Applied Catalysis A: General 386 (2010) 109–115

Contents lists available at ScienceDirect

Applied Catalysis A: General

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

Extractor-type catalytic membrane reactor with nanocomposite MFI-alumina membrane tube as separation unit: Prospect for ultra-pure para-Xylene production from m-Xylene isomerization over Pt-HZSM-5 catalyst

M.O. Daramola[a,*], A.J. Burger[a], A. Giroir-Fendler[b], S. Miachon[b,1], L. Lorenzen[a,2]

[a] *Department of Process Engineering, Stellenbosch University, Private Bag X1, Matieland 7602, Stellenbosch, South Africa*
[b] *Université de Lyon, Institut de Recherches sur la Catalyse et l'Environnement de Lyon (IRCELYON), UMR 5256 CNRS - Université Lyon 1, 2, Av. A. Einstein, 69626 Villeurbanne Cedex, France*

ARTICLE INFO

Article history:
Received 15 February 2010
Received in revised form 20 July 2010
Accepted 21 July 2010
Available online 30 July 2010

Keywords:
m-Xylene
Isomerization
Reactors
Membrane
Catalysis

ABSTRACT

This paper is a follow-up on our recent study on the applications of extractor-type zeolite catalytic membrane reactor (herewith referred to as e-ZCMR) for m-Xylene isomerization. In this paper, results of a preliminary investigation on the possibility of producing ultra-pure p-Xylene (PX) (purity > 99%) via m-Xylene (MX) isomerization over Pt-HZSM-5 catalyst in an e-ZCMR with a "defect-free" nanocomposite MFI-alumina membrane tube as the separation unit is presented. Unlike "film-like" architectures, in nanocomposite architectures zeolite crystals are embedded within the pores of the supports. During m-Xylene isomerization conducted at a temperature range 473–573 K, liquid meta-Xylene (99% purity) saturated in N_2 gas was fed into the reactor and N_2 gas was swept over the outer surface of the membrane on the shell side of the reactor. Analysis of results was based on permeate-only mode (products in permeate stream only) and combined mode (products in both permeate and retentate) operations. At 473 K, e-ZCMR gave a maximum p-Xylene yield of 2.7% at permeate-only mode and 19.0% at combined mode. Throughout the temperatures investigated, the purity of PX approached 100% in the permeate and the membrane displayed 100% PX selectivity. These results indicate that there is a possibility of cutting down operational costs through a reduction in energy consumption during ultra-pure PX production and that this becomes feasible with the application of e-ZCMR having nanocomposite MFI-alumina membrane as separation unit. However, high flux defect-free nanocomposite MFI-alumina membranes are necessary to make this technology attractive and competitive with those currently in use.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

This paper presents a further study on the development and evaluation of energy-efficient process for p-Xylene production based on the application of MFI-type zeolite membranes in extractor-type zeolite catalytic membrane reactors (e-ZCMRs). In the last decades, zeolite membranes have attracted great attention, with special emphasis on the synthesis of MFI membranes. These membranes have found their applications in e-ZCMR for selective extraction of targeted product from the reaction zone. Previous researches have compared the efficiency of e-ZCMR with that of conventional fixed-bed reactors (FBRs) in the production of p-Xylene (PX) from meta-Xylene (MX) via isomerization reaction over suitable catalysts [1–5]. Their evaluations also have been based on permeate-only mode operation [1] and combined mode operations [1–5]. The authors of these studies evaluated their reactor performance in term of p-Xylene selectivity, p-Xylene yield and conversion of the reactant (m-Xylene, p-Xylene or o-Xylene (OX)). The authors reported and concluded that advantages of e-ZCMR over conventional fixed-bed reactor (FBR) for PX production include enhancement of PX yield and MX conversion due to selective extraction of PX from the reaction zone as soon as it is produced. This in turn forward-shifts the equilibrium position of the reaction, allowing more MX to react to produce more p-Xylene.

