Characteristics of Cellulose Acetate/Polyethylene Glycol membrane with the addition of Graphene Oxide by using surface coating method

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Abstract. Fabrication of membrane for desalination process with high performance is the objective of this study. The addition of Graphene Oxide (GO) in cellulose acetate/polyethylene glycol (CA/PEG) with a ratio of 80:20 was prepared by a surface coating method. In this study, the composition of GO was compared, with composition ranging from 0.0025 to 0.0125 wt% to the mass of the solvent. The characteristics of membranes were observed by using Scanning Electron Microscopy (SEM), Fourier-Transformed Infra-Red (FTIR), contact angle analysis and Dynamic Mechanical Analysis (DMA). While its performance was observed by analyzing salt rejection, flux permeate and permeability of the membranes. SEM analysis shows that membrane exhibits asymmetric membrane with sponge-like structure. FTIR results show that the peak area of –OH bonding decreases with the addition of GO. This result is in agreement with the contact angle analysis. The experiment results shows that membrane with the best performance is CA/PEG membrane with the addition of 0.01 wt% GO with salt rejection (%R) of 37%, flux permeate (F) of 1356 L/m^2.h, and permeability (P) of 0.0013 L/m^2.h.kPa.

1. Introduction

One of the most important insights in providing clean water is saline water desalination, which is considered capable of meeting global clean water needs. The separation technique with the principle of Reverse Osmosis using membranes, is currently a rapidly developing technique compared to thermal desalination with the evaporation process. Reverse Osmosis is a separation process technology by using a semipermeable membrane by the process of forcing solvents from areas with high solute concentration to areas with low concentrations by applying pressure over osmotic pressure.

Membrane research currently being developed aims to improve membrane performance such as flux, salt rejection and surface morphology, and hydrophilicity. All parameters are influenced by the type, composition of membrane-forming raw materials and the methods used [1].

Fabrication of membrane has a variety of processes, in this study the phase inversion method is used, which is a method where the mixture of solvents and polymers is drained on a glass plate and immersed in non-organic substances, generally water. The material used for membrane synthesis is a mixture of polymeric substances, this was chosen because the polymer is easily formed and strong [2]. Of the

several types of polymers, Cellulose Acetate (CA) was chosen for use in the desalination process because it has the potential to produce high flux, has an economical price, and high hydrophilicity [3]. The effect of the addition of GO by surface coating method has been studied in this study by characterized the membrane property and performance.

2. Materials and method

2.1. Materials
Cellulose acetate (CA, 30.000 Da, with acetyl content 39,8%), polyethylene glycol-400 (PEG, 400 Da) are purchased from Sigma Aldrich. Graphene oxide (GO) with bulk density of 1,8 g/cm^3, was purchased from Sigma Aldrich. Aceton and Dimethylformamide (DMF) was used as solvent and water as non-solvent.

2.2. Method
Polymeric membrane of CA/PEG was prepared by adding 2 gram of CA and 0.5 gram of PEG were dissolved in 17 mL of acetone/DMF (50/50 vol%). Then, the solution heated at 70°C for 12 hours. After that, membrane was casted and immersed in water, resulting CA/PEG membrane. After that, CA/PEG membrane dip-coated in GO solution. GO solution was made by dissolving GO in distilled water with variable composition, which are 0, 0.0025, 0.0050, 0.0075, 0.0100, and 0.0125 wt% of membrane solvent and named as CGS 0, CGS 1, CGS 2, CGS 3, CGS 4, and CGS 5, respectively.

2.3. Membrane characterization
CA/PEG/GO membrane characterization was observed by its hydrophilicity, with Fourier-Transformed Infra-Red (FTIR), and Contact Angle analysis, while its morphology was observed by Scanning Electron Microscopy (SEM).

2.4. Membrane performance
Percentage of salt rejection is the efficiency of membrane and its ability to remove contaminates and calculate by using this equation: $R=(1-C_p/C_f)$ x 100%, where R is the percentage of salt rejection, C_p (in ppm) is the salt concentration of the permeate, C_f (in ppm) is the salt concentration in the feed water. The permeate flux (J) represents the amount of pure water collected per unit time and per unit area at variable pressures. It was calculated by: $J = Q/t \times A$, where J is the permeate flux ($mL/h.m^2$), Q is the amount of permeate (mL), t is the time and A is the area (m^2).

3. Results and discussions

### 3.1. Fourier Transformed Infra Red (FTIR) characterization
The results of FTIR analysis for CA / PEG membrane with the addition of GO using the surface coating method can be seen in Figure 1. From the results of the FTIR analysis the area of peak -OH cluster calculation can be seen in Table 1.

