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Effect of Dope Solution Temperature on Characteristic and Performance of Cellulose Diacetate Membrane Based on Cellulose of Sengon Wood (*Paraserianthes falcataria sp*)

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***Abstract*— Cellulose diacetate (CA) based on cellulose of sengon wood (*Paraserianthes falcataria sp*) have content of acetyl as much as 39,66 % have be done as polymer material on preparation of membrane by phase inversion methode. Solution dope is done by blending cellulose acetate in 18% wt of polymer concentration with dimethyl formamide as solvent at room temperature, 40^{o}C, and 50^{o}C. After casting and evaporating on the glass plate, the thin layer of dope solution was coagulated in water at room temperature until formed the membrane. FTIR's analysis shows typical group absorption of carbonyl C=O in the number of wave 1751 cm-1 and group of Acetyl C-O in number wave of 1250 cm-1 . The resulting membrane morphology by Scanning Electron Microscope (SEM) JSM-5000 showed asymmetric membrane consist of top layer with dense surface skin and bottom layer are more porous. The performance of membranes tested on 1 bar trans membrane pressure. Varying of dope solution temperature affected the membrane flux and rejection produced. Highest flux obtained at a temperature of 50°C for dope solution was 362.459 L/m2.hour, while the lowest flux generated in solution at room temperature dope was 244.34 L/m2.jam. Rejection of the standard solution dextran 9500 Da range 62.13% - 69.39%, while for 12 000 Da dextran standard solution at various temperatures dope solution produces rejection> 90%.**

Keywords: **Cellulose Wood Sengon; Cellulose Diacetate Membranes; Phase Inversion; Dope Solution Temperature.**

I. INTRODUCTION

Membrane is one medium that is now rapidly expanding its use in the process of separation and purification technologies such as in the pharmaceutical, biotechnology and food industries [2] as well as in water treatment and for wastewater treatment [3]. The advantages of using membrane technology include low energy required, requires no additional chemicals, produces no contaminants or pollutants, require a relatively small area and is modular so easily combined with other technologies [5]. However, the use of membranes in the industry in Indonesia is constrained in terms of the high cost and scarcity of imported membrane local membrane [6]. Some researchers have been conducting research and development on making the membrane in an attempt to overcome the obstacles faced.

Cellulose acetate is one of polymer that is widely used in the manufacture of membranes. It was produced by the acetylation of cellulosic biomass such as Sengon wood (*Paraserianthes falcataria sp*). Production process of cellulose acetate polymer is more environmentally friendly than the production of polymers derived from fossil fuels because the product is produced from biomass [7].

In general, the phase inversion method of membrane preparation involves three main components: a polymer, solvent, and non-solvent. There are several factors that affect the characteristics of the resulting membrane, such as dope solution composition, polymer concentration, choice of solvent / non-solvent and evaporation time [5]. Solution viscosity of the mixture components are also dependent on the polymer molecular weight and concentration, type of solvent, and additives. While the performance of the membrane is determined by the value of the flux. Flux values are very dependent on the viscosity of the dope solution, where the value is closely related to the molecular weight of the polymer membrane materials used. The larger the molecular weight of the polymer, the more viscous polymer solution and it is very influential on the formation of the membrane and the resulting performance. In this study, the raw material used is cellulose diacetate from wood sengon pulp cellulose by acetylating process with a molecular weight of 130 221 and acetyl content of 39.66% [10]. In general, the manufacture of cellulose acetate membranes with polymers having 30,000 – 50,000 of molecular weight at room temperature to obtained homogeneous solution.

The aim of this paper to investigate the effect of the dope solution temperature variation in order to obtain a balanced dope solution viscosity to yield a well capable of forming membranes.

II. MATERIALS AND METHODS

Cellulose diacetate (acetyl content 39.66%) was the polymer used which was produced by acetylating cellulose of sengon wood in earlier research [10], N,N, dimethylformamide (DMF) (Merck, Germany) was used as the solvent, distilled water, dextran (MW 9,500 Da and 12,000 Da) (Aldrich, UK) were used as feed solution. The preparation method of cellulose diacetate (CA) membrane ultrafiltration made by a phase inversion. The flow diagram of the formation of membrane showed in Fig. 1.

