Journal of Membrane Science 322 (2008) 349–359

Contents lists available at ScienceDirect

Journal of Membrane Science

journal homepage: www.elsevier.com/locate/memsci

Pectin recovery from model solutions using a laboratory-scale ceramic tubular UF membrane module

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ARTICLE INFO

Article history:
Received 9 January 2008
Received in revised form 5 May 2008
Accepted 31 May 2008
Available online 11 June 2008

Keywords:
Ceramic tubular ultrafiltration module
Citrus pectin
Laboratory-scale plant
Limiting permeation flux
Rheology
Solute concentration
Superficial velocity
Transmembrane pressure difference

ABSTRACT

In this work the feasibility of the recovery of a commercial sample of citrus pectin from model solutions using a temperature- and pressure-controlled bench-top ultrafiltration (UF) plant equipped with a ceramic tubular, 20 kDa nominal molecular weight cut-off (NMWCO), UF membrane module, was assessed.

Several total recycle tests allowed the assessment of the effects of transmembrane pressure difference (ΔP), feed superficial velocity (v_S) and solute concentration (c_{BR}) in the ranges of 0.4–4.2 bar, 4–6 m s^{-1}, and 2.2–30.4 kg m^{-3}, respectively, on the permeation flux under a constant process temperature of 50 °C.

As c_{BR} increased from about 2.2 to 10 kg m^{-3} the limiting permeation flux ($J_{P\infty}$) almost linearly decreased from 125–195 to about 25–40 dm^3 m^{-2} h^{-1}, these values depending on v_S. For c_{BR} rising from ~10 to 31 kg m^{-3}, $J_{P\infty}$ was almost constant or slightly tended to decline. The change in slope of the plot $J_{P\infty}$-vs.-log(c_{BR}) was attributed to the transition from turbulent to laminar regime.

Two dimensionless empirical regressions among the modified Sherwood, Reynolds and Schmidt numbers, valid in the laminar or turbulent flow regime, enabled the estimation not only of the $J_{P\infty}$ values detected here in the total recycle mode, but also of the permeation fluxes observed by Pritchard et al. [M. Pritchard, R. Field, J.A. Howell, The ultrafiltration of viscous liquids, J. Membr. Sci. 102 (1995) 223–235] using a polyethersulphone, 65-kDa NMWCO, tubular module operating at 45 °C, ΔP = 2.5 bar and different feed superficial velocities (1.3–2.7 m s^{-1}) with about 21% deviation. Moreover, such relationships yielded satisfactory simulation of two other independent validation tests performed in the batch mode with an average percentage error between the theoretical and experimental fluxes of 15%.

1. Introduction

Pectin for commercial use has been defined by the Joint FAO/WHO Expert Committee on Food Additives [1] as composed mainly of partial methyl esters of polygalacturonic acids and their sodium, potassium, calcium and ammonium salts.

In its basic extraction process fresh or dried citrus peel or apple pomace is washed with demineralised water at pH 2 and temperature of 70 °C for not less than 3 h [2].

After removal of exhausted peel or pomace by for instance continuous under vacuum filtration, the extract is clarified and pectin precipitated by using no other organic precipitants than methanol, ethanol, or isopropanol [1], the overall alcoholic titre being higher than 45% (w/v). The alcohol-wet precipitate is washed to remove contaminants in the form of heavy metals, acid, sugars, polyphenols, pigments, and other alcohol-soluble material [3]. Finally, it may be dried and milled directly or de-esterified in alcohol suspension.

The clarified extract may be concentrated in multistage evaporators to 3–5% pectin content to reduce the amount of either alcohol or live steam used to flocculate the biopolymer or re-distillate the precipitant from exhausted alcoholic media, respectively, and thus to minimise pectin specific recovery costs [3,4].

By accounting for the potential application of membrane separation processes in the food industry to re-design classic down-stream operations for relatively low energy requirements, no additives, generally mild operating conditions and easy scaling-up rules [5], pectin thickening might be alternatively achieved by ultrafiltration (UF) membrane processing. In this way, it would be possible not only to reduce the energy consumption from 30–50 to 10–25 kW h/m^3 of water evaporated, but also to improve the overall quality of the final product by resorting to appropriate nominal molecular weight cut-off (NMWCO) UF membrane modules to get rid of either the pectin macromolecules of lower molecular mass or the aforementioned contaminants [4]. Finally, further UF treatment

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doi:10.1016/j.memsci.2008.05.066

of wastewaters might enhance the overall pectin extraction yields and, consequently, minimise their pollution charge.

To this end, it is worth citing the use of UF hollow-fibre, 500 kDa NMWCO modules to increase the xanthan gum present in the exhausted culture broths of *Xanthomonas campestris* from about 3 to 7% (w/w) with a reduction in the operating costs from US$ 2.7 to US$ 1.5 per kg of biopolymer recovered [6].

Several authors [4,7–9] have so far investigated the recovery of pectin by using 20–100 kDa NMWCO, organic or ceramic hollow fibre, plate-and-frame or tubular membrane modules at 45–75 °C and average permeation fluxes ranging from 20 to 35 $dm^3\,m^{-2}\,h^{-1}$ [4].

The aim of this work was to model mathematically the UF recovery of a target citrus pectin from model solutions using a laboratory-scale plant, appropriately assembled and equipped with a ceramic tubular membrane module, as a preliminary step in the assessment of an alternative UF process flow scheme for thickening clarified pectin extracts.