There was a slight increase in the peak area of the -OH group on the CA / PEG membrane variable by the addition of GO using a surface coating method. There are also some variables that experienced a significant decrease in the peak area of the -OH group, namely the CGS 1 variable and the CGS variable 4. The decrease in the peak area of the -OH group shows the hydrophilicity of the CA / PEG / GO membrane with a decreased surface coating method.

The decrease of CA / PEG / GO membrane hydrophilicity by surface coating method on the addition of GO variable 0.0025 wt% solvent to the addition of 0.0125 wt% solvent GO can occur because the morphological structure of the surface coating method shows a tendency towards wider sponge-like pores compared to CA / PEG membranes without the addition of GO.

Surfaces that tend to be dominated by sponge-like pores are caused because sponge-like pores are formed due to the process of solvent displacement when evaporation occurs slowly [4]. The slow

evaporation process occurs due to the synthesis of CA / PEG / GO membrane by surface coating method through a longer immersion process.
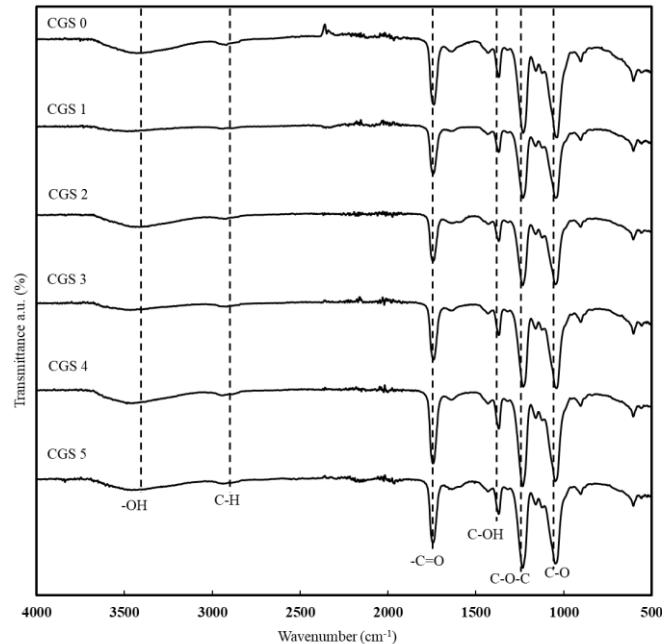
Figure 1. Results of Fourier Transform Infrared Spectroscopy Membrane CA / PEG / GO surface coating method.

Table 1. Peak -OH Area on the FTIR graph CA / PEG / GO surface coating method

| Sample code	-OH peak area (area unit)
CGS 0	2319.67
CGS 1	1036.67
CGS 2	2107.00
CGS 3	1181.00
CGS 4	2337.00
CGS 5	2155.00

3.2. Contact angle analysis

Hydrophilicity analysis also plays an important role in determining membrane performance in the desalination process. The decrease in the contact angle value that occurs in almost all surface coating variables shows that GO affects the membrane surface which causes the membrane surface to be more hydrophilic. In Table 2 the CGS 4 variable has the lowest contact angle value. This can be attributed to the results of sponge-like pore area measurements where the CGS 4 membrane has increased pore area.

Table 2. Contact angle values on CA / PEG / GO membrane surface coating method

Sample code	Contact angle (o)
CGS 1	43.210
CGS 2	42.940
CGS 3	43.190
CGS 4	40.363
CGS 5	42.208

