UNIVERSITI SAINS MALAYSIA

Laporan Akhir Projek Penyelidikan Jangka Pendek

Characterization and Development of Indigenous Membrane for Industrial Application

by

Prof. Abdul Latif Ahmad
Prof. Subhash Bhatia
Dr. Syamsul Rizal Abd Shukor
Dr. Tan Soon Huat
Assoc. Prof. Dr. W. J. N. Fernando

2011

UNIVERSITI SAINS MALAYSIA

UNIVERSITY RESEARCH GRANT FINAL REPORT

Geran Penyelidikan Universiti Laporan Akhir

A.	**PARTICULARS OF RESEARCH** / *MAKLUMAT PENYELIDIKAN:*
(i)	**Title of Research:** Characterization and Development of Indigenous Membrane for Industrial Application *Tajuk Penyelidikan:*
(ii)	**Account Number:** 1001/PJKIMIA/814002 *Nombor Akaun:*
B.	**PERSONAL PARTICULARS OF RESEARCHER** / *MAKLUMAT PENYELIDIK:*
(i)	**Name of Research Leader:** Profesor Abdul Latif Ahmad *Nama Ketua Penyelidik:*
	Name of Co-Researcher *Nama Penyelidik Bersama:* 1. Profesor Subhash Bhatia 2. Dr. Syamsul Rizal Abd Shukor 3. Dr. Tan Soon Huat 4. Profesor Madya Dr. W. J. N. Fernando
(ii)	**School/Institute/Centre/Unit** : School of Chemical Engineering *Pusat Pengajian /Institut/Pusat/Unit :*
C.	**Research Platform (Please tick (/) the appropriate box):** *Pelantar Penyelidikan (Sila tanda (/) kotak berkenaan):* ☐ A. **Life Sciences** *Sains Hayat* ☐ B. **Fundamental** *Fundamental* ☑ C. **Engineering & Technology** *Kejuruteraan & Teknologi* ☐ D. **Social Transformation** *Transformasi Sosial* ☐ E. **Information & Communications Technology (ICT)** *Teknologi Maklumat & Komunikasi* ☐ F. **Clinical Sciences** *Sains Klinikal* ☐ G. **Biomedical & Health Sciences** *Bioperubatan Sains Kesihatan*

D. **Duration of this research :**
Tempoh masa penyelidikan ini :

***Duration :** .....42 months...............
Tempoh :

From : ....October......... 2007 **To** : ....April...........2011
Dari: *Ke :*

E. **ABSTRACT OF RESEARCH**

(An abstract of between 100 and 200 words must be prepared in **Bahasa Malaysia and in English**.
This abstract will be included in the Annual Report of the Research and Innovation Section at a later date as a means of presenting the project findings of the researcher/s to the University and the community at large)

Abstrak Penyelidikan
(Perlu disediakan di antara 100 - 200 perkataan di dalam **Bahasa Malaysia dan juga Bahasa Inggeris**.
Abstrak ini akan dimuatkan dalam Laporan Tahunan Bahagian Penyelidikan & Inovasi sebagai satu cara untuk menyampaikan dapatan projek tuan/puan kepada pihak Universiti & masyarakat luar).

Membrane processes are widely being used in various industries applications nowadays. This project consists of five research. The research for synthesis and development of nitrocellulose membrane for medical biotechnology application has succesfully fabricated the lateral flow nitrocellulose membrane via dry phase inversion. For the research of development of nanoporous gamma-alumina functional membrane using vanadium oxide nanotubes as template and its application for styrene oxidation reactions, the impregnation of vanadium oxide on membrane has been done. The studies for composite membrane/adsorption separation for sequestration of CO_2 has sucessfully set-up a membrane/absorber unit. The research for development of catalytic zeolite membrane for simultaneous conversion and separation of para-xylene from xylene isomers, the specific zeolite membrane has been synthesised and characterized. Lastly, the research for synthesis of multiwalled carbon nanotube/poly(vinyl alcohol) membranes and their application to pervaporation dehydration of isopropanol and water mixture is also successfully being carried out.

Membran proses telah diguna dalam pelbagai industri. Projek ini mengandungi lima penyelidikan. Penyelidikan untuk sintesis dan pembangunan membran nitroselulosa untuk aplikasi bioteknologi perubatan telah berjaya menghasilkan nitroselulosa membran aliran lateral melalui inversi fasa kering. Untuk penyelidikan pembangunan membran gamma-alumina nanoporous berfungsi menggunakan nanotube oksida vanadium sebagai templat dan aplikasinya untuk reaksi oksidasi stirena, impregnasi vanadium oksida pada membran telah dilakukan. Kajian untuk membran komposit / pemisahan jerapan untuk penyerapan daripada CO_2 telah berjaya mendirikan sebuah unit membran / penyerap. Kajian untuk pembangunan membran zeolite mangkin untuk penukaran serentak dan pemisahan para-xilena dari isomer xylena, membran zeolite khusus telah disintesis dan disifatkan. Akhirnya, kajian untuk sintesis carbon nanotube multiwalled / poli (vinil alkohol) membran dan aplikasinya untuk dehidrasi pervaporasi isopropanol dan campuran air juga berjaya dilaksanakan.

F.

SUMMARY OF RESEARCH FINDINGS
Ringkasan dapatan Projek Penyelidikan

The five research projects are carried out in line with the project objective. The study on nitrocellulose membrane synthesis, development and application has been performed by the reserarch team. Nanoporous gamma-alumina membrane were also developed using vanadium oxide nanotubes. The membrane/absorber unit was set up with membrane-synthesized hydrotalcite doped with alumina using sol gel technique and adsorbent of hydrotalcite. Zeolite membrane was also synthesized and characterized for conversion and recovery of para-xylene from its isomers. Multiwalled carbon nanotube/poly(vinyl alcohol) membranes were synthesized for pervaporation dehydration application.

G.

a) Results/Benefits of this research
Hasil Penyelidikan

No. *Bil:*	**Category/Number:** *Kategori/ Bilangan:*	**Promised**	**Achieved**
1.	**Research Publications (Specify target journals)** *Penerbitan Penyelidikan (Nyatakan sasaran jurnal)*		42
2.	**Human Capital Development**		
	a. Ph. D Students		5
	b. Masters Students		0
	c. Undergraduates (Final Year Project)		1
	d. Research Officers		0
	e. Research Assisstants		0
	f. Other: Please specify **i)Graduated phD Students** **ii) Graduated MSc Students** **iii) Graduated Final Year Students**		 3 4 3
3.	**Patents** *Paten*		1
4.	**Specific / Potential Applications** *Spesifik/Potensi aplikasin*		3
5.	**Networking & Linkages** *Jaringan & Jalinan*		10
6.	**Possible External Research Grants to be Acquired** *Jangkaan Geran Penyelidikan Luar Diperoleh*		1
7.	**Awards** *Anugerah*		10
8.	**Proceedings** *Prosiding*		32

- Kindly provide copies/evidence for Category 1 to 6.

Category 1 (Research publication)

1. A.L. Ahmad; S. C. Low; S.R. Abd. Shukor; A. Ismail, Synthesis and Characterization of Polymeric Nitrocellulose Membrane: Influence of Additives and Pore Former on Membrane Morphology, *Journal of Applied Polymer Science* 108 (2008) 2550–2557.
2. Fong, Y.Y.; Abdullah, A.Z.; Ahmad, A.L.; Bhatia, S., Development of functionalized zeolite membrane and its potential role as reactor combined separator for para-xylene production from xylene isomers, *Chemical Engineering Journal*, 139 (2008)172-193.
3. Y.F. Yeong, A.Z. Abdullah, Ahmad, A.L, S. Bhatia, Synthesis, characterization of phenethyltrimethoxysilane (PE) modified organic-inorganic hybrid silicalite-1 molecular sieves and its transformation into solid acid materials, *Advanced Materials Research* 47-50 PART 1 (2009) 238-241.
4. A.L. Ahmad, C. P. Leo and S. R. Abd Shukor, Tailoring of a γ-alumina membrane with a bimodal pore size distribution for improved permeability, *Journal of the American Ceramic Society*, 91 (1) (2008) 246-251.
5. A.L. Ahmad, C. P. Leo and S. R. Abd Shukor, Preparation of γ-alumina thin layer with bimodal pore size distribution for diminution of transport resistance in bi-layered membrane, *Thin Solid Films*. 516 (12) (2008) 4319-4324.
6. A.L. Ahmad, C. P. Leo and S. R. Abd Shukor, Vanadium oxide supported γ-alumina with bimodal porous structure for intra-particle diffusion enhancement in styrene oxidation reaction, *Journal Porous Material*, 16 (2009) 1:33-40.
7. A. L. Ahmad, S. C. Low, S. R. Abd Shukor, and A. Ismail. Effects of Membrane Cast Thickness on Controlling the Macrovoid Structure in Lateral Flow Nitrocellulose Membrane and Determination of Its Characteristics, *Scripta Materialia*, 57(2007) 743-746.
8. A. L. Ahmad, S. C. Low, S. R. Abd Shukor, and A. Ismail. Morphological and Thermal Mechanical Stretching Properties on Polymeric Lateral Flow Membrane, *Industrial & engineering Chemistry Research*, 48(2009) 7: 3417-3424.
9. A. L. Ahmad, P. C. Oh, and S. R. Abd Shukor. Sustainable Biocatalytic Synthesis of L-homophenylalanine As Pharmaceutical Drug Precursor, *Biotechnology Advances*, 27(2009) 3:286-296.
10. A. L. Ahmad, P. C. Oh, and S. R. Abd Shukor. Biocatalytic Synthesis of Angiotensin-Converting Enzyme (ACE) Inhibitor Drug Precursor Using Membrane Bioreactor: A Review, *Journal of Applied Membrane Science & Technology (JAMST)*, 9(2009) 19-26.
11. A. L. Ahmad, S. A. Rahman, K. T. Lee, and W. J. N. Fernando. Effect of DEA In Membrane Gas Absorption System Using Polymeric Flat Sheet membrane. *Journal of Applied Membrane Science & Technology (JAMST),* 8(2008) 9-14.
12. A. L. Ahmad, and H. N. A. Halim. Protein-Membrane Interactions in Forced-Flow Electrophoresis of Protein Solutions: Effect of Initial pH and Initial Ionic Strength, *Separation and Purification Technology*, 66(2009) 273-278.
13. A. L. Ahmad, S. A. Rahman, K. T. Lee, and W. J. N. Fernando. Development of Thin Film Composite for CO_2 Separation In Membrane Gas Absorption Application, *Asia Pasific Journal of Chemical Engineering,* (2009) 4: 787–792.
14. Y.F. Yeong, A.Z. Abdullah, A.L. Ahmad, S. Bhatia. Process Optimization Studies of P-Xylene Separation from Binary Xylene Mixture over Silicalite-1 Membrane using Response Surface Methodology, *Journal of Membrane Science*, Vol. 341, Issues 1-2, pp. 96-108, 2009.
15. Y.F. Yeong, A.Z. Abdullah, A.L. Ahmad, S. Bhatia. Separation of *p*-xylene from Ternary Xylene Mixture over Silicalite-1 Membrane: Process Optimization Studies, *Journal Chemical Technology and Biotechnology*, Vol. 85, Issues 2, pp. 216-225, 2010.
16. Y.F. Yeong, A.Z. Abdullah, A.L. Ahmad, S. Bhatia. Synthesis, Characterization and Reactive Separation Activity of Acid-functionalized Silicalite-1 Catalytic Membrane in *m*-xylene Isomerization, *Journal of Membrane Science,* 360 (2010) 109–122.
17. T.L. Chew, A.L. Ahmad, S. Bhatia. Rapid Synthesis of Thin SAPO-34 Membranes Using Microwave Heating, *Journal of Porous Materials*, (In press).

18. Y.T. Ong, A.L. Ahmad, S.H.S. Zein, S.H. Tan. A Review on Carbon Nanotubes in Environmental Protection and Green Engineering Perspective, *Brazilian Journal of Chemical* Engineering, Vol. 27, Issue 2, 2010.
19. A. L. Ahmad, S. C. Low, S. R. Abd. Shukor, A. Ismail. Optimization of Membrane Performance by Thermal-Mechanical Stretching Process Using Responses Surface Methodology (RSM), *Separation and Purification Technology*, Vol. 66, 177-186, 2009.
20. A. L. Ahmad, S. C. Low, S. R. Abd. Shukor, A. Ismail. Development of Lateral Flow Membranes for Immunoassay Separation, *Desalination and Water Treatment*, Vol. 5, 99-105, 2009.
21. A. L. Ahmad, S. C. Low and S. R. Abd. Shukor. Optimization of Membrane Formulation and Process Variables via Crossed-Design Concept in design of Experimental (DOE), *Separation Science and Technology*, 44(12), 2870 – 2893, 2009.
22. K.K.Lau, M.Z. Abu Bakar, A.L. Ahmad, T.Murugesan. Feed spacer mesh angle: 3D modeling, simulation and optimization based on unsteady hydrodynamic in spiral wound membrane channel. *Journal of Membrane Science*, 343(2009) 1-2 : 16-33.
23. A.L. Ahmad, M.F. Chong, S. Bhatia. A comparative study on the membrane based palm oil mill effluent (POME) treatment plant. *Journal of Hazardous Materials*, 171(2009) 1-3: 166-174.
24. Y.F. Yeong, A.Z. Abdullah, A.L. Ahmad, S. Bhatia. Modeling and simulation of an acid-functionalized silicalite-1 membrane reactor for xylene isomerization, *Separation and Purification Technology*, 75 (2010) 183–192.
25. T.L. Chew, A.L. Ahmad, S. Bhatia. Ordered mesoporous silica (OMS) as an adsorbent and membrane for separation of carbon dioxide (CO_2), *Advances in Colloid and Interface Science*, 153 (2010) 43–57.
26. Y.F. Yeong, A.Z. Abdullah, A.L. Ahmad, S. Bhatia. Propylsulfonic acid-functionalized partially crystalline silicalite-1 materials: synthesis and characterization, *Journal Porous Material*, Vol. 18, Number 2, 147-157.
27. Y.F. Yeong, A.Z. Abdullah, A.L. Ahmad, S. Bhatia. Separation of p-xylene from binary xylene mixture over silicalite-1 membrane: Experimental and modeling studies, *Chemical Engineering Science*, 66 (2011) 897–906.
28. Y.F. Yeong, A.Z. Abdullah, A.L. Ahmad, S. Bhatia. Xylene isomerization kinetic over acid-functionalized silicalite-1 catalytic membranes: Experimental and modeling studies, *Chemical Engineering Journal*, 157 (2010) 579–589.
29. Ahmad A. L., Low S. C., Shukor S. R. A., Fernando, W.J.N., Ismail, A. Hindered diffusion in lateral flow nitrocellulose membrane: Experimental and modeling studies, *Journal of Membrane Science*, 357 (1-2) (2010) pp. 178 – 184.
30. Ahmad A. L., Low S. C., Abd Shukor S. R. and Ismail, A. Preparation and Characterization of Various Membrane Morphologies for Lateral Flow Immunoassay Development, *Journal of Applied Membrane Science and Technology*, 5(2007) 23-29.
31. Ahmad A. L., Low S. C., Abd Shukor S. R., Ismail, A. and A.R. Sunarti, Development of Lateral Flow Membrane for Immunoassay Separation, *Desalination and water treatment*, 5(2009) 99-105.
32. Sani, N.A.A., Ahmad A. L., Zein, S.H.S. Preparation and properties of perovskite-dispersed titania membranes, *Journal of Applied Sciences*, 10 (12) (2010) pp. 1097 – 1103.
33. A.L. Ahmad, A.R. Sunarti, K.T. Lee, W.J.N. Fernando, CO_2 removal using membrane gas absorption, *International Journal of Greenhouse Gas Control*, 4 (3) (2010) pp. 495 – 498.
34. A.L.Ahmad, P.C.Oh, S.R. Abd Shukor Synthesis of L-homophenylalanine via integrated membrane bioreactor : Influence of pH on yield, *Biochemical Engineering Journal*, Vol. 52 (2-3) (2010) pp. 296 – 300.
35. A.L.Ahmad, B.Koohestani, S.Bahtia, B.S.Ooi Synthesis of vanadium oxide nanotubes via an ultrasonic method, *Journal of Sol-Gel Science and Technology*, Vol. 56 (2010) pp. 327 – 332.
36. A.L. Ahmad, M.A.T. Jaya, C.J.C. Derek, 2010, The effects of organic binders on palladium impregnated in TiO2 membrane synthesis: X-Ray diffraction analysis, *Journal of the American Ceramic Society*, Vol 93 (1), (2010) pp. 3595 - 3599.

37. A. L. Ahmad, P. C. Oh and S. R. Abd Shukor, L-homophenylalanine Production by Reductive Amination of 2-oxo-4-phenylbutanoic Acid via Integrated Membrane Bioreactor, *Journal of Materials Science and Engineering* [Accepted]
38. Ahmad, A.L., Jaya, M.A.T., Derek, C.J.C., Ahmad, M.A., Synthesis and characterization of TiO2 membrane with palladium impregnation for hydrogen separation, *Journal of Membrane Science*, Vol. 366 (1-2), (2011), pp. 166 – 175.
39. Ong, Y.T., Ahmad, A.L., Zein, S.H.S., Sudesh, K., Tan, S.H., Poly(3-hydroxybutyrate)-functionalised multi-walled carbon nanotubes/chitosan green nanocomposite membranes and their application in pervaporation, *Separation and Purification Technology*, Vol 76 (3), (2011), pp. 419-427.
40. A.L. Ahmad, N.A. Abdullah Sani, S.H.S. Zein, Influence of Calcination Temperature and Perovskite Concentration on the Morphology, Gas Permeance and Selectivity of Perovskite-Titania Membranes, *Journal of Ceramic Science and Technology*, (2010) [In press].
41. A.L. Ahmad, N.A.A. Sani, S.H.S. Zein, Synthesis of a TiO_2 ceramic membrane containing $SrCo_{0.8}Fe_{0.2}O_3$ by the sol-gel method with a wet impregnation process for O2 and N2 permeation, *Ceramics International* [In press].
42. A.L. Ahmad , P.C. Oh and S.R. Abd Shukor, Synthesis of 2-oxo-4-phenylbutanoic acid: Parameter Optimization using Response Surface Methodology, *Chemical Engineering Journal*, [In Press].

Category 2 (Human Capital Development)

Graduated Ph.D. students

1. Ms Leo Choe Peng

   Thesis title: *Bimodal porous ceramic membrane via nanosized polystyrene templating: synthesis, characterization and performance evaluation*

2. Ms. Low Siew Chun

   Thesis title: *Lateral Flow Nitrocellulose Membrane for Diagnostic Kit Application: Synthesis, Characterisation and Performance Evaluation*

3. Ms. Yeong Yin Fong

   Thesis title: *Functionalized and Nonfunctionalized Zeolite Membranes: Synthesis, Characterisation and its Role in Separation and Catalysis*

Graduated M.Sc. students

1. Mr. Muhammad Azan b. Tamar Jaya

   Thesis title: *Synthesis and Characterization of Palladium-zplionia Ceramic Membrane For Hydrogen Separation*

2. Ms. Nur Aimie Abdullah Sami

   Thesis title: *Development of Perovskite Zirconia Ceramic Membrane Via Sol-Gel Method: Synthesis and Characterization*

3. Mr. Ong Yit Thai

   Thesis title: *Study of the Dispersion and Alignment of Carbon Nanotubes in Polymeric Pervaporation Membranes*

4. Ms. Nadia bt. Isa

Thesis title: *The use of Adsorption / Desorption and Membrane Separation Methods Based on Hydrotalcite for Removal of Carbon Dioxide*

Current Ph.D. Student (Research is on-going)

1. Ms. Oh Pei Ching
2. Mr. Chew Thiam Leng
3. Mr. Behnam Koohestanti
4. Ms. Norhidayah Ideris
5. Mdm. Sunarti Abd. Rahman

Category 3 (Patents)

1. Malaysian Patent Pending No. PI 20092840

   Date of Filing: 6 July 2009

   Applicant: Universiti Sains Malaysia

   Entitled: A System for Producing L-Homophenylalanine and a Process for Producing L-Homophenylalanine

   Inventors: Abdul Latif Ahmad; Oh Pei Ching; Syamsul Rizal Abd Shukor

2. PCT International Application No. PCT/MY2010/000001

   Date of Filing: 5 January 2010

   Applicant: Universiti Sains Malaysia

   Entitled: A System for Producing L-Homophenylalanine and a Process for Producing L-Homophenylalanine

   Inventors: Abdul Latif Ahmad; Oh Pei Ching; Syamsul Rizal Abd Shukor

Category 4 (Specific / Potential Applications)

1. Malaysian Technology Development Corporation (MTDC)

   Biomedical approach to produce nitrocellulose membrane for diagnostic kits

2. Yayasan Felda

   Treatment of palm oil mill effluent (POME) using innovative membrane based technology

3. Patent

   Synthesis of L-Homophenylalanine as a drug precursor for treatment of Cardiovascular disease

Category 5 (Networking & Linkages)

1. Universiti Kebangsaan Malaysia (UKM)
2. Universiti Teknologi Malaysia (UTM)
3. Universiti Putra Malaysia (UPM)
4. Universiti Malaysia Sabah (UMS)
5. University of Wales, Swansea, United Kingdom
6. The Hong Kong University of Science and Technology
7. Institut Teknologi Bandung (ITB)
8. Monash University
9. The University of Notthingham
10. Universiti Tunku Abdul Rahman (UTAR)

Category 6 (Possible External Research Grants to be Acquired)

1. Grant Penyelidikan daripada Kerajaan Oman

Category 7 (Awards)

1. Award Name: **Silver Medal**

   18th International Invention, Innovation & Technology Exhibition (ITEX 2007), 18th – 20th May 2007, KLCC, Malaysia.

   Title of invention: StretchTech: Novel Uni-Axial Membrane Stretcher for Membrane Morphology Modification

   Awarded by: Malaysian Invention and Design Society (MINDS)

2. Award Name: **Gold Medal**

   Malaysian Technology Expo-Global Invention & Innovation (MTE 2008), 20th – 22nd February 2008, PWTC, Malaysia.

   Title of invention: Innovative and Novel Membrane Morphology Modification

   Awarded by: Malaysia Association of Research Scientists (MARS)

3. Award Name: **Bronze Medal**

   37th International Exhibition of Inventions, New Techniques and Products 2009, 1st – 5th April 2009, Geneva, Switzerland.

   Title of invention: StretchTech: Innovative and Novel Membrane Morphology Modification

   Awarded by: International Juries of Salon International des Inventions

4. Award Name: **Gold Medal**

   20th International Invention, Innovation & Technology Exhibition (ITEX 2009), 15th – 17th May 2009, KLCC, Malaysia.

   Title of invention: NCmem: Indigenous Biomedical Lateral Flow Membrane

   Awarded by: Malaysian Invention and Design Society (MINDS)

5. Award Name: **Bronze Medal**

International Exposition of Research and Invention of Institutions of Higher Learning (PECIPTA 2009), 9-10 October 2009, KLCC, Malaysia

Title of Invention: Synthesis of L-homophenylalanine as Drug Precursor for Treatment of Cardiovascular Diseases via Innovative Integrated MBR

Awarded By: Ministry of Higher Education and University of Malaya

6. Award Name: **Bronze Medal**

Malaysia Technology Expo (MTE 2010), 4-6 February 2010, PWTC, Malaysia

Title of Invention: Cardiovascular Drug Precursor Production via Novel Integrated EMBR

Awarded By: Malaysian Association of Research Scientists

7. Award Name: **Gold Medal**

21st International Invention, Innovation & Technology Exhibition (ITEX 2010), 14-16 May 2010, KLCC, Malaysia

Title of Invention: L-HPA: Cardio Drug Precursor

Awarded By: Malaysian Invention and Design Society (MINDS)

8. Award Name: **Special Award – Winner of INST Invention Award for Excelence in Invention & Innovation**

Taipei International Invention Show & Technomart (INST 2010), 14-16 May 2010, KLCC, Malaysia

Title of Invention: L-HPA: Cardio Drug Precursor

Awarded By: INST Taipei

9. Award Name: **Gold Medal**

International Trade Fair: Ideas-Inventions-New Products (IENA 2009), 5-8 November 2009, Nuremberg Exhibition Center, Nuremberg, Germany

Title of Invention: γ-Alumina with bimodal pore size distribution: transport resistance diminution in bi-layered membrane

10. Award Name: **Gold Medal**

International Exposition of Research and Inventions of Institutions of Higher Learning 2007, organized by Ministry of Higher Education and USM, 10-12 August 2007, Kuala Lumpur Conventional Centre (KLCC), Malaysia

Title of Invention: Gamma-Alumina with Bimodal Pore Size Distribution: Transport Resistance Diminution in Bi-layered Membranes

Category 8 (Proceeding)

1. Abdul Latif Ahmad, Low Siew Chun and Syamsul Rizal Abd Shukor. Effects Of Stretching Temperature Orientation On Membrane Roughness And Performance. 21st Symposium of Malaysian Chemical Engineers (SOMCHE 2007), 12-14 December 2007, Universiti Putra Malaysia, Selangor, Malaysia.
2. A.L. Ahmad, A.R. Sunarti,, K. T. Lee And W.J.N. Fernando, Review of liquid absorbent behaviours in membrane gas absorption process, 12th Asian Pacific Confederation of Chemical Engineering Congress, 4-6 August 2008, Dalian, China.

