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Ultrafiltration of Raw Indian Sugar Solution Using Polymeric and Mineral Membranes

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ABSTRACT

The traditional sugar industry still faces problems from the disposal of final wastes, which generally have concentrated organic materials, because of the use of chemicals for liming, sulphitation, or phosphatation in the treatment of raw cane juice and for the removal of color. In the present work, ultrafiltration and coagulation coupled with ultrafiltration are used for clarification as well as for decolorization of raw brown sugar obtained from an Indian sugar industry. Polyethersulphone (PES) membranes (5–100 kDa MWCO) and mineral Carbosep membranes (15–50 kDa MWCO) are used for decolorization of raw brown sugar solutions. Using a feed solution of 50°Brix, about 50% reduction in color at a permeate flux of 1.4×10^{-5} $m^3/m^2 \cdot s$ (about 50 lmh) was obtained both for PES and mineral membranes with MWCO between 30–50 kDa. The ultrafiltration process provided a juice of higher purity and lower color; thus this process could be an economic option for the sugar industry. Extended time experiments (which are under way) need to be carried out to study the long-term behavior of such membranes for application in the sugar industry.

Key Words. Cane sugar; Ultrafiltration; Decolorization; Mineral membrane; Polymeric membrane

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INTRODUCTION

Raw sugarcane juice obtained by crushing sugarcane contains 12–15% sucrose and various impurities, such as reducing sugars, organic acids, amino acids, proteins, starch, gums, coloring matter, and other suspended matters that impart a dark color and high turbidity to the juice (1, 2). Clarification of the juice in the manufacture of raw sugar is mainly done by liming, carbonation, sulphitation, or phosphatation, followed by sludge separation (3). The affination step removes 40–50% of the colored impurities, while carbonation/phosphatation eliminates 40–60% of the remaining color (4, 5). Hence, the traditional treatment method does not, to a large extent, remove macromolecular substances like dextrans, starch, waxes, bacteria, etc. (6). The presence of such macromolecules along with the sucrose significantly hinders the successive purification stages (7–9).

Membrane ultrafiltration (UF) and microfiltration (MF) have been applied for raw juice clarification and for the removal of the macromolecular species in the beet sugar and cane sugar industries (10–13). These studies showed that UF/MF could be effective in the partial removal of the macromolecular species. Further development was slow because of low ($<2.8 \times 10^{-6}$ $m^3/m^2{\cdot}s$) permeate fluxes and the life span of the polymeric membranes (14, 15).

In a generalized refining scheme the raw sugar is conveyed to the affination station, mixed with water, centrifuged, and washed with water to give a low-color sugar liquor of about 70% solid content. It is further clarified on powdered carbon or by phosphatation/carbonation treatment to produce SVLC (super very low color) sugar. A new process, NAP (new Applexion process) has been integrated in a cane sugar mill (Hawaiian Commercial and Sugar company, HC&S, Hawaii) in which the remelted raw sugar is decolorized in an ion exchanger, then crystallized in a single- or double-stage batch pan (16). This new process can produce a very good SVLC sugar completely free of turbidity and sulfites. The ion-exchange resins absorb color from cane sugar liquor, and regeneration is carried out by passing through an alkaline brine solution. The regeneration effluent contains a high concentration of sodium chloride and colored organic matter. The nanofiltration (NF) process has been found satisfactory for the recovery of sodium chloride and the removal of organic matter from the regeneration effluent (17).

The development of improved polymeric and mineral UF membranes has rekindled interest in the treatment of raw clarified juice (of around 20°Brix) because of potentially high permeate fluxes and extended membrane life. A comprehensive survey of present day applications of membranes in the sugar industry has been nicely presented by Cheryan (18).

In the Indian sugar industry, following the primary purification stage, the raw juice of around 20°Brix is concentrated to 50°Brix using a five stage heating system. This heat treatment induces further color formation due to the presence of high-molecular-weight species in the sucrose solution. Membrane UF is hence a potential application for removal of color from such a concentrated stream.

