



ELSEVIER

Available online at www.sciencedirect.com

ScienceDirect

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

Structure-controlled mesoporous SBA-15-derived mixed matrix membranes for H₂ purification and CO₂ capture

Hui-Hsin Tseng^{a,b}, Hao-Wei Chuang^c, Guo-Liang Zhuang^c,
Wen-Hsiung Lai^c, Ming-Yen Wey^{c,*}

^a School of Occupational Safety and Health, Chung Shan Medical University, Taichung 402, Taiwan, ROC

^b Department of Occupational Medicine, Chung Shan Medical University Hospital, Taichung 402, Taiwan, ROC

^c Department of Environmental Engineering, National Chung Hsing University, Taichung 402, Taiwan, ROC

ARTICLE INFO

Article history:

Received 4 December 2016

Received in revised form

25 February 2017

Accepted 4 March 2017

Available online xxx

Keywords:

Mesoporous filler

Shape

Size

Loading weight

Mixed matrix membrane

ABSTRACT

The composite mixed matrix membranes (MMMs) were prepared by incorporating mesoporous SBA-15 as a filler to discuss the effects of its particle shape, particle size, and loadings on the organic–inorganic interfacial morphology. The SBA-15 was synthesized by template method and its particle shape and size was adjusted by adding electrolyte. The results indicated that the spherical SBA-15 can improve the dispersion and have better adhesion with organic phase, which showed better permselectivity than the rod-like one. The SBA-15 filler also could increase the diffusion selectivity of MMMs by the addition of different particle sizes. The permeabilities of H₂ and CO₂ were 1207.9 and 552.86 Barrer, respectively, with selectivities of H₂/CH₄ and CO₂/CH₄ reached 247.0 and 112.8, respectively, when 1.6 μm spherical SBA-15 was added at 3 wt. %. The dissimilarity occurring in the permselectivity values with changes made in the particle shape and size are much more pronounced at the lower SBA-15 loading, which exceeded the 2008 Robeson's upper bound limited.

© 2017 Hydrogen Energy Publications LLC. Published by Elsevier Ltd. All rights reserved.

Introduction

Nowadays, approximately 95% of hydrogen produced comes from biomass, like carbonaceous raw materials, primarily fossil in origin and so on [1–3]. In these processes, the hydrogen can be produced via steam reforming and water gas shift reaction, and the carbon is converted to CO₂ and released to the atmosphere [4–8]. Therefore, the H₂-selective and CO₂-selective separation process are recognized as the most important processes in the clean energy power

generation system. The clean energy H₂ and the greenhouse-gas CO₂ can be simultaneously concentrated and collected, which will be a benefit to high energy efficiency and greenhouse-effect control. The separation membrane technologies have generally been acknowledged as the most efficient way because of their low equipment costs, lower energy requirements, and easy operating techniques [9–14].

Currently, the research about membrane development is based on achieving a high permeability and selectivity. There are many polymer membranes that have been developed and

* Corresponding author. Fax: +886 4 22862587.

E-mail address: mywey@dragon.nchu.edu.tw (M.-Y. Wey).

<http://dx.doi.org/10.1016/j.ijhydene.2017.03.026>

0360-3199/© 2017 Hydrogen Energy Publications LLC. Published by Elsevier Ltd. All rights reserved.

commercialized due to their economical processing capabilities [15,16]. However, the trade-off relationship between gas permeability and selectivity is one of the biggest problems limiting their application in the gas separation field, as suggested by Robeson [14,17]. Although the inorganic membranes have good thermal stability and permselectivity, they are difficult to fabricate and commercialize. Therefore, to overcome this limitation, mixed matrix membranes (MMMs) by adding inorganic materials into polymer membrane have started to emerge as an alternative approach in membrane technology [18–21].

The performance of the MMMs is based on the combination characteristics of the inorganic and organic materials, such as (1) the aggregation of fillers in matrix [22,23] and (2) The adhesion between filler and polymer matrix [24–27]. The gas permeability was limited at low filler loadings; however, the fillers aggregation early occurred when the filler loadings were increased [22]. The fillers aggregation causes defects between the two phases or inter-filler void, which have a strong influence on gas performance [11,12]. Mahajan and Koros [24] have attributed weak polymer-particle adhesion to non-uniform stress directions around the particles causing non-selective voids and reducing gas resistance instead of the gas passing through the pores in the particle. Shahid et al. [23] also indicated that filler at higher loadings (<30 wt%) could occur the less adhesion between filler and matrix, resulting in the low gas selectivity.