3. Abdul Latif Ahmad, Low Siew Chun and Syamsul Rizal Abd Shukor, Polymeric Membrane Morphology and Surface Roughness Modification through Stretching Process, 12th Asian Chemical Congress (12ACC 2007), 23-25 August 2007, Putra World Trade Centre, Kuala Lumpur, Malaysia.
4. A.L. Ahmad, A.R. Sunarti,, K. T. Lee And W.J.N. Fernando, Thin Film Composite (TFC) Membrane in Membrane Gas Absorption System, 12th Asian Pacific Confederation of Chemical Engineering Congress, 4-6 August 2008, Dalian, China.
5. A. L. Ahmad, S. C. Low, S. R. Abd Shukor, and A. Ismail. A Preparation and Characterization of Various Membrane Morphologies for Lateral Flow Immunoassay Development. Proceeding of the 6th Regional Symposium on Membrane Science & Technology (MST 2008), 13-15 August 2008, Phuket Graceland Resort & Spa, Phuket, Thailand.
6. A. L. Ahmad, S. C. Low, S. R. Abd Shukor, and A. Ismail. Development of Lateral Flow Membrane for Immunoassay Separation. Proceeding of the Desalination Cooperation among Mediterranean Countries of Europe and the MENA Region (EUROMED 2008), 9-13 November 2008, Dead Sea, Jordan.
7. A. L. Ahmad, P. C. Oh, and S. R. Abd Shukor. Enhanced Biotransformation and Removal of Biocatalytic Compounds In Water By Membrane Bioreactor Technology. Proceeding of the Desalination Cooperation among Mediterranean Countries of Europe and the MENA Region (EUROMED 2008), 9-13 November 2008, Dead Sea, Jordan.
8. A. L. Ahmad, S. A. Rahman, K. T. Lee, and W. J. N. Fernando. Use of AMP As Liquid Absorbent For The Removal of CO_2 From Flue Gas using Polymeric Flat Sheet Membrane Contactor. Proceeding of the Desalination Cooperation among Mediterranean Countries of Europe and the MENA Region (EUROMED 2008), 9-13 November 2008, Dead Sea, Jordan.
9. Y. F. Yeong, A. Z. Abdullah, A. L. Ahmad, and S. Bhatia. Optimization of p-Xylene Separation from Ternary Xylene Mixture Over Silicalite-1 Membrane Using Response Surface Methodology. 7th International Conference on Membrane Science and Technology (MST 2009), 12-15 May 2009, Kuala Lumpur, Malaysia.
10. T. L. Chew, A. L. Ahmad and S. Bhatia. SAPO-34 Membrane for CO_2 Separation from Gaseous Mixtures. 7th International Conference on Membrane Science and Technology (MST 2009), 12-15 May 2009, Kuala Lumpur, Malaysia.
11. A. L. Ahmad, S. A. Rahman, K. T. Lee, and W. J. N. Fernando. Absorption of Carbon Dioxide Through Flat Sheet Membranes Using Various Aqueous Liquid Absorbents. 7th International Conference on Membrane Science and Technology (MST 2009), 12-15 May 2009, Kuala Lumpur, Malaysia.
12. A. L. Ahmad, M. A. T. Jaya, and C. J. C. Derek. Synthesis of Palladium/Titania membrane by Sol-Gel Method. 7th International Conference on Membrane Science and Technology (MST 2009), 12-15 May 2009, Kuala Lumpur, Malaysia.
13. A. L. Ahmad, N. A. A. Sani, and S. H. S. Zein. Preparation of Perovskite Titania Ceramic Membrane by Sol-Gel Method. 7th International Conference on Membrane Science and Technology (MST 2009), 12-15 May 2009, Kuala Lumpur, Malaysia.
14. A. L. Ahmad, S. A. Rahman, K. T. Lee, and W. J. N. Fernando. Review of Liquid Absorbent Behaviours In Membrane Gas Absorption Process. 12th Asia Pacific Confederation of Chemical Engineering Congress, 3-6 August 2009, Dalian, China.
15. P.C. Oh, A.L. Ahmad and S.R. Abd Shukor. L-homophenylalanine Production by Reductive Amination of 2-oxo-4-phenylbutanoic Acid via Integrated Membrane Bioreactor. *Proceedings of 2010 International Conference on Process Engineering and Advanced Materials, 24th Symposium of Malaysian Chemical Engineers (ICPEAM 2010/ SOMCHE 2010)*, 15-17 June 2010, Kuala Lumpur Convention Centre, Malaysia.
16. Y.F. Yeong, A.Z. Abdullah, A.L. Ahmad, S. Bhatia. Optimization of p-xylene Separation from Ternary Xylene Mixture over Silicalite-1 Membrane using Response Surface Methodology. *7th International Conference on Membrane Science and Technology (MST 2009*), 12 – 15 May 2009, Kuala Lumpur, Malaysia.
17. T.L. Chew, A.L. Ahmad, S. Bhatia. Sapo-34 Membrane For CO_2 Separation From Gaseous Mixtures. *7th International Conference on Membrane Science and Technology (MST 2009),* 12-15 May 2009, Kuala Lumpur, Malaysia.

18. Y.F. Yeong, A.Z. Abdullah, A.L. Ahmad, S. Bhatia. Acid-Functionalized Silicalite-1 Membranes: Synthesis, Characterization and Its Performance in m-Xylene Isomerization. *Proceedings of 2010 International Conference on Process Engineering and Advanced Materials, 24th Symposium of Malaysian Chemical Engineers (ICPEAM 2010/ SOMCHE 2010)*, 15-17 June 2010, Kuala Lumpur Convention Centre, Malaysia.
19. T.L. Chew, A.L. Ahmad, S. Bhatia. Microwave Heating for Rapid Synthesis of SAPO-34 Zeolites. *Proceedings of 2010 International Conference on Process Engineering and Advanced Materials, 24th Symposium of Malaysian Chemical Engineers (ICPEAM 2010/ SOMCHE 2010)*, 15-17 June 2010, Kuala Lumpur Convention Centre, Malaysia.
20. Y.T. Ong, S.H. S. Zein, A.L. Ahmad, S.H. Tan. Functionalization of Short Multiwalled Carbon Nanotubes with Poly(3-hydroxybutyrate). *The 2nd International Conference on Bio-Based Polymers (ICBP 2009)*, 11-13 November 2009, Universiti Sains Malaysia, Penang, Malaysia.
21. Y.T. Ong, S.H. S. Zein, A.L. Ahmad, S.H. Tan. Application of Carbon Nanotubes in Environmental Engineering. *International Conference on Environment (ICENV 2008)*, 15-17 December 2008, G-Hotel, Penang, Malaysia.
22. N. Isa, W.J.N. Fernando, A.L. Ahmad, M.R. Othman. A Study of Rate of Reaction of Carbon Dioxide on Hydrotalcite Pallets of Different Size with Temperature. *Proceedings of 2010 International Conference on Process Engineering and Advanced Materials, 24th Symposium of Malaysian Chemical Engineers (ICPEAM 2010/ SOMCHE 2010)*, 15-17 June 2010, Kuala Lumpur Convention Centre, Malaysia.
23. Ong, Y. T., Ahmad, A. L., Zein, S. H. S., Sudesh. K. & Tan, S. H. (2010) Poly(3-hydroxybutyrate) Functionalized Multiwalled Carbon Nanotubes-Chitosan Membranes and Their Application in Pervaporation. *Chemeca Conference 2010*, Adelaide, Australia, 26th – 29th September 2010.
24. T.L. Chew, A.L. Ahmad and S. Bhatia, "Rapid Synthesis of SAPO-34 Zeolite membrane using Microwave Heating for CO2/N2 Binary Gas Separation", *Chemeca Conference 2010*, Adelaide, Australia, 26th – 29th September 2010.
25. S. C. Low, A. L. Ahmad, S. R. Abd Shukor, A. Ismail (2010) Development of Lateral Flow Immunoassay via Various Membrane Morphologies, *8th International Conference on Membrane Science and Technology*, Institut Teknologi Bandung, Bandung, Indonesia, 30th Nov. – 1st Dec. 2010.
26. T.L. Chew, A.L. Ahmad and S. Bhatia, "Thin SAPO-34 zeolite membrane for separation of CO2/CH4 Mixture Gas", *8th International Conference on Membrane Science and Technology*, Institut Teknologi Bandung, Indonesia, 29th Nov – 1st Dec 2010.
27. A.R Sunarti, A.L Ahmad, K.T. Lee and W.J.N Fernando. Membrane gas absorption performances using PVDF microporous membrane. *Proceeding of Postgraduate School of Chemical Engineering Colloquium 2009*, Bukit Jawi Resort, Penang, 31 Oct- 1 Nov 2009.
28. A.L. Ahmad, M.A.T. Jaya, and C.J.C. Derek. Effects of pva/hpc towards unsupported and supported palladium-doped titania membrane. *Symposium of USM Fellowship Holders 2009*, School of Housing, Building and Planning, Universiti Sains Malaysia, Pulau Pinang.
29. A.L. Ahmad, M.A.T. Jaya, and C.J.C. Derek. X-ray diffraction analysis of titania membrane impregnated with Pd^{2+} by sol-gel method at different acid concentration. *School of Chemical Engineering Postgraduate Colloquium 2009*, 31st October – 1st November 2009, Bukit Jawi Golf Resort, Penang.
30. A.L. Ahmad, N.A. Abdullah Sani and S.H.S. Zein. Preparation of Perovskite Titania Membrane for Oxygen Separation Material by Sol-Gel Method.*Symposium of USM Fellowship Holders 2009*, School of Housing, Building and Planning, Universiti Sains Malaysia, Pulau Pinang.
31. A.L. Ahmad, N.A. Abdullah Sani and S.H.S. Zein. Perovskite Titania Ceramic Membrane by Sol-gel Process: Preparation and Characterization. *School of Chemical Engineering Postgraduate Colloquium 2009*, 31st October – 1st November 2009, Bukit Jawi Golf Resort, Penang.
32. A.R Sunarti, A.L Ahmad, K.T Lee and W.J.N Fernando. Hydrophobic TFC Membrane for MGA application. *Proceeding of Malaysian Technical Universities Conference on Engineering and Technology*, 28-29 June 2010, Bayview Hotel, Melaka.

b) Equipment used for this research.
Peralatan yang telah digunakan dalam penyelidikan ini.

Items *Perkara*	Approved Equipment	Approved Requested Equipment	Location
Specialized Equipment Peralatan khusus	1. Haze Meter 2. Heating Tape 3. Conductivity Meter	1. Membrane Bioreactor	MTDC lab, School of Chemical Engineering, USM Engineering Campus
Facility Kemudahan			
Infrastructure Infrastruktur			

- Please attach appendix if necessary.

H. **BUDGET / *BAJET***

Total Approved Budget : RM 466, 330
Total Additional Budget : RM 0
Grand Total of Approved Budget : RM 466, 330

Yearly Budget Distributed
Year 1 : RM 205, 100
Year 2 : RM 135, 600
Year 3 : RM125, 600

Additional Budget Approved
Year 1 : RM -
Year 2 : RM -
Year 3 : RM -

Total Expenditure : RM 466,027.19
Balance : RM 302.81

- **Please attach final account statement from Treasury**

25/4/11

Signature of Researcher
Tandatangan Penyelidik

Date
Tarikh

© Division of Research & Innovation – Universiti Sains Malaysia – **2011331**

I.	**COMMENTS OF PTJ'S RESEARCH COMMITTEE** *KOMEN JAWATANKUASA PENYELIDIKAN PERINGKAT PTJ*
	General Comments: *Ulasan Umum:* .... **Signature and Stamp of Chairperson of PTJ's Evaluation Committee** *Tandatangan dan Cop Pengerusi Jawatankuasa Penilaian PTJ* **Date** : .... *Tarikh* :... **Signature and Stamp of Dean/ Director of PTJ** *Tandatangan dan Cop Dekan/ Pengarah PTJ* **Date** : .... *Tarikh* :.....................................

APPENDICES

Synthesis and Characterization of Polymeric Nitrocellulose Membranes: Influence of Additives and Pore Formers on the Membrane Morphology

A. L. Ahmad,[1] S. C. Low,[1] S. R. Abd. Shukor,[1] A. Ismail[2]

[1]School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan (SPS), Penang, Malaysia
[2]Institute for Research in Molecular Medicine, Health Campus, Universiti Sains Malaysia, 16150 Kubang Kerian, Kelantan, Malaysia

Received 21 May 2007; accepted 5 October 2007
DOI 10.1002/app.27592
Published online 20 February 2008 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The control of the membrane surface and cross-section morphology is extremely important in the enhancement of the wicking and binding ability of the lateral flow membrane, which is one of the processing materials in medicine and health care analysis devices. The lateral flow rate and protein-binding performance is based on the thin layers of the membrane. The challenge of this study was to combine the influences of additives and pore-former materials to obtain a thin lateral flow nitrocellulose membrane with controlled membrane morphologies. Water was found to be an effective pore former for enhancing the porosity and pore size of the membrane. However, too high of a water content increased the surface roughness and decreased the membrane protein-binding ability. Different properties of the individual plasticizers/additives contributed to the disparity of membrane performance in binding and solute lateral wicking time. The correlations between the effects of additives and pore former toward the final membrane structure and performance of the membrane-forming system are discussed extensively. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 108: 2550–2557, 2008

Key words: membranes; microstructure; morphology; synthesis

INTRODUCTION

The membrane is a key element in the production of rapid diagnostic test strips. The study of the behavior of the surface and internal layers of membrane is fundamental in the development of the lateral flow membrane as one of the processing materials in medicine and health care analysis devices. Controlled membrane surfaces and cross-section structures are important in all types of membrane applications. At present, thin-film nitrocellulose (NC) membranes with high mechanical stability and tensile strengths are often described as universal blotting surfaces for protein research,[1,2] lateral flow immunochromatography testing,[3] and immobilization of proteins.[4] If the membrane surface and internal layer structure could be controlled precisely, various kinds of immunological analysis could be performed effectively and accurately.

Membrane production is, nonetheless, a very sensitive process. The most commonly used and important class of membrane preparation method is the phase-inversion technique.[5–7] *Phase inversion* refers to the process in which a polymer solution (liquid phase) inverts into a swollen, three-dimensional macromolecular network (solid state).[7,8] At a particular stage during solvent evaporation, the high-polymer-concentration phase solidifies and forms a solid matrix.[7] In the synthesis of NC membranes, the phase-inversion technique is deemed to be the most suitable method for modifying the surface and internal layer morphology of the membrane.[9] In this process, NC membranes are made from solutions to form a porous solid film.

The microstructure of a membrane is known to depend on various rheological factors, such as the composition of the casting materials; the choice of polymer, solvent, nonsolvent, and additives; and the gelation and crystallization behaviors of the polymer.[6] The addition of organic or inorganic additives, such as glycerol and poly(ethylene glycol) (PEG), as third components to a casting solution has been proven capable of enhancing the permeation properties of membranes.[10,11] Hence, by the manipulation of the initial stage of phase transition and rheological factors, the membrane morphology can be controlled, and porous membranes can be prepared at the desired pore size, porosity, thickness, and surface roughness.[12–14]

Technologically speaking, the solute-spreading trend or liquid-wicking trend reflects the surface

---

Correspondence to: A. L. Ahmad (chlatif@eng.usm.my).

Journal of Applied Polymer Science, Vol. 108, 2550–2557 (2008)
© 2008 Wiley Periodicals, Inc.

WILEY
InterScience
DISCOVER SOMETHING GREAT

ELSEVIER

Chemical Engineering Journal 139 (2008) 172–193

Chemical Engineering Journal

www.elsevier.com/locate/cej

Review

Development of functionalized zeolite membrane and its potential role as reactor combined separator for *para*-xylene production from xylene isomers

Yeong Yin Fong, Ahmad Zuhairi Abdullah, Abdul Latif Ahmad, Subhash Bhatia *

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau Pinang, Malaysia

Received 17 July 2007; received in revised form 11 October 2007; accepted 17 October 2007

Abstract

Zeolite membranes are relatively new class of advanced materials where its characteristics have opened up the new opportunities of their application in separator, sensitive chemical sensors, and catalytic membrane reactor. The present review focuses on the development of functionalized zeolite membrane and subsequently its usage in the catalytic membrane reactor for combined separation and reaction of xylene isomers in a single unit. Synthesis of functionalized zeolite microporous crystal, development of functionalized zeolite membrane and xylene isomerization are presented and discussed. The models representing combined separation and reaction using functionalized zeolite membrane are also suggested.

© 2007 Elsevier B.V. All rights reserved.

Keywords: Functionalized zeolite membrane; Synthesis; Xylene isomerization; Catalytic membrane reactor; Modeling

1. Introduction

The industrial production and recovery of *para*-xylene is an important step in a large petrochemical plant. Xylene has three isomers namely *para*-xylene (molecular size ~0.58 nm), *ortho*-xylene and *meta*-xylene (molecular size ~0.68 nm) and are used as industrial solvents or intermediates for many derivatives [1–5]. Of the three xylene isomers, *para*-xylene has the largest commercial market. As reported by Ministry of Economy, Trade and Industry (MITE), Japan, the worldwide demand for *para*-xylene will increase to 27.8 million tonnes in year 2008 from 18.7 million tonnes in year 2002. *Para*-xylene is the feed for the production of pure terephthalic acid (PTA), which in turn is used in the production of polyester resin and fibers.

In order to meet the *para*-xylene demand, *para*-xylene is currently produced in the petrochemical industry through two different routes: (1) separation of *para*-xylene from its isomers and (2) conversion of less used *ortho*- and *meta*-xylenes to *para*-xylene through xylene isomerization reaction. Separation of *para*-xylene from its isomers is an important operation in the petrochemical industry, but it is difficult to separate the isomers due to their close boiling points, *para*-xylene (boiling point = 138.4 °C), *meta*-xylene (boiling point = 139 °C) and *ortho*-xylene (boiling point = 144.4 °C). Thus, the xylene isomers are currently separated by cryogenic crystallization, or selective adsorption process Parex, which is highly energy intensive. The current technology for xylene isomerization process such as XyMax (ExxonMobil) also consumes high energy. Therefore, there is a need to develop an efficient and energy saving technology to recover and separate *para*-xylene from its isomers.

Zeolite membranes or films have been in focus in recent years because of their well-defined micropore structure, good thermal and structural stability, high mechanical strength, feasible for steady-state operation, low energy consumption, resistance to relatively extreme chemical environment and great potential for combined steps of reaction/separation [2,6–10]. Zeolites are crystalline, microporous aluminosilicates which find extensive industrial usage as catalysts, adsorbents, and ion exchangers with high capacities and selectivities [9,11,12]. When zeolites are grown as films, zeolite membrane is formed. The characteristics of zeolite membrane have found its new application in gas, vapor and liquid separation especially in petrochemical industry based on their properties adsorption, preferential diffusion,

---

* Corresponding author. Tel.: +60 4 5996409; fax: +60 4 5941013.
E-mail address: chbhatia@eng.usm.my (S. Bhatia).

1385-8947/$ – see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.cej.2007.10.019

Advanced Materials Research Vols. 47-50 (2008) pp 238-241
online at http://www.scientific.net
© (2008) Trans Tech Publications, Switzerland
Online available since 2008/Jun/12

Synthesis, Characterization of Phenethyltrimethoxysilane (PE) Modified Organic-Inorganic Hybrid Silicalite-1 Molecular Sieves and Its Transformation into Solid Acid Materials

Y. F. Yeong, A. Z. Abdullah, A. L. Ahmad and S. Bhatia*

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia,
Seri Ampangan, 14300 Nibong Tebal, S.P.S. , Pulau Pinang. Malaysia
E-mail: chbhatia@eng.usm.my

Keywords: Organic-inorganic hybrid materials, Synthesis, Characterization, Solid Acid Materials

Abstract. Organic-inorganic hybrid nanoporous materials with silicalite-1 structure were synthesized in one step co-condensation technique and subsequent transformed into solid acid materials. The tetraethoxysilane (TEOS) was used as the primary inorganic silica source with the phenethyltrimethoxysilane (PE) as an organic modifier. The effect of the PE presence in the initial synthesis mixture was studied by varying the molar ratio of PE to TEOS in the range of 0.05 to 0.20. The resulting organic-inorganic hybrid materials were characterized for its crystallinity (X-ray diffraction, XRD), surface morphology (scanning electron microscopy, SEM) and elemental composition (elemental analysis). The degree of chemical interactions between the organic and inorganic phases was determined by Fourier transform infrared spectroscopy (FTIR). The acid strength of the organic-inorganic hybrid solid acid materials was also obtained by titration technique.

Introduction

In the recent year, the increasing demand of nanostructured materials with tailored chemical and physical characteristic has resulted in the development of new class of organic-inorganic hybrid nanoporous materials [1]. Various organic moieties, including thiol, amine, phenyl, or allyl groups, have been successfully incorporated into mesoporous structure and subsequently transformed into organosulfonic acid group (M41S and SBA-15),following different synthesis methods [2]. The combination of inorganic and organic fragments inside the structure of porous materials significantly modify the physical and chemical properties of inorganic materials; resulting into new desirable advanced properties [3].

The weak hydrothermal stability and inhomogeneous distribution of active sites in the mesoporous hybrid materials have limit their practical and potential applications [1]. Nevertheless, the modification of microporous zeolites with organic group can be of interest and achieved little success [4-5]. Combining inorganic and organic moieties to form well-defined hybrid materials is a challenging task for microporous zeolite. In the present study, synthesis of organic-inorganic hybrid microporous materials having silicalite-1 structure is reported. The effect of the organosilanes at various concentrations present in the initial synthesis mixtures on the formation of hybrid microporous materials was systematically studied. The organic moieties were subsequently transformed into organosulfonic acid solid via sulfonation. The resulting hybrid materials are characterized by a number of physical and chemical techniques to provide detailed information about the chemical nature of the incorporated organic moieties and their effect on the microstructure of the parent silicalite-1.

Experimental

Samples preparation. Organic-inorganic hybrid materials having silicalite-1 structure were synthesized by mixing tetrapropylammonium hydroxide (TPAOH, 1 M, Merck), deionized (DI)

All rights reserved. No part of contents of this paper may be reproduced or transmitted in any form or by any means without the written permission of the publisher: Trans Tech Publications Ltd, Switzerland, www.ttp.net. (ID: 202.170.56.250-28/08/08,11:28:04)

J. Am. Ceram. Soc., 00 [0] 1–6 (2007)
DOI: 10.1111/j.1551-2916.2007.02095.x
© 2007 The American Ceramic Society

journal

Tailoring of a γ-Alumina Membrane with a Bimodal Pore Size Distribution for Improved Permeability

A. L. Ahmad,[†] C. P. Leo, and S. R. Abd. Shukor

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, S.P.S., Penang, Malaysia

Porous γ-alumina with a bimodal pore size distribution has been developed by adding nanosized polystyrene beads to boehmite sol as templating units. The primary pore diameter is in the range of 4–6 nm and the secondary pore diameter is ca. 50 nm with minor pore shrinkage. The unsupported γ-alumina with different porous structures are characterized using thermogravimetric analysis, Fourier transform infrared spectra, X-ray diffraction, N_2 adsorption/desorption, and transmission electron microscopy. γ-alumina with a bimodal porous structure shows reduced transport resistance compared with γ-alumina with a unimodal porous structure in the dye adsorption test. Although the thickness of γ-alumina thin layer increases when more secondary pores are generated, a γ-alumina membrane with a bimodal pore size distribution shows diminution of transport resistance in the water permeability study also.

Nomenclature

C_e	equilibrium concentration (mg/L)
C_t	dye concentration in solution at time t (mg/L)
C_0	initial dye concentration in solution (mg/L)
D_0	diffusion coefficient (m/s)
ΔP	applied pressure difference across the membrane (bar)
Δq	standard deviation
F	fractional uptake of solute
η	liquid viscosity (bar · s)
J	volumetric flux (m/s)
k_1	rate constant of pseudo-first-order model (L/min)
k_m	permeability coefficients of overall membrane (m)
k_α	permeability coefficients of the α-alumina support (m)
k_γ	permeability coefficients of the γ-alumina thin layer (m)
L_m	membrane thickness (m)
m	mass of unsupported γ-alumina (g)
Q	maximum adsorption capacity (mg/g)
q_e	adsorption capacity in equilibrium (mg/g)
q_t	amount of adsorption at time t, (mg/g)
r	particle radius (mm)
r_m	specific resistance (m^2)
t	time (min)
V	volume of solution (L)

I. Introduction

POROUS materials with a highly defined pore distribution are imperative in chemical engineering, especially catalysis. Among the porous materials, nanoporous ceramics are crucial because of their large surface area and great resistance against severe operating conditions.[1] Consequently, nanoporous ceramics have been engineered into thin layers that have broad applications such as gas-separating membranes and catalytic membrane reactors.[2–4] Nanoporous ceramic membranes are usually prepared by the sol–gel method using colloidal sols of nanometer-sized metal oxide particles, formed by hydrolysis and condensation of metal-organic precursors in aqueous media. Sol is then deposited on an adequate macroporous support and calcined to form a thin layer of membrane with a narrow pore distribution and a high surface area.[5]

Nanoporous ceramics with a narrow pore size distribution is not always a definite option. The formation of bimodal porosity in nanoporous material is more practical as the presence of secondary pores considerably enhances the diffusion of molecules. In this regard, several researchers have reported that bimodal porous materials exhibit reduced diffusion resistance and enhanced catalytic function in heterogeneous reactions.[6–9] Until now, incorporation of different types of templates into the sol–gel method has been prevalent in the preparation of bimodal porous ceramics. The templates include emulsion droplets,[10] foam,[11] and solid templates.[12,13] For creating symmetrical secondary macropores, however, scientists often select polymer beads such as polystyrene and polymethyl methacrylate.[14–17] This is the reason why orderly arranged and uniform secondary pores can be generated after assembling of polymer beads using a centrifuge or a filter.[18] Besides this, the control of the primary pore size distribution is independent of additional templating units in the sol. Li–La–Ti–O, ZrO_2, TiO_2, and SiO_2 with well-ordered secondary macropores have been successfully produced using this method.[16,19–21]

Although the preparation of ceramics with a bimodal porous structure is greatly reported, not much work was published on the synthesis of ceramic membranes with a bimodal pore size distribution. Alonso *et al.*[22] only described the preparation of a porous silicon–titanium oxycarbide membrane with a bimodal pore size distribution. Meanwhile, Tsuru *et al.*[23] synthesized a bimodal porous α-alumina membrane for steam reforming of methane. Owing to the lack of related work, it is important to synthesize thin layers of ceramics with a bimodal pore size distribution and to understand their characteristics for further applications in catalysis. In this work, secondary mesopores are tailored in γ-alumina, which plays important roles in industrial chemical processes such as hydrodesulfurization, metathesis, and cracking. From a chemical viewpoint, γ-alumina constitutes a metastable transition alumina phase, resulting in the dehydration of boehmite. Alumina in the γ-phase possesses a large surface area, high pore volume, and great catalytic properties.[24] The catalytic properties of γ-alumina strongly depend on its crystalline structures and textural characteristics.[25] Hence, bimodal porous γ-alumina is synthesized with intermediate crystallization of boehmite. Nanosized polystyrene beads (diameter ca. 50 nm) are selected in this research for tailoring secondary mesopores in γ-alumina with the aim of achieving minor pore shrinkage. This is because severe pore shrinkage usually occurs when macrosized polymer beads are utilized as a template.[26]

Furthermore, a simple dye adsorption test is used to compare the diffusion kinetics of unsupported γ-alumina with a unimodal pore size distribution and unsupported γ-alumina with a

---

A. Krell—contributing editor

Manuscript No. 22986. Received March 26, 2007; approved August 21, 2007.

