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Separation of lactose from raw goat's milk by cross-flow hollow fiber ultrafiltration membrane

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Abstract

An extensive amount of research has reported on the use of ultrafiltration (UF) membrane, particularly in the improvement of membrane performance efficiency on cow's milk. However, a very limited number of researches reported on using UF for producing low-lactose goat's milk due to inherently low lactose. Nonetheless, goat's milk is still not suitable to be consumed in a large amount by people who are lactose intolerant, especially among Asians, where over 90% of the populations are suffering from lactose intolerance. Until today, fouling and concentration polarization (CP) on membrane surface in cross-flow hollow fiber UF unit are the major problems in the dairy industry. Discovery on how to overcome the problem is still in a hot debate due to the nature's complex composition in milk. One way to overcome this problem is by evaluating the effects of processing parameters such as trans-membrane pressure (TMP) and feed-flow rate on flux (J), lactose rejection (Ri), concentration factor (CF), and accumulation rate (AR) during the fractionation of lactose. In terms of lactose fractionation for 5 KDa and 10 KDa UF membranes, the TMPs examined were 0.41, 0.55, and 0.69 bars, while feed flow-rates examined were 0.18, 0.34, 0.54, and 0.74 L/min. 5 KDa membrane shows that feed flow-rate and flux behave in a direct relationship, while an inverse relationship in 10 KDa membrane. Both membranes showed that TMP 0.55 bar exhibit the best flux value without reaching the limiting flux region, but with feed flow rate of 0.74 L/min in 5 KDa, while 0.18 L/min in 10 KDa membrane. Lactose rejection percentage (%Ri) is the lowest with 77.71% in 5 KDa membrane while 66.28% in 10 KDa membrane. This can be summarized that the best parameters for 5 KDa membrane is at TMP 0.55 bar with feed flow-rate of 0.74 L/min, while for 10 KDa membrane is at TMP 0.55 bar with feed flow-rate of 0.18 L/min. Due to higher flux value and lowest lactose rejection obtained from low feed flow-rate, 10 KDa UF membrane size was chosen over 5 KDa. As a conclusion, a high degree of lactose removal from goat's milk could be achieved by 10 KDa UF membrane in a cross-flow hollow fiber system, which proved that different outcomes between 5 KDa and 10 KDa membranes and feed flow-rate required is closely associated to UF pore size and molecular weight of feed solute particles.

Introduction

Mahmood and Usman (2010) reported that goat's milk contained slightly lower lactose, which was 4.39 ± 0.34%, compared to 4.51 ± 0.38% of lactose in cow's milk. This is supported by most previous studies, provided that goat's milk was found to have 4.08% of lactose, which is lower in comparison to 4.78% of lactose content in cow's milk (Posati and Orr, 1976; International Dairy Federation, 1991; Saini and Gill, 1991).

Lehr and Chang (2010) reported that the average daily intake of milk in Malaysia was far less than in Scandinavia as a majority of Malaysians were found sensitive to lactose and this sensitivity is called 'lactose intolerance'. Moreover, Asmawi (2006) also proved that over 80% of Malaysians suffered from lactose intolerance. Thus, as reported by Pouliot (2008) and Robinson and Tamime (1991), since in the early 1970s, a huge idea to separate milk components at the molecular level via separation technique to better utilize each component have been implemented in the dairy industry.

At present, the membrane technology has become widely used because it does not require phase change in dewatering, unlike condensation and evaporation, hence an energy saving process, a non-thermal technique, higher separation efficiency, and

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organoleptic characteristics of the milk product may be retained (Humphrey and Siebert, 1992).

Ultrafiltration (UF) is a medium pressure-driven membrane filtration process that emerged in the 1970s, which can separate components on a molecular basis. In UF, water, minerals, sugars (lactose), urea, amino acids, organic acids, and vitamins pass through the membrane (Nielsen, 2000; Shakeel-Ur-Rehman, 2009), while retaining 20-40% of non-protein nitrogen compounds, up to 10% lactose and minerals, and other ions attached to proteins (calcium, magnesium, phosphate, and citrate). UF was mainly used for producing low lactose dairy products from cow's milk (Kosikowski, 1979).

Generally, the membrane technique method possesses two possible phenomena that can affect the accuracy or the operation effectiveness, which are concentration polarization (CP) and fouling at the membrane surface (Edelsten *et al*., 1983; Patel *et al*., 1991; Castro and Gerla, 2005; Ochando-Pulido *et al*., 2015; Norafifah *et al*., 2015). Membrane fouling is the accumulation of soil, or foulant, on the surface or within the pores of a membrane. Fouling prolongs processing times, increases energy and cleaning costs, decreases separation efficiency, and may lead to irreversible clogging of the membrane (Choi, 2003; Brans *et al*., 2004). The severity of fouling may be controlled and reduced, but it is impossible to be completely vanished (Howell and Finnigan, 1991). Meanwhile, CP is the accumulation of excess particles in a thin layer adjacent to the membrane surface and is inherent of all membrane filtration processes. It may increase resistance to solvent flow, and thus, reduce the permeate flux (Song and Elimelech, 1995).

The scopes of work in this research had been limited to determine the performances of two sizes of MWCO 5 KDa and 10 KDa UF membranes in a cross-flow filtration unit by means of lactose concentration, permeate flux (J), and lactose rejection percentage (%Ri), and accordingly, to select the best processing parameters examined, which were TMP and feed flow-rate.

