PURIFICATION OF TWIN-SCREW EXTRUDER-PRESSED SUNFLOWER OIL USING POLYETHERSULFONE ULTRAFILTRATION MEMBRANES

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ABSTRACT

This study focused on the purification of crude sunflower oil wing ultrafiltration membranes. The experiment was conducted with twin-screw extruder-pressed sunflower oil and three polyethersulfone (PES) membranes cassette with a molecular weight cut-off (MWCO) of 10, 30 and 50 kDa and a permeation area of 0.09 m^2. Several tests were carried out to define the best performance (permeate flux, phospholipids rejection, free fatty acid and color reductions) by studying the influence of operating conditions transmembrane pressure, temperature and tangential flow rate.

Permeate flux was low for all membranes rested but remained stable for a long period No long-term fouling appeared during the membrane processing. The permeate flux increased with MWCO of membrane, temperature, transmembrane pressure and tangential flow rate. The highest permeate flux of 26 l/m^2.h was obtained under operating conditions of 3.5 bar, 50°C and 0.1 m/s with membrane of 50 kDa. Furthermore, ultrafiltration membranes were found to have a positive effect on phospholipids and undesirable components removals, particularly with membrane of 10 and 30 kDa.

Keyword: Ultrafiltration; Polyethersulfone; Sunflower oil; Phospholipids; Purification

INTRODUCTION

Industrial oil extraction from oleaginous seeds is commonly realized through mechanical pressing with a hydraulic or single expeller press, followed by solvent extraction. Today, extensive studies on continuous oil extraction of oilseeds using a twin-screw extruder to generate oil have been successfully carried out (Guyomard, 1994; Bouvier and Guyomard, *1997;* Lacaze-Dufaure et al., 1999a, 1999b; Amalia Kartika et al., 2003a, 2003b, 2004a, 2005, 2006; Amalia Kartika, 2005). *Twin-screw* extruder is gradually more used for production of a wide variety of food products due to its great capability to conduct diverse functions and processes.

Despite the quality of pressed *oil* is usually better than the quality of oil obtained from solvent extraction, a refining process is required to remove undesirable components that impact on flavour, odour and appearance. The main contaminants and impurities are phospholipids, pigments, free fatty acids (FFA) and oxidation products. Conventional edible oil refining is commonly realized through many *steps* including degumming, neutralisation, bleaching and deodorisation. These processes consume *large* amount of energy, water and chemical with much loss of neutral oil, nutrients and natural antioxidants, and the creation of large amount of undesired by products (Snape and Nakajima, 1996).

Owing to its environmental benefits and other advantages including *the* simplicity of the processes, the gentle nature of the separation, the usual low *energy* requirement, no addition of chemicals, easy change of scale and low capital and operating cost (Wankat, 1990), the membrane technology has showed a promising alternative for oil refining (Iwama, 1987; Lin et al., *1997;* Subramanian and Nakajima, 1997; Subramanian et al., 1998a, 1998b, 2001a; Ong et al., 1999; Ochoa et al., 2001; Pagliero et al., 2001,2004; In-Chul Kim et al., 2002; Alicieo et al., 2002; Koris and Vatai, 2002; Miyagi et al., 2003; Amalia Kartika et al., 2003c, 2003d, 2004b; Hafidi et al., 2003, 2004, 2005a, 2005b, 2005c; Bhosle et al., 2005; Artz et al., 2005). The application of this technology can furthermore retain nutrients and other desirable components in oil.

Among the steps of oil refining, the application of oil degumming using membrane technology has attracted attention from a few researchers (Subramanian and Nakajima, 1997; Subramanian et al., 1998a; Ong et al., 1999; Ochoa et al., 2001; In-Chul Kim et al., 2002; Koris and Vatai, 2002: Pagliero et al., 2001, 2004). It concerns the elimination of the phospholipids, also called gums, contained in oil. Classic oil degumming *is* usually realized by

gums/ phospholipids precipitation using a small quantity of water or steam (2-4%) and separation by centrifugation. Most of the studies focused on gum removal from an oil/organic solvent mixture (Iwama, 1987; Snape and Nakajima, 1996; Kwiatkowski and Cheryan, 2005), but only few studies were related to oil degumming without organic solvent. The major limitation in this case comes from low permeate flux as a consequence of high oil viscosity.

