EQUILIBRIUM JOURNAL OF CHEMICAL ENGINEERING

*Homepage:*https://jurnal.uns.ac.id/equilibrium

Synthesis and Characterization of Cellulose Acetate Membranes from Kepok Banana Stem *(Musa acuminata x balbisiana)* for Microfiltration Process

Fitria Nur Anissa*, Selastia Yuliati, Adi Syakdani

Industrial Chemical Technology Study Program, Department of Chemical Engineering, Sriwijaya State Polytechnic, Palembang, Indonesia

*Corresponding author: fitria.nur.anissa14032002@gmail.com

DOI: https://doi.org/10.20961/equilibrium.v7i2.78825

Article History
Received: 14-09-2023, Accepted: 12-10-2023, Published: 16-10-2023

Keywords:

Membrane, Microfiltration, Cellulose Acetate, Banana Frond, Acetone

ABSTRACT. The membrane is a separation technology between permeate and feed. The challenge in applying this technology is related to the substances that can cause fouling. The cellulose acetate membrane is a porous membrane as a solution to overcome fouling problems. The selectivity level of cellulose acetate membranes in the microfiltration process for water and wastewater treatment is very high. Utilization of kepok banana stem waste as an alternative material for making cellulose acetate membranes that are biodegradable can be decomposed quickly by microorganisms with a limited shelf life. The aim of this research is to apply cellulose acetate membranes from kepok banana stem (*Musa acuminata x balbisiana)* for the microfiltration process in water and wastewater treatment with varying concentrations of acetone using the phase inversion method. The cellulose content contained in the kepok banana stem is 54.3%. The composition for making the membrane uses 2 grams of cellulose acetate; 20 ml PVA ; and 1.7 ml of PEG then mixed with acetone varying the concentration. Membrane A 15 ml, membrane B 20 ml, and membrane C 25 ml. The results of the SEM test of the membrane with a concentration of 25 ml of acetone solvent had the best characteristics with a pore size of 0.0932 μm; thickness of 1.778 mm, and swelling index of 9.87%. The highest average flux value was owned by the membrane with a concentration of 20 ml, namely 71.3444 l/m^2.hour The lowest flux value was owned by the membrane with a concentration of 25 ml, namely 55.5549 l/m^2.hour.

1. INTRODUCTION

Separation using membrane technology is far superior to previously used methods. This is a modern alternative to separating substances. Membrane technology provides many advantages, such as the ability to carry out separations continuously, saving energy, and reducing production costs. Another advantage is the membrane's ability to be adapted to a variety of different compounds, so this technology can be applied in various contexts [1]. The challenge in applying membrane technology lies in the materials that can cause fouling. One alternative is to use cellulose acetate, which can reduce fouling during the separation process [2]. Separation using a membrane is basically a process of selective material transfer caused by the presence of forces related to certain parameters between the two media being separated, due to differences in electrochemical potential and pressure [10]

The advantage of cellulose acetate as a raw material for making membranes lies in its asymmetric structure, which includes a very thin active layer. This membrane has the ability to hold dissolved materials in its rough supporting layer, as well as reliability in dealing with precipitation. In addition, cellulose acetate is also able to create a balance between hydrophilic and hydrophobic properties, which makes it more efficient in the separation or filtration process [3]. The synthesis of cellulose acetate membranes from banana fiber is an alternative that can be used in the separation process. The results of the analysis by the researchers showed that the dried banana stem contained very high levels of cellulose and glucose. More than 50%, is cellulose. Kepok banana (*Musa acuminata x balbisiana)* stem contains 64% cellulose, 19% hemicellulose, 5% lignin and 11% water content [4].

