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Concentration and Desalination of Protein Derived from Tuna Cooking Juice by Nanofiltration

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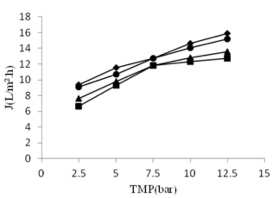
Graphical abstract



Abstract

Tuna cooking juice is a co-product of tuna canning industry. It riches in protein, currently used for production of feed meal as well as protein hydrolysate. The finish products are usually in the form of concentrate, produced by evaporation process. However, evaporation is energy consumable process and the salt content level of the concentrate is often over the standard, thus required additional process for lowering salt content e.g. crystallization. The use of membrane technology, therefore, is of interest, since it required less energy and footprint compared with evaporation and is also able to reduce salt content of the concentrate. The aim of this study were to employ and select the membrane filtration process, and optimize the operating condition for protein concentration and desalination of tuna cooking juice. The results indicated that nanofiltration (NF) was more suitable than the ultrafiltration (UF) process, regarding the ability in protein recovery and desalination. The NF performance was evaluated in terms of permeation flux and protein and salt retentions. The protein and salt rejections of NF were 96 % and 5 %, respectively. The permeate flux(J) increased as transmembrane pressure (TMP) or cross flow rate (CFR) increased and the highest flux was obtained at TMP of 10 bar and CFR of 800 L/h. Operating with batch mode, the permeate flux was found to decrease as protein concentration increased, and at volume concentration factor about 4, the protein concentration about 10% while salt removal was aproximately 70 % of the initial value. This work clearly showed that NF was successfully employed for concentration and desalination of protein derived from tuna cooking juice.

Keywords: Tuna cooking juices; nanofiltration; protein; desalination; fouling

■1.0 INTRODUCTION

Tuna cooking juice is co-product genrerated from canned tuna processing. Its contains high amount of protein ranging from 2.0-5.5% and aproximantely 0.2-1.0 % of salt[1]. Since tuna cooking derived from tuna cooking process, protein is partially hydrolysed by thermal hydrolysis. Nowaday, most of tuna cooking juice is used for producing either feed meal or value added products, protein hydrolysate. It is possible to find for small manufacturers that tuna cooking juice is directly discharged to wastewater treatment system. Traditionally tuna cooking juice is pre-concentrated prior drying process to obtain finish product, a feed meal. Tuna cooking juice can be also hydrolysed by proteinase to obtain protein hydrolysate. This hydrolysate is required to concentrated prior filling process. However, there are two major challenges to utilize tuna cooking juice for tuna industry. Firstly, finish products including feed meal and protein hydrolysate contain high amount of salt affecting animal and human health and consequently limits their applications. Secondly, tuna cooking juice and protein hydrolysate are usually concentrated by evaporation process. However, the evaporation is operated at relatively hig temperature and vacuum and also time consuming process. As a result, it is quite energy consumable process, and may cause environmental pollution. Thus the alternative concentrating and desalting process is urgently needed. The use of pressure driven membrane filtration processes are of interest since, in principle, it offers not only to reduce salt content in the finish products but also is able to concentrate the products. In addition, they have been widely employing due to certain advantages, such as: low temperatures, absence of phase transition and low energy consumption[2]. Among them ultrafiltration(UF) and nanofiltration(NF) are promissing processes that permit to tackle such problems. They have been enployed in various applications, such as protein urification and concentration, solvent recovery from filtered oil, exchange of solvents in the chemical industry[3], concentration and purification of ethanolic extracts of xantophylls[4], desalination of soy sauce[5], concentration of wastewaters from the fish meal industry[6]. However, different types of membranes lead to different process performances especially, in this case, protein and salt rejections. UF is usually used for protein purification, fractionation and concentration while. Its separation abiltiy depends on miolecular weight (MWCO) and feed properties e.g. molecular weigh of protein, pH and ionic strength as well as operating conditions. Its separation mechanism is solely size exclusion or sieve mechanism. NF is employed to separate low molecular weigh species, e.g. salt , surgar, peptides. In addition to the size of the pores, the charge of the feed and membrane play important role in separation ability. The objective of this study was to concentrate and remove salt from tuna cooking juice by membrane filtration. The effect of membrane (type/molecular weight cut of (MWCO)), cross flow rate (CFR) and transmembrane pressure (TMP) on permeates flux, salt and protein rejections were sutudied to determine the optimum condition.