The permeate flux can be improved by optimizing operating parameters (transmembrane pressure, temperature, feed velocity) and by applications of tangential flow and suitable membrane module. In process application, tangential or crossflow filtration is most often used due to its ability to create turbulence at the membrane surface and minimize concentration polarization and fouling. In addition, this system can be well adapted when it is applied at different membrane module designs.

The most common designs include flat sheet, spiral wound, tubular, capillary and hollow fiber. Flat sheet, tubular and capillary designs are preferred for tangential filtration of suspensions containing macromolecules and these have intermediate to very low tendency to clog and exhibit high permeation fluxes (Belfort et al., 1994).

The objective of this work was to evaluate different ultrafiltration membranes under different operating conditions for twin-screw extruder-pressed sunflower oils purification. The performance of ultrafiltration membrane was characterized with the permeate flux, the phosphotipids rejection but also the color and the free fatty acid removal. The influence of operating conditions (transmembrane pressure, temperature, tangential flow rate) on these parameters was investigated with three polyethersulfone (PES) membranes cassette with a molecular weight cut-off (MWCO) of 10, 30 and 50 kDa and a permeation area of 0.09 m^2.

MATERIALS AND METHODS

Sunflower oils and chemicals

Crude sunflower oils were produced by pressing oleic sunflower seeds in a CLEXTRAL BC 45 co-rotating twin screw extruder. The characteristic of the crude oils extracted was presented in Table 1. All solvent and chemicals were analytical grades that were obtained from Sigma-Aldrich, Fluka, Prolabo and ICS.

Membrane and membrane module

Experiments were conducted using three membranes from PALL-FILTRON with a molecular weight cut-off (M_{WCO}) of 10.30 and 50 kDa. The membranes were in a cassette configuration which is composed of multiple layers of polyethersulfone (PES) membranes with polyester separators (screens) placed between them. The membranes have a total permeation area of 0.09 m^2. The initial pure water permeability of 10, 30 and 50 kDa membranes was respectively 174, 534 and 540 $l/m^2.h.bar$.

Table 1. Characteristic of crude twin-screw extruder-pressed sunflower oil

Parameters	Value
Acid value (mg KOH/ g oil)	1.55 - 2.60
Phosphorus content (mg/kg)	16 - 76
Phospholipids content (mg/kg)	486 - 2288
Color (CIE):	
L* (lightness)	79 - 88
a* (green-red color)	-0.8 - (- 0.5)
b* (blue-yellow color)	58 - 77

Experimental

Tangential ultrafiltration was conducted with a diaphragm pump (QUATRO S/N: 1270, PALL-FILTRON) for the PALL membranes under continuous recycling permeate (*Figure* 1). The transmembrane pressure (TMP) was calculated from following relationship:

$$[(P_{feed} + P_{retentate})/2] - P_{permeate}$$

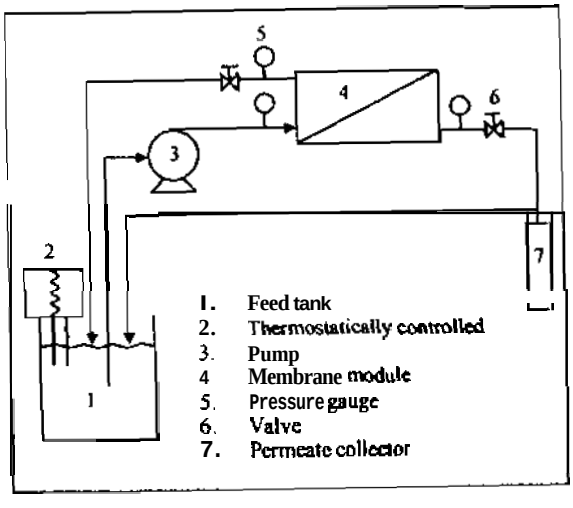

Figure 1. Schematic flow diagram of membrane filtration

The feed tank was charged with 5 liters of the crude sunflower oil and regulated at the desired temperature. Influence of MWCO membrane and operating parameters including temperature, transmembrane pressure and

tangential flow rate on ultrafiltration performance were evaluated by comparing permeate flux, phospholipids rejection, color and free fatty acid reduction. In this study, the membrane retention was calculated from following relationship:

R (%) = $(1 - C_p/C_f) \times 100$ where C_p and C_f are phospholipids concentration in permeate and feed, respectively.