Cellulose acetate membranes made from banana tree trunks are the latest update. Because the cellulose content in Kepok banana stems is more than 50%, it can be used as the main material for making membranes and has biodegradable properties. However, their mechanical properties are not optimal, causing the membrane's service life to be shorter [5]. The simplest and most economical way to improve its mechanical properties is to add a solvent. The membrane manufacturing process is influenced by the type of solvent used, and this affects the characteristics of the membrane formed. In this case, the solvent used is acetone. The amount of solvent used also plays a crucial role in membrane formation [6]. The use of acetone solvent in membranes causes the formation of smaller and more regular pores in the membrane [7]. The quantity of solvent used has a significant role in the membrane formation process. Using too little solvent can cause the dope solution to become inhomogeneous, which ultimately results in an uneven cellulose acetate membrane morphology. On the other hand, if the solvent is used in the right amount, it will produce a membrane with a more even surface [8]. Apart from that, the addition of acetone solvent can also increase the flux value of the membrane [9].

In the water treatment process and liquid waste processing, microfiltration can be carried out. Microfiltration is a process that can be applied to separate dissolved particles with sizes in the range of 0.1 to 10 μm and has a standard flux value of >50 l/m2.hour [10]. Apart from that, the material used in this microfiltration process is cellulose acetate.

This study was conducted to make cellulose acetate membranes from banana fronds with variations in the composition of acetone solvents reviewed from the characterization of the membranes. It obtained optimal pore sizes for the microfiltration process and can be applied to water and liquid waste separation processes. Using the phase inversion method when printing membranes involves the process of extracting cellulose, synthesizing cellulose acetate, and printing membranes. Then the membrane was tested for its characteristics through the SEM (scanning electron microscope) test, swelling index, and permeability through the flux test.

2. MATERIALS AND METHODS

2.1 Research Materials

The main substances used are kepok banana stem, sodium hydroxide (NaOH), 12% sodium hypochlorite (NaClO), sulfuric acid (H_2SO_4), 98% glacial acetic acid (CH_3COOH), acetate anhydride ($C_4H_6O_3$), Acetone (C_3H_6O), distilled water, polyethylene glycol (PEG), and polyvinyl alcohol (PVA).

2.2 Research Tools

The equipment used during the research was a series of microfiltration membrane devices: reflux, 250 beakers; 500; and 1000 ml; conical flask 100; 250; 500; 1000 ml; 25 measuring flasks; and 50 ml; a measuring pipette; 25 ml; an analytical balance, an oven, a hot plate, a spatula, filter paper, a stirrer, a blender, and a glass plate measuring 22 x 22 cm.

2.3 Research Steps

2.3.1 Quantitative Analysis of Cellulose Content in Kepok Banana Stems

Analysis of cellulose content using the SNI 14-0444-1989 method. 3 grams of dried Kepok banana stem powder was soaked in 35 ml of NaOH solution, then stirred for 15 minutes. then add NaOH after 3, 6, 9, and 12 minutes respectively (10 ml). After stirring, let stand for 30 minutes then add 100 ml of distilled water. After that, rinse until the pH is normal. then added 12.5 ml of 2N acetic acid and stirred for 5 minutes. After that, rinse again until the pH is normal. filter the sediment which is then dried at 70ºC for 60 minutes. after that, cool and weigh the dried results. Calculate the cellulose content obtained using the SNI 14-0444-1989 method formula in equation (1).

$$cellulose\ content(\%) = \frac{\text{Weight of cellulose precipitate}}{\text{Dry banana fronds weight}} x\ 100\% \qquad \text{..............................(1)}$$

2.3.2 Extraction and Isolation (Purification) of Cellulose from Kepok Banana Fronds

The extraction process uses reflux as a medium for separating cellulose and lignin from Kepok banana stems. dry and grind 10 grams of kepok banana stem powder, then reflux with 250 ml NaOH for 120 minutes. then rinsed with distilled water until the pH returns to normal. Followed by adding 250 ml of 12% NaClO, refluxing again for 240 minutes until the sample changed color from brown to white. Rinse again with distilled water until the pH returns to normal. And dried in the oven at 70oC for 240 minutes. The dried samples were ground using a blender.. The ratio of the resulting powder is 1:10 grams.