In this work the decolorization of 50°Brix solutions of raw Indian sugar using polymeric polyethersulfone (PES) membranes (5–100 kDa MWCO) and mineral Carbosep tubular membranes (15–50 kDa) was investigated. The temperature of UF was kept constant at around 80°C, which is the process temperature at which the 50°Brix stream is available in the sugar industry. Coagulation using a polyelectrolyte as a method of pretreatment was also examined to determine the efficacy of coupling coagulation with membrane UF.

EXPERIMENTAL

Membranes and Ultrafiltration Testing Loop

Polyethersulfone membranes (Omega) in the form of circular discs of 47-mM diameter were purchased from Pall Filtron, France. Ultrafiltration was carried out using a stirred cell unit marketed by Osmonics (Sepa ST membrane cell) with an effective membrane diameter of 39 mm. The UF cell was immersed in a 50–50% mixture of ethylene glycol and water that was heated using a magnetic stirrer/heater to maintain a constant temperature of 80°C. The permeate flux was measured as a function of time by data acquisition into a microcomputer from a Sartorius balance having 1-mg accuracy. The stirring speed was kept constant for all runs at 800 rpm. The UF cell was pressurized using compressed air. The pressure was kept constant at 0.29 MPa for all stirred-cell UF runs. The pressure was monitored using a Bourdon pressure gauge having an accuracy of 9.8 kPa. Typically, an initial volume of 100 mL was introduced into the stirred cell. The cell was then pressurized to the desired pressure using compressed air. The permeate flux was monitored by measuring the weight of the permeate produced as a function of time.

Cross-flow UF was carried out using a tubular (Tech-Sep) Carbosep mineral membrane with a skin layer of zirconium oxide on carbon support of 6 mm i.d. and 1.2 m long. The feed reservoir was heated to maintain a constant temperature at around 80°C. The feed was pumped through the membrane tube using a four-head diaphragm pump (equipped with a pulsation damper) at a constant velocity of 2.5 m/s. The retentate stream was recycled back into the feed reservoir while the permeate was removed continuously. The permeate flux and the operating pressure were measured as described for the stirred-cell UF. The operating pressure was maintained constant using a back pressure valve at the exit of the membrane module. Initially, 2 L of solution was taken into the feed reservoir. This was then circulated through the membrane circuit while keeping the permeate valve closed. The feed reservoir was heated electrically so as to bring the entire testing circuit to the desired temperature (~80°C). Once the desired temperature was achieved, the permeate valve was opened while simultaneously the back pressure valve was closed at the exit of the membrane module until the desired feed pressure was achieved. All runs were carried out at constant cross-flow velocity and feed pressure.

Analytical Measurements

°Brix was measured using an Abbe refractometer. Color was measured according to the conventional ICUMSA (International Commission for Uniform methods of Sugar Analysis) method (4, 19). The sample was diluted to 5°Brix, adjusted to pH 7 using dilute alkali and filtered through a 0.45-μm membrane. The optical density was measured at 420 nm using a HACH DR/2000 direct-reading spectrophotometer.

Coagulation

Coagulation of raw Indian sugar solutions was carried out using a cationic polyelectrolyte (polyacrylamide, molecular weight $> 10^7$ Da) at 80°C. To determine the optimum dosage, addition of the polyelectrolyte was carried out in steps of 0.5 ppm, and the color reduction after each addition was monitored.

RESULTS AND DISCUSSION

Figure 1 shows the % color reduction as a function of dosage of polyacrylamide for three different initial concentrations of raw Indian sugar solutions. Surprisingly, the optimum dosage is around 1.5 ppm irrespective of the initial concentration. The maximum color reduction is about 25% at the optimum dosage of 1.5 ppm of polyelectrolyte irrespective of the initial concentration.