The membranes were conditioned in glycerol for 90 minutes and then soaked overnight in refined sunflower oil to reduce the moisture content of membrane before its introduction into the module. To ensure a stable permeate flow, the tangential ultrafiltration was operated for 60 minutes before processing the actual samples. Upon achieving steady operation, permeate samples were immediately collected over certain period. Permeate flow was measured using graduated cylinder. To maintain the performance of the membrane, the membrane was cleaned using sodium dodecyl sulfate (SDS) detergent after the test and then rinsed with water.

The tangential flow rate of membrane cassette was calculated considering the feed channel to be completely open. This is therefore a rough approximation since we should have to consider the screen in the channel. However, the hydrodynamic description is not yet elucidated and can not be introduced at this time.

Analysis

The phosphorus content was measured using the AOCS method Ca 12-55 and the total phospholipids obtained by multiplying phosphorus content by 30. The free fatty acids content was analyzed using the French normalized method (NF T60 204). The color was determined by CIE method with a spectrocolorimeter (Minolta Seri CM-500i). The color values measured are presented as L*, a* and b* corresponding to lightness, the green-red and the blueyellow components, respectively. Those results are the average of five consecutive measurements.

RESULTS AND DISCUSSION

Variation with time

The ultrafiltration was conducted with membrane of 10 kDa under operating conditions of 1.8 - 3.0 bar, 23°C and 0.026 m/s. As shown in Figure 2, the permeate flux did not decrease with filtration time. No long term fouling was observed. However, the permeate flux was low compare to water permeability of membrane (174 l/m^2.h.bar). The steady state permeate flux is obtained less than 60 minutes, as observed by Nabi et al. (2000), Pagliero et al. (2001) and Hafidi et al. (2005a, 2005b).

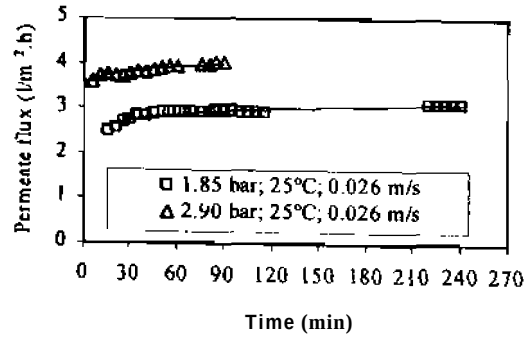

Figure 2. Permeate flux as a function of time for membrane of 10 kDa

Factors affecting permeate flux

The results (Figure 3) indicate that the permeate flux increased with the transmembrane pressure for all membranes tested. There was no compaction of membranes for the transmembrane pressure in the range tested in this study, hence the permeate flux increased rapidly as observed by Lin et al. (1997) and Nabi et al. (2000). At a very low pressure, the permeate flux is low, so the effect of concentration polarization is small and a gel layer does not form on the membrane surface. As the applied pressure is increased, the concentration polarization and the concentration of retained solutes at the membrane surface increase as the flux rises. If the pressure is increased further, concentration polarization becomes enough for the retained solutes at the membrane surface to reach the gel concentration and form the secondary barrier layer. This is the limiting flux for the membrane (Baker, 2000), which was not reached under the operating conditions tested in this study.

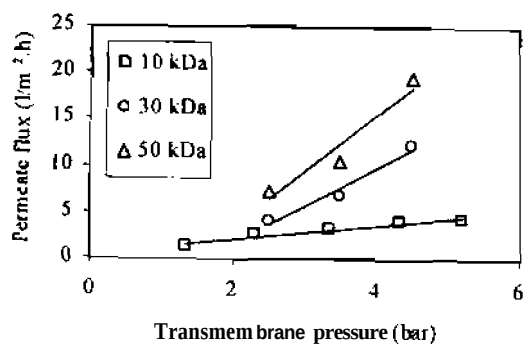

Figure 3. Influence of transmembrane pressure on permeate flux as function of MWCO of membranes (50°C and 0.04 m/s)

The permeate flux of the 30 and 50 kDa membranes was higher than the 10 kDa membrane. The difference in pore size distribution, as well as the membrane-solute interaction, caused the variation in permeate flux. As MWCO of 30 and 50 kDa membranes were higher than 10 kDa membrane, in consequence the triglycerides release easily through both these membranes. In addition, the interaction occurring between the oil components, particularly the phospholipids, and the membranes and the structure formed by oil components at membrane surfaces or within the pores changed physicochemical properties of membrane and caused an increase in membrane resistance. Higher MWCO of membranes should reduce the membrane resistance and thus allows better permeate flux.