### 3.3. Scanning Electron Microscopy characterization (SEM)

Results of SEM analysis for CA / PEG membranes with the addition of GO using a surface coating method can be seen in Figure 2. The pore morphological structure of the CGS membrane that shows a dominant sponge-like pore is due to the addition of GO carried out after the CA / PEG membrane has been made, so the GO effect to the formation of pores on the inside of the membrane is reduced, as evidenced by the reduction in finger-like pores on several membrane variables. Likewise, the pore structure of the outer layer tends to experience thickening which is dominated by sponge-like pores. A sponge-like pore diameter measurement was also carried out on the CA / PEG / GO membrane surface coating method. The sponge-like pore diameter tends to decrease in size, but in the CGS variable 4 the sponge-like pore diameter size increases.
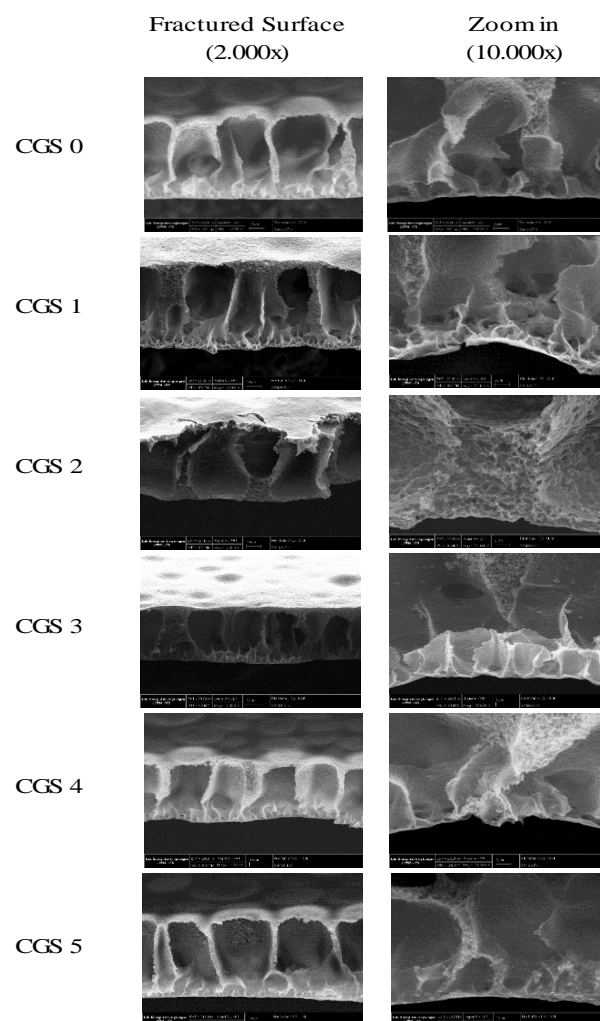
Figure 2. Results of Scanning Electron Microscopy Membrane CA / PEG / GO surface coating method.

3.4. Salt rejection and flux permeate

Results of the analysis of the performance of salt rejection on CA / PEG membranes and the addition of GO by using a surface coating method can be seen in Figure 3 and Figure 4.
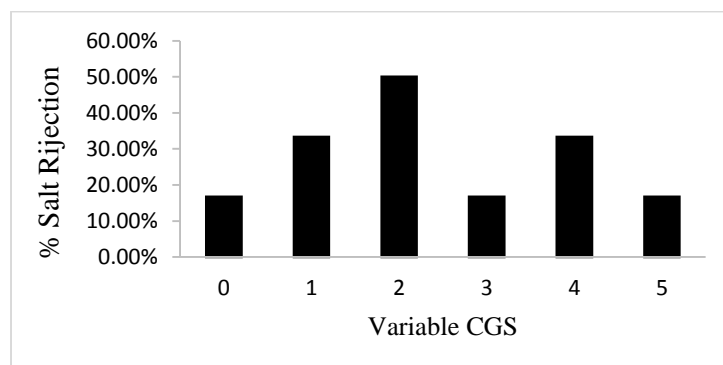
Figure 3. Salt rejection of CA / PEG / GO membranes.

Salt rejection in CA / PEG / GO membrane surface coating method achieves the highest rejection of CGS 2 membrane variable and decreases in CGS 3 variable, it because adding too much GO will cause an increase in finger like pore and sponge like pore on the membrane [5] and on the CA / membrane PEG with the addition of GO through the surface coating method there is a tendency to increase the flux value along with the increase of GO added to the membrane. However, there is a decrease in flux in the addition of GO in the highest amount in the CGS variable 5. This can occur due to the addition of additives using a coating method which can cause additional obstacles caused by the coating layer on the membrane [6].
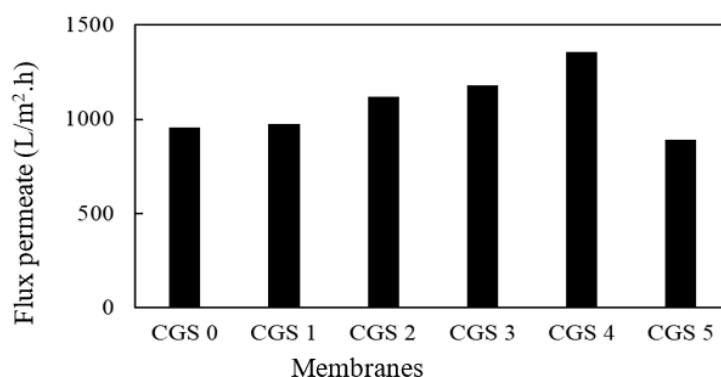
Figure 4. Permeate flux of CA / PEG / GO membranes.

4. Conclusion
The addition of GO with a surface coating method has an effect on increasing the hydrophilicity and performance of CA / PEG membranes with the best results on the CGS 4 variable with the addition of GO at 0.0100 wt% which results in the value of% R = 37%, F = 1356 L / m^2.h, and P = 0.0013 L / m^2.h.kPa. Regarding the salt rijection, this value is still very low, need more study to improve it.

Acknowledgments

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