Figure 2 shows the variation of permeate flux as a function of the volume concentration factor (VCF) for an initial feed solution of 50°Brix at 0.29 MPa ultrafiltered in the stirred cell using 30 and 50 kDa PES membranes. Even though the initial flux is much higher for the 50 kDa membrane, the flux soon drops to stabilize at a value of around 1.4×10^{-5} $m^3/m^2 \cdot s$ (50 lmh) for both membranes. The steady (quasi-time invariant) flux corresponded to a volume concentration factor of around 2. A similar observation was made by Punidadas and Decloux (15) during cross-flow MF of sugar solutions on mineral membranes. Similar steady flux values were also obtained when the 100 kDa membrane was used. The permeate flux became quasi-steady for a volume concentration factor of around 2 for three different initial volumes (50, 100, and 200 mL) using a 30 kDa membrane. For the 100 kDa membrane however, a quasi-steady flux was obtained at a volume concentration factor of about 3. This seems to indicate that the UF process is sensitive to a critical

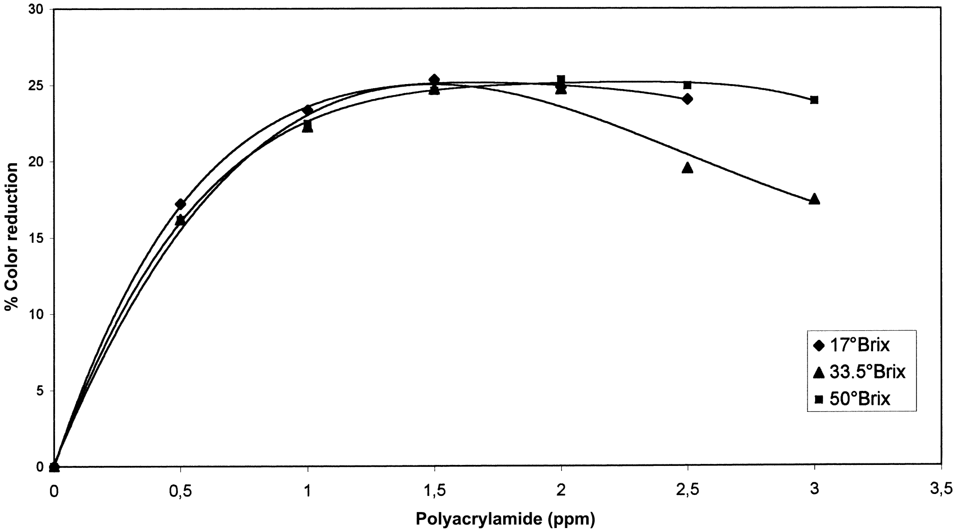

FIG. 1 Percentage of color reduction by coagulation using cationic polyacrylamide at 80°C as a function of dosage of polyacrylamide for raw Indian sugar solutions of different initial °Brix.

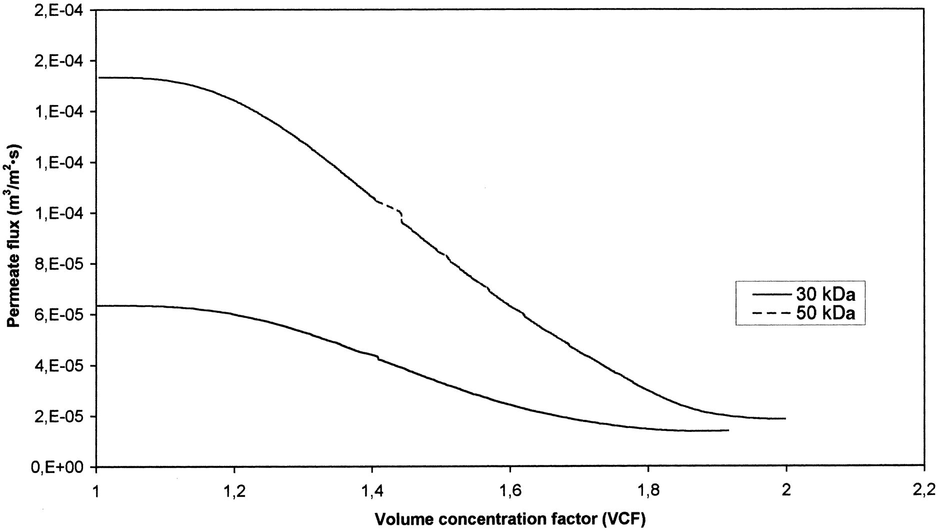

FIG. 2 Variation of permeate flux as a function of volume concentration factor using 30 and 50 kDa PES membranes (frontal filtration) for uncoagulated 50°Brix raw Indian sugar solutions; 80°C, 0.29 MPa.