Materials and Methods

Membrane materials

Hollow fiber cartridges 5 KDa and 10 KDa (Xampler Cartridge, GE Healthcare Bio-Science, Westborough, USA) with polysulfone (PS) materials and surface area of 140 cm^2 were used for lactose fractionation process and being compared in terms of permeate flux and lactose rejection at varying parameters.

Preparation of raw goat's milk

Raw goat's milk was collected from Taman Dagang in Ampang, where the supplier obtained the source from their own farm in Pahang. The raw goat's milk was cooled down to a holding temperature that ranged between 2 and 5°C immediately after milking. The milk was then directly packed in 500 ml bottles with an-airtight sealed container, and stored in a freeze (Lassele LRF-1382PC, 1014 Litre, Kozyair, Australia) temperature; -18°C, The milk was then thawed by water cooling and was kept still until it reached the room temperature; 27°C, prior to using.

Goat's milk quality analysis using MilkoScan FT2

Firstly, the physiochemical constituents of raw goat's milk samples were analyzed using MilkoScan (FT2, Foss Electric, Denmark) based on AOAC, 2000 methods. The analysis was performed at the Department of Veterinary Services in Malacca. At least 50 ml of the milk sample was prepared for the analysis to be experimented approximately 3 minutes. The digital indicator (IED display) showed the specified results.

Experimental setups for lactose fractionation

Next step is lactose fractionation process which aims to remove lactose from raw goat's milk by fractionate it using two different UF membrane sizes 5 KDa and 10 KDa. Figure 1 shows the laboratory scale of cross-flow hollow fiber unit (Quixstand-Benchtop system, GE Healthcare Bioscience, USA) equipped with a peristaltic pump (Quixstand-Benchtop system, GE Healthcare Bioscience, USA) used for lactose fractionation. The desired yield from the filtration was in the retentate stream, which was the concentrated milk.

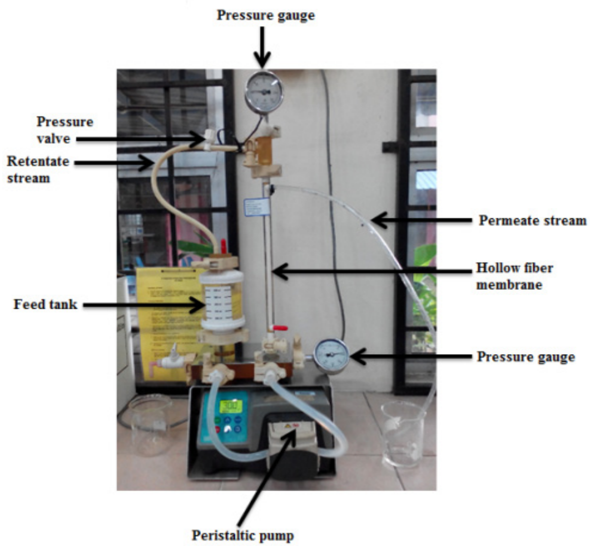

Figure 1. Cross-flow hollow fiber separation unit

The feed flow-rate and the TMP were set prior starting the unit. The feed flow-rates examined were 0.18, 0.34, 0.54, and 0.74 L/min. Meanwhile, TMPs were experimented at 0.41 bar, 0.55 bar, and 0.69 bar. This range was selected based on a preliminary study done by the researcher.

Each run was performed in duplicate by starting with varying cross-flow rate at a constant TMP, followed by varying TMP at a constant cross-flow rate. The separation process began with constant TMP (0.55 bar) at different feed flow-rates (0.18, 0.34, 0.54, and 0.74 L/min) to investigate the effect of feed flow rate on cross-flow filtration process. The obtained results were observed and the feed flow-rate that produced the highest permeating flux, J, had been considered as the best operating flow-rate and hence, was selected as the operating flow-rate used throughout the research.

Then, the separation process was continued with operation at different TMPs (0.41 bar, 0.55 bar, and 0.69) with a constant feed flow-rate (according to each membrane size) to determine the effects of TMP on the separation process, and thus, the best TMP value was obtained by observing the permeate flux and lactose rejection obtained for each parameter condition.

Firstly, the feed was allowed to run through the unit until the feed entered the membrane cartridge. Beakers were prepared at the permeate stream outlet to collect the samples that flowed into the centrifuge tube. When filtrate began to flow out, the stopwatch began and the sample was collected in every 5 minutes for 60-minutes duration. After 5 minutes, immediately a new container was replaced to collect the filtrate. The permeate flux was measured at every 5-minutes interval using equation (1). This was a continuous process until the designated processing time was completed. Similar steps were applied to both UF sizes used. All samples of retentate and permeated streams were collected in order to calculate permeate flux (J), concentration factor (CF), and accumulation rate using following equations:

Permeate flux, J (*L/m²*.*hr*) (Limsawat and Pruksasri, 2011).

a) Effect of TMP on flux
b) Effect of feed flow-rate on flux

$$J = \frac{V_P}{A.t} \quad (1)$$

where; V_p = permeate volume (L); A = membrane effective area (m^2); t = time (minute)

Concentration Factor, CF (Limsawat and Pruksasri, 2011)

$$\mathrm{CF} = \frac{Retentate\ mass + Permeate\ mass}{Retentate\ mass} \quad (2)$$

Accumulation rate (Shirazi, Lin, and Chen, 2010)

$$\text{Accumulation rate} = \frac{Permeate\ mass + Retentate\ mass}{Permeate\ mass} \quad (3)$$

The total volume of each retentate (ml), permeated (ml), and unrecoverable holdup volume (ml) had been recorded. Ideally, these amounts summed to the total feed amount (Aspelund, 2010). If they fall short, there were likely due to some adsorption and solubility losses of foulant such as protein and fat on UF membrane materials during the process.