Figure 4 shows that an increase in temperature enhanced the permeate flux of 30 and 50 kDa membranes while the permeate flux of 10 kDa membrane remained relatively stable with the temperature. In *the* pressure-controlled region, the influence of temperature *on* permeate flux is due to its effect on fluid density and viscosity (In-Chul Kim et al., 2002). Diffusivity also increases with temperature, e.g. phospholipids diffusivity increases. Increasing the temperature reduces the feed viscosity which increases the convective flow rate and reduces polarization concentration at membrane surface, thus increasing the permeate flux, as described by Pagliero et al. (2001), Subramanian et al. (2001a) and Amalia Kartika et al. (2003c, 2003d, 2004b). According to the Darcy's law, decreasing of permeate viscosity contributes also to the increased permeate flux. In this work, the reduction of the feed viscosity is up to 14% for an increase of 5°C. Fw all these reasons, it is best to operate the ultrafiltration of crude sunflower oils at the highest possible temperature consistent with the limits of the *feed* solution and the membrane, particularly *for* membrane of 30 and 50 kDa.

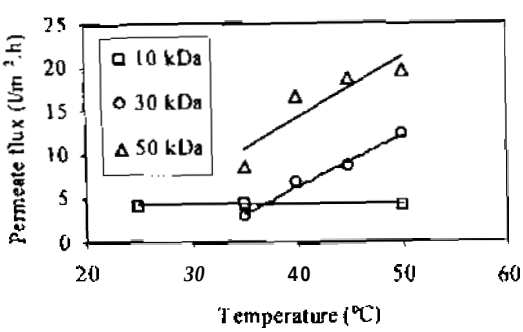

Figure 4. Influence of temperature on permeate flux as function of MWCO of membranes (4.5 bar and 0.04 m/s)

A high permeate flux was obtained when the tangential flow rate increased, as observed in Figure 5. At high tangential flow rate, agitation and mixing of the fluid near the membrane surface sweep away the accumulated solutes and reduce the thickness of the boundary layer *on* the membrane surface (In-Chul Kim et al., 2002). The high tangential flow rate limits therefore the deposition and the formation of a cake layer on the membrane *surface* and allow better permeate flux, as observed on ultrafiltration of crude sunflower oils conducted with tangential flow rate of 0.04 - 0.07 m/s for membrane of 30 and *50* kDa, and *0.026* - 0.033 m/s for membrane of 10 kDa. However, further increases in tangential flow rate did not *enhance* permeate flux. This phenomenon could be attributed to the effect *of* the tangential flaw rate being limited in the close vicinity of the membrane and therefore no longer affecting the concentration of solutes or particles at the oil/membrane interfaces (Pioch et al., 1998). In other words, the concentration polarization layer has already reached a limit value (thickness) at 0.07 m/s for membrane of 30 and *50* kDa and 0,033 m/s for membrane of 10 kDa.

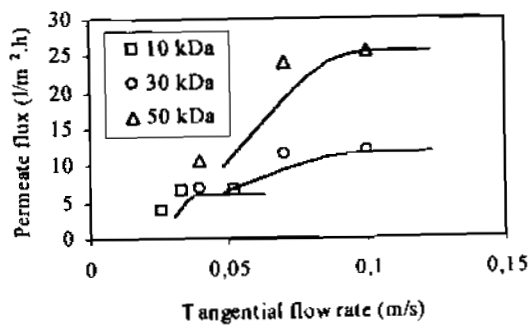

Figure 5. Influence of tangential flow rate on permeate flux as function of MWCO of membranes (3.5 bar and *50°C)*

Factors affecting phospholipid rejection

The selectivity of ultrafiltration mainly depends on membrane type, molecular size of solute and micro-environmental condition as temperature and concentration of solutes (Lin et al., 1998). The average molecular weight of phospholipids (700 Da) is lower than triglycerides (900 Da). However, phospholipids are surfactant with both hydrophobic and hydrophilic groups which cause their preference for self assembly with hydrophilic groups on *one* side and hydrophobic groups *on* the other side into a wide variety of aggregate (micelles, liquid crystals or crystals). *They* tend also to form reverse micelles with imbedded ends inside non-polar media like hexane or neutral oil. The micelles formed can have a molecular weight of

