



## Research Paper

## Effect of Ethylene Glycol as Pore Former on Polyphenylsulfone Hollow Fiber Membrane for Crude Palm Oil Deacidification through Membrane Contactor

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## Article info

Received 2016-06-28

Revised 2016-08-29

Accepted 2016-09-02

Available online 2016-09-02

## Keywords

Ethylene glycol  
Polyphenylsulfone  
Membrane contactor  
Sodium hydroxide  
Free fatty acid  
Crude palm oil

## Highlights

- EG assisted in the formation of more uniform interconnected finger-like membrane structure
- Increasing EG concentration had reduced the hydrophobicity of 14PPSU membrane
- 14PPSU membrane contactor allow the oil to wet the membrane layer and prevent the NaOH (liquid extractant) from penetrate into CPO stream for FFA removal
- No soap formation detected in the permeate sample collection

## Abstract

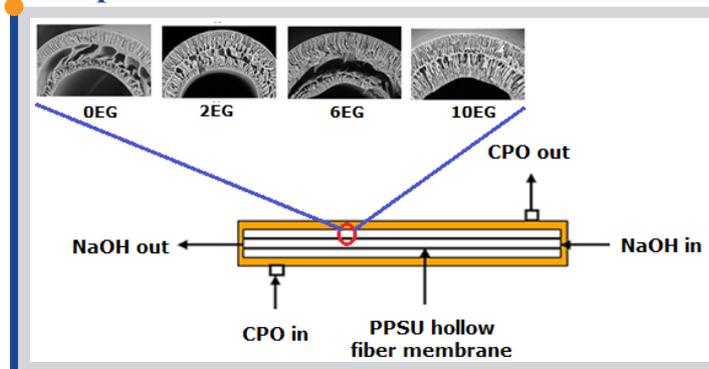
In this study, polyphenylsulfone (PPSU) hollow fiber membrane was fabricated via a wet spinning method with the addition of ethylene glycol (EG) in the range of 0-10 wt% as the additive. The EG was added to improve the membrane pore formation and interconnectivity for better membrane contactor performance in extracting and removing the free fatty acid (FFA) from crude palm oil (CPO). Four different hollow fiber membranes consisted of 14PPSU-0EG, 14PPSU-2EG, 14PPSU-6EG and 14PPSU-10EG were characterized for their morphologies, wettability and pore sizes. From the characterization, it was proven that the presence of EG has promoted the formation of more uniform interconnected finger-like structure with double-layer membrane morphology. In membrane contactor performance testing, sodium hydroxide (NaOH) was used as liquid extractant due to the high affinity of free fatty acid (FFA) towards NaOH. Unfortunately, the new developed PPSU hollow fiber membrane with the presence of EG has significantly ( $P < 0.05$ ) reduced the membrane contactor performance from 16.54% FFA removal using 14PPSU-0EG to less than 5% FFA removal in the treated CPO permeate after 3 hours operation. It may be due to a significant ( $P < 0.05$ ) reduction in membrane hydrophobicity which reduced the oil wettability and interfacial contact between CPO and NaOH at the membrane interface. Nevertheless, no soap formation was detected in the permeate collection.

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## 1. Introduction

Membrane separation is a mature technique and has been successfully applied in various food industries for separation of undesirable fractions from valuable components. The developing membrane application in vegetable oils processing includes solvent recovery, degumming, deacidification, pigment removal, wax removal and extraction of minor components [1]. The application of membrane technology in vegetable oils refining has been receiving much more attention in the light of various inherent advantages

## Graphical abstract



associated with membrane processes such as: low energy consumption, mild operating temperature, no addition of harmful/toxic chemicals, retention of nutrients and other desirable components that contributes to cost and energy effectiveness and eco-friendliness [2-4].

Free fatty acid (FFA) removal or also known as deacidification has a big impact in vegetable oil refining process. If deacidification is not carried out properly, there will be more oil loss in the subsequent processing steps and

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DOI: 10.22079/jmsr.2016.21596

unfavourably affects the quality of the final refined product [5]. Conventionally, chemical and physical deacidification methods have been employed in palm oil processing industries. In chemical refining, deacidification is accomplished by neutralising the FFA with sodium hydroxide (NaOH) and removing in the form of soap-stock [6]. Any residual soap will be removed by the addition of hot water and subsequent centrifugation. FFA content in the crude oil has direct impact in the loss of significant amounts of triglycerides due to saponification [7]. On the other hand, in physical refining deacidification is accomplished with the used of steam distillation where the FFA and other volatile components are distilled off from the oil by effective stripping agent under suitable processing conditions [8]. Despite the advantages of reducing the loss of neutral oil and simplified process operation, physical refining consumes high energy due to the high deodorization temperature in the range of 240-260 °C [5].

