Data Set

The Utilization of Micro-Mesoporous Carbon-Based Filler in the P84 Hollow Fiber Membrane for Gas Separation

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Table S1 Mixed gas performance of the MMMs. Number in parentheses corresponds to the performance increase (+) or decrease (-) in percent

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Membrane** | **Binary Gas (Barrer)** | | | | **Selectivity** | |
| CO2/CH4 | | H2/CH4 | | CO2/CH4 | H2/CH4 |
| CO2 | CH4 | H2 | CH4 |
| Neat | 9.09±0.01  (-32.09) | 1.15±0.01  (-25.92) | 19.92±0.31  (-12.31) | 1.43±0.07  (-7.89) | 7.90 ± 0.53  (-8.60) | 13.93 ± 0.18  (-4.97) |
| P84/ZTC 0.5 | 11.42±0.11  (-23.80) | 1.51±0.00  (-22.27) | 28.96±0.21  (-18.30) | 1.68±0.01  (-13.51) | 7.56 ± 0.16  (-2.02) | 17.24 ± 0.50  (-5.58) |
| P84/ZTC 1 | 18.67±0.20  (-23.89) | 1.77±0.03  (-21.59) | 36.61±0.01  (-22.83) | 1.85±0.02  (-18.04) | 10.55 ± 0.49  (-2.94) | 19.79 ± 0.74  (-5.84) |
| P84/ZTC 1.5 | 13.92±0.06  (-20.68) | 1.91±0.01  (-16.80) | 21.28±0.04  (-39.06) | 1.83±0.08  (-20.29) | 7.29 ± 0.12  (-4.76) | 11.63 ± 0.37  (-23.54) |

The Data for Robeson Upper Bound 1991 and 2008 was available elsewhere [1,2].

Table S2 CO2/N2 Reference

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Membrane** | **Filler Loading** | ***P*CO2 (Barrer)** | **αCO2/N2** | **Ref** |
| PES | 0 | 0.12 | 22.5 | [3] |
| PES/MWCNTs | 5 wt% | 0.2 | 22.5 |
| PES/CNT | 5 wt% | 0.2 | 22.1 | [4] |
| PES/CNT-Ru Metal | 5 wt% | 0.13 | 26.5 |
| P84 | 0 | 0.89 | 20.23 | [5] |
| P84/SiO2 | 4 wt% | 0.92 | 19.57 |
| BPPO | 0 | 2.6 | 30 | [6] |
| BPPO/MWCNTs | 5 wt% | 4.7 | 31 |
| BPPO/SWCNTs | 5 wt% | 2.6 | 30 |
| SEBS/ZIF-8 (S) |  | 14.71 | 10.6 | [7] |
| SEBS/ZIF-8 (M) |  | 15.23 | 12 |
| Matrimid/CSM-18.4 |  | 1.32 | 38.1 | [8] |
| Matrimid | 0 | 9.6 | 30 | [9] |
| Matrimid/PIM-1 Blend | 10 wt% PIM-1 | 17 | 30 |
| P84 | 0 | 3.05 | 13.6 | [10] |
| P84 anne 180°C |  | 1.09 | 9.5 |
| P84 anne 200°C |  | 0.97 | 7.7 |
| CA |  | 13.8 | 39.5 | [11] |
| P[CA](Tf2N) |  | 8.9 | 26.8 |
| P[DADMA](Tf2N) |  | 6.8 | 18.4 |
| PI/AAMSN | 3 wt% | 54 | 28.1 | [12] |
| PI | 0 | 66.7 | 27.8 |
| PEBAX/ZIF-8 | 3 wt% | 150 | 41.9 | [13] |
| P84 | 0 | 3.55 | 0.87 | [14] |