2.3.3 Synthesis of Cellulose Powder into Cellulose Acetate

15 grams of cellulose were dissolved in 150 ml of 98% CH_3COOH and stirred for 50 minutes at 40ºC. After 50 minutes, the sample was mixed with 4.5 ml of concentrated H_2SO_4 and stirred at 40ºC. Then added 100 ml of $C_4H_6O_3$ was stirred at the same time and temperature. After that, add 15 ml of distilled water and 30 ml of 98% CH_3COOH mixed into the acetylated product and stirred for 30 minutes at 50ºC. After the synthesis process is complete, the sample is precipitated in 1000 ml of distilled water to obtain a white powder resulting from the synthesis of cellulose acetate. The cellulose acetate flakes were precipitated and rinsed until the pH returned to normal, then dried in an oven at 60ºC for 210 minutes. The cellulose acetate powder obtained was filtered and ground until smooth.

2.3.4 Preparation of Cellulose Acetate Membrane from Kepok Banana Fronds

Mould the membrane by weighing 2 grams of cellulose acetate, which is then dissolved in 15; 20; 25 ml of C_3H_6O and stirred until evenly mixed. A total of 1.7 ml of PEG was dissolved in 20 ml of PVA solution until it became homogeneous. After both are homogeneous, add a homogeneous mixture of PVA and PEG solutions to the mixture of acetone and cellulose acetate and stir until thoroughly mixed. After being homogeneous, print using the phase inversion method, where the polymer solution is poured on a glass section measuring 22 x 22 cm, and then a thin layer in the form of a membrane is obtained that is ready to be tested.

2.3.5 Characterization Test of Cellulose Acetate Membrane

Membrane morphology was examined using a SEM (scanning electron microscope) tool This test was carried out to determine the pore size of the cellulose acetate membrane. Next, test the thickness of the membrane using a caliper to make it easier to determine the thickness of the cellulose-acetate membrane. Lastly, the swelling index test using the gravity method was carried out by soaking the membrane in distilled water for 15 minutes to determine the ability of the, cellulose acetate membrane to absorb water. The equation used to determine the swelling index value is in equation (2).

$$SI = \frac{B_K - B_A}{B_A} X\ 100\% \qquad ....(2)$$

where B_k is the final weight of the sample and B_A is the initial weight of the sample.

2.3.6 Cellulose Acetate Membrane Permeability Test Using Microfiltration Equipment

The permeability testing process uses a series of microfiltration tools which can be seen in Figure 1.

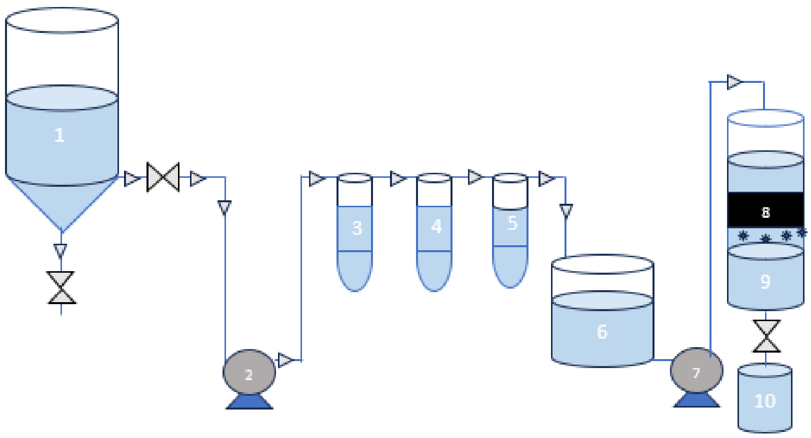

Figure 1. Microfiltration Tool for Permeability Test

Free-contaminated water flow rate measurements were carried out to evaluate the performance and permeability of the membrane in facilitating the movement of pure water over a specified time period. Flux value measurements were carried out by holding the permeate volume every 4 minutes in a measuring cup. Distilled water was used as bait in the test. Each membrane will be given the same pressure treatment. To test the suitability of the membrane, pressure variations are used, namely 0.2, 0.4, 0.6, 0.8, and 1.0 bars. The following is a description of the microfiltration tool for flux testing: (1) water storage tank (feed), (2) pump, (3) activated carbon, (4) zeolite, (5) cartridge filter, (6) feed tank, (7) pump booster, (8) cellulose acetate membrane, (9) membrane module, (10) permeate