Some researchers have improved the adhesion of polymer and fillers by chemical methods such as using coupling agents [28], plasticizers [29] or copolymers [30]. However, some types of coupling agents may penetrate into the porous fillers resulting in reduced gas permeability [28]. Additionally, some plasticizers will change the inherent characteristics of the polymer, such as PI polymer. The enhancement of the polymer mobility decreased the inherent PI selectivity, but it created good adhesion between the inorganic-organic phases [29]. Furthermore, to find the appropriate copolymer for a specific polymer is not economical and efficient.

The dispersion of fillers in the polymer matrix not only depends on the interaction between the organic and inorganic phases but also on the filler's morphology, such as particle size and shape and pore properties and so on all have important effects on the gas separation performance [10]. Vu et al. [31] indicated that smaller particles can prevent precipitation behavior during the membrane casting, and thus, a defect-free membrane can be obtained. Moreover, they also indicated that the spherical shape would minimize the agglomeration at the polymer matrix because it limits the contact between particles. Reid et al. proposed that the spherical particle shape of fillers such as zeolite MCM-41 and MCM-48 can reduce the agglomeration of each particle because it has good compatibility with the polymer matrix [32], and thus, it can also result in good dispersion [33,34]. Another challenge is that pore blockage is related to porous fillers. Li et al. [35] demonstrated that the solvent or polymer entered into the particle pores when the porous fillers were added into the polymer matrix. Although the gas permeability decreases, the reduced pore size of zeolite 5A allowed the gas selectivity to increase. Therefore, adjusting the physical properties of fillers directly seems to be a simpler method.

Incorporation of inorganic mesoporous SBA-15 silica as a filler into MMMs has been evaluated in our previous studies, and it showed higher gas permselectivity than other microporous silica [22,36]. The SBA-15 silica has a large pore volume, high surface area and better thermal stability than the MCM-41 [37,38]. The pore structure of SBA-15 silica is neat hexagonal columnar, and the pore size can be adjusted according to the synthetic conditions. However, there is less information about the influence of the morphology and particle size of SBA-15 silica filler on gas separation performance. SBA-15 silica is generally synthesized by hydrolysis-condensation hydrothermal reaction. SBA-15 crystals are formed and grown in a solution containing a template, such as block copolymer Pluronic P123 ($\text{EO}_{20}\text{PO}_{70}\text{EO}_{20}$) surfactant [22,36,39]. In this solution, the morphology of SBA-15 is controlled by condensation rate. The morphology of SBA-15 can further be modified by adding co-surfactant (e.g., CTAB) [40,41], cosolvent (e.g., DMF) [42], a swelling agent (e.g., TMB) [43], or inorganic salt [42–44]. Newalkar and Lomarneni [44] investigated the effect of NaCl on the textural properties of SBA-15 in detail. Results indicated that the salt can affect the micellar properties of block copolymer P123, such as reduced micelle polarity and size, thereby forming small pore-sized SBA-15. Wan et al. [29] reported the synthesis of SBA-15 spheres by using tetraethyl orthosilicate (TEOS) silica source and adding KCl electrolyte to an aqueous solution. The particle size of SBA-15 was found to decrease from 4.5 μm to 2.0 μm with increased KCl concentration.

In the present study, we used electrolyte and adjusted its amount to accelerate the effects of reaction rate, as well as to control the shape and particle size of SBA-15 silica filler. The crystallized samples were characterized by X-ray diffraction (XRD), nitrogen adsorption, and field-emission scanning electron microscopy (FESEM) analyses. With the aid of salt-modification method for SBA-15 filler synthesis, we demonstrated the effects of textural parameters of modified SBA-15 fillers on the morphology and gas separation performance of membrane.

Experimental

Materials

For the synthesis of SBA-15 mesoporous materials, TEOS (tetraethyl orthosilicate, 98%, Aldrich) and Pluronic P123 ($\text{EO}_{20}\text{PO}_{70}\text{EO}_{20}$, Aldrich) were used as the Si source and template, respectively. An aqueous solution of HCl (37%, Sigma Aldrich) was also used to control the pH of the reaction system, and the electrolyte KCl (Sigma Aldrich) was used to control the ionic strength of the solution.

A porous α -alumina support disk with an average pore size of 74.2 nm, diameter of 1.6 cm and porosity of 40–48% was purchased from the Ganya Fine Ceramics Co., LTD (Taiwan). To modify the surface roughness of Al_2O_3 support and increase the adhesion between polymer selective layer and support layer, the support was coated with a carbon thin film. The detail procedure has been mentioned in our previously study [45]. Gases for permeation tests were supplied from

Download English Version:

<https://daneshyari.com/en/article/5145741>

Download Persian Version:

<https://daneshyari.com/article/5145741>

[Daneshyari.com](https://daneshyari.com)