2.0 EXPERIMENTAL

2.1 Materials and Methods

Tuna cooking juice was obtained from Chotiwat Manufacturing Company Limited, Hat Yai, Songkhla, Thailand. The pH, COD, salt and protein contents of pretreated sample were 6.47, 33,427 mg/L, 1.4% and 3.2 %, respectively.

2.2 Characteristic of Membranes

Various types of membranes were used to select the most suitable one and their characteristics are shown in Table 1**.**

Table 1 Membrane characteristics and system

Membrane	Module	Supplier	Material	Nominal solution rejection/MWCO	Active area (m^2)
UFP-1-L-3M	Hollow fiber	GE Healthcare	Polysulphone	1 kDa	0.014
UFP-5-E-3MA	Hollow fiber	GE Healthcare	Polysulphone	5 kDa	0.011
UFP-10-E-3MA	Hollow fiber	GE Healthcare	Polysulphone	10 kDa	0.011
KO 1 B	Tubular	Kerasep	Ceramic	15 kDa	0.245
DL 2540	Spiral wound	Osmonics	Thin film	96 % $MgSO_4$	1.27

2.3 Analysis of Sample

The salt and protein contents of the tuna cooking juice were determined, according to AOAC (1999) and Lowry method (1951), respectively and COD was analysed using Titrimetric Method.

2.4 Experiment

2.4.1 Effect of Membrane (Type/MWCO) on Permeates Flux, Salt and Protein Rejection

The membranes used for this experiment are shown in Table 1. All membrane processes were crossflow systems and operated under total recycle mode in which, the retentate and permeate were recycled to the feed tank. The permeate flux was calculated using the following equation (1);

$$J = \frac{V}{At} \quad (1)$$

where *V*, *A* and *t* are the volume of permeate (L), membrane area (m^2) and time (h), respectively. The percentage of rejection (*R*, %) of protein and salt were calculated using equation (2);

$$R\,(\%) = \left(1 - \frac{Cp}{Cf}\right) \times 100 \quad (2)$$

where C_p and C_f are the concentration of species in the permeate and feed, respectively. The most suitable membrane then was selected regarding their permeate flux, and protein and salt rejections.

2.4.2 Effects of Transmembrane Pressure (TMP) and Crossflow Rate (CFR) on Permeate Flux, Salt and Protein Rejections

The membrane, selected according the result obtained in previous experiment was used to study the effect of CFR and TMP at operating temperature of 40°C on permeate flux, salt and protein rejections. Note that the ranges of TMP and CFV for study were selected according the membrane system as recommended by the manufacturer.

2.4.3 Effect of Cross Flow Rate on Permeate Flux, Salt and Protein Rejections Under Batch Concentration Mode

The membrane, CFR and TMP were selected according the result obtained in the previous experiments to study the permeate flux, protein and salt rejection, the volume concentration factor (VCF) using batch concentration mode. The VCF was determined using the following equation (3);

$$VCF = \frac{V_0}{(V_0 - V_p)} \quad (3)$$

where V_o and V_p are the initial feed volume and permeate volume, respectively.

■3.0 RESULTS AND DISCUSSION

3.1 Effect of Membranes on Salt and Protein Rejections Using NF and UF

Both UF and NF membranes were used to study their ability in recovery protoein as well as desalination. The NF-DL 2540 membrane was found to gave the highest protein and salt rejections compared to other membranes as show in Table 2. These results were as expected since when the pore size of membrane increase, the protein and salt rejection would decrease (Cheryan, 1998). Although the highest salt rejection was obtained using NF- DL2540 membrane but it was considered as the most suitable membrane because of its protein retention.

Table 2 Salt and protein rejection of different membranes

Module membrane	Protein Rejection (%)	Salt Rejection(%)
Hollow fiber (1 kDa)	$93.44^{b}\pm0.28$	$2.23^{b}\pm0.20$
Hollow fiber (5 kDa)	$85.06^{c}\pm0.15$	$0.31^{d}\pm0.004$
Hollow fiber (10 kDa)	$76.24^{d}\pm0.09$	$0.73^{c}\pm0.01$
Tubular (15 kDa)	$68.10^{e}\pm0.08$	$2.16^{b}\pm0.4$
Spiral wound (96 %$MgSO_4$)	$97.42^{a}\pm0.47$	$5.13^{a}\pm0.10$

Same letters in the same colume present no statistical differences according to Duncan's multiple range test at P<0.05

