

Highly Selective H₂ Separation Zeolite Membranes for Coal Gasification

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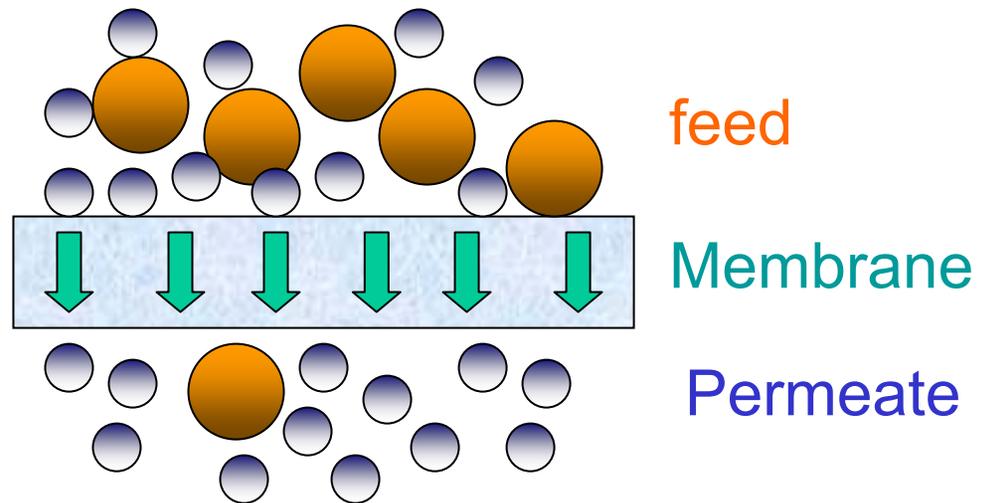
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Membrane process has the potential to separate H_2 with high energy efficiency

- Coal Gasification
 - Coal converted in synthetic natural gas
 - Need to separate H_2 from CO_2 , CO , CH_4 and H_2O
- Membrane process
 - Energy efficient
 - Flexible on demand
 - Continuous



Zeolite membranes have the potential to separate H₂ from coal gasification products

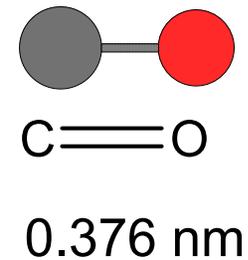
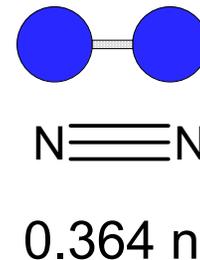
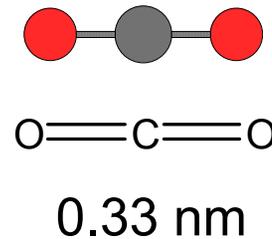
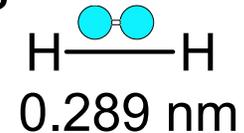
- Zeolites

