Selection of Tubular Membrane Separation based on the Resistance Performance

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Abstract: Processing of fruits (such as pink guava) to produce fruit juices results in high amount of waste materials that still contain valuable by-products (e.g. antioxidants or polyphenols). Analysis of hydrodynamic resistances that considers gel layer formation as the main fouling mechanism and permeate flux decline was studied. Using tubular membrane FP 100 and ES 404 performed the experiments, with a molecular weight cut-off of 100 kDa and 4 kDa respectively. Results showed that the permeate fluxes for both of the membranes increased by increasing the Trans Membrane Pressure (TMP) and it would decrease with time. All of the resistances increased with TMP meanwhile the mass transfer of polyphenols did not affect the TMP. All the TMP resulted in similar fouling values for both membranes.

1 INTRODUCTION

The waste-to-wealth approach for management of the residuals will result in the recovery of valuable by-products as well as solving the waste disposal problem. In the case of pink guava processing waste, the recovery of bioactive compounds is a profitable venture that can result in the recovery of polyphenols and other antioxidants. Polyphenols have good properties because they have anti-oxidant activity properties that are beneficial for enhancing health effects for humans (Friedman, 2002). Polyphenols can counteract the attacks of free radicals, therefore polyphenols can avoid the body from hereditary diseases such as cancer and other diseases (Gökmen, 2003).

In general, polyphenol compounds can be isolated or taken from fruit or vegetable processing wastes by the extraction process (D'Alvise, 2000). The use of filtration membranes for recovery, purification or concentration of fruits and vegetables has been extensively studied for the past 25 years. (Czekaj, 2000) identify restoration of polyphenols from pink guava manufacture residual pulp, and these processes can be done at low temperature; do not involve a high energy usage and makes it possible to separate bacterial and spore cells and to completely remove suspended solids. However, the decrease in permeate flux along with the increase in reaction time will occur, this is because some material will clog the membrane pores while others will thicken on the membrane surface and will shape a gel. Membrane contamination resulting in membrane fouling has been explored by many membrane separation researchers because it can make lower the productivity and age of membranes. (Nilsson, 1990). However, the decrease in flux caused by increasing the concentration of the solution on the surface of the membrane so that gel formation caused by complex polyphenols substances needs to be studied further. Furthermore, the basic mechanism so that the occurrence of fouling which causes a decrease in flux during the ultra filtration process of polyphenols is still not known correctly.

Fouling membrane that occurs in membrane filtration is influenced by three main factors, namely, the nature of the material or membrane material used, characteristics of the feed or sample and parameters of the operation process. In most studies related to ultrafiltration separation membranes (UF) a model is made using the hydrodynamic theory based on the formation of polarization concentrations and the formation of solids or gels on the membrane surface,

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which produce hydrodynamic resistance to absorb flow (Fane, 1987). This model can also be applied to the process of separating polyphenols from pink guava because this process also forms a layer of cake and gel on the surface of the separating membrane. Polarization of the concentration and layer of cake can facilitate irreversible membrane contamination by revising interactions between solvents, solutes and membranes. Therefore, to understand the phenomenon of the fouling process which results in membrane process failure is by analyzing the surface chemistry of the membrane, the interaction of solutes to the membrane and the interaction between the solute. So, the interaction between separation membrane and solution can determine the occurrence of fouling caused by adsorption of dissolved polyphenols on the membrane surface.

The paper is part of the research that has been done on the recovery of polyphenols from the processing of pink guava waste (Sukeksi & Sarah, 2016), (Sukeksi et al., 2016). This paper will discuss the effects of differences in operating pressure or TMP on membrane fouling and permeate flux in two types of membrane separation during the polyphenol recovery process.

Permeate flux decline over time is the main limiting factor that influence the membrane process. The permeate flux decline because of feed components increases inside the pores. This process results in membrane fouling. The feed component also increases on the membrane surface that result forming concentration polarization or gel layer. Some researchers have learned about fouling that occurs in membranes, this is done because fouling can reduce the productivity and lifetime of the membrane (Nilsson, 1990). However, a decrease in flux due to polarization and concentration of the solution resulting in fouling, and the complex effects of polyphenol substances need to be investigated further.

