

EFFECT OF HYBRID $\text{Mg}(\text{OH})_2$ /CHITOSAN ON THE HYDROPHILICITY AND ANTIFOULING OF POLYETHERSULFONE (PES) MEMBRANE

U. Fathanah[✉], I. Machdar, M. Riza, N. Arahman, M. Y. Wahab, S. Muchtar, C. M. Rosnelly, S Mulyati, Y. Syamsuddin, S. Juned and F. Razi

Department of Chemical Engineering, Universitas Syiah Kuala, 23111, Banda Aceh, Indonesia

[✉]Corresponding Author: umifathanah@unsyiah.ac.id

ABSTRACT

This work presents the preparation and modification of hybrid $\text{Mg}(\text{OH})_2$ /chitosan blended PES membranes with the aim of improving the hydrophilic, antifouling and performance of the resulting membranes. The deposition of additives by blending the polymer with a concentration of 1%, 3%, 5%, and 7% on PES membranes. The influences from varying concentrations of additives were evaluated by analyzing changes in chemical groups, changes in morphological structure, hydrophilicity degree, porosity, filtration properties, and the antifouling propensity of the fabricated membrane. PES membranes with the addition of 5% additive (MC-5) showed the best performance biggest degree of hydrophilicity compared to the other membranes. In terms of performance, the MC-5 membrane produces a PWP of almost 15 folds bigger than that of unmodified PES, with a rejection value of 68.2% and a water flux recovery ratio (FRw) of 82.7%.

Keywords: Polyethersulfone (PES), Hybrid $\text{Mg}(\text{OH})_2$ /Chitosan, Blending Polymer, Antifouling.

RASĀYAN J. Chem., Vol. 15, No.2, 2022

INTRODUCTION

In recent decades, the separation process based off membrane technology has been widely utilized for application in water purification and liquid waste treatment process. This process is broadly employed to remove many kinds of unwanted contaminants in water.^{1,2} Membrane separation technology has been adopted in various industries as it offers advantages, including low usage of energy, uncomplicated setup and produces permeate of high quality.^{3,4}

Amongst uncountable polymers that are commonly employed in the manufacture of membranes for nanofiltration (NF), ultrafiltration (UF) and microfiltration (MF) process is Polyethersulfone (PES). Impressive characteristics of PES such as outstanding mechanical strength, exceptional chemical stability, high glass transition temperature (T_g), and high resistance to oxidation processes^{5,6} make PES one of the macromolecular polymers that really attract the attention of researchers to be exploited in the manufacture of asymmetric membranes. But, unfortunately, PES is hydrophobic, so it is prone to causing fouling which can reduce membrane performance⁷. During filtration, solutes that fail to infiltrate the PES film will be easily ingested on the surface of the hydrophobic polymer network resulting from the decline in permeability and general performance of the membrane.

Efforts to improve membrane performance have been always studied and developed. In recent years, various attempts at membrane modification have been reported, for instance by incorporating polymers such as polyethylene glycol (PEG) and nanoparticles such as Ag, TiO_2 , SiO_2 .⁸⁻¹⁰ The membranes prepared through such modifications were reported to have increased hydrophilicity, porosity, and increased water flux, as well as anti-fouling capabilities. Studies regarding the fouling resistance of PEG against a large number of biofoulants have been reported. Likewise, the nanoparticles Ag and TiO_2 have also been utilized to improve membrane characteristics. However, there are concerns about their toxicity during washing. Therefore, it is necessary to explore the safe and inexpensive hydrophilic additives for membranes, such as chitosan and $\text{Mg}(\text{OH})_2$.

Chitosan is a polyaminosaccharide derived from chitin which is generally used in the synthesis of hybrid materials because of its biocompatibility, pH sensitivity and easy, inexpensive and environmentally friendly

gel formation.^{11,12} Chitosan incorporates a positive charge in an acid solution which can be ascribed to its atomic chain which is rich in glucosamine.^{13,14}

Moreover, $\text{Mg}(\text{OH})_2$ is also considered a good material with structurally bound OH groups.¹⁵ $\text{Mg}(\text{OH})_2$ is a chemical that is easy to afford in terms of availability and cost, harmless, often found as an additive in medications and foods^{16,17}

The development of hybrid additives such as chitosan and $\text{Mg}(\text{OH})_2$ is very interesting to be researched and utilized in membrane technology. Previous work reported that the superposition effect caused by the chemical interaction between $\text{Mg}(\text{OH})_2$ and chitosan resulted in increased antifouling properties and membrane hydrophilicity.¹⁸ However, the influences increasing the concentration of hybrid $\text{Mg}(\text{OH})_2$ /chitosan (MC) on the traits as well as the performance of membranes need further investigation. In this work of research, we aim to investigate the influence of MC additive on the traits and performances of the modified PES membrane. The modification of membrane with additive was conducted by means of polymer blending technique. Characterizations of the membrane are carried out by analyzing the membrane's morphological structure, chemical composition, water contact angle, porosity, tensile strength. Meanwhile, membrane performance is evaluated by measuring the permeability of pure water, humic acid removal and the antifouling properties of the resulting membrane.

EXPERIMENTAL

Materials

Fabrication of membrane was carried out using chemicals such as PES (Ultrason E6020 BASF Co, Mw 65000, Ludwigshafen, Germany) as the supporting polymer, and solvent N-methylpyrrolidone (NMP) (Merck, Hohenbrun, Germany). For additives, magnesium hydroxide ($\text{Mg}(\text{OH})_2$) which was obtained from Xinglu Chemical Tech Co., China and chitosan (Sigma Aldrich, Germany) was employed. Humic acid (HA, Sigma Aldrich, Germany) was used as a foulant model, meanwhile Deionized (DI) water was employed for nonsolvent and other use.