[†] Author to whom correspondence should be addressed. e-mail: chlatif@eng.usm.my

Available online at www.sciencedirect.com
ScienceDirect
Thin Solid Films 516 (2008) 4319–4324
www.elsevier.com/locate/tsf

Preparation of γ-alumina thin layer with bimodal pore size distribution for diminution of transport resistance in bi-layered membrane

A.L. Ahmad *, C.P. Leo, S.R. Abd. Shukor

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, S.P.S., Penang, Malaysia

Received 5 March 2007; received in revised form 19 November 2007; accepted 6 December 2007
Available online 15 December 2007

Abstract

Porous γ-alumina with bimodal pore size distribution (average pore size of primary mesopores is 4–5 nm and diameter of secondary mesopores is ca. 50 nm) was introduced into bi-layered membranes as an intermediate layer for diminution of transport resistance. The secondary mesopores were tailored in γ-alumina using nanosized polystyrene beads (ca. 50 nm) as templating units. Surfactant templated silica was stacked on top of the γ-alumina with different pore structures to form bi-layered membranes. Besides characterizing using X-ray diffraction, transmission electron microscope, scanning electron microscope and X-ray microanalysis, bi-layered membranes with different types of intermediate layers were also studied using a dead-end stirred cell. The water permeabilities of the bi-layered membranes with secondary mesopores in intermediate layers are much higher than the permeability of the conventional bi-layered membrane. The molecular weight cut-off of the improved membrane was estimated at 400 Da, same as the molecular weight cut-off of the conventional membrane.

© 2008 Elsevier B.V. All rights reserved.

Keywords: γ-alumina; Bimodal; Bi-layer membrane; Scanning electron microscopy; Transport through membranes

1. Introduction

Generally, inorganic nanofiltration membranes are susceptible to low retention of electrolytes at pH values which are near to their iso-electric points (IEP), as this corresponds to a high membrane potential [1]. Without addition of acid or base, most of the ceramic membranes perform unsatisfactorily as the pH of most aqueous electrolyte solutions are usually close to the IEPs of common ceramics membranes (IEP of titania ≈6, zirconia ≈7 and γ-alumina ≈8) [2]. Apparently, the application of ceramic membranes to selectively remove differently charged species at a wide pH range is less practical.

A probable solution of this hitch is to create a stack of layers of materials with different IEPs [3,4]; for example, bi-layered membranes with silica layer supported on the γ-alumina layer. Recently, De Lint et al. [2] showed that the retention of NaCl was maintained higher than 90% in the range of pH 4 to pH 10 by using silica/γ-alumina membrane in the electrolyte separation. The sustainability of NaCl retention in a wide pH range using the bi-layered membrane is considerably better than systems with only a γ-alumina layer [5] or TiO_2 layer [6]. However, the permeate flux is largely reduced when a bi-layered membrane is utilized. Chowdhury et al. [7] demonstrated that pure water permeability of a silica/γ-alumina membrane is far lower than a silica membrane or a γ-alumina membrane. It is evident that the intermediate layer contributes significant transport resistance which reduces the applicability of bi-layered membranes. Fuelled by these reasons, porous γ-alumina with bimodal pore size distribution is introduced in this work as an intermediate layer in silica/γ-alumina membranes for diminution of the transport resistance. Using porous ceramic with a bimodal pore size distribution, it has been shown that secondary macropores enhanced the intraparticle diffusion rate drastically in heterogeneous catalytic reactions [8,9]. However, not much work was reported on the application of porous materials with secondary pores in membrane separation. Alonso et al. [10] described the preparation of porous silicon–titanium oxycarbide membrane with bimodal pore size distribution. Meanwhile, Tsuru et al. [11] reported the preparation and

---

* Corresponding author. Tel.: +60 604 594 1012; fax: +60 604 5941013.
E-mail address: chlatif@eng.usm.my (A.L. Ahmad).

0040-6090/$ - see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.tsf.2007.12.078

J Porous Mater
DOI 10.1007/s10934-007-9165-8

Vanadium oxide supported γ-alumina with bimodal porous structure for intra-particle diffusion enhancement in styrene oxidation reaction

Abdul Latif Ahmad · Choe Peng Leo ·
Syamsul Rizal Abd. Shukor

© Springer Science+Business Media, LLC 2007

Abstract Porous γ-alumina with well arranged secondary mesopores has been contrived using nanosized templating units. The pore size of templated mesopores is precisely controlled as the pore shrinkage is insignificant. The primary pore diameter is ca. 4 nm and the secondary pore diameter is ca. 50 nm. The porous material was characterized using N_2 adsorption/desorption, TEM, XRD and FT-IR. γ-alumina with bimodal pore size distribution shows improved intra-particle diffusion compared to γ-alumina with unimodal pore size distribution in a simple dye adsorption test. γ-alumina with different porous structures were then impregnated with vanadium oxide for catalytic effect comparison. It was perceived that secondary pores improve the styrene oxidation rate after the conversion of styrene reaches 30%.

Keywords γ-Alumina · Vanadium oxide · Template · Bimodal · Oxidation

Abbreviation

C_t	Dye concentration in solution at time t (mg l^{-1})
C_e	Equilibrium concentration (mg l^{-1})
q_e	Adsorption capacity in equilibrium (mg g^{-1})
C_0	Initial dye concentration in solution (mg l^{-1})
D_0	Diffusion coefficient (m s^{-1})
F	Fractional uptake of solute
k_i	Rate parameter of intraparticle diffusion model (mg dye g^{-1} particle $min^{-0.5}$)
m	Mass of unsupported γ-alumina (g)
q_t	Amount of adsorption at time t (mg g^{-1})
r	Particle radius (mm)
t	Time (min)
Q	Maximum adsorption capacity (mg g^{-1})
V	Volume of solution (l)
Δq	Standard deviation

1 Introduction

Porous materials with highly defined pore distribution are imperative in chemical engineering, chiefly catalysis, adsorption and separation. Among the porous materials, nanoporous ceramics are crucial because of their large surface area and great resistance of severe operating conditions [1]. Consequently, nanoporous ceramics such as alumina and silica have found great utility as catalysts support [2, 3] and membrane reactor in chemical industry [4, 5]. Nanoporous ceramics can be prepared by sol–gel method using colloidal sols of nanometer-sized metal oxide particles, formed by hydrolysis and condensation of metal–organic precursors in aqueous media. After drying and calcinations, nanoporous ceramics with narrow pore distribution and high surface are formed [6].

Nanoporous ceramics with narrow pore size distribution is not always a definite option. In catalysis, the formation of bimodal porosity in nanoporous material is more practical as the presence of secondary pores greatly enhances the diffusion of molecules to active sites. Considerable numbers of researchers have reported that bimodal porous materials exhibit reduced diffusion resistance and enhanced catalytic function in heterogeneous reactions [7–10]. Until now, incorporating different types of templates in sol–gel method is prevalent in the

---

A. L. Ahmad (✉) · C. P. Leo · S. R. Abd. Shukor
School of Chemical Engineering, Universiti Sains Malaysia,
Seri Kembangan, 14300 Nibong Tebal, Penang, Malaysia
e-mail: chlatif@eng.usm.my

Available online at www.sciencedirect.com
ScienceDirect
ELSEVIER
Scripta Materialia 57 (2007) 743–746
Scripta MATERIALIA
www.elsevier.com/locate/scriptamat

Effects of membrane cast thickness on controlling the macrovoid structure in lateral flow nitrocellulose membrane and determination of its characteristics

A.L. Ahmad,* S.C. Low and S.R.Abd. Shukor

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, SPS, Penang, Malaysia

Received 15 March 2007; revised 13 June 2007; accepted 16 June 2007
Available online 16 July 2007

Lateral flow in nitrocellulose membrane is widely used as the transport medium in immunological test strips. This study investigates the effect of cast thickness on membrane morphology and its performance. Results show that macrovoid structures only appear in membranes with an initial cast thickness of 800 μm. For membrane performance, it was observed that thicker membranes with more porous structure have a higher lateral wicking rate but simultaneously lower protein-binding ability on the membrane surface.

© 2007 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.

Keywords: Casting; Polymer; Scanning electron microscopy (SEM); Surface structure; Macrovoids

Lateral flow nitrocellulose (NC) membranes with high mechanical stability and tensile strength constitute important elements in rapid diagnostic test strips [1,2]. This thin film membrane functions as a transport medium to carry target analyte, and binds the target analyte together with the immobilizing antigen on the membrane surface control line [1,3]. In diagnostic test strip applications, different membrane materials, surface properties, structure and dimensions are needed for varying diagnostic analyses. Hence, the membrane surface properties and cross-sectional morphology are important parameters in the production of effective immunological assays for diagnostic and healthcare analysis. If the membrane surface and internal layer structure could be controlled precisely, various types of immunological analysis could be performed effectively and accurately.

The mechanism of membrane formation depends on the membrane fabrication method and the casting conditions. The microstructure of a membrane is known to depend on rheological factors such as the choice of casting materials, casting material composition, cast thickness as well as the gelation and crystallization behaviour of the polymer [4]. Membrane morphology can vary depending on rheological factors.

Macrovoid formation and membrane separation performance, which depend strongly on membrane cast thickness, have been widely studied to date [5–9]. However, most investigations have focused on the membrane cross-flow separation performance. There have been no reports on membrane lateral flow (in-plane) liquid distribution performance, which depends on cast thickness. In addition, the formation of "channel"-like macrovoids in the membrane by the dry phase inversion method has not been reported. It is a challenging task to study the effects of membrane thickness by the dry phase inversion method. This is due to the fact that membrane thickness is highly sensitive to the surrounding factors such as humidity and room temperature during the casting process. Thus, in the present study, the membrane casting process was performed in a temperature- and humidity-controlled environment.

In this study, experimental investigations were systematically carried out to investigate the role of cast thickness on membrane morphology and its performance as transport medium in an immunological membrane application. The improved understanding of membrane formation mechanism obtained via this study can be used for the future development of higher-efficiency membranes for immunological analyses.

---

* Corresponding author. Tel.: +60 4 5941012; fax: +60 4 5941013; e-mail: chlatif@eng.usm.my

1359-6462/$ - see front matter © 2007 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.
doi:10.1016/j.scriptamat.2007.06.037

I&EC research
Industrial & Engineering Chemistry Research

Subscriber access provided by UNIV SAINS MALAYSIA

Article

Morphological and Thermal-Mechanical Stretching Properties on Polymeric Lateral Flow Nitrocellulose Membrane

A. L. Ahmad, S. C. Low, S. R. Abd. Shukor, and A. Ismail

Ind. Eng. Chem. Res., **2009**, 48 (7), 3417-3424• DOI: 10.1021/ie801282t • Publication Date (Web): 09 March 2009

Downloaded from http://pubs.acs.org on March 25, 2009

More About This Article

Additional resources and features associated with this article are available within the HTML version:

- Supporting Information
- Access to high resolution figures
- Links to articles and content related to this article
- Copyright permission to reproduce figures and/or text from this article

View the Full Text HTML

ACS Publications
High quality. High impact.

Industrial & Engineering Chemistry Research is published by the American Chemical Society. 1155 Sixteenth Street N.W., Washington, DC 20036

Biotechnology Advances 27 (2009) 286–296

Contents lists available at ScienceDirect

Biotechnology Advances

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

Research review paper

Sustainable biocatalytic synthesis of L-homophenylalanine as pharmaceutical drug precursor

A.L. Ahmad *, P.C. Oh, S.R. Abd Shukor

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia

ARTICLE INFO

Article history:
Received 19 September 2008
Received in revised form 2 January 2009
Accepted 14 January 2009
Available online 22 January 2009

Keywords:
L-homophenylalanine
Pharmaceuticals
ACE inhibitor
Drug precursor
Membrane bioreactor
Biocatalysis

ABSTRACT

Over the past decade, L-homophenylalanine is extensively used in the pharmaceutical industry as a precursor for production of angiotensin-converting enzyme (ACE) inhibitor, which possesses significant clinical application in the management of hypertension and congestive heart failure (CHF). A number of chemical methods have been reported thus far for the synthesis of L-homophenylalanine. However, chemical methods generally suffer from process complexity, high cost, and environmental pollution. On the other hand, enantiomerically pure L-homophenylalanine can be obtained elegantly and efficiently by employing biocatalytic methods, where it appears to be the most attractive process in terms of potential industrial applications, green chemistry and sustainability. Herein we review the biocatalytic synthesis of vital L-homophenylalanine as potentially useful intermediate in the production of pharmaceutical drugs in environmentally friendly conditions, using membrane bioreactor for sustainable biotransformation process. One envisages the future prospects of developing an integrated membrane bioreactor system with improved performance for L-homophenylalanine production.

© 2009 Elsevier Inc. All rights reserved.

Contents

1. Introduction

Modern biotechnology has vast applications in nearly all industrial sectors, particularly medical and pharmaceutical industries. Advances and breakthroughs in target-oriented biotechnology

---

* Corresponding author. Tel.: +60 4 5941012; fax: +60 4 5941013.
E-mail address: chlatif@eng.usm.my (A.L. Ahmad).

0734-9750/$ – see front matter © 2009 Elsevier Inc. All rights reserved.
doi:10.1016/j.biotechadv.2009.01.003

J. Applied Membrane Science & Technology, Vol. 9, June 2009, 19–26
© Universiti Teknologi Malaysia

Biocatalytic Synthesis of Angiotensin-Converting Enzyme (ACE) Inhibitor Drug Precursors Using Membrane Bioreactor: A Review

Ahmad A.L.[1*], Oh P.C.[2] & Abd. Shukor S.R.[3]

[1,2&3]School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia.

ABSTRACT

Over the past decade, the production of angiotensin-converting enzyme (ACE) inhibitor drug intermediates has become increasingly important in the pharmaceutical industry. The most significant clinical application of ACE inhibitors is in the management of hypertension and cardiovascular diseases, particularly congestive heart failure (CHF). A number of chemical methods have been reported thus far for the synthesis of ACE inhibitor precursors. However, chemical methods generally suffer from process complexity, high cost, and environmental pollution. On the other hand, enantiomerically pure ACE inhibitor drug precursors can be obtained elegantly and efficiently by employing biocatalytic methods, where it appears to be the most attractive process in terms of potential industrial applications and green chemistry. In the recent past, membranes are expeditiously used in the enzyme-catalyzed synthesis of optically active pharmaceutical compounds on laboratory and industrial scales, being incorporated in membrane bioreactor system. This technique can be applied for the repeated use of the enzymes in the synthesis via retention or immobilization, especially when dealing with costly biocatalysts. Herein we review the enzymatic synthesis of vital ACE inhibitor drug precursors as potentially useful intermediates in the synthesis of pharmaceutical products, in environmentally friendly condition, using membrane bioreactor for biotransformation.

Keywords: Membrane bioreactor; ACE inhibitors; drug precursors; enzyme-catalyzed processes; pharmaceuticals

1.0 INTRODUCTION

Over the last 20 years, angiotensin-converting enzyme (ACE) inhibitors have achieved wide-spread usage as one of the first line drugs for treatment of hypertension and congestive heart failure [1-3]. ACE inhibitors have long been the cornerstone for therapy of cardiovascular disorders, chronic heart failure, hypertension, left ventricular dysfunction as well as prevention of nephropathy in diabetes mellitus. Recent clinical trials have shown that ACE inhibitors are superior to other classes of drugs in the reduction of morbidity and mortality [4], hence establishing its importance in the pharmaceutical industry. Virtually all ACE inhibitors with therapeutic significance such as enalapril, delapril, lisinopril, quinapril, ramipril, trandolapril, cilazapril and benzapril (Figure 1) [5,6], refer to (S)-2-amino-4-phenylbutanoic acid and (R)-2-hydroxy-4-phenylbutanoic acid or their esters as a common building block, due to the presence of (S)-homophenylalanine moiety as the central pharmacophore unit [7].

Thus far, there exist many conceivable methods of producing enantiomerically pure ACE inhibitor drug precursors, more commonly via chemical [8, 9], microbial or enzymatic routes by way of kinetic resolution or asymmetric synthesis. Chemical methods generally suffer from process complexity, high cost, and environmental pollution. The advantages of microbial or enzyme-catalyzed reactions over chemical synthesis are that reactions involving biocatalysts are often highly stereo-

---

* *Corresponding Author: chlatif@eng.usm.my*

J. Applied Membrane Science & Technology, Vol. 8, December 2008, 9–14
© Universiti Teknologi Malaysia

Effect of DEA in Membrane Gas Absorption System using Polymeric Flat Sheet Membrane

Ahmad A.L.[1*], Sunarti A.R.[2], Lee K.T.[3] & Fernando W.J.N.[4]

[1,2,3&4]School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Pulau Pinang, Malaysia.

[1&2]Faculty of Chemical Engineering and Natural Resources, Universiti Malaysia Pahang, 25000 Kuantan, Pahang, Malaysia.

ABSTRACT

The objective of this study was to investigate the potential process of the removal of carbon dioxide (CO_2) from flue gases by using developed membrane gas absorption system. The experiments were performed in membrane gas absorption system consisted 0.45 μm pore size microporous polyvinylidenefluoride (PVDF) flat sheet membrane. Diethanolamine (DEA) solution was employed as the liquid absorbent. The operating parameters such as the gas and liquid flow rate were setup at 100 cm^3/min and 25 cm^3/min and liquid absorbent was at room temperature. The effect of DEA concentration was studied with variation in range 1M to 5M. In addition, the experiments were carried out with 20%, 40% and 100% gas ratio CO_2 to N_2. The results show that hydrophobic polymeric membranes such as PVDF can be efficiently used for gas absorption with optimized concentration of DEA.

Keywords: Membrane gas absorption, CO_2 removal, DEA, polymeric membrane

1.0 INTRODUCTION

Membranes have become an established technology for carbon dioxide (CO_2) removal since their first use in this application in 1981. Initial acceptance was slow and limited to smaller streams, mostly because of the economic risks involved in treating larger streams, but also because many process design parameters were largely unknown. A further factor was the general downturn in the oil and gas industry in the 1980s. The multiple benefits of membrane technology promised by early innovators have since been proven in a wide variety of installations in many locations around the world, and vendors of traditional CO_2 removal technologies have been quick to acquire or develop membrane-based processes to supplement their older processing routes. In some cases, the most economical approach is to combine membranes with existing technologies or use membranes to debottleneck existing solvent-based plants. Currently, the only commercially viable membranes used for CO_2 removal are polymer based, for example, cellulose acetate, polyimides, polyamides, polysulfone, polycarbonates, and polyetherimide. Polyimide has some potential in certain CO_2 removal applications, but it has not received sufficient testing to be used in large applications.

Flue gas is a gas is released to the atmosphere via a flue and quite often refers to the combustion exhaust gas produced at power plants. Its composition generally depends on what is being burned, but usually consist of mostly carbon dioxide (CO_2) and water vapor as well as nitrogen (N_2) and excess oxygen. It also contains a small percentage of pollutants such as particulate matter, carbon monoxide (CO), nitrogen oxides (NOx) and sulfur oxides (SOx). Lately, the removal of CO_2 from flue gases has received

---

* *Correspondence to*: chlatif@eng.usm.my

Journal of Membrane Science 341 (2009) 96–108

Contents lists available at ScienceDirect

Journal of Membrane Science

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

Process optimization studies of *p*-xylene separation from binary xylene mixture over silicalite-1 membrane using response surface methodology

Yin Fong Yeong, Ahmad Zuhairi Abdullah, Abdul Latif Ahmad, Subhash Bhatia *

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau Pinang, Malaysia

ARTICLE INFO

Article history:
Received 26 February 2009
Received in revised form 22 May 2009
Accepted 27 May 2009
Available online 6 June 2009

Keywords:
Silicalite-1 membrane
Xylene mixture separation
p-Xylene flux
Central composite design
Optimization

ABSTRACT

The process optimization of *p*-xylene separation from *p*-/*o*-xylene binary mixture through silicalite-1 membrane using statistical design of experiments (DoE) is reported in the present study. The silicalite-1 membrane was synthesized and characterized using different analytical techniques. The effect of three important process variables, temperature (150–250 °C), *p*-xylene feed partial pressure (0.04–0.50 kPa) and *p*-xylene feed composition (0.20–0.80) on the separation performance of the membrane was studied. The response surface methodology (RSM) coupled with central composite design (CCD) was used to develop three models to correlate the effect of process variables to three responses: (i) *p*-xylene flux, (ii) *o*-xylene flux and (iii) *p*-/*o*-xylene separation factor. The most influential factor on each of the response was identified using the analysis of variance (ANOVA). The interaction between the three variables was systematically investigated based on three-dimensional response surface plots. The optimum operating condition for the process was determined by setting the optimization criteria to maximize the *p*-xylene flux and *p*-/*o*-xylene separation factor, and to minimize the *o*-xylene flux. The optimum *p*-xylene flux of 3.83×10^{-6} mol/m^2 s and *p*-/*o*-xylene separation factor of 46 were obtained at a temperature of 198 °C, *p*-xylene feed partial pressure of 0.15 kPa and *p*-xylene feed composition of 0.80. The simulated values obtained from the statistical model were in agreement with the experimental results within an average error of ±2.70%. The mass transport of xylene isomers and its separation in the silicalite-1 membrane was related with the characteristics of the membrane.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

The industrial production and recovery of *p*-xylene is an important operation in a large petrochemical plant. Xylene has three isomers namely *p*-xylene (molecular size ~0.58 nm), *o*-xylene and *m*-xylene (molecular size ~0.68 nm), and is used as industrial solvents or intermediates for many derivatives [1–5]. Of the three xylene isomers, *p*-xylene has the largest commercial market. The isomer *p*-xylene is the feed for the pure terephthalic acid (PTA) production, which in turn is used in the production of polyester resin and fibers.

Separation of *p*-xylene from its isomers is an important operation in petrochemical industry. However, the process is difficult because their boiling points (*p*-xylene: 138 °C, *m*-xylene: 139 °C and *o*-xylene: 144 °C) are close. Xylenes isomers are currently separated by cryogenic crystallization, or selective adsorption process Parex, which is highly energy intensive. Therefore, there is a need to develop an efficient and energy saving technology to recover and separate *p*-xylene from its isomers.

Zeolite membranes have been in focus in recent years due to their potential application in wide range of industrial processes especially in petrochemical industry [6–10]. Zeolite membrane with well-defined micropore structure, good thermal and structural stability have potential for its application in membrane reactor, catalytic membrane reactor, sensitive chemical sensor and gas sensor [6,8,11–15]. Currently, zeolite membrane is reported its application in corrosion protection and antimicrobial coatings [16,17].

MFI-type zeolite membranes (ZSM-5 and Silicalite-1) are the most common membranes reported by the researchers. MFI type membranes have the pore structure of straight (*b*-oriented), circular pores (0.54 × 0.56 nm) interconnected with sinusoidal (*a*-oriented), elliptical pores (0.51 × 0.54 nm) and a tortuous path along the *c*-direction [1], which are near to the sizes of many industrially important organic molecules. Therefore, these membranes can be used in the separation of organic compounds with kinetic diameters close to their pores. Moreover, zeolite membrane might offer significant advantage compared to existing technology for the separation

---

Abbreviations: ANOVA, analysis of variance; CCD, central composite design; DF, degrees of freedom; DoE, design of experiments; *F* value, measurement of distance between individual distributions (fit value); MFI, Mobil-Five (Zeolite Socony Mobil-Five, ZSM-5); Prob, probability; RSM, response surface methodology; SEM, scanning electron microscope; XRD, X-ray diffraction.

* Corresponding author. Tel.: +60 4 5996409; fax: +60 4 5941013.
E-mail address: chbhatia@eng.usm.my (S. Bhatia).

0376-7388/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.memsci.2009.05.042

Research Article

Received: 30 July 2009 Revised: 22 September 2009 Accepted: 24 September 2009 Published online in Wiley Interscience: 4 November 2009

(www.interscience.wiley.com) DOI 10.1002/jctb.2291

Separation of *p*-xylene from ternary xylene mixture using silicalite-1 membrane: process optimization studies

Yin Fong Yeong, Ahmad Zuhairi Abdullah, Abdul Latif Ahmad and Subhash Bhatia*

Abstract

BACKGROUND: The design of experiments (DoE) is applied to the process optimization of *p*-xylene (pX) separation from its isomers *m*-xylene (mX) and *o*-xylene (oX) mixture using silicalite-1 membrane supported on α-alumina. A central composite design (CCD) coupled with response surface methodology (RSM) was used to correlate the effect of two separation process variables, temperature (150–250 °C) and pX feed partial pressure (0.10–0.26 kPa) to three responses: (i) pX flux; (ii) pX/oX separation factor; and (iii) pX/mX separation factor. The significant factors affecting each response were elucidated from the analysis of variance (ANOVA). The interaction between two variables was investigated systematically based on three-dimensional response surface plots.

RESULTS: The optimization criteria were used to maximize the value of pX flux, pX/mX separation factor and pX/oX separation factor. The optimum pX flux of 5.94×10^{-6} mol m^{-2} s^{-1}, pX/oX separation factor of 19 and pX/mX separation factor of 20 were obtained at a temperature of 198 °C and pX feed partial pressure of 0.22 kPa.

CONCLUSIONS: The experimental results were in good agreement with the simulated values obtained from the proposed models, with an average error of $\pm$2.90%. In comparison with the conventional approach, DoE provides better flexibility of the process studies and a useful guideline for the membrane process operation for pX separation.
© 2009 Society of Chemical Industry

Keywords: separation; ternary xylene mixture; silicalite-1 membrane; design of experiments (DoE); optimization

INTRODUCTION

Zeolite membranes have been the focus of attention in recent years because of their characteristics (well defined micropore structure, good thermal and structural stability), and have found new applications in gas, vapor and liquid separation, especially in the petrochemical industry.[1–5] One particular process in which zeolite membranes offer significant advantages over existing technology is the separation of close-boiling point hydrocarbons, xylene isomers, which are difficult to separate by distillation or other complex and energy-intensive processes.[6–10]

Separation of para-xylene (pX) from its isomers, meta-xylene (mX) and ortho-xylene (oX) is an important operation in the petrochemical industry, but is difficult due to the close boiling points of xylene isomers (pX: 138 °C, mX: 139 °C and oX: 144 °C). MFI-type zeolite membranes (ZSM-5 or Silicalite-1) could be utilized for energy-efficient xylene separation because their pore openings are close to the kinetic diameters of the isomers (0.58 nm for pX and 0.68 nm for both mX and oX)

Various research groups have studied the separation of xylene isomers using zeolite membranes. Keizer *et al.*[4] obtained separation factors of pX/oX <1.0 at 25 °C and >200 at 102–142 °C, for 0.31 kPa pX and 0.26 kPa oX binary mixture. The results showed that the separation factors were significantly dependent on the operating temperature. Sakai *et al.*[11] reported on a self-supporting MFI-type zeolite membrane for the separation of ternary mixtures of xylene isomers. The separation studies were performed at temperatures between 30 °C and 400 °C, and feed partial pressures between 0.30 kPa and 5.1 kPa. The separation factors pX/mX and pX/oX showed the same maximum value of 250 at 200 °C.