3. RESULTS AND DISCUSSIONS

3.1 Test Results for the Cellulose Content of Kepok Banana Fronds

Testing the cellulose content of the kepok banana midrib is an important process in analyzing the fiber content in this part of the banana plant. Making cellulose acetate is made from cellulose, so testing the cellulose content is very important to determine the content contained in the kepok banana stem. The test results for the cellulose content of Kepok banana fronds were 54.3%.

In the tests carried out, the cellulose content obtained was 54.3%, and the method used was in accordance with SNI 14-0444-1989. The cellulose content obtained is included in good cellulose standards, namely 45–60%. So that the cellulose of banana stems can be used to manufacture cellulose acetate membranes.

3.2 Characteristic Test Results of the Cellulose Acetate Membrane

Membrane characteristics include several important parameters such as pore size, thickness, membrane diameter, and swelling. To identify the size of the pores in the membrane, the SEM (scanning electron microscope) Thermo Fisher Phenom Pro, which is very relevant in analyzing the membrane surface structure The results of SEM tests carried out on the three membranes can be seen in Figure 2 as follows:

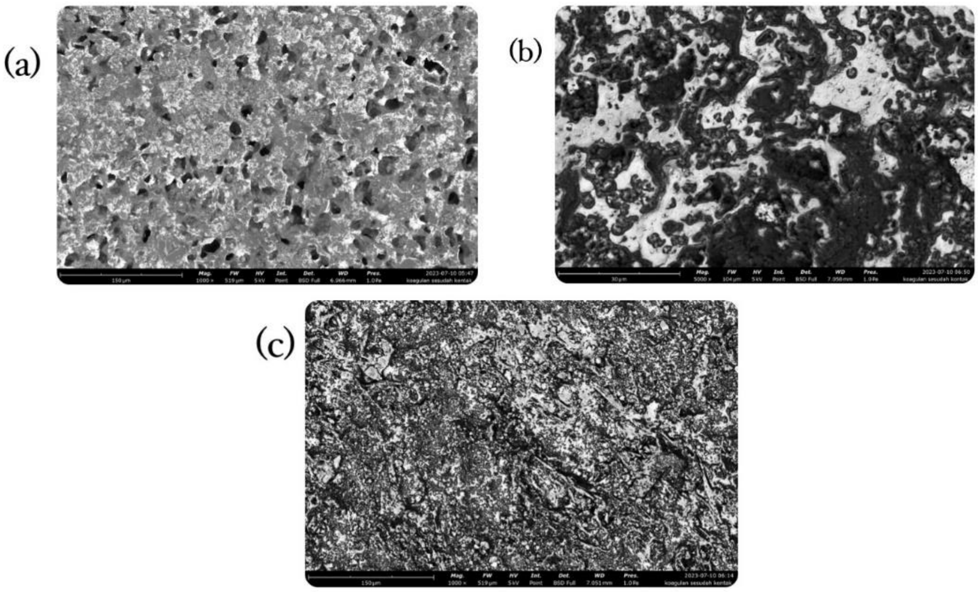

Figure 2. SEM images of (a) Membrane A; (b) Membrane B and (c) Membrane C

From the results of the SEM test, membrane A was carried out with a concentration of 15 mL of acetone, resulting in a membrane with a pore size of 1.345µm. From the image of the membrane, it can be seen that there are holes or voids. This is because the dope solution does not completely dissolve during the manufacturing process [11]. Thus causing pores to form.has a large size. The pores formed on the A membrane become wider and irregular. Addition inappropriate solvents produce membranes with uneven, thin, and wavy surfaces [12].

