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Decoloration of sugarcane molasses by tight ultrafiltration: Filtration behavior and fouling control

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ABSTRACT

Economically viable recovery of sucrose from sugarcane molasses is of great interest to academia and industry, and membrane technology is promising to achieve this goal, especially the separation of pigments and sucrose. A suitable membrane should have higher permeate flux and color removal, as well as lower sugar loss and membrane fouling during the decoloration of the sugarcane molasses. Compared with polyether sulphone ultrafiltration (UF) membranes, a polyamide UF membrane with a molecular weight cut-off of 2 kDa was preferred for this purpose. To further control membrane fouling, alkali treatment on such membrane and different pretreatments on molasses were also examined. Alkaline treatment on the membrane could further increase its anti-fouling performance at the sacrifice of some color removal. The pretreatment with ceramic membrane filtration greatly increased the permeate flux of the polymeric membrane, but it only transferred the fouling from the polymeric membrane to the ceramic membrane, unfortunately, from reversible to irreversible fouling. Moreover, the UF permeate flux was improved at higher pH (neutral) and temperature (60 °C), and these conditions also could retard microbial growth and sucrose conversion. Higher dilution times on molasses enhanced the permeate flux of the UF and reduced the sugar loss, but the filtration time was prolonged. Moreover, the sucrose and reducing sugar retentions by nanofiltration (NF) membrane at 60 °C kept around 96% and 60% respectively, implying that separation of sucrose and reducing sugar after the decoloration of molasses could be realized. This work not only provides an alternative method to efficiently utilize sugarcane molasses, but also serves as a valuable guide for process design and practical operation in subsequent industrial application.

1. Introduction

Cane molasses is one of the main byproducts in cane sugar production industry. It is a big waste of sucrose and is difficult to handle due to its large production amount and complicate components (e.g. pigments and salts). The components of cane molasses vary with cane type, growing condition, ripeness degree and refining process. Its main components include sucrose, reducing sugar, pigments, collides, inorganic salts and other trace amount minerals, etc. Thereinto, sucrose and reducing sugar are the major part of molasses accounting for 30–50% and 15–20% (dry weight) respectively [1,2]. Thanks to its high content of valuable substance (e.g. fermentable sugars), low price and easy utilization, molasses has been applied as animal feed supplement [3], substrate for fermentation [4,5], slashing agent for concrete [6,7], etc. However, these utilization approaches can not fully exploit the potential value of molasses. It is well known that a large amount of sucrose remains in the mother liquid (i.e. molasses) during the crystallization process due to the presence of many impurities especially the pigments and inorganic salts. If this residual sucrose can be recovered, the sugar plant will gain great economic benefit and also reduce the waste generation. Thus, many technologies have been applied to purify the molasses and recover the sugars.

Many methods such as barium sulfate method, Steffen process and chromatography have been used for sugar recovery from molasses. Among these methods, the barium sulfate and Steffen methods suffer from toxicity residue, difficulty of operation and high cost. As for the chromatographic technology, it has been successfully applied in the sucrose recovery from beet molasses [8,9]. However, it is not cost-efficient for the cane molasses because of the higher contents of reducing sugar, inorganic ions, colloids and other impurities in the cane molasses than in the beet molasses (resulting in severer fouling and shorter lifespan of the resin for the former) [10]. Membrane filtration, which is regarded as a simple, mild, energy-efficient and scalable process, has been widely used in food industry [11–13]. Membrane filtration includes microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO) according to the difference of membrane pore

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size. MF was utilized to remove suspended solids and collides in the crude feed stream; macromolecular and micromolecular solutes in the clarified solution can be separated by UF, NF and RO in sequence. Actually, it was reported that membrane filtration has been extensively applied for sugar refining, for example, cane juice purification and concentration [14], refining of remelted raw sugar as well as clarification and decolorization of molasses. Regarding the cane juice purification, Jegatheesan et al. applied ceramic MF and UF membranes to further purify the limed sugarcane juice [15,16]. Balakrishnan's group conducted serial investigations on the purification of raw sugarcane juice by polymeric UF membranes from bench-scale to pilot-scale trials in sugar mills [17,18], and also elaborately studied the fouling mechanism of cane juice on UF membranes [19]. In order to further improve color removal, Luo et al. developed a two-stage UF process for refining sugarcane juice including a tubular loose UF membrane for clarification and a spiral-wound tight UF for decoloration, and the color removal kept more than 95% (the color value of the final juice was below 800 IU) [20]. Moreover, an integrated UF and ion exchange (IE) for the production of colorless liquid sugar from sugarcane juice was evaluated by Susanto et al. [21]. As for remelted raw sugar, after carbonated and filtrated by plate-and-frame filter press, the clarified remelted raw sugar was further purified by a ceramic MF membrane [22]. With respect to molasses purification by membrane technology, the objectives of most studies were to improve the subsequent fermentation efficiency and reduce the wastewater generation [23]. Besides, Geanta et al. examined the feasibility of micellar-enhanced UF for the recovery of lactic acid and citric acid from the beet molasses previously decolorized with activated charcoal [24]. Decoloration of beet molasses by activated charcoal followed by UF was also investigated by Bernal [25], where the membrane was mainly used for the removal of powdered activated charcoal, and a high color reduction of 96.5% was achieved.