2. Materials and methods

2.1. Raw materials

In this work a sample of pectin (Sigma, EC no. 232-553-0, batch no. 900-69-5), extracted from citrus fruits, was used. It had a galacturonic acid content of 93.5% with a degree of methyl esterification (DE) of 63–66% and a methoxy content of 9.4%. The unesterified carboxyl groups are predominantly in the un-ionised (acid) form, giving solutions of pH ~3.4 at 1 $kg\,m^{-3}$ or lower (pH 3.2) at 10 $kg\,m^{-3}$. In all tests the sample having a moisture content of $6.8 \pm 0.2\%$ (w/w) was dissolved in 0.1 $kmol\,m^{-3}$ NaCl, this solvent being regarded as a good one for minimising pectin aggregation whatever the DE between 30 and 72% [10].

2.2. Rheometrical determinations

A #50-Cannon-Fenske capillary viscometer [11] was used to determine the kinematic viscosity (ν) at 25 and 50 °C of pectin dispersions with solute concentrations (c_B) ranging from 0 to ~10 $kg\,m^{-3}$ using the ASTM procedure [11]. Once their corresponding density (ρ) had been measured with calibrated volumetric flasks, it was possible to estimate their dynamic viscosity ($\eta = \nu\rho$).

The rheological behaviour of pectin dispersions for c_B varying from 4 to ~56 $kg\,m^{-3}$ was assessed at 50 °C by using a dynamic stress rheometer type RS200 (Rheometric Scientific Inc., Piscataway, NJ, USA) equipped with plate and cone sensors with smooth surfaces (type LS-PELT-IC40.04) with a cone diameter of 40 mm and a cone angle of 0.0405 rad. The dynamic stress sweep tests were performed by applying a shear stress increasing exponentially with time from 0.1 to ~25 Pa while monitoring the resulting strain, as described previously [12].

One-min delay was assumed to be sufficient to remove practically all the shear history and to reconfigure the unperturbed state of any sample before any testing.

2.3. Laboratory-scale UF membrane plant

Fig. 1 shows the process flow sheet of the temperature- and pressure-controlled bench-top laboratory-scale UF plant used here.

The membrane module (MM) was a ceramic (ZrO_2–TiO_2) monochannel tubular one (US Filter, Warrendale, PA, USA) with 10-mm external diameter, 500-mm module length, 6-mm channel diameter, 20-kDa cut-off and 0.00942-m^2 effective surface area. Digital pressure transducers (TP), with a maximum scale pressure (P_{ts}) of 5 bar were attached at the MM feed inlet and retentate outlet; while a Bourdon manometer (P_{ts} = 8 bar) was connected to the MM permeate one.

The process temperature was monitored and automatically controlled by an on-off temperature-controller (TC), that regulated the flow of heating water (WW), as provided by a thermostat type LTD6 (Grant Instrument Ltd., Cambridge, UK), through the heat exchanger (HE).

A rotameter (F1), equipped with a float, type SK 11 (Georg Fischer, Schaffhausen Schweiz, CH), was used to measure the feed volumetric flow rate (Q_F) in the range of 0.1–1.0 $m^3\,h^{-1}$ (as referred to a liquid of relative density of 1.05 at 20 °C). Another rotameter (F2) in Trögamid-T (nominal diameter of 10 mm and a polypropy-

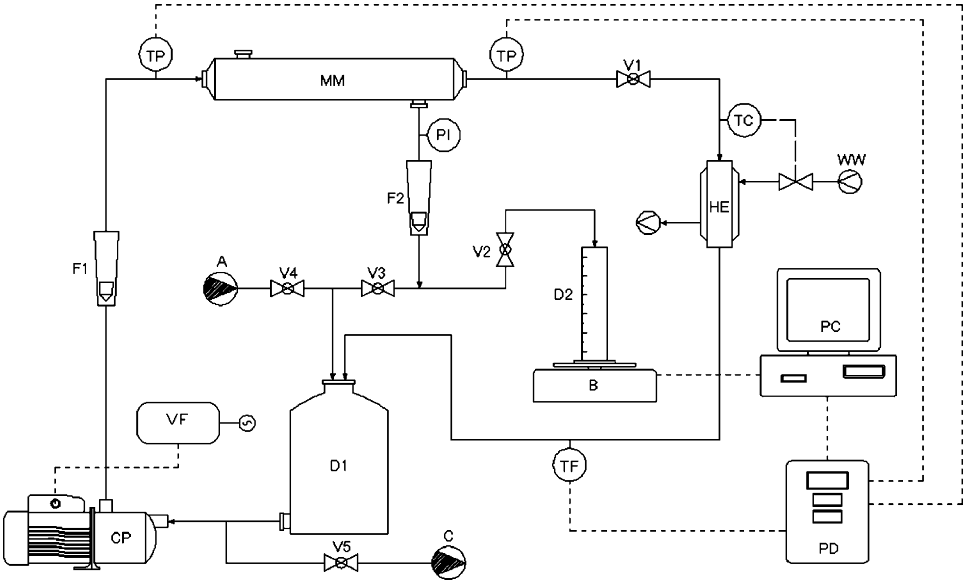

Fig. 1. Process flow sheet of the laboratory-scale UF membrane plant used in this work. Equipment identification items: A, feed; B, balance; C, concentrate; CP, centrifugal pump; D, tank; F, rotameter; HE, heat exchanger; MM, UF membrane module; PC, personal computer; PD, control and display cabinet; PI, manometer; TC, temperature controller; TF, digital flowmeter transducer; TP, digital pressure transducer; V, manual valve; VF, frequency inverter; WW, heating water.