concentration of certain moderately high molecular-weight species. The rejected species quickly build up in concentration at the feed surface of the membrane forming a highly viscous additional barrier to the filtration. Following the standard filtration theory, the presence of this barrier would lower the flux. Furthermore, since the thickness of this additional barrier is dependent on the transmembrane flux, the system stabilizes to a unique steady flux value. Such self-regulatory behavior has been experimentally reported for UF where there is a formation of a "gel" layer of rejected particles (20). A detailed analysis of these results within the framework of the boundary-layer theory is underway and will be presented in a forthcoming publication.

The effect of coupling coagulation with UF can be seen in Fig. 3, which shows the permeate flux as a function of VCF for stirred-cell UF using a 30 kDa PES membrane at 0.29 MPa for 50°Brix feed. It must be emphasized here that the floc formed after coagulation by addition of the optimum dosage of coagulant was not removed before UF. Even though the initial flux is higher after coagulation, it soon stabilizes to around the same value of 1.4×10^{-5} $m^3/m^2 \cdot s$ (50 lmh) both with and without coagulation. This is as expected, because even in the case of the coagulated feed a viscous barrier layer of rejected species would develop, causing the UF to become self regulatory as in the case without coagulation. This phenomenon of self regulation was seen irrespec-

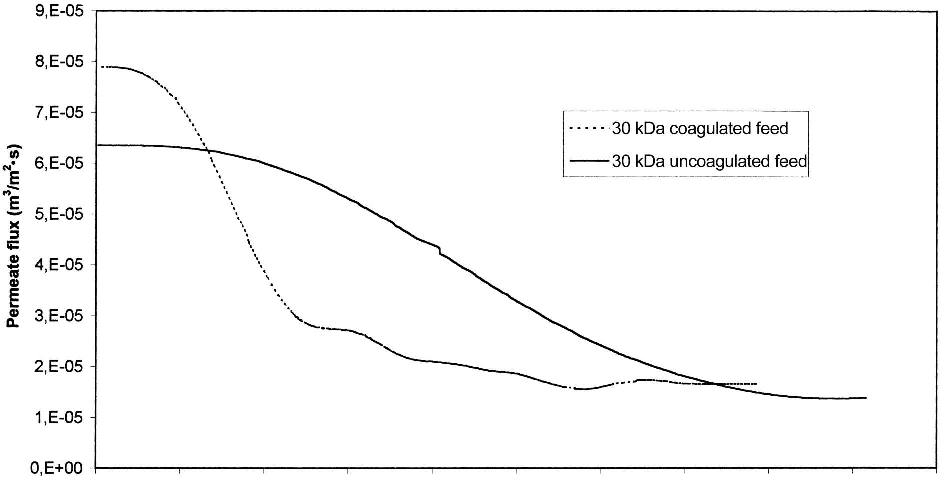

FIG. 3 Variation of permeate flux as a function of volume concentration factor using a 30 kDa PES membrane (frontal filtration) for coagulated and uncoagulated 50°Brix raw Indian sugar solutions; 80°C, 0.29 MPa.

