Open access • Journal Article • DOI:10.1021/ACS.IECR.6B04868

Zeolitic Imidazolate Framework-8 (ZIF-8) Membranes for Kr/Xe Separation

— Source link

Ting Wu, Xuhui Feng, Sameh K. Elsaidi, Praveen K. Thallapally ...+1 more authors

Institutions: Colorado School of Mines, Pacific Northwest National Laboratory

Published on: 01 Feb 2017 - Industrial & Engineering Chemistry Research (American Chemical Society)

Topics: Zeolitic imidazolate framework, Imidazolate and Membrane

COLORADO SCHOOL OF MINES
EARTH ● ENERGY ● ENVIRONMENT

Zeolitic Imidazolate Framework-8 (ZIF-8) Membranes for Kr/Xe Separation

Ting Wu
Chemical Engineering Department

Xe and Kr Separation in Nuclear Reprocessing

Separating Krypton (Kr) from Xenon (Xe) is a critical step during treatment of spent nuclear fuel

- 85Kr -> **radioactive**
- **10Xe:1Kr**

Need to store 85Kr for 100 years

- Separating Xe and Kr can save space and money!

Xe used in lighting, medical applications, including neuroprotection, imaging, and anesthesia

- Currently expensive

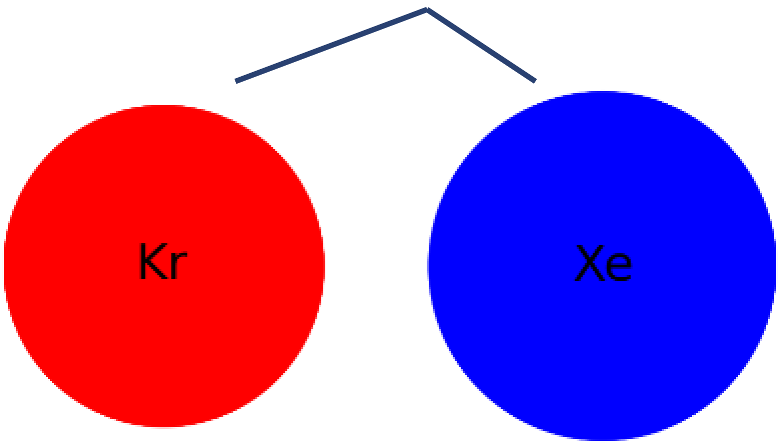
Benchmark separation technology: cryogenic distillation

- Energy intensive and expensive process.

Distinguishing Xe and Kr

Membrane Separation
(size)

← No phase transformation!



Diameter = 0.37 nm
Boiling point = -153.4 °C

Diameter = 0.41 nm
Boiling point = -108.1 °C

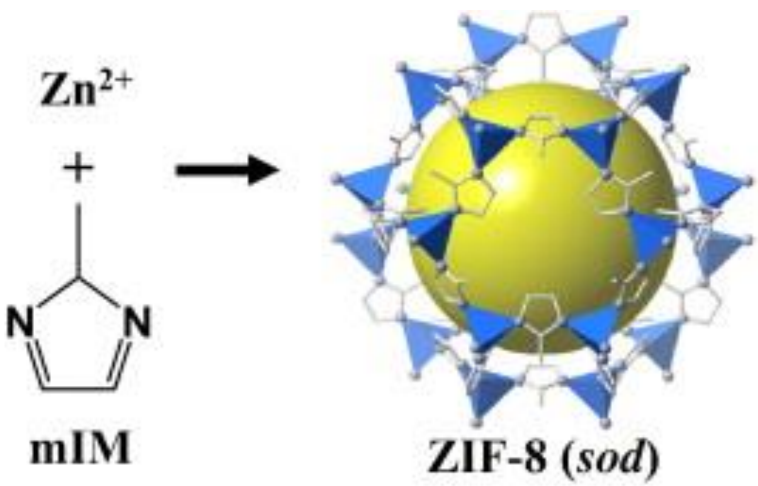
Cryogenic Distillation
(temperature)

Physical properties	Kr	Xe
Kinetic diameter (nm)	0.366	0.405
Polarizability (10^{-25} cm^3)	24.844	40.44
Dipole moment (esu cm)	0	0
Quadruple moment (esu cm^2)	0	0

