

Article

Low-Fouling Plate-and-Frame Ultrafiltration for Juice Clarification: Part 2—Module Design and Application

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Abstract: The purification and concentration of orange juice are crucial to remove undesirable materials, such as pectin, which is responsible for juice clouds; or limonene, which is responsible for bitter taste. Membrane-based juice clarification is preferred due to its capability to separate specific targeted molecules, while still maintaining the clarified juice's nutritional content. In this study, a novel designed bench-scale plate-and-frame membrane module composed of low fouling cellulose acetate membrane sheets was manufactured to facilitate orange juice clarification. The experimental results demonstrated the effectiveness of the developed module to be used for juice clarification. After incorporating the functional and structural design parameters, the final module had the following specifications: dimensions of 125 × 168 mm, an effective volume of 0.9–9.4 L, a total active membrane area of 1088 cm², and a transmembrane pressure of 0.3–0.55 MPa. The results of the juice clarification show no difference in the value of pH, viscosity, total acid, water content, color L* (brightness), and color a* (reddish) of the feed, the permeate, and the retentate streams. The clarified juice had slightly higher total dissolved solids (°Brix), ash content, vitamin C, and color (b* yellowish). Overall, our findings demonstrated that the developed plate-and-frame module could effectively be used to clarify orange juice without altering the quality, i.e., reducing the nutritional contents.

Keywords: composite membrane; anti-biofouling; plate-and-frame module; juice clarification; stability; vitamin C



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1. Introduction

Fruit juice is a drink made from the extraction of the natural liquid contained in fruit, which contains vitamins and antioxidants that are beneficial for health [1]. A popular example of fruit juice is orange juice, which contains several micronutrients, such as vitamin C, folate, and polyphenols [2]. Orange juice is a natural source of antioxidants, including flavonoids (hesperidin and naringenin predominantly as glycosides), carotenoids (xanthophylls, cryptoxanthin, and carotenes), vitamin C, and folate [3].

Orange juice generally has a pale yellow color, and it contains pectic and cellulosic substances that cause turbidity, which requires a clarification process [4]. Decantation is a simple way to clarify an orange juice, but it is less effective because it requires a long period [5]. Centrifugation is much faster than clarification, but it consumes high energy and cost [6,7]. Filtration has gained attention in regard to orange juice clarification due to its simple operation, and especially due to the ability to tune the filter's pore size according to the process objective [8,9]. The aroma of the fruit juice will not disappear during the concentration process using a membrane [10]. Besides turbidity removal, juice clarification

should also consider the possible removal of antimicrobial components of orange juice, such as the essential oils present in the cloud fraction [11].

The quality of orange juice is influenced by several factors: low levels of compounds that cause a bitter taste, soluble solids, acidity, Brix/acid ratio, color, and flavor [12]. In fruit-juice processing, purification and concentration processes play essential roles that dictate the characteristics of the produced fruit juices, such as texture, taste, color, solutes, decreased water levels, and an increase in essential substances, such as vitamin C.

Membrane technology is a potent approach for clarifying and concentrating juices, and it offers several advantages over traditional separation processes [13]. In the food-processing industry, membrane technology has been widely applied, especially in clarifying and concentrating agricultural-based beverages. Compared to other conventional technology (such as evaporation), the membrane process offers improved product quality, requires less energy, has a high efficiency, requires a short processing time, and operates in the ambient temperature. The concentration by evaporation process can reduce the quality of the produced juice due to heat exposure, which damages the nutrition [14]. Ultrafiltration and microfiltration membranes with tangential or crossflow filtration can replace conventional filtration methods for clarifying fruit juices [15,16].

Membrane filtrations for clarification and concentration of fruit juices have been carried out, e.g., orange juice with polymeric membranes (polyvinylidene difluoride/PVDF and poly(methyl methacrylate)/PMMA) [17], bitter orange juice with mixed cellulose ester membrane [18], apple juice with polysulfone and polyethyleneimine membranes [19], prickly pear juice with polysulfone membrane [20], orange juice with polyether sulfone and polysulfone membranes [21], passion fruit juice [8], commercial and pure orange juice [22], and citrus lemon juice [23]. Membranes can be applied to clarify and concentrate apple, strawberry, orange, black currant, pear, hawthorn, pineapple, and date juices. Clarification by membrane produces high-quality products with low operating costs and minimum use of an additive. However, clarification using the membrane suffered from membrane fouling [24–26].

This study explores the design and application of a plate-and-frame membrane for juice clarification. The plate-and-frame module was selected for its simplicity and facile assembly from a flat sheet membrane. The elements of plate-and-frame membrane are flat sheet membrane. The membrane used in this study was predeveloped from cellulose acetate (CA) polymer and phenolic substances from garlic bulb to pose a low biofouling vulnerability. Cellulose acetate membranes enriched with phenolic substances derived from garlic bulbs have been prepared and showed low-fouling properties against bacterial adhesion [27]. The module components include a support plate, spacer, and feed distribution plate [28]. A plate-and-frame module configuration includes sets of two membrane sheets sandwiching a spacer that acts as permeating channel. A plate-and-frame stack consists of several membrane sets that are sealed by using rings and end plates [29]. The module provides low packing density and easy assembly without requiring delicate gluing. A plate-and-frame module is also superior to other modules in resisting particulate blocking during filtration, considering the high concentration of particulates in orange juice [30].

The first commercially successful ultrafiltration systems were based on tubular modules and plate-and-frame modules. Plate-and-frame units compete with tubular units in several applications. These modules are not as resistant to fouling as tubular modules but are less expensive. Most consist of a flat membrane envelope with a rubber gasket around the outside edge. The membrane envelope, together with the appropriate spacers, forms the plates contained in stacks of 20–30 plates. Typical feed channel heights are 0.5–1.0 mm, and the system operates under high-shear conditions [31].

The advantages of plate-and-frame systems are that they can be used for highly fouling solutions and can operate at high temperatures with relatively aggressive feed solutions, conditions under which spiral-wound modules might fail. Furthermore, it can also operate at higher pressures than tubular or capillary modules—operating pressures up to 150 psi are not uncommon. The compact design, small holding volume, and no stagnant areas make

sterilization easy. For this reason, plate-and-frame units are used in several food-industry operations, e.g., in the production of cheese, in the production of apples and other juices, and more recently, in the production of beer and wine [32,33].

The drawbacks of plate-and-frame units are that they are expensive compared to the alternatives and that there is leakage through the gaskets required for each plate, creating a serious problem [31]. Moreover, the plate-and-frame membrane module requires the product stream to spread across the entire surface of the individual sheets prior to recirculation. This causes the flow path to be non-uniform and slower, thus accelerating the occurrence of concentration polarization (laminar). Consequently, it is not directly scalable [34]. In addition, the process of cleaning the plate-and-frame modules of the membrane is quite time-consuming. Equipment needs to be removed to perform cleaning of each membrane after the clarification process [35].

In this study, a plate-and-frame membrane module was selected and designed for orange juice clarification. The optimal design of a plate-and-frame module is required to enhance the filtration efficiency. The design of the plate-and-frame module in this study uses ear bolts to facilitate the disassembling process when cleaning or changing the membrane. On top of that, the design of this module is equipped with two straight-blade impellers that provide radial flow to help spread the material across the membrane surface and reduce the formation of fouling. The ease of manufacture and production is a distinct advantage of such a straight-blade impeller when compared with other impellers [36]. The scope of this study was limited to assessing the impact of the membrane-based clarification on the quality of the produced orange juice. The hydraulic performance, especially related to membrane fouling, was not addressed in this work.

2. Materials and Methods

2.1. Materials

A predeveloped CA-based flat-sheet membrane was employed in this study. The membrane was optimized to minimize biofouling by incorporating phenolic substances from garlic extract; the details can be found elsewhere [27]. Poly(methyl methacrylate) plates were used as supported plate and frame, holding baffle, feed container, and electronic control box (power supply 5 A, 12 V; potentiometer, on/off controlled system). Silicon rubber was used as the rings and gasket for the membrane stack to prevent leakage. A nylon-based 100 mesh spacer was used as a spacer to set two membrane sheets apart.

A straight-blade impeller was installed to induce fluid radial flow, provide fouling control, and prevent sedimentation of juice component in the bottom of feed container. The impeller was powered by a motor reduction gear turbine worm self-locking endorser signal feedback with DC 12 V/150 rpm. A diaphragm vacuum pump (60 W, 12–24 V, 1.6–3.0 A, max flow 3.6 L/min) was used to suck liquid from the feed juices, and vacuum suction was measured by a manometer (vacuum: -1.5 – 0.5 bar pressure). All materials were of food-grade quality.