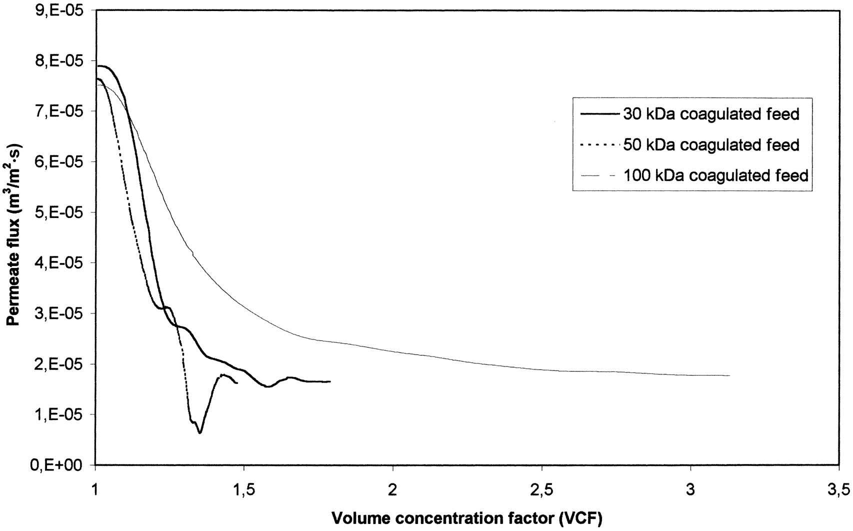

FIG. 4 Variation of permeate flux as a function of volume concentration factor using 30, 50, and 100 kDa PES membranes (frontal filtration) for coagulated 50°Brix raw Indian sugar solutions; 80°C, 0.29 MPa.

tive of the MWCO as shown in Fig. 4 for coagulated 50°Brix feed solution at 0.29 MPa; although it takes much longer for the 100 kDa membrane compared to the 30 and 50 kDa membranes.

The steady-state color reduction for 50°Brix feed solutions using each of the PES membranes with and without coagulation is shown in Fig. 5. As expected, the color rejection decreases as the MWCO of the membrane increases. For the 5 kDa membrane, the color reduction is greater than 80%, but the corresponding steady-state flux (~1.9×10^{-6} m^3/m^2·s; ~7 lmh) is not economically viable, as discussed by Nielsen et al. (14). For the 30 and 50 kDa membranes, the reduction with coagulation is around 45%, which, coupled with a flux of around 1.4×10^{-5} m^3/m^2·s (50 lmh), makes them interesting candidates for industrial application. As discussed by Nielsen et al., a membrane capable of 50% color reduction along with 1.4×10^{-5} m^3/m^2·s (50 lmh) steady permeate flux would be an interesting economic option for the sugar industry.

Of course, this color reduction by membrane can replace the actual process of color reduction using chemicals, and the retentate (wastes) can also be valorized and the total processing time can be reduced. Also, as membrane prices fall, the economic feasibility of applying membranes to this process increases.