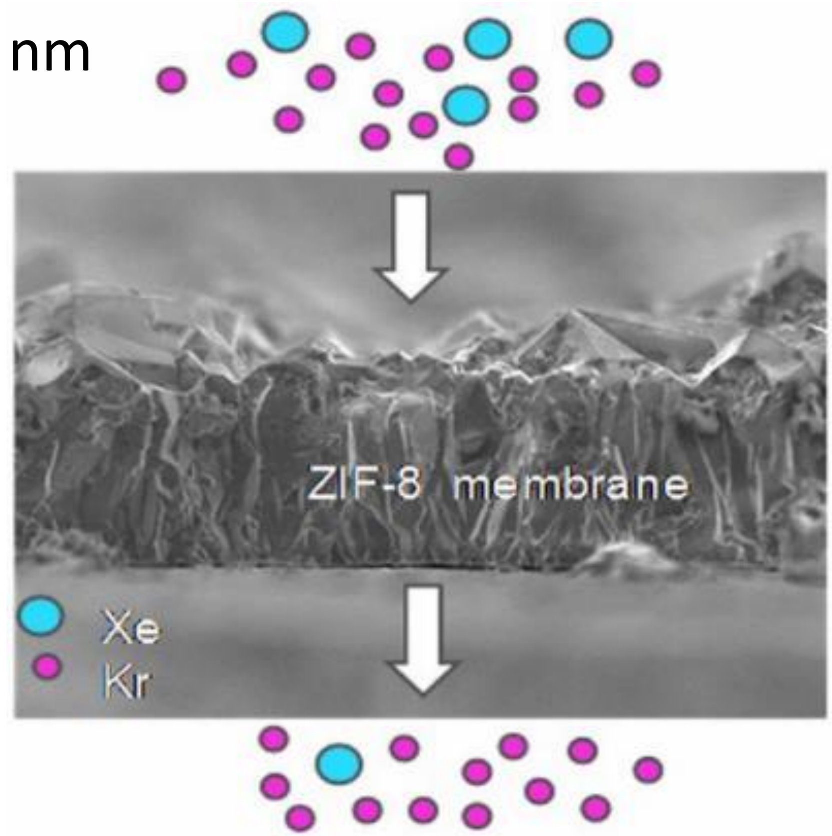
Wu, T., Feng, X., Elsaidi, S. K., Thallapally, P. K., & Carreon, M. A. (2017). *Industrial & Engineering Chemistry Research*, *56*(6).

ZIF-8

- Family of Metal–organic frameworks (MOFs)
- Zeolitic imidazolate framework, thermally and chemically stable
- Effective aperture size of 0.4–0.42 nm



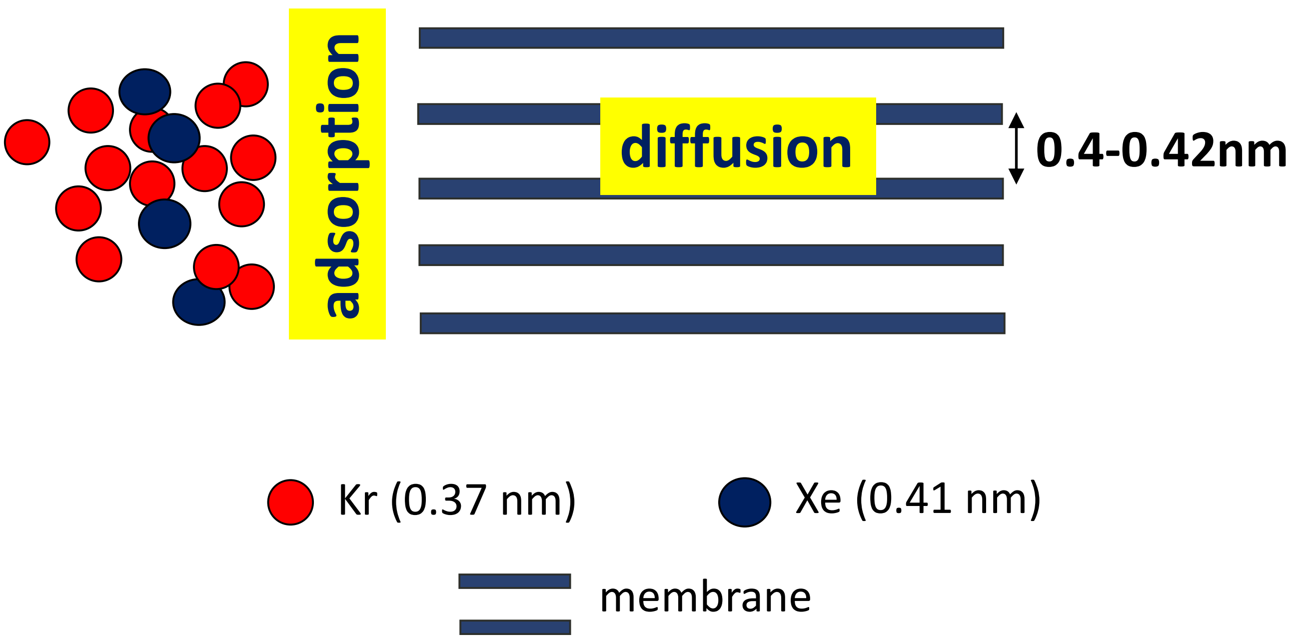
Crystal structure of ZIF-8: Zn (polyhedral), N (sphere), and C (line)



