Impurity Removal of Waste Cooking Oil Using Hydrophobic Polypropylene Hollow Fiber Membrane

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Abstract. Removal of impurities from cooking oil is an important step in providing the possibility of WCO reuse to extend the life cycle of cooking oil, leading to a reduction of WCO disposal. This study was conducted to investigate the performance of a polypropylene (PP) hollow fiber ultrafiltration (UF) membrane for removal of impurities from WCO. The results showed that the membrane could remove water content up to 95% (at 0.1 MPa and 30 °C), but the color improvement was only 9.5% as indicated by the absorbance reduction. Within the range of the operation conditions (i.e. a trans-membrane pressure of 0.1-0.2 MPa and a temperature of 30-50 °C), the oil flux varied from 0.3 $L.m^{-2}.h^{-1}$ to 1.3 $L.m^{-2}.h^{-1}$. In long-term operation, the membrane wettability was improved as shown by the oil contact angle decreasing from 28.2 ± 1.5° to 14.4 ± 0.5°. This resulted in a higher oil flux. At the same time, the hydrophobicity was also increased, as indicated by an increase in the water contact angle from 95.4 ± 0.7° to 97.3 ± 1.1°.

Keywords: *hydrophobicity; polypropylene; reuse; used oil; waste oil; water removal.*

1 Introduction

A large quantity of waste cooking oil (WCO) is generated daily, which may pose environmental problems if not handled properly. Cooking oil is exposed to high temperatures of up to 200 °C during the cooking process, which leads to the formation of hazardous substances. These hazardous substances are potentially dangerous to human health and can have a detrimental effect on the environment [1]. The impurities of WCO are derived from hydrolysis of triglycerides, oxidation, and dimerization or polymerization during frying [2]. The triglyceride is hydrolyzed to form diacylglycerol and free fatty acids (FFA) due to the presence of water in WCO [3]. The values of FFA, viscosity, total polar matter, and color of WCO are higher than those of fresh cooking oil, but

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the oxidative stability index is lower [4,5]. WCO has higher viscosity than fresh oil due to heat-driven polymerization during the cooking process [6]. Combined with particulate impurities formed from solids in cooked or fried foodstuffs, WCO exhibits higher turbidity and a darker color than fresh cooking oil. Cooking and especially frying produce complex reaction products that contribute significantly to the change in color. Melanoidines are identified as one of the major responsible components [7].

Removal of impurities is an attractive way to improve WCO quality. It can provide the possibility of WCO reuse to extend the life cycle of cooking oil, leading to a reduction of WCO disposal [7,8]. In addition, the treated WCO may be used as feedstock for fatty acid methyl ester production [9,10]. Clarification of WCO can be carried out using extraction [1,11,12], adsorption [4,13-15], and the latest membrane technology [16,17]. Extraction using supercritical fluids allows separating WCO into non-polar triglycerides that are dissolved in the non-polar fluid and undissolved polar compounds from oil degradation during the frying process [11,18]. This technique is operated at low temperature, has a relatively low energy requirement and has good separation performance [18]. Commonly used supercritical fluids for WCO treatment are carbon dioxide [1,11,18] and ethane [12,19]. High triglyceride removal can be achieved by two-stage supercritical carbon dioxide extraction [18]. The extraction using supercritical carbon dioxide shows good separation of triglycerides from polymeric compounds, but cannot well separate low molecular weight polar compounds from the triglyceride fraction [12].

In the adsorption process, many types of adsorbents are used, namely bentonite [14,15], calcium silicate, SiO_2, $Al(OH)_3$+water, aluminum silicate, alumina, silica and citric acid, porous rhyolite, citric acid and water, activated carbon, magnesium silicate [20,21], silica gel, magnesium oxide, activated clay, and aluminum hydroxide gel [13], and sodium silicate films [8]. Silica gel is the most effective adsorbent for treating WCO, achieving the highest value of FFA removal reached (98%) [14]. However, adsorption has disadvantages, such as contamination of the oil by foreign powders and metals, legal issues, and high cost [22].

Membrane filtration is an interesting alternative for WCO clarification. Using a membrane has attractive features such as low-temperature operation, high separation efficiency, scalability, high packing density, low footprint, and high product quality [23-26]. Membrane filtration can effectively reduce polar compounds in WCO, including oxidation products and polymers, resulting in fewer polar compounds, improved color and viscosity, while the oil stability deteriorates [7]. Polar compounds such as color, polymers, oxidation products, phosphatides, and nitrogen compounds are selectively rejected. On the other hand, nonpolar compounds such as triglycerides are permeated due to their solubility and the hydrophobic properties of the membrane [16].

