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The effect of apple cider characteristics and membrane pore size on membrane fouling

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ABSTRACT

Microfiltration (MF) of apple cider using four membrane pore sizes was investigated. MF with 0.2 and 0.45 μm pore sizes, and with 0.8 and 1.4 μm pore sizes, respectively, resulted in similar flux behavior and MF juice characteristics. Clear juice was obtained using all four pore sizes. MF did not cause any significant changes in pH and °Brix, regardless of pore size. Viscosity of the MF juice was lower than of the unfiltered apple cider for all membranes, but the viscosity decrease was the largest for the 0.2 and 0.45 μm membranes, due to the low transmission of pectin into the MF juice. Pectin transmission was 3–9% of the initial pectin content for the 0.2 μm membrane, 6–51% for the 0.45 μm. 45–100% for the 0.8 μm, and 60–100% for the 1.4 μm membrane. Apple cider haze particles that were smaller or of size comparable with the membrane pore size were deemed responsible for fouling, while particles much larger than the pores did not have a significant contribution to fouling. The fouling of 0.2 μm and 0.45 μm membranes was dominated by cake layer formation (external fouling), while fouling of the 0.8 μm and 1.4 μm membranes was dominated by pore constriction and pore blocking (internal fouling).

1. Introduction

Microfiltration (MF) has received increased interest in recent years as a method for juice clarification and microbial removal, since this processing method is able to maintain high nutritional and sensory quality of the processed juice (Zhao et al., 2015). One of the challenges associated with the large scale adoption of MF by the juice industry is the fast decline in permeate flux due to membrane fouling. Typically, membrane fouling mechanisms include pore constriction, pore blocking and cake layer formation (de Barros, Andrade, Mendes, & Peres, 2003). Particles in the feed that are smaller than the membrane pore openings can enter the pores; if they get adsorbed onto the membrane channels they will cause pore constriction, thus reducing the effective diameter of the membrane pores. When feed particles are comparable in size with the membrane pore size, pore blocking occurs as the permeate flux creates a convective drag toward the membrane, and particles are adsorbed and/or deposited onto the membrane pores and surface. Particles larger in size than the membrane pores can be retained onto the membrane surface as a cake layer. A significant reduction of the permeate flux will occur in all of these cases. The specific mechanisms of membrane fouling depend on the feed composition and the interactions between the membrane material and the feed components. Several studies examined the effect of small pore MF (0.1–0.2 μm membrane pore size) on the permeate flux and the composition of MF apple juice (Ben Amar, Gupta, & Jaffrin, 1990; Fukumoto, Delaquis, & Girard, 1998; Padilla-Zakour & McLellan, 1993; Su, Liu, & Wiley, 1993; Vladisavljević, Vukosavljević, & Bukvić, 2003; Wu, Zall, & Tzeng, 1990; Zárate-Rodríguez, Ortega-Rivas, & Barbosa-Cánovas, 2001). Wu et al. (1990) showed that MF membranes with a 0.1 μm pore size had a higher permeate flux than UF membranes with molecular weight cut-off of 5 kDa and 50 kDa, and the MF juice had significantly higher total soluble solids, was visually darker and was preferred over UF juice by a sensory panel. This suggests that increasing the membrane pore size above 0.2 μm, which is the pore size typically used by the juice industry for clarification purposes, can lead to a substantial increase in flux and transmission of nutritional components, which will benefit both the consumers and the juice industry. However, very little information exists about the MF behavior and fouling mechanisms in large pore MF of apple cider, and these cannot be predicted based on what is known for small pore MF. Therefore, the objective of this study is to evaluate the effect of apple cider components and membrane pore size on the flux and fouling in

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large pore MF of apple cider.

2. Materials and methods

2.1. Materials

Cold, raw apple cider was obtained from Cornell Orchards (Ithaca, NY) and Red Jacket Orchards (Geneva, NY). Each batch of cider was stored at 4 °C for maximum two weeks before being processed. Due to the seasonal production, some batches of cider were stored frozen at −20 °C and thawed at 4 °C before use.

2.2. Microfiltration experiments

The pilot-scale microfiltration unit consisted of a feed tank with a capacity of about 190 L connected to a variable-speed centrifugal pump, a tubular heat exchanger and a ISOFLUX™ tubular ceramic (made of α-Al_2O_3) membrane of TAMI design (GEA Filtration, WI) placed inside a stainless steel housing. The membrane had an outside diameter of 25 mm, a length of 1200 mm, 23 internal channel of 3.5 mm hydraulic diameter each, and a total membrane surface area of 0.35 m^2. Four membrane pore sizes were used in this study: 0.2 μm, 0.45 μm, 0.8 μm, and 1.4 μm.

The MF of raw apple cider was conducted at a cross-flow velocity of 5.5 m/s and a temperature of 6 ± 1 °C, which was maintained by circulating chilled water in the counter current tubular heat exchanger. This temperature was chosen to preserve the quality of apple cider and MF juice and limit browning during processing.