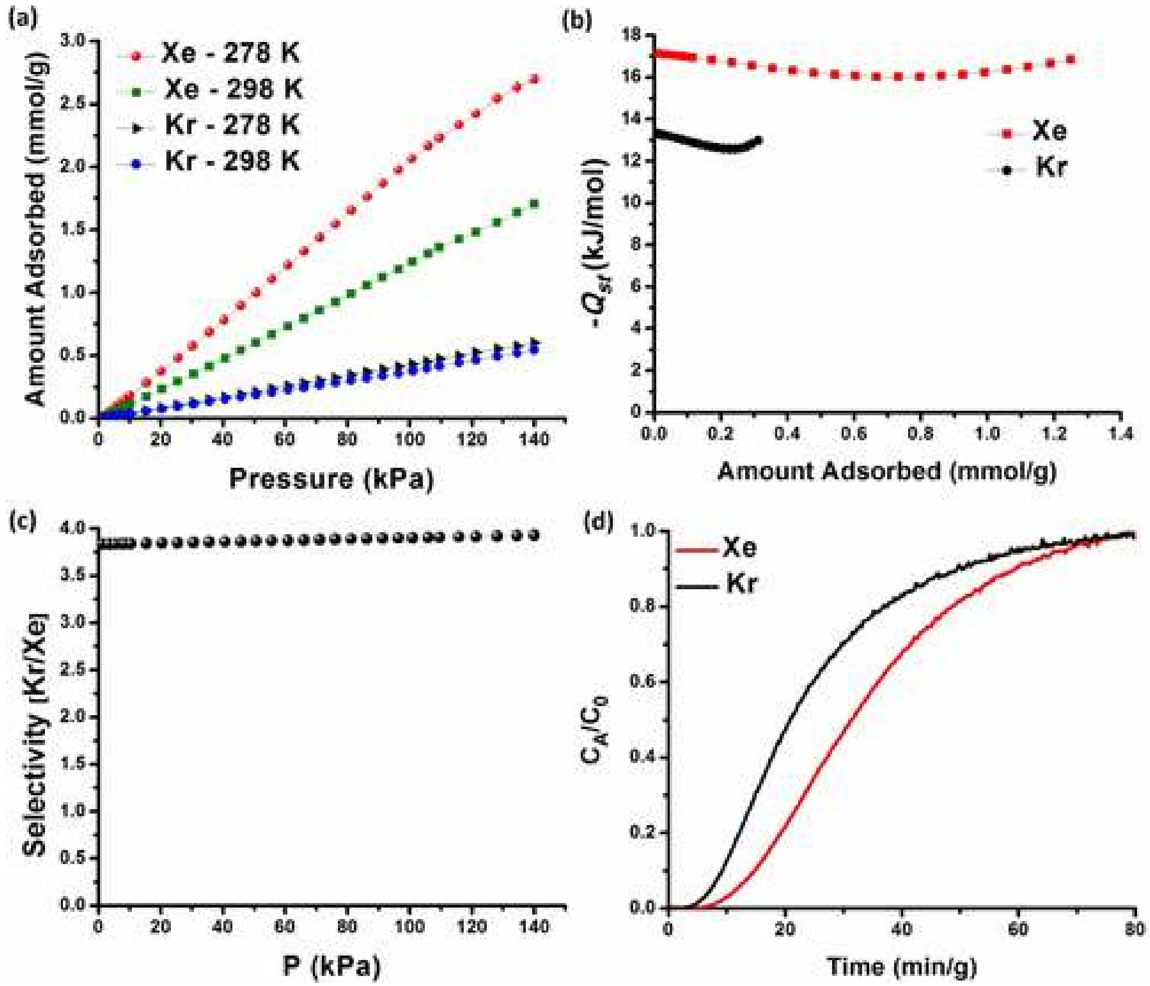
Wu, T., Feng, X., Elsaidi, S. K., Thallapally, P. K., & Carreon, M. A. (2017). *Industrial & Engineering Chemistry Research*, *56*(6).