Preparation of PES Membrane (MC-0)

The preparation of the membrane was carried out by NIPS technique. Initially, PES with a concentration of 18% by weight was dissolved into 82%w/w NMP solvent. The polymer and solvent were dissolute altogether by vigorous stirring. After the PES-NMP solution reached homogeneity, the casting of membrane film was carried out on the support media using a laboratory flattening knife at a thickness of 254 μm . The cast media was afterward dipped in the bath containing non-solvent, which in this experiment we used DI water. Furthermore, the membrane was kept in DI water before being used for filtration tests and membrane characterization.

Preparation of Hybrid $\text{Mg}(\text{OH})_2$ /Chitosan (MC)

The preparation of the MC solution was conducted by referring to the procedure done by Munnawar et al.¹⁹ 1 gram of each $\text{Mg}(\text{OH})_2$ and chitosan were dissolved in acetic acid 1% (volume 100 ml) then stirred to homogeneity at a temperature of 50°C. This homogeneous solution is then used as an additive in the modification of membranes.

Modification of Membranes

18% (of total solution weight) PES was added with hybrid MC with a varied concentration of 1%, 3%, 5% and 7% (of total solution weight) into NMP solvent in separate vials accordingly. After that, the solution was stirred at 50°C and once homogenous it was then cast on the support media followed by the membrane solidification process by inserting the cast media into the DI water-containing bath. Following that, the membranes were put in a container containing DI water for future use. The variation of membranes prepared in this research is detailed based on dope composition as shown in Table-1.

Characterization of Membranes

To understand the effect of hybrid MC addition on the PES membrane, basic membrane characterizations such as spectral analysis to determine changes in the membrane functional groups after modification using FTIR-ATR (Thermo Scientific iD5 ATR-Nicolet iS5 FTIR Spectrophotometer, Japan), morphological structure of the membrane using scanning electron microscopy (FE-SEM, JSF-7500F, JEOL Co., Ltd.,

Japan), membrane's hydrophilicity degree assessment using Drop Master 300 Kyowa Interface Science Co. (Japan) instrument, membrane's strength measurement using Autograph AGS-J, Shimadzu Co. (Japan) tensile machine (adapting the ASTM D 638-14 standard) were carried out. Meanwhile, membrane porosity was determined using Eq.-1.¹³

$$\varepsilon = \frac{(\omega_w - \omega_d)}{\rho \times A \times l} \times 100\% \quad (1)$$

Where, ω_w and ω_d are the weights of membrane specimens before and after drying (g), ρ denotes the default water density (1 g/cm³), A and l symbolize the area (cm²) and thickness (cm) of the membrane film, respectively.

Tabel-1: Membrane Identity based on Composition Ratio

Membrane	PES (%)	Mg(OH ₂)/Chitosan (%)	NMP (%)
MC-0	18	0	82
MC-1	18	1	81
MC-3	18	3	79
MC-5	18	5	77
MC-7	18	7	75

Membrane Performance Evaluation

The membrane filtration performance was determined from the membrane pure and HA water permeability as well as HA rejection by using an ultrafiltration cell with a dead-end type. Before the actual experiment, the membrane was initially compacted until the water flux was stabilized. After that, for the measurement of pure water permeability (PWP), DI water was infiltrated into the membrane, then the resulting permeate was weighed using a digital scale. The PWP (L/m².h.bar) was mathematically computed using the following Eq.-2:

$$PWP = \frac{Q}{A \times \Delta t \times \Delta P} \quad (2)$$

The Q in the equation refers to the amount of filtered water (L), A for the membrane's surface area (m²), Δt is the duration of the process (hour), and ΔP is the given pressure during filtration (bar).

To test the membrane's selectivity properties, experiment for solute rejection experiment was performed using an artificial HA solution at 50 ppm, in which 0.05 grams of HA solids were dissolved in 1 liter of DI water. The HA rejection coefficient (R) can be calculated using Eq.-3:

$$R (\%) = \left(1 - \frac{C_p}{C_f}\right) \times 100 \quad (3)$$

Given, R as the coefficient of rejection (%), C_p as the concentration of filtered HA (ppm) and C_f as the concentration of original HA solution (ppm).

Antifouling Evaluation

The experiment for evaluation of antifouling performance was carried out using UF module dead-end type in which the feeds used were with DI water and artificial HA solution. The procedure was initially started by membrane compaction in which DI water flowed through the membrane sheet at a pressure of 1 bar until constant data of water flux were acquired. Subsequently, the measurement of PWP (J_{w1}) was taken place for one hour at a pressure of 1 bar followed by switching the feed from DI water to a 50 ppm HA solution and flux measurement was continued following the same procedure. The generated permeate from HA filtration was collected for determination of concentration using UV-Vis Spectrophotometer. After 1 hour of HA filtration, the membrane used for that experiment was cleaned through the backwash technique by adapting the procedure reported in earlier published work.³ Afterward, the cleaned membrane was used again for repeated flux measurement of DI water and HA solution in order to obtain the after-cleaning flux (J_{w2}) data. Data from this whole set of the process were utilized to calculate the recoverable value of water flux loss (FRw). FRw is a parameter used to indicate the resistance degree of the membrane to fouling through the effectivity of cleaning. Equation-4 was used to determine FRw:²⁰