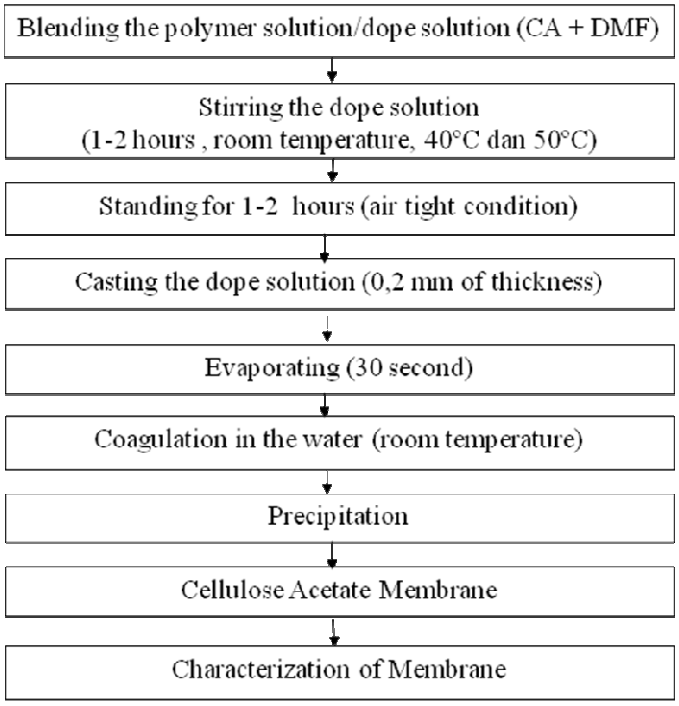

Fig. 1. Flow diagram of the formation of membrane

Cellulose acetate (CA) with a polymer concentration of 18% was mixed with the solvent DMF using a magnetic stirrer in the vessel for 1-3 hours at room temperature , 40ºC and 50 ºC to obtain a homogeneous solution called dope solution. The dope solution was subsequently allowed to stand about 1-2 hours in an airtight condition in order to remove air bubbles. Membrane obtained by pouring a solution of dope on the smooth glass plate, and casted using the applicator YBA 3 (brand Yoshimitsu, Japan) at 0.2 mm thickness as cast. The thin layer is left at least 30 seconds for evaporating of DMF followed by immersion in the water coagulation bath at room temperature to form a thin layer of membrane. The result of membrane were washed with distilled water in large quantities and stored in water before characterized (modification [10,4,8]. The characterization of membrane include the morphology, molecular weight cut off (MWCO), flux, and rejection.

III. RESULTS AND DISCUSSION

A.Membrane Morphology

The formation of CA membrane by phase inversion using DMF solvent and water as non-solvent will follow instantaneous demixing mechanism. The shortes membrane formation mechanism (instantaneous) will provide the membrane with bottom layer are more porous and the top layer with dense pore. The results of Scanning Electron Microscope (SEM) JSM-5000 LV, Jeol-JAPAN analysis of the membrane morphology produced can be seen in Fig. 2.

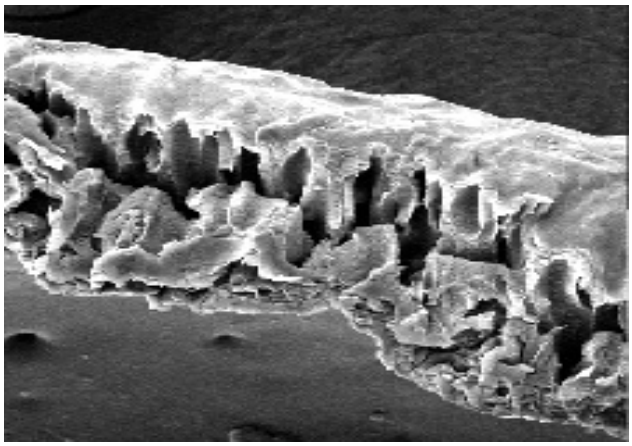

Fig. 2. Cross section structure of the CA membrane 18% dope solution at room temperature at 100x magnification.

From Fig. 2 showed that cross section structure membrane produced was an asymmetric membrane because the structure of the bottom had pore size larger than the top. The result of SEM analysis for CA membrane morphology on *dope* solution temperature variations was seen in Fig. 3 and Fig. 4.

