

Contribution of membrane fouling during ultrafiltration of *Hylocereus Polyrhizus* Peel Extract

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Abstract— Betacyanin pigment is red purple colour that can be produced from the peels of *Hylocereus polyrhizus* fruit. The separation of betacyanin from fruit peels with high pulp content is very tedious due to the high concentration of fouling substances in the fruit peel that is also concentrated in the retentate during processing. The suspended solids usually responsible for this fouling are polysaccharides that come from cellular walls in the fruits, such as insoluble pectins, cellulose, hemicelluloses, and lignin. The high concentration of these macromolecules on the surface of the membrane produces a dynamic layer that, in some cases, forms an impermeable gel-like layer. In this study, ultrafiltration membrane was used as a tool to separate betacyanin pigment from *Hylocereus Polyrhizus* peel extract. Separation of betacyanin from *Hylocereus polyrhizus* peel extract was assessed by employing an ultrafiltration membrane reactor system. The effect of transmembrane pressure and crossflow velocity were studied. The fouling membrane can effectively control by changing the crossflow velocity and transmembrane pressure.

Keywords— betacyanin; *hylocereus polyrhizus* ; ultrafiltration

1. INTRODUCTION

Replacing synthetic dyes with natural colorants offers a challenge because the colour and stability of pigments are dependent on several factors like pH, temperature, light intensity, oxygen, and product degradation. Currently, *Hylocereus polyrhizus* is one of the new focuses for the next source of red natural dye because it is rich with betacyanin which is a similar array of colour pigments found in beetroot. The interesting colour of betacyanin, which is red-purple, can potentially be commercialised for various applications such as textile industry. Synthetic dye is used in textile industries for colouring fabrics such as cotton, silk, leather, nylon, leather and wool. The waste water from textile industries carries a large amount of dyes and other additives that are used during the colouring process [1]. Compounds from dye effluents are toxic and potentially carcinogenic [2] and can get into the system of an aquatic organism, pass through the food chain and ultimately reach humans, causing various physiological disorders such as hypertension, sporadic fever, renal damage and cramps [3]. Since *Hylocereus polyrhizus* have a high betacyanin pigment content, it has the credibility to be a sustainable source for natural dye for substituted synthetic dye. The *Hylocereus polyrhizus* peel is usually red, constitutes 22% of the whole *Hylocereus polyrhizus* fruit [4], contains considerable amounts of pectin, betacyanin pigment as well as dietary fibre, and is typically discarded by the *Hylocereus polyrhizus* juice processing industry. These discarded peels may cause environmental problems, particularly water pollution.

Ultrafiltration, which is widely used for the separation, purification and concentration of water-soluble solutes or water dispersible materials, is considered to be a versatile separation process. However, one of the biggest problems of using membranes is the decay of the permeate flux, which is caused by the phenomenon known as fouling. Serious membrane fouling easily occurs on membrane surfaces, thus limiting its wide application. The separation of betacyanin from fruit peels with high pulp content is very tedious due to the high concentration of fouling substances in the fruit peel that is also concentrated in the retentate during processing. The high concentration of these macromolecules on the surface of the membrane produces a dynamic layer that, in some cases, forms an impermeable gel-like layer [5].

Membrane such as ultrafiltration (UF) is widely used for removal of macromolecule [6] from kiwi [7], mosambi [8], apple [9], orange [10], passion fruit [11], pineapple [12], cactus pear [13] and carrot juice [14]. The presence of macromolecules needs to be removed because they potentially degrade the betacyanin structure and make the colour turn into brown. One of the biggest problems of using membranes to separate betacyanin from fruit peels is the decay of the permeate flux, which is caused by the phenomenon known as fouling. Fouling on the membrane surface as well as inside the membrane pores happens due to the accumulation of macromolecular or colloidal species at these sites.

In this research, betacyanin from *Hylocereus polyrhizus* peels was separated using UF membrane. The UF membranes used for separating the macromolecules, suspensions and colloids which are associated with betacyanin in *Hylocereus polyrhizus* peels. Thus the purpose of this research is to determine the affected factors which are transmembrane pressure (TMP) and crossflow velocity (CFV) during the separation process of betacyanin.

2. METHODOLOGY

2.1 Extraction Process

Betacyanin was prepared using water extraction. The purpose of using water extraction is to make sure that the pigment is safe from chemicals and other harmful substances. *Hylocereus polyrhizus* fruit peels were immersed in water with a solid loading ratio of 1:20, and then betacyanin was extracted at $50^{\circ}\text{C} \pm 5$, with a heating time of 10 minutes, in a 20 L tank. This method was modified from a previous study [15],[16]. Finally, the extraction was filtered in order to remove residual compounds using a metal sieve.