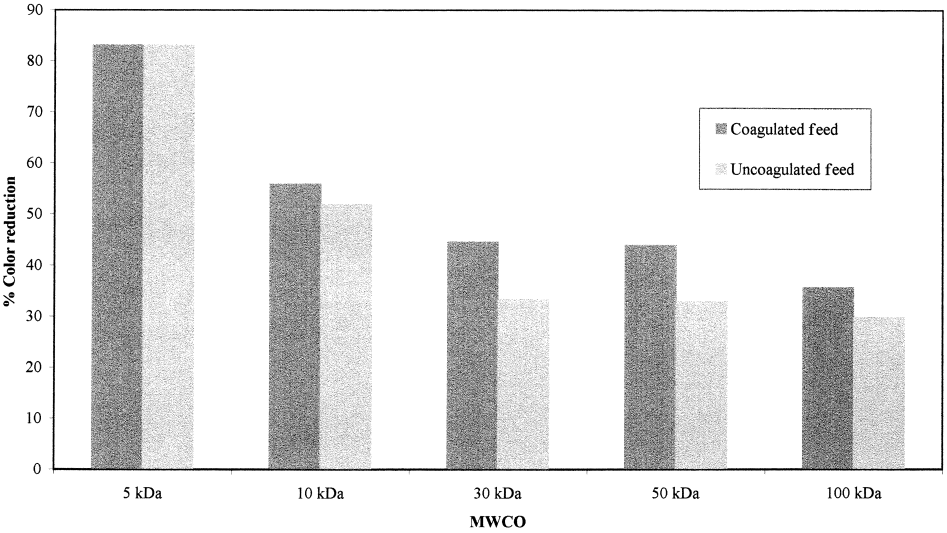

FIG. 5 Steady state percentage of color reduction using various MWCO PES membranes (frontal filtration) for coagulated and uncoagulated 50°Brix raw Indian sugar solutions; 80°C, 0.29 MPa.

Figure 6 shows the variation of permeate flux with time with mineral membranes for a cross-flow velocity of 2.5 m/s, transmembrane pressure of 0.44 MPa, and at the indicated temperatures. The cross-flow filtration was operated in a concentration mode, i.e., the retentate was recycled back into the feed reservoir while the permeate was continuously removed. In all cases, the permeate flux becomes quasi-steady after a volume concentration factor of between 2 and 3. The 50 kDa membrane was used for both coagulated and uncoagulated 50°Brix feed solutions. The flux for the coagulated feed was slightly lower as compared to that for the uncoagulated feed.

The 50 kDa membrane at 80°C is seen to give a steady permeate flux of around 1.4×10^{-5} $m^3/m^2{\cdot}s$ (50 lmh) at 0.44 MPa. All other runs in the cross-flow mode were conducted at 70°C because of the heating limitations of the installation. This data generated in the study is currently being analyzed using a new unsteady-state theory for UF of macromolecules. This detailed analysis will be presented in a forthcoming publication.

Figure 7 shows the color reduction in cross-flow UF (at 70°C) for the various membranes used. As expected, the color reduction is extremely good using a 15 kDa membrane. The flux however is economically uninteresting ($<$10 lmh) as discussed by Nielsen et al. (14). The reduction for the 30 and 50 kDa membranes is similar to the 50-kDa membrane giving a higher permeate

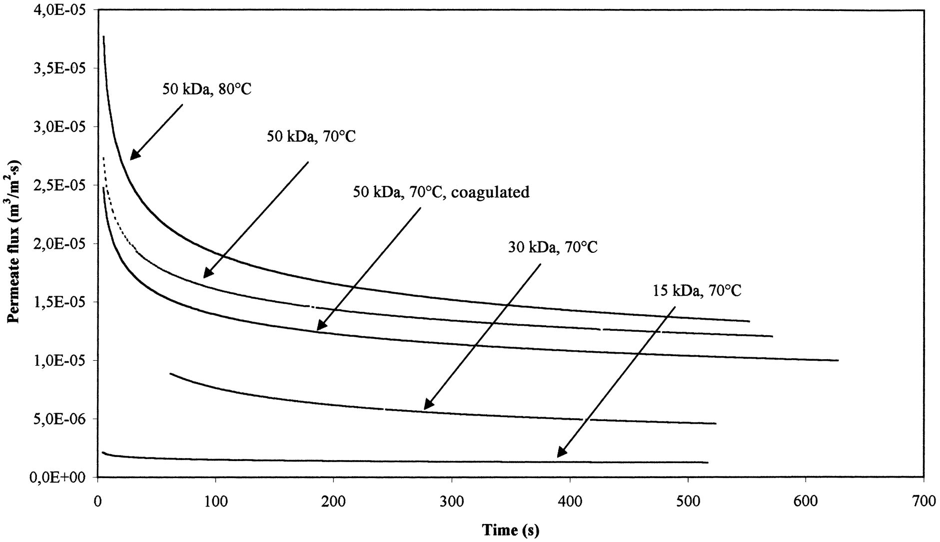

FIG. 6 Variation of permeate flux as a function of time for various MWCO mineral (CARBOSEP) membranes for 50°Brix raw Indian sugar solutions at indicated temperatures; 2.5 m/s cross-flow velocity, 0.44 MPa.

