Determination of ultrafiltration resistance using series resistance model in inulin purification from red fruit (Pandanus conoideus L.) pedicel extract

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Abstract. The crude inulin extracted from red fruit pedicels using hydrodynamic cavitation contained inulin and other impurities such as reducing sugars and proteins. Membrane technology is an alternative technology for separating inulin from its impurity. This study aimed to purify the inulin extract and to evaluate the membrane resistance using a series of Membrane Titania (5 × 10^3 Da pore diameter) with 1–2.6 bar pressure variation on ultrafiltration. The steady-state on the inulin crude filtration at 0 scales and 1 bar occurred at the 25th minutes with a flux value 3.06 L m⁻² h⁻¹. The optimum pressure for the transmembrane was at 2.2 bar with permeate flux of 7.75 L m⁻² h⁻¹. The amount of flux in the membrane filtration was determined by the resistance of internal membrane, fouling, and polarization.

1. Introduction
Inulin is a fructans compound consisting of oligosaccharides or fructose-containing polysaccharides with linear β-2,1 bond and one glucose terminal unit [1]. Fructans, type of inulin and their derivatives are potential and widely used in the food, feed and pharmaceutical industries [2]. Chicory (Cichorium intybus) and Jerusalem artichokes (Helianthus tuberosus) are the inulin raw material on an industrial scale, which each contains 15–20% and 14–19% inulin, while domestic sources only come from Dahlia and Gembili tubers [3–5]. Research [6] has proven that red fruit pedicels (Pandanus conoideus L) which are by-products from the red fruit oil industry have the potential to be a source of raw materials with 3.11% (b. k) inulin content. Pedicel is a by-product of red fruit oil production with the highest percentage of weight (51–61%) compared to drupa (39–49%) and seeds (27–36%) [7]. At present, pedicel has not been used only as animal feed and only disposed of.

The crude inulin extract (filtrate) from several sources and extraction methods, is reported to contain impurities such as carbohydrates, reducing sugars, proteins, gums, fibers, and pigments [8–11]. These impurities can affect the inulin quality, especially in the drying process. The previous study [11] reported that agglomeration in purified inulin powder was lower compared to inulin powder without purification. Therefore, a purification process is needed to separate the inulin from its impurity so the quality improved.
The conventional purifying method requires a long stage (calcification, one carbonation stage, one filtration stage, two carbonation stages, and two filtration stages) and needs high temperature (80–90 °C) [8,10]. On the other hand, the high temperatures caused hydrolysis in inulin molecules which affect their functional properties [12]. Therefore, membrane technology is one alternative method for inulin purifying with the advantages of high product quality, high productivity, and low costs [9, 13, 14].

However, there is a major problem in membrane filtration, such as the emergence of fouling and concentration polarization, which can reduce the performance of membrane fluxes. Fouling is affected by membrane characteristics and their interaction with the filtered material, while the polarization intensity is influenced by the operating parameters such as transmembrane pressure, flow rate, and temperature [15,16]. The formation of the particle layer becomes a barrier to the permeate flow rate, which is characterized by a decrease in flux. The relationship can predict the flux performance and membrane characteristics hence the occurrence of fouling phenomena and concentration polarization can be approached using a quantitative model, such as the series resistance model [16]. This model uses the principle of forming layers on the surface and inside the membrane, which will cause certain resistance and contribute to total membrane resistance including internal membrane resistance (Rm), fouling resistance (Rf) and polarization resistance concentration (Rp).

The research on the component of crude inulin extract from red fruit pedicels and its refining process has never been reported. This study aimed to (1) separate inulin in crude extracts of red fruit pedicels from impurity components using ultrafiltration membranes with a pore size of 5 × 10⁻⁷ Da; and (2) evaluate the phenomena of fouling and concentration polarization formation on the purification of crude fruit red inulin pedicel extract using an ultrafiltration membrane.

2. Materials and methods

2.1. Materials and equipment
The raw material was red fruit obtained from the experimental garden of Papua University (UNIPA) Manokwari. The chemicals used were standard inulin (Merck), water for HPLC as the mobile phase, H₂SO₄, NaOH, Na₂SO₄, CuSO₄, H₃BO₃, methyl red and distilled water indicators. The equipment used were HPLC (High Performance Liquid Chromatography) Shimadzu type UFLC Prominence LC 20 AD with Aminex ion exclusion column (250 × 4.6 mm), refractive index detector, hydrodynamic cavitation extractor circuit equipped with Showfou 0.5 hp centrifugal pump, ultrafiltration ceramic membrane Titan 5 × 10⁻⁷ Da with diameter 0.7 cm and length 25 cm obtained from MEMBRALOX™ and some glassware for preparation and analysis needs.

2.2. Extraction and analysis of inulin filtrate
The extraction process began with the preparation of red fruit pedicel powder. The preparation consisted of pedicel separation from the red fruit Drupa, then washing with running water to remove impurities. Furthermore, red fruit pedicels were chopped with a thickness of ± 5 mm and dried using sunlight to obtain ± 5% moisture content. After drying, the size reduction was conducted using a hammer mill with a size of ± 80 mesh.