Feed inlet pressure (P_1), retentate outlet pressure (P_2) were recorded and transmembrane pressure was calculated as

$$TMP = \frac{(P_1 + P_2)}{2} - P_p \quad (1)$$

The permeate pressure (P_p) equaled the atmospheric pressure. The transmembrane pressure in all MF runs was 159 kPa.

The permeate flux data was obtained gravimetrically using an electronic scale. The permeate flux (J) was calculated as:

$$J = \frac{M}{A \times t \times \rho} \quad (2)$$

where: J: permeate flux (L/m^2h); M: amount of permeate (kg) collected in the time interval t (h); A: surface area of the membrane (m^2); ρ: density of the permeate at the filtration temperature (kg/m^3).

The duration of each MF experiment was 1 h.

In order to compare the rate of flux drop among different experimental conditions, the relative flux was calculated as:

$$Relative\ flux = \frac{J}{J_0} \times 100 \quad (3)$$

where: J: permeate flux at a given time point (L/m^2h); J_o: initial flux (L/m^2h).

The "initial" flux value, J_0, was taken at 7 min after starting the pump, after the system was fully stabilized. The value of the relative flux relates to membrane fouling, i.e. a lower J/J_0 value indicates more pronounced fouling of the membrane.

2.3. Membrane cleaning

After each MF experiment, a chemical cleaning cycle was carried out. The cleaning procedure consisted of a rinse with reverse osmosis (RO) water for 10 min, followed by alkaline cleaning with Ultrasil-25 at a concentration of 20 g/L at 80 °C for 30 min and a second RO water rinse for 10 min or until neutrality. Acid cleaning with 5 mL/L HNO_3 at 50 °C for 20 min was then performed, followed by a third RO water rinse for 10 min or until neutrality. The effectiveness of cleaning and change in the membrane performance with time were monitored by determining the water flux of the membrane before and after the MF experiments. The cleaning process was deemed satisfactory if the water flux did not change by more than 5%.

2.4. Physicochemical analysis of cider and juice

pH was measured at 20 °C using a Fisher Scientific Accumet Excel XL20 pH meter, (Fisher Scientific, Pittsburgh, PA). °**Brix** was measured with a MISCO® digital probe refractometer (MISCO® Products Division, Cleveland, OH), at room temperature.

Viscosity was measured at 6 °C using a Brookfield DV-II+ Pro viscometer with a ULV adapter, in triplicate.

Turbidity of the unfiltered apple cider and the MF juice was measured using a 2020wi turbidimeter (LaMotte Company, USA) in Formazin Nephelometric Units (FNU). Measurements were duplicated.

The **suspended insoluble solids (SIS)** content was determined by centrifuging 10 mL of apple cider at 2200 × g for 15 min. After discarding the supernatant, the sediment (SIS) was weighed and SIS was expressed in g/L. All experiments were carried out in duplicate.

The **pectin** content was determined with a colorimetric assay using m-hydroxydiphenyl for analysis of galacturonic acid (Kintner & van Buren, 1982), in duplicate.

The **particle size distribution** in the apple cider and MF juice, without dilution, was measured by dynamic light scattering, using a Brookhaven 90Plus Particle Size Analyzer equipped with a Peltier temperature control system (Brookhaven Instruments Corporation, Holtsville, NY). Measurements were conducted at 20 °C, a fixed angle of 90°, and a wavelength of 658 nm. Data collection and analysis were performed using the BIC software (Brookhaven Instruments Corp., Holtsville, NY) and size distribution was obtained from the experimental data. The dust filter cut-off was set at 30, which improves the quality of the measurements by rejecting measurement resulted from random particles, such as air bubbles or dust. This value was selected based on the manufacturer recommendation for scenarios where the average particle size is expected to be in the hundreds of nm range. Each particle size measurement consisted of 8 individual runs for duration of 30 s per run. The relative particle size distribution and the intensity weighted effective average diameter were determined for each sample. At least one measurement for each sample was conducted and measurements were taken within 24 h from processing. It is important to note that this method of particle size measurement works on the assumption that all particles have spherical shapes.

Zeta potential of the ceramic membrane particles used in this study was measured using a Malvern Zetasizer nano-ZS (Malvern Instruments Ltd., Malvern, Worcestershire, United Kingdom) with disposable folded capillary cells (Malvern Instruments Ltd., Malvern, Worcestershire, United Kingdom). Two grams of ceramic membrane was ground into a fine powder using a pestle and mortar. The powder was then suspended into 10 mL deionized water (Mili-Q; Merck Millipore Ltd., Billerica, MA), vortexed thoroughly, and allowed to settle for 40 min without disturbance. Next, 0.1 mL of the supernatant was transferred into 0.9 mL of pH 3.5 buffer to make a 1:10 dilution. A 1 mL sample from the resulting suspension was aliquoted into a cuvette, and zeta potential were measured at 20 °C, in triplicate, using 100 cycles per analysis for each of the triplicates.

Scanning electron microscopy (SEM). A clean membrane