Membrane-based processes have been used in filtration of vegetable oil as reported in the literature [27-30]. However, membranes have lower flux due to the high viscosity of vegetable oil [31,32]. Increasing the operating temperature and pressure and employing a solvent is usually done to improve the flux [28,33]. Commonly, there are several modes of membrane filtration, namely microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). The membranes used in these filtration methods have pore sizes according to the following order: MF > UF > NF > RO [34]. MF membranes allow a higher flux due to their large pore size (1-5 μm) [34] but cannot improve the color of the WCO [7]. On the other hand, dense or nonporous membranes, e.g. as used in nanofiltration, yield better color removal than porous membranes but require high operating pressure and allow only very low permeate flux [7]. Therefore, it is necessary to find a membrane that allows a high oil flux and high impurity rejection.

UF membranes offer better rejection of impurities than MF and higher oil flux than NF or dense membranes. In a previous study, it has been demonstrated that a polypropylene (PP) membrane with high hydrophobicity showed good performance in clarification of crude palm oil [35]. This work investigated the performance of a hollow fiber polypropylene (PP) UF membrane for the removal of WCO impurities. The effects of operating conditions and membrane wettability on membrane performance were studied.

2 Materials and Methods

The sample used in the experiment was obtained from cooking oil that was used for cooking chicken meat for 5-8 times at 170-200 °C. The characteristics of the WCO are tabulated in Table 1. The PP membrane was supplied by GDP Filter Indonesia and had an effective surface area of 0.95 m^2, a pore size of 50 nm, an outside diameter of 0.4 mm, and an inside diameter of 0.3 mm.

Table 1 Specifications of waste cooking oil (WCO).

Parameter	Percentage (wt %)
Moisture	0.20
FFA	0.64
Insoluble solids	0.54

Figure 1 illustrates the experimental set-up for testing the UF membrane. In this experiment, 1 liter of WCO was placed into a beaker glass and fed into the membrane module from the shell-side while the permeate flowed through the lumen-side (outside-in configuration). The temperature was adjusted by placing the beaker glass in a water-bath. Trans-membrane pressure (TMP) was controlled by adjusting the opening valve in the feed and retentate stream. The retentate was recirculated to the feed while the permeate was collected in a tank. Filtration was conducted in batch mode at a TMP of 0.1-0.2 MPa and an operating temperature of 30-50 °C. Air scouring was conducted for 1-2 minutes in every run before the next filtration to remove the residual oil in the membrane pores. Long-term filtration was also conducted to analyze the stability of the membrane flux.

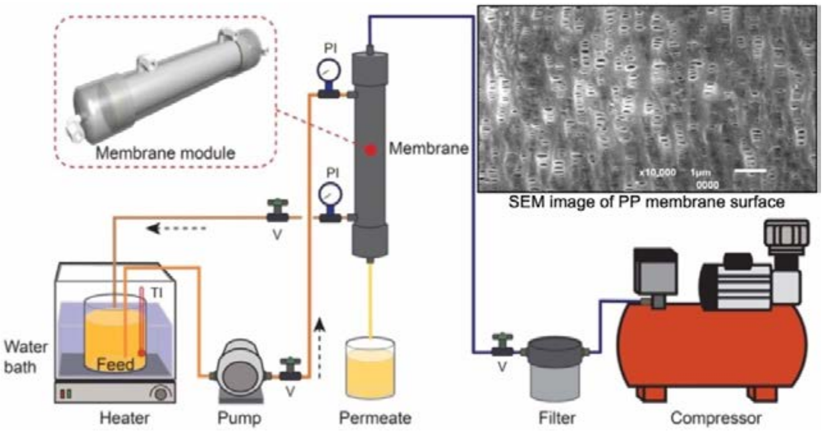

Figure 1 Experimental set-up of WCO filtration.

Membrane performance is expressed as the reduction percentage of impurities and the flux of the permeate. In this work, the ability of the membrane to remove water and to improve oil color was evaluated. The analysis of water content was based on the AOCS Ca 2c-25 standard using the air-oven method. The test was carried out by heating 5-10 gram of sample at 110 °C for 1 hour. After that, the oil was cooled in a desiccator and then weighed. This procedure was repeated until the mass of the oil was constant and the water content in the permeate was calculated using Eq. (1).

$$Water\ content = \frac{m_1 - m_2}{m_1}\ x\ 100\% \tag{1}$$

where m_1 is the initial sample mass (g), and m_2 is the mass of oil after being dehydrated and having reached a constant value (g).

Water removal or rejection (R) was determined using Eq. (2):

$$R = \frac{C_F - C_P}{C_F}\, x\ 100\% \tag{2}$$

where C_F and C_P are the concentrations of each impurity in the feed and permeate, respectively (wt%).

The permeate flux was determined using Eq. (3).

$$J = \frac{\Delta V}{\Delta t . A} \tag{3}$$

where J represents the permeate flux ($L.m^{-2}.h^{-1}$), ΔV and Δt are the volume change (L) and duration (h), while A is the effective area of the membrane (m^2). The changes in membrane properties during filtration were evaluated by conducting filtration in long-term operation. The changes of the flux during filtration were observed for 75 minutes at TMP 0.1 MPa and room temperature (~26 °C).