The inulin extraction used the hydrodynamic cavitation method with a venturi device equipped with nozzles and throat each 4 mm in diameter and 100 mm in length. The extraction process was carried out using 650 mL of liquid (a mixture of distilled water and powdered powder) with material to solvent ratio of 1:50 (b/v), circulated by the pump through the extraction tank following the main path for 15 minutes with an initial temperature of ± 28 °C. The flow on the hydrodynamic cavitation device consisted of two lines, namely the mainline and the by-pass line. The by-pass line was intended to control the flow rate of the liquid through the mainline, but because the flow rate of the pump was fixed, the by-pass line was not used. The fluid flow rate was generated by a centrifugal pump and was made constant, while the gas flow rate was fixed at 4.5 LPM. The gas used was UHP nitrogen (Ultra
High Purity). Analysis of the chemical components of crude red pedicel inulin filtrate included total inulin [17] and total N (Kjeldahl method) [18].

2.3. Inulin purification using membrane filtration
At this stage, the membrane permeability, the steady-state, and the influence of the pressure on the flux were determined. Membrane permeability was determined by circulating pure water, whereas the steady-state and the influence of transmembrane pressure on flux were determined using crude inulin extract. The circulated feed passed through the membrane as much as 300 mL by regulating the transmembrane pressure and flow rate. The system was pressurized with nitrogen gas and the temperature maintained constant at room temperature of 30 ± 1 °C. The operating flow rate was adjusted using the scale on the membrane device (0–9).

Determination of membrane permeability was carried out at 0 scale flow rate in transmembrane pressure (1–1.8 bar). The flux measurements were carried out by measuring the amount of permeate produced for 5 minutes. The steady-state determination was carried out in a 1 bar transmembrane pressure operation on 0 scales at several times t (minutes) until the steady-state was reached. The flux measurement at steady-state determination was carried out every 5 minutes. The time when flux starts was constant, and it's used in the next process to determine the performance of the membrane. The flux was operated in a 1.2–6 bar the transmembrane pressure range at 0 scales to determine the influence of the pressure on. For all treatments, permeate was taken to measure its flux after a steady-state was reached and the retentate was returning to the feed tank.

After a membrane process was completed, the membrane was washed with pure water and a solution containing 1% NaOH at 50 °C for 15 minutes with the permeate channel close before continuing to the next operation. After washing with 1% NaOH, the membrane was washed by circulating pure water until the washing water was neutral. Furthermore, the membrane flux was measured again using pure water at 1 bar transmembrane pressure.

3. Results and discussions

3.1. Characteristics of crude inulin extract
The crude inulin extract was in the form of a cloudy brown filtrate extracted by the hydrodynamic cavitation method. The crude inulin extract contained 3.57% inulin, 1.12 reducing sugars, and 3.34% proteins (table 1).

Table 1. Components of inulin crude extract.

| Component    | Amount (% d. b) | Molecular size (Da) |
|--------------|-----------------|---------------------|
| Inulin       | 3.57            | 5.0 × 10^2–1.3 × 10^3 |
| Reducing sugar | 1.12          | 1.8 × 10^2         |
| Total N      | 3.34            | 1.4 × 10^4–6.6 × 10^4 |

A study by [20] showed that purification of inulin from chicory extract with 50 × 10^3 Da pore size ultrafiltration membrane produced purity of 90.9%, whereas 5 × 10^3 Da membrane produced less pure filtrate (85.5%). The purity was calculated as the mass percentage ratio of inulin to the total soluble solids (◦Brix) of the filtrate or juice. The average molecular weight of Jerusalem artichoke and Chicory ranges between 3.4 × 10^3 and 6.2 × 10^3 Da [21], the use of a 5 × 10^3 Da membrane makes many inulin molecules cannot pass. However, in the case of a 5 × 10^3 Da membrane, a protein with a molecular weight of more than 1.4 × 10^5 Da (table 1) does not pass through the membrane because the protein can reduce the quality of the inulin.

3.2. Determination of membrane permeability
Membrane permeability is a parameter that determines the ability of the membrane to pass the fluid, and the value depends on the nature of the membrane. Figure 1 shows the relationship between flux
and transmembrane pressure with pure water as feed. An increase in transmembrane pressure from 1 to 1.8 will increase the bar flux linearly from 6.55–13.54 L m⁻² h⁻¹. The results showed that permeate flux increased with increasing transmembrane pressure, which also showed an increase in membrane permeability. Under Darcy's law, the flux will increase in proportion to the increase in membrane pressure given [22]. The membrane permeability is influenced by its pore size hence the larger the pore, the higher the permeability. A study [23] showed that the membrane permeability of 5 nm and 10 nm pore size in the separation of beta-carotene and alpha-tocopherol palm oil in isopropanol were respectively 1.2921 L bar⁻¹ m⁻² h⁻¹ and 4.0099 L bar⁻¹ m⁻² h⁻¹.