To the best of our knowledge, there has been no systematical report yet regarding the recovery of sucrose from sugarcane molasses through membrane technology. This is possibly due to the complex and high content impurities in cane molasses which are difficult to remove and prone to cause serious membrane fouling. It was reported that membrane fouling is the main limitation for the successful application of membrane technology in the juice clarification [26,27]. Therefore, fouling control is the most important issue in the membrane filtration of sugarcane molasses. In cane molasses, the main foulants for the membrane are colloids, hydrophobic pigments and multivalent salts (SO_4^{2-}, PO_4^{3-}, Mg^{2+}, Ca^{2+}). A possible strategy for fouling control is to separate these foulants in different steps. Therefore, an integrated process mainly consisting of pretreatment, tight UF and NF was proposed, as shown in Fig. 1. Suspended solids and collides could be eliminated by pretreatment, and the majority of the pigments could be removed by tight UF, and then inorganic salts and reducing sugar in the UF permeate were washed away using NF. In such process, the UF decoloration step plays a vital role in the sucrose recovery since the pigments have a prominent negative effect on the subsequent sucrose crystallization.

In this study, the UF decoloration of sugarcane molasses was systematically investigated by a spiral-wound membrane module, focusing on the improvement of fouling control, permeate flux and color removal. First, a suitable UF membrane was selected mainly in terms of permeate flux and anti-fouling performance. Then a special alkaline treatment on the chosen membrane was carried out to further improve its performance. Moreover, effect of pretreatment methods (i.e. centrifugation, chitosan flocculation and ceramic membrane filtration) on the UF behavior was studied, and the process parameters including pH and temperature were optimized. Finally, effect of molasses dilution time on UF and subsequent NF performance was estimated in terms of permeate flux and retention.

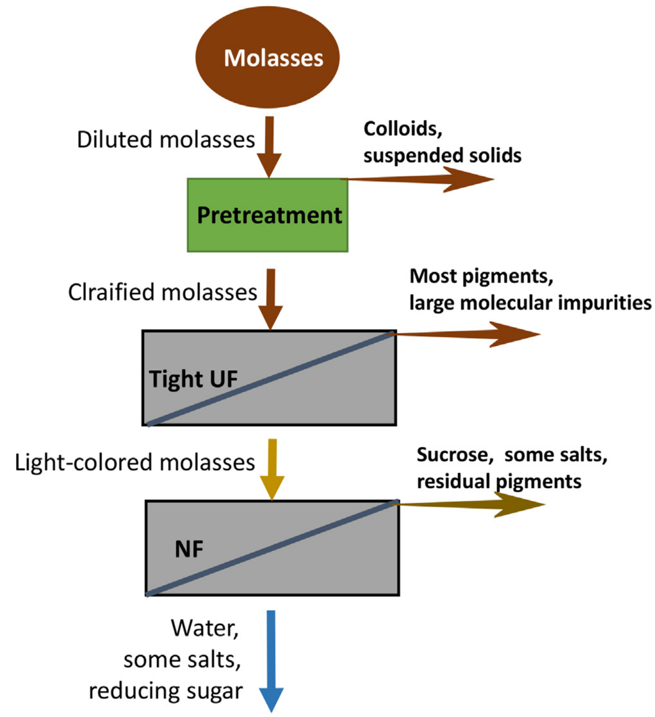

Fig. 1. Schematic diagram of molasses refining by an integrated membrane process.

2. Experimental

2.1. Materials

Sugarcane molasses was kindly provided by a local sugar mill in Zhanjiang, Guangdong Province, China. The composition of the diluted raw molasses with different dilution ratios was listed in Table 1. Chitosan (CTS) (deacetylation degree ≧ 95%, viscosity 100–200 mpa s) was obtained from Macklin (Shanghai, China). All the other chemical reagents used were purchased from Beijing Chemicals Reagent Company, China.

Ceramic membrane (pore size 50 nm) was supplied by TAMI industries, France. The tubular UF ceramic membrane consists of seven channels with an area of $1.1 \times 10^{-2}\,m^2$. Main properties of all the tested polymer UF membranes were summarized in Table 2. For the alkaline treatment on membrane, one of tight UF membrane module was rinsed by an alkaline solution of 0.5% (w/v) NaOH at 35 °C for 1 h, and then soaked in 0.5% (w/v) $NaHSO_3$ solution for at least one week. The polyamide (PA) NF membrane (Desal-5 DL) was obtained from Osmonics (GE) with a molecular weight cut-off (MWCO) of 150–300 Da.

2.2. Molasses pretreatments

The raw molasses in storing tank was strongly stirred before sampling. After the molasses became homogeneous, a certain amount of

Table 1
Main characteristics of sugarcane molasses at different dilution ratios.

Molasses: water (g/g)	Brix (%)	Conductivity ($mS\,cm^{-1}$)	Absorbance at 420 nm	Sucrose ($g\,L^{-1}$)	Reducing sugar ($g\,L^{-1}$)
1: 2	27.6 ± 0.2[a]	57.3 ± 1.1	36.9 ± 0.9	159 ± 28	55 ± 3
1: 3	20.5 ± 0.1	44.1 ± 0.3	29.6 ± 1.8	113 ± 11	38 ± 9
1: 4	16.1 ± 0.2	34.0 ± 1.1	20.9 ± 3.2	89 ± 23	30 ± 5

[a] Standard deviation.