2.2. Module Design

The plate-and-frame module was designed by using AutoCAD software (Autodesk, San Rafael, CA, USA). Two design approaches, namely structural and functional, were conducted. The functional states that a design is a function of its probability of successfully achieving the required functional requirements and constraints. Figure 1 shows schematic overview of the designed plate-and-frame module.

Meanwhile, the structural states that a design is a function of its representation and provides quantitative measures [37]. The goal was to develop an effective instrument to control, contain, and support the juice clarification process. Liquid–solid transfer, sterility, applied pressure, and other practical considerations were considered. The module was fabricated on a laboratory scale, considering the requirement for a proof of concept. Further consideration is required to provide an upscaled membrane module customized for fruit juice clarification.

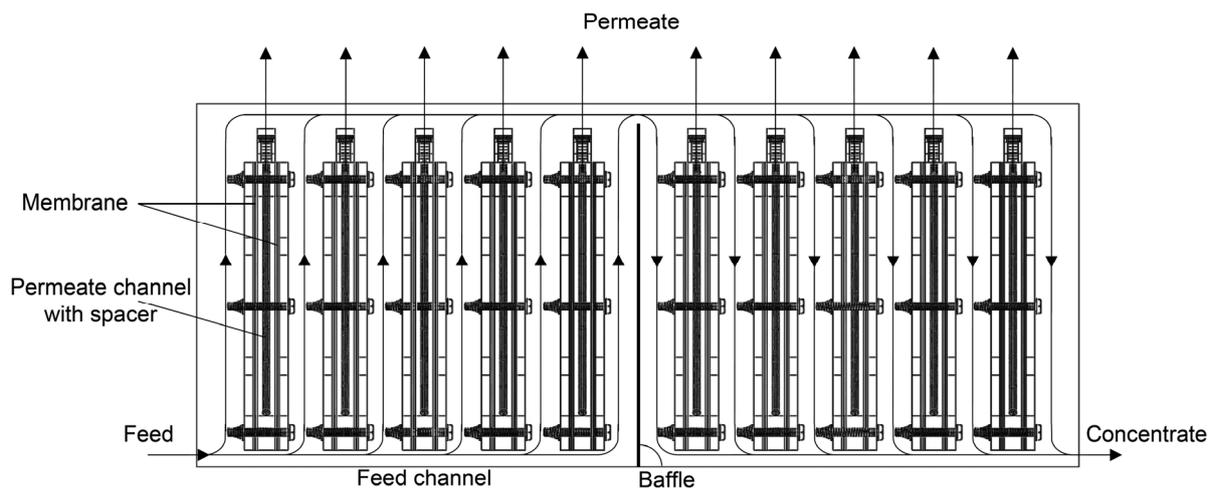


Figure 1. Schematic overview of the designed plate-and-frame membrane module.

2.2.1. Plate-and-Frame Elements

The plate-and-frame elements were from PMMA plate, with the size of 125×168 mm, as shown in Figure 2. The size of the membrane frame was 125×168 mm, in rectangular shape, and two holder plates were located on the side (Figure 2a). Two membrane sheets were separated by a spacer sheet, also acting as the permeate flow channel (Figure 2b,d).

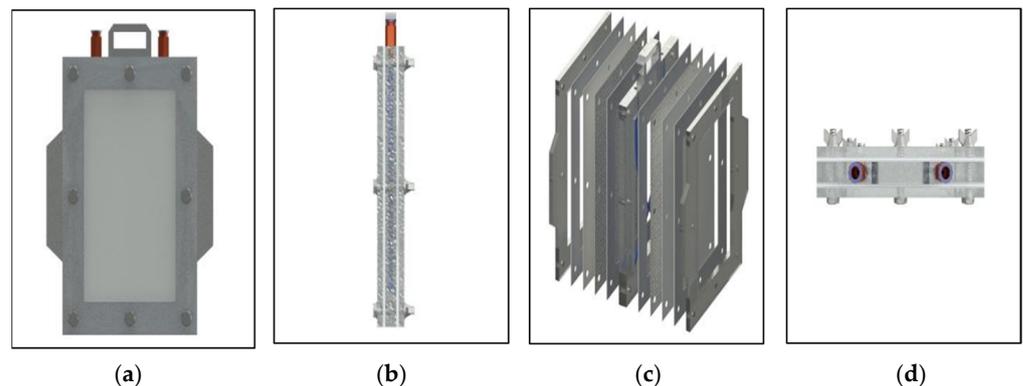


Figure 2. Membrane plate-and-frame element: (a) front view, (b) side view, (c) isometric, and (d) top view.

The active area of the membrane sheet and spacer sheet was 85×128 mm (Figure 3a,b). The silicon rubber had a similar size, but only in the perimeter of the plate (Figure 3c). The membrane element configuration was as follows: front plate–silicon rubber–membrane sheet–spacer–silicone rubber–middle plate–silicone rubber–spacer–membrane sheet–silicone rubber–rear plate (Figure 2c).

The front and the rear plates acted as the cover and the support for the configuration, while the middle plate acted as the support for two 4 mm diameter outlet tubes, as shown in Figure 4. The 4 mm diameter tube was connected to a 6 mm diameter tube to collect the permeate from each membrane element. The plate-and-frame elements were fitted with 8 ear bolts.

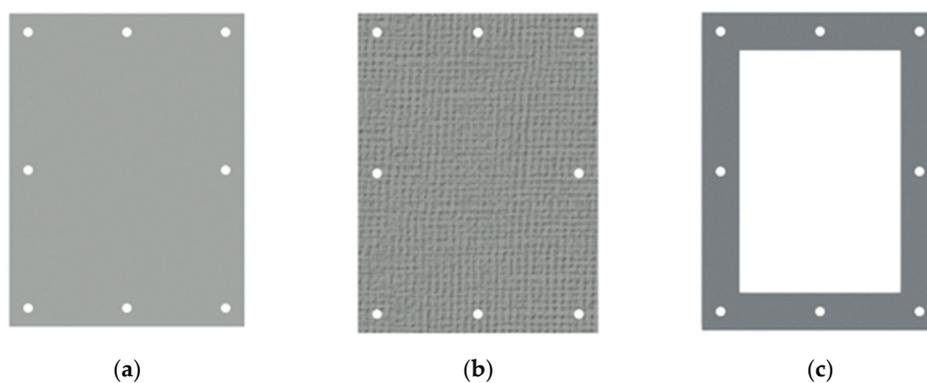


Figure 3. (a) Membrane sheet, (b) spacer sheet, and (c) silicon rubber.

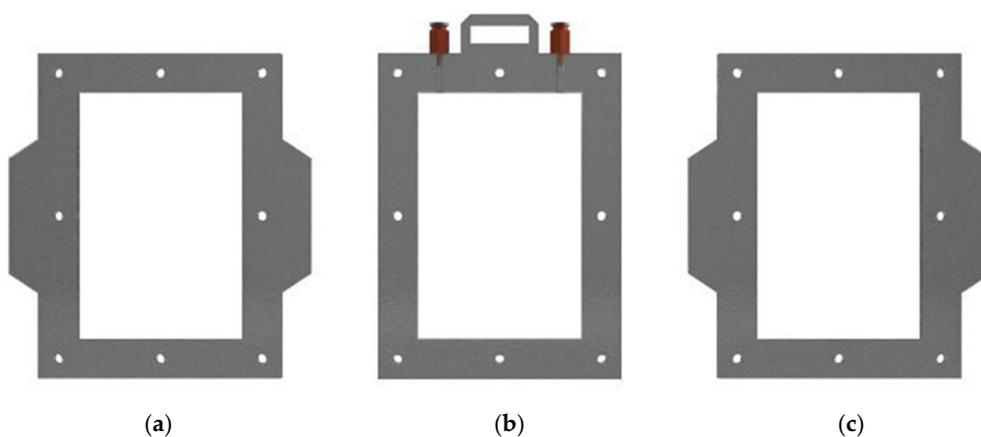


Figure 4. Membrane plates: (a) front plate, (b) middle plate, and (c) rear plate.

2.2.2. Membrane Module

As shown in Figure 2, five plate-and-frames were stacked, connected, and combined into a module, as shown in Figure 5. The plate-and-membrane elements were placed in the feed container. The membrane stack was fully immersed in the feed solution. The feed container capacity was 9–12 L, with the dimension of $345 \times 185 \times 220$ mm (Figure 5c). The permeate tube was used to collect permeates from each plate-and-frame membrane element (Figure 5d).

