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The potential impact of membrane cascading on downstream processing of oligosaccharides

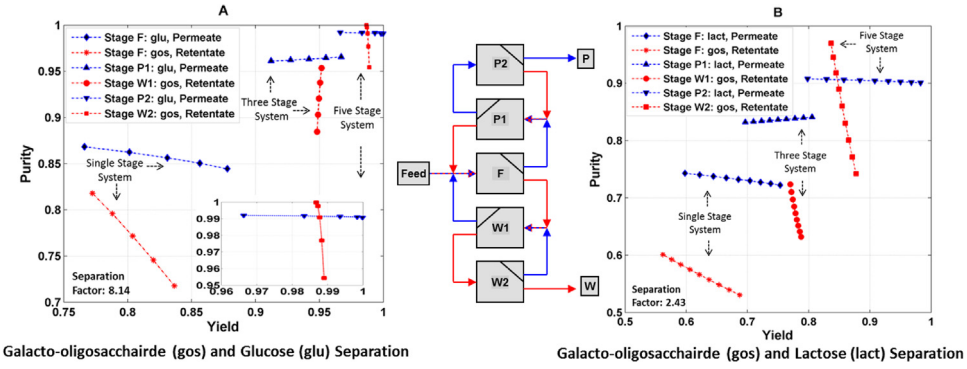
Nirmal V. Patil [a,b,*], Anja E.M. Janssen [b,**], Remko M. Boom [b]

[a] Institute for Sustainable Process Technology, P.O. Box 247, 3800 AE Amersfoort, The Netherlands
[b] Wageningen University, Food Process Engineering Group, P.O. Box 17, 6700 AA Wageningen, The Netherlands

HIGHLIGHTS

- Nanofiltration cascades attractive for the industrial fractionation of oligosaccharides.
- Up to five stage ideal cascade performance simulated with diafiltration, high yield and high purity obtained in five stage systems.
- In contrast to three stage, five stage cascade gives inverse relation of yield and purity.
- Ratio of sieving coefficients plays a significant role on cascade shape and design.
- Membrane area evaluation for each stage gives good process design handle.

GRAPHICAL ABSTRACT



Galacto-oligosacchairde (gos) and Glucose (glu) Separation

Galacto-oligosacchairde (gos) and Lactose (lact) Separation

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ABSTRACT

To assess the potential use of ideal nanofiltration cascades for the industrial fractionation of oligosaccharides, simulations of single, three and five stage NF cascades were carried out. Three and five stage ideal cascades show significant improvement in separation with diafiltration compared to single stage systems. The calculations do imply different membrane areas in each stage of the cascade. The ratio of the sieving coefficients of a binary mixture over the membrane plays an important role in determining the relation between yield and purity in a cascade system. At high sieving coefficient ratios, both yield and purity increase concurrently in a three stage system, whereas at a low ratio of the sieving coefficients, the yield and purity become inversely proportional on the retentate side. In a five stage systems, both yield and purity become inversely proportional at high and low sieving coefficient ratios. A five stage cascade system installation would be optimal for most applications since at very low local separation factors sufficient separation and yield could be achieved.

1. Introduction

Oligosaccharides such as Fructo-Oligosaccharides (FOS) and Galacto-Oligosaccharides (GOS) have received much attention because of their functionality as prebiotic compounds (Gibson and Roberfroid, 1995) in foods. Prebiotic oligosaccharides, usually consisting of 2–10 linked sugar monomers, can be produced with

* Corresponding author at: Wageningen University, Food Process Engineering Group, P.O. Box 17, 6700 AA Wageningen, The Netherlands.
Tel.: +31 317 483770; fax: +31 317 482237.
** Corresponding author. Tel.: +31 317 482231; fax: +31 317 482237.
E-mail addresses: nirmal.patil@wur.nl, nirmalpatil02@gmail.com (N.V. Patil), anja.janssen@wur.nl (A.E.M. Janssen).

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enzymatic transglycosylation synthesis reactions catalyzed by glycosidase (Gibson and Roberfroid, 1995). The product mixtures generally contain next to the oligosaccharides themselves, low molecular weight sugars such as glucose, fructose, sucrose, lactose, and galactose. These low-molecular weight sugars do not contribute to the beneficial properties of the mixture (Goulas et al., 2002), but do influence the sensory perception of the product (e.g., sweetness), and increase the caloric content of the products mixture. Removal of these low-molecular weight components adds to the value of the product, while recycling of some of these sugars to the reaction mixture could possibly enhance the overall conversion.

Various methods have been explored to purify oligosaccharides such as adsorption to activated charcoal (Lee et al., 2004), ultrafiltration (UF) (Nabarlatz et al., 2007), nanofiltration (NF) (Catarino et al., 2008; Goulas et al., 2003, 2002; Li et al., 2004), reverse osmosis (RO), and Simulated moving bed (SMB) technology (Geisser et al., 2005; Masuda et al., 1993). NF membranes have potential in the purification and concentration of oligosaccharide mixtures because their upscaling is straightforward (Catarino et al., 2008; Goulas et al., 2003, 2002; Li et al., 2004, 2005; Lopez Leiva and Guzman, 1995; Wang et al., 2002; Pinelo et al., 2009). NF is efficient in energy, and the operational parameters (pressure, temperature etc.) can be optimized easily. More fundamentally, pressure driven membrane separations have significantly increased transport rates compared to the diffusion dominated chromatographic separations (Lightfoot, 2005; Lightfoot et al., 2008).