3.2 Effect of TMP and CFR on Permeate Flux

The NF-DL2540 is a pilot scale of NF membrane system with membrane area of 1.77 m^2. The maximum operating temperature suggested by the manufacturer is 40°C. The minum volume required for the system be able to operate is approximately 20 liters. The effect of TMP and CFR on permeate flux for tuna cooking juice under total recycle mode for selected condition are show in Figure 1. Generally, the permeate flux increases with pressure which is so call pressure dependent region and no further increases with pressures as it reaches pressure independent region or mass transfer control region. The effect of concentration polarization and fouling and operating at pressure independent region provides favorable conditions for fouling[7]. The results showed that the permeate flux increased as TMP increased for all CFRs (from 8.71 to 14.3 L/m^2h). It was likely that the permeate flux was still in pressure dependent region. The permeate flux also increased as CFR increased. At higher CFR, the rate of removal of retained material by shear force is high and would reduce reversible fouling and enhance the mass transfer rate that benefits permeate flux[8,9]. Similar result was found during nanofiltration to concentrated of flavonoids and phenolic compounds in aqueous and ethanolic propolis extract and licorice aqueous solution[10,11]. Although the highest flux was obtained at CFR of 800 L/h and TMP of 12 bar, operating condition at CFR of 800 L/h and TMP of 10 bar was selected for studying during batch concentration mode to avoid or reduce the impact of concentration polarization and fouling.

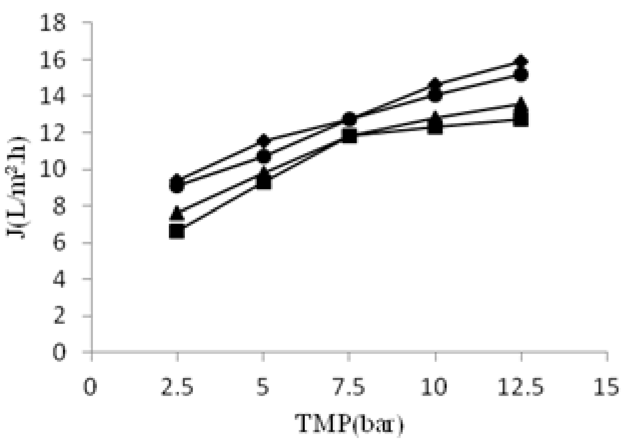


Figure 1 Permeate flux of tuna cooking juice at various CFR and TMP under total recycle mode at temperature of 40 °C ■ 500 L/h ▲600L ●700L/h ◆800L/h

3.3 Effect of TMP and CFR on Salt and Protein Rejection

The effect of pressure on salt and protein rejections is shown in Figure 2. The rejection of salt and protein for all applied TMP and CFR ranged from 4 to 5% and 93 to 97 %, respectively. The results indicated that both salt and protein rejections were almost constant as varying TMP and CF. It was possible that the operating condition employed during this study was still in pressure dependent zone in which the fouling formed did not severe and change the membrane rejection characteristic [11].

3.4 Permeate Flux and VCF Under Batch Concentration Mode

In batch concentration mode using NF-DL2540 membrane, it was expected that most of protein would retain in the concentrate while the amount of salt was remarkably reduced, compared to those found in the original feed. Thus, operating under diafiltration mode did not need to employ. The performance of nanofiltration of tuna cooking juice was evaluated at TMP 10 bar, CFR of 800 L/h and temperature of 40 °C. The permeate flus is usually used to indicate the capacity and be used to calculated membrane area for scaling up the plant. The permeate flux decreased from 12.5 to 6.9 L/m^2h and the VCF increased from 1 to 4 after 180 min of operation (Figure 3). The flux reduction was due to the impact concentration polarization and fouling as well as back transfer rate of the retained molecules[11]. The feed bulk concentration increased as VCF increased resulted in reduction of concentration difference between those at the membrane wall and bulk feed. In addition, in higher concentration fouling tendency components, protein in this case, it was likely to induce severe fouling.