$$FRw(\%) = \left(\frac{J_{w2}}{J_{w1}}\right) \times 100 \quad (4)$$

The water flux that was a loss in the cause of membrane fouling consists of reversible flux loss (R_r) which is defined as the flux that is restore-able flux with membrane cleaning. The other one is irreversible flux loss (R_{ir}) or the loss of flux that cannot be restored even after the membrane has undergone physical cleaning. The combination of both of these fouling-induced flux losses is called total flux loss (R_t). These three fouling behaviors can be determined using Equations 5-7:²⁰

$$R_t(\%) = \left(\frac{J_{w1} - J_{HA}}{J_{w1}} \right) \times 100 \quad (5)$$

$$R_r(\%) = \left(\frac{J_{w2} - J_{HA}}{J_{w1}} \right) \times 100 \quad (6)$$

$$R_{ir}(\%) = \left(\frac{J_{w1} - J_{w2}}{J_{w1}} \right) \times 100 \quad (7)$$

RESULTS AND DISCUSSION

Chemical Functional Groups

One of the ways to confirm the successful incorporation of MC hybrid additive on the membrane is by observing the chemical components using FTIR which results are shown in Fig.-1. As a comparison, the IR spectra of pure chitosan and $Mg(OH)_2$ samples are also displayed in Fig.-1a. The spectra of chitosan show some important transmission bands to identify characteristic functional groups such as that recorded in the range of wavenumber between 500 cm^{-1} to 4000 cm^{-1} . The most visible characteristic of chitosan is denoted by a wide peak at a wavenumber of $3400\text{--}3600\text{ cm}^{-1}$ which is the stretching vibration of the NH_2 and $-OH$ groups. Furthermore, the small peak at 2900 cm^{-1} is the band of the $-CH$ group. at 1656 cm^{-1} shows the stretching of the $C=O$ group of the amide (I) band, and NH group can be observed at 1571 cm^{-1} which is mainly sourced from the N-acetylated residue of the amide (II) band. Several other peaks with weak intention are also seen at 1422 , 1378 , and 1157 cm^{-1} representing the bending of CH group, OH , and asymmetric stretching of the COC band.^{21,22}

Whereas, in the spectra of the $Mg(OH)_2$ sample, the majority of the visible peaks are an indication of the presence of the OH group. As can be seen from the transmission peak with strong intensity at 3600 cm^{-1} . At the wavenumber of 3450 cm^{-1} , the stretching vibration of the hydroxyl (OH) group from the water molecule on the $Mg(OH)_2$ particle is also visible. In addition, there are also present several small peaks at 1012 cm^{-1} and 1634 cm^{-1} belonging to the bending and stretching vibration of the OH group.^{16,23}

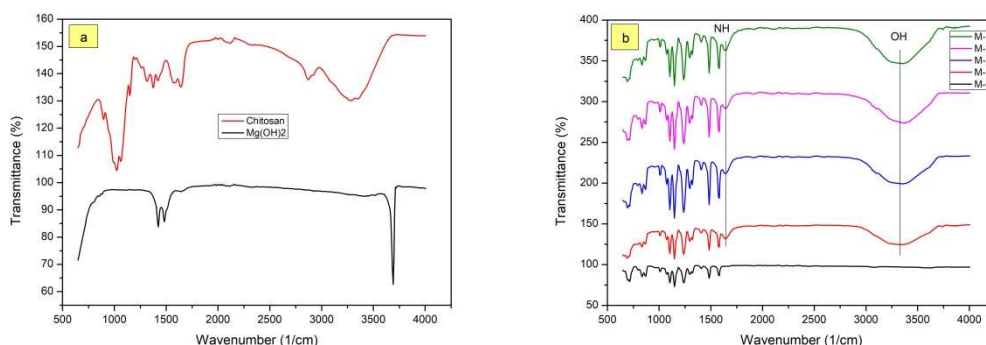


Fig.-1: IR Spectra of (a) Chitosan and $Mg(OH)_2$, and (b) Pure and modified PES membranes with Different Additive Concentrations

Figure-1b shows the IR spectra of chitosan and $Mg(OH)_2$ and the membrane modified with the hybrid MC additive at different concentrations. When compared with pure PES membranes, all modified membranes (MC-1 ~ MC-7) show slightly different FTIR spectra results. On the original PES membrane, it can be seen that the FTIR generated in the range of wavenumber of $1700\text{--}4000\text{ cm}^{-1}$ is a straight line without peaks,

meanwhile, after modification, several new peaks appear in the membranes specifically at wavenumbers of 1640 cm^{-1} and 3346 cm^{-1} which indicate the existence of NH and OH groups. These two groups are components obtained from the addition of chitosan and $\text{Mg}(\text{OH})_2$ where these functional groups have a vital part in improving the degree of membrane hydrophilicity.^{16,24} The emergence of these new groups also confirms that the hybrid MC additive has been successfully added to the PES membrane.

Membrane Morphology and Porosity

Figure-2 shows that the pure PES membranes and PES membranes modified with the hybrid MC at various concentrations have the same cross-sectional morphological structure as membranes produced via NIPS method in general. The morphology MC-modified membranes consist of two parts, namely the thin top layer (selective layer) and the macroporous bottom layer distinguished by finger-like or macrovoid-shaped pores.

