The performance of double layer structure membrane prepared from flowing coagulant

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Abstract. Membrane with double layer structure is favourable as it exhibits smooth surface and macrovoids free structure. However, its' performance in terms of permeability, porosity and strength has not been studied thoroughly. Additionally, the effect of flowing coagulant on the formation of double layer membrane has not been reported. Thus, the objective of this study is to investigate the performance of double layer membranes, which were prepared using flowing coagulant. Results showed that when the coagulant flow changed from laminar to turbulent, the pure water permeation of the membrane increased. It was due to the higher porosity in the membrane, which prepared by turbulent flow (CA-Turbulent) compared to the membrane which fabricated under laminar condition (CA-Laminar). This can be explained by the rapid solvent-coagulant exchange rate between the polymer solution and the turbulent coagulant. In term of strength, the tensile strength of the CA-Turbulent was ~32 MPa, which was 100% higher compared to CA-Laminar. This may due to the presence of large amount of nodules on its surface, which reduced the surface integrity. In conclusion, flowing coagulant altered the membrane properties and adopting turbulent coagulant flow in membrane fabrication would improve the porosity, surface roughness and the strength of the membrane.

1. Introduction

Polymeric membranes are widely used for separation purpose due to low cost [1], well developed module technology and available in varied materials [2]. However, polymers such as polyethersulfone (PES) and polyvinylidene fluoride (PVDF) are hydrophobic in nature, which exhibit high fouling tendency. Natural polymer like cellulose acetate (CA) is weak in mechanical strength which limits its usage in high pressure operating condition. Thus, efforts have been done to improve the membrane properties by adding new additives into the formulation [3-4], introducing new fabrication technique [5] and surface modification [6-7].

Double pass casting technique was first introduced by Dang et al. [8] and this technique improved the surface roughness of hydrophobic PES membrane. Besides, the researchers also observed macrovoids free structure in the modified hydrophilic PES membrane. However, this fabrication technique has not been studied extensively by the researchers, especially on different types of material.

The effect of the flowrate of bore fluid in fabricating hollow fibres membrane has been widely studied by the researchers. The finding from Alsalhy et al. [9] showed that high bore fluid rate produced PES membranes with high water flux property by altering the pore size and the porosity of the membranes. Similar finding also reported by Qin et al. [10] where the flowrate of the bore fluid affected the selectivity of gas membrane. In flat sheet membrane fabrication, the coagulant plays the same role as the bore fluid in introducing the phase inversion process. Nevertheless, the effect of flowing coagulant on the performance of flat sheet membranes has not be studied.

Therefore, the objective of this study is to bridge the gap in the existing research literature by investigating the performance of double layer structured CA membranes, which were fabricated using flowing coagulant. The membranes were evaluated in terms of water permeation, hydrophilicity and tensile strength.

2. Materials
CA (average molecular weight, 100,000, ACROS Organics™) was used as the membrane forming material and acetic acid glacial (Fisher Scientific) was used as the solvent. All the chemicals were used as received.

3. Methodology

3.1. Membrane fabrication
CA solution was prepared by mixing 23 wt% of CA in 77 wt% of acetic acid at 65 °C for 4 hours at 250 rpm. The solution was cast by using the self-fabricated casting machine at 4.74 m/s [11] once the bubbles was removed. The cast CA solution was placed the flowing coagulant to form the membrane. RO water was used as the coagulant. Three types of coagulant flow were studied, which were laminar, transition and turbulent. The type of flow was determined by calculating the Reynolds number. The membrane was labelled according to the type of flow. CA-Laminar indicates that the CA membrane was immersed in a laminar flow of RO water during the phase inversion process.

3.2. Pure water permeation (PWP)
The flux of membrane was determined by conducting the ultrafiltration experiment at 3 bar using Milipore stirred cell. The effective membrane area was 13.4 cm^2. The experiment was conducted for 3 hours to collect the permeate.

3.3. Tensile test
The mechanical strength of the membrane was determined by using Shimadzu AG-X Universal Tensile Machine, according to the ASTM D882-09 [12].

3.4. Contact angle measurement
3 µL of water was placed on the surface of the membrane using the sessile drop method. Ten contact angle data was collected for each membrane using the Attension Theta Automatic Goniometer.

