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Technical and Economic approach of bioethanol production from nanofiltration of biomass chemical hydrolysis solutions

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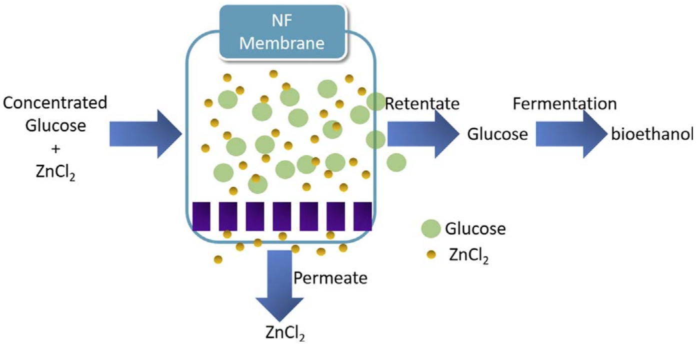
HIGHLIGHTS

- Nanofiltration achieves *SF* up to 10 for 120 g/L $ZnCl_2$ and 30 g/L glucose.
- Retention of glucose reaches 95% in a solution of 120 g/L $ZnCl_2$ by nanofiltration.
- The rejection of $ZnCl_2$ depends on Donnan exclusion and dielectric exclusion.
- Desalinated bagasse hydrolysis sugar solution achieved 82% yield of bioethanol.
- The availability of the membranes makes the industrial process scale-up easily.

GRAPHICAL ABSTRACT



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ABSTRACT

The effects of pH, transmembrane pressure, and $ZnCl_2$ concentration on the purification of glucose from solution through nanofiltration were investigated. The solutions used contained $ZnCl_2$ and glucose in concentrations as high as 180 and 80 g/L, respectively. A high concentration of glucose was discovered in the retentate as the product, and $ZnCl_2$ was filtered out into the permeate; the highest separation factor achieved was 10. The derivative Hagen–Poiseuille equation was applied to model the experimental data and calculate the effective thickness and pore radius of the membranes used. Moreover, an integrated multiple-nanofiltration system was designed to process a real bagasse hydrolysis solution. The fermentability of the purified sugar solution was confirmed, with an 82% total alcohol yield obtained. The multiple-nanofiltration system is both economically and environmentally sustainable for the production of bioethanol through chemical hydrolysis of biomass.

1. Introduction

Lignocellulosic biomass is one of the most abundant materials on earth [1] and approximately 200 billion metric tons is produced worldwide each year [2]. Lignocellulose is also a sustainable alternative energy source to fossil fuel products such as fuel and chemicals. Researchers at the National Renewable Energy Laboratory of the US Department of Energy have defined five platforms of conversion technologies for using biomass to generate base chemicals that can subsequently be employed by industry to produce a wide range of fuels, chemicals, and materials [3]. Among these, the sugar platform is one of the most promising for the production of fuels such as ethanol and other building block chemicals.

Currently, bioethanol is produced from biomass mainly through enzymatic processes [4], which have the advantages of lower temperatures and higher selectivity [5]. However, the conversion involved

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Nomenclature

Ak	porosity of the membrane
C_p	concentration in the permeate (mol m^{-3})
C_r	concentration in the retentate mol m^{-3}
$D_{i,\infty}$	bulk diffusivity of solute i (m^2/s)
G	hydrodynamic enhanced lag coefficient
J	volume flux ($\text{m}^3\,\text{m}^{-2}\,\text{s}^{-1}$)
k	mass transfer constant
$K_{i,c}$	hindrance factor for convection
$K_{i,d}$	hindrance factor for diffusion
Pem	Peclet number
R_{obs}	observed rejection
r_p	mean pore radius (m)
R_{real}	real rejection
r_s	solute Stokes radius (m)
Δ_x	membrane thickness (m)
$\Delta\pi$	the osmotic pressure difference
λ	ratio of solute to pore radius
μ	viscosity of solution (kPa s)
δ	film thickness
Φ	steric partition term

is extremely sensitive to the feedstock used [6]. In addition, many enzymatic processes must be improved and their energy input reduced [7]. These issues are being addressed by research on, for example, standardization of enzymatichydrolysis [8], feedstock pretreatments [9–11], and thermostable hydrolytic enzymes [12,13].

