Separation and Purification Technology 209 (2019) 211–222

Contents lists available at ScienceDirect

Separation and Purification Technology

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

Optimisation of interfacial polymerization factors in thin-film composite (TFC) polyester nanofiltration (NF) membrane for separation of xylose from glucose

K.H. Mah[a], H.W. Yussof[a], M.N. Abu Seman[a,*], A.W. Mohammad[b]

[a] Faculty of Chemical & Natural Resources Engineering, Universiti Malaysia Pahang, Lebuhraya Tun Razak, 26300 Kuantan, Pahang, Malaysia
[b] Department of Chemical and Process Engineering, Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia

ARTICLE INFO

Keywords:
Nanofiltration
Interfacial polymerisation
Separation factor
Optimisation
Xylose
Glucose

ABSTRACT

A tailored thin-film composite (TFC) NF membrane may offer alternative separation technique to widely used chromatographic techniques in separating two monosaccharides with similar properties. The aim of this paper is to pinpoint the optimum condition in preparing TFC membranes with the highest xylose separation factor. To achieve this, curing time, curing temperature, and reaction time were optimised using central composite design (CCD). Polyethersulfone (PES) was used as a support membrane for interfacial polymerisation (IP) of two active monomers, namely triethanolamine (TEOA) and trimesoyl chloride (TMC). The xylose separation factor was chosen as the response for this study. In addition, occurrence of IP reaction was verified by visual interpretation using field emission scanning electron microscope (FESEM). The chemical elements in TFC membrane and its functional groups were determined using FESEM equipped with energy dispersive X-ray and Attenuated total reflectance–Fourier transform infrared (ATR-FTIR), respectively and compared to the initial PES membrane. A quadratic model was developed and tested with analysis of variance (ANOVA). The model was used to simulate and locate the optimum point. The optimum point was within the studied region and validation tests were conducted to confirm this point. The tests showed little error of less than 2% from the predicted optimal points. The optimum IP conditions for xylose separation were 45.25 min, 15.53 min, and 58.4 °C for reaction time, curing time, and curing temperature, respectively. Under these optimum conditions, a maximum xylose separation factor of 1.334 ± 0.007 was achieved. The optimised TFC membrane exhibited comparable xylose separation factor to commercial membranes.

1. Introduction

Biomass generated from agriculture waste contains high content of monosaccharides, such as glucose and xylose, that can be harnessed through thermochemical conversion and subsequent purification steps [1]. Conventionally, monosaccharide purification is performed by chromatography due to the similar physicochemical properties between monosaccharides. For example, xylose and glucose are both non-charged components with small dissimilarities in molecular weight which are 150 and 180 g/mol, respectively. In this recent decade, studies on improving chromatography process have been carried out by investigating the adsorption behaviour of monosaccharides on resins to determine the highest separation factor that can be achieved for each resin [1–4]. Chen and colleagues [1] explored the feasibility to separate major monosaccharides (glucose, xylose, and arabinose) from hydrolysates of lignocellulosic biomass using column chromatography process with cation exchange resin Amberlite IR120 and Amberlite IRP69 in Na^+ and Ca^{2+} forms. Column with Amberlite IRP69 Ca^{2+} form had the highest xylose/glucose selectivity at 1.31. Bi and colleagues [2] studied different imidazolium stationary phases with xylose/glucose selectivity (separation factor) between 1.02 and 1.07. Lei and colleagues [3] studied the adsorption equilibrium of glucose, xylose, and arabinose on five gel-type strong-acid cation exchange resins loaded into identical dimension column. Column loaded with resin crosslinked with 8% Ca^{2+} achieved 1.4 xylose/glucose selectivity, which is the highest. Saari and colleagues [4] studied the four main types of gel-type

Abbreviations: ANOVA, analysis of variance; ATR, attenuated total reflectance; CCD, central composite design; EDX, energy dispersive X-ray; FESEM, field emission scanning electron microscope; FTIR, fourier transform infrared; HPLC, high performance liquid chromatography; IP, interfacial polymerisation; NF, nanofiltration; OFAT, one-factor-at-a-time; RSM, response surface methodology; TFC, thin-film composite; Std, standard run

* Corresponding author.
E-mail address: mazrul@ump.edu.my (M.N. Abu Seman).

https://doi.org/10.1016/j.seppur.2018.07.041
Received 3 March 2018; Received in revised form 14 July 2018; Accepted 14 July 2018
Available online 17 July 2018
1383-5866/

ion-exchange resins: strong acid cation (Na^+), strong base anion (SO_4^{2-}), weak base anion (Na^+), and weak acid cation (SO_4^{2-}), with similar crosslinking and bead sizes using static method. Both weak base anion and strong base anion resins demonstrated good selectivity for xylose. Weak base anion and strong base anion resins had xylose/sucrose selectivity of 1.36 and 1.31, while glucose/sucrose selectivity of 1.00 and 1.12, respectively [4]. This can translate to xylose/glucose selectivity of 1.36 and 1.17 weak base anion and strong base anion resins, respectively.