Total feed = Retentate + Permeated + Unrecoverable holdup volume (4)

Statistical analysis for effects of TMP and feed flow-rate on permeate flux

Next, the statistical method SAS (version 9.3 SAS institute Inc., Cary, USA) was used for statistical analyses. The significant differences between the mean values were calculated in one way analysis of variance (ANOVA) form using Proc Print procedure, Duncans multiple-range test at probability $P < 0.05$.

HPLC analysis on lactose concentration of goat's milk

In this study, the concentrations of lactose in sample solutions were determined via HPLC based on the method from ISO 22662:2012 (IDF 198: 2012), which stated the determination of lactose content by HPLC for milk and milk products. A monosaccharide analysis column (Phenomenex, Rezex RCM-Monosaccharide column, 7.8 x 300 mm) was used in the HPLC system (1200 series, Agilent Technologies, USA), which consisted of a refractive index (RI), detector (S 3580), a pump (Isocratic LC Pump 250, Pelkin Elmer), a column oven (CH-30 column heater, Eppendorf), and a system for data analysis. The lactose standards for calibration (D Lactose monohyhdrate) were purchased from SIGMA.

The permeated and retentate samples obtained from each separation process were filtered through syringe filters (ChromTech, Nylon membrane, nominal pore size 0.22µm, and filter size 25mm) prior to injection into the HPLC system. The sample injection volumes were 20 µL for all samples. The eluent used was pre-degassed with distilled water at a temperature of 80°C and it was fed at a flow-rate of 0.6 ml/min. The column detection at 40°C with retention time or peak efficiency for sugar lactose was at minutes 9.03.

Lactose concentration in each samples were measured in order to analyze the data in terms of lactose rejection percentage (%Ri) by using the following equation:

Lactose rejection percentage, %Ri (Limsawat and Pruksasri, 2011)

$$\%R_i = \left(1 - \frac{C_p}{C_f}\right) X\ 100 \quad (5)$$

where; C_p = concentration of solute in the permeate (mg/mL); C_f = concentration of solute in the feed stream (mg/mL)

Lactose concentration was determined via calibration plot of absorbance of standard lactose solution, as suggested by Chollangi and Hossain (2007). The absorbance of each sample was measured using HPLC. Five samples of standard lactose were diluted to prepare aqueous solutions with concentrations of 20, 40, 60, 80, and 100 mg/ml.

Results and Discussion

Effects of feed- flow rate

Figures 2 (a) and (b) represent permeate flux at different feed flow-rates for 5 KDa and 10 KDa membrane pore size, respectively. Both of the graphs show that permeated flux decreased as the separation time proceeds regardless of flow rate. At the onset of filtration from 5 to 15 minutes, the steep decline slope was observed but only for a transient period. At the end of the separation within 50 to 60 minutes, the flux shows the slowest filtration. This is because, the increase in feed flow-rate increasing the permeate flux at the tube inlet, but as the feed flows through the length of the tube, the permeate flux decreases. The initial increase in permeate flux is due to the decrease in CP when operating at high feed flow-rate which is also agreed by Yeh *et al*. 2003. As concentration proceeds, the onset decrease in CP incorporated with better retention of proteins at the entrance of the tube gradually increases the viscosity (total solid) of the feed solution as it flows along the tube (Cheryan, 1998). These results in an increase of protein concentration along the tube, hence increases CP along the tube, resulting in a decrease in permeate flux down the tube (Yeh, 2002; Yeh, 2003).

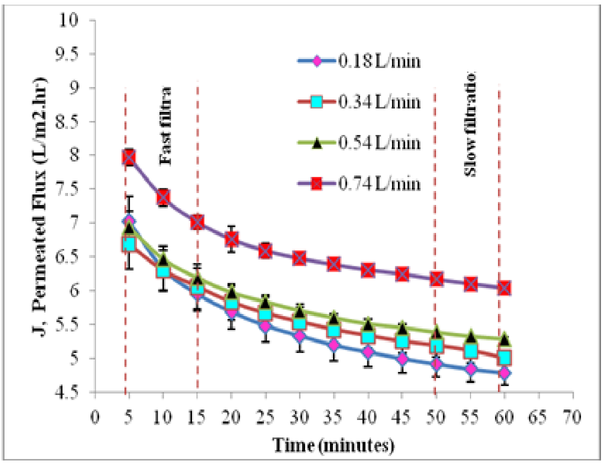

Figure 2. (a) Effect of feed flow rate at constant TMP in 5 KDa ultrafiltration membrane

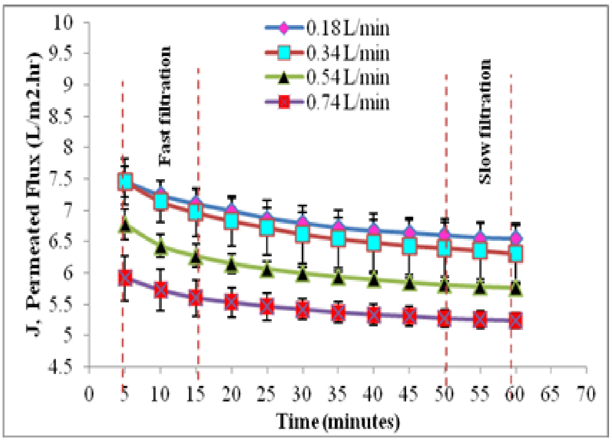

Figure 2. (b) Effect of Feed flow rate rate at constant TMP in 10 KDa ultrafiltration membrane