Xomeritakis *et al.*[12] investigated the separation of xylene isomer vapors with oriented MFI membrane in the temperature range 22 °C to 275 °C and xylene feed partial pressures up to 0.7–0.9 kPa. They found that the separation performance of these membranes is directly related to the synthesis conditions and membrane microstructure. Hedlund *et al.*[13,14] synthesized ultra thin MFI-membrane on porous α-alumina support by a two-step masking technique. High pX permeance value of 3×10^{-7} mol m^{-2} s^{-1} Pa^{-1} was obtained together with pX/oX separation factors varying from 3 to 17 in the temperature range 100–390 °C. Tsapatsis and co-workers[3,6] reported the best results for vapor permeation separation of xylene isomers using MFI-type zeolite membranes.

---

* *Correspondence to: Subhash Bhatia, School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau Pinang, Malaysia. E-mail: chbhatia@eng.usm.my*

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau Pinang, Malaysia

Journal of Membrane Science xxx (2010) xxx–xxx

Contents lists available at ScienceDirect

Journal of Membrane Science

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

Synthesis, characterization and reactive separation activity of acid-functionalized silicalite-1 catalytic membrane in *m*-xylene isomerization

Yin Fong Yeong, Ahmad Zuhairi Abdullah, Abdul Latif Ahmad, Subhash Bhatia*

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau Pinang, Malaysia

ARTICLE INFO

Article history:
Received 26 January 2010
Received in revised form 29 April 2010
Accepted 1 May 2010
Available online xxx

Keywords:
Synthesis
Acid-functionalized silicalite-1 membrane
m-Xylene isomerization
Catalytic activity
p-Xylene yield
Reactive separation

ABSTRACT

Propylsulfonic acid-functionalized silicalite-1 membrane and arenesulfonic acid-functionalized silicalite-1 membrane were synthesized over α-alumina support via one-step *in situ* hydrothermal crystallization and subsequent post-synthesis modification. Propylsulfonic acid-functionalized silicalite-1 membrane was synthesized using 3-mercaptopropyltrimethoxysilane (3MP) as an organosilane source whereas for arenesulfonic acid silicalite-1 membrane, phenethyltrimethoxysilane (PE) was used as an organosilane source. The acid capacity of the membrane was varied by adjusting the concentration of organosilane from 5 mol% to 20 mol%. The membranes were characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM) and nitrogen gas permeation. Ammonia temperature-programmed desorption (NH_3-TPD) and Fourier transform infrared spectroscopy (FT-IR) showed the presence of strong Brönsted acid sites in both membranes. The total acid capacity increased with increase in organosilane concentration in the synthesis mixture. Both membranes were tested for their catalytic activity in *m*-xylene isomerization reaction in the temperature range of 355–450 °C. Due to higher acid density, arenesulfonic acid-functionalized silicalite-1 membrane gave higher catalytic activity compared to propylsulfonic acid-functionalized silicalite-1 membrane. At 450 °C, *m*-xylene conversion of 57% with 33% *p*-xylene yield was achieved using arenesulfonic acid-functionalized silicalite-1 membrane with 15 mol% of phenethyltrimethoxysilane, while *m*-xylene conversion of 46% with 28% *p*-xylene yield was achieved using propylsulfonic acid-functionalized silicalite-1 membrane with 15 mol% of 3-mercaptopropyltrimethoxysilane. The enhancement in *p*-xylene yield was due to the simultaneous isomerization reaction and separation of the reaction products through the catalytic membrane. Both catalytic membranes exhibited good structural stability after subjected to isomerization reaction study for 120 h.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Para-xylene (*p*-xylene), meta-xylene (*m*-xylene) and ortho-xylene (*o*-xylene) are the three isomers of xylene and used as industrial solvents or intermediates for many derivatives. Among the three isomers, *p*-xylene has the largest commercial market. *p*-Xylene is the feed for pure terephthalic acid (PTA) production, which is generally produced in the petrochemical industry following: (1) separation of *p*-xylene from its isomers (cryogenic crystallization or selective adsorption process Parex) and (2) conversion of *o*-xylene and *m*-xylene to *p*-xylene through xylene isomerization (XyMax, ExxonMobil) [1–3]. The current technologies for *p*-xylene production are highly energy intensive, and therefore, continuous efforts have been directed to reduce the production cost of *p*-xylene and to improve *p*-xylene yield and selectivity [4–6].

Zeolite membranes have been in focus in recent years because of their well-defined micropore structure, good thermal and structural stability. These membranes are suitable for their application in membrane reactors at high temperature [7–10]. Most of these studies reported MFI type (ZSM-5 and silicalite-1) membranes, due to their pore structure near to the sizes of many important organic molecules [7,8]. The application of zeolite membrane as catalytic membrane reactors has improved catalytic activity, i.e., conversion, selectivity and yield by selective removal of the product from the reactor [11,12]. The combined separation and reaction in a single unit has also provided better flexibility of operation and enhancement in the reaction process [13].

Xylene isomerization is an acid-catalyzed reaction and the use of H-ZSM-5 as catalyst in the membrane reactor as well as H-ZSM-5 catalytic membrane has been reported by a number of researchers. van Dyk et al. [4] reported that *p*-xylene yield of about 10% was enhanced in an extractor-type membrane reactor as compare to the conventional fixed bed reactor. Haag et al. [10] also reported 15% higher *m*-xylene conversion and 10% more *p*-xylene selectivity in H-ZSM-5 catalytic membrane reactor as compared to the conven-

---

* Corresponding author. Tel.: +60 4 5996409; fax: +60 4 5941013.
E-mail address: chbhatia@eng.usm.my (S. Bhatia).

0376-7388/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.memsci.2010.05.009

Please cite this article in press as: Y.F. Yeong, et al., Synthesis, characterization and reactive separation activity of acid-functionalized silicalite-1 catalytic membrane in *m*-xylene isomerization, J. Membr. Sci. (2010), doi:10.1016/j.memsci.2010.05.009

J Porous Mater
DOI 10.1007/s10934-010-9385-1

Rapid synthesis of thin SAPO-34 membranes using microwave heating

**Thiam Leng Chew · Abdul Latif Ahmad ·
Subhash Bhatia**

© Springer Science+Business Media, LLC 2010

Abstract SAPO-34 membranes were prepared by microwave (MW) heating method using a colloidal solution containing tetraethylammonium hydroxide as a template. SAPO-34 in the form of seed and membranes were investigated for their properties such as morphology, pore characteristic, crystallinity and thickness, using characterization method of scanning electron microscope (SEM), X-ray diffraction (XRD), high resolution transmission electron microscopy (HRTEM), FT-IR and nitrogen adsorption-desorption. SAPO-34 membrane was also prepared using conventional hydrothermal heating and studied for its comparison with those formed by MW heating. SAPO-34 membrane containing homogenous SAPO-34 crystals with average size of ~0.7 μm was formed during MW heating. Compare to the conventional hydrothermal heating, MW heating facilitates formation of SAPO-34 crystals with narrower size distribution due to the highly uniform volumetric heating provided by microwave heating. MW heating was able to produce thinner SAPO-34 membrane (1–2 μm) where as hydrothermal heating formed thicker SAPO-34 membrane (~3.6–5.5 μm). The synthesis time for membrane formation was significantly shortened from 24 h for conventional hydrothermal heating to 2 h for microwave heating at 200 °C.

Keywords Hydrothermal synthesis · Microwave heating · SAPO-34 · Membrane

T. L. Chew · A. L. Ahmad · S. Bhatia (✉)
School of Chemical Engineering, Engineering Campus,
Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal,
Seberang Perai Selatan, Pulau Pinang, Malaysia
e-mail: subhashbhati@gmail.com

1 Introduction

Zeolites is microporous and crystalline zeolite with well defined pore structure which has exhibited excellent performance in the field of catalysis, adsorption and separation [1]. In recent years, there have been increasing interest on the study of SAPO-34 (a silicoaluminophosphate zeolite) in gas separation application due to its small pore size, medium acidic strength and high thermal stability [2, 3]. The chabazite framework with pore diameter of 0.38 nm of SAPO-34 [4], which is close to the sizes of gas molecules such as CH_4 and CO_2, has been a promising material for CO_2 separation [2, 4].

There are number of studies reported on the SAPO-34 membrane for gas permeation and separation [2–5]. Although high separation factors were obtained in these studies but the SAPO-34 membrane exhibited low gas permeance. An ideal membrane should perform with high gas permeance as well as separation factor, and thus require the membrane to be thin enough. Reduction in zeolite crystal size, is an important issue in recent years because the properties of the materials are greatly influenced by the change in zeolite crystal dimensions [6]. Zeolite crystal dimension plays impact on the thickness and morphology of the membrane formed.

Conventionally, SAPO-34 is synthesized by time-consuming hydrothermal heating method. There was also effort in synthesizing SAPO-34 film using vapor phase transport (VPT) method [7]. Microwave (MW) heating appears to be an attractive method for the membrane synthesis due to its number of advantages over conventional hydrothermal heating. These advantages are shorter synthesis time, production of small zeolite crystals with narrow size distribution and high purity, rapid and uniform heating [8–10]. Venna and Carreon [11] reported the

Published online: 28 May 2010

Brazilian Journal
of Chemical
Engineering

ISSN 0104-6632
Printed in Brazil
www.abeq.org.br/bjche

Vol. 27, No. 02, pp. 000 - 000, April - June, 2010

A REVIEW ON CARBON NANOTUBES IN AN ENVIRONMENTAL PROTECTION AND GREEN ENGINEERING PERSPECTIVE

Yit Thai Ong, Abdul Latif Ahmad, Sharif Hussein Sharif Zein and Soon Huat Tan*

School of Chemical Engineering, Engineering Campus, Phone: +60-4-599-6475, Fax: +60-4-594-1013,
Universiti Sains Malaysia, Seri Ampangan, 14300, Nibong Tebal, SPS, Pulau Pinang, Malaysia
E-mail: chshtan@eng.usm.my

(Submitted: November 19, 2009 ; Revised: February 20., 2010. ; Accepted: March 21, 2010)

Abstract - Recent developments in nanotechnologies have helped to benchmark carbon nanotubes (CNTs) as one of the most studied nanomaterials. By taking advantages of CNTs extraordinary physical, chemical and electronic properties, a wide variety of applications has been proposed in various engineering fields. In this short review, the contribution of CNTs is addressed in terms of sustainable environment and green technologies perspective, such as waste water treatment, air pollution monitoring, biotechnologies, renewable energy technologies, supercapacitors and green nanocomposites. Consideration of CNTs for large scale application from the aspect of cost and potential hazards are also discussed. Based on the literature studied, CNTs pose a great potential as a promising material for application in various environmental fields.

Keywords: Carbon nanotubes; Environment; Waste water treatment; Air pollution monitoring; Biotechnologies; Renewable energy; Supercapacitors.

INTRODUCTION

Since the discovery of carbon nanotubes (CNTs), they have eventually revolutionized the future nanotechnologies area. CNTs as reported by Iijima (1991) and Bethune et al. (1993), are seamless cylinder-shaped macromolecules with a radius as small as a few nanometers, and up to several micrometers in length. The walls of these tubes are constructed of a hexagonal lattice of carbon atoms and capped by fullerene-like structures. The unique structure of CNTs can be divided mainly into multi-walled carbon nanotubes (MWCNTs) and single-walled carbon nanotubes (SWCNTs). MWCNTs are composed of two or more concentric cylindrical shells of graphene sheets coaxially arranged around a central hollow area with spacing between the layers. In contrast, SWCNTs are made of a single cylinder graphite sheet held together by van der Waals bonds (Balasubramanian and Burghard, 2005; Daniel et al., 2007). Current synthesis techniques including electric arc discharge (Journet et al., 1997), laser ablation (Guo et al., 1995) and chemical vapor decomposition (CVD) (Dai et al., 1996) are used commercially to produce large quantities of CNTs.

CNTs mutable hybridization states and sensitivity of the structure to perturbations in synthesis conditions exploit their unique physical, chemical and electronic properties (as stated in Table 1) which inspire innovation in new technologies and applications. Moreover, these unique and tunable properties offer potential advances in environmental systems from proactive (prevention of environmental degradation, optimizing energy efficiency) to retroactive (waste water reuse, pollutant transformation) (Mauter and Elimelech, 2008).

In this short review, current applications of CNTs in waste water treatment, air pollution monitoring, biotechnology, renewable energy and supercapacitors are explored and a proposal for green nanocomposite design that embraces the 3R (reduce, reuse and recycle) concept has also been discussed. At the end, consideration of CNTs in large scale applications is surveyed from the aspect of cost and potential hazards.

---

*To whom correspondence should be addressed

Separation and Purification Technology 66 (2009) 177–186

Contents lists available at ScienceDirect

Separation and Purification Technology

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

Optimization of membrane performance by thermal-mechanical stretching process using responses surface methodology (RSM)

A.L. Ahmad[a],*, S.C. Low[a], S.R. Abd Shukor[a], A. Ismail[b]

[a] *School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, S.P.S., Penang, Malaysia*
[b] *Institute for Research in Molecular Medicine, Health Campus, Universiti Sains Malaysia, 16150 Kubang Kerian, Kelantan, Malaysia*

ARTICLE INFO

Article history:
Received 20 May 2008
Received in revised form 29 October 2008
Accepted 10 November 2008

Keywords:
Membranes
Morphology
Stretching
Response Surface Methodology (RSM)
Central composite design (CCD)

ABSTRACT

Thermal-mechanical stretching process was proposed to modify membrane morphology for enhancing membrane performances. In this study, Response Surface Methodology (RSM) was used to achieve satisfactory responses of lateral flow membrane performances. The factors considered in this study were the stretching elongation (4–22% of the total membrane sample's length), stretching speed (0.04–0.1 mm/s) and stretching temperature (25–75 °C). The response variables included the lateral flow membrane performances in terms of porosity, lateral wicking time and protein binding ability. These responses were presented graphically based on the central composite design (CCD) of RSM. Atomic Force Microscope (AFM), Field Emission Scanning Electron Microscopy (FESEM) and statistical analysis showed that all the three stretching factors significantly affected the final membrane structures and performances. The optimum stretching elongation of membrane was found to be at 18% with the stretching rate and temperature of 0.07 mm/s and 35 °C respectively. This optimum stretching condition is proven to have high membrane porosity (78.63%), high protein binding ability (4496.5 μg/cm^3) and fast lateral wicking rate (579.0 s/4 cm) where deviation between predicted and actual responses fell within 5%.

© 2008 Elsevier B.V. All rights reserved.

1. Introduction

Thin film nitrocellulose membrane is a well-known transport medium in diagnostic test strip and protein immobilization due to its specific binding affinity to amino acids [1–3]. In lateral flow membrane tests, a solution containing target analyte (either antigen or antibody) is moved by capillary force along a narrow rectangular membrane strip. When the analyte moves along this flow path, it will come in contact with the immobilized detecting reagent that had been dried on the membrane capture zone [4]. The target analyte will be separated from the sample solution via binding with the immobilized detecting reagent. The test result is clearly visible due to the appearance of red colored bands on the membrane strip.

Functionality of the membrane is greatly influenced by its physical morphology and chemical composition [5,6]. Hence, membrane surface properties and cross-sectional morphology are important parameters in developing lateral flow membrane as one of the processing materials in medical and healthcare analysis devices. If membrane surface and internal layer structure could be controlled precisely, various types of immunological analysis could be effectively and accurately performed.

However, the formation mechanism was said to be rather complex as there are various fabrication factors to be considered during the casting stage [7]. It is nearly impossible to mull over all the varieties of mixture components and casting processes with conventional experimental study on membrane formation. Thus, an idea on modification and improvement of membrane morphology through mechanical stretching technique coupled with heat treatment is proposed.

Thermal mechanical stretching is a technique that applies mechanical strength and heat treatment to synthesize membrane with desired morphology [8–10]. Synthesized membrane is uniaxially stretched parallel to the casting direction under optimum heating temperature and stretching speed. The purpose of stretching is to modify the thin film structure while appropriate level of heat treatment is given to ensure that the polymer of membrane is ductile enough to be stretched.

Positive correlations between controlling membrane pore morphology and stretching operation were shown by Morehouse et al. [11,12]. According to their observation, modifying membrane pore structure by stretching operation corresponded to increasing the membrane flux for cross-flow separation [11,12]. This finding was in close agreement with Kurumada et al. [13], who also intentionally modified the PTFE membrane pore dimensions in order to improve the overall membrane performances by membrane stretching operations.

---

* Corresponding author. Tel.: +60 4 5941012; fax: +60 4 5941013.
E-mail address: chlatif@eng.usm.my (A.L. Ahmad).

1383-5866/$ – see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.seppur.2008.11.007

Desalination and Water Treatment
www.deswater.com
5 (2009) 99–105

1944-3994/1944-3986 © 2009 Desalination Publications. All rights reserved

Development of lateral flow membranes for immunoassay separation

A.L. Ahmad[a]*, S.C. Low[a], S.R. Abd Shukor[a], A. Ismail[b], A.R. Sunarti[a]

[a]*School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, S.P.S, Penang, Malaysia*
Tel: +6 04 594 1012, Fax: +6 04 594 1013; email: chlatif@eng.usm.my
[b]*Institute for Research in Molecular Medicine, Health Campus, Universiti Sains Malaysia, 16150 Kubang Kerian, Kelantan, Malaysia*

Received 9 September 2008; Accepted 10 March 2009

ABSTRACT

The lateral flow nitrocellulose membrane is one of the commonly used separation media for bacteria detection in drinking water treatment facilities. In order to enhance its performance, control of the membrane surface and cross-section morphology is primarily important. The challenge of this study is to combine various formulations and casting variables to obtain lateral flow nitrocellulose membranes with the desired morphologies. Through the dry-phase inversion method, the drying temperature was found to be an important parameter in synthesizing membranes as it affects pore structures. A high drying temperature causes agglomeration of the polymer matrix, and thus smaller pores were observed on the membrane surface. This further decreases the membrane lateral liquid migration rate, besides reducing the membrane binding ability for bacteria detection. Results show that by decreasing polymer concentration, membrane surface pores became apparently larger, thus creating a faster lateral migration speed of the water solution. A larger pore size increases the chance for the bacteria detecting agent to bind onto the pore layers, which ultimately enhances the bacteria detection ability of the device.

Keywords: Membrane; Morphology; Microfiltration; Binding ability; Lateral flow

1. Introduction

Virus and bacteria detection in drinking water has brought water quality analysis to a whole new perspective due to waterborne outbreaks [1] such as typhoid fewer [2], *Escherichia coli* O157 [3], *Giardia* and *Cryptosporidium* [4]. These typical waterborne viruses, even in very low concentration, may be lethal [4]. Hence, there is an urgent need to detect the presence of viruses and bacteria in water by a sensitive, reliable and efficient method.

Membrane separation technologies such as ultrafiltration (UF), microfiltration (MF) and reverse osmosis (RO) are known as effective ways to detect low concentrations of viruses in wastewater treatment processes for the past decades [4]. A simple and rapid test strip was developed to detect the presence of selected viruses or bacteria in drinking water [5,6]. However, the developed immunological test strips are not able to cater to applications since different membrane materials, surface properties, structure and dimensions are required for different applications and different analyses.

In membrane technology, the most commonly used and important class of membrane fabrication method is the phase inversion technique [7,8]. Phase inversion refers to the process in which a polymer solution (liquid phase) inverts into a swollen, three-dimensional macromolecular

---

*Corresponding author.

Presented at EuroMed 2008, Desalination for Clean Water and Energy Cooperation among Mediterranean Countries of Europe and the MENA Region, 9–13 November 2008, King Hussein Bin Talal Convention Center, Dead Sea, Jordan.

Separation Science and Technology, 44: 2870–2893, 2009
Copyright © Taylor & Francis Group, LLC
ISSN: 0149-6395 print/1520-5754 online
DOI: 10.1080/01496390903014573

Taylor & Francis
Taylor & Francis Group

Optimization of Membrane Formulation and Process Variables via Crossed-Design Concept in Design of Experimental (DOE)

A. L. Ahmad,[1] S. C. Low,[1] S. R. Abd Shukor,[1] and A. Ismail[2]
[1]School of Chemical Engineering, Engineering Campus, Universiti Sains
Malaysia, Seri Ampangan, Nibong Tebal, Penang, Malaysia
[2]Institute for Research in Molecular Medicine, Health Campus, Universiti
Sains Malaysia, Kubang Kerian, Kelantan, Malaysia

Abstract: A mixture-process design methodology, i.e., the crossed design, is proposed for experimental analysis and optimization. Five mixture materials for membrane formulation and two process factors for casting condition were fixed in the design methodology. The study was to generate a regression model for each of the responses, based on the experimental data and analysis variance of the study. Based on the response models, the optimal blend and casting condition were predicted. The highest desirability function, D, which prevailed from the optimization was 0.66. This optimal blend composition of the cast solution and the process condition in crossed design is proven to increase the membrane performance through the high membrane porosity, the high protein binding ability, and the fast lateral wicking rate.

Keywords: Crossed design, membrane, methodical analysis, mixture, optimization

INTRODUCTION

Lateral flow polymeric membrane is the key element in producing medical and healthcare analysis devices, such as rapid diagnostic test strip (1,2). In diagnostic test strips, different polymer materials, surface

Received 7 January 2009; accepted 30 March 2009.

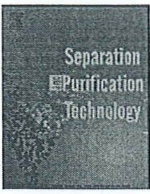
Address correspondence to A. L. Ahmad, School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, S.P.S, Nibong Tebal, Penang 14300, Malaysia. Tel.: +6-04-5941012; Fax: +6-04-5941013. E-mail: chlatif@eng.usm.my

Separation and Purification Technology 75 (2010) 183–192

Contents lists available at ScienceDirect

Separation and Purification Technology

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



Modeling and simulation of an acid-functionalized silicalite-1 membrane reactor for xylene isomerization

Yin Fong Yeong, Ahmad Zuhairi Abdullah, Abdul Latif Ahmad, Subhash Bhatia*

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau Pinang, Malaysia

ARTICLE INFO

Article history:
Received 22 April 2010
Received in revised form 2 July 2010
Accepted 31 July 2010

Keywords:
Modeling
Simulation
Acid-functionalized silicalite-1 membrane
m-Xylene isomerization
Membrane reactor

ABSTRACT

A mathematical model representing *m*-xylene isomerization reaction and subsequent separation of xylene isomers in a catalytic membrane reactor is proposed. Propylsulfonic acid-functionalized silicalite-1 and arenesulfonic acid-functionalized silicalite-1 membranes were used as the catalytic membranes. A triangular reaction scheme was used as the reaction model while *m*-xylene conversion, *p*-xylene selectivity, *p*-xylene yield, *p*-xylene flux and *p*-/*o*-xylene separation factor under isothermal condition were calculated from the model. The simulated results were analyzed and showed good agreement with the experimental data within an error of ±5%. The effects of *p*-xylene diffusivity on *m*-xylene conversion, *p*-xylene flux and *p*-/*o*-xylene separation factor were investigated. Simulated results showed that the increase in *p*-xylene flux enhanced *m*-xylene conversion. *p*-Xylene yield increased with an increase in *m*-xylene conversion. *m*-Xylene conversion by arenesulfonic acid functionalized silicalite-1 membrane was higher compared to that of propylsulfonic acid functionalized silicalite-1 membrane due to higher diffusion rate of *p*-xylene. The removal of *p*-xylene through the membrane contributed to higher *m*-xylene conversion in both of the membranes.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

In recent years, attention has been focused on greater economy and energy efficiency in the chemical industry. This has promoted process intensification strategies such as combined separation and catalytic reaction in a single step of operation [1,2]. Catalytic membrane reactor (CMR) permits simultaneous reaction and separation. Thus, conversions higher than the thermodynamic equilibrium could be achieved by selective removal of reaction products from the reactor [3]. The subsequent separation offers energy savings as compared to conventional separation processes.

Production of *p*-xylene is one of the processes in which a great deal of energy is used in the reaction and separation stages. *p*-Xylene (para-xylene) has the largest commercial market compared to other isomers i.e. *m*-xylene (meta-xylene) and *o*-xylene (ortho-xylene). *p*-Xylene is the feed for pure terephthalic acid (PTA) production. It is generally produced in the petrochemical industry through separation of *p*-xylene from its isomers (cryogenic crystallization or selective adsorption process Parex) and conversion of *o*-xylene and *m*-xylene to *p*-xylene through xylene isomerization (XyMax, ExxonMobil) [4–6]. The current technologies used for *p*-xylene production are highly energy intensive. Therefore, efforts have been directed towards reduction in the production cost of *p*-xylene and to improve *p*-xylene yield and selectivity [7–9] so that better overall process economy can be achieved.

Zeolite membranes have been in focus in recent years because of their well-defined micropore structure, good thermal and structural stability. The use of H-ZSM-5 zeolite as a catalyst in a packed bed membrane reactor (PBMR) as well as H-ZSM-5 catalytic zeolite membrane in catalytic membrane reactor (CMR) for the xylene isomerization has been reported by a number of researchers [7,9–11]. Despite showing enhanced conversion, H-ZSM-5-based membrane reactor for xylene isomerization usually shows moderate to low selectivity to *p*-xylene [12,13]. In our previous paper [14], we reported the synthesis and characterization of acid-functionalized silicalite-1 membranes as well as their activities for *m*-xylene isomerization reaction. An increase in *m*-xylene conversion in the membrane reactor was observed and these values exceeded the equilibrium values due to the continuous removal of reaction product through the membrane. However, the contribution of permeation properties such as reactant diffusivity and products permeation flux needs to be investigated.

In order to develop a scale-up procedure for potential application of catalytic membrane reactor in industrial processes, modeling studies have been reported by a number of researchers. Sousa and coworkers [1,15,16] reported the mathematical modeling of polymeric catalytic membrane reactor by assuming different reactor configurations i.e. plug-flow and well-mixed flow patterns for different kinds of equilibrium reaction under isothermal condi-

---

* Corresponding author. Tel.: +60 4 5996409; fax: +60 4 5941013.
E-mail address: chbhatia@eng.usm.my (S. Bhatia).

1383-5866/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.seppur.2010.07.018

Advances in Colloid and Interface Science 153 (2010) 43–57

Contents lists available at ScienceDirect

Advances in Colloid and Interface Science

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

Ordered mesoporous silica (OMS) as an adsorbent and membrane for separation o carbon dioxide (CO$_2$)

Thiam-Leng Chew, Abdul L. Ahmad, Subhash Bhatia *

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Pulau Pinang, Malaysia

ARTICLE INFO

Available online 4 January 2010

Keywords:
Carbon dioxide
Adsorption
Ordered mesoporous silica
Gas separation
Membrane

ABSTRACT

Separation of carbon dioxide (CO$_2$) from gaseous mixture is an important issue for the removal of CO$_2$ i natural gas processing and power plants. The ordered mesoporous silicas (OMS) with uniform pore structur and high density of silanol groups, have attracted the interest of researchers for separation of carbon dioxid (CO$_2$) using adsorption process. These mesoporous silicas after functionalization with amino groups hav been studied for the removal of CO$_2$. The potential of functionalized ordered mesoporous silica membrane fo separation of CO$_2$ is also recognized. The present paper reviews the synthesis of mesoporous silicas an important issues related to the development of mesoporous silicas. Recent studies on the CO$_2$ separatio using ordered mesoporous silicas (OMS) as adsorbent and membrane are highlighted. The futur prospectives of mesoporous silica membrane for CO$_2$ adsorption and separation are also presented an discussed.

