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Morphological and Permeability Fraction of Mixed Matrix Polysulfone Membrane on Batik Palembang Wastewater Treatment

Morphological and Permeability Fraction of Mixed Matrix Polysulfone Membrane on Batik Palembang Wastewater Treatment

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Abstract. High performance polysulfone (PSf) membrane has been prepared for treating of batik palembang wastewater. Many investigations attempted the use of polysulfone (PSf) membranes to filtrate dilute contaminant in textile wastewater. The matrix membrane is a mixture of PSf which is produced by adding inorganic materials to obtain a polymer that functions to manufacture the membrane by increasing hydrophilicity and impurity resistance. MMM were fabricated from a dope solution containing PSf/LiCl/DMF at different additives composition. Antifouling properties of membranes could be enhance with 0.0 - 10 wt.% LiCl that has been added into dope solution. Pure water permeation flux analysis (PWP), AFM and FESEM. From the results of research on FESEM, the PSf / LiCL membrane showed a fairly symmetrical cross-sectional structure containing two layers. The layers of the tiny finger-like structure on the edge of the perforated fibers and the holes like the larger fingers are mixed with the macrovoid layer in the middle section. PWP flux is not proportional to mean rough outer surface but higher water permeation. Therefore, PSf/LiCl exhibited a better performance that resulted a much lower fouling tendency than the naked membrane in batik Palembang wastewater treatment.

Keywords: mixed matrix membrane, textile wastewater, polysulfone, lithium chloride.

1. Introduction

The treatment of water organic wastes is becoming ultrafiltration has been important in finding applications in this area [1]. Tools used in water and waste treatment use membranes. the reason for using the membrane is the density in the strong packaging, as well as providing convenience in the manufacture and operation of the membrane module. The hollow fibers in the membrane are implanted directly in the feed reservoir by drawing seepage through the fibers by applying vacuum to the lumen fiber outlet [2]. Textile wastewater treatment according to previous research, uses particulate, inorganic and organic materials (for example sulfides, phenols) because these materials are a potential contributor to membrane fouling which can be useful in textile wastewater treatment [3][4]. The structure of the morphological membrane which has the properties of a fouling layer is useful for controlling the structure. whereas for the performance of the membrane using a modified PSf membrane, as shown in Figure 1. Preventing the formation of fouling layers using cross-sectional structure analysis or create this layer more reversible, this method can improve the performance of the membrane process.

Hydrodynamic conditions and physicochemical properties are important tools for membranes in the impurity layer and membrane morphology [5]. Membrane properties and flux are influenced by a wide variety of drugs.

Some authors have suggested that to improve the morphology and properties of membranes, it is necessary to add inorganic organic additives and different high molecular weight to the anesthetic solution. The mechanism of the anesthetic solution mixing process used in the formation of membranes is based on changes in the properties of the kinetic and thermodynamic phases during the phase inversion process and when additives occur. The categories of additives used are classified as follows:

a. Polymer additives (PVP.PEG).
b. Chemicals that have a low molecular weight include satl (LiCl), inorganic acids (acetic acid and phosphoric acid), organic acids (propionic acid).
c. Weak common solvents (ethanol, propanol and acetone).
d. Non-strong solvents such as water

Fontananova et al stated that the function of aacetamide dopene additive is as a pore formation, increasing viscosity of the solution, accelerating phase inversion process. The addition of LiCl in dope PVDF / dimethylacetamide is useful in increasing the flux of the casted membrane at low LiCl concentrations (2.5 wt%) but the formation of high macrovoid concentrations (7.5 wt% LiCl) can result in decreasing membrane permeation.

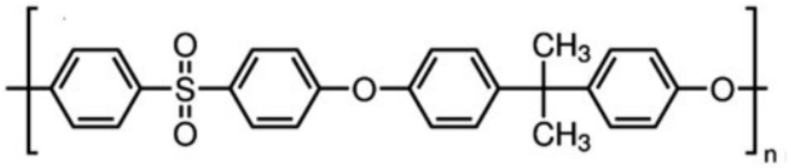

Fig. 1. Chemical structure of polysulfone.

Research by Zodrow et al. carried out mixing of the nanosilver particles onto the PDf membrane matrix with the aim of increasing the hydrophilicity of the membrane and reducing biofouling on virus penetration. If according to Kang et al. [6] using the phase inversion method, aims to prepare more hydrophilic PSf / SGO (sulfonated graphene oxide) membrane. After testing, the researcher was able to explain that mixing the 1.5 w / w.% SGO into the created PSf matrix the membrane achieves a higher flux in comparison to using neat PSf..