Although they have similar morphology and the images are taken at the same magnification, if observed closely, there are differences in the characteristics of the cross-sectional structures of each membrane, especially in terms of the shape and size of membrane's finger-like pores. The cross-sections of the MC-1, MC-3, and MC-5 membranes show that there is improvement in pore properties such as size and number of finger-like macrovoid with increasing hybrid additive concentration. This is brought by the addition of hydrophilic MC particle in the casting solution resulting in thermodynamic instability which causes instant demixing in the phase inversion process in a non-solvent bath.²⁵ The thermodynamics of the polymer-additive-solvent-non-solvent system as well as the kinetics of the demixing process greatly influence the morphological structure of the membrane. The formation of macrovoid with a larger size is caused by instant demixing where the solvent enters at a higher rate into the membrane through the fracture formed due to the breaking of the skin layer. In addition, in the precipitation process in the coagulation bath, there is an aggregation between the PES polymer and hybrid $\text{Mg}(\text{OH})_2$ /chitosan particles to release the solvent from the system so that the pore size increases to become finger-like macrovoid on the membrane.^{25,26} However, there are detectable differences in the cross-sectional morphology of the MC-7 membrane prepared by adding a 7% concentration of $\text{Mg}(\text{OH})_2$ /chitosan hybrid additive. The number of finger-like and macrovoid pores on the MC7 membrane appeared to be less with a shorter size and there is also a visible dense area on the membrane matrix. This is due to the delay in the phase separation process which disrupts pore formation due to the increased viscosity barrier of the solution with the addition of too high a concentration of additives.²⁶

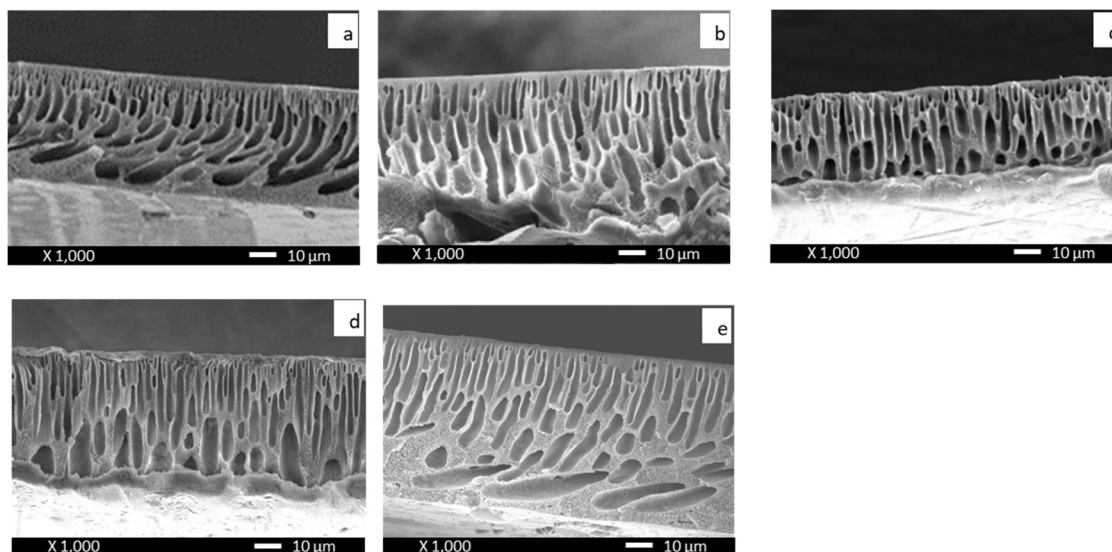


Fig.-2: Cross-sectional Morphology of (a) MC-0, (b) MC-1, (c) MC-3, (d) MC-5 dan (e) MC-7 Membranes (Mag. 1000x)

To further study the concentration effect of MC additive on the pore characteristics of the membrane, an evaluation of membrane porosity was also conducted. The obtained results are presented in Fig.-3. It can

be observed from Fig.-3 that in general the porosity of the PES membrane improves after being modified with MC at a concentration of 1-5%, but the porosity decreases when the additive is exceeding 5%. This result is in agreement with the results of SEM analysis (Fig.-1) and the discussion in the previous subsection.

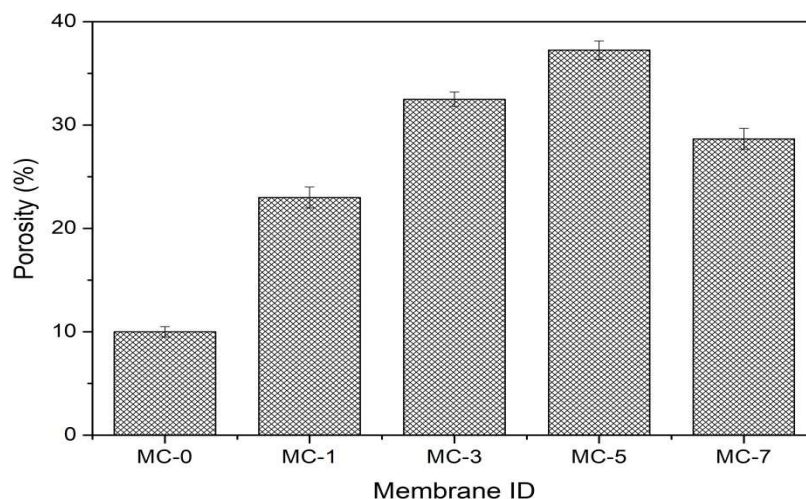


Fig.-3: Porosity of PES Membranes before and after modification with a hybrid of $\text{Mg}(\text{OH})_2$ /Chitosan at Various Concentration