Meanwhile, membrane B with a solvent concentration of 20 ml has a pore size of 1.134µm. Based on the SEM test results, there are membrane pores that have a non-uniform shape. This indicates that membrane B can be classified as an asymmetric membrane, which displays differences in pore structure between the two sides of the membrane [13]. The interaction between the solvent (acetone) and the polymer (cellulose acetate) used in making the membrane affects the formation of membrane pores. During the membrane manufacturing process, these interactions can cause contraction and expansion of the polymer, which in turn affects the porosity and structure of the membrane. These factors cause membrane B to have a non-uniform or asymmetric pore shape on its surface.

And for membrane C with an acetone solvent concentration of 25 ml, a membrane with a pore size of 0.823µm was obtained. Membrane C has the tightest and smallest pore size compared to membranes A and B. This condition is caused by the composition of the acetone solvent, which is mixed with cellulose acetate more than membranes A and B. As the amount of polymer in solution increases, the number of pores present in the membrane increases. Becomes more limited, and the pore dimensions also decrease. The results obtained show that the higher the polymer concentration, the lower the flow value of the membrane and the higher the rejection rate of the substance [14]. From the results of the SEM test on three cellulose acetate membranes for the microfiltration process, it can be concluded that the three membranes meet the pore size according to microfiltration standards. The standard pore size in microfiltration membranes is in the range of 0.05 µm to 10 µm [10]. The characteristic test results of the three Cellulose Acetate Membranes can be seen in Tabl.1.

Table 1 Characteristic Test Results for Cellulose Acetate Membranes

Membrane Type	Membrane Composition	Membrane Characteristics			
		Thickness (mm)	Pore Size (µm)	Membrane Diameter (cm)	*Swelling* (%)
A	Acetone 15 ml; 1,7 ml PEG	0.762	1.345	11	55.56
B	Acetone 20 ml; 1,7 ml PEG	1.27	1.134	11	24.44
C	Acetone 25 ml; 1,7 ml PEG	1.778	0.823	11	9.87

Based on the results of the swelling test measurements obtained, it was found that the acetone composition showed a lot of high swelling results. The composition of membrane A with 15 ml of acetone produces a value of 55.56%, membrane B with 20 ml of acetone produces 24.44% acetone, and membrane C with 25 ml of acetone produces 9.87% acetone. The effect of the composition ratio on the swelling index shows that the higher the ratio of acetone used, the lower the swelling index. Thus, the membrane will be more hydrophobic if the swelling index is smaller [15].

The thickness of the membrane is measured using a caliper. The thicknesses of membranes A, B, and C are respectively 0.762 mm, 1.27mm, and 1,778 mm. The thickness of the membrane is one of the key parameters in membrane manufacture, which can be adjusted by adjusting the amount of acetone in the dope solution. Membranes with thinner thicknesses tend to have larger pores and higher porosity. Conversely, membranes with thicker thickness tend to have smaller pores and lower porosity.

3.3 Test Results of Cellulose Acetate Membrane on Permeability in Microfiltration Equipment

The determination of pure water flux is carried out to measure the performance and permeability of the membrane in passing pure water in a certain time unit. The factor that influences the flux value is pressure (driving force). To test the suitability of the membrane, pressure variations are used, namely 0.2, 0.4, 0.6, 0.8, and 1.0 bars. Flux value measurements were carried out by holding the permeate volume every 4 minutes in a measuring cup. Aquades was used as bait in the test. Each membrane will be given the same pressure treatment. The following is a table of cellulose acetate membrane permeability test results. Cellulose Acetate Membrane Test Results on Permeability in Microfiltration Equipment can be seen in Table.3.

Table 3. Test Results of Cellulose Acetate Membrane on Permeability in Microfiltration Equipment

Pressure	Average flux Membrane A (liter/$m^2.hour$)	Average flux Membrane B (liter/$m^2.hour$)	Average flux Membrane C (liter/$m^2.hour$)
0,2 bar	52.377	53.446	55.555
0,4 bar	59.164	57.873	60.08
0,6 bar	62.86	59.47	61.341
0,8 bar	63.427	64.583	52.293
1,0 bar	67.98051	71.34449	66.52274

The effect of flux on pressure can be seen in Figure 3.