Measurement of the acid value (FFA) was conducted by titrating the oil with alcoholic potassium hydroxide solution. The acid value was calculated using Eq. (4).

$$Acid\ Value = (Mr \times V) \times (\frac{N}{W}) \tag{4}$$

where Mr, N, and V are the molecular weight (g/mol), normality (eq/L), and volume (L) of potassium hydroxide, respectively, while W is the weight of the sample taken (g).

Generally, insoluble impurities in oils are defined as materials that remain insoluble and can be filtered out [36]. The insoluble impurities in the oil were determined according to ISO 663: 2000(E). After being treated with an excess of n-hexane or light petroleum, samples of WCO/solvent mixture were filtered. The filter and residue were washed with the same solvent and dried at 103 °C and then weighed to determine the number of insoluble impurities present in each oil sample [37].

The absorbance of the feed and permeate were determined by analyzing the absorption spectra in the visible range (400 to 700 nm) using a UV-Vis spectrophotometer (Shimadzu UV-120-02). The presence of impurities results in a darker color of the WCO, which can be detected by visible wavelengths. In addition, the absorbance value of the WCO has greater values at a lower wavelength range [7]. The absorbance investigation this study was conducted at a wavelength of 430-480 nm.

Meanwhile, membrane wettability was analyzed by measuring the contact angle between water and oil droplets dropped onto the membrane surface before and after use. The water contact angle (WCA) and oil contact angle (OCA) indicate the hydrophobic and oleophilic properties of the membrane, respectively [38-

40]. Membrane surface wettability was determined by measuring the static contact angles of liquid droplets [41,42]. In this study, the contact angles were determined by dropping 1 μL of deionized water and fresh oil, respectively, onto the membrane surface. The contact angles formed by the droplets and the membrane surface were determined by using an image processing software application.

3 Result and Discussion

3.1 Color of Fresh and Treated WCO

Color is the main property to determine the quality of frying oil. WCO has 10-20 times higher absorbance than fresh cooking oil [6,7]. The presence of unsaturated carbonyl components and browning pigment from the fried food is polymerized and dissolved in the cooking oil, resulting in the dark color of WCO [43]. Figure 2 shows the absorbance value of WCO before and after ultrafiltration at 0.1 MPa and 30 °C. The color improvement achieved by the UF membrane was relatively insignificant, as expressed by the absorbance value of the permeate. The improvement was only 5.2% at a wavelength of 460 nm, while at a wavelength of 480 mm it reached 9.5%. This value is higher than that of fresh oil, which is only 0.088 at 460 nm.

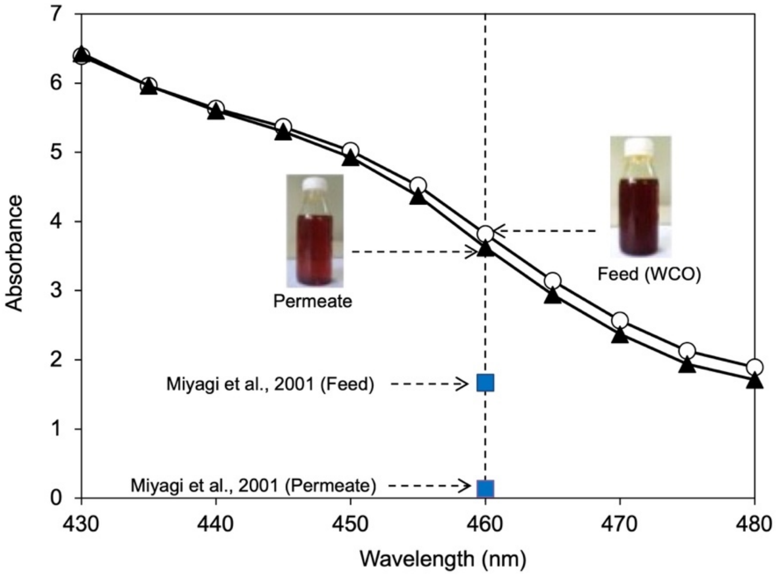

Figure 2 Absorbance values of WCO at 430-480 nm wavelengths.

The low color removal could be because the majority of the polar compounds, e.g. oxidation products, polymers, and color compounds, cannot be rejected by a porous membrane [7]. This may imply that the UF membrane was only able to remove particulate matter. Therefore, to improve WCO color, more post-treatment is required.

3.2 Effect of Applied Pressure

Generally, the flux increases with the applied pressure while impurity rejection decreases. Figure 3 shows the effect of applied pressure (TMP) on the flux and water removal. The flux varied in the range of 0.85 $L.m^{-2}.h^{-1}$ to 1.33 $L.m^{-2}.h^{-1}$ at operating conditions of 0.1-0.2 MPa and 40 °C. The hollow fiber PP-UF membrane reduced the water content up to 83% at 1 bar and 40 °C. Water rejection is more effective at low pressure due to the more intensive contact between the membrane and the oil. A higher TMP also increases the drag force for both oil and water onto the membrane surface. This condition enhances the wetting and coalescence of the water droplets on the membrane surface as an effect of increased convection and forces some of the water to pass through the membrane pores, leading to a decrease in water removal [44]. On the other hand, the color or absorbance value of treated WCO did not change by increasing TMP.