Membrane Hydrophilicity

As given in Fig.-4, the original PES membrane has a contact angle of the water (WCA) reaching up to 84.2° . The addition of 1% hybrid MC decreased the membrane contact angle to 68° . When the hybrid additive concentration increased to 5%, the membrane contact angle decreased to 60.2° . This phenomenon explains that the presence of the MC additive increases the hydrophilicity of the membrane thanks to OH and NH groups which are known as hydrophilic groups which can increase the membrane interaction with water.^{27,28} It was reported that hydrophilic additives tended to migrate to the water-polymer interface during the membrane formation process, increasing the amount of hydroxyl groups on the surface causing an increase in hydrophilicity.²⁵

Moreover, the hydrophilicity degree of a membrane is complementary to the morphology and pore characteristics of the membrane. A membrane with a higher number or a larger size of pores will cause water to be adsorbed or scattered more easily thus the contact angle formed when measuring the contact angle will be smaller.^{3,29}

However, increasing concentration actually increases the WCA to 63.8° on the MC-7 membrane. The WCA of MC-7 is higher than that of MC-5, which is 60.2° . The MC-7 membrane has a pore structure with a smaller number of finger-like pores than those on the MC-5 membrane, as shown in the SEM analysis results (Fig.-2). This is the reason for the increase in the degree of MC-7 contact angle.

Overall, the modified membranes found have a lower WCA than the pure PES membrane. A conclusion can be taken that hybrid MC brought about the improvement in the hydrophilic nature of the membrane which is expected to benefit permeation performance and the declining propensity of fouling during the filtration process.

Filtration Performance

The effect of MC additive concentration on membrane performance for filtration applications was assessed in regards to water permeability and rejection of common water contaminants, namely HA. The resulting pure water permeability (PWP) and HA rejection from filtration experiments using PES membranes modified at various concentrations of $\text{Mg}(\text{OH})_2$ /chitosan hybrid additives are shown in Fig.-5 and Fig.-6, respectively.

In Fig.-5, it is displayed that increasing the concentration of MC can significantly increase the permeability of pure water. This finding is greatly related to the morphological and surface properties of the membranes.

Since the pure PES membrane has a nonporous morphology and hydrophobic surface, the generated PWP is only 4.1 L/m².h.bar.

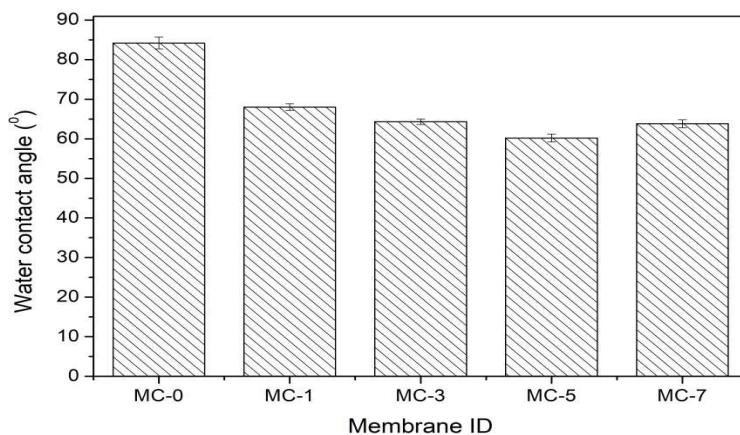


Fig.-4: Water Contact Angle of PES Membranes before and after modification with hybrid Mg(OH)₂/Chitosan at Various Concentration

The addition of 1% MC brought about 10 times increment of PWP, namely to 40 L/m².h.bar. Furthermore, the permeability of water increased significantly to 59.2 L/m².h.bar with the addition of 5% additive (MC-5). However, the PWP decreased when the additive concentration was added to 7%, which was around 45.23 L/m².h.bar. The performance of water permeability flux is greatly affected by the morphology and characteristics of the membrane, particularly those related to pore properties.²⁸ The MC-5 membrane produced the highest PWP because the number of pores is higher than the MC-1, MC-3 and MC-7 membranes as evidenced by the SEM analysis results and porosity in Fig.-1 and Fig.-3.

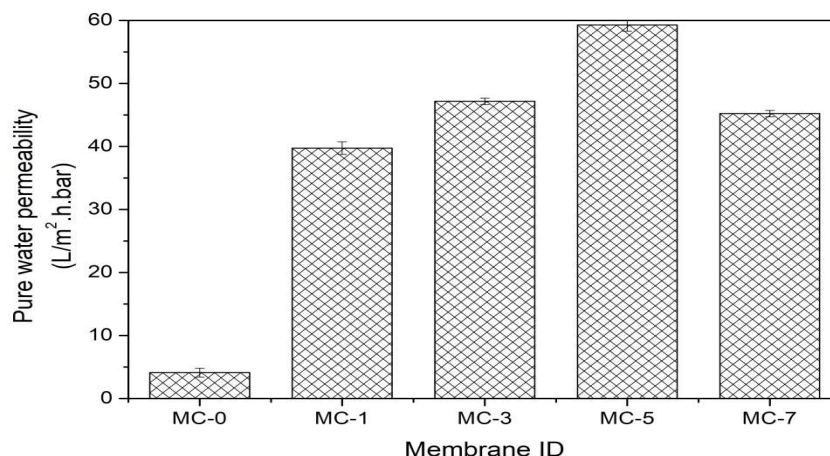


Fig.-5: Pure Water Permeability of PES Membranes before and after modification with hybrid Mg(OH)₂/Chitosan at Various Concentration