© 2009 Elsevier B.V. All rights reserved.

Contents

1. Introduction

The emission of carbon dioxide (CO$_2$) is one of the serious environmental problems due to its significantly increased concentration in the past five decades and most notably in last 15 years [1]. CO$_2$ has been identified as greenhouse gas, which contributes to the global climate change and global warming. According to the report by Intergovernmental Panel on Climate Change (IPCC), there has been global increment of the atmospheric concentration of CO$_2$ by about 100 ppm (36%) over the last 250 years, from a range of 275–285 ppm in the pre-industrial era (1000–1750) to 379 ppm in 2005. The highest average growth rate of atmospheric CO$_2$ concentration was

---

* Corresponding author. Tel.: +60 4 5996409; fax: +60 4 5941013.
E-mail address: chbhatia@eng.usm.my (S. Bhatia).

0001-8686/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.cis.2009.12.001

J Porous Mater (2011) 18:147–157
DOI 10.1007/s10934-010-9365-5

Propylsulfonic acid-functionalized partially crystalline silicalite-1 materials: synthesis and characterization

Yin Fong Yeong · Ahmad Zuhairi Abdullah ·
Abdul Latif Ahmad · Subhash Bhatia

Published online: 20 February 2010
© Springer Science+Business Media, LLC 2010

Abstract Propylsulfonic acid-functionalized partially crystalline silicalite-1 materials were synthesized via one step co-condensation technique by varying the molar ratio of organosilane source, 3-mercaptopropyltrimethoxysilane (3MP) to tetraethylorthosilicate (TEOS) in the range of 0.05–0.30, and subsequent oxidation of thiol group to propylsulfonic acid using hydrogen peroxide (H_2O_2). These materials were characterized by X-ray diffraction (XRD), high resolution transmission electron microscopy (HRTEM), scanning electron microscopy (SEM) and nitrogen adsorption–desorption method. The structure of these materials was determined by Fourier transform infrared spectroscopy (FT-IR) and ^{29}Si and ^{13}C solid state NMR. XRD results show that % crystallinity of the materials decreased with the increase in 3MP concentration in the synthesis mixture. Selected area electron diffraction (SAED) showed the presence of crystalline and amorphous phases in the samples. An amorphous phase was formed when 3MP concentration was 30 mol% of the total silica source. After elimination of the structure directing agent (SDA) by calcination at 420 °C, thermogravimetric analysis (TGA) shows that the structure was thermally stable up to 550 °C. Ammonia temperature-programmed desorption (NH_3-TPD) shows that the acid capacity of these materials was in the range of 1.19–1.83 mmol H^+/g, which shows that these materials could be used as potential heterogeneous acid catalyst.

Keywords Synthesis · Characterization · Propylsulfonic acid-functionalized · Partially crystalline silicalite-1 · 3-mercaptopropyltrimethoxysilane · Oxidation · Characterization

1 Introduction

Zeolites are crystalline microporous aluminosilicates materials with well-defined pore structure which generated considerable interest in their application in the field of catalysis, adsorption and separation [1]. The application of zeolites as catalysts for industrial processes has been widely reported in the literature [2, 3]. From a catalytic point of view, the efficiency of zeolites as catalyst is related to their crystalline structure, strength and nature of the acid sites present. These properties are important to the shape selectivity catalysis for reactions occurring within the micropore system [4, 5].

A variety of modification methods over ZSM-5 zeolite (MFI-type zeolite) such as by varying Si/Al ratio, crystallite size and morphology, or modifications of extra-framework by cation exchange, pore blockage, isomorphous substitution, metal substitution and functionalization with organic group [3, 6–9], have been reported in order to obtain high selectivity of desired products in the catalytic reactions such as toluene disproportionation, isomerization and alkylation reaction [10]. There are reports to synthesize zeolites with smaller crystal sizes [4, 5] which shorten the intracrystalline diffusion path for the reactants and products and at the same time increase in the surface area provides a large number of external active sites [11–14].

In the synthesis of ZSM-5 by varying Si/Al ratio, the increase in Al increases the acid capacity of the catalyst,

Y. F. Yeong · A. Z. Abdullah · A. L. Ahmad · S. Bhatia (✉)
School of Chemical Engineering, Engineering Campus,
Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal,
Seberang Perai Selatan, Pulau Pinang, Malaysia
e-mail: chbhatia@eng.usm.my

Chemical Engineering Science 66 (2011) 897–906

Contents lists available at ScienceDirect

Chemical Engineering Science

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

Separation of *p*-xylene from binary xylene mixture over silicalite-1 membrane: Experimental and modeling studies

Yin Fong Yeong, Ahmad Zuhairi Abdullah, Abdul Latif Ahmad, Subhash Bhatia *

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau Pinang, Malaysia

ARTICLE INFO

Article history:
Received 13 February 2010
Received in revised form
17 November 2010
Accepted 20 November 2010
Available online 1 December 2010

Keywords:
Xylene mixture separation
Dual-site Langmuir adsorption
Maxwell–Stefan diffusion
Flux
Silicalite-1 membrane
Modeling

ABSTRACT

The permeation of single component and binary mixture containing *p*-xylene and *o*-xylene through silicalite-1 membrane was studied experimentally in the temperature range of 150–250 °C at feed partial pressure of 0.26 kPa for *p*-xylene and 0.22 kPa for *o*-xylene. The model for single component flux based on the combination of dual-site Langmuir isotherm and Maxwell–Stefan formulation was derived. The adsorption parameters were estimated by minimizing the difference between the experimental flux and simulated flux. The heat of adsorption and entropy values obtained were in good agreement with the reported values. The effect of feed partial pressure in the range of 0.20–1.50 kPa on xylene flux was predicted using the adsorption parameters and compared with the experimental values. The Maxwell–Stefan diffusion model, in combination with the ideal adsorbed solution (IAS) theory and single-component adsorption parameters was used to predict the permeation flux of *p*-xylene and *o*-xylene for binary xylene mixture through the silicalite-1 membrane. The simulated results were in good agreement with the experimental data. The simulated adsorption isotherm in higher temperature range of 150–250 °C using the model and derived adsorption parameters could provide useful information for adsorption of xylene molecules on silicalite-1 membrane at a higher operating temperature.

© 2010 Elsevier Ltd. All rights reserved.

1. Introduction

Separation of *p*-xylene from its isomer, *m*-xylene and *o*-xylene using MFI-type zeolite membrane has attracted much attention in the recent years (Gu et al., 2006; Lai and Tsapatsis, 2004; Tarditi et al., 2006a, 2006b; Xomeritakis et al., 2001). MFI-type zeolite membranes, ZSM-5 and silicalite-1 with the uniform dimensions of circular ($0.54\times0.56\ \mathrm{nm}^2$) and elliptical pores ($0.51\times0.54\ \mathrm{nm}^2$) (Choudhary et al., 1997), are capable to separate close-boiling point xylene isomers, which are currently separated by energy intensive technology such as cryogenic crystallization or selective adsorption process Parex. Although highly selective separations of xylene isomers could be achieved based on molecular sieving effect, it is reported in the literature that *p*-xylene adsorbs selectively on MFI zeolite membrane compared to its isomers and its permeation behavior is controlled by an adsorption–diffusion mechanism (Gu et al., 2006; Lai and Tsapatsis, 2004; Tarditi et al., 2006a, 2006b, 2008; Xomeritakis et al., 2001). The two factors that influence the selectivity of the separation are: (i) differences in the adsorption characteristics of the individual species, and (ii) differences in the mobilities (diffusivities) of the components (Krishna, 2001a). Thus, the characteristics of the zeolite membrane dominate the transport of xylene molecules through the membrane (Tarditi et al., 2008).

Extensive studies of adsorption and diffusion of hydrocarbon on silicalite-1 membrane have been reported by Krishna and coworkers (Calero et al., 2001; Kapteijn et al., 2000; Krishna and Paschek, 2000a, 2001b; Krishna and van Baten, 2005; Vlugt et al., 1999). For the description of the equilibrium adsorption in silicalite-1 membrane, the molecular shape and size of the component significantly affect molecular locations in the channels and intersections of silicalite-1, resulting in different adsorption behaviors (Zhu et al., 2000). Therefore, various adsorption models, i.e. Langmuir isotherm, dual site Langmuir isotherm, extended Langmuir isotherm, Ideal Adsorbed Sorption Theory (IAST) and Real Adsorbed Sorption Theory (RAST) have been applied to describe the adsorption isotherms for single component, binary mixture and multicomponents mixture. Krishna et al. (1999) reported that single component adsorption isotherm of benzene, *p*-xylene, *n*-hexane, *n*-heptane and branched alkanes in silicalite-1 membrane is adequately modeled using a dual-site Langmuir model due to the preferential location of molecules at certain sites in the silicalite-1 structure. A distinction between two sites with different sorption characteristics was accounted in this model: site A, refers to the intersections between the straight channels and the zig-zag channels, and site B, refers to the channel interiors (straight or zig zag channels) (Krishna et al., 1999; Krishna and Paschek, 2000a). Narkiewicz-Michalek et al. (1997) confirmed and reported that the two types of adsorption sites were involved in the adsorption of benzene and *p*-xylene on ZSM-5 zeolites.

It is generally accepted that the Maxwell–Stefan formulation offers the most nearest quantitative prediction for diffusion of

---

* Corresponding author: Tel.: +60 45996409; fax: +60 45941013.
E-mail address: chbhatia@eng.usm.my (Y.F. Yeong).

0009-2509/$ - see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ces.2010.11.035

Chemical Engineering Journal 157 (2010) 579–589

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Chemical
Engineering
Journal

Xylene isomerization kinetic over acid-functionalized silicalite-1 catalytic membranes: Experimental and modeling studies

Yin Fong Yeong, Ahmad Zuhairi Abdullah, Abdul Latif Ahmad, Subhash Bhatia*

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau, Pinang, Malaysia

ARTICLE INFO

Article history:
Received 7 October 2009
Received in revised form
25 December 2009
Accepted 11 January 2010

Keywords:
Synthesis
Acid-functionalized silicalite-1 membrane
m-Xylene isomerization
Catalytic activity
Kinetic modeling
Activation energy

ABSTRACT

m-Xylene isomerization kinetics has been studied using acid-functionalized silicalite-1 catalytic membrane in the temperature range of 355–450 °C. Two types of catalytic membranes: (1) propylsulfonic acid-functionalized silicalite-1 membrane and (2) arenesulfonic acid-functionalized silicalite-1 membrane were synthesized on α-alumina support via one-step *in situ* hydrothermal crystallization and subsequent post-synthesis modifications. The membranes were characterized by scanning electron microscopy (SEM), ammonia temperature-programmed desorption (NH_3-TPD) and Fourier transform infrared spectroscopy (FT-IR). Arenesulfonic acid-functionalized silicalite-1 membrane with its higher acidity gave better catalytic activity as compared to propylsulfonic acid-functionalized silicalite-1 membrane. The continuous removal of reaction products over the membrane contributed in the higher *p*-xylene yield. A triangular reaction scheme based on time on stream (TOS) model was used to analyze the experimental data. The simulated results were in good agreement with the experimental results, within an error less than ±5%. The estimated activation energies indicated that conversion of *m*-xylene to *p*-xylene in both acid-functionalized silicalite-1 membranes is affected by the mass transfer rate through the membrane, while conversion of *m*-xylene to *o*-xylene is controlled by the reaction rate.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Para-xylene (*p*-xylene) is the feed for pure terephthalic acid (PTA) production which has the largest commercial market as compared to its isomers, meta-xylene (*m*-xylene) and ortho-xylene (*o*-xylene). With increasing demand of *p*-xylene, selective production of *p*-xylene by *m*-xylene isomerization using zeolite catalyst has gained considerable interest over the years and much attention has been focused on ZSM-5 zeolite as catalyst due to its high activity and shape selectivity [1,2]. Recently, the application of zeolite membrane as catalytic membrane has been reported by number of researchers [3–8] to improve *p*-xylene yield by selective removal of the product from the reactor. The application of catalytic membrane reactor has proven flexibility of its operation and improvement in product selectivity in Knoevenagel condensation reaction between benzaldehyde and ethyl acetoacetate [9–11].

van Dyk et al. [6] reported that *p*-xylene yield of about 10% was enhanced in an extractor-type membrane reactor as compare to the conventional fixed bed reactor. An increment in *p*-xylene production of 28% was observed by Tarditi et al. [5] using 100% exchanged Ba-ZSM-5 in the membrane reactor. Haag et al. [7] also reported 15% higher *m*-xylene conversion and 10% more *p*-xylene selectivity in H-ZSM-5 catalytic membrane reactor as compared to the conventional packed-bed reactor. Recently, Zhang et al. [8] reported that an increment of 26% *p*-xylene yield could be achieved in a silicalite-1 membrane reactor packed with H-ZSM-5 catalyst. However, H-ZSM-5 in membrane reactors gave moderate to low selectivity of *p*-xylene [12,13].

There is a need to develop a catalytic membrane in order to improve *p*-xylene yield, selectivity and separation rate. Due to the higher diffusion rate of *p*-xylene compared to *m*-xylene and *o*-xylene, *p*-xylene could be separated though silicalite-1 membrane [14–16]. However, silicalite-1 is catalytically inactive in its pure form (an aluminum-free analogue of ZSM-5 (Si/Al = ∞)). It is reported in the literature that selective and continuous removal of *p*-xylene from the reaction system could enhance xylene isomerization and thus higher selectivity and yield. Therefore, it has drawn an interest in the synthesis of silicalite-1 membrane with catalytic acid sites.

To best of our knowledge, synthesis of acid-functionalized silicalite-1 membrane and its performance in *m*-xylene isomerization has not been reported. Earlier, we reported the introduction of acid sites in silicalite-1, by adding organic-functional groups into the synthesis mixture and subsequent transformation of organic-functional group into acid-functionalized silicalite-1 [17]. The catalytic activity of these membranes in xylene isomerization

---

* Corresponding author. Tel.: +60 4 5996409; fax: +60 4 5941013.
E-mail address: chbhatia@eng.usm.my (S. Bhatia).

1385-8947/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.cej.2010.01.022

Journal of Membrane Science 357 (2010) 178–184

Contents lists available at ScienceDirect

Journal of Membrane Science

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

Hindered diffusion in lateral flow nitrocellulose membrane: Experimental and modeling studies

A.L. Ahmad[a,*], S.C. Low[a], S.R. Abd. Shukor[a], W.J.N. Fernando[a], A. Ismail[b]

[a] *School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, S.P.S, Penang, Malaysia*
[b] *Institute for Research in Molecular Medicine, Health Campus, Universiti Sains Malaysia, 16150 Kubang Kerian, Kelantan, Malaysia*

ARTICLE INFO

Article history:
Received 2 November 2009
Received in revised form 10 April 2010
Accepted 13 April 2010
Available online 20 April 2010

Keywords:
Lateral flow membrane
Protein diffusion
Hindered diffusion
Modeling
Pore connectivity

ABSTRACT

Lateral flow systems in nitrocellulose (NC) membranes are widely used for transport media applications related to immunoassays. The most important aspect of membranes is their ability to control the diffusion rate of the medium. This study investigated the effects of membrane pore size on lateral diffusion of protein molecules in a NC membrane. Both experimental work and mathematical modeling were carried out in this study. In modeling, the driving force for transportation of the diffusing molecule was the concentration gradient in lateral diffusion cell under unsteady diffusion. Diffusion experiments for Lysozyme and BSA were carried out by using different pore sizes of NC membranes. Good agreement was observed between the developed model and experimental results with correlation coefficients of more than 0.98. Effects of diffusion length and different diffusing molecules toward the lateral diffusion performance in NC membranes were also discussed extensively. The good fit between the model and experimental results has proven the reliability and flexibility of the lateral diffusion model developed in this study. The understanding of the diffusion phenomenon would be a useful tool for membrane properties design and customization of specific membrane applications in immunoassay.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Diffusion of protein molecules in fibrous structures is important in biomedical as well as pharmaceutical industries for the applications such as hydrophobic interaction chromatography, gel permeation chromatography, ultrafiltration, microfiltration and drug release [1–3]. One important example of the biomedical application is the production of diagnostic kit. This application requires detailed knowledge of protein diffusion in order to predict the membrane efficiency and to control the separation during immunoassay processing. The development work on immunoassay often requires substantial input from experimental work, where a general approach is needed to describe the molecular transportation in membrane layers. For performance improvement of immunoassays, it is important to gain substantial understanding of the mass transfer process and the effects of hindered diffusion. For this reason, a number of researchers have derived and elaborated several mathematical models that described the membrane transportation by taking into account the effects of hindered diffusion, pore size, and the adsorption of biological molecules [4–7].

Due to the need of extensive understanding of protein diffusion in membranes, mathematical models are continuously being developed [8–11]. The general conclusion drawn is that the diffusion process through a polymeric membrane can be regarded as the rate-determining step [12]. In the process, the physical resistance exerted by 3D pore structures heading the diffusing molecules to take a longer path, thus affecting the diffusion mass transfer. Due to this reason, it is of immense importance to carry out theoretical modeling in parallel with the experimental work for a better understanding on the underlying phenomena that have taken place in the membrane.

Research to study the phenomena of solution-diffusion in polymeric membranes has started more than a century ago. Some basic insights on the mass transport phenomenon of liquid solution through the membrane layers were derived from the diffusion studies presented by Crank [13] and Neogi [14]. In a dilute solution, the diffusivity rate derived by Fick's law is expected to be independent of concentration. However, at high concentrations, interaction between the diffusing molecules and the solvent can cause the diffusion coefficient to be dependent of the concentration [13]. A typical diffusion coefficient of a dilute solution in polymer can be estimated theoretically, that is at the quantum of 10^{-8} $\mathrm{cm^2/s}$ [15].

In this paper, a modeling study of membrane lateral flow (in-plane) diffusion, which depends on the pore size and diffusion length of membrane, is presented. Lateral diffusivity of protein solution (Lysozyme and BSA) in each membrane plane is determined and correlated with the membrane pore sizes as well as the

---

[*] Corresponding author. Tel.: +60 4 5941012; fax: +60 4 5941013.
E-mail address: chlatif@eng.usm.my (A.L. Ahmad).

0376-7388/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.memsci.2010.04.018

J. Applied Membrane Science & Technology, Vol. 5, June 2007, 23–29
© Universiti Teknologi Malaysia

Preparation and Characterization of Various Membrane Morphologies for Lateral Flow Immunoassay Development

Ahmad A. L.[1*], Low S. C.[1], Abd Shukor S. R.[1] & Ismail A.[2]

[1]School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, S.P.S, Penang, Malaysia.

[2]Institute for Research in Molecular Medicine, Health Campus, Universiti Sains Malaysia, 16150 Kubang Kerian, Kelantan, Malaysia.

ABSTRACT

Lateral flow in nitrocellulose membrane is widely used as the transport medium in immunological test strips. The immunoassay performance depends greatly on the binding mechanism on membrane surface. If the membrane structures could be controlled precisely, various kinds of immunological analysis could be performed effectively and accurately. This study is aimed at a quantitative understanding on (1) the sensitivity of protein quantification on membrane surface by using densitometer and (2) comparison of the protein binding ability on different membrane surface pore structures. Concentration of immobilised protein on membrane surface was measured as a function of colour intensity, which is initially stained with Ponceau S. The findings showed that membrane morphologies significantly affect the sensitivity of membrane protein binding ability. Depending on the membrane pore structures, different red spot sharpness and intensities were observed from the stained protein dot. Membrane with smaller pore creates higher protein immobilization density and thus, increases the sensitivity level of the assay. However, larger membrane pore structure is necessary to induce rapid migration rate for faster assay.

Keywords: Nitrocellulose Membrane; Morphology; Immunoassay; Microstructure; Pore size.

1.0 INTRODUCTION

Lateral flow immunoassays have been widely introduced in the field of food science and healthcare [1, 2], due to their sensitivity, rapid testing, inexpensive manufacturing cost and easy to operate procedure [3]. A large number of testings are available for the detection of certain infectious diseases [4], food hygiene [5, 6], fertility test and etc. This will then translate to swift detection of hazardous substance in food prior to human consumption, which then helps to save lives.

Both immunoassay performance and its effectiveness depend greatly on the transport rate of aqueous solution through membrane pores and the binding mechanism on membrane surface [7]. If the membrane surface and internal layer structures could be controlled precisely, various kinds of immunological analysis could be performed effectively and accurately [8]. Thus, membrane plays an important role on the efficiency of a diagnostic test strip.

Morphology of the selected membrane is crucial to attain the sensitivity of a diagnostic assay [9, 10]. In the production of an immuno diagnostic test strip, microporous lateral flow nitrocellulose (NC) membrane is known as the popular transport medium due to its specific binding affinity to amino acids. Different pores structure is used to immobilize various detecting reagents with varying molecular size. Lower membrane pore structure has higher membrane surface area and able to bind more detecting

---

* *Correspondence to*: chlatif@eng.usm.my

Journal of Applied Sciences 10 (12): 1097-1103, 2010
ISSN 1812-5654
© 2010 Asian Network for Scientific Information

Preparation and Properties of Perovskite-Dispersed Titania Membranes

N.A. Abdullah Sani, A.L. Ahmad and S.H.S. Zein
School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia,
Seri Ampangan, 14300 Nibong Tebal, SPS, Penang, Malaysia

Abstract: Defect-free $SrCo_{0.8}Fe_{0.2}O_3$ perovskite-dispersed titania (perovskite-titania) membrane was successfully prepared by sol-gel method combined with wet impregnation process. In this experiment, titania membrane was immersed in a perovskite solution, dried and calcined to obtain perovskite dispersed in titania membrane. The phase composition and membrane morphology of the membrane were characterized by thermogravimetric analysis (TGA), X-Ray Diffraction (XRD) spectroscopy, Transmission Electron Microscopy (TEM) and Scanning Electron Microscopy (SEM). The TEM result demonstrated that perovskite particles were well-dispersed among the titania particles. The SEM analysis showed that a uniform membrane approximately 0.5 μm thick on titania support was obtained after multiple coatings and calcination at 400°C. O_2 and N_2 permeability across the perovskite-titania membrane was measured to investigate membrane performance. From the permeability result, both gases appeared to decrease as the operating temperature increased, indicating that surface diffusion gradually reduced. The separation of O_2 from its binary mixtures containing N_2 was possible because the value of separation factor N_2/O_2 was slightly higher than the theoretical Knudsen diffusion.

Key words: Titania membrane, Perovskite, sol-gel method, characterization, morphology, gas permeability

INTRODUCTION

Titania is a well-known ceramic material employed in many industrial applications because of its superior physical and chemical properties. Titania membranes have been widely used in ultrafiltration processes, catalytic/photocatalytic membrane reactors, gas separations/reactions and gas sensoring (Ahmad *et al.*, 2004; Tan *et al.*, 2007; Bayraktar *et al.*, 2007; Mohammadi *et al.*, 2007; Ding *et al.*, 2008; Wang *et al.*, 2008). In recent years, many papers have been published on the preparation of membranes derived from sol-gel method (Sekulic *et al.*, 2004), reversed micelle method (Ju *et al.*, 2002), chemical vapor deposition (Lin *et al.*, 1992) and anodic oxidation (Zhao *et al.*, 2005). In this study, sol-gel method is considered the most practical method. This method has demonstrated a homogenous microstructure and a feasibility to produce thin films on complex shapes when dip-coating is used; additional elements can also be added to further modify its properties (Pierre, 1998; Mohammadi *et al.*, 2007; Agoudjil and Benkacem, 2007).

Following the concept of a metal dispersed in a porous membrane, recent investigations have led to the development of metal-ceramic membranes such as zirconia/titania (Sekulic *et al.*, 2004), lanthana/titania (Sibu *et al.*, 2002) and silica/titania (Okada *et al.*, 2001). The abovementioned membranes were fabricated by sol-gel method. In this light, the present study investigates the addition of perovskite particle dispersions within a titania layer formed over a macro-porous titania support.

A perovskite-dispersed alumina membrane was prepared by Irusta *et al.* (1998). The produced catalytic membranes were successfully used as combustors of volatile organic compounds contained in air streams. However, limited information exists about the synthesis and characterization of $SrCo_{0.8}Fe_{0.2}O_3$ perovskite dispersed in titania membranes

In study, therefore, the aim of this study is to develop and characterize perovskite-titania membranes. The titania layer was prepared by sol-gel method, whereas the coating of perovskite materials on the surface of the titania layer was conducted by the wet-impregnation process. Membranes were characterized by thermogravimetric analysis (Tga)/Differential Thermal Analysis (DTA), X-Ray Diffraction Spectroscopy (XRD), Transmission Electron Microscopy (TEM) and Scanning Electron Microscopy (SEM). Permeability of O_2 and N_2 across the perovskite-titania membrane was conducted to measure permeation performance of the membrane.

MATERIALS AND METHODS

Preparation of Titania membrane: Titania sol was prepared using titanium tetra-isopropoxide as a precursor

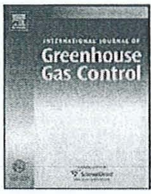
Corresponding Author: A.L. Ahmad, School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, SPS, Penang, Malaysia

International Journal of Greenhouse Gas Control 4 (2010) 495–498

Contents lists available at ScienceDirect

International Journal of Greenhouse Gas Control

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



CO_2 removal using membrane gas absorption

A.L. Ahmad[a,]*, A.R. Sunarti[a,b], K.T. Lee[a], W.J.N. Fernando[a]

[a] *School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Pulau Pinang, Malaysia*
[b] *Faculty of Chemical Engineering and Natural Resources, Universiti Malaysia Pahang, 25000 Kuantan, Pahang, Malaysia*

ARTICLE INFO

Article history:
Received 6 August 2009
Received in revised form 19 October 2009
Accepted 7 December 2009
Available online 12 January 2010

Keywords:
Membrane gas absorption
CO_2 removal
AMP
PVDF membrane

ABSTRACT

The objective of this study is to investigate the potential process for the removal of carbon dioxide (CO_2) from flue gas using fundamental membrane contactor, which is a membrane gas absorption (MGA) system. The experiments consisted of microporous polyvinylidenefluoride (PVDF) flat sheet membrane with 0.1 μm (as module I) and 0.45 μm (as module II) pore size. 2-Amino-2-methyl-1-propanol (AMP) solution was employed as the liquid absorbent. The effect of AMP concentration was studied with variation in the range 1–5 M. In addition, the experiments were carried out with 10%, 20%, 30% and 40% gas ratio of CO_2 to N_2 and pure CO_2 as well. Through contact angle measurement, membranes for module I and module II were obtained with CA values of around 130.25° and 127.77°, respectively. The mass transfer coefficients for module II are lower than those of module I for 1–5 M of AMP. Furthermore, the increase in CO_2 concentration in the feed gas stream enhanced the CO_2 flux as the driving force of the system was increased in sequence from 1 M to 5 M of AMP. However, after the particular percentage (40%) of CO_2 inlet concentration, the CO_2 fluxes seem saturated. The combination of AMP as liquid absorbent and PVDF microporous membrane in MGA system has shown the potential to remove the CO_2 from flue gas. In addition, the higher AMP concentration gave higher mass transfer coefficient at low liquid flow rates.