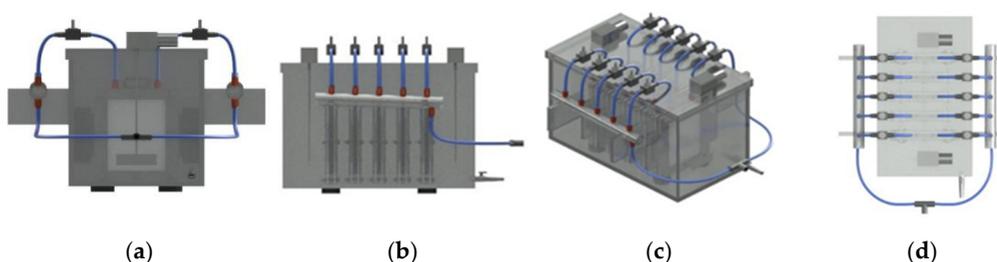


Figure 5. Membrane module: (a) front view, (b) side view, (c) isometric, and (d) top view.

Two straight-blade impellers, with a blade size of 60×20 mm thickness 1 mm and height of 210 mm, were installed in the left and the right side of the module stack to promote mixing and prevent decantation of the solid components in the bottom of the feed container (Figure 6).



Figure 6. A straight-blade impeller.

2.3. Orange Juice Clarification

The plate-and-frame module was used to clarify the orange juice. Orange juice was obtained from a local farmer in Malang city, East Java, Indonesia. The juice clarification was conducted under dead-end mode, with a suction pressure of -0.2 bar, a feed volume of 12 L, and 2 membrane elements (4 membrane sheets). The schematic diagram of the orange-juice clarification process is shown in Figure 7.

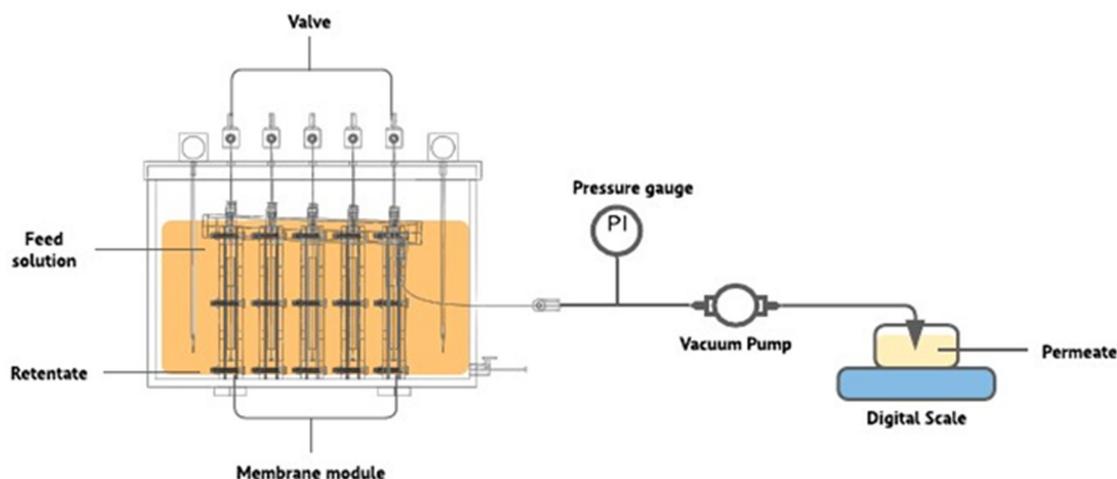


Figure 7. Schematic diagram of orange juice clarification and concentration using a plate-and-frame ultrafiltration membrane system.

The orange juice clarification was conducted based on the modification of previous research [38]. Some main parameters were assessed. They included water content, ash content, pH (pH Meter, Crison type 20, Barcelona, Spain), viscosity (Vibro viscometer, AND/SV-10, Tokyo, Japan), total soluble solids ($^{\circ}$ Brix) with an Abbe digital refractometer (WAY 2S, Wincom Company Ltd., Hunan, China), total acidity titrated (NaOH 0.1 N, with phenolphthalein as an indicator), vitamin C (by using titration), and color with a color reader (Minolta, Tokyo, Japan) [39].

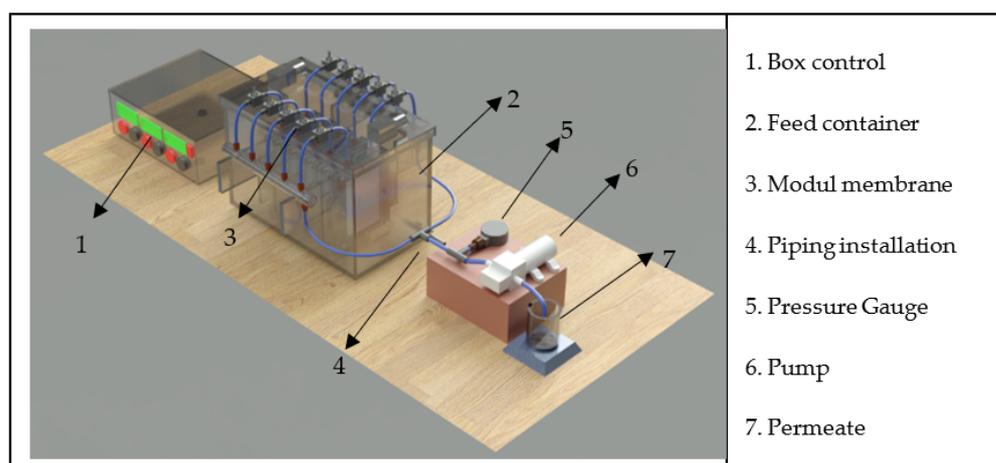
3. Results and Discussion

3.1. Plate-and-Frame Module

Details of plate-and-frame membrane specifications are summarized in Table 1, and the schematic of the plate-and-frame module and filtration membrane system is shown in Figure 8.

Table 1. Plate-and-frame membrane specification.

No.	Instruments	Specification	Dimensions
1.	Feed container	Capacity: 12 L	345 × 185 × 220 mm
2.	Membrane process	Batch ultrafiltration using a submerged module	-
3.	Type of membrane	Flat sheet	Active area: 85 × 128 mm Thickness: 0.3 mm
4.	Membrane additive	Phenolic compound from garlic extract	-
5.	Plate and frame	5 elements	125 × 168 mm with a thick cover of 5 mm and the middle 10 mm
6.	Food-grade nylon spacers	10 sheets	100 mesh (125 × 168 mm) Thickness: 0.1 mm
7.	Food-grade silicon rubber	20 sheets	125 × 168 mm Thickness: 1 mm
8.	Straight-blade impeller	2 pieces	Blade: 60 × 20 mm Height: 202 mm
9.	Manometer	1 piece	−1–0.5 bar
10.	Diaphragm pump	Voltage: 12–24 V Max Power: 60 W Current: 1.6–3.0 A Max Flow: 3.6 L/min Max Pressure: 0.3–0.55 MPa Max Liquid Temperature: 100°	157 × 100 × 60 mm
11.	Electronic box control	- Push ON/OFF AC: 1 piece - Push ON/OFF DC: 3 pieces - Potentiometer: 3 pieces - LCD V/A: 3 pieces - Power supply: 5 A; 12 V	280 × 240 × 100 mm

**Figure 8.** Plate-and-frame module and membrane filtration systems.

3.2. Clarification of Orange Juice

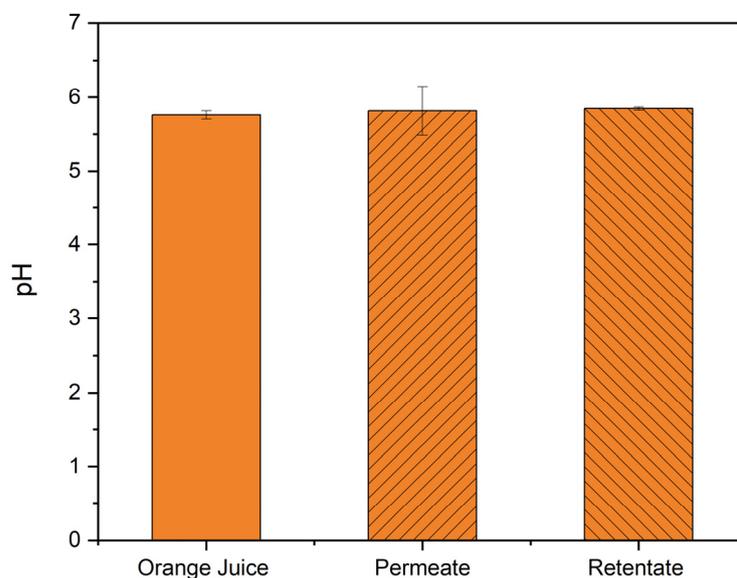
Some of the molecular weights of the constituents of the orange-juice feed are summarized in Table 2. The soluble solids in orange juice consisted of several components, e.g., carbohydrates, 76%; organic acids, 9.6%; free amino acids, 5.4%; inorganic ions, 3.2%; vitamins, 2.5%; liquid constituents, 1.2%; nitrogen-based and glutathione, 0.9%; flavonoids, 0.8%, volatile constituents, 0.38%; carotenoids, 0.38%; and enzymes, 0% [40]. The fruit juice was clarified by using a predeveloped membrane detailed in the previous study [27].