- Nanoporous crystalline aluminosilicates

- **Molecular diameter pores**

- Uniform pore size for a given type

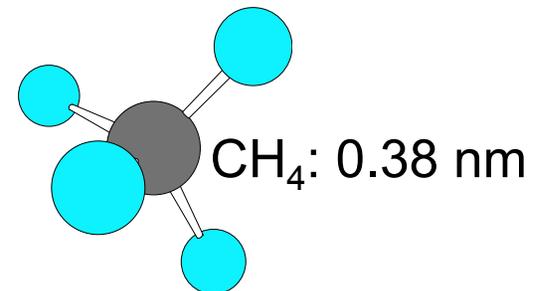
- Discrete pore size



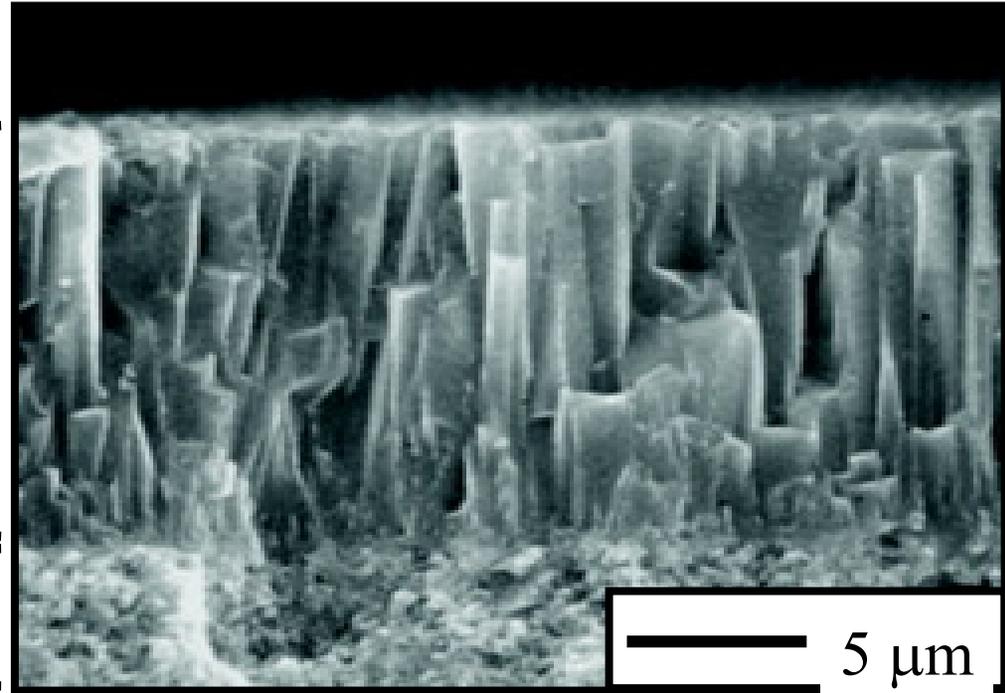
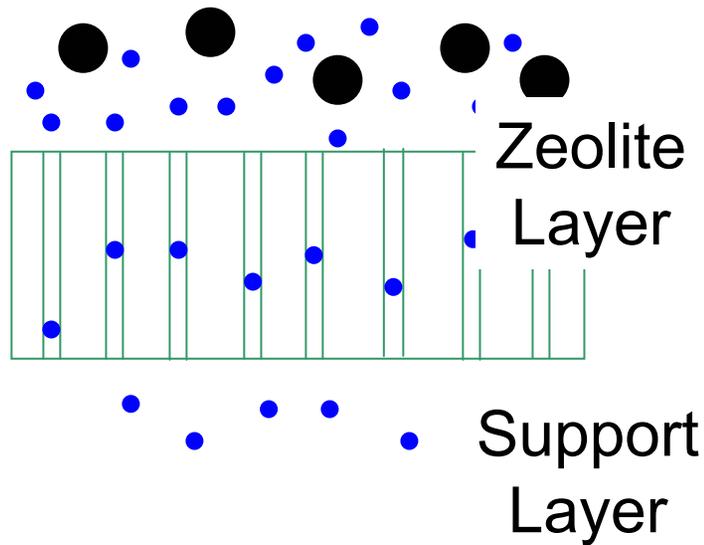
- Zeolite membranes