© 2009 Elsevier Ltd. All rights reserved.

1. Introduction

Flue gas is a gas that exits to the atmosphere via a flue, which has a pipe or channel for conveying exhaust gases from a fireplace, oven, furnace, boiler or steam generator. Quite often, it refers to the combustion exhaust gas produced at power plants. Its composition depends on what is being burned, but it usually consists of mostly nitrogen (N_2), derived from the combustion air, carbon dioxide (CO_2) and water vapor as well as excess oxygen (http://en.wikipedia.org/wiki/Flue_gas). Even though CO_2 is only 10–40 percent from the total post-combustion outlet into atmosphere, it is believed to contribute to undesired global warming. In order to capture CO_2 from power plant flue gas, the developed technology just should not combat climate change but should be cost effective, also have the potential to store the CO_2 for a cleaner and more secure energy in the future. Today's technologies for CO_2 captures are gas absorption processes using solvents, solid sorbents, cryogenic processes, adsorption processes, such as pressure swing adsorption (PSA), thermal swing adsorption (TSA) and iron sponge as well as membrane separation technology.

Traditionally, the gas absorption process for capturing CO_2 can be carried out in many reactors such as bubble columns, packed towers, venturi scrubbers and sieve trays. Although the traditionally packed bed absorbers have been used in the chemical industry for decades, there are several disadvantages such as flooding at high flow rates, unloading at low flow rates, and channelling and foaming, which lead to difficulties in mass transfer of CO_2 between gas and liquid phase. Recently, an alternative technology to capture CO_2 that overcomes these disadvantages and also offers more interfacial area than the conventional approaches is a non-dispersive contactor via a microporous membrane or better known as membrane gas absorption (MGA). The MGA played the biggest role starting late 1990s when global warming issues and green house gases are no more looked as safe to our world. The essential element in MGA is a porous, hydrophobic and polymeric membrane as well. The gas phase remains separated from the liquid absorbent as a result of the hydrophobicity of the membrane. A flue gas stream is fed along one side of the membrane. The components to be removed from the gas stream will diffuse through the gas-filled pores of the membrane. On the other side of the membrane they will be absorbed into the absorption liquid (Paul et al., 2002). The biggest difference between gas separation membrane and MGA is the separation potential. In gas separation membrane process, to get higher permeability usually selectivity is lower and vice versa. However,

---

* *Corresponding author.* Tel.: +60 4 594 1012; fax: +60 4 594 1013.
E-mail address: chlatif@eng.usm.my (A.L. Ahmad).

1750-5836/$ – see front matter © 2009 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ijggc.2009.12.003

Biochemical Engineering Journal 52 (2010) 296–300

Contents lists available at ScienceDirect

Biochemical Engineering Journal

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

Short communication

Synthesis of L-homophenylalanine via integrated membrane bioreactor: Influence of pH on yield

A.L. Ahmad *, P.C. Oh, S.R. Abd Shukor

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Penang, Malaysia

ARTICLE INFO

Article history:
Received 17 March 2010
Received in revised form 19 August 2010
Accepted 20 August 2010

Keywords:
L-homophenylalanine
Membrane bioreactor
Biocatalysis
Biotransformation
Reductive amination
pH

ABSTRACT

L-homophenylalanine can be used as a precursor for production of angiotensin-converting enzyme (ACE) inhibitor, which possesses significant clinical application in the management of hypertension and congestive heart failure (CHF). Pure chiral L-homophenylalanine can be synthesized efficiently by employing enzymatic methods. The biocatalysts used for biotransformation must be retained and reused due to their costs. Conventional processes for L-homophenylalanine production utilized dialysis bag for biocatalysts retention. Nevertheless, this could only be efficiently adopted for laboratory scale processes which possess low scale-up potential. In order to overcome this problem, integrated membrane bioreactor is highlighted for sustainable biocatalytic synthesis of L-homophenylalanine in environmentally friendly conditions which has not been explored by other researchers. The yield of biocatalytic reactions is greatly affected by the pH of the reaction solution, as enzymes are highly sensitive to pH change. Thus in this study, the influence of pH on the product yield at ambient temperature and atmospheric pressure was evaluated. By using L-phenylalanine dehydrogenase and formate dehydrogenase as the biocatalysts, it was found that pure L-homophenylalanine could be synthesized with >80% yield and an enantiomeric excess of over 99% in the integrated membrane bioreactor by adjusting the pH of the reaction solution. The optimal pH was found to be 8.5, when independent of other parameters.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Over the past decade, the production of angiotensin-converting enzyme (ACE) inhibitor drug intermediates has become increasingly important in the pharmaceutical industry. This is mainly due to the fact that ACE inhibitors have achieved wide-spread usage as one of the first line drugs for treatment of hypertension and congestive heart failure [1–3]. Virtually all ACE inhibitors with therapeutic significance such as enalapril, delapril, lisinopril, quinapril, ramipril, trandolapril, cilazapril and benzapril [4,5], refer to L-homophenylalanine as a common building block, due to the presence of L-homophenylalanine moiety as the central pharmacophore unit [6–8].

Thus far, there exist many conceivable methods of producing enantiomerically pure ACE inhibitor drug precursors, more commonly via chemical or biocatalytic routes by way of kinetic resolution [9] or asymmetric synthesis [10]. Today, compound biotransformation which commonly deals with biocatalysts is an extremely important research area for pharmaceutical drug discovery and development [11]. Biocatalysis is a relatively green and stereoselective technology compared to chemical processes which generally suffer from process complexity, high cost, and environmental pollution. Enzyme-catalyzed reactions are often highly stereoselective [12], can be carried out at ambient temperature and atmospheric pressure thus avoiding the use of more extreme conditions. This minimizes problems of isomerization, racemization, epimerization or rearrangement [13]. Nevertheless, biocatalysts used in biotransformation need to be retained for simple downstream processing, and improve process sustainability.

It is possible to prepare L-homophenylalanine from the corresponding 2-oxo-4-phenylbutanoic acid [14,15] via various stereoselective methods including enzymatic reduction. To date, asymmetric reduction of prochiral ketone remains one of the most investigated methods for production of chiral L-homophenylalanine. Many successful advances have been published thus far, with reproducible results of satisfactory yield. One of the most established methods for synthesizing L-homophenylalanine on a laboratory scale was carried out via enzyme-catalyzed asymmetric synthesis of keto acids [16,17]. In this method, prochiral ketone was converted via reductive amination to enantiopure products [18] with bulky side chains by addition of biocatalysts such as L-phenylalanine dehydrogenase [19] in the presence of cofactor.

Since the biological cofactor is too expensive to be used in stoichiometric amounts, efficient and cost-effective *in-situ* regeneration of the compound is prerequisite in order to meet the

---

* Corresponding author. Tel.: +60 4 5941012; fax: +60 4 5941013.
E-mail addresses: chlatif@eng.usm.my, ohpeiching@gmail.com (A.L. Ahmad).

1369-703X/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.bej.2010.08.010

J Sol-Gel Sci Technol (2010) 56:327–332
DOI 10.1007/s10971-010-2309-3

ORIGINAL PAPER

Synthesis of vanadium oxide nanotubes via an ultrasonic method

A. L. Ahmad · B. Koohestani · S. Bhatia · B. S. Ooi

Received: 30 April 2010 / Accepted: 12 August 2010 / Published online: 21 August 2010
© Springer Science+Business Media, LLC 2010

Abstract Vanadium oxide nanotubes were synthesized using V_2O_5 powder as the precursor and hexadecylamine as the structure-directing template using a sol-gel reaction method followed by a one-step hydrothermal treatment. The effect of ultrasonics on the formation of nanotubes is reported. The structure and morphology of the nanotubes were investigated by X-ray diffraction, scanning electron microscopy, and transmission electron microscopy. The inner and outer diameters of the nanotubes varied from 20 to 40 nm and 80 to 100 nm, respectively. The nanotubes measured several micrometers in length.

Keywords Vanadium oxide · Nanotubes · Ultrasonic · Hydrothermal synthesis

1 Introduction

The discovery of carbon nanotubes (CNT) [1] led to determining a unique structure for novel products, in addition to their physical properties and interesting shapes. A comprehensive development in synthetic routes to nanostructured materials was reported in 1991 [2, 3]. The synthesis of different types of inorganic nanomaterials became the motivation for chemists, physicists, and material scientists to focus their attention on the design of a variety structures containing nanoparticles, such as nanocomposites [4–8], whether tubular or otherwise. Vanadium oxide nanotubes (VOx-NTs) are useful materials, due to their high potential applicability in electrochemical devices and catalytic processes, especially widely used for dehydrogenations or partial oxidations of alkanes to olefins [9–14]. Among several Magneli phases of vanadium oxide, VO_2 has received most attention because of its reversible magnetic, electric and optical properties at temperatures around 70 °C [15].

The first successful approach to producing tubular vanadium oxide used CNT as a template [16]. Different types of vanadium oxide precursor may consist of vanadium penta oxide (V_2O_5), vanadium dioxide (VO_2), vanadium oxytrichloride ($VOCl_3$), and vanadic acid (HVO_3) as vanadium source, in addition to alkyl amines ($C_nH_{2n+1}NH_2$) ($4 \leq n \leq 22$) with alkyl chain lengths of a large size range or other types of amines ($H_2N\ [CH_2]n\ NH_2$) ($12 \leq n \leq 20$). Aromatic amines, as direct structural agents, can be used for the successful synthesis of VOx-NTs [17]. Unfortunately, it remains unclear how organic molecules serve as structure-directing templates and cause the deep structural evolution of bulk crystalline V_2O_5 into nanoscrolls [18]. With the proper choice of structure-directing templates involved in the hydrothermal process, VOx-NTs can be prepared with high yields and the unique structures of multilayer scrolls, the interlayer distances of which can easily be controlled. In a hydrothermal synthesis, the reaction path is sensitive to experimental conditions such as temperature, pH, and hydrothermal treatment time [19]. Beginning with a vanadium penta oxide precursor and amine, the sol-gel reaction, combined with a subsequent hydrothermal treatment, could lead to the formation of VOx-NTs. This highly ordering procedure is repeatable and leads to excellent tube yields. The organic template molecules are fixed among vanadium oxide layers in the interior the tube walls. The synthesis of VOx-NTs can be approached via three methods: (1) Adding a

A. L. Ahmad (✉) · B. Koohestani · S. Bhatia · B. S. Ooi
School of Chemical Engineering, Engineering Campus,
Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal,
S.P.S., Penang, Malaysia
e-mail: chlatif@eng.usm.my

J. Am. Ceram. Soc., 93 [11] 3595–3599 (201
DOI: 10.1111/j.1551-2916.2010.04128.
© 2010 The American Ceramic Societ

journal

The Effects of Organic Binders on Palladium Impregnated in TiO Membrane Synthesis: X-Ray Diffraction Analysi

Abdul Latif Ahmad,† Muhammad Azan Tamar Jaya, and Derek Chan Juinn Chie

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan
14300 Nibong Tebal, Penang, Malaysi

This paper reports the effects of organic binders on the crystal microstructure of unsupported, Pd^{2+}-impregnated TiO_2 membranes. The organic binders used in this study were polyvinyl alcohol and hydroxypropyl cellulose. The use of the binders is important in the production of crack-free membranes. However, the use of the binders influenced the microstructure of the unsupported TiO_2 membranes. X-ray diffraction analysis utilized showed that the addition of binders has caused aggregation of palladium oxide (PdO) during its formation in the TiO_2 network. This phenomenon directly affects the effectiveness of PdO presence in the suppression of anatase–rutile phase transition. N_2 adsorb/desorption analysis was conducted to investigate the Pd^{2+}–TiO_2 membrane morphology at different Pd^{2+} loading. Based on the observation, the presence of 2 wt% Pd^{2+} in the TiO_2 microstructure has significant impacts on the increment of surface area, pore volume, and average pore width from the original TiO_2. However, the surface area shows decrement after the Pd^{2+} loading was further increased from 2 to 6 and 10 wt%.

I. Introduction

INORGANIC microporous and mesoporous materials are considered to be good candidate materials for full utilization in the areas of industrial catalysis, separation and purification technology, membranes, sensors, and optical devices. For membrane applications in gas separation and purification, amorphous silica is commonly investigated.[1–3] Other types of ceramic materials such as ZrO_2, TiO_2, and Al_2O_3 are given less attention. Although amorphous silica shows promising performance, its moderate hydrothermal and chemical stabilities are not suitable for certain harsh environments and long-term exposure.[4] TiO_2 gives the highest chemical stability out of the following ceramics: ZrO_2, SiO_2, and Al_2O_3. TiO_2 can withstand phosphoric acid and concentrated hot ammonia.[5] These advantages mean that TiO_2 can be used in both liquid[6] as well as gas separation[7] applications.

Palladium has the ability to absorb hydrogen into its crystal lattice.[8] Decades of research has been conducted on the synthesis of thin, dense palladium membranes for hydrogen purification.[9] Although the selectivity of palladium toward hydrogen is high, the flux is too low for mass production.[10] Palladium membranes are also very expensive. The high cost has driven membrane researchers to find alternatives through the introduction of new materials such as Ti-, Zr-, and Cu-based alloys[11] and through impregnation and doping. Impregnation and doping decrease the amount of palladium that is used.[12] Metal impregnation has caught the attention of researchers because it offer the most simple and effective way to reduce the cost and retai the excellent catalytic performance of palladium on hydrogen.[1 Other elements that are being incorporated into TiO_2 throug the sol–gel technique are Ag, Au, and Ru.[14–16]

The present work reports the preparation and characterization of unsupported TiO_2 membranes that have been impregnated with Pd^{2+} via the sol–gel method. A homogeneou distribution of the impregnating element is obtained throug sol mixing of the synthesized TiO_2 sol and a palladium chlorid solution. X-ray diffraction (XRD) was used to investigate th effects of inorganic binders. A scanning electron microscop (SEM) was used to observe whether or not the impregnation o the metal element caused any defects in the membrane coatin and film. SEM was also used to estimate the thickness of th membrane on top of the supporting surface.

II. Experimental Procedure

(1) Materials

Reagent-grade titanium (IV) isopropoxide (TTIP) (Sigma-Aldrich, Malaysia, 97%), isopropanol (Merck), distilled water nitric acid (HNO_3) (Merck, Malaysia, 65%), palladium chlorid (Merck, 59%), polyvinyl alcohol (PVA) (MW 80 000), hydroxypropyl cellulose (HPC) (MW 88 000), hydrochloric aci (HCl), and titanium dioxide powder (Sigma-Aldrich, technica grade) were used as received.

(2) Preparation of the TiO_2 Colloidal Sol

The experimental preparation of the sol was based on a previou work that was reported by Zaspalis *et al.*[17] The aqueous precipitate solution was stirred for 30 min. HNO_3 was added with final H^+/Ti^{4+} molar ratio of 0.5. The solution was gently stirre and heated at 70°C for approximately 1.5 h. The final solutio was a semiopaque blue with an approximate pH of 1.3 and concentration of 0.3–0.35 mol Ti^{4+}/dm^3. Then, the final solutio was diluted to 0.05 mol Ti^{4+}/dm^3.

(3) Preparation of the HPC and PVA Solutions

One hundred milliliters of distilled water was used to dissolv 0.7 g of HPC and 0.1 g of PVA, respectively. The HPC solutio was stirred for 1 h at 70°C. The PVA solution was stirred an refluxed at 80°C for 2 h. After this, 20 mL of the HPC solutio was added to 30 mL of the sol and followed by 10 mL of th prepared PVA solution.

(4) Preparation of the Palladium Chloride Solution

Palladium chloride powder was weighed and diluted with 30 mL of water. Seven drops of HCl was added into the solution o palladium chloride and water. The solution was stirred an refluxed at 80°C for 2 h. A 2 mL palladium solution was the

---

J. I. C. Diez—contributing editor

Manuscript No. 28022. Received May 16, 2010; approved August 12, 2010.
This work was financially supported by the Universiti Sains Malaysia fellowship and IPS short-term grant.

†Author to whom correspondence should be addressed. e-mail: chlatif@eng.usm.my

Journal of Membrane Science 366 (2011) 166–175

Contents lists available at ScienceDirect

Journal of Membrane Science

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

Synthesis and characterization of TiO_2 membrane with palladium impregnation for hydrogen separation

A.L. Ahmad*, M.A.T. Jaya, C.J.C. Derek, M.A. Ahmad

School of Chemical Engineering, Universiti Sains Malaysia, Engineering Campus, Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau Pinang, Malaysia

ARTICLE INFO

Article history:
Received 10 March 2010
Received in revised form
27 September 2010
Accepted 29 September 2010
Available online 8 October 2010

Keywords:
Ceramic membrane
TiO_2
Palladium
Impregnation
Hydrogen separation

ABSTRACT

This paper reports the synthesis and characterization of TiO_2 membranes impregnated with elemental palladium (Pd–TiO_2) for hydrogen separation at elevated temperatures and pressures. The membranes were prepared by sol–gel method where the elemental Pd was introduced through sol-mixing of titania sol and Pd precursor solution. The Pd–TiO_2 membranes were characterized by X-ray diffraction (XRD), transmission electron microscope (TEM), scanning electron microscope (SEM), thermogravimetric analysis (TG/DTA), N_2 adsorb/desorption analysis, and single permeation test of H_2 and N_2. The XRD analysis revealed that the presence of Pd nanoparticles has retarded the crystallization and anatase-rutile phase transition. The TEM images indicated that the Pd–TiO_2 membranes are composed of non-uniform particle size as compared to that of TiO_2 membranes alone. For four times dip-coating, the membrane thickness was measured to be approximately 634 nm. By impregnating small concentration of Pd, it significantly enlarged the pore size of the original TiO_2 membrane structure besides increasing its surface area and pore volume. However, a reverse trend was observed when the Pd concentration was increased further. Single permeance of H_2 and N_2 for both TiO_2 and Pd–TiO_2 membranes showed that the transport mechanism to be dominated by Knudsen diffusion and Poiseuille flow. The ideal selectivity of H_2/N_2 crossing through the Pd–TiO_2 membrane was calculated and found to be less than the theoretical value of Knudsen diffusion based separation. The selectivity of Pd–TiO_2 membrane increased with the temperature increment, whereas the same trend was not observed for TiO_2 membrane. These results show that, there were other transport mechanisms involved apart from Knudsen diffusion and Poiseuille flow.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Many studies have recently been conducted on the development of membranes for hydrogen separation on an industrial scale. Cost and ease of fabrication, stability and sustainability under industrial operating conditions (pressure, temperature and contaminants), flux and other relevant specifications must be considered in commercial membranes [1]. Inorganic membranes derived from ceramic materials are able to separate hydrogen gas from heavier gases such as CO_2, N_2 and CH_4. Ceramic membranes possess considerably higher thermal, chemical, and mechanical stabilities than polymeric membranes, and are suitable for gas separation and purification [2].

Numerous studies have shown that ceramic membranes comply with industrial specifications. Besides excellent physical and chemical properties, ceramic membranes are relatively inexpensive to produce and can be synthesized by simple fabrication procedures, such as the sol–gel method. An early sol–gel method used to develop ceramic membranes for hydrogen separation was reported by de Lange et al. [3].

Pd metal has gained considerable attention in membrane research because it is capable of dissociating hydrogen into protons, and it allows for the diffusion and reassociation of hydrogen gas. Furthermore, Pd membranes offer high selectivity by preventing other gases from crossing through. Despite the fact that these membranes are commercially available, improvements in thickness [4], alloy [5,6], permeability [7], support resistance [8], defects and degradation [9–11], module design [12] and fabrication methods [13,14] have been extensively researched. However, the high cost of this material is a serious limitation on the use of palladium membranes. Thus, impregnation of Pd into cheap and abundant materials such as ceramic matrices is desirable [15].

To improve hydrogen selectivity or physical properties, ceramic membranes have been impregnated with a variety of metals including boron [16], Pd/YSZ [17], Pd/alumina [18,19], Pd/silica [20], V–Ti–Ni [21], nickel [22], and Ni–Ba($Zr_{0.1}Ce_{0.7}Y_{0.2}$)O_3 [23]. However, there are limited examples of doped titania membranes for hydrogen separation. According to Kluiters [24], titanium dioxide (TiO_2) is a potential material for hydrogen separation membranes. Furthermore, Zaspalis et al. [25] reported that TiO_2 membranes pos-

---

* Corresponding author. Tel.: +60 4 593 7788x6414; fax: +60 4 594 1013.
E-mail address: chlatif@eng.usm.my (A.L. Ahmad).

0376-7388/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.memsci.2010.09.056

Separation and Purification Technology 76 (2011) 419–427

Contents lists available at ScienceDirect

Separation and Purification Technology

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

Poly(3-hydroxybutyrate)-functionalised multi-walled carbon nanotubes/chitosan green nanocomposite membranes and their application in pervaporation

Yit Thai Ong[a], Abdul Latif Ahmad[a], Sharif Hussein Sharif Zein[a], Kumar Sudesh[b], Soon Huat Tan[a,*]

[a] *School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, SPS, Pulau Pinang, Malaysia*
[b] *Ecobiomaterial Research Laboratory, School of Biological Sciences, Universiti Sains Malaysia, 11800 Penang, Malaysia*

ARTICLE INFO

Article history:
Received 24 August 2010
Received in revised form
10 November 2010
Accepted 11 November 2010

Keywords:
Multi-walled carbon nanotube (MWCNT)
Chitosan
Green nanocomposite membrane
Pervaporation

ABSTRACT

The aim of this research was to align poly(3-hydroxybutyrate) (PHB)-functionalised multi-walled carbon nanotubes (MWCNTs) into a chitosan matrix. The MWCNTs were first functionalised with PHB and aligned into a membrane filter template through a filtration process. A solution casting technique was then applied to cast the chitosan onto the template to form PHB-MWCNT/chitosan nanocomposite membranes. The functionalised MWCNTs and resulting membranes were characterised using thermogravimetric analysis (TGA), Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The TGA demonstrated that approximately 40 wt.% of the PHB was successfully functionalised in the MWCNTs, and TEM showed that the polymer wrapped on the nanotubes surface. The presence of the PHB functional moieties helped to improve the dispersion and compatibility of the MWCNTs in the chitosan matrix. When we applied the PHB-MWCNT/chitosan nanocomposite membranes in the pervaporation process of 1,4-dioxane dehydration, the nanocomposite membrane showed a relatively high permeation flux and selectivity towards water, compared to existing membrane.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

1,4-Dioxane ($C_4 H_8O_2$) is a colourless synthetic heterocyclic organic compound that is widely used as a solvent in the pharmaceutical and chemical industries. It is highly soluble in water and forms an azeotropic mixture at a water composition of 18 wt.%. Additionally, the boiling point of 1,4-dioxane (101 °C) is close to that of water causing the compound to form close boiling mixtures at various compositions with water [1]. Unlike conventional distillation, pervaporation is a better alternative because of its ability to separate azetropic liquid mixtures without the use of entertainer and it is effective in removing the minor component of the liquid mixtures [2]. Pervaporation is a membrane-based process to separate liquid mixtures, which involves a phase change from a liquid to a vapour. Generally, a pervaporation set-up consists of a polymeric membrane that serves as a separating barrier, and a vacuum pump is used at the downstream side to create a driving force so that the component with a higher affinity is transported across the membrane. In pervaporation, proper selection of membrane materials is essential because the materials critically affect the overall performance of the process.

Recently, there has been a growing interest in employing biodegradable polymers in pervaporation membranes. Biodegradable polymers, which are usually derived from natural renewable resources, possess a great potential commercial value to replace synthetic polymers since the detrimental effects of synthetic polymers have become increasingly evident in recent decades. Because of its significant hydrophilicity, good film-forming characteristics and excellent chemical resistance, chitosan has become one of the most studied biodegradable pervaporation membranes. It is a natural polymer that is derived through the deacetylation of chitin under alkaline conditions. However, pure chitosan membranes often suffer from low selectivity because of excessive swelling of the membrane in aqueous solutions. Techniques to overcome the swelling issue include cross-linking and blending with various additives. In addition, incorporation of a nano-filler to form a "green" nanocomposite membrane has recently drawn considerable attention. This nanocomposite membrane, which is comprised of natural renewable resources (chitosan) and inorganic filler (carbon nanotubes (CNTs)), has been viewed as an environmentally friendly material because it embraces the 3R (reduce, reuse and recycle) concept [3].

In the fabrication of such a "green" nanocomposite membrane, the dispersion and alignment of CNTs are crucial in the quality of the resultant nanocomposite membrane. It has been reported that well dispersed and aligned CNTs are required to induce a

---

* Corresponding author. Tel.: +60 4 599 6475; fax: +60 4 594 1013.
E-mail address: chshtan@eng.usm.my (S.H. Tan).

1383-5866/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.seppur.2010.11.013

Journal of
Ceramic Science
and Technology

J. Ceram. Sci. Tech., 02 [xx] xx-xx (2010)
DOI: 10.4416/JCST2010-00026
available online at: http://www.ceramic-science.com
© 2010 Göller Verlag

Influence of Calcination Temperature and Perovskite Concentration on the Morphology, Gas Permeance and Selectivity of Perovskite-Titania Membranes

A.L. Ahmad*, N.A. Abdullah Sani, S.H.S. Zein

School of Chemical Engineering, Engineering Campus, Universiti Sains
Malaysia, Seri Ampangan, 14300 Nibong Tebal, S.P.S, Penang, Malaysia

receive July 02, 2010; received in revised form August 10, 2010; accepted September 24, 2010

Abstract

Defect-free titania membranes doped with different concentrations of $SrCo_{0.8}Fe_{0.2}O_3$ perovskite were successfully prepared using a sol-gel technique combined with a wet impregnation process. The titania membranes were immersed in a perovskite solution, dried, and calcined to obtain perovskite affixed inside the porous titania membranes. The phase transformation of titania and perovskite was investigated by varying the calcination temperature from 300 to 500 °C. The effect of perovskite concentration on the titania membrane morphology is described in detail. The results showed that calcination at 400 °C was preferable for preparing a perovskite-titania membrane with fully crystallized anatase and perovskite phases. The performance of the perovskite-titania membranes with various perovskite concentrations in terms of the O_2 and N_2 permeances and O_2/N_2 selectivity was also studied. We determined that the perovskite concentration affected the gas permeation performance of the membrane and that the P3/titania membrane provided the highest gas permeance and selectivity.