Gas Separation Process

- Adsorption: electrostatic interaction
- Molecular sieving: membrane and gas size
- Diffusion: molecular radii and weight

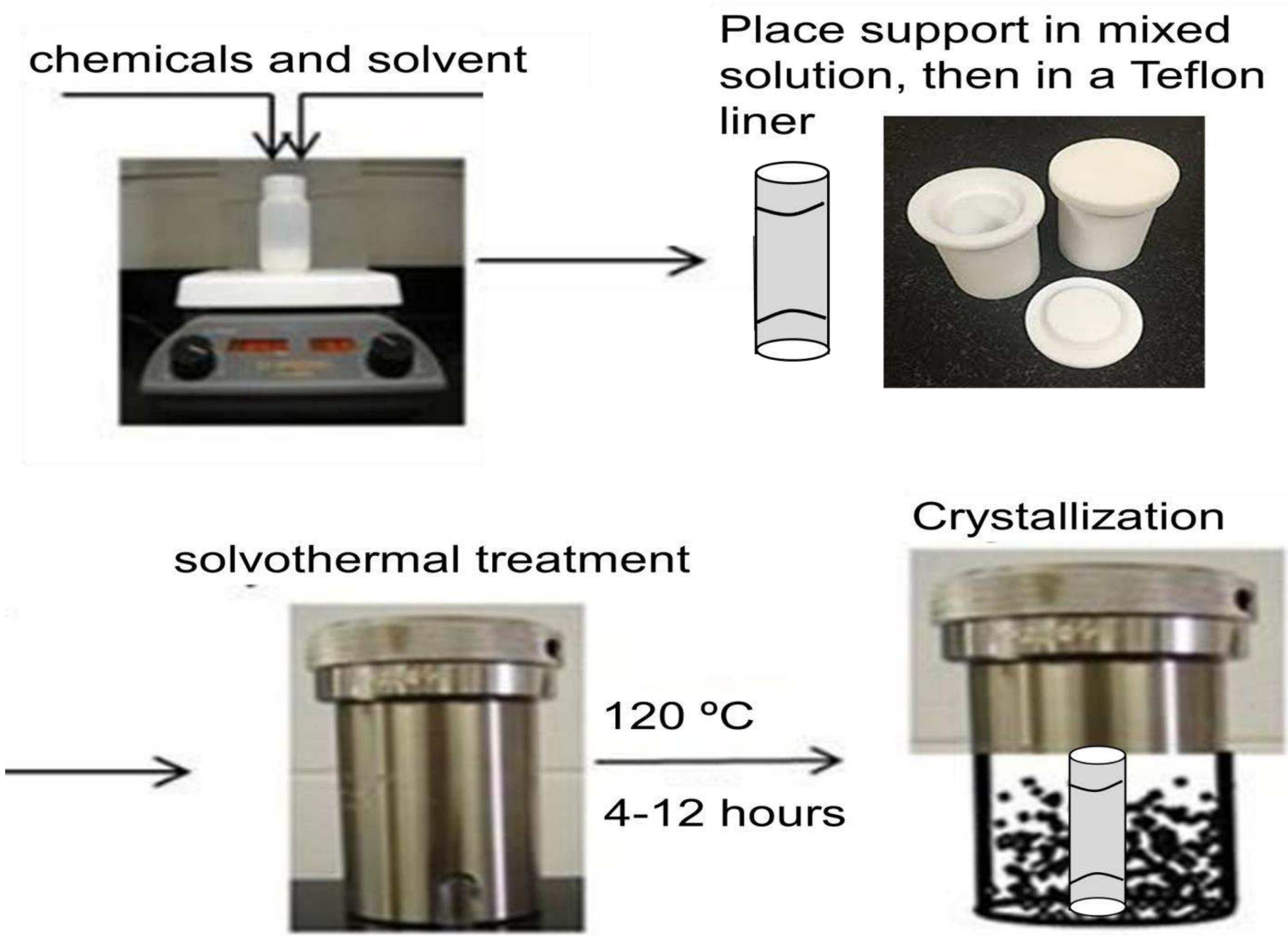


ZIF-8 Adsorption and Diffusion Selectivity