**Figure 1.** The relationship between flux and transmembrane pressure with pure water as feed.

Besides, an increase in flux is proportional to the increase in transmembrane pressure, which can be affected by the nature of the hydrophilic membrane. Permeability measurements in this study were obtained by passing pure water on the ceramic membrane made from titania. Titanium dioxide ceramic membrane (TiO₂) resistances to acid and various types of solvents, stable at high temperatures, insoluble in water, and has a preference for water (hydrophilicity) [24]. Hydrophilic feed ideally uses hydrophilic membrane material (water-attracting) [15].

3.3. **Determination of the steady state**

Steady-state is the time required by each membrane to obtain a constant flux value in one particular condition and reached in a few minutes after the operation takes place. Figure 2 shows that steady-state on filtration of crude inulin extract at 0 scales in 1 bar occurred in the 25th minute with a flux value 3.06 L m⁻² h⁻¹. At the beginning of the filtration process, the permeate flux decreases rapidly and then gradually decreases until steady conditions are reached. A sharp decrease in flux after the ultrafiltration process begins through the membrane feed and membrane interactions will occur due to fouling [25].
3.4. The influence of pressure on flux
The examination of the effect of transmembrane pressure on flux was carried out on 5 × 10^3 Da membranes at a pressure range 1–2.6 bar. Figure 3 shows that the changes in flux values tend to increase with an increase in transmembrane pressure of 1–2.2 bar at 3.17 to 7.75 L m⁻² h⁻¹, then at 2.2 bar until 2.6 bar the flux tends to stable at 7.75 L m⁻² h⁻¹. At 2.2 bar, the value of flux obtained is called limiting flux, which is an increase in pressure that did not affect the permeate flux. The minimum pressure in the limiting flux area can be expressed as the optimum transmembrane pressure.

Figure 3. The relationship between flux and transmembrane with inulin extract as feed.

Transmembrane pressure is the driving force for a feed at the membrane surface, but at certain pressures after reaching a certain point, the flux value is no longer influence the increase in flux [23, 26]. At low pressure, there is no material accumulation on the membrane surface so that the flux is directly proportional to the transmembrane pressure, but if the pressure increases it will increase in material accumulation on the membrane surface resulting in fouling and flux no longer increases linearly with an increase in transmembrane pressure [27]. The decrease in initial flux is associated with the membrane-solute interactions and correlates with the fouling model, and at stable flux conditions, there is an increase in the concentration of solutes in the feed stream and is correlated with a concentration polarization mechanism [25].
3.5. Determination of membrane resistance components with series resistance models

The series resistance model is an approach to predict flux whose performance can decrease because of the particle layer on the membrane surface prevent flow rate. In this model, the membrane resistance observed was internal membrane resistance ($R_m$), fouling resistance ($R_f$) and concentration of polarization resistance ($R_p$) [28]. Internal membrane resistance ($R_m$) is determined using pure water as feed and calculates the gradient value of the relationship between pressure 1/transmembrane (1/∆P) and 1/flux (1/J). Data on transmembrane pressure and flux is shown in figure 1. Figure 3 shows the relationship between transmembrane pressure and flux with inulin extract as feed. The results showed that: 1) flux increased with an increase in transmembrane pressure at $1$–$2.2$ bar, and 2) flux tended to be stable with an increase at $2.2$–$2.6$ bar. The gradient value of the relationship between 1/transmembrane pressure (1/∆P) and 1/flux (1/J) in the flux area will increase along with the increase in transmembrane pressure so that the $R'm$ value (the sum of the internal membrane resistance and fouling resistance) will be obtained. Fouling resistance is expressed as a reduction of $R'm$ to $R_m$.

The polarization resistance index concentration ($\phi$) obtained by calculating the gradient of the relationship between 1/transmembrane pressure (1/∆P) and 1/flux (1/J) in the area where there is no increase in flux as the transmembrane pressure increases. The $R_p$ value was determined by multiplying the polarization resistance concentration index with transmembrane pressure ($R_p = \phi \Delta P$), so that, the series prisoner model can be expressed as in the equation:

$$J = \frac{\Delta P}{R'm + R_p}$$  \hspace{1cm} (1)

$$R'm = R_m + R_f$$  \hspace{1cm} (2)

$$R_p = \phi \Delta P$$  \hspace{1cm} (3)

4. Conclusions

The Steady-state on inulin crude extract filtration in 1 bar at 0 scales occurred in the 25th minute with a flux value of 3.06 L m$^{-2}$ h$^{-1}$. The optimum purification of crude inulin extract occurred at a transmembrane pressure of 2.2 bar with permeate flux 7.75 L m$^{-2}$ h$^{-1}$. The Series resistance models measured and calculated the values and components of membrane resistance, including internal membrane resistance ($R_m$), fouling resistance ($R_f$) and polarization resistance concentration ($R_p$) based on the relationship between transmembrane pressure (1–2.6 bar) and flux with crude inulin extract as feed.

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