- Polycrystalline

- Mechanically, chemically and thermally stable



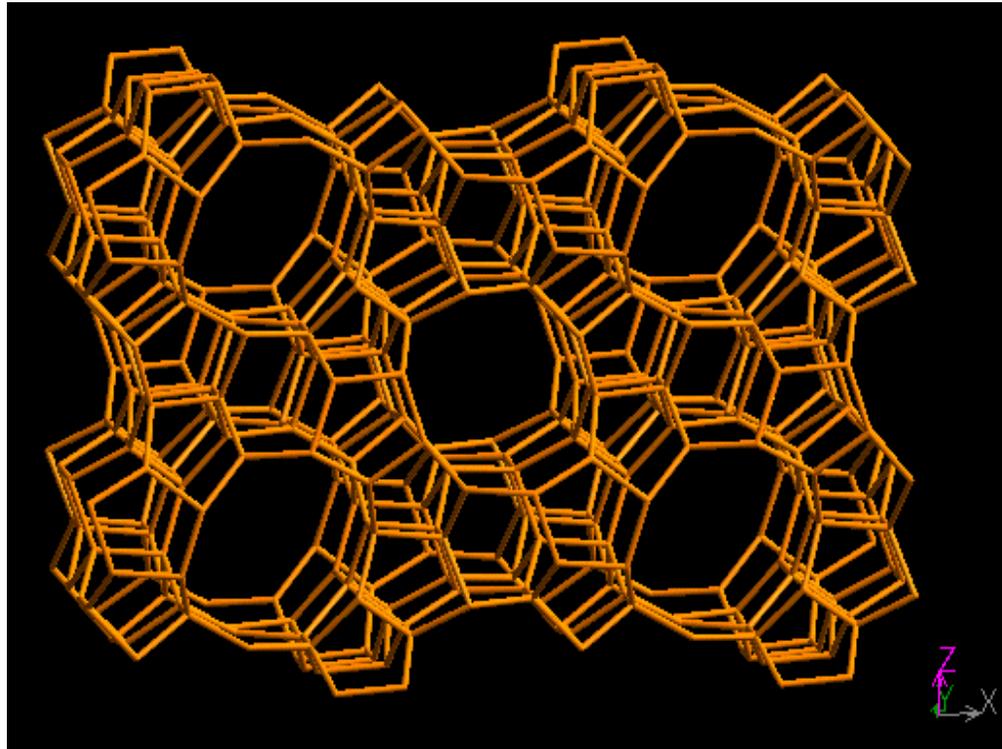
Good quality zeolite membranes should have a negligible amount of defects