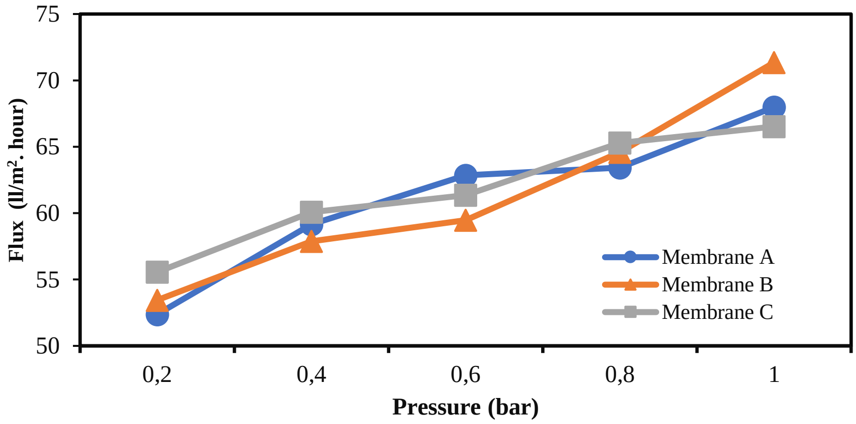

Figure 3. Effect of Flux on Pressure

Based on the graph, it can be observed that there is a correlation between pressure and the average pure water flux. The higher the operating pressure provided, the higher the resulting water flux value. This happens because the influence of the driving force causes more molecules in the solution to pass through the membrane. Thus, the higher the pressure, the greater the water flux coming out of the membrane [10].

Membrane B shows a high flux value with the average flux of membrane B at each pressure of 0.2 bar, namely 53.4457 l/m^2. hour, 0.4 bar is 57.6726 l/m^2. . hour: 0.6 bar, which is 59.4695 l/m^2. hour; 0.8 bar, which is 64.5835 l/m^2. hour; 1.0 bar, which is 71.3444 l/m^2. hour. Meanwhile, membrane A shows an average flux value for membrane A at a pressure of 0.2 bar, namely 52.3765 l/m^2. hour; 0.4 bar, which is 59.1637 l/m^2. hour; 0.6 bar is 62.8602 l/m^2. hour; 0.8 bar is 63.4269 l/m^2. hour; 1.0 bar is 67.9405. Meanwhile, membrane C has a low average flux with a respective pressure value of 0.2 bar, namely 55.5549 l/m^2. hour; 0.4 bar is 60.0804 l/m^2. hour; 0.6 bar is 61.3412 l/m2. hour; 0.8 bar is 65.2928 l/m^2.hour; 1.0 bar is 66.5230 l/m^2. hour.

Increasing the flux value for increased pressure results in deformation, i.e., the membrane pores enlarge. These three membranes meet microfiltration standards and can be used for water treatment processes. In accordance with the standard, which states that the microfiltration membrane flux value is >50 l/m^2.hour [10].

4. CONCLUSION

Based on the findings from research that has been conducted regarding the manufacture of cellulose acetate membranes from banana kepok fronds for the microfiltration process, there are several conclusions, as follows:

1. The cellulose acetate membrane from the kepok banana fronds that meets the microfiltration standard is membrane C (2 gr Cellulose Acetate; 25 ml Acetone; 1.7 ml PEG; 20 ml PVA).
2. Optimum characterization by adding acetone solvent to the membrane permeability properties for the microfiltration process, namely membranes with high concentrations tend to have low and constant flux values, namely membrane C with a flux value at 1 bar pressure of 66.5230 l/m2.hour. Thicker layers tend to have smaller (tighter) pores and are hydrophobic. Meanwhile, low concentrations have high flux values, namely membrane B, with a flux value at 1 bar pressure of 71.3444 l/m2. hour also has a large pore size, contains voids, and is thin and hydrophilic.
3. Characteristics of cellulose acetate membranes from Kepok banana stemsthat meet microfiltration standards for clean water processing and liquid waste processing, namely membrane C, which has a tight pore size of 0.823 µm, low swelling of 9.87%, and a thickness of 1.778 mm.