2 MATERIAL AND METHODS

2.1 Materials

Two commercial tubular membranes with FPDF FP 200 type with nominal MWCO 200,000 and ES 404 membranes with a nominal 4,000 MWCO with a pH operating range ranging from 1.5 - 12, and a maximum operating pressure of 10 bar, and a maximum operating temperature of 80˚ C. Membranes are supplied and manufactured by PCI, UK. The membrane housing used was supplied by local supplier, with 14 mm of inner diameter and 325 mm of length, with the module configuration contained two tubular membranes. Prior to use is soaking overnight in 0.3% HNO_3 to eliminate impurities left from the mechanized process or additives used for stabilization washes the membranes. Membrane equipment modules used for polyphenol recovery from pink guava processing waste consist of one diaphragm pump, 10 liter capacity feed reservoir, permeate collection reservoir, two inlet and outlet pressure gauges, valves for control and balance pressure and equipped with monitors for data processing. The all material of equipment such as, pump, feed reservoir and all connection tubing are used material base of stainless steel. Folin-Ciucalteu and Gallic acid were from Sigma-Aldrich (Germany) and Sodium Carbonate powder, Nitric Acid is supplied by Fluka (Germany). Processing of pink guava waste is collected from Sitiawan Perak, which is produced from a Decanter separator and Refiner separator with a composition of 50%. If this pink guava waste is stored in an improper manner it will result in a rapid loss of polyphenols, so the extraction process cannot be carried out. Therefore, the waste must be stored properly in the refrigerator to prevent fungal growth and oxidation.

2.2 Methods

2.2.1 Extract Preparation

Extraction methods using solvents are the most common way to isolate a compound from various fruits, as well as vegetables, such as polyphenol compounds. To isolate the substance in the extract is very dependent on the type of solvent used, because each type of polyphenols compound has a different polarity. Waste pink guava processing extract for total polyphenols content analyses are prepared by following method of Swain and Hillis (1959), with some modifications. Base in our study before, the best solvent for extraction to recovery polyphenol from pink guava wastes processing are Methanol/Water at 60% and the second is water. The best composition ratio between the sample wastes and solvent is 1:40. In this project the polyphenols within the pink guava wastes processing is extracted using water as a solvent. The choice of water as a solvent is based on the information that water more saves for human than organic solvent. Solid pink guava waste and water are then stirred using a blender constantly for 10 minutes until a homogeneous slurry or solution is produced. After 12 hours, the aqueous extract is separated from the solid by removed the upper of solution to reduce

the suspended solid content. The clear solution produced will be used for the recovery process of polyphenols using FP 200 and ES 404 membrane UF.

2.2.2 Recovery Polyphenols

Membrane separation is an alternative method for the solvent separation process from polyphenol extract. The system consists of PVDF 200 FP and ES 404 membrane connected to a feed reservoir and a diaphragm pump. The steps involve in experiment are:

To determine the flux of water by entering tap water into the membrane by turning on the pump at a certain TMP and calculating the volume of permeate (Vp) generated at a certain time (t) passing through the surface area of the separation membrane (A) using the equation below:

$$J = \frac{V_p}{t \, x \, A}$$

First Cleaning.

The cleaning involved by initial water flushing for ten minutes and nitric acid 0.3% 30 minutes followed by water flushing again for ten minutes, to remove impurities left from the mechanized process or additives used for stabilization. The end of water flushing flux was measured by using data storing from the data lodging.

Ultra Filtration.

In operation, the feed stream extract of pink guava processing wastes is pumped using a diaphragm pump through the both of tubular PVDF membrane. The process of ultra filtration tubular membranes begins with the permeate port being closed, this is to allow cross speed before permeating out, with both inlet valves for feed solution and retentate solution in wide open conditions. After the pump is run, the valve for the inlet channel is opened and the valve for the solution on the retentate is slowly closed to produce the preferred Trans Membrane Pressure (TMP). With the increase in volume of solution at the permeate, the concentration of polyphenols will also increase. Ultra filtration experiment is carried out with continuous retentate recycling. The result permeate is continuously removed, until the desired volume concentration ratio (VCR) is achieved.

$$\text{VCR} = \frac{V_f}{V_R}$$

Where:
VCR or (Volume of Concentration Ratio),
V_f (m^3) is initial volume of the feed
V_R (m^3) is retentate volume

All data is collected and record by computer via a data logger. The samples that are resulted from permeate and retentate are provided to analyze total polyphenols content.

Second Cleaning.

The cleaning procedure is the same with the first cleaning procedure before ultra filtration processing, and the water flushing flux was also measured by using data storing from the data lodging.

All the processing procedures were repeated for three times, by using the same membrane which are, each step operation until VCR = 4 were reached and at TMP = 1, 2 and 3 bar, respectively.

Determination of Total Polyphenols Content.