Table 2. Typical orange juice components and their molecular weight.

Orange Juice Content	Molecular Weight	Source
Pectin commercial (Hereford UK)	212,000 Da	[41]
Pectin Methyl Ester	54,000 Da	[42]
Protein	12–72 kDa	[43]
Citric Acid	210.14 Da	[44]
Citric Acid	192.12 Da	[44]
Ascorbic Acid	176.12 Da	[45]
Carotenoid	536.87 Da	[46,47]
Malic Acid	134.09 Da	[48]
Benzoic acid	122.22 Da	[49]
Oxalic Acid	90.03 Da	[50]
Tartaric Acid	150.09 Da	[51]
Succinic Acid	118.09 Da	[52]
Fructose	180.16 Da	[53]
Sucrose	342.30 Da	[53]
Glucose	180.16 Da	[53]

3.3. pH Value

The measurement of the pH of the orange juice clarification by a plate-and-frame membrane module is shown Figure 9. The pH values of the feed, the permeate, and the retentate were similar. As reported elsewhere, the pH value and total acid did not significantly change due to the filtering process that used an ultrafiltration membrane [21]. Acids' molecules could pass through the pores of the membrane. Hence, the feed and the permeate pH values were expected to be similar. In addition, the pH value increased due to the higher acid value gained from long storage, as reported earlier [38].

**Figure 9.** The pH of the feed (orange juice), the permeate, and the retentate streams.

3.4. Viscosity

The measurement of the viscosity of orange juice clarified by the developed plate-and-frame module is shown Figure 10. The viscosity value of the clarified juice decreased after the clarification via ultrafiltration. The decrease in viscosity can be attributed to the removal of suspended solids from the feed. On the other hand, the viscosity value increased with the decreasing amount of water content (due to concentration). The decrease in viscosity for the retentate stream can be attributed to the accumulated suspended solid in the system or the attachment on the membrane surface.

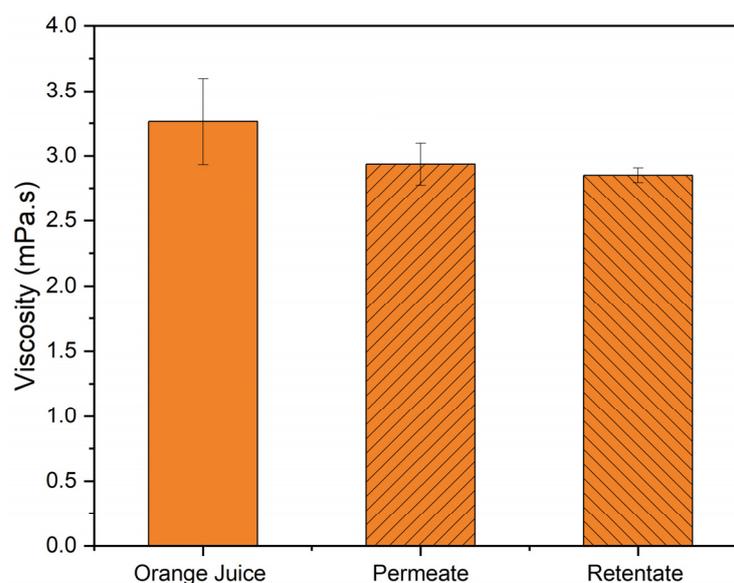


Figure 10. The viscosity of the feed (orange juice), the permeate, and the retentate streams.

Table 2 shows that the molecular weight of the pectin in orange juice was 212,000 and 54,000 Da, and the protein was 12–72 kDa, while the pore size of the membrane and MWCO was smaller. The rejection of pectin was then expected and caused a gel formation on the surface of the membrane. In addition, the carbohydrate content (sucrose, glucose, and fructose) could pass through the membrane pores.

The viscosity and density of the filtered juice (permeate) were significantly reduced due to the membrane's retention of the suspended solids, such as orange pulp and pectin materials, as reported elsewhere [21,54]. The retention of some juice components on the membrane increased the rejection by forming a dynamic membrane in the form of a cake layer. The cake can later undergo gelation, induce concentration polarization, and increase the filtration resistance [55]. The decrease in the permeate's viscosity can enhance the filtration's hydraulic performance [56]. An addition of cyclodextrin can increase the viscosity, while cellulose acetate decreases it. Cyclodextrin is soluble in the juice, while cellulose acetate is insoluble.

A previous study [57] employed two membranes with an MWCO of 30 and 100 kDa to clarify pineapple juice in addition to an enzymatic. It was found that there was an insignificant difference in the pH, acidity, total soluble solids, total solids, total sugar, and reducing sugar between. However, the suspended solids, viscosity, and color decreased significantly in the presence of enzymes [57]. The change in the property of the juice product was due to the initial treatment of pineapple juice with enzymes, causing pectin degradation and decreasing viscosity. The viscosity of the juice was almost constant when the total soluble solids were increased as a result of membrane-based clarification [58].

3.5. Total Soluble Solid

The total soluble solids for the feed, permeate, and retentate streams during the orange juice clarification by plate-and-frame membrane are shown Figure 11. The total soluble solid of the retentate was the highest at 9.6 °Brix. This finding suggests that part of the soluble solid was retained by the membrane, considering the formation of additional dynamic layer on the membrane surface. The decrease in °Brix of the permeate would slightly affect the taste of the juice product.

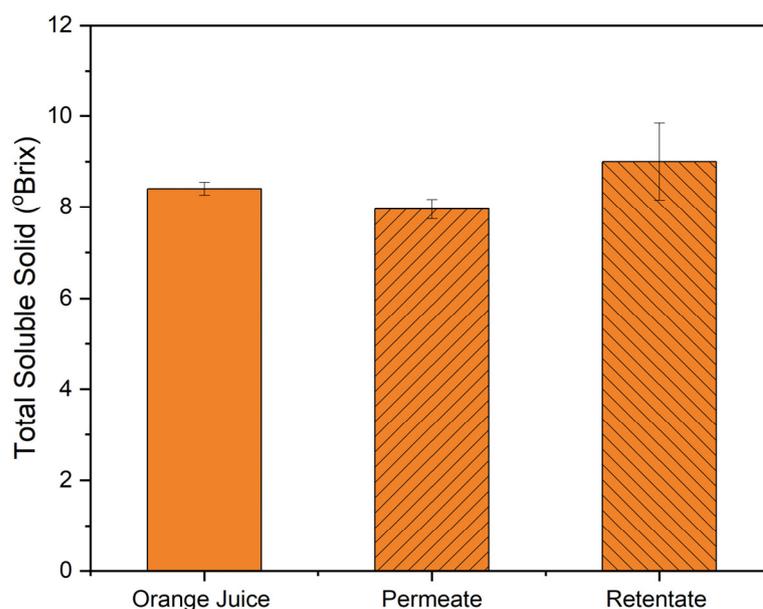


Figure 11. The total soluble solid of the feed (orange juice), the permeate, and the retentate streams.

The total carbohydrates in the orange juice had the highest value in the soluble fraction of 76%, consisting of sucrose, glucose, and fructose (2:1; 1). It was known that the pore size of the membrane was larger than the molecular weight of the carbohydrate materials listed in Table 2, e.g., sucrose (342.30 Da), glucose (180.16 Da), and fructose (180.16 Da). Those carbohydrates were expected to pass through the membrane pore. In addition, the molecular weight of pectin and protein was around the pore size of the membrane; hence, they were partly rejected (up to 90%) and accumulated in the retentate stream. Another study reported that the °Brix value of an orange juice increased by a factor of two by ultrafiltration and increased from 24 to 65 °Brix when using a direct contact membrane distillation [58]. The °Brix of a clarified fruit juice can reach 25–30 through a concentration process, using a reverse-osmosis membrane [59]. The increase in total soluble solids also occurred due to reduced water content due to the clarification process with the membrane. The °Brix in the clarified Valencia orange juice via ultrafiltration decreased slightly [21]. The soluble solids in the clarified orange prickly pear juice with ultrafiltration membrane increased from 10.8 to 11.4 °Brix [20]. Pomelo fruit has a total soluble solids value of 7.14–9.10 °Brix [38].