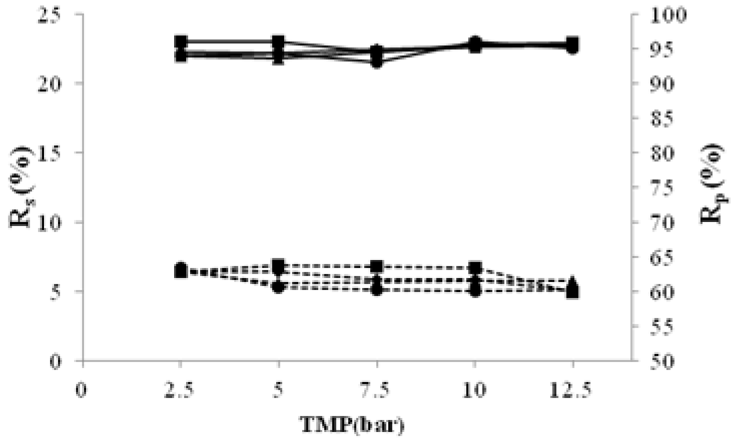


Figure 2 Salt and protein rejections during nanofiltration of tuna cooking juice as varying CFR and TMP under total recycle mode at temperature of 40° C (R_s: salt rejection, R_p: protein rejection). ■ 500 L/h ▲600L ●700L/h ◆800L/h

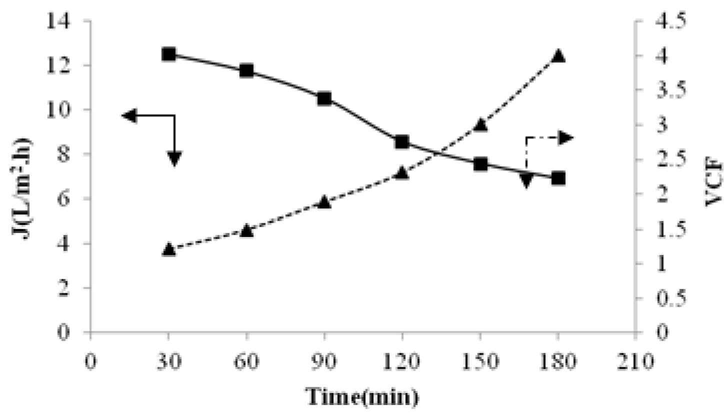


Figure 3 Permeate flux and VCF during nanofiltration of tuna cooking juice under batch concentration mode (CFR 800 L/h, TMP 10 bar and temperature of 40 ◦ C)

3.5 Salt and Protein Rejections Under Batch Concentration Mode

Salt and protein rejections during nanofiltration is of interest since they are also key indicators to indicate the process performace and the quality of the finish product. Salt and protein rejections during nanofiltration of tuna cooking juice at CFR 800 L/h, TMP 10 bar and temperature of 40 °C are shown in Figure 4. The protein rejection significantly increased from 94 to 97 % with operating time as well as VCF. The protein rejection tend to constant when the VCF was higher than 4. The salt rejections were in the ranges of 5.3-5.5. The average of protein and salt rejection through out this experiment were approximately 97 and 5.3 %, respectively.

Traditionally, the plot of permeate flux vs protein concentration can be used to calculate the constants in film theory Equation as shown in Equation 4[12];

$$J = kln\frac{C_G - C_p}{C_b - C_p} \tag{4}$$

where k is the mass transfer coefficient (m/s), C_B the bulk protein concentration of the rejected solutes, C_G the maximum possible solute concentration at which flux becomes zero or the "gel" concentration and C_p is the protein concentration in the permeate. Since the protein rejection for this case was very high (>95%), It was assumed that C_p is negligible). The plot permeate flux vs protein cocnetration for this study is shown in Figure 5. It can be seen that the relationship between permeate flux vs protein cocnetration in semi log plot was linear with reasonable regression coefficienct (0.97). This result suggest that mass transfer coeffiecienct was constant. The mass transfer coefficienct estimated from the slope was 26.02 (m/s) and the gel concentration was 30 %. Thus the empirical Equation to predict the permeate flux for this case can be formulated as follows;