Investigation of the potential application of various kinds of configurations of MFI-type zeolite membranes prepared using various hydrothermal synthesis techniques has been carried out [6–10]. The first preliminary investigation was reported by van Dyk et al. [1]. At 577 K, the authors reported about 10% increase in p-Xylene yield – compared to a conventional FBR – in combined mode operation with 65% para-selectivity. In their work, m-Xylene isomerization was conducted in an e-ZCMR equipped with inert MFI-type zeolite membrane supported on alumina tube and prepared using a pore-plugging synthesis technique [11–13]

---

[*] Corresponding author.
E-mail address: daramola@sun.ac.za (M.O. Daramola).
[1] Deceased.
[2] Current address: Snowden Group, West Perth, Australia.

0926-860X/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.apcata.2010.07.040

as separation unit. Regarding the application of e-ZCMR equipped with a "film-like" tubular MFI-type zeolite membrane supported on porous stainless steel as separation unit, Tarditi et al. [2] reported a p-Xylene production increase of 26% above that of a FBR during m-Xylene isomerization over a commercial Pt/silica–alumina catalyst at 643 K.

Furthermore, Zhang et al. [3] reported m-Xylene isomerization over HZSM-5 catalyst in an e-ZCMR. The authors used a disk-shaped silicalite-1 membrane that displayed p-Xylene/o-Xylene (p/o) separation factor of 16 as the separation unit. The authors reported 26% increase in PX yield over a conventional FBR. On the modelling studies of this promising system, Deshayes et al. [5] reported on the simulation and modelling study of xylene isomerization reaction in an industrial FBR and the effect of incorporation of multi-tubes containing Na-ZSM-5/SS membranes upon the catalytic performance of the reactor. These authors predicted an increase of about 12% in p-Xylene production over a conventional FBR.

In the case of the application of catalytically active MFI-type zeolite membranes for xylene isomerization, the only example in the open literature was reported by Haag et al. [4]. In this study, the membrane consisted of a HZSM-5 film grown on top of a porous stainless steel disk. These authors reported an increase of 15% of m-Xylene conversion at 673 K compared to a FBR, while p-Xylene selectivity was enhanced by 10%.

All the above-mentioned studies involved the use of supported film-like MFI membranes; except the study reported by van Dyk et al. [1]. One of the shortcomings of "film-like" MFI-type zeolite membranes is a mismatch between the thermal expansion coefficients of the support and the zeolite material at higher operation temperatures. This allows permeation of undesirable isomers through the intercrystalline defects, contributing to a reduction of the membrane selectivity, thereby resulting in reduction PX purity in the permeate stream. The permeation of the impurities or undesirable products becomes more pronounced if the membrane support is a stainless steel because of the higher disparity between the thermal expansion coefficient of stainless steel and that of MFI zeolite. This limitation ascribed to "film-like" MFI-type membranes could retard or even hinder the industrialization of this energy-efficient technology, especially, for industrial production of p-Xylene production via isomerization reaction (involving catalytic conversion and separation) that usually occurs at temperatures between 623 K and 673 K [2]. Drastic reduction of p-Xylene purity at pronounced permeation of impurities such as m-Xylene and o-Xylene at these temperatures during the application of "film-like" MFI-type zeolite membranes in e-ZCMR might have discouraged researchers in this area to report the p-Xylene purity of their membranes during xylene isomerization in e-ZCMR.

As pointed out in a series of previous studies by Dalmon and his group, shortcoming like this can be overcome by using nanocomposite MFI-ceramic membranes [11–13]. Contrary to their "film-like" counterparts, in nanocomposite architectures, zeolite crystals are embedded within the pores of the supports rather than formation of zeolite film layers on the surfaces of the supports. Advantages of nanocomposite architectures compared to zeolite films include defect control and higher mechanical and thermal stability. Defect control with nanocomposite architecture is possible because the zeolite crystals are embedded within the support pores, thereby limiting the maximum defect size to the pore sizes of support.