Keywords: titania membrane, perovskite, calcination temperature, concentration, morphology, phase structure

I. Introduction

The perovskite-type oxide $SrCo_{0.8}Fe_{0.2}O_3$ is a typical mixed ionic electronic conducting membrane material that has garnered a significant amount of attention and has been applied in the chemical and petroleum industries. Owing to its high oxygen ion and electron conductivity, it can operate steadily for long periods of time at high temperatures and has been used widely as a membrane for oxygen separation in the oxidative coupling of methane and the partial oxidation of methane [1]. Usually, the perovskite membrane is used independently and is prepared with a conventional solid state method to form a perovskite powder. This powder is compressed into a disc and sintered before application in gas-separation processes [2].

Ceramic membranes made with a variety of materials, such as alumina, titania, silica, zeolites, and zirconia, have been extensively studied. Among them, titania has gained considerable attention owing to its high chemical resistance and several unique characteristics, including semiconductivity, catalytic and photocatalytic behaviour, and high water flux. The potential applications of titania membranes are numerous. They include ultrafiltration processes, catalytic/photocatalytic membrane reactors, gas separations/reactions, and gas sensors [3-6].

Titania membranes are usually prepared in porous form with the sol-gel process. This method is considered the most practical one. The sol-gel process is based on the controlled hydrolysis of alkoxides and condensation-polymerization reactions at low temperature. The produced membrane has a very high specific area, and additional elements can be added to further modify its properties. This method has demonstrated excellent compositional control, homogeneous microstructure, and feasibility in producing thin films on complex shapes when dip-coating is used [6, 9, 10].

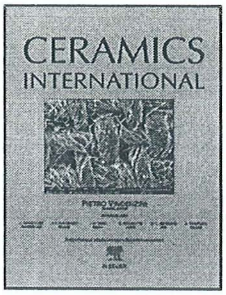
Although titania membranes can offer various advantages, they possess one drawback with regard to their anatase-rutile phase transformation. This phase transformation causes large volume and energy changes, leading to large variations in particle size and, therefore, in the membrane pore size and porosity [11]. The transformation from the anatase to the rutile phase is not favoured because the anatase phase has a higher specific surface area compared to the rutile phase. Thus, the anatase phase is more useful as a membrane material in gas-separation applications [12].

Another problem relating to titania membranes in gas-separation processes is their selectivity. Titania membranes with porous structures have a high permeability but low selectivity. Hence, one of the objectives of this study was to improve the selectivity of the membrane by affixing perovskite materials in the membrane pores in a wet impregnation process. This is a well-known method in the development of heterogeneous catalysts and has been utilized to deposit nano-sized particles into the electrode structures of solid oxide fuel cells [13]. Thus, we believed that it would impart highly uniform perovskite into the titania matrix.

---

* Corresponding author: chlatif@eng.usm.my

Accepted Manuscript



Title: Synthesis of a TiO_2 ceramic membrane containing $SrCo_{0.8}Fe_{0.2}O_3$ by the sol-gel method with a wet impregnation process for O_2 and N_2 permeation

Authors: Abdul Latif Ahmad, Nur Aimie Abdullah Sani, Sharif Hussein Sharif Zein

PII: S0272-8842(11)00158-1
DOI: doi:10.1016/j.ceramint.2011.03.036
Reference: CERI 4062

To appear in: *Ceramics International*

Received date: 24-1-2011
Revised date: 27-3-2011
Accepted date: 27-3-2011

Please cite this article as: A.L. Ahmad, N.A.A. Sani, S.H.S. Zein, Synthesis of a TiO_2 ceramic membrane containing $SrCo_{0.8}Fe_{0.2}O_3$ by the sol-gel method with a wet impregnation process for O_2 and N_2 permeation, *Ceramics International* (2008), doi:10.1016/j.ceramint.2011.03.036

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

Chemical Engineering Journal xxx (2011) xxx–xxx

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

ELSEVIER

Chemical
Engineering
Journal

Synthesis of 2-oxo-4-phenylbutanoic acid: Parameter optimization using response surface methodology

A.L. Ahmad*, P.C. Oh, S.R. Abd Shukor

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Penang, Malaysia

ARTICLE INFO

Article history:
Received 26 November 2010
Received in revised form 31 March 2011
Accepted 6 April 2011

Keywords:
Synthesis
Design of experiment
Optimization
Response surface methodology

ABSTRACT

A simple approach for the formation of 2-oxo-4-phenylbutanoic acid (OPBA), which might find important use as substrate for production of angiotensin-converting enzyme (ACE) inhibitor drug precursor, was studied. With the appropriate choice of solvent and base, it was possible to produce pure OPBA in good yields. In this study, optimization of OPBA synthesis was investigated. Response surface methodology (RSM) and 4-factor central-composite rotatable design (CCRD) were employed to evaluate the effects of synthesis parameters, such as reaction temperature (40–70 °C), reaction time (5–13 h), choice of solvent (dried tetrahydrofuran or diglyme) and base (sodium methoxide or sodium hydride) on product yield. On the basis of numerical optimization, the optimum conditions for synthesis were: reaction time 6 h, reaction temperature 65 °C, using dried tetrahydrofuran as solvent and sodium methoxide as the base. The yield obtained was 98%. Comparison of predicted and experimental values revealed good correspondence, implying that empirical models derived from RSM can be used to adequately describe the relationship between the factors and response in OPBA synthesis.

© 2011 Published by Elsevier B.V.

1. Introduction

2-Oxo-4-phenylbutanoic acid (OPBA) is the common substrate used to produce various angiotensin-converting enzyme (ACE) inhibitor drug precursors, including (S)-2-amino-4-phenylbutanoic acid and (R)-2-hydroxy-4-phenylbutanoic acid. ACE inhibitors are a class of antihypertensive drug which has shown to reduce morbidity and mortality in chronic heart failure. The corresponding ACE inhibitor drug precursors are generally produced from their keto acid counterparts [1]. To date, several methodologies have been developed for chemical synthesis of the α-keto acid esters, including preparation from 2-phenylethylmagnesium bromide and diethyl oxalate in the presence of mild base [2]. However, a more convenient and straightforward synthetic method for the synthesis of 2-oxo-4-phenylbutanoic acid is needed.

In this work, OPBA was prepared to be used as the starting material for producing ACE inhibitor drug precursors. The reaction time, temperature, type of solvent and base used in the synthesis of the title compound were varied, in order to produce OPBA in sufficient yield. The synthesized OPBA was subsequently characterized using NMR. This study also aimed to better understand the relationships between parameters (reaction time, temperature, choice of solvent and base) and the response result (yield); and to determine the optimal conditions for OPBA synthesis using a central-composite rotatable design (CCRD) and response surface methodology (RSM) analysis.

The classical method of optimization for chemical synthesis involves varying one parameter at a time and keeping the others constant. Nevertheless, this method is inefficient as it fails to understand relationships between the variables and the response [3,4]. RSM is an effective statistical technique to quantify the relationships between the response and the input factors in which the objective is to find a desirable location in the design space. The main advantage of RSM is the reduced number of experimental runs needed to provide sufficient information for statistically acceptable result. It is a faster and less expensive method for gathering results than the classical method [5–8], and can be applied for a wide range of chemical reactions involving more than one parameter or response.

2. Materials and methods

2.1. Chemicals

3-Phenylpropionic acid, sulphuric acid, magnesium sulphate, tetrahydrofuran, sodium, benzophenone and hydrochloric acid were purchased from Sigma–Aldrich. Methanol, ethyl acetate, sodium hydrogen carbonate, sodium hydride, calcium chloride anhydrous powder, 2-propanol, dimethyl oxalate, absolute methanol, diethyl ether, ethanol gradient grade, deuterated

---

* Corresponding author. Tel.: +60 4 6533090; fax: +60 4 6584149.
E-mail addresses: chlatif@eng.usm.my, ohpeiching@gmail.com (A.L. Ahmad).

1385-8947/$ – see front matter © 2011 Published by Elsevier B.V.
doi:10.1016/j.cej.2011.04.015

Please cite this article in press as: A.L. Ahmad, et al., Synthesis of 2-oxo-4-phenylbutanoic acid: Parameter optimization using response surface methodology, Chem. Eng. J. (2011), doi:10.1016/j.cej.2011.04.015

Perbadanan Harta Intelek Malaysia
Intellectual Property Corporation of Malaysia
(Diperbadankan)

MyIPO

Aras 27, 29, 30 dan 32, Menara Dayabumi, Jalan Sultan Hishamuddin, 50623 Kuala Lumpur
Tel: 603-2263 2100 Fax: 603-2274 1332 Website: www.myipo.gov.my

---

CERTIFICATE OF FILING

APPLICANT	:	UNIVERSITI SAINS MALAYSIA
APPLICATION NO	:	PI 20092840
REQUEST RECEIVED ON	:	06 July 2009
FILING DATE	:	06 July 2009
AGENT'S/APPLICANT'S FILE REF.	:	RD/P1861/USM/09/SL

Please find attached, a copy of the Request Form relating to the above application, with the filing date and application number marked thereon in accordance with Regulation 25(1).

Date : 06 JULY 2009

(ABDUL RAHMAN RAMLI)
For Registrar of Patents
✉rahman@myipo.gov.my
☎03 – 22632117

To : RAMAKRISHNA A/L DAMODHARAN
KASS INTERNATIONAL SDN BHD,
SUITE 8-7-2, MENARA MUTIARA BANGSAR,
JALAN LIKU, OFF JALAN BANGSAR,
59100 KUALA LUMPUR,
MALAYSIA

FF 09

PATENT COOPERATION TREATY

From the RECEIVING OFFICE

To:

DAMODHARAN , Ramakrishna
KASS INTERNATIONAL SDN. BHD.,
Suite 8-7-2, Menara Mutiara Bangsar,
Jalan Liku, Off Jalan Riong,
Bangsar,
59100 Kuala Lumpur
MALAYSIA

PCT

NOTIFICATION OF THE INTERNATIONAL APPLICATION NUMBER AND OF THE INTERNATIONAL FILING DATE

(PCT Rule 20.2(c))

Date of mailing *(day/month/year)* 15 January 2010 (15.01.2010)

Applicant's or agent's file reference P1861-USM

IMPORTANT NOTIFICATION

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/MY2010/000001	05 January 2010 (05.01.2010)	06 July 2009 (06.07.2009)

Applicant Universiti Sains Malaysia et al

Title of the invention

A SYSTEM FOR PRODUCING L-HOMOPHENYLALANINE AND A PROCESS FOR PRODUCING L-HOMOPHENYLALANINE

1. The applicant is hereby notified that the international application has been accorded the international application number and the international filing date indicated above.

2. The applicant is further notified that the record copy of the international application:

☒ was transmitted to the International Bureau on 15 January 2010 (15.01.2010)

☐ has not yet been transmitted to the International Bureau for the reason indicated below and a copy of this notification has been sent to the International Bureau*:

  ☐ because the necessary national security clearance has not yet been obtained.

  ☐ because *(reason to be specified)*:

* The International Bureau monitors the transmittal of the record copy by the receiving Office and will notify the applicant (with Form PCT/IB/301) of its receipt. Should the record copy not have been received by the expiration of 14 months from the priority date, the International Bureau will notify the applicant (Rule 22.1(c)).

Name and mailing address of the receiving Office
Intellectual Property Corporation of Malaysia
Level 32, Menara Dayabumi, Jalan Sultan Hishamuddin, 50623 Kuala Lumpur MALAYSIA
Facsimile No. 603-22741332

Authorized officer
Mohd Amran Abas
Telephone No. 0322632123

Form PCT/RO/105 (July 2008)

Temporary Save (Not Complete)   Complete (Ready to mail)

MINISTRY OF SCIENCE,
TECHNOLOGY & INNOVATION

ITEX MALAYSIA
Invention - Innovation
Industrial Design - Technology

MINDS

Certificate of Award

This is to certify that

PROF ABDUL LATIF AHMAD, LOW SIEW CHUN, DR SYAMSUL RIZAL ABD SHUKOR

has been awarded the

ITEX SILVER MEDAL

for the invention

STRETCHTECH : NOVEL UNI-AXIAL MEMBRANE STRETCHER FOR MEMBRANE MORPHOLOGY MODIFICATION

at the

18th International Invention, Innovation & Technology Exhibition
ITEX 2007
Kuala Lumpur, Malaysia

held from

18th – 20th May 2007

Tan Sri Datuk Dr Augustine S. H. Ong
President
Malaysian Invention and Design Society

MARS
MALAYSIAN ASSOCIATION OF RESEARCH SCIENTISTS

Gold Medal

This Certificate of Award is presented to

Prof. Abdul Latif Ahmad
Dr. Syamsul Rizal Abdul Shukor
Low Siew Chun
Prof. Asma Ismail

For the invention/innovation of

Innovative and Novel Membrane Morphology Modification

Malaysia Technology Expo 2008
21 - 23 February 2008
Kuala Lumpur

(DATUK DR. DAUD MOHAMAD)
President
Malaysian Association of Research Scientists

(DR. WAN MANSHOL B. W. ZIN)
Chairman
Organizing Committee MTE 2008

MALAYSIA TECHNOLOGY EXPO 2008

DIPLÔME

SALON INTERNATIONAL DES INVENTIONS

GENÈVE

Après examen, le Jury International a décidé

de remettre à: **ABDUL LATIF AHMAD, LOW SIEW CHUN, SYAMSUL RIZAL ABD SHUKOR, ASMA ISMAIL**

pour l'invention: **STRETCHTECH - Modification innovative de la morphologie d'une membrane**

MÉDAILLE DE BRONZE
BRONZE MEDAL
BRONZEMEDAILLE

Genève, le 3 avril 2009

Le Président du Jury: David Taji

Le Président du Salon: Jean-Luc Vincent

MINISTRY OF SCIENCE,
TECHNOLOGY & INNOVATION

ITEX
MALAYSIA
Invention - Innovation
Industrial Design - Technology

MINDS

Certificate of Award

This is to certify that

**PROFESSOR ABDUL LATIF AHMAD, LOW SIEW CHUN,
DR. SYAMSUL RIZAL ABD SHUKOR, DR. OOI BOON SENG,
PROFESSOR ASMA ISMAIL**

has been awarded the

ITEX GOLD MEDAL

for the invention

NCMEM: INDIGENOUS BIOMEDICAL LATERAL FLOW MEMBRANE

at the

**20th International Invention, Innovation & Technology Exhibition
ITEX 2009, Kuala Lumpur, Malaysia**

held from

15th – 17th May 2009

Academician Tan Sri Emeritus Professor
Datuk Dr Augustine S. H. Ong
President
Malaysian Invention and Design Society

pecipta 2009
MALAYSIA
INTERNATIONAL EXPOSITION OF RESEARCH AND INVENTION OF INSTITUTIONS OF HIGHER LEARNING 2009

Jointly Organised By:

MINISTRY OF HIGHER EDUCATION

UNIVERSITY OF MALAYA

BRONZE MEDAL

awarded to

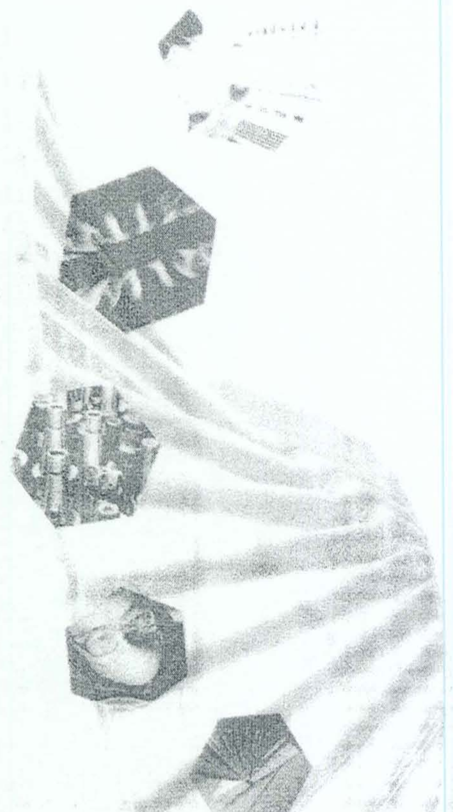
Abdul Latif Ahmad
Syamsul Rizal Abd Shukor
Oh Pei Ching

for the project entitled

Synthesis of L-homophenylalanine As Drug Precursor For Treatment Of Cardiovascular Diseases Via Innovative Integrated MBR

at **the International Exposition of Research and Invention of Institutions of Higher Learning 2009**

held at the **Kuala Lumpur Convention Centre, 8 - 10 October 2009**



PROF. DATO' IR. DR. RADIN UMAR BIN RADIN SOHADI
Director General, Department of Higher Education

GHAUTH JASMON
Vice Chancellor

MARS

MALAYSIAN ASSOCIATION OF RESEARCH SCIENTISTS

Bronze Medal

This Certificate of Award is presented to

Abdul Latif Ahmad
Oh Pei Ching
Syamsul Rizal Abd Shukor

For the invention/innovation of

Cardiovascular Drug Precursor Production via Novel Integrated EMBR

Malaysia Technology Expo 2010
4 - 6 February 2010
Kuala Lumpur

(DATUK DR. DAUD MOHAMAD)
President
Malaysian Association of Research Scientists

(DR. WAN MANSHOL B. W. ZIN)
Chairman
Organizing Committee MTE 2010

MINISTRY OF SCIENCE,
TECHNOLOGY & INNOVATION

ITEX MALAYSIA
INTERNATIONAL
INVENTION, INNOVATION
AND TECHNOLOGY EXHIBITION

MINDS

Certificate of Award

This is to certify that

ABDUL LATIF AHMAD,OH PEI CHING,SYAMSUL RIZAL ABD SHUKOR

has been awarded the

ITEX GOLD MEDAL

for the invention

L-HPA: CARDIO DRUG PRECURSOR

at the

21st International Invention, Innovation & Technology Exhibition
ITEX 2010
Kuala Lumpur, Malaysia

held from

14th – 16th May 2010

Academician Emeritus Professor Tan Sri Datuk
Dr Augustine Ong Soon Hock
President
Malaysian Invention and Design Society

DIPLOMA

TAIPEI INTERNATIONAL INVENTION SHOW & TECHNOMART

INST 2010

RECOGNIZES & HONORS

Inventor

L-HPA: CARDIO DRUG PRECURSOR

Invention

ABDUL LATIF AHMAD, OH PEI CHING, SYAMSUL RIZAL ABD SHUKOR

AS THE WINNER OF INST INVENTION AWARD

FOR

EXCELLENCE IN INVENTION & INNOVATION

ORGANIZER OF INST 2010
KUALA LUMPUR, MALAYSIA, MAY 15, 2010

URKUNDE/CERTIFICATE

INTERNATIONALE FACHMESSE

»IDEEN - ERFINDUNGEN - NEUHEITEN«

NÜRNBERG

INTERNATIONAL TRADE FAIR

»IDEAS-INVENTIONS-NEW PRODUCTS«

NUREMBERG

Erfindung Universität / Hochschule

Prof. Abdul Latif Ahmad; Dr. Syamsul Rizal Abd Shukor;Dr. Leo Choe Peng
Universiti Sains Malaysia
11800 USM, PENANG

MALAYSIA

wurde für hervorragende Leistungen eine
Goldmedaille
verliehen.

Erfindung/ Neuheit
γ-Aluminiumoxid mit bimodaler Porengrößenverteilung: Transportwiderstand-Minderung in zweischichtigen Membranen
γ-Alumina with bimodal pore size distribution: transport resistance diminution in bi-layered membrane

International Jury of iENA 2009

Kom. Rat Volkwin Hoffelner
Vorsitzender der Jury/Chairman of the jury

7. November 2009

International Exposition of Research and Inventions of Institutions of Higher Learning 2007
10 - 12 August 2007 | Kuala Lumpur Convention Centre (KLCC)

pecipta 07
MALAYSIA
EKSPO PENYELIDIKAN DAN CIPTAAN INSTITUSI PENGAJIAN TINGGI ANTARABANGSA 2007
INTERNATIONAL EXPOSITION OF RESEARCH AND INVENTIONS OF INSTITUTIONS OF HIGHER LEARNING 2007

Certificate of Award

GOLD

EXHIBIT

Y-Alumina With Bimodal Pore Size Distribution: Transport Resistance Diminution in Bi-Layered Membrance

EXHIBITOR(S)

Prof. Abdul Latif Ahmad, Dr. Shamsul Rizal Abdul Shukor, Leo Choe Peng

DATO' MUSTAPA MOHAMED
MINISTER OF HIGHER EDUCATION
MALAYSIA

PROFESSOR DATO' DZULKIFLI ABDUL RAZAK
CHAIRMAN
ORGANISING COMMITTEE

Jointly Organised by:

KEMENTERIAN PENGAJIAN TINGGI

USM
UNIVERSITI SAINS MALAYSIA

12th Asia Pacific Confederation of Chemical Engineering Congress
August 3-6, 2008
Dalian, China
http://apcche2008.ecust.edu.cn/
E-mail: apcche2008@ecust.edu.cn

To Sunarti Abd Rahman
School of Chemical Engineering, Universiti Sains Malaysia
Malaysia

Date: June 8, 2008

Dear Sir or Madam,

It is our pleasure to invite you to participate in the 12th Asia Pacific Confederation of Chemical Engineering Congress (APCChE 2008) which will be held in Dalian, China from August 3rd to 6th, 2008.

Your contribution entitled "Review of liquid absorbent behaviours in membrane gas absorption process" has been accepted as oral /poster presentation.

Yours sincerely,

Qian Xuhong

QIAN, Xuhong, Ph.D.
President of Asian Pacific Confederation of Chemical Engineering
President of East China University of Science and Technology,
Professor of Bioorganic Chemistry and Engineering
130 Meilong Road, Shanghai 200237, China
Tel/Fax: +86-21-64252390
xhqian_apcche@ecust.edu.cn

Review of liquid absorbent behaviours in membrane gas absorption process

ABDUL LATIF AHMAD[a,*], SUNARTI BINTI ABD RAHMAN[a,b], LEE KEAT TEONG[a] AND W JAMES NOEL FERNANDO[a]
[a]School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, 14300, Nibong Tebal, Seberang Perai Selatan, Pulau Pinang. Malaysia.
[b]Faculty of Chemical Engineering and Natural Resources, Universiti Malaysia Pahang, 25000 Kuantan, Pahang, Malaysia.
*Corresponding Author: Tel: +60 (4) 593-7788; Fax: +60 (4) 594-1013;
Email: chlatif@eng.usm.my

ABSTRACT

Emission of CO_2 becomes a major problem to all people everywhere and a famous debate title among scientists and politicians worldwide. Uncontrollable of CO_2 emission will be caused the climate change or global warming. Recently, CO_2 removal from flue gases of power plant using membrane gas absorption (MGA) technology has attracted many researchers throughout the world. The advantages of MGA are offer higher performance compared the conventional approaches, environmental friendly, easy to handle as well as low cost of maintenances. In this paper, criteria, type, scenario and trends of liquid absorbents for MGA will be reviewed. Strategies on further development of this process are also proposed.

Keywords: carbon dioxide, membrane gas absorption, liquid absorbents, flue gas

1. Introduction

Although membrane technology is new, the scientific discovery of membrane phenomena can be traced back to the eighteen century where Abbe Nollet (Mulder, 1996) invented the word of osmosis to describe water permeation through a diaphragm in 1748. Although this technology emerged a long time ago, technical viability has only occurred over the last 40 years. The foundation for gas separation was laid by Thomas Graham in 1829, which performed the first recorded experiment on the transport of gases and vapours in polymeric membranes (Pandey and Chauhan, 2001). However, the academic studies and industrial researches of membrane gas absorption (MGA) have started even as early as 1980's. The MGA's concept was commercialized in late 1980's with the scale up of liquid/ liquid membrane contactor using microporous polypropylene membrane capillary membrane modules to separate of water from salt solutions. However, in term of energy savings, liquid/ liquid membrane contactor does not

Thin Film Composite (TFC) Membrane in Membrane Gas Absorption System

ABDUL LATIF AHMAD[a], SUNARTI BINTI ABD RAHMAN[a,b*], LEE KEAT TEONG[a] AND
W JAMES NOEL FERNANDO[a]
[a]School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, 14300,
Nibong Tebal, Seberang Perai Selatan, Pulau Pinang. Malaysia.
[b]Faculty of Chemical Engineering and Natural Resources, Universiti Malaysia Pahang, 25000
Kuantan, Pahang, Malaysia.
*Corresponding Author: Tel: +60 (9) 5492388; Fax: +60 (9) 5492399;
Email: sunarti@ump.edu.my

ABSTRACT

Flue gas is a gas is released to the atmosphere via a flue and quite often refers to the combustion exhaust gas produced at power plants. Its composition generally depends on what is being burned, but usually consist of mostly carbon dioxide (CO_2) and water vapor as well as nitrogen (N_2) and excess oxygen. Lately, the removal of CO_2 from flue gases has received intense attention due to the properties of CO_2. It is widely known that CO_2 is a one of the major greenhouses gases. Membrane gas absorption (MGA) is one technology used to remove the CO_2. The present research focuses on the separation of CO_2 from N_2 using a thin film composite (TFC) membrane based on glutaraldehyde in MGA system. The THC membrane with glutaraldehyde as the selective layer on the top of different microfiltration supports with varying properties based on polypropylene (PP) and polyvinylidene fluoride (PVDF) were prepared. The effects of crucial parameters that effected TFC membrane performance such as coating solution concentration, membrane support porosity and nature of materials were investigated. Deionized water was employed as the liquid absorbent. The TFC membrane in combination with deionized water as an absorbent liquid was found to be a suitable for bulk CO_2 removal using MGA system.