$$J = 1.0 \times 10^{-6}\, ln(\frac{40}{C_b}) \tag{5}$$

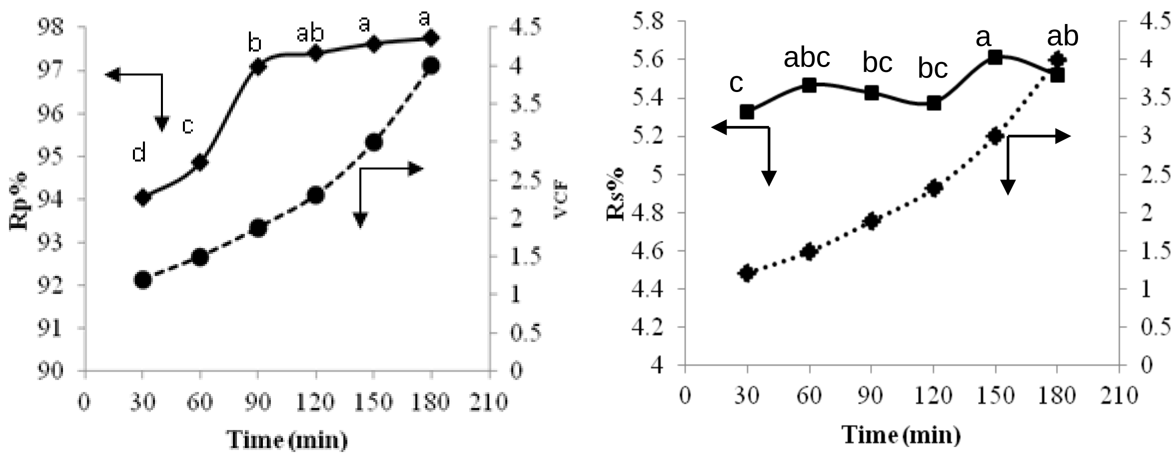


Figure 4 Salt and protein rejections during nanofiltration of tuna cooking juice under batch concentration mode (CFR 800 L/h, TMP 10 bar and temperature of 40 ◦ C)

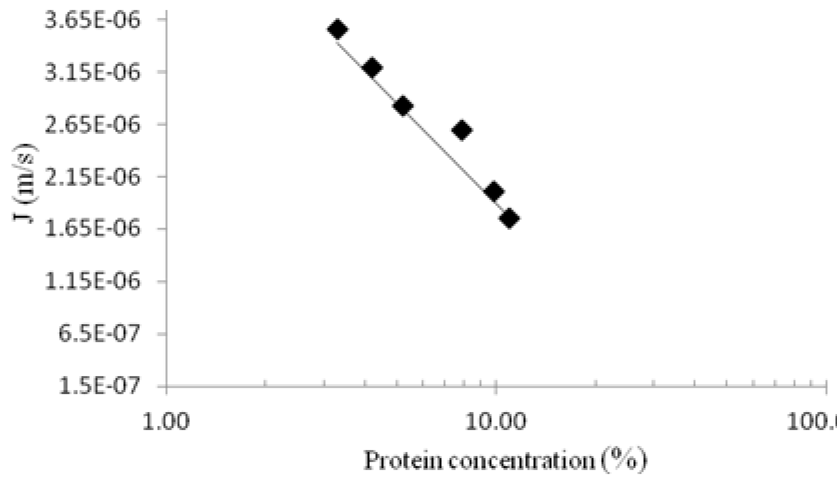


Figure 5 Plot of protein concentration vs permeate flux during nanofiltration of tonu cooking juice under batch concentration mode 800 L/h, TMP 10 bar and temperature of 40 ◦ C)

The salt and protein contents in the permeate and retentate obtained and their rejection at VCF of 4 are presented in Table 3. The cocentration of protein and salt in the concentrate increased upto approximately 11 % while the salts removal was about 70 %. These result indicate that NF could effectively concentrate protein and remove salt from tuna cooking juice. This cocentrate juice is suitable for further processing e.g. enzymatic hydrolysis. Thus the reactor tank volume and energy consumption for this process is expected be reduced. After hydrolysis, protein hydrolysate will be concentrated by evaporation. The protein content of hydrolysate generally is about 3.1-3.5 %. For this case, protein content was about 11 % , thus less water to be evaporate by evaporation is expected.

Table 3 Salt and protein content in the retentate and permeate at the end of expereiment (VCF = 4)

Components	Retentate	Permeate
Protein(%)	10.97	0.26
Salt (%)	1.79	1.64

■4.0 CONCLUSION

Various types of membranes were tested for concentrating and desalting of tuna cooking juice. The NF-DL2540 was found to be the most suitable membrane. Protein and salt rejections of NF-DL2540 were approximately 97 and 5.0 %, respectively. The NF could be effectively employed to concentrate and desalting tuna cooking juice under batch concentration mode at CFR 800 L/h, TMP 10 bar and temperature of 40 °C. The constants in film theory equation, mass transfer coefficient and C_G were also calculated to obtain an empirical equation. At VCF of 4 , protein concentration of tuna cooking juice was increased from approximately 2 to 11% while more than 70 % the salt in the cooking juice was removed.

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