Diffusion is the dominant mechanism

Membrane Preparation



Membrane Separation Results

Permeance = pressure - normalized flux, membrane property

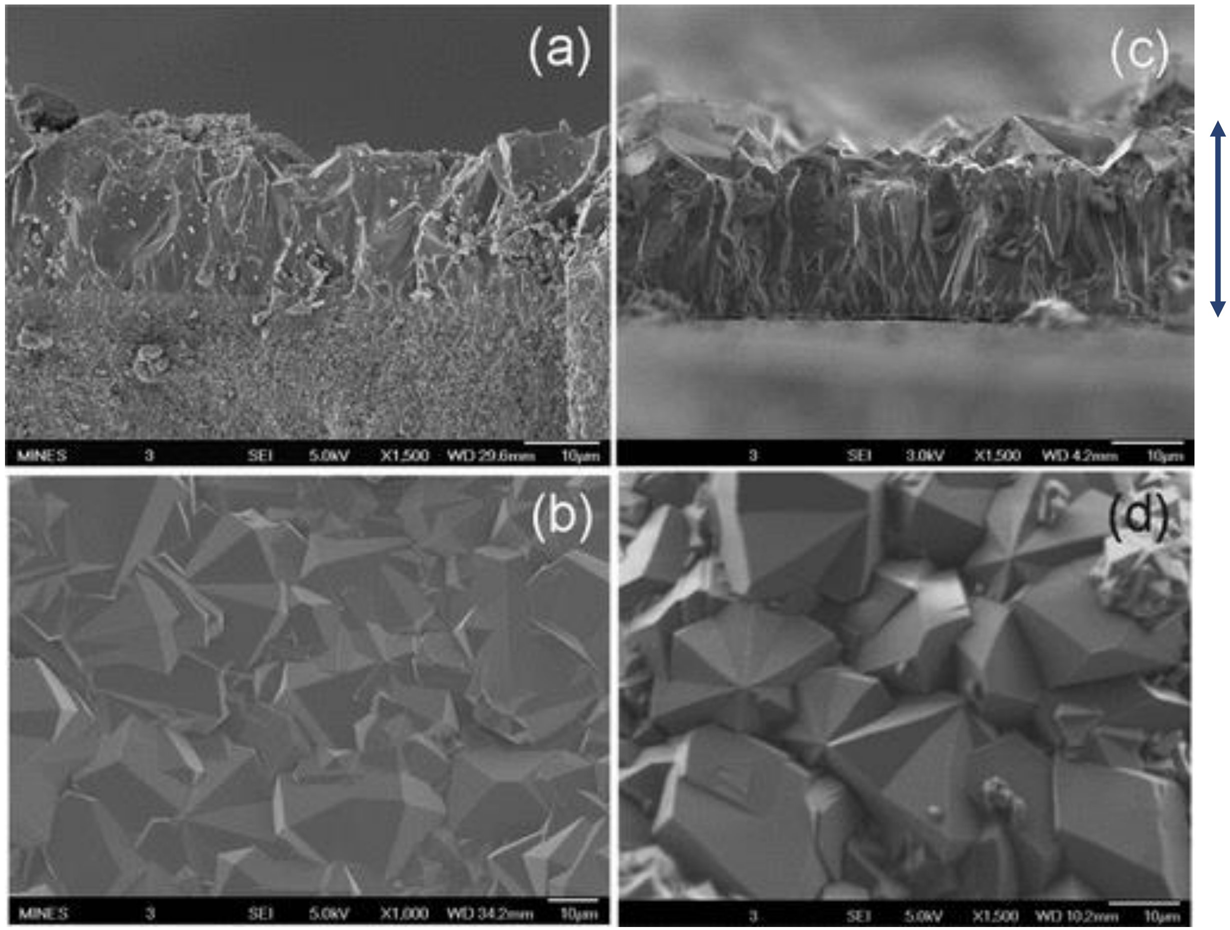
Separation factor α for a ratio of Kr permeance/Xe permeance.