From Figure 2 (a), for 5 KDa membrane, as the feed flow-rate increased the permeate flux also increased except for flow rate 0.18 L/min which at the beginning the permeated was slightly higher compared to 0.34 and 0.54 L/min. In 5 KDa, feed-flow rate of 0.74 L/min resulted in the highest permeated flux followed by 0.54, 0.34, and 0.18 L/min. Meanwhile, based on Figure 2 (b), for 10 KDa UF membrane, the result obtained was contrary to 5 KDa pore size. The permeate flux responded in inversely proportional against feed flow-rate where the flux decreased as the feed-flow rate increased. In this membrane, feed flow-rate of 0.18 L/min gave the highest permeate flux, followed by feed flow-rate of 0.34 L/min, 0.54 L/min, and 0.74 L/min.

These different patterns of flux between 5 KDa and 10 KDa UF membranes are most likely due to different pore size since UF is a size-based separation membrane and hence lead to different limiting flux between each membrane. The contrary filtration results between 5 and 10 KDa membrane in this study are also supported by many past literature, which agreed that fouling depends on the membrane pore size (Belfort *et al*., 1994; Kelly and Zydney, 1995; Guell and Davis, 1996; Singh and Cheryan, 1997).

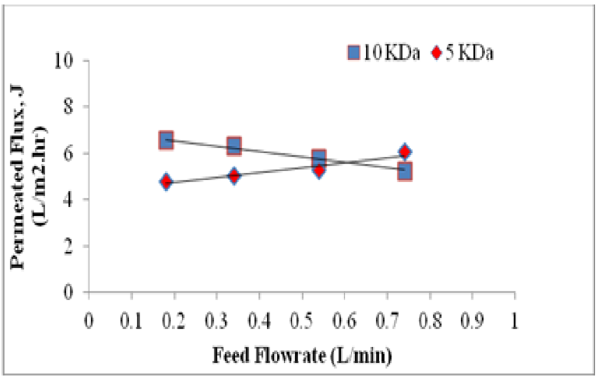

Figure 2. (c) Flux comparison between 5 KDa and 10 KDa membrane at different feed flow-rate, constant TMP 0.55 bar, and 60 minutes separation time;

Figure 2 (c) illustrates the flux comparison between 5 KDa and 10 KDa UF membrane at different feed-flow rates, constant TMP 0.55 bar, and at 60 minutes separation time. At the beginning, 10 KDa membrane operated at 0.18 L/min achieved the fastest filtration, but showed the flux decay as the flow rate increased. The flux continued to decline gradually when operated in the sequence of 0.34, 0.54, and 0.74 L/min. Interestingly, at highest flow-rate (0.74 L/min), 5 KDa membrane appeared to have a faster filtration compared to 10 KDa, although it resulted in an initial lower flux when operating at flow rate of 0.18 L/min up to 0.54 L/min. The increment of flow rate can be said to have been greater for 5 KDa membrane, but not for 10 KDa.

The result obtained agreed with the report by Kanani, (2015) that stated that membrane with larger pore sizes resulted in an initial higher flux compared to membrane with smaller pore sizes. However, as the separation proceed (Figure 2(c)), it appears that the flux declined rapidly and eventually stopped at lower flux. Many studies also agreed that the resolution for this is the indication of the optimum pore size, below which flux are substantially reduced by membrane resistance and cake fouling layer, whereas above which a severe degree of fouling deposited into the membrane pores where the process was irreversible (Marshall *et al*., 1993).

It can be seen from Figure 2 (c) that in 10 KDa membrane, flux of 6.54 L/m^2.hr is the limiting flux where further increase in feed flow-rate above 0.18 L/m^2.hr caused an over-fouling in membrane and decline in flux (Choi, 2003). As feed-flow rate increased, permeate flux also increased in 5 KDa membrane caused by the increased of sweeping effect (Hwang and Hwang, 2006). On the other hand, as feed flow-rate increased, permeate flux decreased in 10 KDa membrane due to the increase in membrane transport resistance (Baker *et al*., 1985; Tanaka *et al*., 1998; Tanaka *et al*., 2001).

In 5 KDa UF membrane, this result is due to the high feed flow-rate which reduced the deposition of boundary layer cakes or other clogs at membrane surface that may affect the permeation of particles, resulting in the higher permeate flux (Matthiason, 1980). As reported by Song and Elimelech (1995), the major hindrance in CFF processes over time is the demonstration of CP phenomena effect. Accordingly, high feed flow-rate is required to avoid blocking due to CP at the inlet (Wagner, 2001). This is due to fouling and CP problem that might hindere the transmission of lactose through the membrane, thus make it difficult for the lactose molecule to penetrate through smaller membrane size. This justifies why in 5 KDa UF membrane, the highest flux was obtained when operating at feed flow-rate of 0.74 L/min.

Meanwhile, in 10 KDa UF membrane, increment in feed flow-rate made the membrane boundary layer thicker and resulted in high membrane resistance and lower permeate flux. This is because at the higher feed flow-rate, as the concentration of the retained molecule increased at the membrane surface and formed a filter cake, there was a higher resistance to flow through the membrane, leading to a decrease in the permeate flux and difficulty for the smaller permeable species to pass through the membrane (Tanaka *et al*., 1998).