20000 or more in hexane-oil miscella (Iwama, 1987). Hence, phospholipids can be separated from triglycerides wing an ultrafiltration membrane of MWCO value 20000 or less. Studies (Subramanian and Nakajima, 1997; Lin et al., 1997, Hafidi et al., 2005b) reported that the phospholipids micelles were formed when the phospholipids content of the oil was above critical micelle concentration (CMC). The CMC values of phosphatidylcholine (PC) in triolein and phospholipids in crude soybean oil were found to be 440 and 1020 mg/kg, respectively, while the size of the PC micelles was determined to be in the range of 3.56 to 4.70 nm (Subramanian et al., 2001b). On the other hand, when phospholipids exist as monomers (below CMC), the rejection of phospholipids was due to low solubility of phospholipids. Another possibility could be due to swollen in the presence of small quantities of water and having affinity for some of the other impurities such as color compounds, which are then rejected by size exclusion (Subramanian et al., 2001b). Subramanian et al. (1999) reported that the degumming performance in membrane process depends on the total amount of phospholipids. The addition of 4% hydratable phospholipids and 1% water into crude oil containing phosphorus content lower than 400 ppm increased the phospholipids rejection up to 98% as result of enhancing the encapsulating ability of phospholipids.

The effect of operating parameters on phospholipids rejection is presented in Figures 6, 7 and 8. Figure 6 shows that the phospholipids rejection of 30 and 50 kDa membranes decreased, respectively, from 60 to 35% and 30 to 15% when the transmembrane pressure was increased from 2.5 to 4.5 bar while the phospholipids rejection of 10 kDa membrane remained high (> 87%) with the transmembrane pressure. The fouling as a result of the adsorption of the phospholipids at the pore walls, cake built-up at the membrane surface and mechanical pore plugging (Baker, 2000; Hafidi et al., 2003) increased phospholipids rejection of 10 kDa membrane when the transmembrane pressure was increased.

The phospholipids rejection decreased for the membranes of 30 and 50 kDa but remained high (> 85%) for the membrane of 10 kDa when the temperature was increased (Figure 7). Under high temperature, the loss of efficiency in selectivity might come either from the well known tendency of molecules to solubilize instead of aggregating when increasing the temperature or also from a change in the interactions between the oil components and the membrane (Pioch et al., 1998).

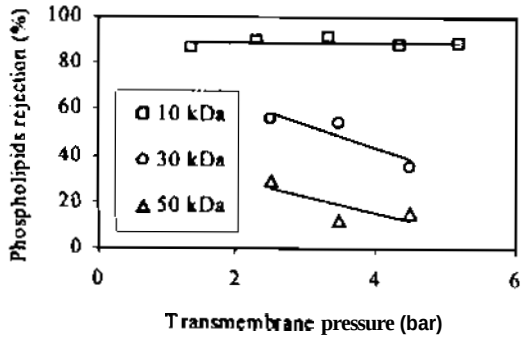

Figure 6. Influence of transmembrane pressure on phospholipids rejection as function of MWCO of membranes (50°C and 0.04 m/s)

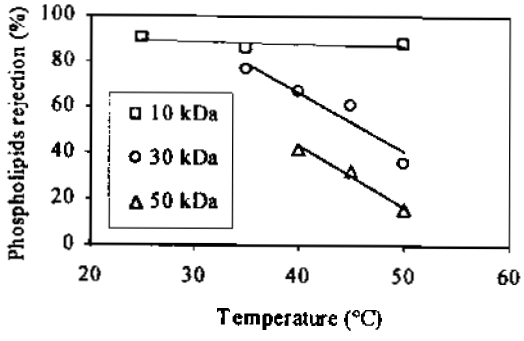

Figure 7. Influence of temperature on phospholipids rejection as function of MWCO of membrane (4.5 bar and 0.04 m/s)

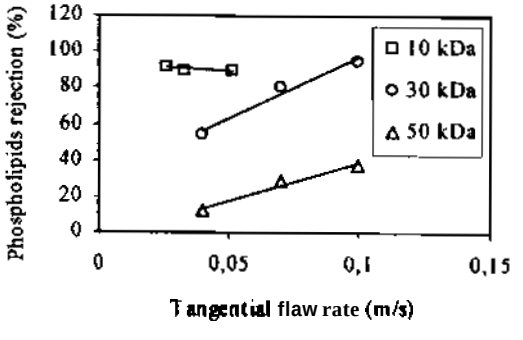

Figure 8. Influence of tangential flow rate on phospholipids rejection as function of MWCO of membranes (3.5 bar and 50°C)

In addition, selectivity of the membrane could be due to a structure formed by the micelle at membrane surface. This structure would appear under specific hydrodynamic conditions (trans-membrane pressure, tangential flow rate), which occur when the temperature is high.