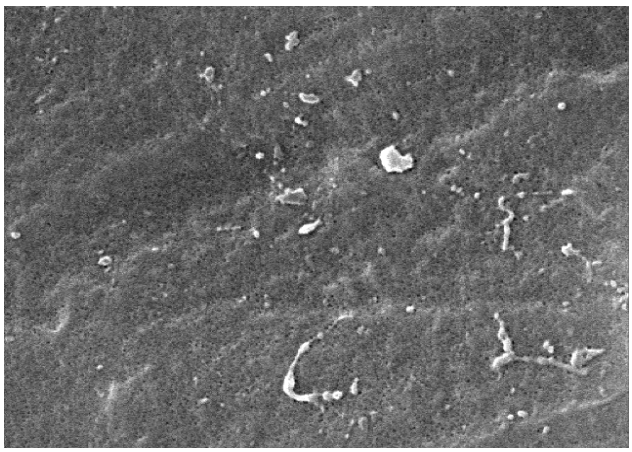

Fig. 3. Surface structure of the CA membrane 18% dope solution at room temperature at 5000x magnification

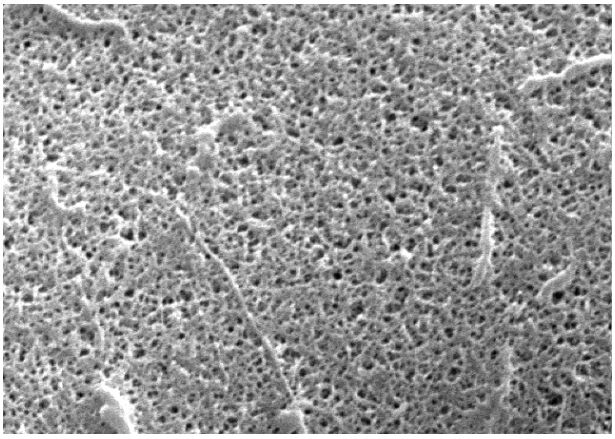

Fig 4. Surface structure of the CA membrane 18% dope solution at 50°C at 5000x magnification.

At 5000x magnification shown in Fig 3, the pore size was relatively smaller produced of SDA membrane 18% dope solution at room temperature than the pore size of polymer at dope solution temperature 50°C. This is because an increase in dope solution temperature causing molecular bond polymer stretching and increasing of solution density so that the solution viscosity is decreased. In these conditions cause diffusion between DMF and non solvent is faster so that pores produced in the membrane is also more. The functional group of actyl in cellulose acetate membrane was analyzed by IR Prestige-21. FTIR's analysis shows typical group absorption of carbonyl C=O in the number of wave 1751 cm-1 and group of Acetyl C-O in number wave of 1250 cm-1, can be seen in Fig. 5 and it is to prove the cellulose acetate was formed.

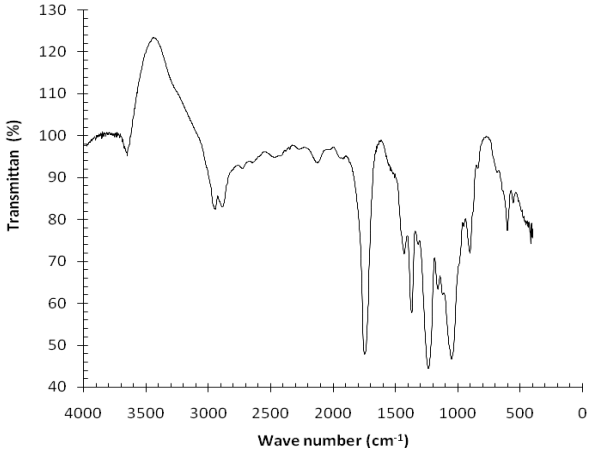

Fig.5. FTIR's analysis of cellulose acetate membrane

B. Membrane Performance (Flux and Rejection)

Flux measurements performed with each feed stream (water and dextran) using ultrafiltration module flow system dead end filtration. Membranes used in the form of a flat sheet with the surface area of 12.56 cm^2 (12.56 x 10-4 m^2) on the trans membrane pressure (TMP) 1 bar. Membrane characteristics of flux (water and dextran) of the membrane produced in dope solution temperature variation with CA concentration 18% can be seen in Fig 6.

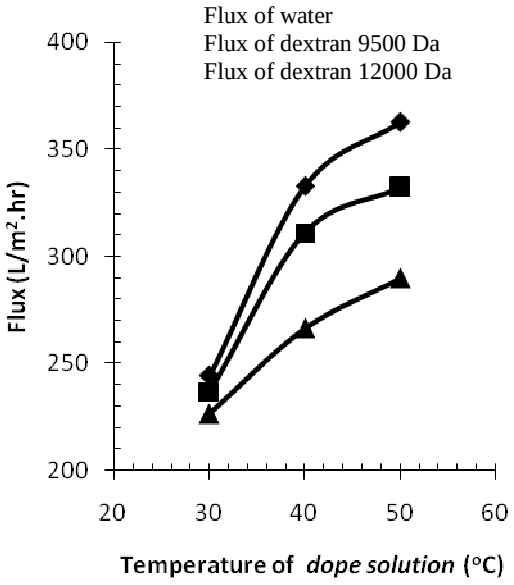

Fig 6. The relation between water flux, the dextran standard solution 9500 Da and 12000 Da (TMP 1 bar) at various temperatures dope solution.