At 60 minutes separation time in 5 KDa, feed flow-rate of 0.74 L/min gave the highest permeate flux, followed by feed flow-rate of 0.54, 0.34, and 0.18 L/min at 6.04, 5.29, 5.00, and 4.79 L/m^2.hr, respectively. Meanwhile, at 60 minutes separation time in 10 KDa, feed flow-rate of 0.18 L/min gave the highest permeate flux, followed by feed flow-rate of 0.34 L/min, 0.54 L/min, and 0.74 L/min at 6.54, 6.31, 5.77, and 5.24 L/m^2.hr, respectively. Feed flow-rate 0.74 L/min and 0.18 L/min produced the highest flux in MWCO 5 and 10 KDa membrane, respectively, thus was selected to be the operating flow rate for the following experiment. Statistical method SAS 9.3 was used to determine the best feed-flow rate in a way of statistical analysis proven data.

Effects of TMP

According to previous experiment, the selected operating feed-flow rate for 5 KDa UF is 0.74 L/min, while for 10 KDa membrane is 0.18 L/min. Therefore, for MWCO 5 KDa, the feed flow-rate was maintained constant at 0.74 L/min, while for 10KDa UF membrane, the feed flow-rate was maintained constant at 0.18 L/min. The performance was measured based on the permeate flux obtained for a given TMP.

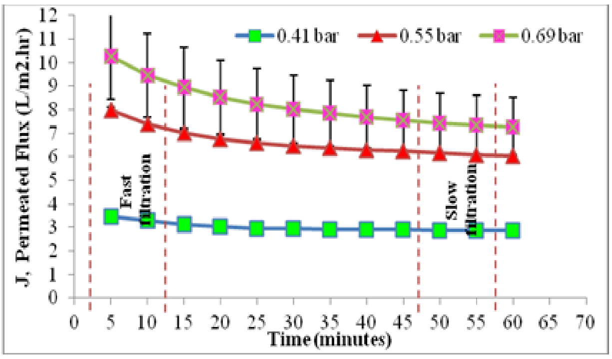

Figure 2. (d) Effect of TMP at constant feed-flow rate in 5 KDa ultrafiltration membrane

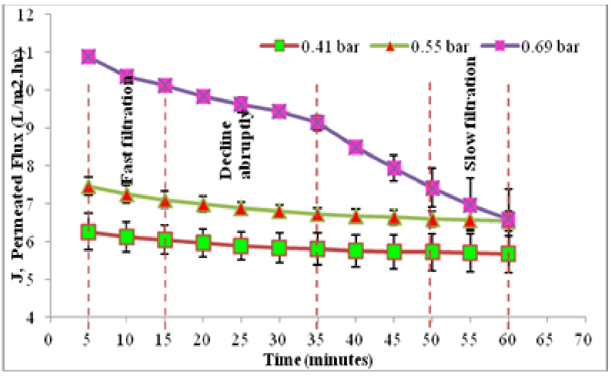

Figure 2. (e) Effect of TMP at constant feed-flow rate in 10 KDa

Figure 2 (d) and (e) show the effect of permeate flux against separation time at various TMP for 5 KDa and 10 KDa UF-membrane, respectively. Both membranes showed that as TMP increased from 0.41 bar to 0.69 bar, the permeated flux also increased. TMP 0.69 bar gave the highest permeate flux, followed by TMP 0.55 and 0.41 bar. This proved that higher TMP resulted in higher permeate flux and likewise, and that there was a parallel correlation between TMP and permeates flux (Cheryan, 1998; Yeh *et al*., 2003).

Figures 2 (d) and (e) also show that permeated flux declines as the processing takes time. This is because when constant feed flow-rate is maintained, permeate flux is allowed to decline as fouling limits membrane permeability. For all replications, as the product flows down the hollow fiber membrane, there is a natural hydrodynamic pressure drop from the inlet to the outlet of the flow channel. The uneven permeate flux distribution along the length of the flow channel can be very significant due to the resulting CP effect particularly at the higher pressure inlet, and decreasing toward the outlet end which is supported by Atkinson, 2005. CP not only offers extra hydraulic resistance to the flow of solvent but also results in the development of osmotic pressure which acts against the applied TMP (Aspelund, 2010). This explains why the permeated flux for all TMP decreased over time.

Based on Figure 2 (d), in 5 KDa UF membrane, all TMP show the highest filtration at the early stage from 5 to 15 minutes, while all present the slowest filtration at the end stage of separation from 50 to 60 minutes. Based on figure 2 (e), in 10 KDa UF membrane, from 5 to 15 minutes, all TMP also performed the highest filtration, but at TMP 0.69 bar, there is a considerably rapid decrement in permeating flux, especially at 35 minutes where the flux started to decline abruptly. This is because when larger pores of membrane (10 KDa) are blocked in the early stage, higher retention observed, hence permeate flux decline sharply (Brans *et al*., 2004). In this case, increasing the pressure initially results in an increase in permeate flux and in the fouling rate (Forman, 1990; Jonsson *et al*., 1995). This fouling interferes with the product transmission through the membrane, decreasing the quality of the separation, increasing membrane retention, shortening running time, and increasing costs.

