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Separation of by-product gases in steel industry for carbon resources utilization by semi-alicyclic polyimide membranes

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Huge amount of by-product gas mixtures (H_2 , CH_4 , CO , CO_2 , N_2 , etc.) has been produced from steel industry in Korea and worldwide. Among the by-product gases, CH_4 and H_2 are included in coke oven gas (COG). CO_2 and CO is included in blast furnace gas (BFG) and Linz Donawitz gas (LDG) in steel industry. If they can be efficiently separated at high purity and recovery, they can be used valuably for carbon capture and utilization (CCU) producing chemical products such as methanol, ethanol, ethylene, acetic acid, etc. and therefore, contributing to reduction of global warming. There are typical separation technologies: cryogenic, adsorption, absorption and membrane. Membrane technology can be a promising new separation technology owing to cheap plant construction, easy operation, environmental friendliness etc. The economy and energy efficiency of membrane process depends mainly upon gas selectivity and gas permeability of membrane materials. This study shows the preliminary result for the gas separation properties of soluble polyimides as membrane materials - CO_2/N_2 (or CO_2/CO) and H_2/CH_4 (or CO_2/CH_4) selectivities and CO_2 and H_2 permeabilities. We have developed an alicyclic dianhydride -5-(2,5-dioxotetrahydrofuryl)-3-methyl-3-cyclohexene-1,2-dicarboxylic anhydride (DOCCA) based homo- and co-polyimides with various dianhydrides and diamines using m-cresol as a solvent, respectively. Thin dense membranes were prepared from the copolyimides to check their gas permeation properties with a time-lag apparatus. All homo- and copolyimides showed good solubility for organic solvents and excellent thermal stability. The synthesized polyimides showed excellent CO_2/N_2 (or CO_2/CO) and H_2/CH_4 (or CO_2/CH_4) selectivities, and high CO_2 and H_2 permeabilities. These results confirmed these polyimides could be used as membrane materials for the separation of CO_2 , CH_4 , H_2 and from by-product gases in steel industry.

Recent Publications

1. L Zhai et al. (2012) Preparation and characterization of highly transparent and colorless semi-aromatic polyimide films derived from alicyclic dianhydride and aromatic diamines. *Polymer* 53:3529-3539.
2. S Xia et al. (2013) Synthesis of soluble polyimide derived from novel naphthalene diamines for liquid crystal alignment layers and a preliminary study on the mechanism of imidization, *RSC Adv.* 3:14661-14670.
3. T Yusuke et al. (2014) Surface wettability controllable polyimides bearing long-chain alkyl groups via phenyl ester linkages by ultraviolet light irradiation. *High Performance Polymers* 27:46-58.
4. A F Ismail et al. (2015) Gas separation membranes: Polymeric and inorganic. Springer, ISBN 978-3-319-01095-3.
5. D Guzmán Lucero et al. (2015) Gas permeation properties of soluble aromatic polyimides based on 4-fluoro-4,4'-diaminotriphenylmethane. *Materials* 8:1951.

Biography

Jeong Hoon Kim is Head of Greenhouse Gas Separation and Recovery Research Group, Carbon Resources Institute, KRICT. He got his BS and MS degree at Hanyang University in 1984 and 1986, respectively, and got his PhD from Korea Advanced Institute of Technology (KAIST) in 1999. He also worked as a Postdoc position at the Department of Chemical Engineering in University of Waterloo, Canada in 2000-2001. He has intensively been studying the synthesis of polymer membranes, fabrication of asymmetric flat and hollow fiber membranes, and development of multi-stage membrane process in the field of gas separation, electro dialysis, pervaporation, water purification, electro dialysis, and fuel cells since 1984. He published about 65 papers in Korean Journals and 30 papers in international Journals, and owns more than 100 Korean and international patents.

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Table 1. Pure gas permeability and gas ideal selectivity of DOCCA based polyimides and four commercial polymeric membrane materials.

Polymer	Permeability (Barrer)							
	P_{CO_2}	P_{N_2}	P_{CO}	P_{H_2}	P_{CH_4}	P_{O_2}	P_{H_2O}	P_{H_2S}
DOCCA-pPDA	81.5	8.0	2.2	8.8	0.44	0.15		
DOCCA-SAP	61.2	6.0	1.8	8.2	0.32	0.12		
DOCCA-NDA	50.0	5.0	1.4	6.7	0.28	0.08		
DOCCA-ODA	71.0	7.0	2.0	8.5	0.38	0.12		
DOCCA-pPEA	136	17.4	3.7	13.0	0.60	0.15		
Matsuyama 132	21.2	7.1	2.1	N/A	0.28	0.12		
PAI(30)	8.8	1.7	0.4	N/A	0.05	0.03		
Polysulfone(4)	31.8	6.1	1.4	N/A	0.34	0.12		
Cellulose acetate(5)	0.40	0.1	0.03	N/A	0.01	0.01		

Table 2. Ideal selectivity of DOCCA based polyimides and four commercial polymeric membrane materials.

Polymer	Ideal selectivity (α)			
	α_{CO_2/N_2}	$\alpha_{CO_2/CO}$	α_{H_2/CH_4}	α_{CO_2/H_2}
DOCCA-pPDA	10.1	11.5	11.0	1.0
DOCCA-SAP	10.5	10.5	14.1	1.5
DOCCA-NDA	14.3	11.3	18.4	1.7
DOCCA-ODA	10.1	11.0	11.3	1.0
DOCCA-pPEA	10.2	10.3	8.2	1.0
Matsuyama 132	12.0	10.0	N/A	7.5
PAI(30)	10.0	10.0	N/A	1.0
Polysulfone(4)	10.0	10.0	N/A	1.0
Cellulose acetate(5)	1.0	1.0	N/A	1.0