3.6. Total Acid

Total acid is defined as the total amount of acid that can be titrated. The total acid in orange juice was expressed as citric acid and measured using titration. Total acid has an inverse relationship with pH and taste. The total acids of orange juice clarification by plate-and-frame membrane in the feed, permeate, and retentate streams are shown in Figure 12. The total acid in the retentate was lower than in the feed. The pH value of orange juice greatly affected the total acid value, so the value remained constant or decreased. Data in Table 2 show that several acid molecules were contained in the orange juice sample. The size of citric acid molecules of 192.12 Da was slightly smaller than the nominal size of the membrane pore of 210.14 Da. It was expected to be partly retained, considering the presence of a dynamic cake layer on the membrane surface. Other acids, such as tartaric (150.09 Da), oxalic (90.03 Da), benzoic (122.22 Da), and malic (134.09 Da), were expected to pass through to the permeate stream because their sizes were far below the MWCO of the membrane.

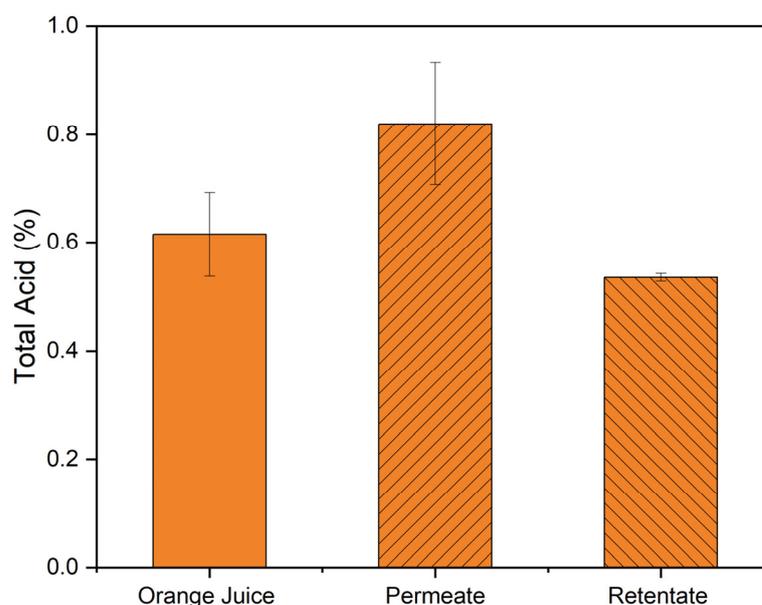


Figure 12. The total acid of the feed (orange juice), the permeate, and the retentate streams.

The decrease in the total acid value of the retentate in this study was attributed mainly to acids with small molecules thoroughly permeating through the membrane and that were probably neutralized or oxidized. For instance, the total acid decreases under prolonged heating at high temperatures due to the oxidation of ascorbic acid. The total acid value (represented by citric acid) was about 0.54%, exceeding the minimum acidity content in orange juice, i.e., 0.35%. Total acid content can affect the pH and the taste [56]. The total acid content of orange juice increased with cyclodextrin and decreased with the addition of cellulose acetate. The acidity of fruit juices in some countries determines fruit ripeness because fruit color is an inferior and unreliable guide [40].

The literature reports on the effect of membrane-based juice clarification are inconclusive. A change in total acid value is affected by several factors, namely the pH value, citrus variety, soluble solids content, season, and water content in the juice. In addition, the pH and total acid (citric acid) might change over time due to neutralization or oxidation, which unfortunately could not be confirmed in this study. Microfiltration can remove large particles and reduce the turbidity and acidity of the juice that accumulates in the cake layer [60]. The total acid and pH values remained constant with the filtering process with polyether sulfone and polysulfone membranes [21]. Using an ultrafiltration membrane with an MWCO of 30.50 and 100 kDa for treating orange juice, the pH and total acid were slightly changed during the clarification [61]. Another study found that the ultrafiltration membrane did not affect the pH and the total acid [57].

3.7. Vitamin C

The vitamin C content in orange juice was expressed as ascorbic acid and was measured by using the titration method, as shown in Figure 13. The vitamin C content in the clarified orange juice was 31.85 mg/100 g, as the vitamin C content tends to increase in the retentate. Table 2 shows that the molecular weight of vitamin C (ascorbic acid) was 176.12 Da, slightly lower than the MWCO of the applied membrane of 210.14 Da. Partial retention of vitamin C was then expected and accumulated in the retentate. The formation of a dynamic membrane on the membrane surface enhanced the vitamin C retention.

The findings show that the membrane-based clarification could slightly maintain > 75% of the vitamin C content in the clarified orange juice. The nature of filtration in the ambient temperature helped prevent vitamin C denaturation. In another report, clarification of bitter orange via microfiltration, using a cellulose ester-based membrane, found that ascorbic acid content (mg/100 g) and total acid in the permeate were lower than the feed due to

sensitivity to the oxidation process [18]. Adding cyclodextrin could maintain vitamin C, while adding cellulose acetate could preserve vitamin C [56].

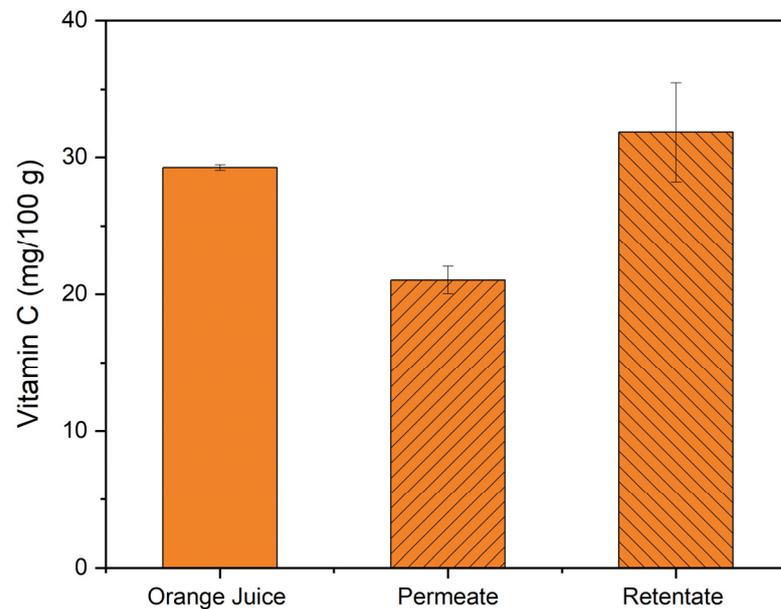


Figure 13. The vitamin C content of the feed (orange juice), the permeate, and the retentate streams.

3.8. Water Content

The water content was measured to evaluate the concentration level of the fresh juice via clarification, and the results are shown in Figure 14. The water content in the feed (fresh orange juice), the permeate, and the retentate are relatively similar. The water content of the clarified juice (the permeate) was slightly higher than the feed and the retentate due to the retention of suspended solid remained.

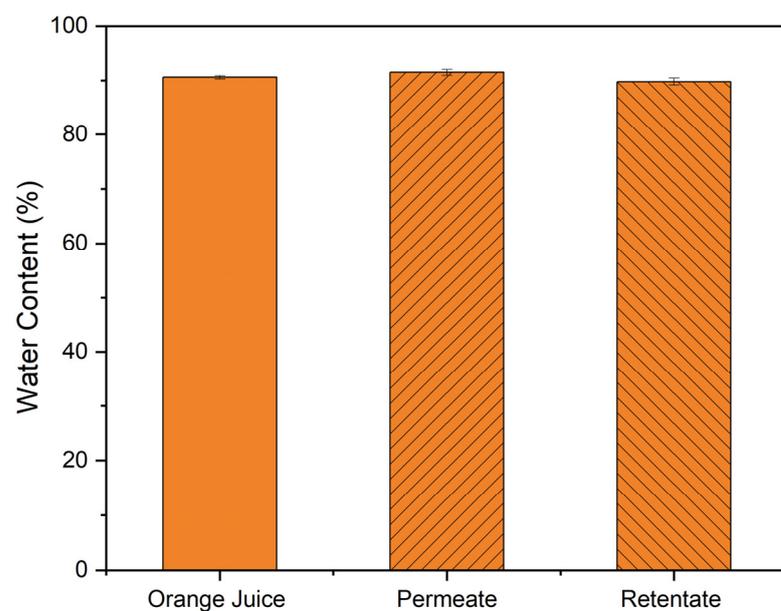


Figure 14. The water content of the feed (orange juice), the permeate, and the retentate stream.