From Fig 6 is seen that at the concentration of polymer solution constant, an increase in flux with increasing temperature of the polymer solution. It can be explained that with increasing dope solution temperature, the viscosity of the polymer be decrease and solvent is more volatile so that the solvent and non-solvent diffusion in the coagulation bath becomes faster. This resulted in a pore that is formed will be greater [1]. This condition makes the flux increases with rising temperature polymer solution. The highest flux produced at a temperature of 50°C dope solution respectively for water, dextran 9500 Da and dextran 12000 Da is equal to 362.459 L/m^2.hour; 332.087 L/m^2.hour, and 289.669 L/m^2.hour.

Membrane performance is also determined from the value of rejection. Measurements rejection was carried out on standard solutions of dextran with a molecular weight of 9,500 Da, and 12,000 Da which initial concentration 500 ppm is passed on the module the membrane with TMP 1 bar. Accommodated permeate final concentration analyzed using a UV-VIS spectrophotometer Shimadzu 1700 Rejection is a measure of the ability of the membrane to withstand a certain species.

Dextran rejection of the membranes with various concentrations and temperatures dope solution can be seen in Fig 7. When viewed from the acquisition of rejection, for the rejection of dextran 12,000 Da dextran obtained higher than 9,500 Da (Figure 7). It is thought at 9,500 Da dextran, many dextran molecules are not able to retained by the membrane due to the membrane pore size is still large. The highest rejection obtained on dope solution room temperature and the lowest rejection on dope solution temperature of 50°C. Effect of dope solution temperature of the membrane rejection coefficient contradicts the flux values, where the higher temperatures used dope solution, the lower the coefficient obtained rejection. This condition shows that the viscosity of dope solution that higher (dope solution

temperature is low) will form a membrane denser pore , resulting in the phenomenon of rejection coefficient decreases as the temperature increases dope solution [1].

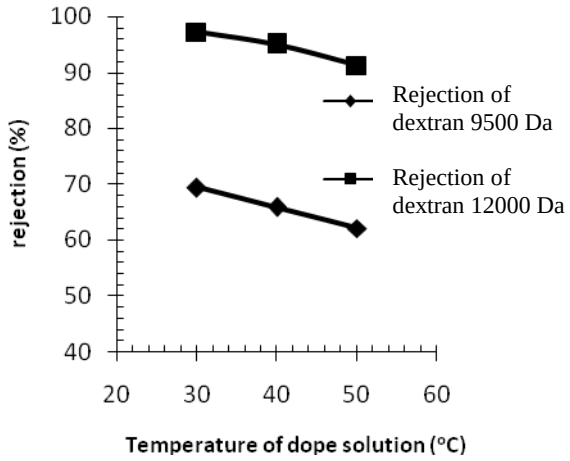

Fig 7. The relation dextran rejection 9500 Da dan 12000 Da (TMP 1 bar) at various dope solution temperature.

C. Molecular Weight Cut Off (MWCO)

Pore size of the membrane is done through MWCO using a standard solution which known molecular weight. Molecular Weight Cut-Off (MWCO) is defined as the molecular weight of a solute that can direjeksi 90% by the membrane [5]. The dextran rejection 9500 Da at various dope solution temperatures is about 62.13% - 69.39%. Membrane selectivity increased to 12000 Da dextran standard solution, where the membrane rejection reaches 91.21% - 97.24%. Therefore, the CA membrane produced is identical to pore size 12000 Da due to capable of holding solute up to 90%. The resulting membrane is included as ultrafiltration according to the Osaga and Nakagawa statement in Rosnelly [11] that the ultrafiltration membrane have a pore size with MWCO about 103-106 Da.

IV. CONCLUSION

Based on SEM analysis was known that the resulting cellulose diacetate membrane including asymmetric membrane. The highest flux was obtained 362.459 L/m^2.hour at a temperature of 50°C for dope solution, while the lowest flux was obtained 244.34 L/m^2.hour in dope solution at room temperature. Instead, the highest rejection was obtained at dope solution room temperature and the lowest rejection at 50^{o}C. The pore size of membrane obtained based MWCO on CA membrane 18% for various dope solution temperature about 12000 Da.

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