This is also supported by Grandison *et al*. (2000) which stated that deposition on the membrane surface at higher TMP is closely packed, hence difficult to remove the deposited layer. When the TMP were too high, convective forces towards the membrane were also too high. Subsequent work showed that fouling was reduced significantly at low values of TMP. Accordingly, though 0.69 bar gave the highest flux, TMP 0.69 bar was not selected as the operating pressure because the high TMP increased the resistance of the cake layer due to the deposited mass that highly accumulates and besides, it involves higher operating cost in the plant scale (Blanpain, *et al*., 1993; Li *et al*., 1996; Ohmori and Glatz, 1999; McCarthy *et al*., 2002).

ultrafiltration membrane

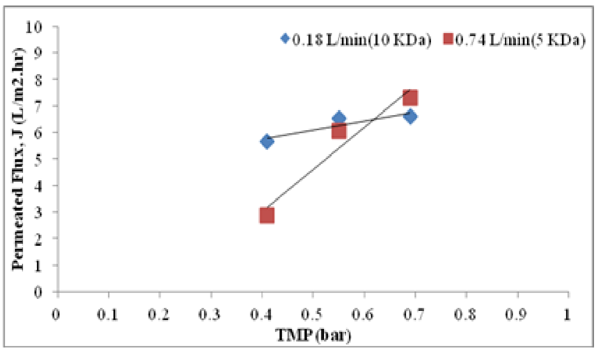

Figure 2. (f) Flux comparison between 5 KDa and 10 KDa membrane with different TMP, constant flow rate, and 60 minutes separation time

Table 1 (a) Mean (n=2) SAS analysis, lactose rejection percentage, concentration factor, and accumulation rate at different feed-flow rate with constant TMP 0.55 bar and 60 minutes separation time

TMP (bar)	MWCO (KDa)	Mean (L/ m^2.hr)	Feed-flow rate (L/min)	Standard Deviation	Lactose Rejection (%Ri)	CF	Accumulation Rate
0.55	5 KDa	6.04	0.74^{A}	0.04	77.71	1.30	4.42
		5.29	0.54^{B}	0.04	78.51	1.25	5.03
		5.00	0.34^{C}	0.08	81.51	1.26	5.22
		4.79	0.18^{C}	0.18	78.76	1.23	5.50
0.55	10 KDa	6.54	0.18^{A}	0.25	66.28	1.33	4.07
		6.31	0.34^{B}	0.46	74.94	1.25	5.00
		5.77	0.54^{B}	0.05	74.81	1.27	4.68
		5.24	0.74^{C}	0.13	71.37	1.24	5.11

$^{A-C}$ Means in the same column not sharing a common superscript are different (P<0.05).

Figure 2 (f) shows value of permeated flux at 60 minutes separation time with different TMP and constant feed flow-rate. Based on the figure, in 5 KDa UF membrane, the permeated flux at 60 minute separation time is in ascending order from for TMP 0.41, 0.55, and 0.69 bar at 2.87, 6.04, and 7.29 L/m2.hr, respectively. Meanwhile, for 10 KDa UF membrane, the permeated flux obtained at 60 minutes separation time was also in ascending order from TMP 0.41, 0.55, and 0.69 bar at 5.66, 6.54, and 6.59 L/m^2.hr, respectively. From the figure, in 10 KDa membrane, flux of 6.54 L/m^2.hr is the limiting flux where further increase in TMP above 0.55 bar would cause the system to be operated in a pressure-independent zone. This means that TMP has a negligible effect on the permeate flux and leads to an irreversible fouling due to firm compaction of foulant in membrane (Choi, 2003). For both membranes performance, the notable increase is from TMP 0.41 bar to TMP 0.55 bar, while starting from TMP 0.55 to TMP 0.69 bar, there is only slightly increment of flux. The results obtained can be summarized that the selected operating TMP in both membrane is 0.55 bar.

This occurrence is because flux may not increase proportionally to pressure at higher TMP due to pore compression as reported by Chollangi and Hossain, 2007 and Pouliot *et al*. 1999 which both membranes (5 and 10 KDa) from this study did not show any signs of compaction caused by pressure effect until TMP 0.69 bar was exerted on the separation system, which reduced diffusivity and dispersion of the polarized layer (Attia, 1991; Scott, 2012). On the other hand, too high TMP may cause over-fouling (Attia, 1991). Hence, fouling on the membrane surface may be reduced by exerting the best TMP to increase the diffusivity and decrease the CP. Statistical method SAS 9.3 was used to determine the best TMP in a way of statistical analysis proven data.

Result of statistical analysis on TMP and feed-flow rate

Based on Table 1 (a), for 5 KDa pore size, feed flow-rate 0.74 L/min had the highest mean score (P < 0.05), compared to feed-flow rate 0.54, 0.34, and 0.18 L/min. This means that feed-flow rate 0.74 L/min resulted in the highest flux, 6.04 ± 0.04, followed by 5.29 ± 0.04, 5.00 ± 0.08, and 4.79 ± 0.18 when performed at 0.54, 0.34, and 0.18 L/min, respectively. Feed flow-rate 0.34 and 0.18 L/min had a comparable flux. Although no change was detected at 0.18 and 0.34 L/min (P > 0.05), feed flow-rate of 0.34 L/min gave flux increment. At this point, the separation process is in the equilibrium state. Feed-flow rate 0.18 L/min resulted in the lowest permeated flux for 5 KDa and thus was rejected. The mean score for permeated flux decreased with a decrease in feed flow-rate. It is clear from the data representing the desired flux rating for 5 KDa is 0.74 L/min.