2.2 Separation Process

After extraction, the mixture was continuously subjected to filtration using an ultrafiltration membrane module. Figures 1 show the schematic diagram of the module during betacyanin filtration. Filtration was run at 0.70 ± 0.02 bars and 0.20 cm/s in order to study the effects of filtration time. At the preliminary filtration stage, the pure water flux (J_{pwp}) was measured by taking the volume of permeates every 5 minutes. After that, the pure water was replaced with the mixture extract and the volume of permeates was collected and measured every 5 minutes until permeation reached a steady state. TMP was varied at 0.50, 0.70 and 0.90 bars with constant CFV of 0.13 cm/s, for the purpose of studying the effects of TMP. In order to study the effects of CFV, experiments were performed at 0.13, 0.16 and 0.20 cm/s at a constant TMP of 0.70 ± 0.02 bars [17].

The parameter used for the quantification of the efficiency of membrane processes was flux (J), in which the flux can be defined as:

$$J = \frac{Q}{A} \quad (1)$$

where Q is the permeate flowrate (L h^{-1}) and A is the membrane area (m^2).

2.3 Analysis UV-Vis Spectroscopy Investigation of Betacyanin

Absorbance values for all samples were measured at 535 nm wavelength using a spectrophotometer to determine total betacyanin concentration (TBC). Resulting aliquots in each experiment were allowed to cool before taking UV-Vis Spectroscopy measurements. The absorbance readings obtained were used to calculate the total betacyanin concentration (TBC) for each sample using the following formula, derived from equation 2 [18].

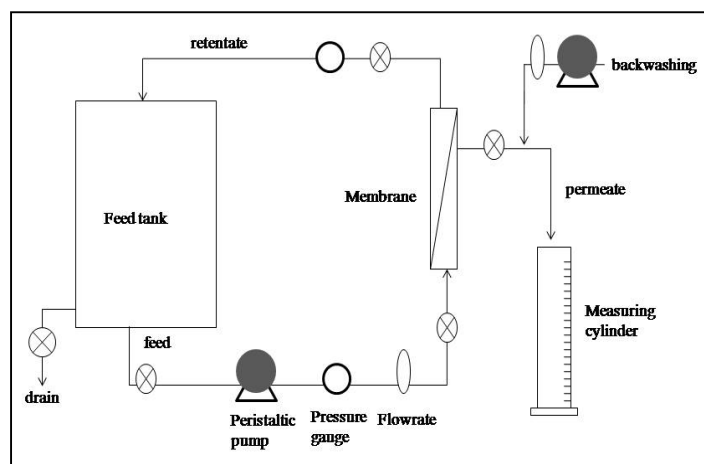


Figure 1: Schematic diagram during separation of betacyanin

$$\text{TBC (mg/L)} = \frac{A \times \text{MW} \times 1000 \times \text{DF}}{C \times l} \quad (2)$$

where A is absorbance, DF is dilution factor, MW is the molecular weight of betanin (550g/mol), C is molar extinction coefficient (60,000 L/mol cm in H₂O) and l is the path length of the cuvette (1cm).

3. RESULT AND DISCUSSION

Figure 2 shows the flux decline as a function of time in regards to various transmembrane pressures (TMP) applied during betacyanin separation. Increment in the operational TMP enhances flux declination and this accelerates the operation to achieve a steady state flux condition. This is due to the effects of compaction of the gel/cake layer [19]. As the flux increased, more solute mass (pectin and betacyanin) were brought to the membrane surface, which increased pore plugging and resistance to flow, thus increasing transmembrane pressure. Therefore, it is worth noting that the gel/cake layer deposited over the membrane plays an important role in betacyanin filtration. High TMP clearly indicates maximal permeate flux but unfortunately it would also increase the rate of membrane fouling, meanwhile low TMP reduces the rate of membrane fouling but does not lead to maximal membrane performance as it produces low concentrations of betacyanin. In fact, severe flux decline was observed at a higher operational TMP. High TMP clearly indicates maximal permeate flux but unfortunately it would also increase the rate of membrane fouling, meanwhile low TMP reduces the rate of membrane fouling but does not lead to maximal membrane performance as it produces low concentrations of betacyanin.

Figure 3 also indicates that the total betacyanin concentration during filtration process was found to be positively related to the operational TMP. Thus, increment of total betacyanin concentration as a correlation to increment in TMP verifies that the operational TMP is one of the most significant parameters that impacts betacyanin filtration. Higher TMP also increased the total betacyanin concentration as well as increasing steady state flux value. Moreover, high betacyanin filtered required high operational TMP in order to maximize membrane performance.