Ideal zeolite membrane

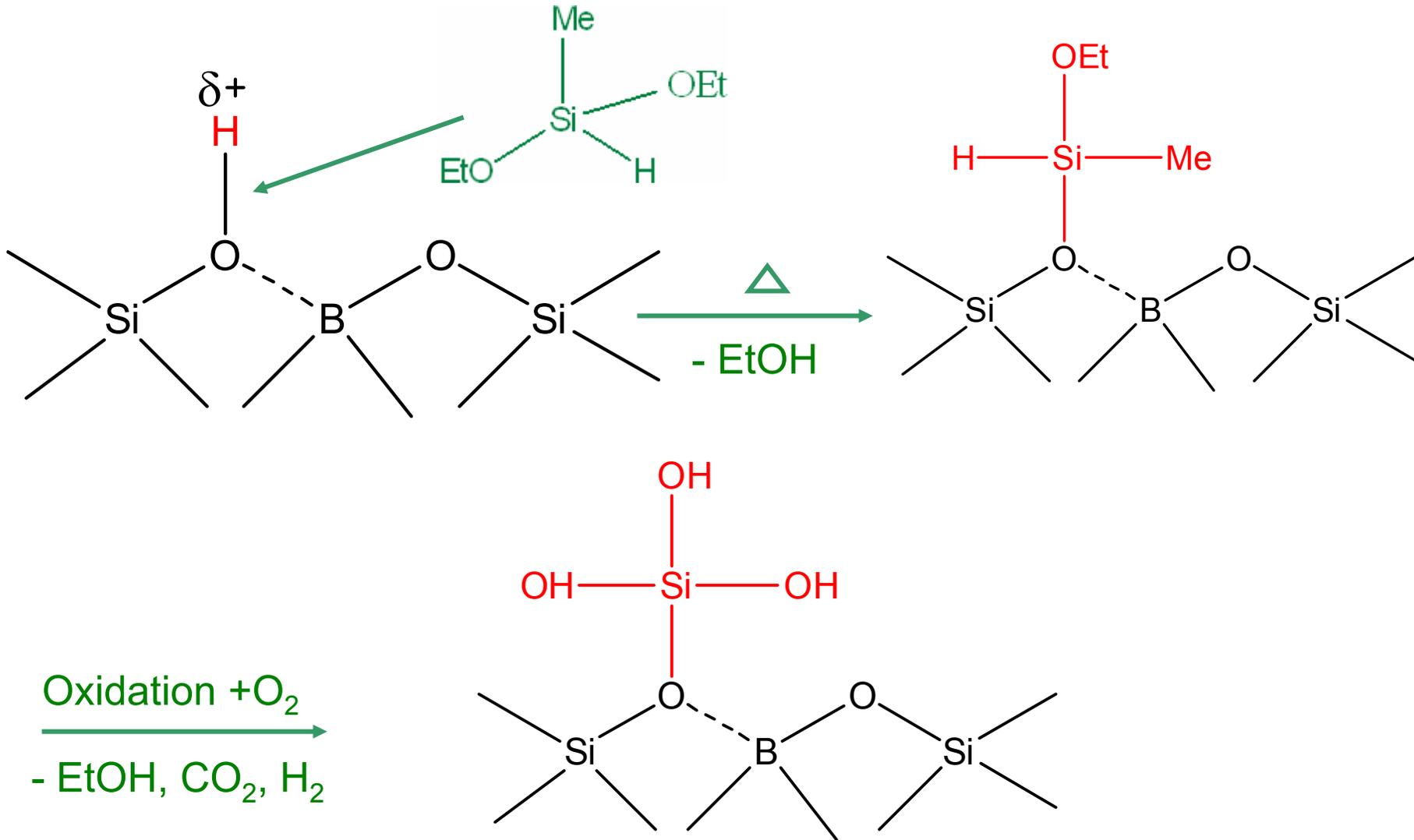
Real zeolite membrane

MFI zeolite membranes not selective for H₂

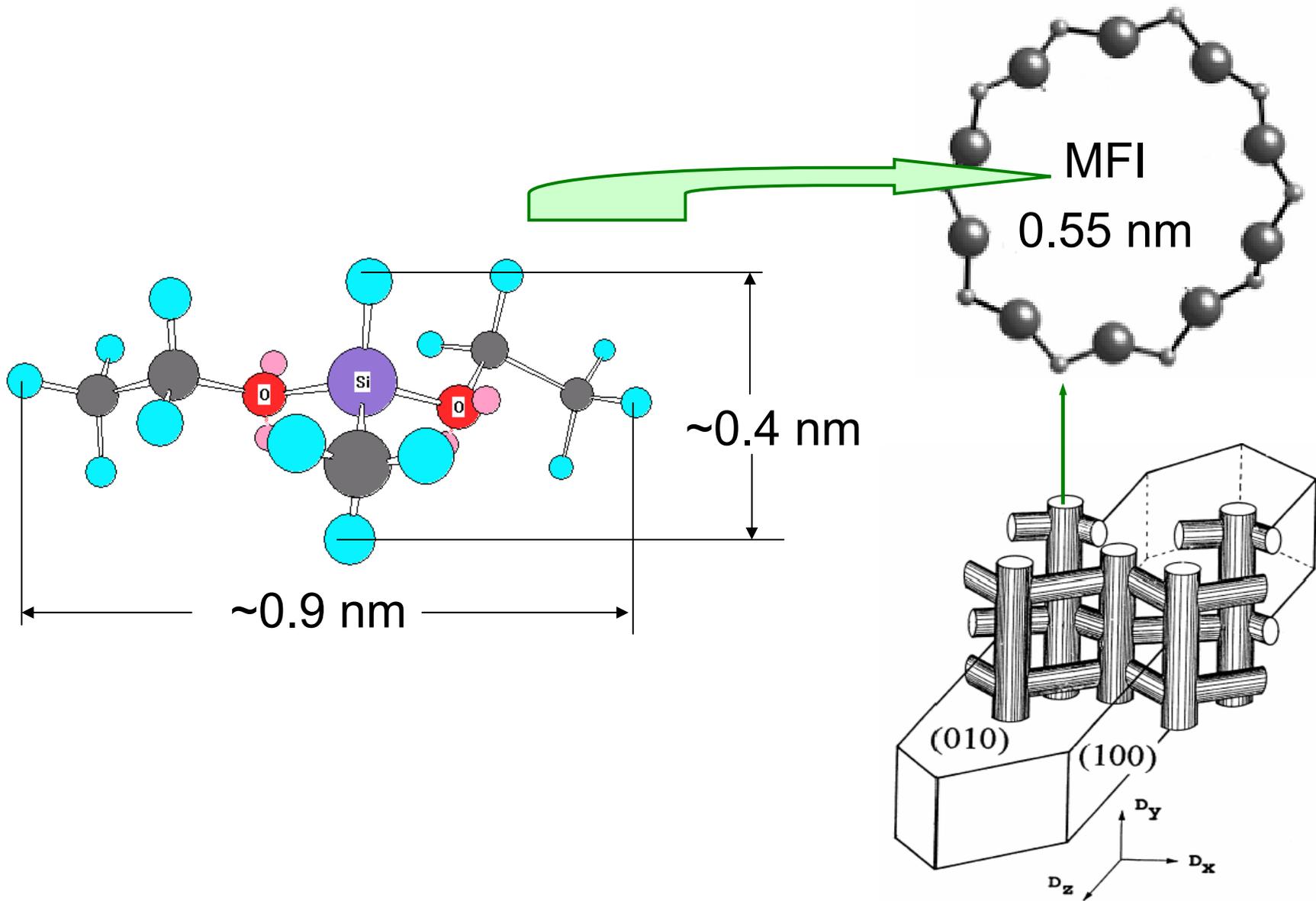


- MFI (e.g., B-ZSM-5)
 - 10-membered ring
 - Pore size ~0.55 nm
 - Pores too big
- Silylation on MFI
 - Reduce pore size
 - Separate H₂