3.5. Porosity
Gravimetric method was adopted to measure the porosity of the membrane. The porosity was calculated by using the following equation (1):

$$\varepsilon = \frac{(w_a - w_b)/\rho_{water}}{(w_a - w_b)/\rho_{water} + w_b/\rho_{membrane}} \qquad (1)$$

where w_a and w_b are the weight of wet membrane and dry membrane respectively. Meanwhile, ρ_{water} refers to the density of water and $\rho_{membrane}$ is the density of membrane

4. Result and discussion

PWP and porosity of the CA membranes are shown in figure 1. When the flow of RO water increased from laminar flow to turbulent flow, PWP of the membrane increased. It was due to higher porosity of the CA-Turbulent membrane compared to CA-Laminar membrane. During phase inversion process, rapid solvent-coagulant exchange between CA solution and RO water resulted a more porous structure in CA-Turbulent [13]. It was due to high solvent concentration gradient between CA solution and RO water [14].
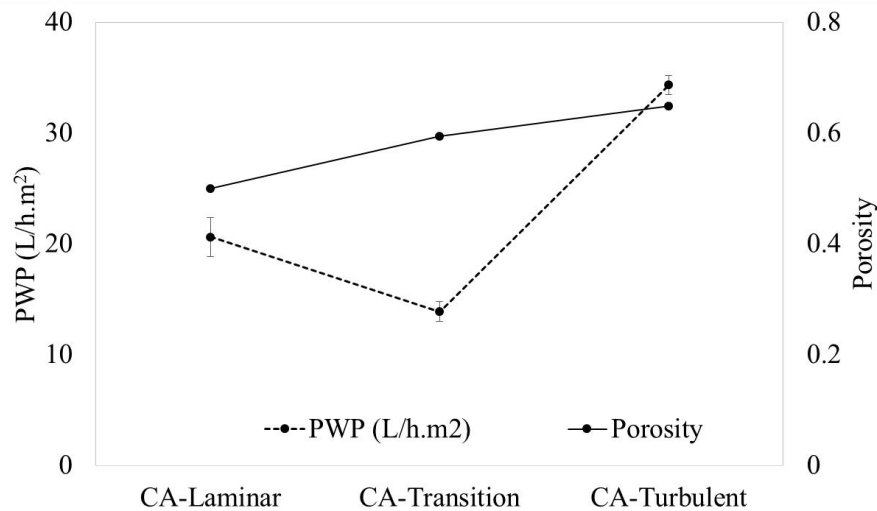

Figure 1. The performance of CA membranes in terms of PWP and porosity.

When the CA solution contacted with RO water, acetic acid would leave the CA solution and mixed with RO due to its high water miscibility. Hence, concentration of acetic acid in the RO water was increased as the contact time increased. This implied that the solvent-coagulant exchange rate would be reduced. However, in turbulent flow, the high concentration gradient between CA solution and RO water was maintained at its optimum condition as fresh RO water was pumped into the system continuously during membrane formation process. Short contact time between CA solution and RO water due to the high velocity of RO water also contributed in maintaining the high concentration gradient condition between these two mediums. Comparatively, during the formation of CA-Laminar membrane, exchange rate was slower and thus porosity was approximately 23% lower compared to CA-turbulent membrane.

Table 1 shows the tensile strength and hydrophilicity of CA membranes. Generally, CA membranes were hydrophilic as the contact angle was less than 90°. Type of flow did not affect the hydrophilicity of membrane as the contact angle was ~73° for all the membranes. In terms of strength, CA-Transition and CA-Turbulent membranes showed ~33MPa where CA-Laminar membrane exhibited the lowest tensile strength, which was ~15MPa. This was due to the presence of large quantity of nodules on the membrane surface, which reduced the surface integrity[14]. Comparatively, strength of the membranes produced in this study was higher than the CA membranes produced by Etemadi et al. [15] and Li et al. [16], which exhibited ~5.2MPa and ~3MPa, due to double layer macrovoids free structure.

Table 1. The physical properties of the CA membranes prepared under different flow conditions.

Membrane ID	Tensile strength (MPa)	Contact angle (º)	Description on the morphology of the membrane [14]
CA-Laminar	15.1886 ± 3.5731	73.0948 ± 3.8080	−Rough, with large quantity of nodules on the

			surface. −Macrovoids free structure
CA-Transition	35.6778 ± 5.8424	72.8586 ± 3.4277	− Rough, with moderate quantity of nodules − Macrovoids free structure
CA-Turbulent	32.2607 ± 4.7077	72.4780 ± 2.7308	− Smooth, with small quantity of nodules − Macrovoids free structure

5. Conclusion
This study showed that the flow of coagulant affect porosity, PWP and tensile strength of CA membranes. Turbulent coagulant produced more porous membrane compared to laminar flow due to rapid solvent-coagulant exchange rate. This explains for the higher PWP in CA-Turbulent membrane compared to CA-Laminar membrane. Results also showed that CA-Turbulent membrane exhibited good strength property due to smooth surface and macrovoids free structure. In conclusion, turbulent coagulant produced smooth porous membrane with good strength property, which is desired for wide range of separation purpose.

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