Single stage nanofiltration is however limited by the relatively low purities that can be achieved. Multiple stage operation may improve its performance significantly, as was proposed by Gunderson et al. (2007), Lightfoot (2005) and Lightfoot et al. (2008). The principle is inspired on multiple stage separations using thermodynamic equilibrium over different aggregation states, such as in distillation. Gunderson et al. (2007), Lightfoot (2005) and Lightfoot et al. (2008) showed improved separation performance by using a three stage ideal membrane cascade combining diafiltration and UF for the fractionation of binary mixtures. This ideal membrane cascade was illustrated for the separation of α-lactalbumin and β-lactoglobulin. Given the moderate retentions and separation factors in NF, this technology seems to be well suited to be applied in a cascade, as this configuration could provide almost complete removal or retention of a component from a mixture of molecules having very similar retention behavior (such as oligosaccharides) in an individual stage. Recently, other authors (Abejón et al., 2012; Alexander Caus et al., 2009; Mayani et al., 2010, 2009; Vanneste et al., 2013) have studied membrane cascade based separation, however they have considered non-ideal cascade approach.

In this paper the potential of ideal nanofiltration cascades was assessed for the fractionation of industrial oligosaccharide mixtures. Simulations of the performance of three and five stage cascades were compared to those of single stage systems, both for the permeate and the retentate products. The effects of the separation factor on the yield and purity were examined and the required dimensionless membrane area were evaluated, which are required for the configuration and design of a practical cascade.

2. Single stage nanofiltration separation of oligosaccharides

Goulas et al. (2002) reported the separation of oligosaccharides from mono-(glucose) and disaccharides (lactose) by using various NF membranes in continuous diafiltration. They studied different types of membranes for the separation of a mixture of raffinose, sucrose and fructose and a commercial mixture of oligosaccharides named Vivinal® GOS. The studied membranes included two flat sheet asymmetric cellulose acetate membranes, NF-CA-50 and UF-CA-1 from Intersep Ltd., and three composite membranes, DS-5-DL, DS-51-HL, and DS-GE from Osmonics. We have based our calculations on the experimental data with NF-CA-50 and DS-5-DL membranes, since these give the most suitable single stage performance (*v.* Table 1).

Table 1
Single stage nanofiltration membrane experimental data used for the nanofiltration membrane cascade simulations obtained from Goulas et al. (2002).

Category	R[d]	S[d]	Φ_L[d]	Feed conc.	
				g/l	mmol/l
Vivinal GOS[a]					
NF-CA-50[b]					
Oligos	0.93	0.07		32.0	61.3
Lactose	0.83	0.17	2.43	31.7	92.7
Glucose	0.43	0.57	8.14	15.5	86.4
DS-5-DL[ce]					
Oligos	0.99	0.01		32.0	61.2
Lactose	0.96	0.04	4.00	31.6	92.4
Glucose	0.43	0.57	57.00	15.5	86.1

[a] Feed concentration composition: Oligosaccharides 40.4%, lactose, 40.0% and glucose 19.6%.
[b] 25 °C, 13.8 bar.
[c] 60 °C, 13.8 bar.
[d] Rejection co-efficient (R), sieving coefficient (S) and local separation factor (ϕ_L) calculated from Eqs. (3) and (8).
[e] Rejection and feed concentration values had been interpreted from Fig. 7 of Goulas et al. (2002).

3. Single stage nanofiltration membrane: simulation theory

3.1. Diafiltration

Diafiltration is filtration while supplying pure solvent to the feed and washing out any component that is not retained by the membrane. An overall diafiltration process may involve a preconcentration step, the diafiltration step and a post concentration step (Dutré and Trägårdh, 1994). The diafiltration process may operate with constant volume (i.e., the inflow of solvent Q_D is just as large as the outflow Q_P) or with variable volume (in which the inflow may be larger or smaller than the outflow).

Diafiltration in a process having a total area of the membrane A (m^2) and total volume of the feed solution V_f, (m^3) for component i with respect to time t (h) and transmembrane flux J_v ($m^3\ m^{-2}\ h^{-1}$) through the membrane is expressed by

$$\frac{d(V_f C_{f,i})}{dt} = -J_v A C_{P,i} \tag{1}$$

where $C_{f,i}$ and $C_{p,i}$ are the molar concentration (mol/m^3) in the feed and permeate of molecule i respectively.

For a constant volume diafiltration process (see Fig. 1A and B), the amount of solvent added ($V_D = Q_D, \ldots, t$, diafiltration volume) is equal to the amount of permeate produced, which yields

$$\frac{dV_f}{dt} = 0 \quad \text{and} \quad \frac{dV_D}{dt} = J_v A \tag{2}$$

In our calculations we assume constant rejection coefficients, R_i, for all components. The sieving coefficients, S_i are related to the retention with

$$R_i = 1 - C_{P,i}/C_{f,i} \quad \text{or} \quad S_i = 1 - R_i \tag{3}$$

Combining Eqs. (1)–(3) and with $C_{f,i} = C_{f_0,i}$ at $t = 0$, we can arrive at the following dimensionless equation:

$$\frac{C_{f,i}}{C_{f_0,i}} = \exp\left[-\frac{V_D}{V_O}(1 - R_i)\right] \quad \text{or} \quad \frac{C_{f,i}}{C_{f_0,i}} = \exp[-\tau s_i] \tag{4}$$