The total polyphenol content produced was determined in all samples using the Folin-Ciocalteau method, which was modified by the theory of (Singleton, 1965) with some modifications. Gallic acid calibration standard solution is prepared for 0.01-0.1 mg/ml by accurately weighing and dissolving with of distilled water as a solvent. The solution mixture consisted of 200 l extract of pink guava waste sample mixed with 1.5 ml of Folin-Ciocalteau reagent and left at room temperature for 5 minutes then 1.5 ml of sodium bicarbonate solution was added to the mixture. After standing 90 minute at room temperature, absorbance is measured at 760 nm. The results obtained are expressed as mg / ml Gallic equivalent (GAE). The gallic acid standard curve can be seen in Figure 1 which is in the range of Gallic acid concentration from 0.025 to 0.1 mg / ml, and this will be used to make a calibration curve.

Calculation of Membrane Performance.

The membrane performance was measured by concentration factor ($C_{R,P}$) and recovery ($C_{R,P}$) of polyphenols content. Concentration factor is the concentrations of polyphenols in either permeate or retentate solution divided by its concentration in the feed solution.

$$R_{(R,P)}\,(\%) = \frac{C_{(R,P)} \, x \, V_{(R,P)}}{C_f \, x \, V_f} \, x \, 100\%$$

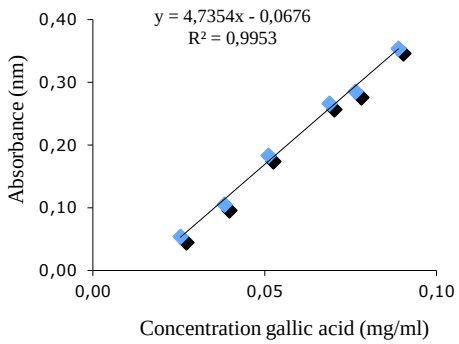

Figure 1: Gallic acid standard curve based on the data collected at absorbance 760 nm.

3 RESULTS AND DISCUSSIONS

3.1 Influence of TMP on the Permeate Volume

The increasing permeates volume as a function of time for tubular membrane at different pressures is shown in Figure 2. The permeate volume collected during recovery of pink guava processing wastes increased with time but at a decreasing rate. For membrane ES 404, the increasing of permeate volume for all transmembrane pressure are similar trend, there is no significant different between them.

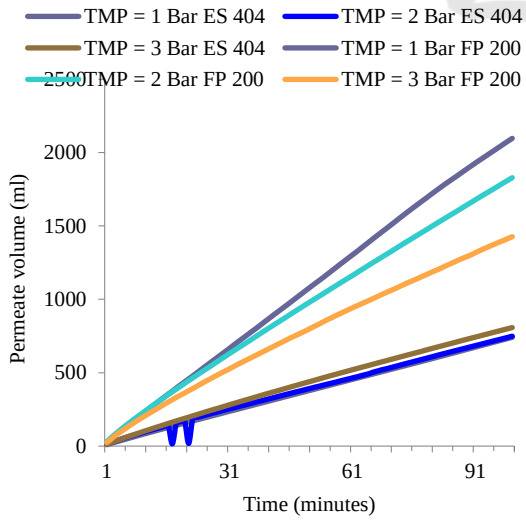

Figure 2: Permeate volume vs time for recovery polyphenols using PVDF membrane FP 200 and ES 404 at different TMP.

But for the membrane FP 100 the permeate volume collected at any time decrease with TMP increase. The lower permeation rates for pink guava processing wastes in comparison with those of water were due to membrane fouling.

For the pure water, the permeate volume collected during ultra filtration increase linearly with time for both of the membrane. In these cases, total resistance (R_t) and membrane resistance (R_m) constant throughout the whole operation and J or flux also constant at TMP constant.

3.2 Influence of TMP on The Permeate Flux

Investigational data related to the permeate flux decline for both of the membrane are presented in Figure 3.

The initial flux decline was 20-37% and 17-29% of the total flux decline for membrane FP 100 and ES 404, respectively at TMP 1-3 Bar.

The steady state was established after 20 minute of operation and the steady state permeate fluxes were 60-80% and 70-80% of their initial values for membrane FP 100 and ES 404, respectively.

Note that the initial flux of the permeate does not depend on the speed of the feed flow in the tubular membrane. This can be explained by the fact that at the start of the separation process, permeate flux is caused more by fouling that occurs on the internal membrane, which is clearly not significantly affected by the feed flow rate. Generally, the permeate flux increases initially with the transmembrane or TMP pressure applied, and then the flux will decrease with increasing pressure from the transmembrane. Contrary to this, the permeate flux for FP 100 membrane decreased with increased the TMP and for ES 404 all the permeate flux for different TMP has similar value.

The flux will reach steady state condition when the cake layer has grown to the equilibrium thickness. From the Figure 4.19 the steady state of flux decreased with the increase of TMP. (Song, 1998), already assumed the equilibrium thickness of cake layer increase with the applied pressure. It is because a thicker cake layer is needed to absorb a higher pressure.