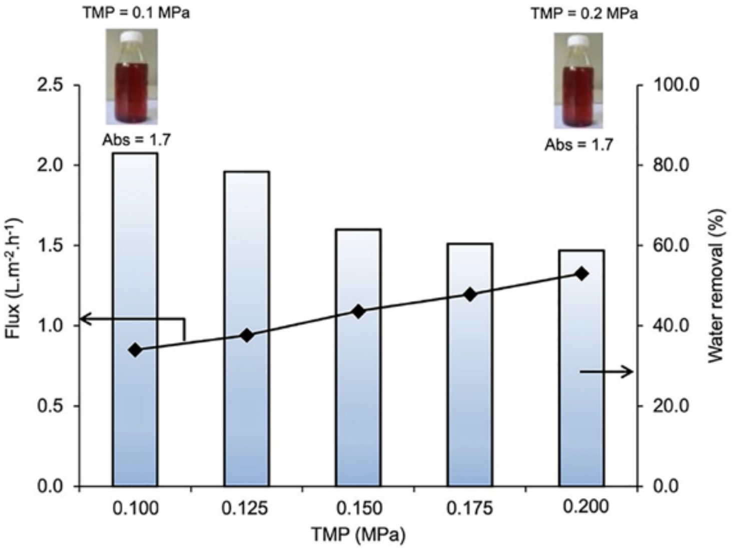

Figure 3 Flux and water removal at various TMP (T = 40 °C)

3.3 Effect of operating temperature

Figure 4 shows the permeate flux and water removal characteristics at various temperatures. Within this range of operating temperatures, the differences in flux are quite large compared to the effect of applied pressure. Impurities in WCO such as fatty acid esters, glycerol, the formation of dimeric and polymeric acids and glycerides result in a higher oil viscosity [45,46]. The increase in flux as a function of temperature is associated with lower viscosity. It has been reported that the viscosity of vegetable oil decreases by 30% for every 10 °C [47]. By increasing the oil temperature, the viscosity becomes lower, resulting in a faster mass transfer of the oil through the membrane [48].

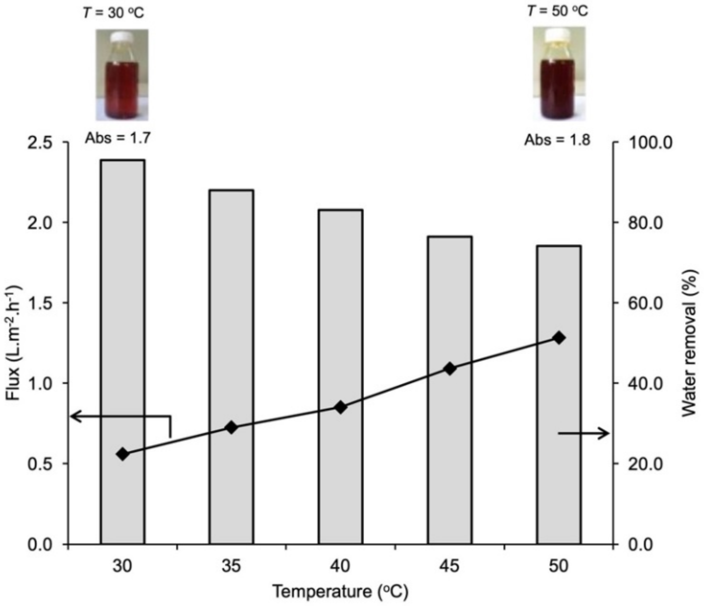

Figure 4 Flux and water removal at various T (TMP = 0.1 MPa).

The best reduction of moisture content in the WCO was 95% at operating conditions of 0.1 MPa and 30 °C. This result indicates that ultrafiltration of WCO using a hollow fiber PP membrane is capable of producing treated oil with a water content of 0.03%, i.e. below the national standard of 0.1% [49]. In contrast, increasing the temperature resulted in lower color reduction, as shown by the absorbance values. This may be due to the increase of the solubility of the dissolved components in the oil that pass through the membrane pores along with the permeate. Miyagi, *et al.* [7] have reported that the increasing temperature in the range of 25-40 °C only results in a 0.6% color improvement.

Table 2 shows the performances of the membrane process in the WCO treatment. Compared to other membranes in several reported studies, the membrane used in this study could achieve a higher oil flux at relatively low pressure as well as high water removal. However, the color removal was still lower than that of other reported membrane performances. Therefore, the PP UF membrane requires additional post-treatment of the WCO for color improvement.

Table 2 Performances of membrane filtration in WCO treatment.

