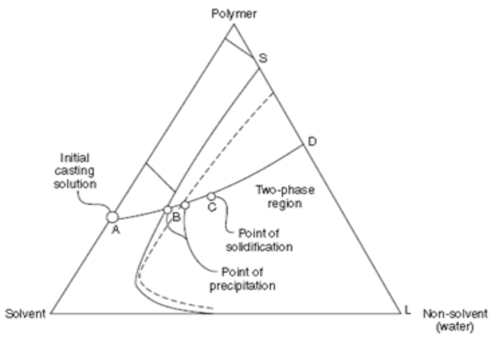

Figure1. phase diagram in phase inversion method.

There are many polymer derivatives that can be used in fabricating membranes including cellulose derivatives i.e. cellulose acetate propionate (CAP), cellulose acetate (CA). CAP is one of the biopolymer materials with the chemical formula $C_{76}H_{14}O_{49}$ (Cid, 2024) which is very potential to be applied to the manufacture of bioplastics because it has biodegradable properties (Faizal et al., 2022; Jiang & To, 2022). CAP has the advantages of excellent transparency and strength, then the incorporation of propionyl groups gives CAP the properties of water resistance, good solubility and compatibility with other polymers, resulting in a balance of hydrophilic and hydrophobic properties (Abdellah et al., 2020; Aburideh et al., 2021; Ashter, 2018).

Many studies on membrane fabrications have been carried out using various types of polymeric materials. Biomaterials are preferably used in fabricating membrane i.e., cellulose acetate, chitosan (Abdellah et al., 2020; Thakur & Voicu, 2016). As time passes, one type of polymer that is rarely found as raw material of membrane is CAP, in spite of cheaper price compared to others. Therefore, this research aims to study the preparation of CAP biopolymer-based membrane by phase inversion method and its application in treatment of textile wastewater containing remazol dye.

MATERIAL AND METHOD

Materials

There were several materials used in fabricating the membrane, including cellulose acetate propionate (CAP) from Sigma Aldrich, acetone (industrial grade), polyethylene glycol 4000 (PEG 4000), formaldehyde (industrial grade), and distilled water. In the application step, remazol dye was used.

Fabrication of cellulose acetate propionate membrane

In the manufacture of CAP membranes, three different compositions were prepared to produce membranes. The composition consisted of a mixture of CAP, PEG 4000, and acetone. The compositions are presented in Table 1.

Table.1. Compositions of CAP, PEG 4000, and acetone in producing CAP membrane.

No	CAP (%)	PEG 4000 (%)	Acetone (%)	Membrane Thickness (μm)
1	15	3	82	200
2	14	3	83	200
3	13	3	84	200

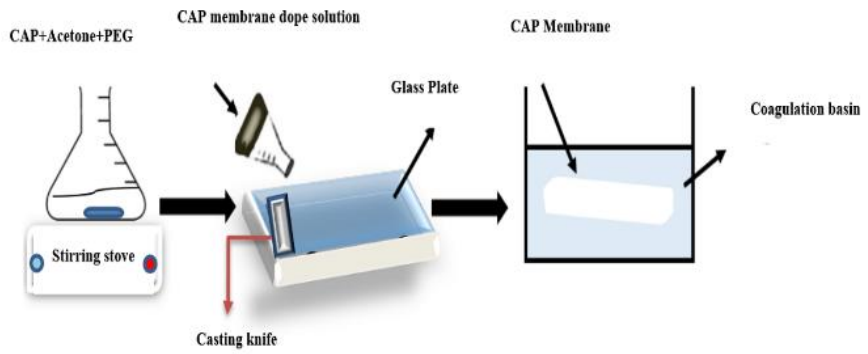

Figure 2. Procedure for CAP membrane preparation with phase inversion method.

The CAP membrane was made in several steps. In the first step, the membrane solution that consisted of CAP, PEG 4000, and acetone was stirred for 4 hours at 30 ° C to obtain the dope solution. The CAP dope solution was allowed to stand for 12 hours. The dope solution was then printed with printing equipment (Four Sided Applicator Frame Type: Wet Film Coater) on a glass plate, then dipped in distilled water until the membrane was peeled off. The CAP membrane was dried at room temperature. The CAP membrane was given a thermal annealing treatment at a time of 60 seconds and at a temperature of 60°C before being applied for wastewater treatment. The CAP membrane manufacturing procedure using the phase inversion method is shown in Figure 2.

Membrane Characterization

Porosity analysis

The porosity analysis was carried out by using gravimetric method (Huang et al., 2017; Li et al., 2009). The porosity ε (%) was calculated by using Eq. (1).

$$\varepsilon = \frac{\omega_w - \omega_d}{A.l.d_w} \quad (1)$$

where ω_w is wet membrane weight (g), ω_d is dry membrane weight (g), A is membrane area (cm^2), l is membrane thickness (cm), and d_w *is* density of water (g/cm^3).

Flux analysis

The flux of CAP membrane was determined by using dead-end filtration unit. The penetration flux J ($L/m^2.h$) is defined as shown in Eq. (2) (Huang et al., 2017; Li et al., 2009).

$$J = \frac{Q}{A.t} \quad (2)$$

where Q is permeate volume (L), A is area of membrane (m^2), t is permeation time (h).

Rejection analysis

The CAP membrane was applied for the filtration process of artificial wastewater containing remazol dye. The experiment was carried out using a dead-end filtration unit. The artificial wastewater was prepared by dissolving remazol dye in the distilled water at concentrations of 5 ppm; 10 ppm; and 15 ppm. The rejection R(%) was calculated using Eq. (3) (Huang et al., 2017).

$$R = \left(1 - \frac{C_P}{C_F}\right) \times 100 \quad (3)$$

where C_P is permeate concentration, C_F is feed solution concentration.