FFA in principle is almost impossible to be removed by membranes itself due to the molecular size of FFA (<300 Da) which is much smaller than that of triglycerides (TG ~ 900 Da) [1]. Theoretically, the ideal process would be the use of a membrane with precise pores so that they could effectively separate the FFAs from the TG [9]. Most of the previous researchers have reported the use of oil/solvent mixture (to form micelle) in membrane deacidification of vegetable oils [3, 10-12]. Only few studies had reported on direct membrane deacidification of vegetable oils. Subramanian et al. [13] studied on the deacidification of crude soybean and rapeseed oils without addition of organic solvents in undiluted oil systems using polymeric membranes. However, the FFA permeated preferentially compared to TG resulting in negative rejection of FFA. Similar results were reported by Bhosle and Subramanian [5] using polymeric nonporous membranes in refined sunflower oil and oleic acid model system. The differences in molecular size, solubility, diffusivity and polarity between TGs and oleic acid were insufficient to achieve direct deacidification. Azmi et al. [14] reported on the modification of the outer surface of PVDF membrane via crosslinking technique with polyvinyl alcohol (PVA) in the present of glutaraldehyde as crosslinking agent. However, the highest FFA rejection recorded was only 5.93% after 3 hours operation. In order to overcome the above limitation with low FFA rejection, membrane contactor technology is one of the preferred approaches that can be further explore in order to exploit the benefit of integrated membrane separation and extraction process.

Membrane contactor is used to identify membrane systems that are employed to kept in-contact at the mouth of membrane pores between two phases without dispersion [15]. However, the chemistry of membrane contactor is relatively unimportant compared to most of other membrane operations. It is because the success of membrane contactors is greatly depends on minimizing the membrane resistance to mass transfer. As a general rule, the chosen membrane must be wetted by the fluid to which the solute has more affinity (higher partition). If the solute partition is more favourable to the organic, a hydrophobic membrane should be used. However, if it partition favourable to the aqueous phase, then a hydrophilic membrane would be the best choice [16].

In this study, polyphenylsulfone (PPSU) membrane was chosen as the membrane for the contactor system because of its hydrophobicity and high tolerance to chemical resistance compared with other polysulfone family members, such as polysulfone and polyethersulfone. PPSU that comprises sulfone moieties, ether linkages and biphenyl group in its repeating group have the highest impact strength compared to other family members [17]. It presents superior resistant to hydrolysis and plasticization of stress cracking. The robustness of the material in terms of physical durability and chemical stability points out that PPSU is a remarkable polymer candidate, which could be potentially developed for further use in CPO deacidification [18].

In general, it is very common to add additives such as polyvinylpyrrolidone (PVP), polyethylene glycols (PEG), tetra ethylene glycol (TEG), etc. to polymer solution which is used in membrane fabrication. Additives also play an important role in the formation of membrane structure by enlarging or preventing the microvoid formation, enhancing pore formation, improving pore interconnectivity, and/or introducing hydrophilicity [19-20]. Particularly, PEG has been used for increasing porosity and reducing the skin layer thickness. In membrane fabrication, PEG is used to control the thermodynamics properties and phase separation kinetics that can affect the final membrane characteristics to enhance the pore size of membrane. In fact, PEG is miscible with most of the membrane materials and is soluble in both aqueous and many organic cultures [21-22].

In the present work, PPSU membrane contactor system was developed by manipulating the amount of ethylene glycol (EG) in membrane dope solution to improve the pore formation and pore interconnectivity of PPSU membrane for membrane contactor system. The improvement of PPSU membrane formation was expected to enhance interfacial contact between CPO (feed oil) and NaOH (3N) stream at the mouth of membrane pores for better extraction

and removal of FFA. The scopes of this study include membrane fabrication, membrane characterization and membrane contactor performance for deacidification of CPO. This is the first attempt which FFA is removed without forming any soap in the permeate oil collection. This is because it is really crucial to avoid oil washing after deacidification to minimize/eliminate wastewater generation from the process.