Meanwhile, for 10 KDa UF membrane, Table 1 (a) shows that feed flow-rate 0.18 L/min had the highest mean flux (P < 0.05) and possibly gave the best condition required for goat's milk separation process, followed by feed flow-rate of 0.34, 0.54, and 0.74 L/min. Mean of feed flow-rate 0.34 and 0.54 L/min each was not significantly different (P > 0.05) from each other and it indicated that the separation process at these feed flow-rate reached the equilibrium permeated flux. Moreover, there was an increase (P > 0.05) in mean flux between 0.74 and 0.54 L/min, while feed flow-rate of 0.74 L/min gave lowest flux (P < 0.05), and thus was rejected. However, as opposed to 5 KDa membrane, the mean score for permeated flux decreased with an increase in feed-flow rate in 10 KDa membrane. It is therefore important to optimize the feed-flow rate used for the purpose of removing lactose efficiently.

Statistical method SAS 9.3 also was used to select which TMP in a range of TMP 0.41, 0.55, and 0.69

Table 1 (b) Mean (n=2) SAS analysis, lactose rejection percentage, concentration factor, and accumulation rate at different TMP with constant feed-flow rate (0.74 and 0.18 L/min for 5 and 10 KDa, respectively) and 60 minutes separation time

Feed-flow rate (L/min)	MWCO (KDa)	Mean	TMP (bar)	Standard Deviation	Lactose Rejection (%Ri)	CF	Accumulation Rate
0.74	5 KDa	7.29	0.69A	1.24	73.45	1. 37	3.78
		6.04	0.55A	0.04	77.71	1.29	4.42
		2.87	0.41B	0.01	89.99	1.13	8.75
0.18	10 KDa	6.59	0.69A	0.80	72.94	1.34	3.95
		6.54	0.55A	0.25	66.28	1.33	4.07
		5.66	0.41B	0.13	74.31	1.28	4.61

A - C Means in the same column not sharing a common superscript are different (P<0.05).

bar that would give the best condition parameter in a way of statistical analysis proven data. From Table 1 (b), for 5 KDa membrane pore size, TMP 0.41 bar gave the lowest permeated flux, and was significantly different ($P < 0.05$) from TMP 0.55 and 0.69 bar, therefore it was rejected. At TMP 0.55 and 0.69 bar, flux did not change ($P > 0.05$) with time, but decreased ($P < 0.05$) when processing at 0.41 bar. In addition, no difference ($P > 0.05$) was detected between TMP 0.55 and 0.69 bar and they have a comparable flux with higher flux in TMP 0.69 bar. However, in this case, TMP 0.69 bar was not selected because of high pressure drop which led to severe mass accumulation and high operating cost in a plant-scale.

From Table 1 (b), for 10 KDa membrane pore size, TMP 0.41 and 0.55 bar were significantly different ($P < 0.05$), provided that TMP 0.55 bar gave a higher permeated flux. Meanwhile, there was no difference ($P>0.05$) detected between TMP 0.55 and 0.69 bar. Though TMP 0.69 bar gave the highest flux, the means score was not significant, hence it was rejected due to high pressure drop leading to severe deposited mass and high operating cost.

For both membranes performance, the notable increase ($P>0.05$) was greater from TMP 0.41 bar to TMP 0.55 bar, while starting from TMP 0.55 to TMP 0.69 bar, there was only slightly and non-significant ($P < 0.05$) increment of flux, indicating that it had reached the limiting flux, J at TMP 0.55 bar. Above this limiting flux, further increment in TMP would not give a substantial permeating flux due to the resistance in the membrane boundary layer, which also increased as the TMP increased (Cheryan, 1998). This indicates that no further improvement in the separation is to be expected by increasing TMP. Accordingly, TMP 0.55 bar was selected as the operating TMP for both 5 and 10 KDa membrane pore size.

Effect of TMP and feed flow-rate on lactose rejection, concentration factor, and accumulation rate

Tables 1 (a) and (b) show the summarized effects of each parameter on lactose rejection percentage, %Ri, CF, and accumulation rate. From the table, the lowest lactose %Ri are 77.71% and 66.28% when run at feed flow-rate of 0.74 $L/m^2.hr$ in 5 KDa, while having 0.18 $L/m^2.hr$ in 10 KDa, respectively. In 5 KDa membrane, lactose %Ri increased from 77.71% to 78.51%, 78.76%, and 81.51% at feed-flow rate of 0.74, 0.54, 0.18, and 0.34 L/min, respectively. Meanwhile, lactose %Ri in 10 KDa membrane increased in the range from 66.28% to 74.94% at feed-flow rate of 0.18, 0.74, 0.54, and 0.34 L/min, respectively. The increment of La %Ri was due to the majority accumulation of protein and minerals near the membrane surface, causing membrane resistance against lactose permeation, thus hinder permeation through the membrane and then left in the retentate line known as ‘retentate’ (Marshall and Daufin, 1995; Kelly and Zydney, 1997). It can be concluded that highest flux resulted in lowest lactose rejection and that lactose %Ri had parallel correlation to permeate flux.