Figure 8 shows that an increase in tangential flow rate enhanced the phospholipids rejection from 50 to 95% for the membrane of 30 kDa and 10 to 40% for the membrane of 50 kDa. The influence of the tangential flow rate for the membrane of 10 kDa was low, therefore the phospholipids rejection remained high (> 88%)

with the tangential flow rate. The high tangential flow rate limits the deposition and the formation of a cake layer on the membrane surface and allows sweeping away the accumulated solutes and reducing the thickness of the boundary layer on the membrane surface.

Free Fatty Acid reductioo

Free fatty acids (FFA) distributions after ultrafiltration are summarized in Table 2. Table 2 shows that membrane process usually could not remove free fatty acids. Similar results have also been reported (Iwama, 1987; Lin et al., 1997; Subramanian et al., 1998b; Ong et al., *1999).*

Transmembrane pressure, temperature and tangential flow rate did not change significantly FFA content. For all trials, the reduction of free fatty acids is lower than 10%. The membrane with smaller molecular weight cut off should be used for crude sunflower oil de-acidification but the flux should be very low.

Table 2. Free fatty acids characteristic of crude and membrane-processed sunflower oils as function of operating conditions and MWCO of membranes

Sample	TMP (bar)	T (°C)	V (m/s)	Acid Value (mg KOH/g oil)		
				10 kDa	30 kDa	50 kDa
Crude oil	/	/	/	1.55	2.53	2.47
Permeate	1.8	25	0.026	1.55	/	/
	2.9	25	0.026	1.55	/	/
	3.9	25	0.026	1.55	/	/
	4.7	25	0.026	1.55	/	/
	1.8	35	0.026	1.55	/	/
	3.0	35	0.026	1.55	/	/
	4.7	35	0.026	1.55	/	/
	5.6	35	0.026	1.55	/	/
	2.3	50	0.033	1.55	/	/
	3.3	50	0.033	1.55	/	/
	4.4	50	0.033	1.55	/	/
	5.2	50	0.033	1.55	/	/
	2.5	50	0.040	/	2.42	7.47
	3.5	50	0.040	/	/	2.47
	4.5	50	0.040	/	2.49	2.45
	2.5	50	0.070	1	2.39	2.45
	3.5	50	0.070	/	2.39	2.44
	4.5	50	0.070	1	2.42	2.44
	2.5	50	0.100	/	2.30	1
	3.5	50	0.100	1	2.27	/
	4.5	35	0.040	/	2.47	2.47
	4.5	40	0.040	1	2.50	2.42
	4.5	45	0.040	/	2.42	2.47

Color

Sunflower oil color is essentially due to carotenoid pigments (yellow color pigment) and a small quantity of chlorophyll (green color pigment) from unripe seed and plant impurities. In this study, the coloring agent in sunflower oil kfore and after membrane ultrafiltration consists of yellow and green color pigments The yellow and green color pigments are indicated by positive values of b and by negative values of a, respectively. For all trials, the values of yellow color pigment increase in permeate, mainly for membrane of 10 and 50 kDa. The use of CIE L*a*b* method for the measurement of color demonstrated excellent repeatability and a significant correlation with the carotenoid content, as reported by Rossi et al. (2001). In addition, this method is simple and rapid and thus can be used as an alternative to the classical Lovibond matching method.

During membrane processing, the values of color characteristic in sunflower oil increase. The oil transmission, which is indicated by positive values of L, increases to 94% thus increasing oil quality. Thesefore, membrane processing may also be used in decolorizing.

CONCLUSION

This study showed that the ultrafiltration membranes were effective in removing phospholipids and increasing color quality of twin-screw extruder-pressed sunflower oil, therefore membrane processing could be used in degumming and decoloration. The best result was given by the *50* kDa membrane with permeate flux of 26 $l/m^2.h$ under operating conditions of 3.5 bar, 50°C and tangential flow rate of 0.1 m/s. However, phospholipids rejection was only 40%. The 30 kDa membrane had 95% phospholipids rejection but permeate flux was only 12 $l/m^2.h$.

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