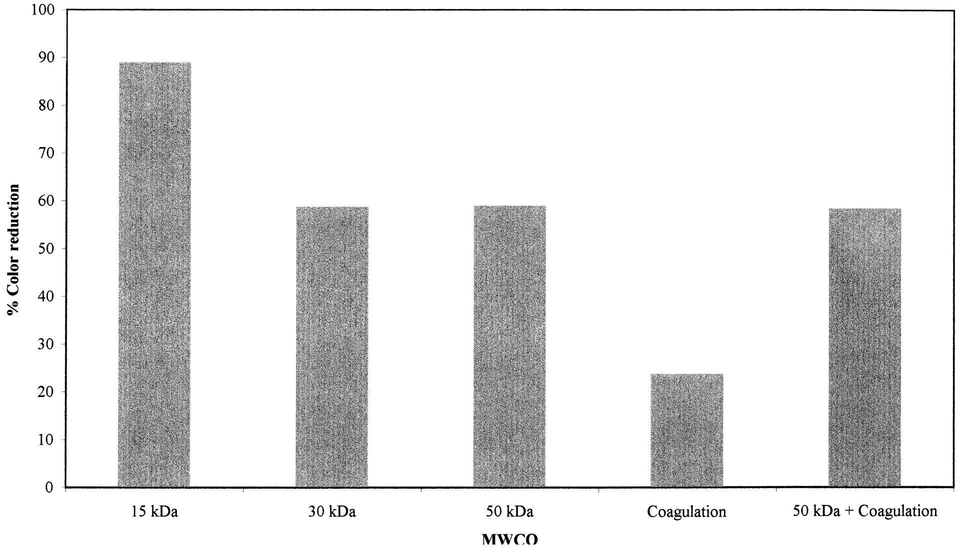

FIG. 7 Steady state percentage of color reduction for various MWCO mineral (CARBOSEP) membranes at 70°C for 50°Brix raw Indian sugar solutions; 2.5 m/s cross-flow velocity, 0.44 MPa.

flux. The color reduction for the 50-kDa membrane both with and without coagulation was almost identical. Hence, coagulation does not seem to increase the efficacy of the decolorization process.

These results seem to indicate that the optimum cutoff for decoloring raw Indian sugar solutions is between 30 and 50 kDa. However, long-term experiments would determine whether is it possible to sustain high enough permeate flux values to make the process technologically or economically interesting. This extended time study is in progress and will be detailed in a subsequent publication.

CONCLUSIONS

The organic (PES) and inorganic (ceramic) membranes used in this study demonstrated satisfactorily qualitative performances for reducing color and for obtaining an economically interesting permeate flux for MWCO between 30 and 50 kDa. The traditional methods used in the sugar industry do not effectively remove substances such as starch, dextran, waxes, bacteria, and sucrose. Membrane ultrafiltration could become an interesting alternative if stable permeate fluxes of around 1.4×10^{-5} $m^3/m^2 \cdot s$ (~50 lmh) can be achieved. These fluxes have been obtained from UF membranes of 30–50 kDa molecular weight cutoff.

The coagulation of raw Indian sugar solutions with only a cationic polyelectrolyte (at 80°C) removed about 25% of the color at an optimum concentration of 1.5 ppm. Furthermore, this dosage was found to remain unchanged, irrespective of the °Brix of the solution. The coupling of coagulation with UF for the treatment of a 50°Brix solution gave a steady permeate flux of 1.4×10^{-5} $m^3/m^2 \cdot s$ (50 lmh) and a reduction in color of about 50% with both types of membranes of 30 and 50 kDa molecular weight cutoff. The filtration seems to be regulated by formation of a concentration polarization layer, which can be a function of the retention of some species of moderate molecular weight and that are not removed by cationic flocculation.

It was observed that coagulation of the solution did not increase the efficacy of the decoloration process. In addition, the UF process uses almost no chemicals for clarification and color removal of raw sugarcane juice; hence, it is possible to partially alleviate the pollution problem for the sugar industry.

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