## 2. Experimental

### 2.1. Materials

PPSU polymer (M.W.=53,000-59,000 g/mol) was purchased from Sigma-Aldrich. It was chosen as a main polymer for membrane contactor system because it's hydrophobicity and high durable resistance to chemical. EG was purchased from Merck and used as additive to improve membrane pore formation and interconnectivity. *N*-methyl-2-pyrrolidone (NMP) was purchased from Merck and used as a solvent in membrane dope solution. Distilled water was used as internal coagulant (bore fluid) and external coagulant bath for membrane fabrication. CPO was obtained from Sime Darby Jomalina Sdn. Bhd. (Telok Panglima Garang, Selangor, Malaysia). NaOH was used as liquid extractant for membrane contactor system. All chemicals used in this work were received without further treatment.

### 2.2. Preparation of dope solution

Four PPSU membrane dope solution were prepared in combination with solvent (NMP) and additive (EG) consisted of 14PPSU-0EG, 14PPSU-2EG, 14PPSU-6EG and 14PPSU-10EG as are shown in Table 1. PPSU, EG and NMP were well-mixed thoroughly under constant mechanical stirring at 60°C for 1 day in order to produce a homogenous solution. Finally, the formulated dope solution was degassed before spinning to remove micro-bubbles that might exist.

**Table 1**  
PPSU membrane dope solution.

Sample	PPSU (wt %)	NMP (wt %)	EG (wt %)
14PPSU-0EG	14	86	0
14PPSU-2EG	14	84	2
14PPSU-6EG	14	80	6
14PPSU-10EG	14	76	10

### 2.3. Fabrication of PPSU hollow fiber membrane

The hollow fiber UF membranes were fabricated according to the well-known wet spinning method. The dope was placed in a stainless steel reservoir (volume: 1 liter) that was subsequently pumped to the spinneret by a gear pump. The dope reservoir that contains polymer solution was kept under nitrogen pressure as a precaution against cavitation in the line to the pump. Then, a pulse-free bore fluid was fed into the inner tube of the spinneret by a syringe pump. Once the spinning dope and the bore fluid met at the tip of the spinneret, they went into the external coagulation bath after going zero air gap. The details spinning parameter of hollow fiber membrane are shown in Table 2. The nascent fibers were then passed through the washing/treatment bath before they were collected by a wind-up drum.

### 2.4. Membrane post-treatment

The as-spun fibers were cut in pieces of approximately 40 cm in length and immersed in water bath at room temperature for 1 day to remove residual solvent from the membrane structure. Afterward, the hollow fibers were post-treated with water/ethanol (1:1) solution for 30 minutes, followed by pure ethanol solution for another 30 minutes to prevent fiber shrinkage. Finally, the hollow fibers hang vertically and air dried at room temperature for about 1 day before membrane characterization and of membrane modulation.

### 2.5. Membrane contactor module

A total of 60 pieces of HF membrane were potted inside the PVC pipe for membrane contactor performance testing. An epoxy adhesive resin (Polyurethane E-30CL Loctite) was used as the potting resin. This resin was chosen due to its suitable cure times, low viscosity and proper adhesion with the PVC and steel tubing. Table 3 shows the detail specification of HF membrane contactor module.

**Table 2**  
Spinning parameters for fabrication of PPSU hollow fiber membrane.

Parameter	Batch 2
Spinning solution	PPSU/NMP/EG
Bore fluid composition	Distilled water
Inner diameter of spinneret (mm)	0.55
Outer diameter of spinneret (mm)	1.25
Air gap (mm)	0
Dope extrusion rate (cm <sup>3</sup> /min)	7.2
Bore fluid flow rate (ml/min)	2.2
Take-up rate (rpm/min)	24
Spinning temperature (°C)	27
External coagulant	water
Coagulation bath temperature (°C)	25

**Table 3**  
Specification of HF membrane contactor module.

Membrane specification	Value
Module i.d. (mm)	35
Module length (mm)	400
Fiber o.d. (mm)	0.55
Fiber i.d. (mm)	1.25
Effective fiber length (mm)	330
No. of fibers	60

## 2.6. Membrane characterization

PPSU membrane morphology was observed using scanning electron microscope (SEM, Model: TM 3000, Hitachi). All specimens were freeze-dried and coated with an ultra-thin layer of gold before conducting analysis. Prior to analysis, a piece of hollow fiber was immersed in liquid nitrogen and fractured carefully in order to create a perfect cross section.

