IOP Conf. Series: Materials Science and Engineering **162** (2017) 012035 doi:10.1088/1757-899X/162/1/012035

Separating xylose from glucose using spiral wound nanofiltration membrane: Effect of cross-flow parameters on sugar rejection

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Abstract. A solution model consisted of two different monosaccharides namely xylose and glucose were separated using a pilot scale spiral wound cross-flow system. This system was equipped by a commercial spiral wound nanofiltration (NF) membrane, Desal-5 DK, having a molecular weight cut off (MWCO) of 150-300 g mol^{-1}. The aim of this present work is to investigate the effect of the cross-flow parameters: the trans-membrane pressure (TMP) and the feed concentration (C_0) on the xylose separation from glucose. The filtration experiments were carried out in total reflux mode with different feed concentration of 2, 5, and 10 g/L at different TMP of 5,8 and 10 bar. The performances of the NF membrane were evaluated by measuring the permeate flux and sugar rejection for each experiment. All the samples were quantified using a high performance liquid chromatography equipped by a fractive index detector. The experimental results indicated an increase in pressure from 5 to 10 bar which was a notable increase to the permeate fluxes from 2.66 x 10^{-3} to 4.14 x 10^{-3}L m^{-2}s^{-1}. Meanwhile, an increase in the C_0 increases the xylose rejection. At TMP of 10 bar and C_0 of 5 g/L, the observed xylose rejection and glucose rejection were measured at 67.19% and 91.82%, respectively. The lower rejection in xylose than glucose suggested that larger glucose molecule were not able to easily pass through the membrane compared to the smaller xylose molecule. The results of this phenomena proved that NF with spiral wound configuration has the potential to separate xylose from glucose, which is valuable to the purification of xylose in xylose production as an alternative to chromatographic processes.

1. Introduction

Chromatography separation method becomes vital in the field of biorefining industry. The separation process involved in this industry was included various sugar separation for example glucose-fructose separation, desugarisation of molasses, and xylose recovery from spent sulphite liquor for production of xylitol. Xylitol becomes a sweetener alternative in the food industry that has equal sweetness as sucrose [1]. Xylose mainly comes from hydrolysis of hemicellulose of agriculture waste, which consists around 55 % of total sugar. Another monosaccharide of interest, which is glucose, also results from the hydrolysis of hemicellulose covering around 25% of the total sugar [2]. Form previous study, the estimated cost of production of xylitol using chemical process and cost of hydrogenation of xylose to xylitol was about $ 350/ton (RM 754/ton) is less than 20% of the total cost of xylitol production. More than 80% of the total cost of xylitol production came from the cost of xylose crystal production which is about $ 2,300 - 2,500/ton (RM 7541 - 8,197/ton). There are few reasons for the high cost of xylose crystal production [3].

Thus, chromatographic separation would be the most suitable technique in sugar biorefinery [4]. However, the performance cost of this technique is relatively higher than that of other separation methods, and selecting an appropriate stationary phase is usually difficult [5] . Among various method of separation currently in study, nanofiltration (NF) offers cost-effective and easy-maintenance alternative separation of xylose from glucose [6,7]. Nanofiltration (NF) membrane separation process is a filtration process employing size exclusion membranes with pore size of 1-10 nm. NF membrane has advantage of low operation pressure, and high permeates flux. Nanofiltration membrane is capable of separation of charged and uncharged substance [8]. Sjoman et al. (2007) observed rejections of xylose were from 0 to 80% and glucose rejections were from 10 to 90% by using NF membrane. The observed rejection was highly dependent on the permeate flux of the membrane affected by pressure. Higher pressure will result higher permeate flux, hence the high rejection of monosaccharide and also few separation parameters that have an effect on nanofiltration beside pressure such as temperature, and total solution composition and concentration have been studied [9].

In the present study, the separation of xylose form glucose was conducted in a pilot scale NF system with Desal-5 DK spiral wound membrane. Experimental variables such as xylose-glucose concentration and transmembrane pressure were studied. In addition, streaming potential of the membrane was measured at various sugar concentration to provide evidence of sugar rejection on separation performance.

2. Materials and Methods

2.1 Model solutions

D-(+)-Xylose (>99%) and D-(+)-Glucose (>99.5%) were purchased from Sigma-Aldrich, Inc. The xylose to glucose concentrations was kept at the ratio 1:1. The feed concentrations were 2,5 and 10 g/L. The transmembrane pressure was tested at 5,8 and 10 bar.