Kr/Xe separation performance over ZIF-8 membranes. Molar gas mixture composition: 92:8 Kr/Xe. Transmembrane pressure 138 kPa.

Membrane ID [a]	Kr permeance ($mol/m^2 \cdot s \cdot Pa$) [b]	Separation selectivity (α)	Separation index (π) [c]	
1A	0.33×10^{-8} (9.9)	5.9	13.6×10^{-4}	2-layer
1B	0.17×10^{-8} (5.1)	10.8	13.9×10^{-4}	2-layer
2A	1.7×10^{-8} (50.8)	12.3	162×10^{-4}	2-layer
2B	1.3×10^{-8} (38.8)	16.1	164×10^{-4}	2-layer
3	0.5×10^{-8} (14.9)	7.9	28.9×10^{-4}	3-layer

[a] 1A, 1B, 2A, 2B are two layer membranes. 3 is three-layer membrane. [b] Numbers in parentheses indicate Gas permeation units (GPU). [c] π= Kr permeance × (selectivity-1) × permeate pressure

SEM Images of ZIF-8 2-Layer Membranes



Conclusion and Future work

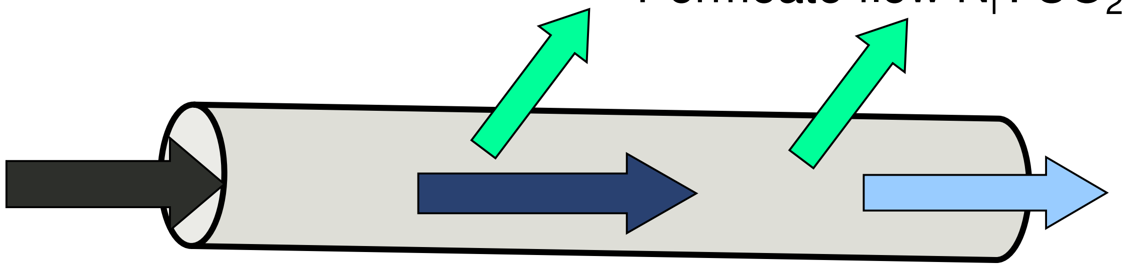
- Successfully synthesized continuous ZIF-8 membranes and first to demonstrate Kr/Xe gas separation with MOF membrane
  - Average Kr permeances as high as 1.5 × 10–8 ± 0.2 mol/m^{2}s Pa and average separation selectivities of 14.2 ± 1.9 for molar feed compositions in air ratio
  - Molecular sieving, competitive adsorption, and differences in diffusivities were the prevailing separation mechanisms.
  - Promising alternative to the benchmark technology cryogenic distillation currently employed to separate Kr/Xe
- Next step:
  - Optimize synthesis parameters to improve selectivity and permeance

Thank you!

- Advisor: Prof. Moises Carreon
- Co-authors: Xuhui Feng, Sameh K. Elsaidi, Praveen K. Thallapally

- Kr: 83.798 g/mol
- Xe: 131.293 g/mol

Membrane Performance Parameters



zeolite membrane inside of an alumina support

Permeance= pressure - normalized flux, membrane property

$$\prod_i (mol \cdot m^{-2} \cdot s^{-1} \cdot Pa^{-1}) = \frac{N_i}{\Delta P_i}$$

Permeability=permeance*membrane thickness, material property

$$P_i (mol \cdot m^{-1} \cdot s^{-1} \cdot Pa^{-1}) = \prod_i l$$

Separation factor α for a binary mixture of component A(CO_2) and B(CH_4):

$$\alpha_{A/B} = \frac{(c_A / c_B)_{permeate}}{(c_A / c_B)_{feed}}$$