Membrane	**Operating conditions**	**Performance**				**Ref.**
		Flux	**Water removal (%)**	**Color removal (%)**	**Viscosity improvement (%)**	
Polyimide NTGS-2200	1-4 MPa; 25 °C.	0.09 kg.m^{-2}.h^{-1}	-	98 (λ: 420nm)		[50]
PTFE-MF (pore size = 0.05 μm) NF MPF-60 (MWCO = 400 Da) Nonporous NTGS-2200	2 -5 MPa; 25-40°C.	- - 0.063-0.121 kg m^{-2}.h^{-1}	- -	- No improvement - Marginally improvement - 91.8 (λ: 400-700 nm *)	- 0.54 - 1.44 - 13.31	[7]
Polysulfone (NTGS-AX, NTGS-BX, NTGS-CX) Polyimide NTGS-220	0.3-4 MPa; <30°C.	0.05-0.56 kg.m^{-2}.h^{-1} 0.12-0.06 kg.m^{-2}.h^{-1}	-	83-93 (Color Lovibond AOCS, Cc 13e-92)	-	[16]
PP membrane (pore size = 0.05 μm)	0.1-0.2 MPa; 26-50°C.	0.3-1.3 L.m^{-2}.h^{-1}	95	5.2 (λ: 460 nm); 9.5 (λ: 480 nm)	-	Current Research

* Wavelength used in UV-Vis spectroscopy (absorbance measurement)

3.4 Membrane Wettability

Wettability is highly affected by the surface characteristics of the membrane. The two main wetting characteristics of the membrane surface toward water are hydrophilicity and hydrophobicity. On the surface of a hydrophilic membrane, a water droplet tends to spread out and has a contact angle of smaller than 90°, while on the surface of a hydrophobic membrane, the water will minimize its contact with the membrane, resulting in a contact angle higher than 90°. In this work, measurement of WCA and OCA was carried out to determine the water and oil wettability of the membrane. The change in membrane properties during the experiments was observed by comparing WCA and OCA of fresh and used membranes. Figure 5 shows the contact angle profiles observed for water and oil droplets on the PP membrane fiber membrane before and after usage.

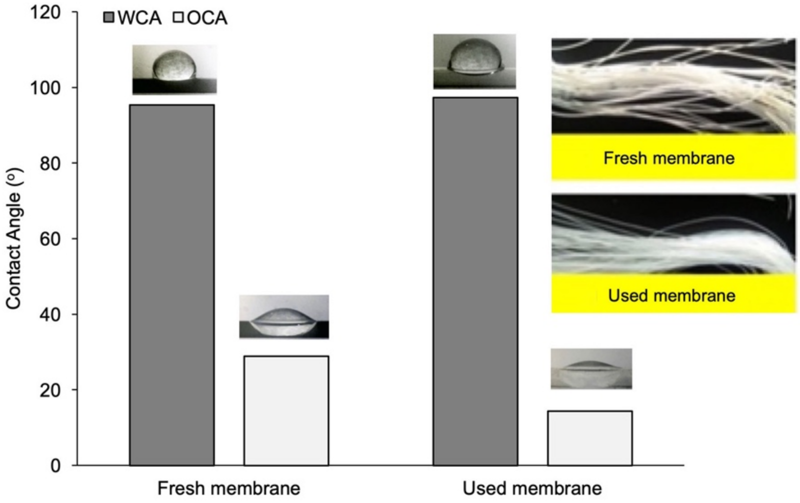

Figure 5 Water contact angle (WCA) and oil contact angle (OCA) on the PP membrane before and after usage.

Based on the experimental results, the membrane that had contact with oil became transparent (inserted image). The OCA profile of the used membrane surface was 14.4 ± 0.5°, much smaller than that of a new membrane, which is 28.2 ± 1.5°. The decrease in OCA indicates that the membrane becomes slightly more hydrophobic. This slight increase in membrane hydrophobicity is highly likely due to the presence of the residues in the cooking oil on top of the membrane or the interaction between oil and PP. Meanwhile, the average value of the WCA of the membrane surface after use was 97.3 ± 1.1°. This is slightly higher than that of a new membrane (95.4 ± 0.7°). This indicates that the membrane became more hydrophobic.

After filtration, the membrane characteristics changed as shown by the values of OCA and WCA. The results indicate that once the membrane has had contact with oil, the membrane becomes more hydrophobic, as shown by the increase of the WCA value, and also becomes more oleophilic, as shown by the decrease of the OCA value. This change of membrane properties leads to a more permeable membrane (Figure 6). The increased membrane permeability is beneficial for long-term operation. As the membrane becomes more hydrophobic and oleophilic, it is expected to obtain better membrane performance in terms of oil flux and water removal. However, further investigation is needed to ensure membrane property stability (preserved hydrophobicity and oleophilicity) as well as to investigate the swelling tendency, which may reduce the membrane's selectivity.