Regarding the application of tubular nanocomposite MFI-alumina membranes for m-Xylene isomerization in e-ZCMR, recently, we reported about 18% increase in p-Xylene yield with PX selectivity in the permeate approaching about 95% at 573 K, over a FBR operated at similar operating conditions [16]. In this study, a tubular nanocomposite MFI-alumina membrane displaying p-Xylene/o-Xylene (p/o) and p-Xylene/m-Xylene (p/m) separation factors >50 and with p-Xylene permeance of about 9.5 nmol m^{-2} s^{-1} Pa^{-1} was used in an e-ZCMR to investigate the influence of operating variables and reactor configuration on the performance of the reactor. In furtherance of this study and with a focus on p-Xylene purity in the permeate side of e-ZCMR, this paper demonstrates the possibility of obtaining ultra-pure p-Xylene in an e-ZCMR with a tubular nanocomposite MFI-alumina membrane displaying p-Xylene/o-Xylene (p/o) >400 as the separation unit. It is noteworthy to mention that for industrial application of p-Xylene, ultra-pure PX purity is essential in most cases and obtaining this through conventional process costs a lot in term of energy and operation.

Also for better understanding of the behaviour of e-ZCMR at lower temperatures, the investigation presented in this study was conducted at temperatures of 473–573 K.

2. Experimental

2.1. Membrane preparation, characterization and separation test

The membrane used in this study was prepared by pore-plugging synthesis technique [11–13]. The membrane was prepared on asymmetric α-alumina support (id 7 mm; od 10 mm; length 150 mm). The support consisted of 3 layers with pore sizes 0.2 μm, 0.8 μm and 12 μm, respectively, from inner to outermost layer.

The MFI zeolite was synthesized by mixing together the structure directing agent (SDA, 1 M tetrapropylammonium hydroxide, TPAOH, supplied by Sigma–Aldrich), and the silica source (Aerosol 380 from Degussa). This mixture was slightly diluted with deionised water to form a clear solution with the molar composition 1.0 SiO_2: 0.45 TPAOH: 27.8 H_2O (pH close to 14) and left to mature for 72 h room temperature under mild stirring to prevent concentration gradient within the solution. The α-alumina support was inserted into a TeflonR-lined autoclave containing about 25 mL of precursor solution, and submitted to an interrupted hydrothermal synthesis at 423 K for 4 days. After the synthesis, the tube was washed repeatedly with deionised water until the pH of the water was 7, dried at 373 K for 12 h, and calcined at 773 K for 4 h under air flow. The formation of a nanocomposite material (namely no formation of continuous MFI film) on the support was confirmed with scanning electron microscope (SEM).

Further characterization of the as-prepared membrane with effective permeation area of 26 cm^2 was done by room-temperature H_2 gas permeation, room-temperature n-butane/H_2 binary mixture separation and xylene ternary vapour mixture separation tests. Xylene ternary vapour mixture separation tests were performed by feeding the inner tube of the membrane with ternary vapour mixture of xylene (p-, m-, and o-Xylene) saturated in N_2 at feed flow rate of 10 mL(STP)/min. Nitrogen gas at 15 mL(STP)/min flow rate was swept over the shell side of the membrane. To ensure proper xylene vapour partial pressures throughout the experimental period, all the lines were heated up to 393 K to prevent xylene condensation.

After attaining a steady-state in about 4 h, composition of the feed, retentate and permeate streams was obtained with a pre-calibrated gas chromatograph equipped with solgel wax capillary column and FID. To evaluate the separation performance of the membrane in term of separation factors, the separation factor was defined as depicted in Eq. (1):

$$Sf_{i/j} = \frac{(y_i/y_j)_{\text{permeate}}}{(x_i/x_j)_{\text{feed}}} \tag{1}$$

where x_i, x_j and y_i, y_j are, respectively, the molar fractions of species i and j in the retentate and permeate streams. In our case, i repre-

Download English Version:

https://daneshyari.com/en/article/41913

Download Persian Version:

https://daneshyari.com/article/41913

Daneshyari.com