3.9. Ash Content

The ash compositions represent orange juice's mineral content, and the analysis results are shown in Figure 15. The ash content slightly increased in the retentate could be attributed to a fraction of soluble solids retained by the membrane.

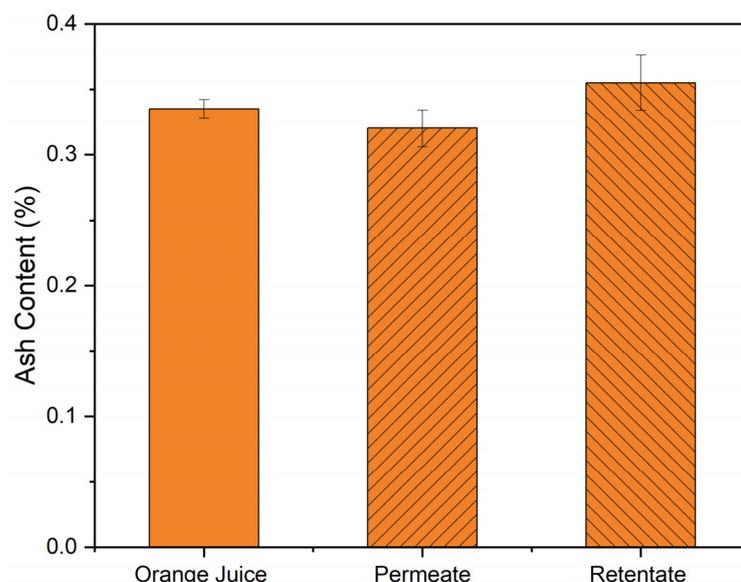


Figure 15. The ash content of the feed (orange juice), the permeate, and the retentate stream.

The clarification increased the ash content due to the increase in viscosity and soluble solids in the orange juice. The carbohydrate fractions in orange juice were in many forms, such as sucrose, fructose, and glucose [40]. The retained pectin would increase the total soluble solids value, eventually increasing the total ash value. In another study, clarification of sugarcane juice via microfiltration resulted in the ash content ranging from 0.32 to 0.34% for the three streams (feed, permeate, and retentate) [62]—not far from the ones obtained in this work.

3.10. Color Space

The results of the CIELAB color space (L^* , a^* , and b^*) orange juice measurement are shown in Figure 16. The measurement was conducted based on the L^* (perceptual lightness) and the a^* and b^* (four unique colors of human vision: red, green, blue, and yellow) of the orange juice. Overall, there was no change in color. The color b^* (yellowish) slightly increased. The L^* color, representing the condition of pure orange juice, had a brightness level of 34.35. The brightness value of the permeate was higher compared to the retentate. It is due to the browning event of the orange juice permeating through the membrane pores. In addition, orange juice contains pectin content, which can make the color paler and cloudier in the retentate stream.

The decrease in the brightness in orange juice occurred due to the natural browning or a particular reaction that darkened the color. Color differences are also influenced by several factors, such as the maturity level, seasonal variations, varieties, and regional developments [40]. In color a^* (reddish level), the value slightly increased in the permeate, reaching 7.9. The red color increment in orange juice tends to be lower than the yellowness because the appearance of orange juice is generally pale yellow. The color b^* (yellowness level) slightly increased in the retentate compared to permeate. The highest value of yellowness level was obtained in the retentate condition of 22.35. This is inversely proportional to the color L^* and a^* , where the color increased in the permeate. It can be attributed to the dominant yellow color compared to other colors in orange juice. Clarification of orange prickly pear juice by using a microfiltration membrane resulted in a clarified juice with a higher luminosity (L^*) value, which was more apparent and less turbid. In addition, the a^* value showed an increase in the red color, and b^* showed an increase in the yellow color [20]. The clarification of pomegranate juice via membrane filtration also led to an increase in the (L^*), (a^*), and (b^*) values, as reported elsewhere [60]. In another study, the membrane-based clarification of apple juice led to a higher permeate color intensity [19]. The clarification of Valencia orange juice with PES and PS membranes increased the color and clarity of

the juice due to the loss of suspended colloidal particles. The color intensity increased in the retentate condition compared to the feed-in in both membranes [21]. The variations of findings in the literature were because of the difference in the feeds and the membrane properties. Clarification using an ultrafiltration membrane resulted in lower turbidity and higher permeate color intensity. The color of the reddish level, the characteristic of blood orange juice, increased more significantly when using the membrane with lower MWCOs. The study found that the dominant color, yellow (b^*), would persist and increase [61] since the colors L^* and a^* became darker due to the concentration and other factors.

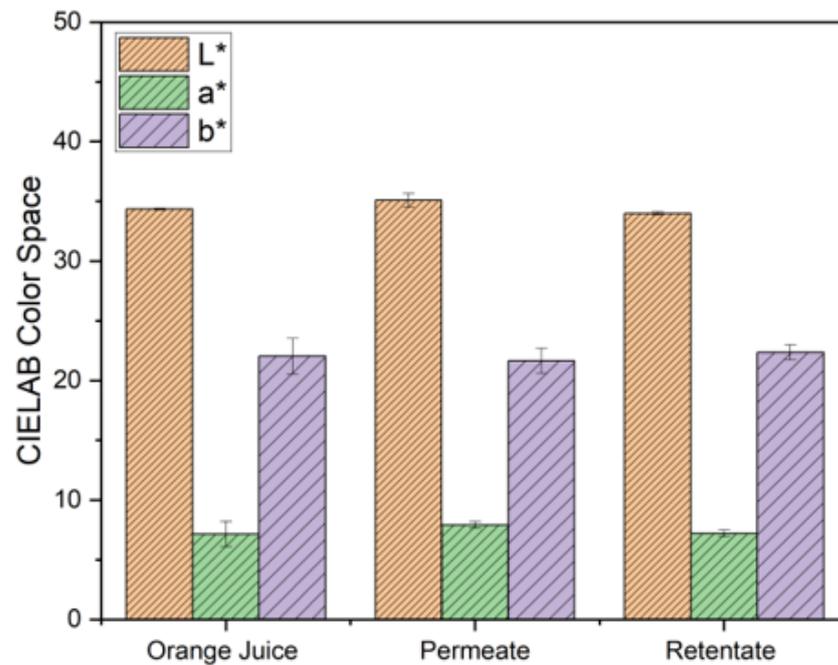


Figure 16. The CIELAB L^* , a^* , and b^* color space of the feed (orange juice), the permeate, and the retentate stream.

3.11. Summary of the Clarified Product Properties

Orange juice clarification and concentration using a plate-and-frame module produced a clarified juice with the properties summarized in Table 3.

Table 3. Properties of the clarified orange juice by using the developed plate-and-frame membrane. Data are presented as average \pm standard deviation.

Sample	pH	Total Soluble Solids ($^{\circ}$ Brix)	Viscosity	Ash Content (%)	Water Content (%)	Total Acid (%)	Vitamin C (mg/100 g)	Color		
								(L^*)	(a^*)	(b^*)
Orange Juice	5.76 \pm 0.06	8.40 \pm 0.14	3.27 \pm 0.33	0.34 \pm 0.01	90.58 \pm 0.32	0.62 \pm 0.08	29.25 \pm 0.21	34.35 \pm 0.07	7.15 \pm 1.06	22.05 \pm 1.48
Permeate	5.82 \pm 0.33	7.95 \pm 0.21	2.94 \pm 0.16	0.32 \pm 0.01	91.48 \pm 0.53	0.82 \pm 0.11	21.02 \pm 1.00	35.10 \pm 0.57	7.90 \pm 0.28	21.65 \pm 1.06
Retentate	5.85 \pm 0.02	9.00 \pm 0.85	2.85 \pm 0.06	0.36 \pm 0.02	89.78 \pm 0.63	0.54 \pm 0.01	31.85 \pm 3.64	34.00 \pm 0.14	7.20 \pm 0.28	22.35 \pm 0.64

4. Conclusions

This study evaluated a plate-and-frame module equipped with a predeveloped cellulose acetate-based ultrafiltration membrane by incorporating phenolic compounds from garlic extract as an additive. The module system was used to clarify orange juice. An analysis of the fresh juice as the feed, the clarified juice as the permeate, and the retentate streams was performed thoroughly. The results show no difference in the value of the pH, viscosity, total acid, water content, color L^* (brightness), and color a^* (reddish). Meanwhile, the total dissolved solids ($^{\circ}$ Brix), ash content, vitamin C, and color (b^* yellowish) were slightly increased in the clarified permeate relative to the fresh juice. The overall findings demonstrated that the developed plate-and-frame module could effectively clarify

orange juice without altering the quality. A follow-up study that assesses the hydraulic performance, especially against membrane fouling, is required to thoroughly assess the performance of the predeveloped membranes in a plate-and-frame module.