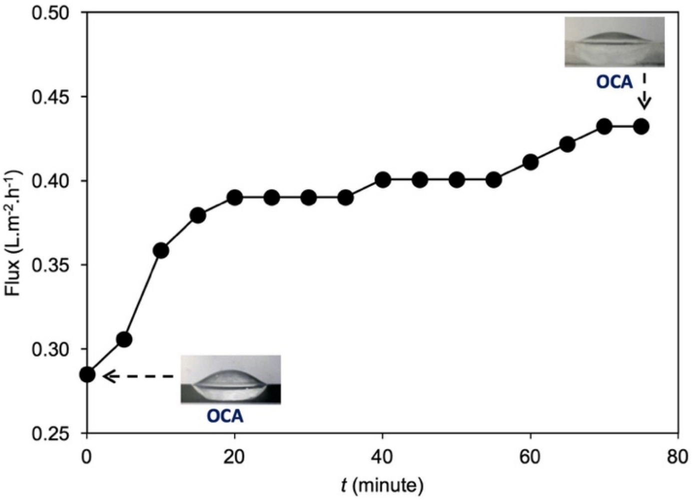

Figure 6 Permeate flux of WCO ultrafiltration in long-term operation (26 °C and 0.1 MPa).

4 Conclusion

In this study, the performance of a PP UF hollow fiber membrane for removal of impurities from WCO was investigated. The results show that the membrane could remove up to 95% water (at 0.1 MPa and 30 °C). Unfortunately, the membrane could only improve WCA clarity by 9.5%, as indicated by the reduction of the absorbance value from 1.89 to 1.7 at 480 nm wavelength. Within the range of the operating conditions used in this study, the oil flux varied from 0.3 $L.m^{-2}.h^{-1}$ to 1.3 $L.m^{-2}.h^{-1}$.

In long-term operation, the membrane showed an increase of wettability towards oil. This phenomenon was indicated by a decreased value of the OCA (from 28.2 ± 1.5° to 14.4 ± 0.5°). On the other hand, the hydrophobicity of the membrane also increased, as shown by the change of the WCA from the initial value of 95.4 ± 0.7° to 97.3 ± 1.1°. These phenomena result in a higher oil flux and are expected to improve the water removal, which is beneficial for long-term filtration operation.

Nomenclature

FFA : free fatty acids

OCA	: oil contact angle
PP	: poly propylene
PTFE	: poly tetra fluoro ethylene
PVDF	: polyvinylidenefluoride
TMP	: trans-membrane pressure
UF	: ultrafiltration
WCA	: water contact angle
WCO	: waste cooking oil

References

[1] Osseo, L.S., Caputo, G., Gracia, I. & Reverchon, E., *Continuous Fractionation of Used Frying Oil by Supercritical* CO_2, Journal of the American Oil Chemists' Society, **81**(9), pp. 879-85, 2004.

[2] Choe, E. & Min, D., *Chemistry of Deep Fat Frying Oils*, Journal of Food Science, **72**(5), pp. R77-R86, 2007.

[3] Dobarganes, M.C., *Formation of New Compounds during Frying-General Observations*, AOCS Lipid Library, http://lipidlibrary.aocs.org/OilsFats/content.cfm?ItemNumber=39209, (13 December 2018).

[4] Bhattacharya, A.B., Sajilata, M., Tiwari, S.R. & Singhal, R.S., *Regeneration of Thermally Polymerized Frying Oils with Adsorbents*, Food Chemistry, **110**(3), pp. 562-570, 2008.

[5] Lin, S., Akoh, C.C. & Reynolds, A.E., *The Recovery of Used Frying Oils with Various Adsorbents*, Journal of Food Lipids, **5**(1), pp. 1-16, 1998.

[6] Paul, S., Mittal, G. & Chinnan, M., *Regulating the Use of Degraded Oil/Fat in Deep-Fat/Oil Food Frying*, Critical Reviews in Food Science and Nutrition, **37**(7), pp. 635-662, 1997.

[7] Miyagi, A., Nakajima, M., Nabetani, H. & Subramanian, R., *Feasibility of Recycling Used Frying Oil Using Membrane Process*, European Journal of Lipid Science and Technology, **103**(4), pp. 208-215, 2001.

[8] Kalapathy, U. & Proctor, A., *A New Method for Free Fatty Acid Reduction in Frying Oil Using Silicate Films Produced from Rice Hull Ash*, Journal of the American Oil Chemists' Society, **77**(6), pp. 593-598, 2000.

[9] Cao, L., Wang, J., Liu, K. & Han, S., Ethyl acetoacetate: *A Potential Bio-Based Diluent for Improving the Cold Flow Properties of Biodiesel from Waste Cooking Oil*, Applied Energy, **114**, pp. 18-21, 2014.

[10] Ullah, Z., Bustam, M.A. & Man, Z., *Biodiesel Production from Waste Cooking Oil by Acidic Ionic Liquid as A Catalyst*, Renewable Energy, **77**, pp. 521-526, 2015.

[11] Rincón, J., Camarillo, R., Rodríguez, L. & Ancillo, V., *Fractionation of Used Frying Oil by Supercritical* CO_2 *and Cosolvents*, Industrial & Engineering Chemistry Research, **49**(5), pp. 2410-2418, 2010.