RESULTS AND DISCUSSIONS

Porosity

Swelling or porosity analysis is carried out to determine the amount of substance absorbed by the membrane (Fadli et al., 2021; Febriasari et al., 2021; Khan et al., 2020; Lubis et al., 2023). The swelling magnitude of the CAP membrane is calculated using Eq. (1) and the results are presented in Figure 3.

Figure 3 shows that the higher porosity value is obtained at a lower CAP concentration. The higher the amount of substance will be absorbed by the CAP membrane at a higher porosity. This result is in accordance with the results of previous research (Hartini et al., 2018) where the greater the porosity value, the greater the substance that can be absorbed by the membrane. According to some other previous research (Ismail et al., 2019; Lusiana & Prasetya, 2020), the higher the polymer concentrations used, the smaller the porosity value of the membrane.

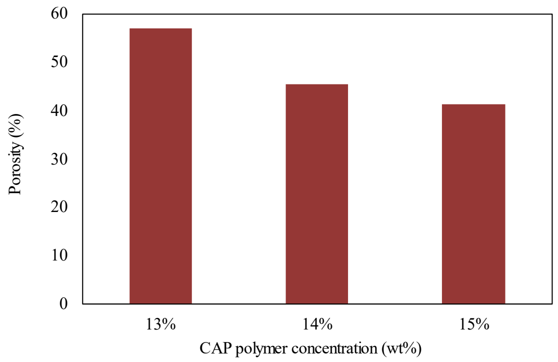

Figure 3. Porosity of membrane with CAP concentration of 13%, 14%, and 15%.

Flux

Flux analysis is carried out using a dead-end filtration device to measure the flow rate of membrane permeate per unit area and time (Bousbih et al., 2021; Indarti et al., 2020; Lusiana & Prasetya, 2020; Asri et al., 2021; Rahmahwati et al., 2021). The following flux analysis results are presented in Figure 4.

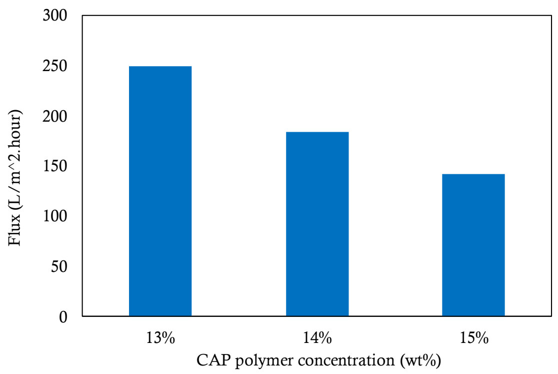

Figure 4. Flux of membrane with CAP concentration of 13%, 14%, and 15%.

From Figure 4, it can be seen that the membrane concentration of 13% CAP has a higher flux value compared to the concentration of 14%, and 15% CAP. This is in line with the porosity results conducted in this study. The flux results are also in accordance with the results that were reported previously (Bhernama et al., 2023; Fathanah & Meilina, 2021; Fransiska et al., 2023) stated that the higher the membrane concentration, the lower the flux value produced.

Rejection

Measurement of the rejection value is carried out to determine the selectivity of the membrane or the ability of the membrane to withstand specimens passing through the membrane (Abu-Zurayk et al., 2023; Bhernama et al., 2023; Lubis et al., 2023; Maulana & Marsono, 2021; Morão et al., 2005; Reddy et al., 2022; Vasagar et al., 2021). The results of the rejection analysis with variations in CAP membrane concentration (13%; 14%; and 15%) and variations in waste dye concentration of 5, 10, and 15 ppm can be seen in Figure 5.

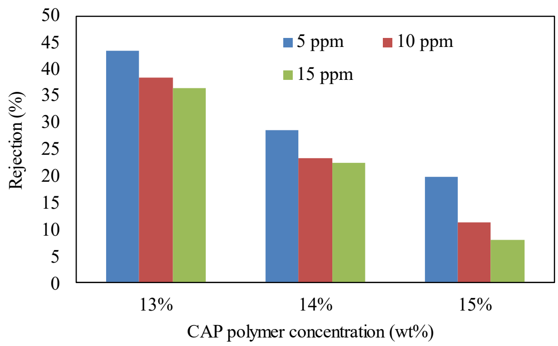

Figure 5. Rejection analysis results with varying CAP membrane concentrations (13%; 14%; and 15%) and varying effluent dye concentrations of 5, 10, and 15 ppm

The value of the rejection yield varies between 0% and 43.48%. Therefore, the rejection efficiency is also largely determined by the membrane pores (Reddy et al., 2022). In Figure 5, it can be seen that the highest rejection result is found in the CAP membrane with a membrane concentration of 13%, while the smallest rejection result is found in the CAP membrane with a concentration of 15%. This is in line with the previous result reported (Lindu et al., 2010) where researchers suggest that the greater the percentage value of membrane rejection is directly proportional to the resulting flux value.

CONCLUSIONS

This research shows that the use of cellulose acetate propionate (CAP) membrane is effective in treating wastewater containing remazol dye. The CAP membrane has a high porosity, as well as flux that varies depending on CAP concentration. The CAP membrane has the effectiveness of reducing the concentration of remazol dye by 43%. The results also show that the membrane with 13% CAP concentration has higher performance than CAP membrane with 14% and 15% CAP concentration.

ACKNOWLEDGEMENTS

The authors would like to thank the members of Material Laboratory, Chemical Engineering Department, Universitas Ahmad Dahlan for their support in conducting this research. The authors also would like to convey their gratitude to Universitas Ahmad Dahlan for financial support trhough a research grant with contract number: PD-250/SP3/LPPM-UAD/VIII/2023

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