2.2 Membrane and module

Table 1 shows the properties of the DK membrane used in this study. A Desal-5 DK NF membrane module was purchased from GE Water & Process Technologies, USA. This membrane is a three-layer thin film composite membrane with a polyamide top layer and a polysulfone support layer. The molecular weight cut-off of the membrane is 150-300 Da. Since the molecular weight of xylose and glucose are ~ 150 - 180 g mol^{-1} respectively [6], it was expected that concentration of xylose could be achieved by this membrane.

Table 1. Properties of Desal-5 DK membrane

Parameter	Characteristics
Manufacturer	GE Osmonics
Support material	Polysulfone
Surface material	Polyamide
Filtration area [m^2]	1.6
Average pore diameter [nm]	0.42 [10],~1 [11]
Maximum Temperature [^{0}C]	50 [12], [13]
Maximum Pressure [bar]	4.6-26.6 [11]
Molecular weight cut-off [g/mol]	150-300 [6], 200 [13]

2.3 Filtration experiment

The Desal-5 DK membrane was flushed with demineralized water at pressure of 5 bar for 30 minutes. Then the pressure was gradually raised to 10 bar and the membrane compaction was done at high pressure for 1 h and 30 minute. Next, the pure water flux of the virgin membranes was monitored every 15 minutes in 90 minutes until the volume of water was constant. This test was done to check the stability of the membrane flow. Before each filtration the pure water flux was measured at 35 ^{0}C to control the cleanliness of the membranes. Pure water flux (J_w) was calculated by using Eq. (1) [14]. The sugar filtration was done in total recycled mode, i.e. permeate and retentate were circulated back to the feed tank to avoid any concentration change in the feed solution. The filtrations pressure used were 5,8 and10 bar. The temperature of the feed solution was monitored and maintained below 35°C to prevent damage to the membrane. The experiment variable was varies into two variables which transmembrane pressure (TMP) and feed concentration (C_0) to determine sugar rejection.

$$J_p = \frac{V}{A \times t} \tag{1}$$

Eq. (1) showed the calculation for permeate flux where V is permeate volume as a function of time, t in hour and A is the area of commercial membrane.

2.4 Chemical analyses

Samples were analyzed by high performance liquid chromatography (HPLC) to determine the amount of xylose and glucose. All samples were filtered through a 0.22-μm filter first. The chromatography system (Agilent 1200 LC) used in this study consisted of a pump with degasser, auto-sampler, chromatography column, column thermal controller and refractive index (RI) detector. The chromatography analysis was performed on a Rezex RHM-monosaccharide H^+, 300 x 7.8 mm column at 85 °C and water was used as mobile phase at flow rate of 0.6 mL min^{-1}. For each analysis, 20 μL of sample solution was injected into the system. The observed retention (R_i^{ob}) was calculated from Eq. (2), where C_f and C_p are the concentration of feed sugar in the tank and permeate, respectively. During the experiments, samples from the tank and permeate stream were taken and concentrations were determined.

$$R_i^{ob} = \left(1 - \frac{C_p}{C_f}\right) \times 100\% \tag{2}$$

Alternatively, the separation factor, X_{xyl} can be used to evaluate the performance of the membrane. In Eq. (3) where C_p (xyl) and C_p (glu) are the concentrations of xylose and glucose in the permeate and C_f (xyl) and C_f (glu) are the concentrations of xylose and glucose in the feed.

$$X_{xyl} = \frac{\left(C_p(xyl) / C_p(glu) \right)}{\left(C_f(xyl) / C_f(glu) \right)} = \frac{1 - R_{xyl}}{1 - R_{glu}} \tag{3}$$

3. Result and Discussion

3.1 Effect of transmembrane pressure (TMP)

The concentration used in this study was 2g/L, 5g/L and 10g/L. The flux for each concentration was increased as the pressure increased. Increasing concentration of xylose in the feed solution with time, increases the concentration polarization of these molecules on the NF membrane surface causing an increase in the osmotic pressure and a corresponding decrease in transmembrane pressure difference across the barrier. This results in a reduction in flux [16]. As shown in Figure 1, the permeation flux at 2g/L, 5g/L and 10 g/L was quite similar for each TMP. The effect of high permeation flux does not affect the sugar rejection and also the separation factor of xylose to glucose.