Although the increase in membrane pore properties has an advantageous role in the permeation performance, at the same time it is also negatively affecting the membrane in terms of selectivity or rejection performance. As shown in Fig.-6, all the membranes after modification produced a permeate with a slightly lower rejection percentage than the pure PES membrane. This is because the enhanced pore properties on the surface of modified membranes ease for more HA particles to pass through into the permeate.²

Fouling Resistance

In filtration processes that employ membranes, the deposition of the feed material into the membrane or widely known as fouling will affect the membrane performance by increasing the resistance to permeation

and decrease in flux. Several factors such as the formation of cake, flow properties of suspension, and the properties of the fouled layer have been extensively studied and it is found that the performance of membrane will decline except if the occurrence of fouling is controlled properly.³⁰

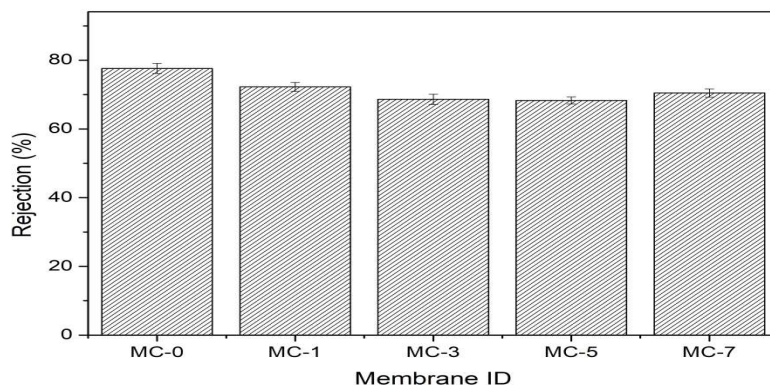


Fig.-6: HA Rejection of PES membranes before and after modification with hybrid MC at Various Concentration

Handling of fouling is generally carried out by cleaning the membrane, and the effectivity of this procedure indicates whether or not the membrane has better antifouling properties. The effectivity is evaluated by the amount of water flux that can be returned from the initial flux after the cleaning process which is known as the water flux recovery ratio (FRw). The high percentage value of FRw means that the after-cleaning water flux (J_{w2}) is not far different from the original (before-cleaning) water flux (J_{w1}) which indicates that not much water flux is lost due to fouling because the fouling formed can be cleaned effectively through cleaning (the membrane has good antifouling).¹¹ The fouling-induced flux losses during the filtration process consist of recoverable (Rr) and non-recoverable (Rir) types. The results of the antifouling evaluation of the modified membranes in this study are shown in Fig.-7 and Fig.-8.

In Fig.-7, the total flux loss due to fouling on MC-0 membrane appeared to be the highest compared to other membranes. Pure PES membrane (MC-0) has a hydrophobic surface characteristic thus the hydrophobic HA particles tend to bond easily to the surface of the membrane. Since the solid interaction between the surface of the PES film and HA particles which are hydrophobic, the fouling that is formed cannot be removed easily even after backwash,³¹ therefore from nearly 87% of the flux lost, only about 1% can be recovered or expressed with a water flux recovery ratio (FRw) of 14% (Fig.-7).

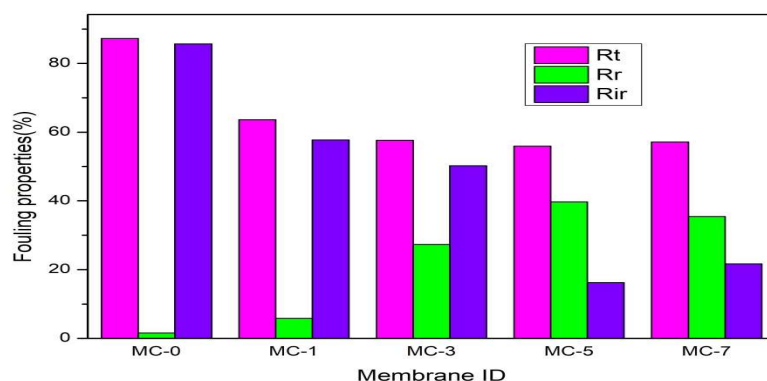


Fig.-7: Fouling-induced Flux Loss

For MC-1, MC-3, MC-5 and MC-7 membranes, the percentage of fouling that occurs are lower due to the enhanced degree of hydrophilicity of membrane surface caused by the presence of hybrid MC additive which the effect is proportional to the increase in concentration. The backwash cleaning of the membrane was successful as shown by the Rr and Rir data in Fig.-7. The MC-5 membrane lost as much as 55% from the initial flux due to the formed fouling. After cleaning, about 39% of the lost flux was successfully returned with a FRw value of 82%.

