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## Synthesis and Performance Evaluation of ELVALOY® 4170 Polymeric Membrane for Separation and Concentration of Omega-3 Fatty Acids

Samaneh Ghasemian<sup>1</sup>, Reza Abedini<sup>2\*</sup>

- 1- Ph.D. of Food Technology, Department of Food Science and Technology, Tarbiat Modares University, Tehran, Iran
  - 2- Assistant Professor, Faculty of Chemical Engineering, Babol Noshirvani University of Technology, Babol, Iran
- \* Corresponding author (abedini@nit.ac.ir)

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### Abstract

Omega-3 is an acid that exists in the structure of some fats and has a vital role in the human health. Thus, the widespread research has been done to concentrate and purify the omega-3 from fish oil. In this research, the performance of membrane process in long chain omega-3 fatty acid concentration of Lantern fish oil which contains 25.23 wt.% omega-3, was investigated. To synthesize the membrane, the ELVALOY®4170 was used and the membrane was prepared through phase inversion method. The morphology of prepared membrane was analyzed using scanning electron microscopy. In concentration process, the effects of three parameters including temperature, pressure and mixing rate were studied through Box-Behnken statistical method. Scanning electron microscopy images showed that the prepared membrane consists of porous structure with thin dense toplayer. The obtained results from concentration process studied through ANOVA method and indicated that between evaluated parameters, temperature had the highest impact on concentration process. Moreover, the fouling behavior of membranes at different mixing rate was studied and outcomes revealed that the lowest fouling occurred at 100 rpm. Among various concentration process parameters, the highest value omega-3 concentration of 37.32% was obtained at temperature of 40 °C, pressure of 5 bar and mixing rate of zero.

**Keywords:** Fouling, Membrane, Omega-3, Pressure, Temperature

### Introduction

$\omega$ 3-fatty acid concentration is increasing interest for both the pharmaceutical and health food industries (Rodriguez *et al.*, 2010; Monroy *et al.*, 2003). As fish oils are a major source of eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA), an adequate intake of sea foods provides levels of  $\omega$ 3-PUFA required for good health. Therefore, research has been focused on concentrating  $\omega$ 3-PUFA to an edible oil form containing the lowest levels of saturated and monounsaturated fatty acids (Ruxton *et al.*, 2005). Thus, a suitable commercial method to concentrate  $\omega$ 3-PUFA as efficiently as possible is desired by the edible oil industry. Vacuum distillation, hexane extraction, urea crystallization, and conventional crystallization

are currently the most common and widely used methods employed for extraction, fractionation and purification of  $\omega$ 3-PUFA (Chen *et al.*, 2007). The use of these conventional methods is limited due to disadvantages such as the use of toxic solvents, high energy consumption and thermal decomposition of labile compounds (Letisse *et al.*, 2006). Consequently, a new method for  $\omega$ 3-PUFA concentration which provides the same or higher level of efficiency but with fewer disadvantages is desirable. Membrane separation has been widely used in different applications including gas separation, desalination, pharmaceuticals and food manufacturing due to its beneficial aspects such as relatively low operating temperatures, requiring less energy and lower investment costs (James *et al.*, 2003; Abedini *et al.*, 2012; Mehrparvar & Rahimpour, 2015). Membrane separation processes are broadly used in the oil and fat industries. Performance of membranes was evaluated in numerous studies including oil deacidification and degumming, color reduction and solvent recovery (Azmi *et al.*, 2015; Stoft *et al.*, 2015). Polymeric membranes are used widely in various membrane separation applications due to ease of fabrication and economic issues (Abedini *et al.*, 2011).

The purpose of this study is to prepare the ELVALOY membranes to concentrate  $\omega$ 3-PUFA of lantern fish oil. The phase inversion method was employed to prepare ELVALOY membranes using a new dry/wet technique. The membrane was analyzed by SEM to evaluate its separation capability under different pressures and temperatures. The performance of prepared in  $\omega$ 3-PUFA rejection was investigated and finally, mechanisms of membrane fouling due to oil filtration were evaluated and discussed.