Table S3 CO2/CH4 Reference

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Membrane** | **Filler loading** | ***P*CO2 (Barrer)** | **αCO2/CH4** | **Ref** |
| P84 | 0 | 3.55 | 0.76 | [14] |
| PI/TiO2 | 5 wt% | 1.97 | 28.14 | [15] |
| PI | 0 | 2.35 | 19.58 |
| PI/TiO2 | 10 | 2.14 | 16.46 |
| PES | 0 | 25.7 | 3.57 | [16] |
| PES/CMS | 15 wt% | 68 | 11.15 |
| PES/Zeolite 4A |  | 6.7 | 28.7 | [17] |
| PC/Zeolite 4A |  | 4.6 | 51.8 | [18] |
| SEBS/ZIF-8 (S) |  | 14.71 | 5.2 | [7] |
| SEBS/ZIF-8 (M) |  | 15.23 | 5.4 |
| SEBS/ZIF-8 (L) |  | 15.59 | 5.2 |
| PSF/SWNT | 10 wt% | 5.19 | 18.53 | [19] |
| PBNPI/MWCNTs | 15 | 6 | 3.37 | [20] |
| PC-PEG/MWCNTs | 10 | 20.32 | 35.64 | [21] |
| Polyimide/MWCNTs | 1 | 37.37 | 16.5 | [22] |
| PSF/NH2-MIL-101 | 25 | 8.5 | 29 | [23] |
| Matrimid/N2-MIL-101 | 15 | 9.5 | 36 |

Table S4 H2/N2 References

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Membrane** | **Filler loading** | ***PH*2 (Barrer)** | **αH2/N2** | **Ref** |
| P84 | 0 | 25.3 | 4.1 | [14] |
| IL-Pebax/ZIF-8 | 8 wt% | 26 | 7.22 | [24] |
| Pebax/ZIF-8 | 8 wt% | 41 | 5.86 |
| PEI/SOD | 10 wt% | 1280 | 3 | [25] |
| MPU4 |  | 1.29 | 15 | [26] |
| PES | 0 | 6.5 | 46.43 | [27] |
| PES/Zeolite 13X | 50 wt% | 8.5 | 70.8 |
| PES/Zeolite 4A | 50 wt% | 14.1 | 56.4 |
| Pebax | 0 | 12.4 | 5.56 | [28] |
| Pebax/ZIF-67 | 2.5 | 9.91 | 4.86 |
| Pebax/ZIF-67 | 10 | 8.20 | 3.71 |
| Matrimid | 0 | 11.7 | 83.57 | [29] |
| P84 | 0 | 33.2 | 6.24 | [30] |

Table S5 H2/CH4 References

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Membrane** | **Filler loading** | ***PH*2 (Barrer)** | **αH2/CH4** | **Ref** |
| P84 | 0 | 25.3 | 3.56 | [14] |
| P84 | 0 | 33.2 | 4.96 | [30] |
| 6FDA-DAM | 0 | 21.4 | 33.96 | [31] |
| 6FDA-DAM/ZIF-11 | 10 wt% | 106.7 | 30.48 |
| 6FDA-DAM/ZIF-11 | 30 wt% | 76.76 | 31.98 |
| Matrimid/GO | 2 wt% | 30 | 130 | [32] |
| PSF/GO | 2 wt% | 12 | 55 |
| PI/MWCNT@GONRs | 2 wt% | 42.5 | 18.5 | [33] |
| P84/ND | 1 wt% | 6.7 | 310 | [34] |
| PI | 0 | 9 | 41 | [35] |
| PI/MOF5 | 5 wt% | 11 | 40 |
| Pebax/ZIF-8 | 8 wt% | 41 | 3 | [24] |
| IPebax/ZIF-8 | 8 wt% | 26 | 3.59 |
| P84 | 0 | 4.44 | 74 | [36] |
| PBNPI/MWCNT | 15 | 14.31 | 8.04 | [20] |
| PES/SAPO-34 |  | 0.206 | 61 | [37] |
| PC/Zeolite 4A |  | 13.4 | 54.4 | [18] |

**References**

[1] L.M. Robeson, Correlation of separation factor versus permeability for polymeric membranes, J. Memb. Sci. 62 (1991) 165–185. https://doi.org/10.1016/0376-7388(91)80060-J.

[2] L.M. Robeson, The upper bound revisited, J. Memb. Sci. 320 (2008) 390–400. https://doi.org/10.1016/j.memsci.2008.04.030.