The HF membrane wettability property was evaluated by sessile drop method using contact angle goniometer (Model: OCA15Plus, Dataphysics, Germany) equipped with images processing software. A total of 10 contact angle measurements were performed for each sample and the average values are reported.

## 2.7. Lab-scale hollow fiber membrane contactor system

Figure 1 shows a schematic diagram of membrane contactor system for CPO deacidification. The pressure was set at 0.1 bar for both CPO stream and NaOH stream. First, CPO was subjected to pre-treatment process to remove gums (phospholipids) before deacidification process. The treated CPO was then heated and maintained at 50 to 60 °C when passing through the membrane contactor module within the membrane housing (shell-side), whereas the NaOH was circulated inside the lumen (tube-side) of PPSU membrane (see Figure 2). Four units of membrane contactor module were tested consisted of 14PPSU-0EG, 14PPSU-2EG, 14PPSU-6EG and 14PPSU-10EG. The interfacial contact between CPO and NaOH (3N) occurred at the mouth of membrane pores for FFA extraction towards the liquid extractant. About 200 ml permeate oil was collected every 1 hr operation time interval for 3 hours to check for FFA and soap content analysis.

## 2.9. Statistical analysis

Statistical analysis software (Minitab 17) was used to perform statistical analysis for *R*-square and *P*-value. Analysis of variance (ANOVA) with Tukey pairwise comparison was performed to determine significance of difference at *P* < 0.05. Analysis was conducted in triplicates.

## 3. Results and discussion

### 3.1. Membrane morphology

EG is hydrophilic in nature. The hydrophilicity of EG in membrane dope affects the exchange rate of solvent and non-solvent during phase inversion

process and influences the precipitation kinetics and the formation of resulting membrane morphology, consequently [21]. The effect of adding EG into the 14PPSU membrane dope solution with respect to membrane morphology can be observed by SEM cross-sectional images and membrane surface images in Figure 3. Three different EG concentrations were added into the 14PPSU membrane from 2 wt% EG (14PPSU-2EG), 6wt% EG (14PPSU-6EG) and 10wt% EG (14PPSU-10EG). The presence of EG in the 14PPSU membrane dope solution was expected to allow formation of bigger and more uniform pore structures (interconnected finger-like) on the membrane surface.

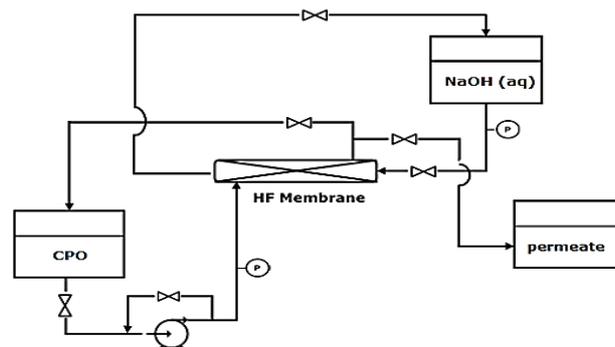


Fig. 1. Schematic diagram of HF membrane contactor system.

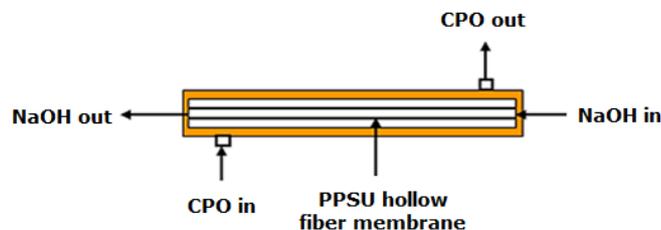


Fig. 2. Flow direction of CPO stream and NaOH stream within a membrane housing.

## 2.8. Oil samples analysis

FFA and soap contents in the oil samples were determined according to the American Oil Chemists' Society (AOCS) Official Method – Ca 5a-40 and Cc 17-95, respectively.

From the SEM micrographs shown in Figure 3 (a, c, e and g), it was proven that the addition of EG into the 14PPSU membrane dope solution has assisted in the formation of more uniform interconnected finger-like structure by suppressing the macrovoids formation. Besides, Figure 3a has clearly shown a change in the 14PPSU membrane morphology from a very large macrovoid at a central membrane layer to a formation of double-layer finger-like structure in the 14PPSU-10EG membrane.