[12] Rincón, J., Martínez, F., Rodríguez, L. & Ancillo, V., *Recovery of Triglycerides from Used Frying Oil by Extraction with Liquid and Supercritical Ethane*, The Journal of Supercritical Fluids, **56**(1), pp. 72-79, 2011.

[13] Miyagi, A. & Nakajima, M., *Regeneration of Used Frying Oils Using Adsorption Processing*, Journal of the American Oil Chemists' Society, **80**(1), pp. 91-96, 2003.

[14] Turan, S. & Yalcuk, A., *Regeneration of Used Frying Oil*, Journal of the American Oil Chemists' Society, **90**(11): pp. 1761-1771, 2013.

[15] Wonglamom, J. & Rakariyatham, N., *Recovery of Used Vegetable Frying Oil by Two Step Adsorbents*, 26th Annual Meeting of The Thai Society for Biotechnology and International Conference, pp. 330-337, 2014.

[16] Subramanian, R., Nandini, K.E., Sheila, P.M., Gopalakrishna, A.G., Raghavarao, K.S.M.S., Nakajima, M., Kimura, T. & Maekawa, T., *Membrane Processing of Used Frying Oils*, Journal of the American Oil Chemists' Society, **77**(3), pp. 323-328, 2000.

[17] Miyagi, A., Nabetani, H. & Subramanian, R., *Purification of Crude Fatty Acids Using A PDMS-Based Composite Membrane*, Separation and Purification Technology, **77**(1), pp. 80-86, 2011.

[18] Yoon, J., Han, B.-S., Kang, Y-C., Kim, K.H., Jung, M.Y. & Kwon, Y.A., *Purification of Used Frying Oil by Supercritical Carbon Dioxide Extraction*, Food Chemistry, **71**(2), pp. 275-279, 2000.

[19] Rincon, J., Canizares, P. & Garcia, M.T., *Regeneration of Used Lubricant Oil by Ethane Extraction*, The Journal of Supercritical Fluids, **39**(3), pp. 315-322, 2007.

[20] Lin, S., Akoh, C.C. & Reynolds, A.E., *Determination of Optimal Conditions for Selected Adsorbent Combinations to Recover Used Frying Oils*, Journal of the American Oil Chemists' Society, **76**(6), pp. 739-744, 1999.

[21] Lin, S., Akoh, C.C. & Reynolds, A.E., *Recovery of Used Frying Oils with Adsorbent Combinations: Refrying and Frequent Oil Replenishment*, Food Research International, **34**(2), pp. 159-166, 2001.

[22] Blumenthal, M.M., *Frying Technology*, Bailey's Industrial Oil & Fat Products, Vol. 3, ed. 5, Hui, Y.H., eds., John Wiley & Sons Inc., pp. 429-481, 1996.

[23] Drioli, E., Stankiewicz, A.I. & Macedonio, F., *Membrane Engineering in Process Intensification-An Overview*, Journal of Membrane Science, **380**(1), pp. 1-8, 2011.

[24] Ariono, D., Khoiruddin, K., Subagjo, S. & Wenten, I.G., *Heterogeneous Structure and Its Effect on Properties and Electrochemical Behavior of Ion-Exchange Membrane*, Materials Research Express, **4**(2), p. 024006, 2017.

[25] Sianipar, M., Kim, S. H., Iskandar, F. & Wenten, I.G., *Functionalized Carbon Nanotube (CNT) Membrane: Progress and Challenges*, RSC Advances, **7**(81), 51175-51198, 2017.

[26] Wenten, I.G., Dharmawijaya, P.T., Aryanti, P.T.P., Mukti, R.R. & Khoiruddin, K. *LTA Zeolite Membranes: Current Progress and Challenges in Pervaporation*, RSC Advances, **7**(47), pp. 29520-29539, 2017.

[27] Subramanian, R., Nakajima, M. & Kawakatsu, T., *Processing of Vegetable Oils Using Polymeric Composite Membranes*, Journal of Food Engineering, **38**(1), pp. 41-56, 1998.

[28] Pagliero, C., Ochoa, N., Marchese, J. & Mattea, M., *Vegetable Oil Degumming with Polyimide and Polyvinylidenefluoride Ultrafiltration Membranes*, Journal of Chemical Technology and Biotechnology, **79**(2), pp. 148-152, 2004.

[29] Koris, A. & Marki, E., *Ceramic Ultrafiltration Membranes for Non-Solvent Vegetable Oil Degumming (Phospholipid Removal)*, Desalination, **200**(1-3), pp. 537-539, 2006.

[30] Mah, S.K., Leo, C.P., Wu, T.Y. & Chai, S.-P., *A Feasibility Investigation on Ultra-Filtration of Palm Oil and Oleic Acid Removal from Glycerin Solutions: Flux Decline, Fouling Pattern, Rejection and Membrane Characterizations*, Journal of Membrane Science, **389**, pp. 245-256, 2012.