Cassano et al. (2008) found the effect of TMP on the steady state of permeate flux which show a linear increase with TMP at lower pressures. Meanwhile at higher pressures the permeate fluxes approach a limiting value independent of further increases in TMP. Their point was considering the pressure independence as the peak of TMP (100 kPa). The first

fluxes decline at their first domain and was 75.8-89.1% of the total flux decline. The steady state value of flux shown after 34-84 minutes of processing and steady state of permeate fluxes were 22-43% from their first value.

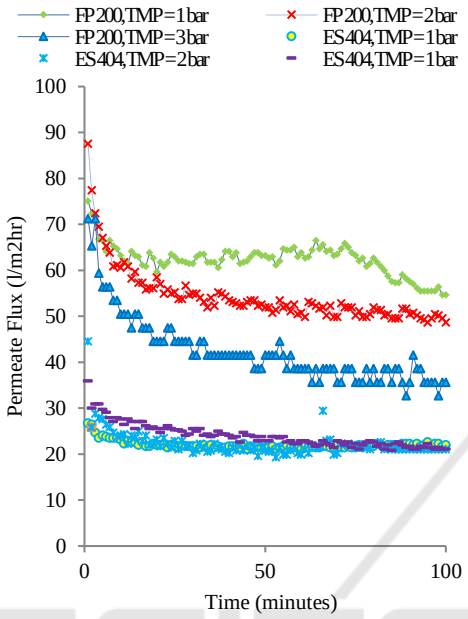

Figure 3: Permeate Flux Decline Vs Time.

3.3 Hydraulic Permeability and Membrane Resistance

The hydraulic permeability of the new clean membrane was 114.9 L/m2hBar for FP 100 membrane; meanwhile for ES 404 membrane was 70.8 L/m2hBar at 25ºC. New membrane resistance, R_m, was calculated from Eq. (3) to be 3.5 x 10^{12} m^{-1} forFP 100 membrane and 5.7 x 10^{12} m^{-1} for ES 404 membrane respectively. The membrane resistance calculated after the cleaning procedure was reported to its original value as observed from the hydraulic permeability data.

3.4 Influence of Transmembrane Pressure on the Total Resistance

As shown in Figure 4 the total resistance (R_t) increased with increased the TMP for both of the membrane. This phenomenon can be explained by assuming that an increase of pressure improved flux and convective flow of the solute towards the membrane. Therefore, the concentration polarization was more evident determining an increase of fouling resistance.

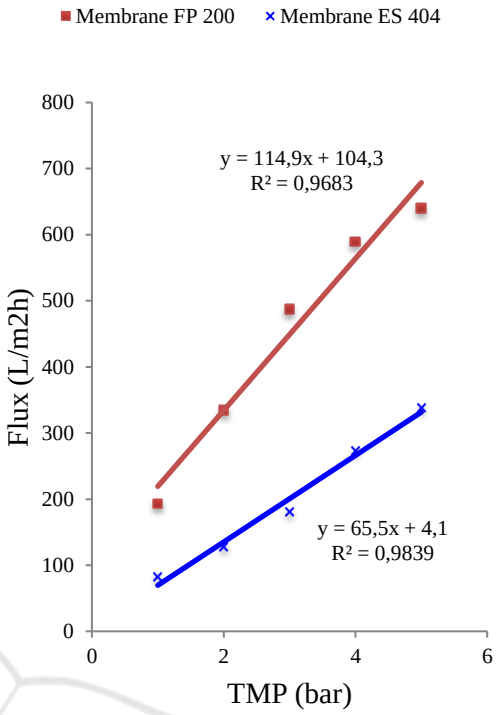

Figure 4: The influence of TMP on total resistance FP 100 membrane and ES 404 membrane.

4 CONCLUSIONS

Permeate volume increase with time for both of membrane ES 404 and membrane FP 200. For membrane ES 404, the increasing permeate volume for all TMP had a similar trend but for membrane FP 200 increasing of TMP resulted in increases of permeate volume. Initial flux decline was achieved 20% for membrane FP 200 and 37% for membrane ES 404 from the total initial flux at TMP 1-3 bar and steady state settled after 20 minutes of processing for both of membranes. The hydraulic permeability of the new clean membrane was 114.9 L/m2hbar for FP 200 membrane. Meanwhile for ES 404 membrane was 70.8 L/m2hbar at 25ºC. New membrane resistance, R_m, was 3.5 x 10^{12} m^{-1} for membrane FP 200 and 5.7 x 10^{12} m^{-1} for ES 404. Total resistance (R_t) increased with increased of TMP for both of the membrane. Meanwhile the R^2 for membrane FP 200 is 0.9719 and for membrane ES 404 is 0.9949.

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