Cross flow velocity (CFV) is one of the operating conditions that is critical for influencing the efficiency of membrane filtration. As seen in Figure 4, the detrimental effect of filtration with lower CFV (0.13 cm/s) on permeate flux was more pronounced as it showed greater flux decline and higher fouling rate than filtration with either 0.16 cm/s or 0.20 cm/s of CFV. This could be related to compression of the fouling layer that covered the membrane surface (at low CFV) as a function of filtration time and afterward this fouling layer has become considerably more concentrated, forming a thick layer. This saturated layer hindered permeates flux through the membrane pores, thus generating lower amounts of betacyanin. Higher CFV implies a higher steady state flux than lower CFV, which suggests that less fouling occurred on the membrane surface and inside the pores. This finding appears to be consistent with the results obtained in Figure 4. This scenario is apparently attributed to the physical scouring effect of CFV on the membrane surface. Higher operating CFV resulted in increased shearing stress at the membrane surface, which redistributed the solute particles on the membrane surface. Filtration with this CFV (0.20 cm/s) produced relatively lower fouling compared to filtration with CFVs of 0.16 or 0.12 cm/s, subsequently making it easier to clean the membrane surface using hydraulic and chemical cleaning. Thus, it is worth noting that high CFV (0.20 cm/s) presumably implies greater membrane efficiency for longer operation time and less frequent membrane replacement.

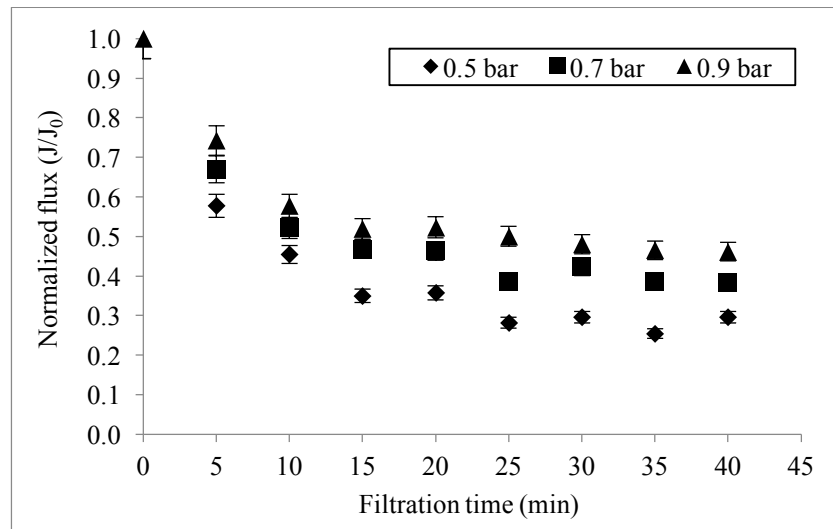


Figure 2: Effect of transmembrane pressure on membrane flux

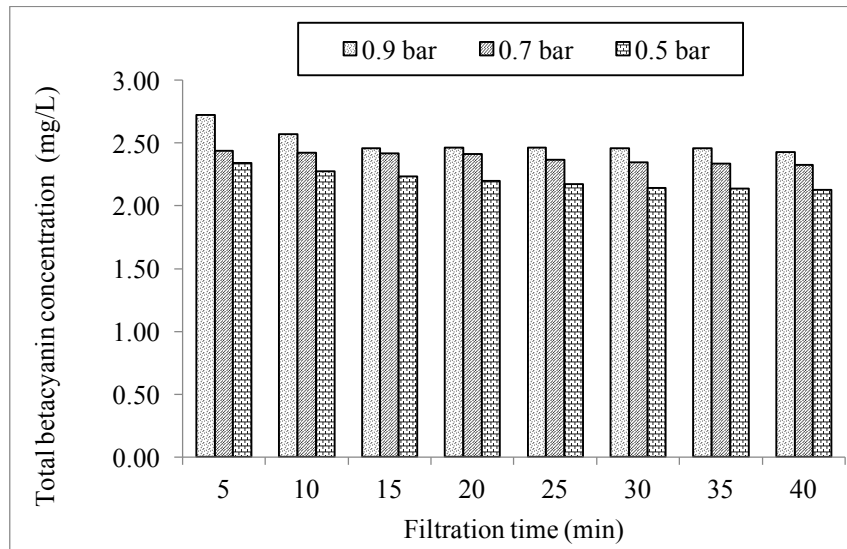


Figure 3: Total betacyanin concentration at different pressures

In addition, the results of Figure 5 show that the total betacyanin concentration slowly decreased with filtration time in respect to operating CFV. This scenario is again presumably related to the deposition of solutes on the membrane surface and subsequent covering of all the pores entries, which consequently resulted in the flux decline as well as low concentration of filtered betacyanin. This phenomenon is more critical for filtration with CFV of 0.13 cm/s. In brief, lower concentrations of betacyanin were filtered and membrane fouling was more severe with lower CFV compared to higher CFV.