Keywords: carbon dioxide, thin film composite membrane, membrane gas absorption

1. Introduction

Hydrophobic microporous membranes have attracted increasing attention in recent years for use as contactors in chemical absorption to remove CO_2, H_2S, and SO_2 from flue and natural gases [1,2]. Although certain downside such as the addition of an extra mass-transfer resistance caused by the membrane, the microporous membrane gas absorption offer a lot of advantages including individual gas and liquid flow channels and operational flexibility to scale up or down. Among various hydrophobic polymers, polypropylene (PP) is the most popular membrane materials. However, since PP membranes are usually produced by stretching or thermal methods, the relatively low porosity of PP membranes restricts a significant increase on absorption flux. Although PP is excluded on account of their insolubility in common liquid absorbents, polyvinylidene fluoride (PVDF), another strong hydrophobic polymer with excellent chemical and thermal resistances, has been successfully employed for membrane gas absorption. In this

6^{th} Regional Symposium on Membr [illegible] & Technology
FOOD ENERGY AND ENVIRONMENT
Patong, Phuket, Thailand, 13 [illegible] August
2008

Preparation and Characterization of Various Membrane Morphologies for Lateral Flow Immunoassay Development

Ahmad A. L. [a*], **Low S. C.** [a], **Abd Shukor S. R.** [a] **and Ismail A.**[b]

[a] *School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, S.P.S, Penang, Malaysia*
Tel 604-5941012, Fax: 6045941013, Email: chlatif@eng.usm.my, siewchun@gmail.com, chsyamrizal@eng.usm.my
[b] *Institute for Research in Molecular Medicine, Health Campus, Universiti Sains Malaysia, 16150 Kubang Kerian, Kelantan, Malaysia.*
Tel 609-7663000, Fax 609-7657267, Email: asma@kb.usm.my

* *Corresponding Author:* chlatif@eng.usm.my

Abstract

Lateral flow in nitrocellulose membrane is widely used as the transport medium in immunological test strips. The immunoassay performance depends greatly on the binding mechanism on membrane surface. If the membrane structures could be controlled precisely, various kinds of immunological analysis could be performed effectively and accurately. This study is aimed at a quantitative understanding on (1) the sensitivity of protein quantification on membrane surface by using densitometer and (2) comparison of the protein binding ability on different membrane surface pore structures. Concentration of immobilised protein on membrane surface was measured as a function of colour intensity, which is initially stained with Ponceau S. The findings showed that membrane morphologies significantly affect the sensitivity of membrane protein binding ability. Depending on the membrane pore structures, different red spot sharpness and intensities were observed from the stained protein dot. Membrane with smaller pore creates higher protein immobilization density and thus, increases the sensitivity level of the assay. However, larger membrane pore structure is necessary to induce rapid migration rate for faster assay.

Keywords: Nitrocellulose Membrane; Morphology; Immunoassay; Microstructure; Pore size.

Introduction

Lateral flow immunoassays have been widely introduced in the field of food science and healthcare [1, 2], due to their sensitivity, rapid testing, inexpensive manufacturing cost and easy to operate procedure [3]. A large number of testings are available for the detection of certain infectious diseases [4], food hygiene [5, 6], fertility test and etc. This will then translate to swift detection of hazardous substance in food prior to human consumption, which then helps to save lives.

Both immunoassay performance and its effectiveness depend greatly on the transport rate of aqueous solution through membrane pores and the binding mechanism on membrane surface [7]. If the membrane surface and internal layer structures could be controlled precisely, various kinds of immunological analysis could be performed effectively and accurately [8]. Thus, membrane plays an important role on the efficiency of a diagnostic test strip.

Development of Lateral Flow Membrane for Immunoassay Separation

Ahmad A.L. [a*], Low S.C. [a], Shukor S.R.Abd [a], Ismail A. [b], Sunarti A.R. [a]

[a] School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia,
Seri Ampangan, 14300 Nibong Tebal, S.P.S, Penang, Malaysia
[b] Institute for Research in Molecular Medicine, Health Campus, Universiti Sains Malaysia, 16150 Kubang Kerian, Kelantan, Malaysia.

* Corresponding Author: Tel: +6-04-5941012, Fax: +6-04-5941013
E-mail: chlatif@eng.usm.my

Abstract

Lateral flow nitrocellulose membrane is one of the commonly used separation medium for bacteria detection in drinking water treatment facility. In order to enhance its performance, control of membrane surface and cross-section morphology is primarily important. The challenge of this study is to combine various formulation and casting variables to obtain lateral flow nitrocellulose membrane with desired morphologies. Through dry phase inversion method, the drying temperature was found to be an imperative parameter in synthesizing membrane as it will affect the pore structures. High drying temperature causes agglomeration of polymer matrix and thus closed pore was observed on the membrane surface. This further decreases the membrane lateral liquid migration rate, beside reduces membrane binding ability for bacteria detection. Results show that by decreasing polymer concentration, membrane surface pore became apparently bigger and this creates faster lateral migration speed of water solution. Bigger pore size increases the chance for bacteria detecting agent to bind onto the pore layers, which ultimately enhance the bacteria detection ability of the device.

Key words: Membrane; Morphology; Microfiltration; Binding Ability; Lateral Flow

1. Introduction

Viruses and bacteria detection in drinking water has brought water quality analysis into a whole new perspective due to waterborne outbreaks [1] such as typhoid fewer [2], *Escherichia coli O157* [3], *Giardia and Cryptosporidium* [4]. These typical waterborne viruses, even in very low concentration, may be lethal [4]. Hence, there is an urgent need to detect the presence of viruses and bacteria in water by a sensitive, reliable and efficient method.

Membrane separation technologies such as ultrafiltration (UF), microfiltration (MF) and reverse osmosis (RO) are the known as effective ways to detect low concentration of viruses in wastewater treatment processes for the past decades [4]. A simple and rapid one step membrane test trip was developed to detect the presence of

---

Enhanced Biotransformation and Removal of Biocatalytic Compounds in Water by Membrane Bioreactor Technology

Ahmad A.L.*, Oh P.C., Uzir M.H., Shukor S.R.Abd., Derek C.J.C. and Low S.C.

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia,
14300 Nibong Tebal, Penang, Malaysia
Tel 604-5941012, Fax 604-5941013,
Email chlatif@eng.usm.my, ohpeiching@gmail.com, chhekarl@eng.usm.my, ddeanasd@eng.usm.my

**Corresponding Author:* chlatif@eng.usm.my

Abstract

Membrane technology has advanced expeditiously in the past decade for water purification, water treatment, desalination, pharmaceutical industry and many other purposes. In recent years, several pharmaceuticals and biocatalytic compounds which are believed likely to induce adverse health effects on aquatic organisms are detected in surface waters in several countries. Membrane-based systems have since then become the opted technology for removal of these compounds in water, both on laboratory and industrial scales. Herein we evaluate the production of vital pharmaceutical drug precursors via biotransformation, simultaneously reviewing on the prospect in utilization of membrane bioreactor for biocatalytic compounds separation and their removal from water.

Keywords: Membrane Bioreactor; Water; Biocatalytic Compounds; Biotransformation

Use of AMP as Liquid Absorbent for the removal of CO_2 from Flue Gas using Polymeric Flat Sheet Membrane Contactor

Ahmad A.L *[a], Sunarti A.R [a,b], Lee K.T [a] and Fernando W.J.N[a]
[a] *School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Pulau Pinang, Malaysia.*
Tel +60 (4) 594-1012, Fax +60 (4) 594-1013, Email chlatif@eng.usm.my, sunartirahman@gmail.com, leekeatteong@gmail.com, fernwn@yahoo.com
[b] *Faculty of Chemical Engineering and Natural Resources, Universiti Malaysia Pahang, 25000 Kuantan, Pahang, Malaysia.*
Tel.: +60 (9) 549-2404, Fax: +60 (9) 549-2399, Email sunartirahman@gmail.com

**Corresponding Author: chlatif@eng.usm.my*

Abstract

Membrane contactor applications that have been developed can be found in both water and gas treatment. The objective of this study was to investigate the potential process of the removal of carbon dioxide (CO_2) from flue gas by using developed membrane contactor system. The experiments were performed in membrane contactor system consisted microporous polyvinylidenefluoride (PVDF) flat sheet membrane with 0.1 and 0.45 μm pore size respectively. 2-Amino-2-methyl-1-propanol (AMP) solution was employed as the liquid absorbent. The operating parameters such as the gas and liquid flow rate were setup at 100 cm^3/min and 25 cm^3/min and liquid absorbent was at room temperature. The effect of AMP concentration was studied with variation in range 1M to 5M. In addition, the experiments were carried out with 10%, 20%, 30% and 40% gas ratio CO_2 to N_2. The results suggest that hydrophobic polymeric membranes such as PVDF can be efficiently used for gas absorption with optimized concentration of AMP.

Keywords: Membrane contactor, CO_2 removal, liquid absorbent, polymeric membrane

1. Introduction

Natural gas, refinery gas or coal gas generates huge quantities of CO_2, when it is employed for industrial and domestic heating and other chemical processes. Emission of CO_2, from these sources is regarded as the most serious potential cause of environmental problems such as global warming and acid rain. In order to utilize these flue gases for chemical processing or energy generation, elimination or limitation of emissions of CO_2 and other greenhouse gases are the current main objectives for all the environmental organizations and establishments in the world.

Recently, membrane contactor processes of CO_2 separation have been developed because of the drawbacks of conventional CO_2 separation processes based on the application of packed towers, spray towers, and bubble columns. In particular, processes employing the use of membrane-based contacting devices have been found promising. In membrane contactor system, the fluids to be contacted flow on the opposite side of the membrane, and the gas–liquid interface forms at the mouth of each membrane pore. The available contact area remains undisturbed even at a high or low flow rate because the two fluid flows are independent. Because the absorbent liquid flows on one side and the gas flows on the other side of the membrane, this method is useful in applications where the required solvent/feed ratio is very high or very low [1]. This type of process offers several practical advantages including high surface area per unit contact volume, independent control of gas and liquid flow rates, compactness, a priori known gas–liquid interfacial area, easiness in scale up, and others. Therefore, in spite of certain disadvantages arising from membrane fouling, membrane

OPTIMIZATION OF *P*-XYLENE SEPARATION FROM TERNARY XYLENE MIXTURE OVER SILICALITE-1 MEMBRANE USING RESPONSE SURFACE METHODOLOGY

Yeong Y. F., Abdullah A. Z., Ahmad A. L. and Bhatia S. *

School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia,
Seri Ampangan, 14300 Nibong Tebal, Seberang Perai Selatan, Pulau Pinang,
Malaysia.

**Corresponding Author: chbhatia@eng.usm.my*

Abstract

The separation of *p*-xylene (pX) from its isomers *m*-xylene (mX) and *o*-xylene (oX) was studied using a silicalite-1 membrane supported on α-alumina. The design of experiment (DoE) is applied to investigate the effect of two process variables: temperature and *p*-xylene feed partial pressure on the separation performance of membrane. The response surface methodology (RSM) coupled with central composite design (CCD) was used to develop five models to correlate the two process variables to the five responses: pX flux, oX flux, mX flux, *p-/o*-xylene (pX/oX) separation factor and *p-/m*-xylene (pX/mX) separation factor. The optimum pX flux of 5.94 x 10^{-6} mol/m^2.s, pX/oX separation factor of 19 and pX/mX separation factor of 20 were obtained at temperature of 198 °C and pX feed partial pressure of 0.22 kPa. The experimental data compared with the simulated results obtained from the model within an average error of ± 2.90 %. In the present work, the DoE provide better flexibility of the process studies as well as to practical considerations of the membrane process for ternary xylene mixture separation.

Keywords: *silicalite-1 membrane, separation, ternary xylene mixture, design of experiment.*

Introduction

Separation of para-xylene (pX) from its isomers is an important operation in the petrochemical industry, but it is difficult to separate because of their boiling points (para-xylene (pX): 138 °C, meta-xylene (mX): 139 °C and o-xylene (oX): 144 °C) are close [1]. pX is the feed for the pure terephthalic acid (PTA) production, which in turn is used in the production of polyester resin and fibers. Therefore, it has the largest commercial market among the three isomers.

In recent years, zeolite membranes have been in focus because of their characteristics such as well defined micropore structure, good thermal and structural stability. The characteristics of zeolite membrane have found its new application in gas, vapor and liquid separation especially in petrochemical industry based on their properties adsorption, preferential diffusion, or molecular sieving (size exclusion) [2-4]. One particular process in which zeolite membrane might offer significant advantage compare to existing technology is the separation of close-boiling point hydrocarbons, i.e. of xylene isomers, which is difficult to separate by distillation or other complex and energy-intensive

SAPO-34 MEMBRANE FOR CO_2 SEPARATION FROM GASEOUS MIXTURES

T.L. Chew [a], A.L. Ahmad [a]. and S. Bhatia *[a]

[a] *School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, Pulau Pinang, Malaysia*

**Corresponding Author: chbhatia@eng.usm.my*

Abstract

Silicoaluminophosphate (SAPO-34) membranes were synthesized onto alumina support. The performance of these membranes was tested in terms of single gas permeation and binary gas separation of CO_2/N_2 gaseous mixtures. The separation selectivities were studied in variation with temperature (302 – 423 K) and partial pressure drop (1-5 bars). CO_2/N_2 separation selectivites of 4.14 was obtained at temperature of 302 K and partial pressure drop of 1 bar.

Keywords: *Zeolite membrane, SAPO-34, gas separation, carbon dioxide*

Introduction

CO_2 separation plays an important role, like in the natural gas and petrochemical industries. Several conventional technologies are used for gas separation such as cryogenic distillation, amine absorption (CO_2 separation) and pressure swing adsorption (PSA) [1]. However, these conventional technologies incur high capital cost due to the installation and operation of the units. As energy costs rise, there is increasing interest in developing membrane technology for separating gases in order to reduce the environmental impact and costs of industrial processes. Membrane separation processes are more energy efficient and easier to be operated compared to the conventional separation unit [2].

Polymeric membranes are widely used in commercial application of the membrane area due to its low energy cost. However, the application of polymeric membranes is limited to its loss in performance stability at high temperature, high pressure and in the highly acidic or alkaline environment [3]. Zeolite membranes are the microporous inorganic membranes which are gaining increasing interest among the researcher as alternative candidates for gas separations, in view of their higher thermal, mechanical and chemical stability compared to organic membranes [4].

SAPO-34, which is a microporous and crystalline zeolite, has attracted much attention in catalytic and gas separation application due to its small pore size, medium acidic strength and high thermal stability [5,6]. SAPO-34 has the chabazite framework with pore diameter of 0.38 nm [7], which is close to the sizes of CH_4 and N_2. Hence, it would be easy to separate CO_2 from CH_4 and N_2 based on molecular sieving. In the studies by Li et al. [6] and Poshusta et al. [8], smaller size of CO_2 and its higher adsorption strength relative to CH_4 and N_2 on SAPO-34 favored the permeation of CO_2. Combination of differences in diffusivity and competitive adsorption is the promising key for SAPO-34 to separate CO_2/CH_4 and CO_2/N_2 binary mixtures.

SYNTHESIS OF PALLADIUM/TITANIA MEMBRANE BY SOL–GEL METHOD

A.L. Ahmad [*a], M.A.T. Jaya [a]. and C.J.C. Derek[a]

[a] School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, S.P.S, Penang, Malaysia, e-mail: chlatif@eng.usm.my, azan85@gmail.com, chderek@eng.usm.my.

*Corresponding Author: chlatif@eng.usm.my

Abstract

A micro thin-layer palladium-doped titania (TiO_2) membrane on titania support is synthesized using sol-gel and sol-solution mixing method. Standard characterizations such as the membrane material phase, morphology, and particle size are studied via Transmission Electron Microscope (TEM), Scanning Electron Microscope (SEM), and X-ray diffraction (XRD). The titania sol prepared with certain amount of organic additives content (Hydroxypropyl Cellulose and Polyvinyl Alcohol) and palladium chloride as palladium precursor has been calcined at various temperature to study the corresponding crystal phase. The particle shape of the calcined TiO_2 is spheroidal. Based on TEM image, the particle size is divided into small (10-15nm) and larger (100-200nm) size. The growth of crystallite size is highly sensitive to the amount of palladium element content. Small amount of palladium element content has favored the anatase-rutile phase transformation but high amount significantly suppressed the phase transformation and crystallite size growth. The corresponding membrane layer thickness is less than 1μm with high homogeneity, crack-free and no pinholes.

Keywords: *ceramic membrane, titania, hydrogen separation, palladium, synthesis*

Introduction

Conventional fuel was the most used energy source today. However, it can be highly expensive depending on the economic situation. Moreover, the non-renewability of it has motivated people to look into other alternative energies like water, wind, hydrogen and etc. Hydrogen is highly abundant, cheap, and considered as clean energy, thus, very promising to replace the conventional fuel in the future [9]. For decades, many efforts relating to H_2 production, separation, purification, storage, transportation, fuel cell process, alternative energy have been studied and advanced [21].

Amongst applications in gas separation most investigated is hydrogen separation ([6],[18],[10]). The potential to become reliable alternative energy shows positive development. Due to mechanical strength, thermal and chemical stability and resistance to microbial degradation, ceramic membrane have been extensively studied for industrial application [16]. Basically, materials to develop membrane for hydrogen separation are polymer, ceramic, and metal. Polymer membrane has high selectivity but unable to endure intense environment. Even though, ceramic membrane gives advantage in terms of physical and chemical stability, it has lower selectivity. Metal membrane, particularly palladium is

PREPARATION OF PEROVSKITE TITANIA CERAMIC MEMBRANE BY SOL–GEL METHOD

A.L. Ahmad *[a], N.A. Abdullah Sani [a]. and S.H.S. Zein [a]

[a] School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, Seri Ampangan, 14300 Nibong Tebal, S.P.S, Penang, Malaysia, e-mail: chlatif@eng.usm.my, nuraimie@gmail.com, chhussein@eng.usm.my.

*Corresponding Author: chlatif@eng.usm.my

Abstract

Ceramic membranes have been applied in many industrial processes mainly for oxygen separation, partial oxidation of hydrocarbons and waste reduction recovery. They have several advantages over the polymeric membranes such as better chemical and thermal stability, longer life, and better defouling properties. These properties have made the ceramic membranes desirable for use in oxidative coupling of methane (OCM) process, partial oxidation of methane (POM) process and fuel cell system. This paper reports a preliminary work for a preparation of perovskite titania ceramic membrane in order to separate oxygen from gas mixtures containing oxygen. A titania disk were prepared using titania powder and used as a membrane support. The membrane was prepared by sol–gel method using titania as a precursor mixed with water and perovskite to form a sol. Then the support were coated with the sol by spin-coating, dried and calcined at different temperature. Transmission electron microscopy (TEM) analysis showed that salts solution homogeneously dispersed in the titania sol and smaller particle size (3.46 nm) was obtained as compared to the titania sol (5.40 nm). Scanning electron microscopy (SEM) analysis confirmed that average particle diameter of perovskite titania membrane increased with increasing calcinations temperature and X-ray diffractometry (XRD) analysis showed that the temperature around 800 °C was suitable to achieve high crystalline structure of perovskite titania membrane containing more anatase phase with less rutile phase. Thermogravimetric analysis (TGA) showed that the volatile materials, the alcohol and the organic additives in uncalcined perovskite titania gel were fully burned out below 650 °C and its total weight loss is 7.1 %. The results from SEM, XRD, and TGA confirmed the formation of perovskite titania membrane.

Keywords: *perovskite, titania membrane, sol–gel method, calcination, morphology.*

1. Introduction

Membranes play an important role in the separation technology, because membrane based separation offers advantageous in terms of low energy use and more economically competitive compared to other conventional separation technologies such as distillation, absorption, adsorption, solvent extraction or cryogenics [1]. Among various types of membranes, ceramic membranes are widely used due to their unique features, such as high temperature stability, high pressure resistance, good chemical stability, high mechanical resistance, long life and good anti-fouling [2]. Nowadays, there are many researchers of developing ceramic membranes for oxygen separation. Applications of oxygen separation including uses of oxygen for oxidative coupling of methane (OCM) [3], partial oxidation of methane (POM) process [4] and fuel cell system [5].

Microwave Heating for Rapid Synthesis of SAPO-34 Zeolites

T. L. Chew, A. L. Ahmad, S. Bhatia
School of Chemical Engineering, Engineering Campus,
Universiti Sains Malaysia,
Seri Ampangan, 14300 Nibong Tebal,
Pulau Pinang, Malaysia.

Abstract- **Microwave (MW) heating was introduced for the crystallization of SAPO-34 zeolites using a colloidal solution containing tetraethylammonium hydroxide as a template. The effects of MW power and synthesis time on the formation of SAPO-34 zeolites were studied and optimized in view of the zeolite properties such as morphology and crystallinity. MW heating was compared with the conventional hydrothermal synthesis for the formation of SAPO-34 zeolites. MW heating formed SAPO-34 zeolites with narrower size distribution within significantly shortened time compared to hydrothermal synthesis. Full crystallization of SAPO-34 zeolites was achieved in only 10 hours by microwave heating instead of 24 hours required by conventional hydrothermal synthesis at 180 °C.**

Keywords- Microwave; hydrothermal synthesis; SAPO-34; zeolites.

I. INTRODUCTION

Silicoaluminophosphates (SAPOs) is microporous and crystalline zeolites that gain increasing attention in catalytic, adsorption and separation application due to their unique characteristics such as well-defined pore structure, surface properties, high thermal and chemical stability [1-3]. SAPO-34 with its framework analog to chabazite and its pore diameter of 3.8 Å [4], is a potential candidate as catalyst for petrochemical industry (i.e. methanol-to-olefin reaction) [5] and as adsorbent for gas separation (i.e. carbon dioxide removal from gas mixtures) [2].

There are several methods for zeolite synthesis. Conventional hydrothermal synthesis (HS) is the common method for synthesis of zeolites where a precursor solution is subjected to heating at required temperature and time under desired pressure. So far, conventional HS has been wise reported for synthesis and studies of SAPO-34 zeolites [1,5,6]. However, the long synthesis time of conventional HS has limited its industrial application. In addition, HS generally form zeolites with broad size distribution. Microwave (MW) heating appears to be an alternative method for significantly reduced synthesis time. MW heating offers advantages such as (1) volumetric and instantaneous heating, (2) higher heating rate compared to those of conventional hydrothermal synthesis, (3) introduction of selective heating and (4) development of molecular sieves with narrower size distribution [5,7]. By incorporating MW radiation to the heating of precursor solution, the formation of high quality and purity zeolites can be achieved within much short time [7].

Venna and Carreon [3] reported the phase transformation of SAPO-5 to SAPO-34 under microwave heating for 500 minutes at 150 °C. However, the crystallization of SAPO-34 zeolite using MW heating is still in early stage of development. It is important to elucidate the factors such as MW power, synthesis temperature and time toward the formation of high purity SAPO-34. Present study reports the rapid formation of high purity SAPO-34 zeolite crystal under MW heating. Emphasis is put on study of the effect of MW power and synthesis time on the growth of SAPO-34 zeolite crystals. MW heating will be compared with conventional hydrothermal synthesis (HS) for the properties of SAPO-34 zeolite crystals synthesized.

II. EXPERIMENTAL

A. Synthesis of SAPO-34 Zeolites

The synthesis precursor was prepared by firstly mixing and stirring deionized water (H_2O), aluminium isopropoxide (>98%, Merck) and phosphorus acid (85 %, Sigma-Aldrich) at room temperature for 12 hours. Tetraethylammonium hydroxide (TEAOH, 35 wt%, Sigma-Aldrich) and Ludox AS-40 colloidal silica sol (40 wt%) were added and the mixture was stirred at room temperature for 24 hours. The final mixture has the molar composition of Al_2O_3:P_2O_5:1.2TEAOH: 0.3SiO_2:55H_2O.

The synthesis precursor was poured into a Teflon-lined vessel and was subjected to microwave (MW) heating using a fabricated MW kitchen oven (Hanabishi, maximum power output 900W). The MW kitchen oven was fabricated so that the MW power and temperature can be controlled simultaneously for heating of the synthesis precursor. The synthesis time and MW power in terms of percentage (%) were varied for the synthesis of SAPO-34 zeolite. For comparison, hydrothermal synthesis (HS) was performed at 180 °C for 24 hours using Teflon-lined stainless steel autoclave. The suspensions from the Teflon-lined vessel after the MW and HS were centrifuged (3000 r.p.m. for 8 minutes) and rinsed with deionised water to obtain the SAPO-34 zeolite crystals. The precipitate was dried overnight in oven.

MODELING OF TRICHLOROETHYLENE DEGRADATION WITH IRON, IRON-REDUCING BACTERIA, AND SUBSTRATE

YEUNOOK BAE[1], * JAE-WOO PARK[1]
[1]Department of Civil Engineering, Hanyang University, 17 Haengdang, Seoungdong, Seoul, Korea
E-mail: jaewoopark@hanyang.ac.kr

ABSTRACT

Trichloroethylene (TCE) removal by permeable-reactive barriers (PRBs) using Zero-valent iron (ZVI) medium have been widely researched, and the effect of iron-reducing bacteria (IRB) has also increasingly researched for enhancing TCE degradation longevity. However, little researches were performed about substrate, which is significant in IRB growth. Present study developed a modeling for an effect of substrate as a first step to address this need. The model was based on transport reaction model including TCE degradation by ZVI and ferrous iron, ferrous iron production by IRB, and IRB growth by substrate. Each injecting TCE concentration and total amount of iron per unit volume was determined 10mg/L and 1061.1mg/L. And each initial IRB concentration and substrate injection concentration was determined from 100mg/L to 1,000mg/L and from 1mg/L to 10mg/L. The model result shows TCE degradation rates did not increase more than approximately 6.1mg/L in all substrate condition. This is because total iron amount per unit volume was limited to 1061.1mg/L so ferrous iron could no produced more than this figure. These results indicate that substrate is required only limited amount to obtain the maximum degradation rate of TCE when the total iron amount in the certain reactor system is limited. This model could offer a framework for determining the optical amount of substrate for treating TCE in the PRBs or other reactor system using iron.

Keywords: Modeling, Trichloroethylene (TCE), Substrate, Iron, Iron-reducing bacteria (IRB)

APPLICATION OF CARBON NANOTUBES IN ENVIRONMENTAL ENGINEERING

YIT THAI ONG, ABDUL LATIF AHMAD, SHARIF HUSSEIN SHARIF ZEIN and SOON HUAT TAN*
School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia
Seri Ampangan, 14300 Nibong Tebal, Pulau Pinang, Malaysia
Email : chshtan@eng.usm.my

ABSTRACT

Many potential applications of carbon nanotubes (CNTs) have been proposed including high-strength composites, field emission devices, sensors, probes, hydrogen storage, biomedicals, adsorbents and etc. The recent study on the application of CNTs in environmental will be presented in this short review. CNTs with excellent mechanical, electronic and chemical properties showed great potential application for environment protection. CNTs with an oxidized treatment surfaces, have been proposed as superior sorbent with an exceptional adsorption capability and high adsorption efficiency to remove heavy metal, dioxin and 1, 2-dichlorobenzene from aqueous solution. Some key results relating to the CNTs characteristics such as surface area, pore size distribution, sorbent mass, pH of the sorbate solution, recovery of metal ions and regeneration of CNTs will also be summarized in this short review.

Keywords: Carbon nanotubes; Environmental; Adsorbent