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References

- Ruxton, C.H.S.; Myers, M. Fruit Juices: Are They Helpful or Harmful? An Evidence Review. *Nutrients* **2021**, *13*, 1815. [[CrossRef](#)] [[PubMed](#)]
- Chanson-Rolle, A.; Braesco, V.; Chupin, J.; Bouillot, L. Nutritional Composition of Orange Juice: A Comparative Study between French Commercial and Home-Made Juices. *Food Nutr. Sci.* **2016**, *7*, 252–261. [[CrossRef](#)]
- Franke, A.A.; Cooney, R.V.; Henning, S.M.; Custer, L.J. Bioavailability and antioxidant effects of orange juice components in humans. *J. Agric. Food Chem.* **2005**, *53*, 5170–5178. [[CrossRef](#)] [[PubMed](#)]
- Dal Magro, L.; Pessoa, J.P.S.; Klein, M.P.; Fernandez-Lafuente, R.; Rodrigues, R.C. Enzymatic clarification of orange juice in continuous bed reactors: Fluidized-bed versus packed-bed reactor. *Catalysis Today* **2021**, *362*, 184–191. [[CrossRef](#)]
- Ackerley, J.; Wicker, L. Floc Formation and Changes in Serum Soluble Cloud Components of Fresh Valencia Orange Juice. *J. Food Sci.* **2003**, *68*, 1169–1174. [[CrossRef](#)]
- Sentandreu, E.; Gurrea, M.d.C.; Betoret, N.; Navarro, J.L. Changes in orange juice characteristics due to homogenization and centrifugation. *J. Food Eng.* **2011**, *105*, 241–245. [[CrossRef](#)]
- Domingues, R.C.C.; Faria Junior, S.B.; Silva, R.B.; Cardoso, V.L.; Reis, M.H.M. Clarification of passion fruit juice with chitosan: Effects of coagulation process variables and comparison with centrifugation and enzymatic treatments. *Process. Biochem.* **2012**, *47*, 467–471. [[CrossRef](#)]
- de Oliveira, R.C.; Docê, R.C.; de Barros, S.T.D. Clarification of passion fruit juice by microfiltration: Analyses of operating parameters, study of membrane fouling and juice quality. *J. Food Eng.* **2012**, *111*, 432–439. [[CrossRef](#)]
- Ghosh, P.; Pradhan, R.C.; Mishra, S. Clarification of jamun juice by centrifugation and microfiltration: Analysis of quality parameters, operating conditions, and resistance. *J. Food Process Eng.* **2018**, *41*, e12603. [[CrossRef](#)]
- Álvarez, S.; Riera, F.A.; Álvarez, R.; Coca, J.; Cuperus, F.P.; Th Bouwer, S.; Boswinkel, G.; van Gemert, R.W.; Veldsink, J.W.; Giorno, L.; et al. A new integrated membrane process for producing clarified apple juice and apple juice aroma concentrate. *J. Food Eng.* **2000**, *46*, 109–125. [[CrossRef](#)]
- Anvarian, A.H.P.; Smith, M.P.; Overton, T.W. The effects of orange juice clarification on the physiology of Escherichia coli; growth-based and flow cytometric analysis. *Int. J. Food Microbiol.* **2016**, *219*, 38–43. [[CrossRef](#)] [[PubMed](#)]
- Fellers, P. Florida's Citrus Juice Standards for Grades and Their Differences from United States Standards for Grades and United States Food and Drug Administration Standards of Identity. *Proc. Fla. State Hort. Soc.* **1990**, *103*, 260–264.
- Li, J.; Chase, H.A. Applications of membrane techniques for purification of natural products. *Biotechnol. Lett.* **2010**, *32*, 601–608. [[CrossRef](#)]
- Hameed, K.W. Concentration of Orange Juice Using Forward Osmosis Membrane Process. *Iraqi J. Chem. Pet. Eng.* **2013**, *14*, 71–79.
- Wu, M.L.; Zall, R.R.; Tzeng, W.C. Microfiltration and Ultrafiltration Comparison for Apple Juice Clarification. *J. Food Sci.* **1990**, *55*, 1162–1163. [[CrossRef](#)]
- Mirsaeedghazi, H.; Mousavi, S.; Mohammad Emam-Djomeh, Z.; Rezaei, K.; Aroujalian, A.; Navidbakhsh, M. Comparison between ultrafiltration and microfiltration in the clarification of pomegranate juice. *J. Food Process Eng.* **2012**, *35*, 424–436. [[CrossRef](#)]
- Pagliero, C.; Ochoa, N.A.; Marchese, J. Orange Juice Clarification by Microfiltration: Effect of Operational Variables on Membrane Fouling. *Lat. Am. Appl. Res.* **2011**, *41*, 279–284.