Lithium chloride (LiCl) states that recently hydrophilic inorganic particles have become one of the most commonly used for polishing mixed membranes [7][8]. This is evidenced by the emergence of the oxygen-containing group including hydroxyl, epoxyl, and carboxyl moment making the graphene oxide mixed membrane. The results show that membrane has a higher flux and better antifouling capacity [9]. The water molecule which consists of oxygen functions into a strong hydrogen network, so as to create a strong hydrophilicity [10][11]. To increase the permeability [12] and the antifouling [13] and anti-bacterial properties [14] for membrane applications it is necessary to combine LiCI into the membrane.

This research makes an efficient way of membrane filtration process, which is generally influenced by several factors during the process, these factors include aeration flow velocity, concentration to mixed fluid suspended density (MLSS), pH, and time of hydraulic retention (HRT). Apart from these factors, in this study chemical oxygen demand, suspended solids, and ammonia nitrogen removal efficiency were considered to be maximized due to environmental regulation. The experimental research was designed to be adapted to synthetic wastewater and a pilot scale membrane system carried out in batches.

Table 1. Composition of standard deviation (S.D) synthetic wastewater and environmental regulatory standards

Constituent, unit	Concentration (S.D)	Standard
pH	6.7 (0.02)	5.5 – 9.0
COD, mg/L	555.0 (0.25)	200
NH3-N, mg/L	29.1 (1.02)	20.0
Suspended Solid, mg/L	213.0 (0.07)	100.0

2. Experimental Methods

2.1. Materials

The PSf membrane (molecular weight 22,000 g, mol-1) was used as the original polymer produced by Sigma-Aldrich in a casting solution. N, N-Dimethylformamide (DMF) purchased from China Science. Co., Ltd is used as a solvent. Lithium Chloride monohydrate purchased by Sigma-Aldrich was used as additive and glycerol purchased by MERCK (Germany) was used as a non-solvent for post membrane treatment. In all cases, tap water is used as an external coagulation bath medium in the spinning process.

2.2. Membrane of Preparations

He PSf membrane was adjusted to room temperature equal to by wet jet dry spinning method. The regulatory the solution is divided into two groups namely membrane prepared from 17 wt.% PSf in DMF at different concentrations of Lithium choloride (0-10 wt.%) than the setting PSf weights as shown in Table 2.

Table 2. Composition of prepared membranes

Sample	PSf wt.%	LiCl wt.%
PSf-0	17	0
PSf-5	17	5
PSf-10	17	10

The general pressure application of the polymer solution is by means of a spinning wheel using speed controlled extrusion, whereas internal coagulant setting is 1.3 mL / minute. The perforated fibers at the end of the spinnerets are arranged through two water baths with a withdrawal speed of 13.5 cm / s, be careful to match desired speed before the waste reaches the final reservoir to complete the compaction process. Then spun hollow fibers immersed in a water bath for 3 days, by doing water changes every day, this was done with the aim of removing the remaining DMF and additives. After the immersion process, the perforated fiber is then treated using a 10% solution by weight of glycerol as a non-solvent exchange for 1 day, this is done in order to minimize fiber shrinkage and pore collapse. After that, it is dried for 3 days, if the fiber is dry then the fiber is ready to be made into a hollow fiber test module.

2.3. Ultafiltration Process

Table 1 explains that the liquid waste from the Palembang batik factory consists of permeation flux and PSf membrane rejection, which are then measured using the ultrafiltration system. The production of fiber modules is carried out at home, using a filtration area of 14.20 dm2, then immersion in a suspension that has been prepared and determined in the membrane reservoir by volume 15 L. Mechanical cleaning of membrane modules is carried out by immersing the membrane using cross flow, namely placing diffuser under the membrane module to produce air bubbles. Air bubble flow circulation per unit in the projected membrane area is regulated constantly at 2.5 mL / minute, it aims to maintain proper turbulence. Filtration pressure is generated from the vacuum pump and controlled by a needle valve. Continuously recorded using a permeate flow meter that occurs. The The lab-scale experimental setup is shown in Figure 1.

Fig. 2. Ultrafiltration Membrane System.