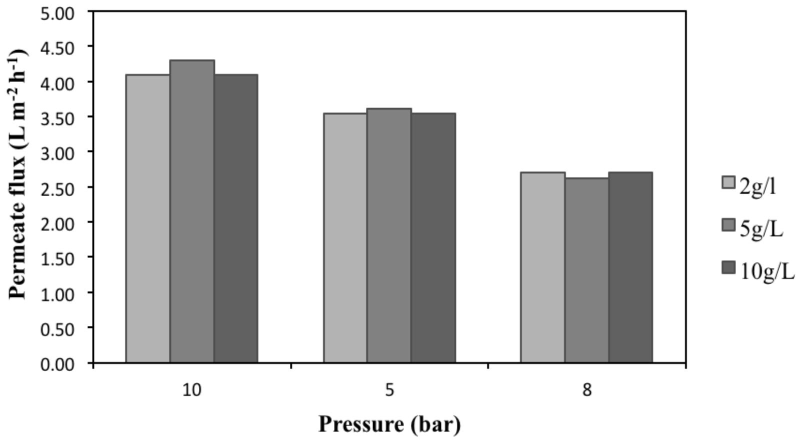

Figure 1. Permeate flux,(L/m^2h) ,J_w vs Pressure (bar)

3.2 Effect of feed concentration and TMP on xylose and glucose retentions

Separation factor is used to determine the performance of the membrane based on sugar rejection. Theoretically, the glucose will retain on the membrane while xylose passes through, according to the properties of xylose-glucose shown in Table 2. The high separation factor with average at 3.25 with 52.57 % xylose retention and and 85.64 % glucose retention was achieved when the feed concentration at 5g/L and at low pressure (5 bar) as shown in Figure 2. However, the results indicate that at 10 bar, the xylose retention was the highest which is 67.19 % but the xylose separation factor for 10g/L and 5g/L was declined. Previous study has been reported that the xylose separation factor will slightly decreased when the pressure increased. The possible causes were membrane compression and concentration polarization occurs at higher applied pressure with concentrated solution [10]. The result in this study was in contradicted to [6], which is the higher separation factor of xylose achieve at higher feed concentration. However, at total feed concentration 2g/L the separation factor was increased when the pressure increases. This might be because of at low concentration with high pressure, less resistance for smaller molecule concentration to pass through the membrane.

Table 2. Properties of Xylose and Glucose [15,10]

Properties	Xylose	Glucose
Molar mass (g/mol)	150.13	180.16
Stokes diameter (nm)	0.64	0.73
Equivalent molar radius (nm)	0.34	0.36

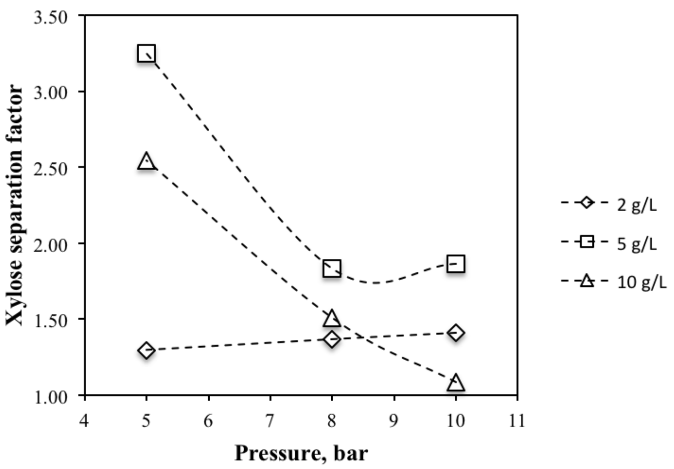

Figure 2. Separation factor xylose to glucose vs pressure at 5,8 and 10 bar

4. Conclusion

Based on the commercial process, sugar was usually separated by chromatography. In this paper, the separation of glucose and xylose was studied by using commercial nanofiltration membranes. Xylose, a pentose sugar, and glucose, a hexose sugar, were chosen as model compounds because xylose purification is an important step in commercial xylitol production. According to the results, nanofiltration has possibilities to enhance the yield and partially replace chromatographic methods in xylose production. There were several factors that will affect the separation of the performance of nanofiltration membranes like pressure, and concentration of feed that will lead to the changes in membrane structure, like compaction and swelling. The performance of Desal-5 DK membranes show that the effects of permeate flux in this study did not led to better xylose separation factor. The high separation factor with average at 3.25 achieved when the feed concentration at 5g/L and at low pressure (5 bar). The decreasing of xylose separation factor from this study indicates that concentration polarization had built a high resistance barrier restricting xylose from passing through the membrane. However, according to the results, nanofiltration showed a promising potential for xylose recovery.

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