18. Mirsaedghazi, H.; Emam-Djomeh, Z. Clarification of Bitter Orange (*Citrus Aurantium*) Juice Using Microfiltration with Mixed Cellulose Esters Membrane. *J. Food Process. Preserv.* **2017**, *41*, e12738. [[CrossRef](#)]
19. Severcan, S.S.; Uzal, N.; Kahraman, K. Clarification of Apple Juice Using New Generation Nanocomposite Membranes Fabricated with TiO₂ and Al₂O₃ Nanoparticles. *Food Bioprocess Technol.* **2020**, *13*, 391–403. [[CrossRef](#)]
20. Mejia, J.A.A.; Yáñez-Fernandez, J. Clarification Processes of Orange Prickly Pear Juice (*Opuntia* spp.) by Microfiltration. *Membranes* **2021**, *11*, 354. [[CrossRef](#)]
21. Qaid, S.; Zait, M.; El Kacemi, K.; Midaoui, A.; el Hajji, H.; Taky, M. Ultrafiltration for clarification of Valencia orange juice: Comparison of two flat sheet membranes on quality of juice production. *J. Mater. Environ. Sci.* **2017**, *8*, 1186–1194.
22. Dahdouh, L.; Delalonde, M.; Ricci, J.; Ruiz, E.; Wisniewski, C. Influence of high shear rate on particles size, rheological behavior and fouling propensity of fruit juices during crossflow microfiltration: Case of orange juice. *Innov. Food Sci. Emerg. Technol.* **2018**, *48*, 304–312. [[CrossRef](#)]
23. Loizzo, M.R.; Sicari, V.; Tundis, R.; Leporini, M.; Falco, T.; Calabrò, V. The Influence of Ultrafiltration of Citrus limon L. Burm. cv Femminello Comune Juice on Its Chemical Composition and Antioxidant and Hypoglycemic Properties. *Antioxidants* **2019**, *8*, 23. [[CrossRef](#)]
24. Hojjatpanah, G.; Emam-Djomeh, Z.; Ashtari, A.K.; Mirsaedghazi, H.; Omid, M. Evaluation of the fouling phenomenon in the membrane clarification of black mulberry juice. *Int. J. Food Sci. Technol.* **2011**, *46*, 1538–1544. [[CrossRef](#)]
25. Gulec, H.A.; Bagci, P.O.; Bagci, U. Clarification of Apple Juice Using Polymeric Ultrafiltration Membranes: A Comparative Evaluation of Membrane Fouling and Juice Quality. *Food Bioprocess Technol.* **2017**, *10*, 875–885. [[CrossRef](#)]
26. Du, N.; Pan, L.; Liu, J.; Wang, L.; Li, H.; Li, K.; Xie, C.; Hang, F.; Lu, H.; Li, W. Clarification of Limed Sugarcane Juice by Stainless Steel Membranes and Membrane Fouling Analysis. *Membranes* **2022**, *12*, 910. [[CrossRef](#)]
27. Wibisono, Y.; Alvianto, D.; Argo, B.D.; Hermanto, M.B.; Witoyo, J.E.; Bilad, M.R. Low Fouling Plate and Frame Ultrafiltration for Juice Clarification: Part 1—Membrane Preparation and Characterization. *Sustainability* **2023**, *15*, 806. [[CrossRef](#)]
28. Scholz, M.; Wessling, M.; Balster, J. Chapter 5 Design of Membrane Modules for Gas Separations. In *Membrane Engineering for the Treatment of Gases: Volume 1: Gas-separation Problems with Membranes*; The Royal Society of Chemistry: Londra, UK, 2011; Volume 1, pp. 125–149.
29. Balster, J. Plate and Frame Membrane Module. In *Encyclopedia of Membranes*; Drioli, E., Giorno, L., Eds.; Springer: Berlin/Heidelberg, Germany, 2015; pp. 1–3.
30. Conidi, C.; Castro-Muñoz, R.; Cassano, A. Membrane-Based Operations in the Fruit Juice Processing Industry: A Review. *Beverages* **2020**, *6*, 18. [[CrossRef](#)]
31. Baker, R.W. *Membrane Technology and Applications*; Wiley: Hoboken, NJ, USA, 2012.
32. Gabelman, A.; Hwang, S.-T. Hollow fiber membrane contactors. *J. Membr. Sci.* **1999**, *159*, 61–106. [[CrossRef](#)]
33. Prasad, R.; Sirkar, K.K. Membrane-Based Solvent Extraction. In *Membrane Handbook*; Ho, W.S.W., Sirkar, K.K., Eds.; Springer: Boston, MA, USA, 1992; pp. 727–763.
34. Curcio, S.; Calabrò, V.; Iorio, G.; de Cindio, B. Fruit juice concentration by membranes: Effect of rheological properties on concentration polarization phenomena. *J. Food Eng.* **2001**, *48*, 235–241. [[CrossRef](#)]
35. de Carvalho, L.M.J.; de Castro, I.M.; da Silva, C.A.B. A study of retention of sugars in the process of clarification of pineapple juice (*Ananas comosus* L. Merrill) by micro- and ultra-filtration. *J. Food Eng.* **2008**, *87*, 447–454. [[CrossRef](#)]
36. Fang, P.; Du, J.; Yu, S. Impeller (straight blade) design variations and their influence on the performance of a centrifugal blood pump. *Int. J. Artif. Organs* **2020**, *43*, 782–795. [[CrossRef](#)] [[PubMed](#)]
37. Braha, D.; Maimon, O. The Measurement of a Design Structural and Functional Complexity. In *A Mathematical Theory of Design: Foundations, Algorithms and Applications*; Braha, D., Maimon, O., Eds.; Springer: Boston, MA, USA, 1998; pp. 241–277.
38. Pichaiyongvongdee, S.; Haruenkit, R. Comparative Studies of Limonin and Naringin Distribution in Different Parts of Pummelo [*Citrus grandis* (L.) Osbeck] Cultivars Grown in Thailand. *Kasetsart J. Nat. Sci.* **2009**, *43*, 28–36.
39. Weaver, C.M.; Daniel, J.R. *The Food Chemistry Laboratory: A Manual for Experimental Foods, Dietetics, and Food Scientists, Second Edition*; Taylor & Francis: Abingdon, UK, 2003.
40. Ashurst, P.R. *Production and Packaging of Non-carbonated Fruit Juices and Fruit Beverages*; Blackie Academic & Professional: Weinheim, Germany, 1995.
41. Corredig, M.; Kerr, W.; Wicker, L. Particle Size Distribution of Orange Juice Cloud after Addition of Sensitized Pectin. *J. Agric. Food Chem.* **2001**, *49*, 2523–2526. [[CrossRef](#)] [[PubMed](#)]
42. Lacroix, N.; Fliss, I.; Makhlof, J. Inactivation of pectin methylesterase and stabilization of opalescence in orange juice by dynamic high pressure. *Food Res. Int.* **2005**, *38*, 569–576. [[CrossRef](#)]
43. Sass-Kiss, A.; Sass, M. Immunoanalytical Method for Quality Control of Orange Juice Products. *J. Agric. Food Chem.* **2000**, *48*, 4027–4031. [[CrossRef](#)]
44. Vandenberghe, L.P.S.; Rodrigues, C.; de Carvalho, J.C.; Medeiros, A.B.P.; Soccol, C.R. 25—Production and Application of Citric Acid. In *Current Developments in Biotechnology and Bioengineering*; Pandey, A., Negi, S., Soccol, C.R., Eds.; Elsevier: Amsterdam, The Netherlands, 2017; pp. 557–575.
45. Kabasakalis, V.; Siopidou, D.; Moshatou, E. Ascorbic acid content of commercial fruit juices and its rate of loss upon storage. *Food Chem.* **2000**, *70*, 325–328. [[CrossRef](#)]
46. Pupin, A.M.; Dennis, M.J.; Toledo, M.C.F. HPLC analysis of carotenoids in orange juice. *Food Chem.* **1999**, *64*, 269–275. [[CrossRef](#)]

47. Meléndez-Martínez, A.J.; Vicario, I.M.; Heredia, F.J. Review: Analysis of carotenoids in orange juice. *J. Food Compos. Anal.* **2007**, *20*, 638–649. [[CrossRef](#)]
48. Li, J.; Zhang, C.; Liu, H.; Liu, J.; Jiao, Z. Profiles of Sugar and Organic Acid of Fruit Juices: A Comparative Study and Implication for Authentication. *J. Food Qual.* **2020**, *2020*, 7236534. [[CrossRef](#)]
49. Lee, H.S. Liquid chromatographic determination of benzoic acid in orange juice: Interlaboratory study. *J. AOAC Int.* **1995**, *78*, 80–82. [[CrossRef](#)] [[PubMed](#)]
50. Siener, R.; Seidler, A.; Voss, S.; Hesse, A. The oxalate content of fruit and vegetable juices, nectars and drinks. *J. Food Compos. Anal.* **2015**, *45*, 108–112. [[CrossRef](#)]
51. Cen, H.; Bao, Y.; He, Y.; Sun, D.-W. Visible and near infrared spectroscopy for rapid detection of citric and tartaric acids in orange juice. *J. Food Eng.* **2007**, *82*, 253–260. [[CrossRef](#)]
52. Shaw, P.E.; Wilson, C.W., III; Hansen, R.W. H.p.l.c. Determination of trace levels of succinic acid in orange juice from Freeze-damaged and undamaged fruit. *J. Sci. Food Agric.* **1987**, *41*, 153–158. [[CrossRef](#)]
53. Pepin, A.; Stanhope, K.L.; Imbeault, P. Are Fruit Juices Healthier Than Sugar-Sweetened Beverages? A Review. *Nutrients* **2019**, *11*, 1006. [[CrossRef](#)]
54. Conidi, C.; Destani, F.; Cassano, A. Performance of Hollow Fiber Ultrafiltration Membranes in the Clarification of Blood Orange Juice. *Beverages* **2015**, *1*, 341–353. [[CrossRef](#)]
55. Abd-Razak, N.H.; Zairossani, M.N.; Chew, Y.M.J.; Bird, M.R. Fouling Analysis and the Recovery of Phytosterols from Orange Juice Using Regenerated Cellulose Ultrafiltration Membranes. *Food Bioprocess Technol.* **2020**, *13*, 2012–2028. [[CrossRef](#)]
56. Fajarika, D.; Noor, E. The Design Process for Entrapping Limonin and Naringin in Siam Juice by Cyclodextrin. *IPTEK J. Proc. Ser.* **2014**, *1*. [[CrossRef](#)]
57. Laorko, A.; Li, Z.; Tongchitpakdee, S.; Chantachum, S.; Youravong, W. Effect of membrane property and operating conditions on phytochemical properties and permeate flux during clarification of pineapple juice. *J. Food Eng.* **2010**, *100*, 514–521. [[CrossRef](#)]
58. Quist-Jensen, C.A.; Macedonio, F.; Conidi, C.; Cassano, A.; Aljlil, S.; Alharbi, O.A.; Drioli, E. Direct contact membrane distillation for the concentration of clarified orange juice. *J. Food Eng.* **2016**, *187*, 37–43. [[CrossRef](#)]
59. Jesus, D.F.; Leite, M.F.; Silva, L.F.M.; Modesta, R.D.; Matta, V.M.; Cabral, L.M.C. Orange (*Citrus sinensis*) juice concentration by reverse osmosis. *J. Food Eng.* **2007**, *81*, 287–291. [[CrossRef](#)]
60. Salehinia, S.; Mirsaedghazi, H.; Khashehchi, M. The effect of laser on the efficiency of membrane clarification of pomegranate juice. *J. Food Sci. Technol.* **2021**, *58*, 1682–1692. [[CrossRef](#)] [[PubMed](#)]
61. Toker, R.; Karhan, M.; Tetik, N.; Turhan, I.; Oziyci, H.R. Effect of Ultrafiltration and Concentration Processes on the Physical and Chemical Composition of Blood Orange Juice. *J. Food Process. Preserv.* **2014**, *38*, 1321–1329. [[CrossRef](#)]
62. Rezzadori, K.; Serpa, L.; Penha, F.; Petrus, R.; Petrus, J. Crossflow microfiltration of sugarcane juice: Effects of processing conditions and juice quality. *Food Sci. Technol.* **2014**, *34*, 210–217. [[CrossRef](#)]

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