[3] L. Ge, Z. Zhu, V. Rudolph, Enhanced gas permeability by fabricating functionalized multi-walled carbon nanotubes and polyethersulfone nanocomposite membrane, Sep. Purif. Technol. 78 (2011) 76–82. https://doi.org/10.1016/j.seppur.2011.01.024.

[4] L. Ge, Z. Zhu, F. Li, S. Liu, L. Wang, X. Tang, V. Rudolph, Investigation of Gas Permeability in Carbon Nanotube (CNT)−Polymer Matrix Membranes via Modifying CNTs with Functional Groups/Metals and Controlling Modification Location, J. Phys. Chem. C. 115 (2011) 6661–6670. https://doi.org/10.1021/jp1120965.

[5] Y. Shen, A.C. Lua, Structural and transport properties of BTDA-TDI/MDI co-polyimide (P84)–silica nanocomposite membranes for gas separation, Chem. Eng. J. 188 (2012) 199–209. https://doi.org/10.1016/j.cej.2012.01.043.

[6] H. Cong, J. Zhang, M. Radosz, Y. Shen, Carbon nanotube composite membranes of brominated poly(2,6-diphenyl-1,4-phenylene oxide) for gas separation, J. Memb. Sci. 294 (2007) 178–185. https://doi.org/10.1016/j.memsci.2007.02.035.

[7] W.S. Chi, S. Hwang, S.J. Lee, S. Park, Y.S. Bae, D.Y. Ryu, J.H. Kim, J. Kim, Mixed matrix membranes consisting of SEBS block copolymers and size-controlled ZIF-8 nanoparticles for CO2capture, J. Memb. Sci. 495 (2015) 479–488. https://doi.org/10.1016/j.memsci.2015.08.016.

[8] M. Waqas Anjum, F. de Clippel, J. Didden, A. Laeeq Khan, S. Couck, G. V. Baron, J.F.M. Denayer, B.F. Sels, I.F.J. Vankelecom, Polyimide mixed matrix membranes for CO2 separations using carbon–silica nanocomposite fillers, J. Memb. Sci. 495 (2015) 121–129. https://doi.org/10.1016/j.memsci.2015.08.006.

[9] W.F. Yong, F.Y. Li, Y.C. Xiao, P. Li, K.P. Pramoda, Y.W. Tong, T.S. Chung, Molecular engineering of PIM-1/Matrimid blend membranes for gas separation, J. Memb. Sci. 407–408 (2012) 47–57. https://doi.org/10.1016/j.memsci.2012.03.038.

[10] L. Sheng, J. Ren, K. Hua, H. Li, Y. Feng, M. Deng, The enhancement of mechanical properties of P84 hollow fiber membranes by thermally annealing below and above Tg, J. Memb. Sci. (2019) 117580. https://doi.org/10.1016/j.memsci.2019.117580.

[11] D. Nikolaeva, I. Azcune, M. Tanczyk, K. Warmuzinski, M. Jaschik, M. Sandru, P.I. Dahl, A. Genua, S. Loïs, E. Sheridan, A. Fuoco, I.F.J. Vankelecom, The performance of affordable and stable cellulose-based poly-ionic membranes in CO2/N2 and CO2/CH4 gas separation, J. Memb. Sci. 564 (2018) 552–561. https://doi.org/10.1016/j.memsci.2018.07.057.

[12] C.-C. Hu, P.-H. Cheng, S.-C. Chou, C.-L. Lai, S.-H. Huang, H.-A. Tsai, W.-S. Hung, K.-R. Lee, Separation behavior of amorphous amino-modified silica nanoparticle/polyimide mixed matrix membranes for gas separation, J. Memb. Sci. (2019) 117542. https://doi.org/10.1016/j.memsci.2019.117542.

[13] P.D. Sutrisna, J. Hou, H. Li, Y. Zhang, V. Chen, Improved operational stability of Pebax-based gas separation membranes with ZIF-8: A comparative study of flat sheet and composite hollow fibre membranes, J. Memb. Sci. 524 (2017) 266–279. https://doi.org/10.1016/j.memsci.2016.11.048.