### Material and methods

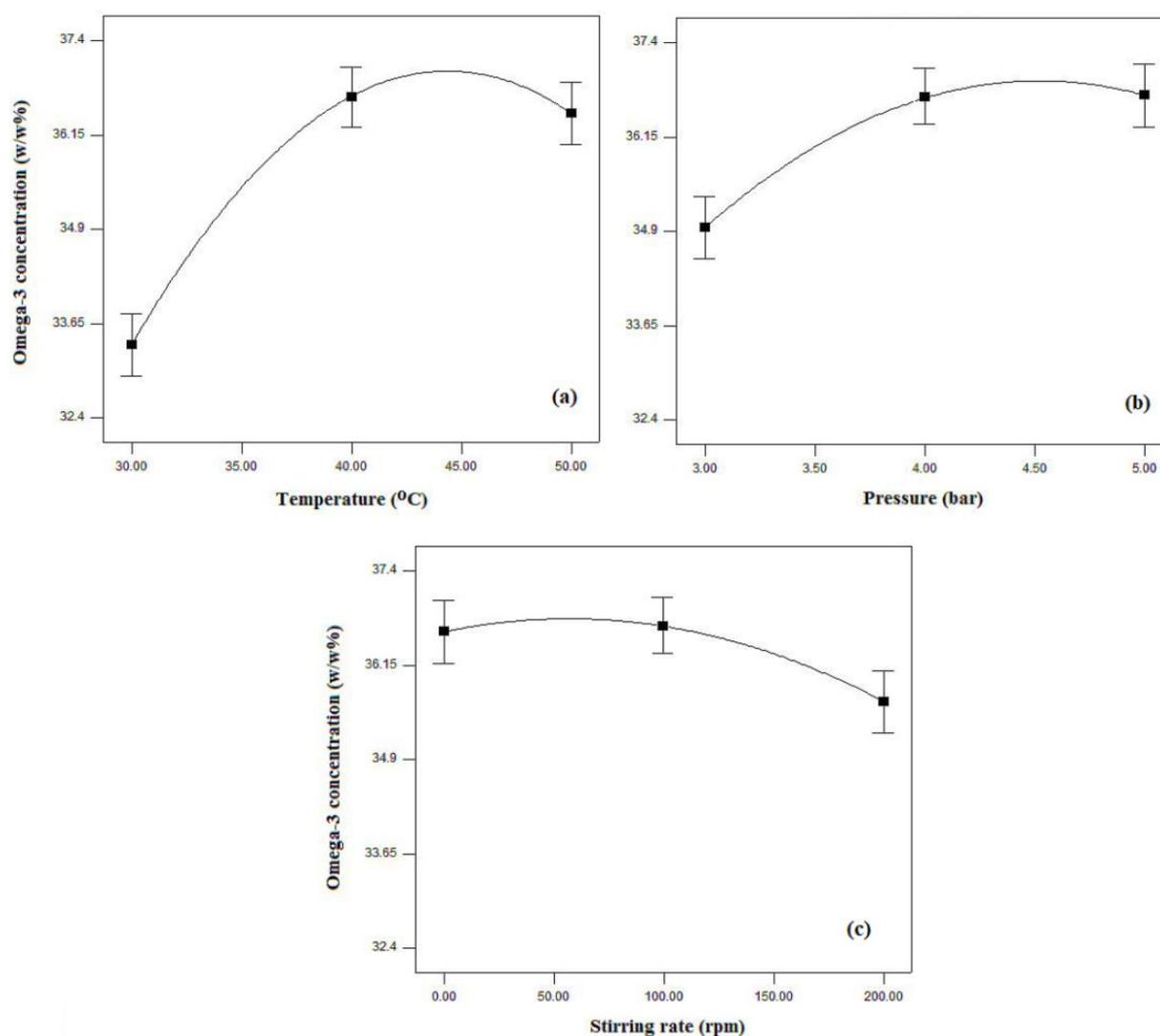
Lantern fish oil was purchased from Qeshm Fish Process Company of South Iran. To prepare fatty acid methyl esters, a Metcalfe method was used. The ELVALOY with density of 0.94 g/cm<sup>3</sup> was purchased from DuPoint company. Asymmetric ELVALOY membranes were prepared via the dry/wet technique along with the phase inversion method. First, a specific amount of ELVALOY was dissolved in toluene. The prepared homogenous solution was cast to a thickness of 300  $\mu$ m on a glass plate substrate. The casted film was placed in a 60 °C vacuum oven for 2 min to allow solvent evaporation. In the next step, for phase inversion, the casted film was moved to a de-ionized water coagulation bath. Finally, the formatted membrane was placed in a vacuum oven to dry the membrane.

### Results and discussion

Figure (1a) shows the effect of temperature on the concentration of  $\omega$ 3-PUFA. From 30 to 40 °C the concentration of  $\omega$ 3-PUFA increased with increasing temperature. However, increasing the temperature beyond 40 °C led a reduction of  $\omega$ 3-PUFA concentration. At 30 °C, which is closer to the melting point of the oil, there were some solid particles suspended in the oil phase. Triglycerides containing higher levels of SFAs in their structure have higher melting points than those with only  $\omega$ 3-PUFA and because of the saturates in their structure, the obtained retentate phase at 30 °C is lower in  $\omega$ 3-PUFA concentration (Hibino *et al.*, 1995). At elevated temperatures, I) the oil is homogeneous liquid with no suspended solids; and II) the mobility of the oil increases leading to improved transport of the linear SFAs through the membrane. Thus more concentrated  $\omega$ 3-PUFA can be attained in the retentate phase.