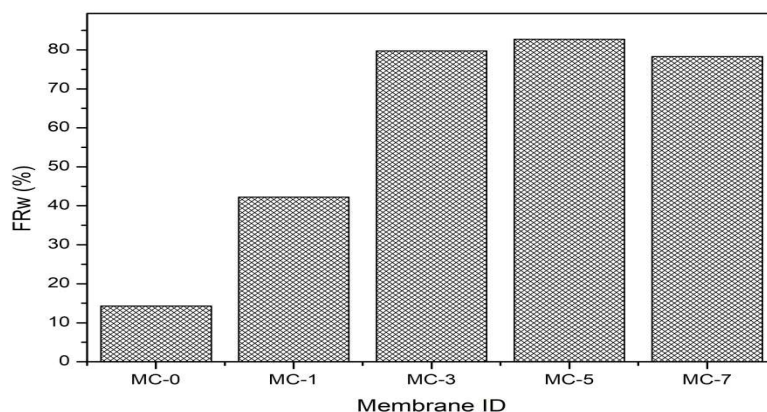
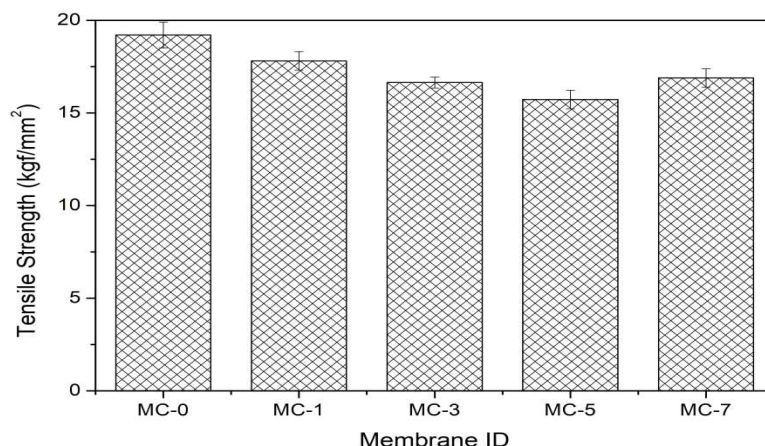


Fig.-8: Water Flux Recovery Ratio (FRw) for all Membranes

Mechanical Strength

Mechanical properties indicate the ability of a membrane to withstand the pressure applied during the filtration process. It is necessary to characterize mechanical traits to determine the strength of the membrane against the forces exerted from outside which can damage the membrane. It is generally reported that the denser the membrane structure, the tighter the distance between the molecules in the membrane matrix hence the better the mechanical characteristics of the membrane.³²

Fig.-9: Effect of Mg(OH)₂/Chitosan on the Tensile Strength of PES Membrane

Based on the results of the membrane tensile test presented in Fig.-9, it is observed that after the addition of MC additive, the PES membrane experienced a decrease in mechanical strength compared to that of the original membrane. This is due to the matrice morphology of MC-1, MC-3, MC-5, and MC-7 membranes which have larger sizes of finger-like and macrovoid pores. The existence of empty spaces from macrovoid and finger-like pores weakens the bonds between the internal structures of the membrane hence it causes the membrane to be broken during tensile measurements.⁹

CONCLUSION

Overall, it can be concluded that the modification of the membrane with an increasing concentration of hybrid MC additive has a positive effect on PES membranes, especially in terms of hydrophilicity, water permeability and antifouling propensity. However, the excessive concentration of hybrid MC (i.e. 7%) brought about a decline in hydrophilicity, permeability and the antifouling propensity of PES membrane. The modified PES membranes made with a casting solution with a composition of 18% PES, 5% hybrid Mg(OH)₂/chitosan and 77% NMP, showed the best performance with a PWP of 59.3 L/m².hr.bar, humic acid rejection of 68.2% and Water Flux Recovery Ratio (FRw) of 82.7% as well as decent tensile strength value.