[14] E.P. Favvas, E.P. Kouvelos, G.E. Romanos, G.I. Pilatos, A.C. Mitropoulos, N.K. Kanellopoulos, Characterization of highly selective microporous carbon hollow fiber membranes prepared from a commercial co-polyimide precursor, J. Porous Mater. 15 (2008) 625–633. https://doi.org/10.1007/s10934-007-9142-2.

[15] H. Ahmadizadegan, M. Tahriri, M. Tahriri, M. Padam, M. Ranjbar, Polyimide-TiO2 nanocomposites and their corresponding membranes: Synthesis, characterization, and gas separation applications, Solid State Sci. 89 (2019) 25–36. https://doi.org/10.1016/j.solidstatesciences.2018.12.016.

[16] M. Farnam, H. Mukhtar, A. Shariff, Analysis of the Influence of CMS Variable Percentages on Pure PES Membrane Gas Separation Performance, Procedia Eng. 148 (2016) 1206–1212. https://doi.org/10.1016/j.proeng.2016.06.449.

[17] A.F. Ismail, T.D. Kusworo, A. Mustafa, Enhanced gas permeation performance of polyethersulfone mixed matrix hollow fiber membranes using novel Dynasylan Ameo silane agent, J. Memb. Sci. 319 (2008) 306–312. https://doi.org/10.1016/j.memsci.2008.03.067.

[18] D. Şen, H. Kalipçilar, L. Yilmaz, Development of polycarbonate based zeolite 4A filled mixed matrix gas separation membranes, J. Memb. Sci. 303 (2007) 194–203. https://doi.org/10.1016/j.memsci.2007.07.010.

[19] S. Kim, L. Chen, J.K. Johnson, E. Marand, Polysulfone and functionalized carbon nanotube mixed matrix membranes for gas separation: Theory and experiment, J. Memb. Sci. 294 (2007) 147–158. https://doi.org/10.1016/j.memsci.2007.02.028.

[20] T.H. Weng, H.H. Tseng, M.Y. Wey, Preparation and characterization of multi-walled carbon nanotube/PBNPI nanocomposite membrane for H2/CH4 separation, Int. J. Hydrogen Energy. 34 (2009) 8707–8715. https://doi.org/10.1016/j.ijhydene.2009.08.027.

[21] A.R. Moghadassi, Z. Rajabi, S.M. Hosseini, M. Mohammadi, Preparation and Characterization of Polycarbonate-Blend-Raw/Functionalized Multi-Walled Carbon Nano Tubes Mixed Matrix Membrane for CO 2 Separation, Sep. Sci. Technol. 48 (2013) 1261–1271. https://doi.org/10.1080/01496395.2012.730597.

[22] M.A. Aroon, A.F. Ismail, M.M. Montazer-Rahmati, T. Matsuura, Effect of chitosan as a functionalization agent on the performance and separation properties of polyimide/multi-walled carbon nanotubes mixed matrix flat sheet membranes, J. Memb. Sci. 364 (2010) 309–317. https://doi.org/10.1016/j.memsci.2010.08.023.

[23] T. Rodenas, M. van Dalen, P. Serra-Crespo, F. Kapteijn, J. Gascon, Mixed matrix membranes based on NH2-functionalized MIL-type MOFs: Influence of structural and operational parameters on the CO2/CH4 separation performance, Microporous Mesoporous Mater. 192 (2014) 35–42. https://doi.org/10.1016/j.micromeso.2013.08.049.

[24] A. Jomekian, B. Bazooyar, R.M. Behbahani, T. Mohammadi, A. Kargari, Ionic liquid-modified Pebax® 1657 membrane filled by ZIF-8 particles for separation of CO2 from CH4, N2 and H2, J. Memb. Sci. 524 (2017) 652–662. https://doi.org/10.1016/j.memsci.2016.11.065.