4. CONCLUSION

In this study the separation of betacyanin from *Hylocereus polyrhizus* peel was successfully carried out using an ultrafiltration membrane. Optimized membrane operating parameters can control the amount of fouling, so that the membrane can be used for a longer time period. The analysis of transmembrane pressure (TMP) and crossflow velocity (CFV) for this study can be used for further studies and experiments for optimizing the process of betacyanin separation using ultrafiltration membrane. Increased transmembrane pressure can lead to a positive effect in flux, since it is the driving force. On the other hand, fouling and polarized layer are more accentuated under higher transmembrane pressure in betacyanin separation. Meanwhile, increasing crossflow velocity was more effective at reducing fouling on the membrane surface.

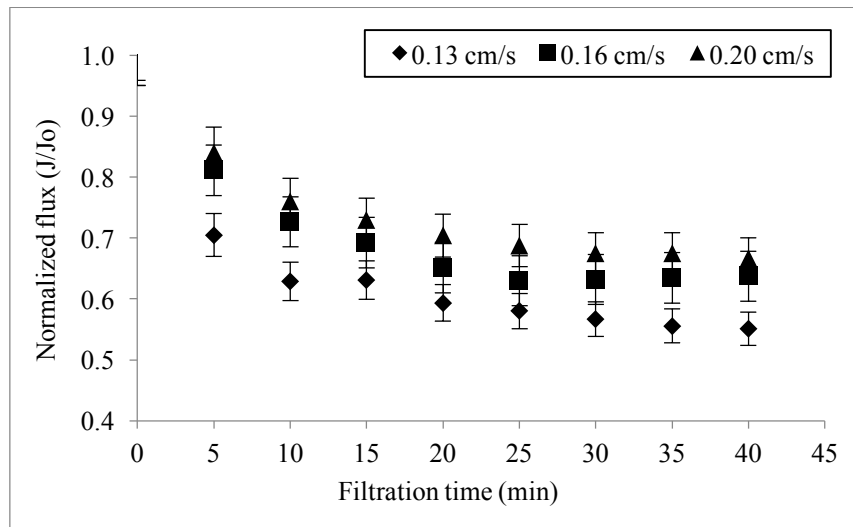


Figure 4: Effect of cross flow velocity on membrane flux

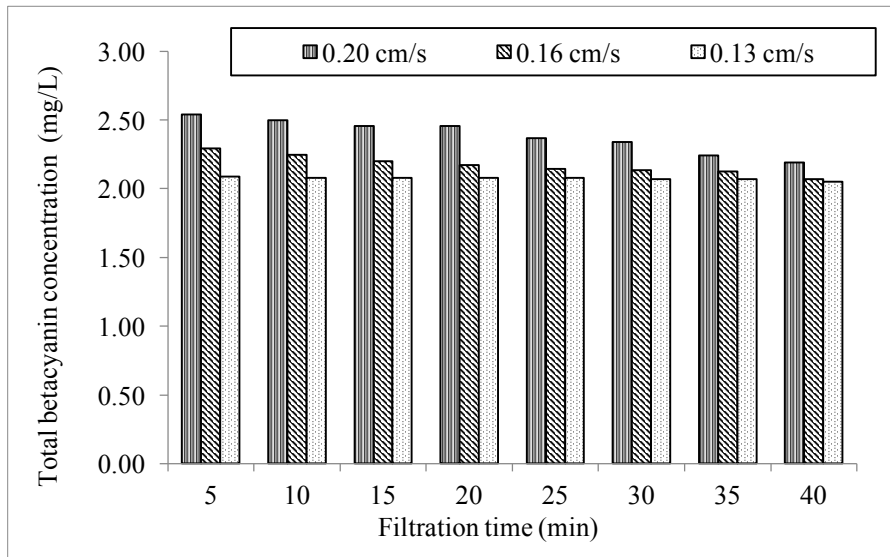


Figure 5: Effect of different CFVs on total betacyanin concentration

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