Moreover, Figure 3-b,d,f and h indicates that increasing the EG concentration promoted the formation of bigger pore structure on the membrane surface. This is in accordance to Pei et al. [23] which reported that as the PEG concentration increased, the surface pores enlarged, and the membrane surface became more porous due to lower thermodynamic stability of the casting solution and thus accelerates phase separation. Besides, Ma et al. [21] reported that the increase in dosage of additive (PEG 400) has increased the inflow rate of water diffusion in the polymer solution film because of its hydrophilicity and led to larger finger-like pores. The developed pores were expected to increase the mass transfer, and better extraction of FFA by NaOH across the membrane barrier, as well.

## 3.2. Membrane wettability

The wettability of membrane was characterized using membrane contact angle analyser. EG is a water soluble polymer. So, addition of EG into the 14PPSU has introduced hydrophilicity into the membrane system. This can be observed in Table 3 where the wettability of the 14PPSU membrane (85.23°)

was decreased significantly ( $P < 0.05$ ) with the addition of 2 wt% EG (82.14°), 6 wt% EG (78.18°) and 10 wt% EG (76.7°) into the membrane dope solution. Nevertheless, there is no significant difference ( $P > 0.05$ ) in the surface contact angle between the 14PPSU-6EG and 14PPSU-10EG membranes.

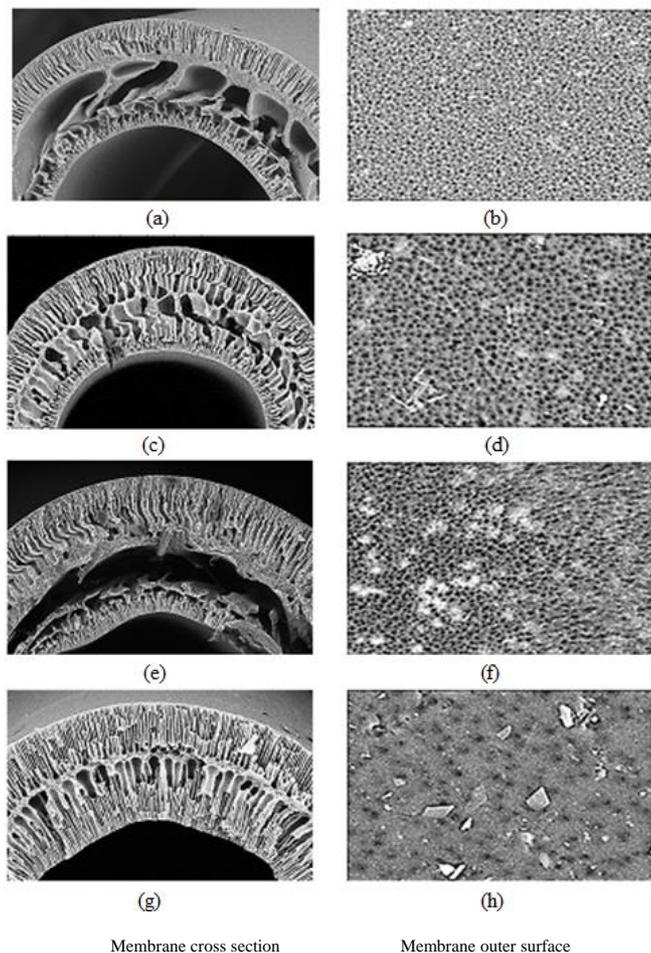


Fig. 3. SEM cross-sectional and outer surface images (400X and 5000X magnifications) of (a, b) 14PPSU-0EG; (c, d) 14PPSU-2EG; (e, f) 14PPSU-6EG; (g, h) 14PPSU-10EG.

Table 3  
Membrane contact angle of PPSU membranes at different EG concentration.\*

Membrane	Contact Angle (°)
14PPSU-0EG	85.23 ± 0.77 <sup>A</sup>
14PPSU-2EG	82.14 ± 1.32 <sup>B</sup>
14PPSU-6EG	78.18 ± 2.78 <sup>C</sup>
14PPSU-10EG	76.70 ± 1.66 <sup>C</sup>

\* Each value in table represents the mean ± standard deviation of sample analysis from triplicate analysis. Mean within column with different superscripts letter A, B, C are significantly different ( $P < 0.05$ ).