5. DAFTAR PUSTAKA

[1] H. Harianingsih, F. Maharani, "Sintesis Membran Selulosa Asetat Cassava Untuk Mikrofiltrasi Fe Pada Limbah Batik Artifisial," *J. Inov. Tek. Kim.* 3 (2018). https://doi.org/10.31942/inteka.v3i2.2489.

[2] H. Nani, M. Mahmud, C. Abdi, "Pengaruh pH Air Gambut Terhadap Fouling Membran Ultrafiltrasi," *Jukung (Jurnal Tek. Lingkungan)*. 1 59–73 (2016). https://doi.org/10.20527/jukung.v1i1.1038.

[3] N. Kumano, A., Fujiwara, "Cellulose triacetate membranes for reverse osmosis," *Dvanced Membr. Technol. Appl.* 21–46 (2008).

[4] S. Ahmad, "Pembuatan Membran Selulosa Asetat Pada Berbagai Variasi Komposisi Polimer, Jenis Pelarut Dan Konsentrasi Aditif," *Pros. Simp. Nas. Polim. V*. 75–80 (2008).

[5] D. Apriana Putri Husni, E. Abd Rahim, J. Soekarno Hatta Km, K. Bumi Tadulako Tondo Palu, "Pembuatan Membran Selulosa Asetat Dari Selulosa Pelepah Pohon Pisang [The Production of Cellulose Acetate Membrane from Stem of Banana Cellulose]," *Kovalen*. 4 41–52 (2018).

[6] S. Darni, Y. Utami, H., Asriah, "Peningkatan Hidrofobisitas dan Sifat Fisik Plastik Biodegradable Pati Tapioka dengan Penambahan Selulosa Residu Rumput Laut Eucheuma spinossum," *Pros. Semin. Has. Penelit. Dan Pengabdi. Kpd. Masy.* (2009).

[7] E.C. Muliawati, "Pembuatan dan karakterisasi membran nanofiltrasi untuk pengolahan air," *Semarang. Magister Tek. Kim.* (2022).

[8] S. Ahmad, "Pembuatan Membran Selulosa Asetat Pada Berbagai Variasi Komposisi Polimer, Jenis Pelarut Dan Konsentrasi Aditif.," *Pros. Simp. Nas. Polim. V*. 75–80 (2008).

[9] P. Yang, L., Hsiao, W., Chen, "Chitosan-cellulose composite membrane for affinity purification of biopolymers and immunoadsorption. Elsevier," *J. Memb. Sci.* 197 185–197 (2001).

[10] M. Mulder, "Basic Principle of Membrane Technology," (1996).

[11] N. Widayanti, "Karakterisasi Membran Selulosa Asetat dengan Variasi Komposisi Pelarut Aseton dan Asam Format.," (2013).

[12] M. Miftahul Jannah, "Pengaruh Penambahan Aseton Sebagai Pelarut Dalam Pembuatan Membran Dari Minyak Nyamplung (Calophyllum Inophyllum)," *Dr. Diss.* (n.d.).

[13] R.Z. Mahdiyah, "intesis dan Karakterisasi Membran Selulosa Asetat dengan Agen Pembentuk Pori Nonilfenol Etoksilat Termodifikasi Silika," (2022).

[14] P. Richa, R., Metty, M., & Slamet, "Sintesis selulosa diasetat dari eceng gondok (Eichhornia crassipes) dan potensinya untuk pembuatan membran.," *J. Teknol. Kim. Dan Ind.* 2(3) 8–16 (n.d.).

[15] F. Kamulyan, B., Hasanah, U., & Matulesi, "Kajian Campuran Pelarut Akuades-Aseton Pada Pembuatan Membran Selulosa Propionat.," *Kpk (Jurnal Kim. Dan Pendidik. Kim.* 3(2) (n.d.).