[31] Noureddini, H., Teoh, B.C. & Davis Clements, L., *Viscosities of Vegetable Oils and Fatty Acids*, Journal of the American Oil Chemists' Society, **69**(12), pp. 1189-1191, 1992.

[32] Majid, R.A., Baharin, B.S., Ahmadun, F.-R. & Man, Y.B.C., *Processing of Crude Palm Oil with Ceramic Microfiltration Membrane*, Journal of Food Lipids, **7**(2), pp. 113-126, 2000.

[33] Bottino, A., Capannelli, G., Comite, A., Ferrari, F., Marotta, F., Mattei, A. & Turchini, A., *Application of Membrane Processes for The Filtration of Extra Virgin Olive Oil*, Journal of Food Engineering, **65**(2), pp. 303-309, 2004.

[34] Cui, Z.F., Jiang, Y. & Field, R.W., *Fundamentals of Pressure-Driven Membrane Separation Processes*, Membrane Technology. A practical Guide to Membrane Technology and Applications in Food Bioprocessing, Cui Z.F., Muralidhara, H.S., eds., Butterworth-Heinemann, pp. 1-18, 2010.

[35] Wenten, I.G., Victoria, A.V., Tanukusuma, G., Khoiruddin, K. & Zunita M., *Simultaneous Clarification and Dehydration of Crude Palm Oil Using Superhydrophobic Polypropylene Membrane*, Journal of Food Engineering, **248**, pp. 23-27, 2019.

[36] Cocks, L.V. & Rede, C.V., *Laboratory Handbook for Oil and Fat Analysts*, Academic Press, 1966.

[37] International Standard, *ISO 663: 2000(E) Animal and Vegetable Fats and oils-Determination of Insoluble Impurities Content*, International Organization of Standard, 2000.

[38] Himma, N.F., Prasetya, N., Anisah, S. & Wenten, I.G., *Superhydrophobic Membrane: Progress in Preparation and Its Separation Properties*, Reviews in Chemical Engineering, **35**(2), 211-238, 2019.

[39] Himma, N.F., Wardani, A.K. & Wenten, I.G., *Preparation of Superhydrophobic Polypropylene Membrane Using Dip-Coating Method: The Effects of Solution and Process Parameters*, Polymer-Plastics Technology and Engineering, **56**(2), pp. 184-194, 2017.

[40] Widodo, S., Ariono, D., Khoiruddin, K., Hakim, A.N. & Wenten, I.G., *Recent Advances in Waste Lube Oils Processing Technologies*. *Environmental Progress & Sustainable Energy*, **37**(6), 1867-1881, 2018.

[41] Mangindaan, D., Kuo, W.H., Wang, Y.L. & Wang, M.J., *Experimental and Numerical Modeling of the Controllable Wettability Gradient on Poly (Propylene) Created by SF6 Plasma*, Plasma Processes and Polymers, **7**(9-10), pp. 754-765, 2010.

[42] Mangindaan, D., Kuo, C.C., Lin, S.Y. & Wang, M.J., *The Diffusion-reaction Model on the Wettability Gradient Created by SF6 Plasma*, Plasma Processes and Polymers, **9**(8), pp. 808-819, 2012.

[43] Yaghmur, A., Aserin, A., Mizrahi, Y., Nerd, A. & Garti, N., *Evaluation of Argan Oil for Deep-Fat Frying*, LWT-Food Science and Technology, **34**(3), pp. 124-130, 2001.

[44] Chakrabarty, B., Ghoshal, A. & Purkait, M., *Ultrafiltration of Stable Oil-In-Water Emulsion by Polysulfone Membrane*, Journal of Membrane Science, **325**(1), pp. 427-437, 2008.

[45] Marmesat, S., Rodrigues, E., Velasco, J. & Dobarganes, C., *Quality of Used Frying Fats and Oils: Comparison of Rapid Tests Based on Chemical and Physical Oil Properties*, International Journal of Food Science & Technology, **42**(5), pp. 601-608, 2007.

[46] Ruiz-Méndez, M., Marmesat, S., Liotta A., Dobarganes, M., *Analysis of Used Frying Fats for Biodiesel Production*, Grasas Y Aceites; **59**(1), pp. 45-50, 2008.

[47] Timms, R., *Physical Properties of Oils and Mixtures of Oils*, Journal of the American Oil Chemists' Society, **62**(2), pp. 241-249, 1985.

[48] Marenchino, R., Pagliero, C. & Mattea, M., *Vegetable Oil Degumming Using Inorganic Membranes*, Desalination, **200**(1-3), pp. 562-564, 2006.

[49] National Standardization Agency of Indonesia, *SNI 7709:2012 Palm Cooking Oil*, BSN, Jakarta, 2012. (Text in Indonesian)

[50] Miyagi, A., Subramanian, R. & Nakajima, M., *Membrane and Additional Adsorption Processes for Quality Improvement of Used Frying Oils*, Journal of the American Oil Chemists' Society, **80**(9), pp. 927-932, 2003.