3.3. Membrane pore size measurement

Table 4 shows the range and mean pore size of the 14PPSU membrane characterized by image analysis from FESEM micrograph at different EG concentration present in the membrane dope solution. It can be observed that 14PPSU added with 2 to 6 wt% EG had increased the range of membrane pore size from 22.3 nm to 51.4 nm, respectively. However, only 14PPSU added with 6 wt% EG has a significant different ( $P < 0.05$ ) in average pore size compared with the 14PPSU-0EG membrane. Whereas, the 14PPSU membrane added with 10 wt% EG has significantly ( $P < 0.05$ ) reduced the average membrane pore size from 31.27 nm (14PPSU-6EG) to 22.84 nm,

which is even lower than the average pore size of the 14PPSU-0EG membrane (24.81nm). It may be due to dissolution of EG and caused dope solution became thermodynamically less stable as explained by Ma et al. [21]. This had resulted in a poor correlation ( $R^2=0.2624$ ) between the EG concentration and the average pore size formation, where no significant difference ( $p > 0.05$ ) was recorded in a present of more than 6 wt% EG in the 14PPSU membrane formation.

The change in membrane pore size was due to the presence of EG in the polymer structure. High mutual affinity of solvent for water and instantaneous demixing is resulted in the formation of finger-like cavities in the membranes' structures. Besides, the dissolution of EG consumed some of the solvent and led to higher polymer concentration and higher viscosity of membrane dope solution, as well. It caused the membrane dope becomes thermodynamically less stable which resulted in rapid instantaneous demixing when the membrane dope was immersed into the coagulation bath. Similar observation was reported by Ma et al. [21].

Table 4  
Membrane pore size of 14PPSU membranes at different EG concentration.\*

Membrane	Range pore size (nm)	Mean pore size (nm)
14PPSU-0EG	22.3 to 31.3	24.81 ± 3.07 <sup>B</sup>
14PPSU-2EG	22.3 to 33.5	26.31 ± 4.45 <sup>AB</sup>
14PPSU-6EG	22.3 to 51.4	31.27 ± 9.16 <sup>A</sup>
14PPSU-10EG	17.86 to 26.8	22.84 ± 3.33 <sup>B</sup>

\* Each value in table represents the mean ± standard deviation of sample analysis from triplicate analysis. Mean within column with different superscripts letter A, B are significantly different ( $P < 0.05$ ).

3.4. The effect of 14PPSU membrane at difference EG concentration on FFA removal

Figure 4 shows the performance of 14PPSU membranes at different EG concentration with regards to the FFA removal via contactor system. The FFA removal was dropped from the highest of 16.54% in 14PPSU-0EG to less than 0.93% in 14PPSU-10EG membrane contactor system. The presence of EG in 14PPSU dope solution unfortunately has reduced the membrane contactor performance, which might be due to the greater reduction in membrane hydrophobicity. Reduction in the oil wettability on the membrane surface may hinder the interfacial contact between CPO and NaOH at the mouth of membrane pores and resulted in lower FFA removal. The initial hypothesis that adding EG into the membrane dope solution can improve the FFA removal due to better interfacial contact area was denied as the reduction in membrane hydrophobicity becomes more dominant rather than improving the membrane pore size.

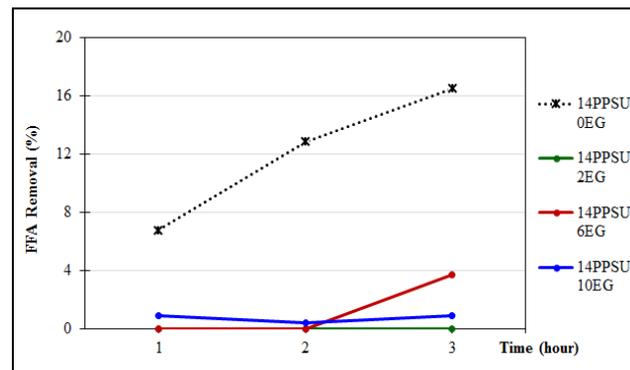


Fig. 4. FFA removal of 14PPSU membranes contactor performance at different EG concentration.