Measurement of the membrane performance of TSS and NH3-N concentrations using a spectrophotometer (DR 5000, HACH) which according to standard procedures, method 8006 (photometric method) and HR TNT 10031 method (Salicylate method). The evaluation of the concentrations was carried out every day as long as the operation was at a high organic load level, and for sampling it was carried out three times a week. Flux, total suspended solids (TSS) and NH3-N removal efficiency are calculated by Eq. (1,2 and 3).

$$J = \frac{1}{A} \cdot \frac{dV}{dt} \quad (1)$$

where V is the permeate volume (L), A is the membrane surface area (m2), and t is time (h).

$$\text{TSS removal (\%)} = \frac{TSS_0 - TSS}{TSS_0} \times 100 \quad (2)$$

where TSS and TDS are initial TSS concentrations from synthetic textile feed wastewater and TSS concentrations from the resulting permeate.

$$NH_3\text{-N removal (\%)} = \frac{NH3\text{-}N_0 - NH3\text{-}N}{NH3\text{-}N_0} \times 100 \quad (3)$$

where NH3-No and NH3-N are the initial NH3-N synthetic concentration textile waste water and the resulting permeate concentration of NH3-N

Table 3. Effluent standard for textile industry [14]

Parameter	Content maximum (mg/L)	Maximum pollution load (kg/ton)	
		Source	Near by river
COD	60	97	67
BOD	150	105	80
TSS	50	0.9	1.0
Phenol	0.5	0.083	0.055
Ammonium Nitrogen	8.0	6.3	0.32
Turbidity	25	32.1	294
Color	50	339.0	120.0
pH		6.0 – 9.0	
Maximum waste discharge in m^3/ton		18	20

This filtration experiment was carried out under vacuum by means of the open end the fibers in the permeate were pulled using a peristaltic pump (Masterflex model 7553-79, Cole Palmer).The constant at the liquid level in the feed tank during the experiment should be maintained. This is done so that the resulting air scoured bubbles are stable because the air scoured bubbles are useful for providing shear stress which serves to minimize particles settling on the membrane surface during the filtration process. The measuring cylinder is used to determine the water permeation volume. After the filtration process is complete, the surface of the membrane is cleaned use a soft sponge to remove the entire layer of particles that may occur during filtration.

2.4. Membrane Characterizations

The function of the electron microscope performs a field emission scan (JEOL JSM-6700F) is to serves to minimize particles settling on the membrane surface during the filtration processfibers. The first thing to do is to immerse the membrane sample in liquid nitrogen and break it down carefully [16]. Before testing the sample was coated first using platinum sputtering. FESEM micrograph is a cross-section and outer surface of the hollow fiber membrane by taking it using a magnifying glass. This is done to observe effect of variations in the concentration of suspended solids from the liquor mixture and the flow rate of air bubbles in the Palembang batik liquid waste process.

3. Results and Discussion

3.1. Membrane Characteristics

Figure 3 showed FESEM image of LiCl nanoparticles, which illustrated that LiCl has a winkled surface with some small pieces.

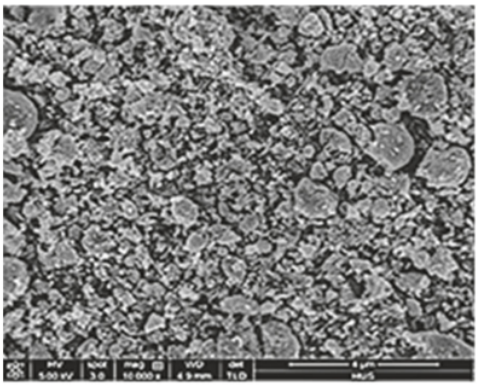

Fig. 3. FESEM image of Lithium Chloride.

Figure 4 provides an explanation that there is an illustrative drawing of a hollow fiber membrane prepared crosswise from different additive compositions. Figure 4 is a representation of the membrane, namely the inner edge layer, inner middle layer, outer middle layer, and outer edge layer of the membrane. The results of the image show that PSf-0 consists of two layers, namely the inner and the edge of the oiter with a small finger-like structure (layers 1 and 4) and two other layers in the middle with a cavity like a large finger mixed with the macrovoid layer (layer 2 and 3). In previous studies the membrane contained four layer structures [15] [16]. This shows a difference with previous studies due to research conducted by observing 2 layers where this layer is shifted to the outermost ego when the composition of lithium chloride is increased. As a result, in Fig. 3 (a-c), the coating thickness is greatly reduced at a lithium chloride composition of 1.0%.