Chromatography allows for excellent separation and purification of monosaccharides with similar physicochemical properties but requires high amount of resources, time, and energy compared to NF [5]. Membrane filtration shows promising separation and purification of monosaccharides with similar physicochemical properties, where commercial NF membrane exhibits excellent performance in the separation of xylose and glucose from a concentrated mixture with a separation factor between 1.5 and 3.0 [6]. The application of these commercial membranes in different hydrolysates [7] shows new possibility in separating xylose and glucose with membrane technology. Another approach in membrane technology is by converting the glucose to gluconic acid through enzymatic reaction to increase the molecular size, thus allowing for easier separation of xylose in an integrated membrane system using commercial NF membranes [8,9]. High xylose/gluconic acid separation factor has been observed using this technique at 34.0 using sugar model [8], 2.2–2.7 using different biomass hydrolysates [9]. This technique [8,9] uses charge effect mechanism as opposed to sieving effect mechanism in an earlier study done by Sjoman and colleagues [6], where the feed solution pH had significant effect on the permeance flux. However, less attention has been paid to develop membranes tailored specifically for xylose separation from glucose. In our previous work, we demonstrated that self-made TFC membrane via IP was able to attain comparable separation factor with commercial NF membrane [10]. Commercial membranes attained separation factors of 1.47, 2.73, and 1.58 for Desal-5 DL, Desal-5 DK, and NF270, respectively at a pressure of 5 bar [6], while self-made TFC membrane [10] attained a 1.64 separation factor at 4 bar.

In general, IP technique has its polymerisation reaction between two monomers occur at the interface of two liquids that are immiscible to each other [11]. Normally, an amine-based active monomer, *m*-phenylenediamine (MPD), is the one widely used in commercial TFC membrane [12] and is still relevant until now for research purposes [13–18]. Besides that, other amine derivatives, piperazine (PIP) [19,20] and triethanolamine (TEOA) [10,21,22] have also been used for TFC membranes. Tang and colleagues [21] first utilised aqueous TEOA, an environmental friendly yet economical monomer, and trimesoyl_chloride (TMC)–hexane organic solution in preparing a novel TFC membrane. As a tertiary amine group, TEOA monomer molecules can be flexibly transferred into a quaternary ammonium group by variation of feed pH. TFC membrane developed using these monomers increases the hydrophilicity on the membrane surface [21,22], influencing rejection of sugar molecules [23]. Although the selection of TEOA as a monomer in aqueous phase has been particularly explained in the past, the selection of TMC as monomer in organic phase is less explained [21,22,24]. However, TMC has been widely used with MPD in developing commercial membranes via IP [12]. A related study by Liu and colleagues [25] showed three different organic monomers in IP, namely TMC, 5-isocyanato-isophthaloyl chloride, and 5-chloroformyloxy-isophthaloyl chloride, can have different structure–property relationship of TFC membrane using the same aqueous monomer. In the study, TFC membrane developed from TMC–MPD was not the best or worst in terms of membrane hydrophilicity, membrane roughness, pure water permeability, salt rejection, and resistance to chlorine compared with the other membranes. This makes TFC membrane developed using TMC as the organic phase monomer versatile and successful in commercial applications.

To attain the optimal TFC membrane, various factors in preparing TFC membrane have been studied in the past. Preceding studies on TFC membrane usually employ one-factor-at-a-time (OFAT) design in their works. This experimental design requires huge amount of resources and time invested to predict the optimal parameters through a high number of trials and errors. Consequently, new studies have introduced a systematic approach in finding the optimal condition for IP in maximising separation performance using common experimental design such as Taguchi [26], Box–Behnken [27], and central composite [28]. Till now, no report is available on systematic approach particularly in optimising factors in TFC preparation through IP, particularly on separating xylose from glucose.

In our previous work, we prepared TFC membrane for xylose separation by manipulating five preparation factors (reaction time, curing, concentration of aqueous and organic monomers, and pH of aqueous monomer) using fractional factorial design and found that the combination of reaction and curing times may boost the separation of xylose from glucose [10]. As continuation, this study aims to pinpoint the optimum conditions for reaction time, curing time, and curing temperature in preparing TFC membrane with maximum xylose separation factor using response surface methodology (RSM). CCD was initially used to design a total of 20 runs to establish a polynomial regression equation that portrays the relationships between the xylose separation factor and the studied factors. Numerical optimisation and validation of model were performed to find the optimum conditions in preparing TFC membrane that have the highest xylose separation factor with the least amount of deviation if prepared repetitively.

2. Materials and method

2.1. Materials

Commercial ultrafiltration PES asymmetric membrane (UF PES50) obtained from AMFOR Inc. (China) was used as support for IP. Based on the specification sheet provided, MWCO of this membrane is 50 kDa with water flux of 260 L/m^{2}h (operating pressure of 50 psi at temperature of 25 °C). The monomers, chemicals, and solvents used for IP were TEOA (R & M Marketing, Essex, UK), TMC (Alfa Aesar, UK), sodium hydroxide, and *n*-hexane were both purchased from Merck, Germany. The chemicals were of analytical grade. The analytical grade xylose and glucose with high purity (> 99%) used in this study were obtained from Sigma Aldrich, USA. Acetonitrile (J.T. Baker, USA) was used in the preparation of mobile phase for high performance liquid chromatography (HPLC).

2.2. Preparation of TFC membrane

The commercial PES support membranes were cut into disc shape and pretreated before initiating the IP process. This pretreatment was done by first immersing the PES membranes in ultrapure water for at least 12 h, then subjected to ultrasonication for 3 min in ultrasonic bath to remove glycerin preserving the pores. The ultrasonication was repeated once with replacement of fresh ultrapure water in the ultrasonic bath prior to any ultrasonication activity. For storage purpose, treated PES membranes were immersed in ultrapure water in a sealed container. Basically, IP takes place at the interface between two immiscible organic and aqueous solutions. The interface was emulated by immersing the support membrane in a series of aqueous and organic phase monomers solutions. The preparation of TFC membranes in this study was based on the work by Tang and colleagues [21] with slight modifications as done in our previous work [10]. Excess liquid on top of PES membrane was removed by rolling with a glass rod, followed by immersion in 4% (w/v) TEOA aqueous monomer solution for 30 min. The TEOA-loaded membrane was then immersed in 0.25% (w/v) TMC solution for a predetermined time. This IP reaction was conducted at room temperature. Finally, the membrane was dried in an oven at different curing temperatures. The general steps for IP process are