Theoretically, the change in the internal layer of 14PPSU membrane from a macrovoids and finger-like microporous structure to a double-layer finger-like microporous structure with the present of EG can increase the capillary driving force for the better FFA extraction process by NaOH. It is proven that adding EG can improve the membrane pore formation and interconnectivity of those pores. Besides, the membrane pore size was slightly increased with

the present of 2 – 6% EG in 14PPSU membrane dope solution. However, Kim and Lee [24] reported that the increase in the ratio of PEG additive to NMP can caused the membrane dope solution becomes less stable, water flux to membrane increase and the solute rejection decreased.

### 3.5. The effect of 14PPSU membrane at difference EG concentration on soap formation

It is an important indicator for the membrane contactor system to remove FFA from feed CPO without contaminating the oil with soap formation. PPSU was chosen as a membrane contactor module due to its hydrophobicity that allows the oil to wet the membrane surface and prevents the NaOH from penetrating into the CPO stream. The selected PPSU membrane was proven highly tolerant to chemical resistance regardless of polymer and chemical concentration as no soap formation was detected in the permeate sample collection. Besides, the setting pressure for both CPO and NaOH streams was carefully established to facilitate FFA extraction but it limits the penetration between the two separated streams at the mouth of membrane pores only. Aside from the low FFA rejection, the robustness of the PPSU membrane with regards to the chemical resistance was maintained. Although the properties of 14PPSU membrane has been altered with the presence of EG in membrane dope solution. No soap formation was detected in a permeate sample throughout 3 hours operation.

## 4. Conclusions

Based on the experimental results obtained in this study, It was proven that the addition of EG into the 14PPSU membrane dope solution has assisted in the formation of more uniform interconnected finger-like structure of membrane internal layer. Increasing the EG concentration into 14PPSU membrane had changed a microporous finger-like morphology with macrovoid structure into a formation of interconnected double-layer finger-like structure in the 14PPSU-10EG membrane.

Besides, the presence of EG in the membrane dope solution had reduced the hydrophobicity of the 14PPSU membrane from the contact angle of 85.23° to 76.7° in 14PPSU-10EG. However, insignificant change in the membrane pore size was observed where poor correlation ( $R^2=0.264$ ) was recorded between the EG concentration and the average membrane pore size. Poor FFA rejection was recorded in the presence of EG with the highest FFA rejection was only 3.7% by the 14PPSU-6EG membrane comparing the membrane without EG (14PPSU-0EG) that performed up to 16.54% FFA rejection.

For future works in this research area, it is recommended to enhance the hydrophobicity of the 14PPSU membrane by blending with other hydrophobic polymer materials or additives to enhance oil absorption/wettability on the membrane internal layer. Hence it will increase the interfacial contact area and mass diffusion of FFA towards the liquid extractant at the lumen side.

## Acknowledgements

The author (N.H. Othman) is thankful to Sime Darby Research Sdn. Bhd. for supplying crude palm oil and other financial support, and to the members of Advanced Membrane Technology Centre (AMTEC), UTM Skudai, Johor for technical support. P.S. Goh and A.F. Ismail would like to acknowledge HiCOE Grants 4J182 and 4J204 under Ministry of Higher Education for the financial support.