Figure (1b) illustrates the variation of  $\omega$ 3-PUFA concentration versus the feed pressure. It was observed that increasing pressure from 3 to 4 bar leads to a rise in  $\omega$ 3-PUFA. Increasing the pressure to 5 bar resulted in trade-off variation in  $\omega$ 3-PUFA concentration. At pressures ranging from 4 to 5 bar, the  $\omega$ 3-PUFA concentration initially increased and then decreased. The variation in  $\omega$ 3-PUFA concentration can be attributed primarily to the unique geometry of each fatty acid structure.  $\omega$ 3-PUFA consists of EPA and DHA, with 5 and 6 cis-double

carbon bonds (C=C) in, respectively (Shahidi, 2005). The presence of these cis-double C=C bonds causes kinks in the arrangement of carbon atoms of EPA and DHA and consequently leads to a 30-40 °C kink. The acyl chains cannot align completely along their length, resulting in and steric hindrance. This kinking increases as the numbers of double bonds increase (Juang *et al.*, 2008). Therefore, triglycerides containing higher level of EPA and DHA in their structure can be rejected at higher pressure due to space prohibition. Increasing pressure enables SFAs to be transported through the ELVALOY membranes with superior permeation. Thus, the concentration of  $\omega$ 3-PUFA increases in retentate phase with the pressures up to 4 bar. Higher trans-membrane pressure results in increasing permeation of both  $\omega$ 3-PUFA and SFA. Figure (1c) illustrates the effect of stirring rate on  $\omega$ 3-PUFA concentration. The concentration of  $\omega$ 3-PUFA at no stirring was 36.41 wt. % and increased to 36.64 wt. % at stirring of 100 rpm. Increasing the stirring rate up to 200 rpm leads to the concentration decline of  $\omega$ 3-PUFA to 35.74 wt.%. The variation in  $\omega$ 3-PUFA concentration due to change in stirring rate can be related to the fouling factor.



**Figure 1.** The effect of (a) temperature, (b) pressure and (c) stirring rate on  $\omega$ 3-PUFA concentration at constant pressure and stirring

As shown in Table (1), the fouling resistance and fouling factor for tests conducted at rate of 100 rpm were lower than that for the cases of no stirring and 200 rpm. Therefore, the  $\omega$ 3-PUFA concentration at 100 rpm was superior to the two another cases. Stirring at 100 rpm

compared to no stirring can eliminate or decrease the agglomeration of oil components at the membrane surface which debilitate the separation process. Thus the  $R_f$  and %F decreased to and  $\omega$ 3-PUFA concentration increased at stirring rate of 100 rpm. Although, stirring enhances the  $\omega$ 3-PUFA concentration, but increasing the stirring rate to 200 rpm resulted in obvious decline of  $\omega$ 3-PUFA concentration compared to stirring rate of 100 rpm and no stirring conditions.

**Table 1.** The fouling resistance and fouling factor under each stirring rate

Stirring rate	$R_f$	%F
No stirring	$1.6 \times 10^7$	27.2
100 rpm	$1.1 \times 10^7$	15.3
200 rpm	$1.9 \times 10^7$	35.1

The different  $R_f$  values of each membrane are a result of the different fouling mechanisms. Table (2) lists the values of the root-mean-square deviations (RMSD) between calculated and experimental data for each proposed fouling mechanism. Table (2) shows the differences in the predominant fouling mechanisms occurring in each membrane. Except for cake formation, which has the lowest contribution to membrane fouling, the RMSD values given in Table (2) indicate that other fouling mechanisms may have accrued during  $\omega$ 3-PUFA concentration.

**Table 2.** Root-mean-square deviation (RMSD) between calculated and experimental flux data for each fouling model

Stirring rate (rpm)	Fouling model			
	Cake formation (n=0)	Intermediate pore blocking (n=1)	Internal pore blocking (n=1.5)	Complete pore blocking (n=2)
0	1.87	0.78	0.89	0.21
100	2.38	0.43	0.74	0.56
200	1.26	0.68	0.85	0.14

## Conclusion

An asymmetric ELVALOY membrane with dense thin selective layers were prepared using phase inversion. The  $\omega$ 3-PUFA concentration performance of membrane was evaluated by filtering lantern fish oil under different temperature and pressure conditions. Increasing the temperature from 30 °C to 50 °C led to enhancement of  $\omega$ 3-PUFA concentration. Increasing pressure from 3 to 5 bar had the same effect on  $\omega$ 3-PUFA concentration. Fouling analysis of the membranes revealed that different mechanisms effect the oil flux variation.

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