REFERENCES

1. N. Arahman, A. Fahrina, M.Y. Wahab, and U. Fathanah, *F1000Research*, **7**, 726(2018), <https://doi.org/10.12688/f1000research.15077.2>
2. S. Mulyati, S. Muchtar, M. Yusuf, N. Arahman, S. Sofyana, C.M. Rosnelly, U. Fathanah, R. Takagi, H. Matsuyama, N. Shamsuddin, and M.R. Bilad, *Polymers*, **10**, 17(2020), <https://doi.org/10.3390/membranes10010017>
3. S. Muchtar, M.Y. Wahab, S. Mulyati, N. Arahman, and M. Riza, *Journal of Water Process Engineering*, **28**, 293(2019), <https://doi.org/10.1016/j.jwpe.2019.02.012>
4. N. Arahman, S. Mulyati, M.R. Lubis, R. Takagi, and H. Matsuyama, *Journal of Water Process Engineering*, **20**, 173(2017), <https://doi.org/10.1016/j.jwpe.2017.10.007>
5. N. Nasrollahi, V. Vatanpour, and S. Aber, *Journal of Water Process Engineering*, **31**, 100891(2019), <https://doi.org/10.1016/j.jwpe.2019.100891>
6. L. Shen, X. Bian, X. Lu, L. Shi, Z. Liu, L. Chen, Z. Hou, and K. Fan, *Desalination*, **293**, 21(2012), <https://doi.org/10.1016/j.desal.2012.02.019>
7. Z. Wang, and C.-y. Wang, *American Society of Mechanical Engineers, Heat Transfer Division, HTD*, **366**, 27(2000), [https://doi.org/10.1016/S0378-7753\(00\)00662-5](https://doi.org/10.1016/S0378-7753(00)00662-5)
8. H.W. Han, W. Liu, J. Zhang, and X.-Z. Zhao, *Advanced Functional Materials*, **15**, 1940(2005), <https://doi.org/10.1002/adfm.200500159>
9. M.Y. Wahab, S. Muchtar, S. Jeon, L.-F. Fang, S. Rajabzadeh, R. Takagi, N. Arahman, S. Mulyati, M. Riza, and H. Matsuyama, *Journal of Applied Polymer Science*, **136**, 47737(2019), <https://doi.org/10.1002/app.47737>
10. S. Liang, K. Xiao, Y. Mo, and X. Huang, *Journal of Membrane Science*, **394-395**, 184(2012), <https://doi.org/10.1016/j.memsci.2011.12.040>
11. X. Zhao, R. Zhang, Y. Liu, M. He, Y. Su, C. Gao, and Z. Jiang, *Journal of Membrane Science*, **551**, 145(2018), <https://doi.org/10.1016/j.memsci.2018.01.039>
12. F. Al-Sagheer, and S. Muslim, *Journal of Nanomaterials*, **2010**, 490679(2010), <https://doi.org/10.1155/2010/490679>
13. Y.-C. Huang, and T.-J. Liu, *Acta Biomaterialia*, **8**, 1048(2012), <https://doi.org/10.1016/j.actbio.2011.12.009>
14. C.K.S. Pillai, W. Paul, C.P. Sharma, *Progress in Polymer Science*, **34**, 641(2009), <https://doi.org/10.1016/j.progpolymsci.2009.04.001>
15. P.T. Jochym, A.M. Oleś, K. Parlinski, J. Łażewski, P. Piekarczyk, and M. Sternik, *Journal of Physics: Condensed Matter*, **22**, 445403(2010), <https://doi.org/10.1088/0953-8984/22/44/445403>
16. C. Dong, Y. Dai, S. Jiang, and G. He, *Journal of Environmental Chemical Engineering*, **5**, 877(2017), <https://doi.org/10.1016/j.jece.2017.01.006>
17. C.M. Jiao, Z.Z. Wang, Z. Ye, Y. Hu, and W.C. Fan, *Journal of Fire Sciences*, **24**, 47(2006), <https://doi.org/10.1177/0734904106053160>
18. U. Fathanah, I. Machdar, M. Riza, N. Arahman, M. Yusuf, S. Muchtar, M.R. Bilad, and N.A.H. Md Nordin, *Journal of Membrane Science and Research*, **6**, 375(2020), <https://doi.org/10.22079/JMSR.2020.124107.1365>
19. I. Munnawar, S.S. Iqbal, M.N. Anwar, M. Batoool, S. Tariq, N. Faitma, A.L. Khan, A.U. Khan, U. Nazar, T. Jamil, and N.M. Ahmad, *Carbohydrate Polymers*, **175**, 661(2017), <https://doi.org/10.1016/j.carbpol.2017.08.036>
20. J.-H. Jiang, L.-P. Zhu, H.-T. Zhang, B.-K. Zhu, and Y.-Y. Xu, *Journal of Membrane Science*, **457**, 73(2014), <https://doi.org/10.1016/j.memsci.2014.01.043>
21. S. Yasmeen, M. Kabiraz, B. Saha, M. Qadir, M. Gafur, and S. Masum, *International Research Journal of Pure and Applied Chemistry*, **10**, 1(2016), <https://doi.org/10.9734/IRJPAC/2016/23315>
22. C. Lustriane, F.M. Dwivany, V. Suendo, and M. Reza, *Journal of Plant Biotechnology*, **45**, 36(2018), <https://doi.org/10.5010/JPB.2018.45.1.036>
23. A. Ansari, A. Ali, M. Asif, and Shamsuzzaman, *New Journal of Chemistry*, **42**, 184(2018), <https://doi.org/10.1039/C7NJ03742B>

- 24.Z. Liu, S. Ma, X. Li, H. Yang, and Z. Xu, *Journal of Solid State Chemistry*, **277**, 493(2019), <https://doi.org/10.1016/j.jssc.2019.07.007>
- 25.M.O. Mavukkandy, M.R. Bilad, J. Kujawa, S. Al-Gharabli, and H.A. Arafat, *Separation and Purification Technology*, **187**, 365(2017), <https://doi.org/10.1016/j.seppur.2017.06.077>
- 26.M. Sadrzadeh, and S. Bhattacharjee, *Journal of Membrane Science*, **441**, 31(2013), <https://doi.org/10.1016/j.memsci.2013.04.009>
- 27.S. Mulyati, M. Armando, H. Mawardi, A. Fahrina, N. Malahayati, and S. Muchtar, *Rasayan Journal of Chemistry*, **11**, 1609(2018), <http://dx.doi.org/10.31788/RJC.2018.1144047>
- 28.S. Mulyati, S. Muchtar, N. Arahman, F. Meirisa, Y. Syamsuddin, Z. Zuhra, C.M. Rosnelly, N. Shamsuddin, N.I. Mat Nawati, M.D. Wirzal, M.R. Bilad, R. Takagi, and H. Matsuyama, *Polymers*, **12**, 1807(2020), <https://doi.org/10.3390/polym12081807>
- 29.S. Mulyati, S. Muchtar, N. Arahman, Y. Syamsuddin, N.I. Mat Nawati, N. Yub Harun, M.R. Bilad, Y. Firdaus, R. Takagi, and H. Matsuyama, *Polymers*, **12**, 2051(2020), <https://doi.org/10.3390/polym12092051>
- 30.Y.Z. Tan, Z. Mao, Y. Zhang, W.S. Tan, T.H. Chong, B. Wu, and J.W. Chew, *Separation and Purification Technology*, **215**, 70(2019), <https://doi.org/10.1016/j.seppur.2018.12.085>
- 31.W. Sun, J. Liu, H. Chu, and B. Dong, *Membranes*, **3**, 226(2013), <https://doi.org/10.3390/membranes3030226>
- 32.A. Nasir, F. Masood, T. Yasin, and A. Hameed, *Journal of Industrial and Engineering Chemistry*, **79**, 29(2019), <https://doi.org/10.1016/j.jiec.2019.06.052>

[RJC-6582/2021]