Hydrolysis alternatives such as concentrated mineral acids (HCl, H_2SO_4, H_3PO_4) [14,15], molten salt hydrates [16], and ionic liquids [17–19] have been investigated in the field. Sugar purification and chemical recycling play technically and economically crucial roles in the hydrolyzation of crystallized polysaccharides [6,20–22]. In 1984, the Purdue Research Foundation patented the hydrolysis of cellulose into glucose using a solution of zinc chloride ($ZnCl_2$) [23]. The Industrial Technology Research Institute (ITRI) has developed an ionic solution, which has a high concentration of $ZnCl_2$, to improve the yield of glucose from cellulose. This ionic solution is much cheaper than conventional ionic liquids and achieves a > 90% yield of glucose from lignocellulosic biomass [24]. The chemical conversion does not require pretreatment and has a short reaction time (total reaction time < 3 h). Most importantly, the chemical process can be employed to treat all types of feedstock without adjustment of its formula. Therefore, this process is more economically competitive than the enzymatic conversion of cellulose. However, the cost of purifying the hydrolysis sugar and recycling chemicals has limited its economic viability.

Nevertheless, a cost-effective process for converting highly concentrated streams is required if chemical hydrolysis for bioethanol production is to be commercialized. Several studies have analyzed the influence of high concentrations of charged solutes on nanofiltration and the effect of salts on glucose retention [25–27]. Nanofiltration membranes have some of the properties of both ultrafiltration and reverse osmosis membranes and have an even wider range of application [28,29]. The commercial application of nanofiltration membranes currently focuses on desalination, especially in food processes. Numerous studies have revealed the effects of solute charge, pH, and temperature on the characteristics of nanofiltration membranes [30–33]. The results have indicated that using salt in the solution may reduce the retention of glucose [34–37] for three main reasons: (1) the membrane swelling effect alters the pore radius of the membrane [35]; (2) the flux effect shifts the pore size distribution; and (3) the charged solutes change the effective molecular size of the neutral component [34].

To the best of our knowledge, this study is the first to desalinate a high concentration of $ZnCl_2$ salt (> 120 g/L) from a real biomass chemical hydrolysis solution using nanofiltration. The present study evaluated the effects of $ZnCl_2$ concentration, transmembrane pressure, and pH on glucose retention in the nanofiltration process. Finally, multiple nanofiltration processes were designed to purify sugar from a real bagasse hydrolysis solution. The nanofiltration successfully desalinated the hydrolysis sugar, and the sugar was subsequently fermented with yeast to produce bioethanol, the quality of which was verified. The optimized nanofiltration parameters can be used with available nanofiltration membranes for industrial use.

2. Theoretical background

To calculate the pore radius of different membranes using a neutral solute solution, the first step is to convert the observed retention into the real retention [38,39]:

$$\ln\left(\frac{1-R_{obs}}{R_{obs}}\right) = \ln\left(\frac{1-R_{real}}{R_{real}}\right) + \frac{J}{k} \tag{1}$$

where the mass transfer constant (k) is defined by [40]

$$k = \frac{D_{i,\infty}}{\delta} \tag{2}$$

$D_{i,\infty}$ is the diffusion coefficient at infinite dilution, and δ is the film thickness, for which 2.0×10^{-5} m was employed in one study [28].

The observed retention is defined as

$$R_{obs} = 1-\frac{C_p}{C_r} \tag{3}$$

where C_p and C_r are the concentration in the permeate and retentate, respectively.

The Nernst–Planck equation can be written in terms of the real rejection of the solute by the membrane:

$$R_{real} = 1-\frac{\Phi K_{i,c}}{1-e^{(-Pe_m)}[1-\Phi K_{i,c}]} \tag{4}$$

where the steric partition term Φ (defined in Eq. (6)) is directly related to the ratio λ of the solute radius to the pore radius (Eq. (7)). The Peclet number Pe_m is defined as

$$Pe_m = \frac{K_{i,c}J}{K_{i,d}D_{i,\infty}}\frac{\Delta_x}{A_k} \tag{5}$$

where $K_{i,d}$ and $K_{i,c}$ are the hindrance factors of convection and diffusion, defined in Eqs. (8) and (9), respectively. $D_{i,\infty}$ is the bulk diffusivity (m^2/s), and Δ_x/A_k is the effective membrane thickness (m).

$$\Phi = (1-\lambda)^2 \tag{6}$$

where the ratio of the solute to the pore radius, λ, is defined as the solute Stokes radius divided by the mean pore radius:

$$\lambda = \frac{r_s}{r_p} \tag{7}$$

Table 1
Pure water permeance, NaCl retention, and MWCOs, as reported in other studies [41,42].

Membranes	(Pure) water permeance ($\text{L m}^{-2}\,\text{h}^{-1}\,\text{bar}^{-1}$)	NaCl retention (%)	MWCO (Da)
NF 40	2.5[a]	≥50 ± 5	300
NF 270	4.5[b]	30–50	200–300 Da

[a] Permeate flow at 225 psi (1.55 MPa) and 77 °F (25 °C), with a membrane surface area of $1.2\,\text{m}^2$.

[b] Permeate flow at 60 psi (0.41 MPa) and 77 °F (25 °C), with a membrane surface area of $1.2\,\text{m}^2$.

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