Transitions occur from finger-like structures to sponge-like structures in the cross-section of the membrane. Along the membrane wall consists of a sponge-like substructure that lies on the thick top layer. This occurs because the higher viscosity of the anesthetic can reduce the solvent and non-solvent exchange rates (water from the coagulation medium), resulting in a higher resistance to non-solvent

diffusion required during polymer aggregation. From the shape of the FESEM section is not lined with dense skin layer pores with a magnification of 40,000. The results showed that the size of the pores on the membrane surface was in the nanometer range. Previous studies have shown that the pore size decreases with increasing concentrations of the polymer. This can occur due to an increase in the viscosity of the solution, which leads to a stronger molecule and a tighter structure [17][18]. Therefore, it is hoped that there will be a shrinkage of the membrane pores along with the polymer concentration. To verify the results of the FESEM form, membrane characterization is required by measuring pure water using the solute transport method carried out in the following section.

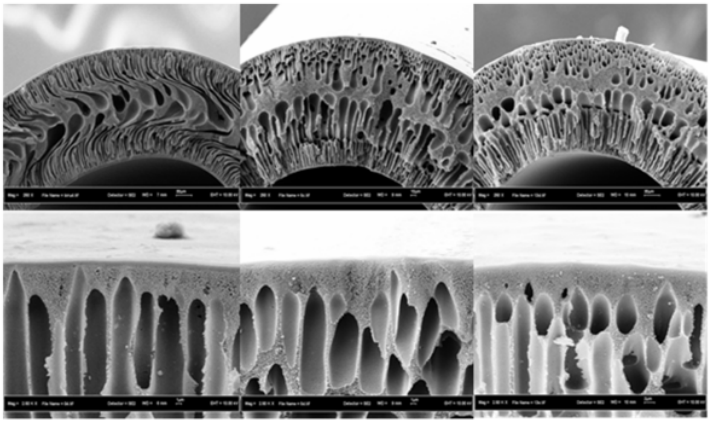

Fig. 4. Cross section and outer surface FESEM images of the modified

3.2. Permeability of Membrane

The relative permeability of the PSf membrane was found to vary the composition of the LiCl portion.

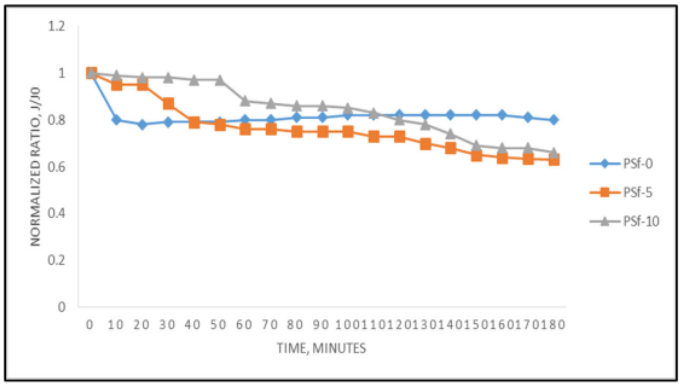

Fig. 5. Relative permeability of PSf membrane with the different LiCl composition.

In Figure 5 above is evidence of a decrease in the relative permeability of the PSf-T membrane with different LiCl concentrations. The occurrence of a rapid reduction in the relative permeability by the investigator was closely observed. This is done in order to determine the consequences of membrane fouling by synthetic wastewater. The performance of the three membranes is shown in Figure 5, PSf-0 membrane had the best performance with removing 91.1%. From this figure, it can be seen that there is a drastic decrease in membrane permeability within 30 minutes of filtration when the initial rejection was high from initial filtration stage as shown in Figure 4(b). This suggests that there is initial resistance

by the membrane to humic acid which is a key factor for the decline at higher fluxes. Thus there is a higher rejection by the membrane, this is because the pore size becomes smaller [17]. The initial rejection of PSf-5 and PSf-10 membranes was 83.6% and 76.1%, this indicates that the pore sizes of both membranes are much larger than humic acid. In fact, larger pores of membranes will cause humic acid much easier to go through the pores and resulting lower solute separations. Figure 4 (b) explains that, there was a decrease in humic acid rejection, this happened due to an increase in the pore size of the outer surface, or by an increase in the air gap distance. In other words, it shows that solute transport can be regulated by the pore size and pore size distribution of the outer surface of the PSf-5 and PSf-10 membranes. In a previous study [14], it was found that the solute separation value was lower for hollow fiber membranes which had a larger pore size. In the case of PSf-5 and PSf-10 membranes, the permeability of the degradation process is much slower than that of PSf-0. However, the PSf-0 membranes keep high permeability compared of PSf-5 and PSf-10 membranes.