## References

- [1] C.d. Coutinho, M. Chiu, R. Basso, A.B. Ribeiro, L. Gonçalves, L. Viotto, State of art of the application of membrane technology to vegetable oils: A review, *Food Res. Int.* 42 (2009) 536 - 550.
- [2] S.S. Koseoglu, D.E. Engelgau, Membrane application and research in the edible oil industry: An assessment, *J. Am. Oil Chem. Soc.* 67 (1990) 239 - 249.
- [3] L.P. Raman, N. Rajagopalan, M. Cheryan, Membrane technology in vegetable oil processing, *Fat and Oils Intern.* 6 (1994) 28 - 38.
- [4] A.R. Ladhe, N.S.K. Kumar, Application of membrane technology in vegetable oil processing, *Membr. Tech.* (2010) 63 - 78.
- [5] B. Bhosle, R. Subramanian, New approaches in deacidification of edible oils—A review, *J. Food Eng.* 69 (2005) 481–494.
- [6] L. Lin, S. Koseoglu, Membrane processing of fats & oils, in: F. Shahidi (Ed.), *Bailey's industrial oils & fats products*, 6<sup>th</sup> ed., John Wiley & Sons, Inc., 2005.
- [7] J.B. Snape, M. Nakajima, Processing of agricultural fats and oils using membrane technology, *J. Food Eng.* 30 (1996) 1-41.
- [8] R. Ceriani, A. Meirelles, Simulation of continuous refiners for edible oil deacidification, *J. Food Eng.* 76 (2006) 261–271.
- [9] M. Cheryan, Membrane technology in the vegetable oil industry, *Membr. Tech.* 2 (2005) 5-7.
- [10] H.J. Zwijnenberga, A.M. Krosse, K. Ebert, V. Peinemann, F.P. Cuperus, F.P. Acetone-stable nanofiltration membranes in deacidifying vegetable oil, *J. Am. Oil Chem. Soc.* 76 (1999) 83–87.
- [11] S. Koike, R. Subramanian, H. Nabetani, M. Nakajima, Separation of oil constituents in organic solvents using polymeric membranes, *J. Am. Oil Chem. Soc.* 79 (2002) 937–942.
- [12] R.R. Jala, Z. Guo, X. Xu, Separation of FFA from partially hydrogenated soybean oil hydrolysate by means of membrane processing, *J. Am. Oil Chem. Soc.* 88 (2011) 1053–1060.
- [13] R. Subramanian, M. Nakajima, T. Kimura, T. Maekawa, Membrane process for premium quality expeller-pressed vegetable oils, *Food Res. Intern.* 31 (1998) 587–593.
- [14] R.A. Azmi, P.S. Goh, A.F. Ismail, W.J. Lau, B.C. Ng, N.H. Othman, A.M. Noor, M.S.A. Yusoff, Deacidification of crude palm oil using PVA-crosslinked PVDF membrane, *J. Food Eng.* 166 (2015) 165–173.
- [15] A. Criscuoli, Membrane contactors, in: E. Drioli, L. Giorno (Eds.), *Membrane operations*, Wiley-VCH, Weinheim, 2009, pp. 449–460.
- [16] J.G. Crespo, I. M. Coelho, R.M.C. Viegas, Membrane contactor: membrane separation, in: I.D. Wilson, E.R. Adlard, M. Cooke, C.F. Poole, 6<sup>th</sup> Eds., *Encyclopedia of separation science*, Academic Press, San Diego, 2000, pp. 3303–3311.
- [17] R. Pethrick, Compositional and failure analysis of polymers: A practical approach, in: J. Scheirs, *Polymer International*, John Wiley & Sons, England, 2000, pp. 1054.
- [18] N. Jullok, S. Darvishmanesh, P. Luis, B. Van der Bruggen, The potential of pervaporation for separation of acetic acid and water mixtures using polyphenylsulphone membranes, *Chem. Eng. J.* 175 (2011) 306 - 315.
- [19] D.B. Mosqueda-Jimenez, R.M. Narbaitz, T. Matsuura, G. Chowdhury, G. Pleizier, J.P. Santerre, Influence of processing conditions on the properties of ultrafiltration membranes, *J. Membr. Sci.* 231 (2004) 209-224.
- [20] B. Jung, J.K. Yoon, B. Kim, H.W. Rhee, Effect of molecular weight of polymeric additives on formation, permeation properties and hypochlorite treatment of asymmetric polyacrylonitrile membranes, *J. Membr. Sci.* 243 (2004) 45-57.
- [21] Y. Ma, F. Shi, J. Ma, M. Wu, J. Zhang, C. Gao, C. Effect of PEG additive on the morphology and performance of polysulfone ultrafiltration membranes, *Desalination* 272 (2012) 51–58.
- [22] Z. Harun, M.R. Jamalludin, M.Z. Yunus, M.F. Shohur, A.F. Ismail, A.F. (2013). The effect of crystalline rice husk silica on polysulfone membrane for wastewater treatment, *Appl. Mech. Mater.* 701 (2013) 319–322.
- [23] L. Pei, W. Zhao, L. Zhang, Preparation and characterization of porous poly(vinylidene fluoride) membranes for dehumidification with poly(ethylene glycol) as an additive, *J. Appl. Polym. Sci.* 118 (2010) 2696–2703.
- [24] J. Kim, K.J. Lee, Effect of PEG additive on membrane formation by surface inversion, *J. Membr. Sci.* 138 (1998) 153–165.