[25] G. Yang, H. Guo, Z. Kang, L. Zhao, S. Feng, F. Jiao, S. Mintova, Green Hydrogen Separation from Nitrogen by Mixed‐Matrix Membranes Consisting of Nanosized Sodalite Crystals, ChemSusChem. 12 (2019) 4529–4537. https://doi.org/10.1002/cssc.201802577.

[26] L.-S. Teo, C.-Y. Chen, J.-F. Kuo, The gas transport properties of amine-containing polyurethane and poly(urethane-urea) membranes, J. Memb. Sci. 141 (1998) 91–99. https://doi.org/10.1016/S0376-7388(97)00293-7.

[27] M.G. Süer, N. Baç, L. Yilmaz, Gas permeation characteristics of polymer-zeolite mixed matrix membranes, J. Memb. Sci. 91 (1994) 77–86. https://doi.org/10.1016/0376-7388(94)00018-2.

[28] S. Feng, M. Bu, J. Pang, W. Fan, L. Fan, H. Zhao, G. Yang, H. Guo, G. Kong, H. Sun, Z. Kang, D. Sun, Hydrothermal stable ZIF-67 nanosheets via morphology regulation strategy to construct mixed-matrix membrane for gas separation, J. Memb. Sci. 593 (2020) 117404. https://doi.org/10.1016/j.memsci.2019.117404.

[29] E.Y. Kim, H.S. Kim, D. Kim, J. Kim, P.S. Lee, Preparation of mixed matrix membranes containing ZIF-8 and UiO-66 for multicomponent light gas separation, Crystals. 9 (2019). https://doi.org/10.3390/cryst9010015.

[30] E.P. Favvas, N.S. Heliopoulos, S.K. Papageorgiou, A.C. Mitropoulos, G.C. Kapantaidakis, N.K. Kanellopoulos, Helium and hydrogen selective carbon hollow fiber membranes: The effect of pyrolysis isothermal time, Sep. Purif. Technol. 142 (2015) 176–181. https://doi.org/10.1016/j.seppur.2014.12.048.

[31] M. Safak Boroglu, A.B. Yumru, Gas separation performance of 6FDA-DAM-ZIF-11 mixed-matrix membranes for H2/CH4 and CO2/CH4 separation, Sep. Purif. Technol. 173 (2017) 269–279. https://doi.org/10.1016/j.seppur.2016.09.037.

[32] S. Castarlenas, C. Téllez, J. Coronas, Gas separation with mixed matrix membranes obtained from MOF UiO-66-graphite oxide hybrids, J. Memb. Sci. 526 (2017) 205–211. https://doi.org/10.1016/j.memsci.2016.12.041.

[33] Q. Xue, X. Pan, X. Li, J. Zhang, Q. Guo, Effective enhancement of gas separation performance in mixed matrix membranes using core/shell structured multi-walled carbon nanotube/graphene oxide nanoribbons, Nanotechnology. 28 (2017) 065702. https://doi.org/10.1088/1361-6528/aa510d.

[34] A. Pulyalina, G. Polotskaya, V. Rostovtseva, Z. Pientka, A. Toikka, Improved Hydrogen Separation Using Hybrid Membrane Composed of Nanodiamonds and P84 Copolyimide, Polymers (Basel). 10 (2018) 828. https://doi.org/10.3390/polym10080828.

[35] H. Aykac Ozen, B. Ozturk, Gas separation characteristic of mixed matrix membrane prepared by MOF-5 including different metals, Sep. Purif. Technol. 211 (2019) 514–521. https://doi.org/10.1016/j.seppur.2018.09.052.

[36] S.-H. Choi, A. Brunetti, E. Drioli, G. Barbieri, H 2 Separation From H 2 /N 2 and H 2 /CO Mixtures with Co-Polyimide Hollow Fiber Module, Sep. Sci. Technol. 46 (2010) 1–13. https://doi.org/10.1080/01496395.2010.487847.

[37] E. Karatay, H. Kalipçilar, L. Yilmaz, Preparation and performance assessment of binary and ternary PES-SAPO 34-HMA based gas separation membranes, J. Memb. Sci. 364 (2010) 75–81. https://doi.org/10.1016/j.memsci.2010.08.004.