3.3. Effect of Additives Concentration on The PSf Membrane Performance

The effect of additive concentration in this study on permeability and rejection was observed by submerged membrane ultrafiltration. Table 3 shows the results of Palembang batik wastewater treatment using the PSf membrane. Firstly, there is an increase and decrease in the flux by increasing the concentration of LiCl. The highest value achieved were 93.82 L/m^{2}h for batik palembang wastewater and 99.67 L/m^{2}h for pure water at 1.0 wt.% LiCl concentration.

Table 4. Properties of PSf membranes

Membrane	Pore size, nm	Porosity (%)	Hydrophilicity.°	Flux, L/m2h	Permeability,%
PSf-0	34.6	65.68	83.5	93.82	80
PSf-5	30.23	72.90	73.2	96.21	84
PSf-10	28.67	75.82	56.0	102.20	91.1

The pore size and porosity that have the membrane will provide performance information of the porosity of the membrane in the range of 65.68 to 75.82% and the pore size of the membrane in the range of 34.6 to 28.67 nm. This membrane has good properties due to its low polymer concentration levels in the spinning anesthetics and additives used. Because the additive is soluble in the solvent, it can escape from the spinning hubs during exchange with the coagulation bath. Therefore, the high porosity membrane structure is given by PSf-10. However, the porosity of the PSf membrane will decrease by increasing the value of the LiCl concentration. This phenomenon can explain the extrusion of LiCl nanoparticles in the porous structure and the aggregation of LiCl nanoparticles in the pores. Furthermore, the interaction between PSf / LiCl described by Chiang et al. has an influence on the porosity of the polymer membrane [18]. The pore size will be reduced by increasing the concentration of LiCl in spinning anesthetics due to the interaction between LiCl and PSf. The small proportion of LiCl in the dope shows that there is an interface tension between the polymer and the LiCl nanoparticles, which forms the interface pores from shrinking the organic phase during the demixing process. A higher concentration of LiCl will result in the formation of a denser substructure, which reduces pore size.

Hydrophilicity measurement resulted the changes of value with adding the LiCl. The results indicated that a significant decrease of contact angle in Table 3. Reduce values from 83.5° to 56.0° was caused by formation of hydroxyl on membrane. The increase in hydrophilic presence in the membrane will result in the value of the contact angle decreasing, but in LiCl there is an increase in the hydrophilicity of the membrane.

The test results show that the LiCl on the membrane surface can reduce the communication between contaminants and the membrane surface. The hydrophilicity of the membrane and the pore size of the membrane present at lower concentrations of LiCl can attract water molecules into the membrane pores, thereby facilitating penetration through the membrane. This phenomenon can increase the flux and decrease the flux reduction coefficient. However, the higher the LiCl concentration (> 1.0 wt.%) Will produce a very thick dope, which will slow down the PSf membrane process and produce a denser

sponge-like substructure. This results in a decrease in the hydrophilicity and diameter of the membrane pores. This shows that the PSf membrane with the addition of LiCl shows a promising flux as needed.

The function of permeability is to characterize the occurrence of changes in the membrane surface for hydrophilic properties. The increase in membrane permeability will increase the value of the membrane to be better than hydrophilic, this can be done by adding LiCl to the dope solution. This process will result in an increase in membrane permeability with LiCl compared to neat PSf. Thus, the presence of the LiCl functional group containing hydroxyl ions causes changes in water permeability. This also corresponds to the hydrophilicity value.

4. Conclusions

The wet jet dry spinning method was used to modify the PSf membrane. Various concentrations of LiCl are used as an inorganic additive in spinning dopes which aims to increase the speed of the inversion phase and provide a well-structured porous asymmetric membrane used for batik Palembang wastewater treatment. Several characterization and measurement techniques that can be used are membrane structure and permeability. It aims to evaluate the details of the fine structure of the membrane and membrane performance. The results of the FESEM analysis state that the addition of 10 wt.% LiCl nanoparticles can produce nanoparticles with a smaller size, so that they can achieve the performance of a higher hydrophilicity, with a small pore size and high porosity. The results of the tests carried out on permeability explained that the LiCl nanoparticles had a high performance effect on the modified PSf membrane, and produced a higher flux of 102.20 L / m2h so that they could treat Palembang batik wastewater.

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