Based on Table 1 (a), in MWCO 5 KDa, goat’s milk was processed under constant TMP and flow rate 0.54 L/min to 1.97X CF, marked as the highest concentration factor, followed by 1.96X CF at 0.74 L/min, while 0.34 and 0.18 L/min had a similar 1.89X CF. In 5 KDa, too high CF leads to higher solute concentration on the membrane surface. Therefore, to maximize separation efficiency, the appropriate CF should be selected, where there was a critical concentration, above which the system would encounter rapid fouling. This is because higher CF resulted in the same effect as increasing feed concentration, which is severe fouling because of solutes that accumulate on the membrane surface as reported by Adams, 2012. For 10 KDa membrane, CF marked the highest value with 1.33X CF at feed-flow rate of 0.18 L/min in the range from 1.24 to 1.33X CF; which correlate with the lactose rejection percentage and flux obtained.

From Table 1 (a), in 5 KDa membrane, accumulation rate has the same correlation to mean flux as the lowest accumulation rate was at feed-flow rate of 0.74 L/min, and increased in the order of 0.54, 0.34, and 0.18 L/min which are 4.42, 5.03, 5.22, and 5.50, respectively. On the other hand, in 10 KDa membrane, accumulation rate is the lowest which is 4.07 at feed-flow rate of 0.18 L/min, and increased in the order 4.68, 5.00, and 5.11 at feed-flow rate of 0.54, 0.34, and 0.74 L/min, respectively. Hence, it can be concluded that the best feed-flow rate in 5 and 10 KDa membrane sizes is 0.74 and 0.18 L/min, respectively.

On the other hand, from Table 1 (b), lactose %Ri is the lowest (73.45%) at TMP 0.69 bar for MWCO 5 KDa, while for MWCO 10 KDa, the lowest lactose %Ri (66.28%) was at TMP 0.55 bar. Although TMP 0.69 bar gave the highest flux for both membrane sizes, lowest lactose %Ri value, highest CF, and lowest accumulation rate, it had no significant changes compared to TMP 0.55 bar. Particularly, in 10 KDa membrane, although TMP 0.55 bar gave lower flux than TMP 0.69 bar, however, the lactose transmission is greater at TMP 0.55 bar. This was due to lower TMP caused more shearing action on the membrane surface to prevent the formation of CP as supported by Bowen, 1992. It should also be noted that in 10 KDa, lactose %Ri increased from 66.28% to 72.94% when operating at TMP 0.55 and 0.69 bar, respectively, due to the increment of lactose concentration in the retentate because processing had exceeded the limiting flux region (Vyas *et al*., 2003). Hence, higher TMP is not recommended as it could cause a faster decline in flux. At TMP 0.41 bar, the highest lactose rejection was obtained due to insufficient pressure force leading to membrane resistance and cake fouling layer as explained by Marshall *et al*. (1993).

In addition, these situations lead to the selection of TMP 0.55 bar as the best TMP in goat's milk lactose fractionation process in both membrane sizes. In conclusion, for 5 KDa UF membrane, feed flow-rate of 0.74 L/min and TMP 0.55 bar was selected as the best condition of processing parameter, meanwhile, for 10 KDa UF membrane, feed flow-rate 0.18 L/min and TMP of 0.55 bar was selected as the best condition parameters as they gave the highest permeate flux, lowest lactose %Ri, highest CF, and lowest accumulation rate without reaching the limiting flux region.

Based on the results analyzed, 10 KDa UF membrane size was chosen as the best membrane size in separating lactose from goat's milk compared to 5 KDa, as the size of lactose molecule is approximately 342.3 Da. The suggested membrane size to separate any molecule component in liquid is by multiply the size of the intended molecule by ten. In this case, 10 times the size of lactose molecule is approximately 3,000 Da. MWCO 5 KDa therefore can separate lactose molecule, but not as efficient as MWCO 10 KDa UF membrane. This is because both membrane sizes have a definite or a diffuse separation limit as the separation accuracy is determined by pore size and the size of particles in the feed solution as agreed by Aspelund (2010).

The difference in severity of fouling occurred was due to goat's milk comprising of solutes with the difference in molecular weight and shape. Therefore, the permeation of lower molecular weight solute (lactose) is influenced by the presence of higher (>342.3 Da) molecular weight solute (protein, minerals, fat). The higher molecular weight solute that retained may block the membrane pores forming a deposition layer as supported by Mulder, 1996, in which is severe in 5 KDa compared to 10 KDa UF size. Hence, 10 KDa UF membrane was selected as the best membrane size in lactose fractionation compared to 5 KDa membrane.

Conclusion

In cross-flow ultrafiltration separation system, both parameters of TMP and feed flow rate affected permeate flux, concentration factor, and accumulation rate in a manner according to each membrane size (5 KDa and 10 KDa) and membrane's behavior. TMP is proportionally related to permeated flux for both membranes, but feed flow-rate is proportionally related to permeate flux in 5 KDa, while inversely related to permeate flux in 10 KDa pore size.

The retentate obtained after each separation run was statistically analyzed on permeated flux. No significant difference ($P>0.05$) in mean flux was observed for TMP 0.55 and 0.69 bar in both membrane. Feed flow-rates of 0.74 L/min and 0.18 L/min were significantly greater ($P < 0.05$) compared to other feed flow-rates imposed in 5 KDa and 10 KDa membrane, respectively. It can be concluded that the feed flow-rate required is associated to UF pore size and molecular weight of feed solute particles.

Lactose %Ri is the lowest with 77.71% in 5 KDa UF, while 66.28% in 10 KDa UF. It can be summarized that the best operating parameter for 5 KDa UF is at TMP 0.55 bar with feed flow-rate of 0.74 L/min, while for 10 KDa UF is at TMP 0.55 bar with feed flow-rate of 0.18 L/min.

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