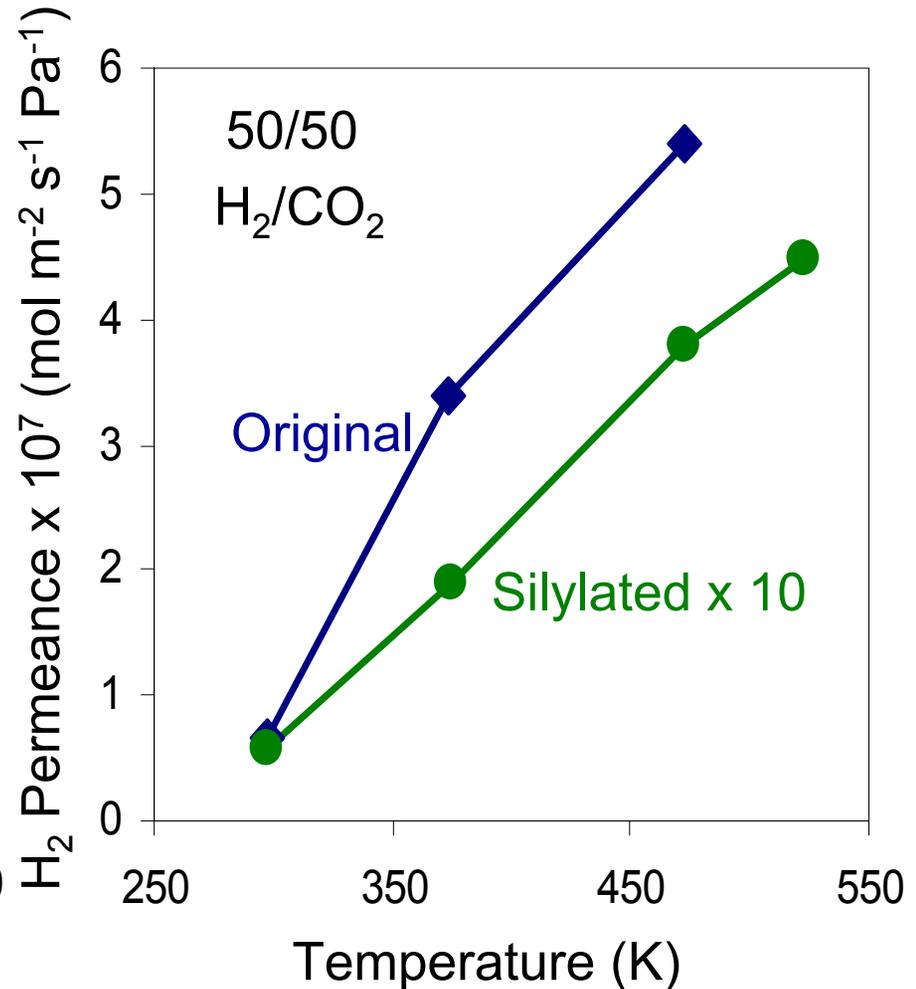
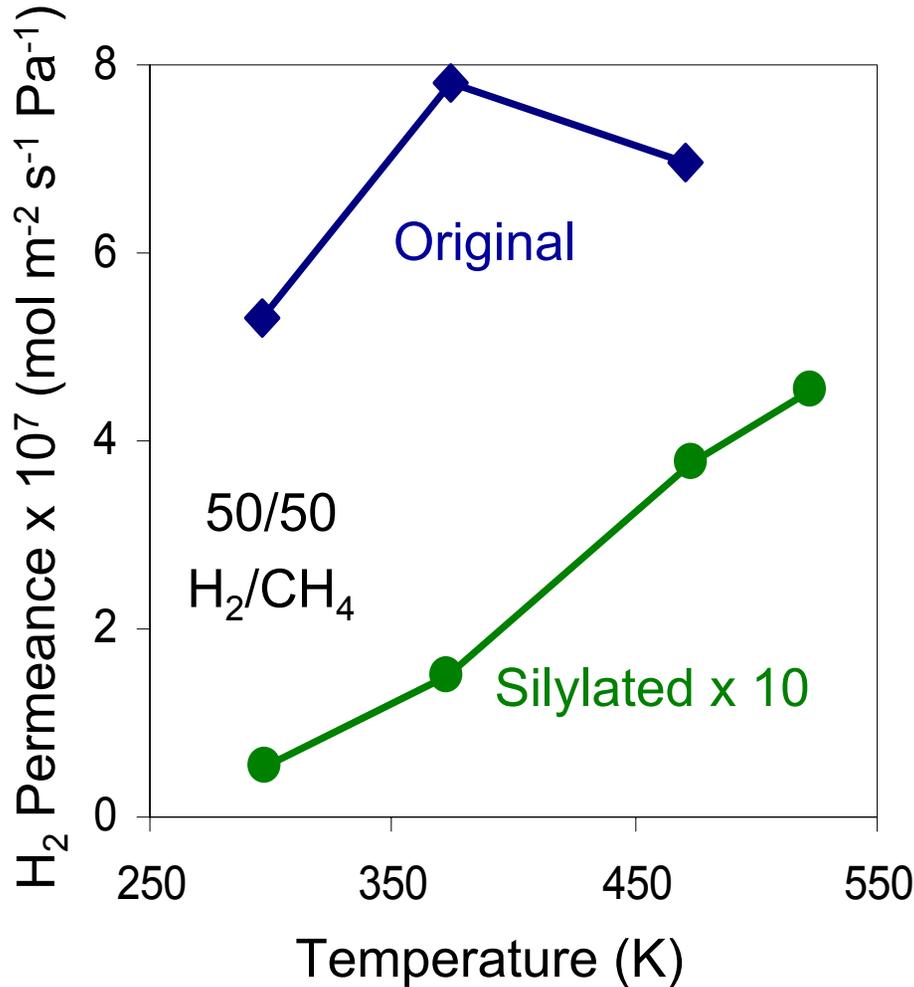
Zeolite membranes were silylated



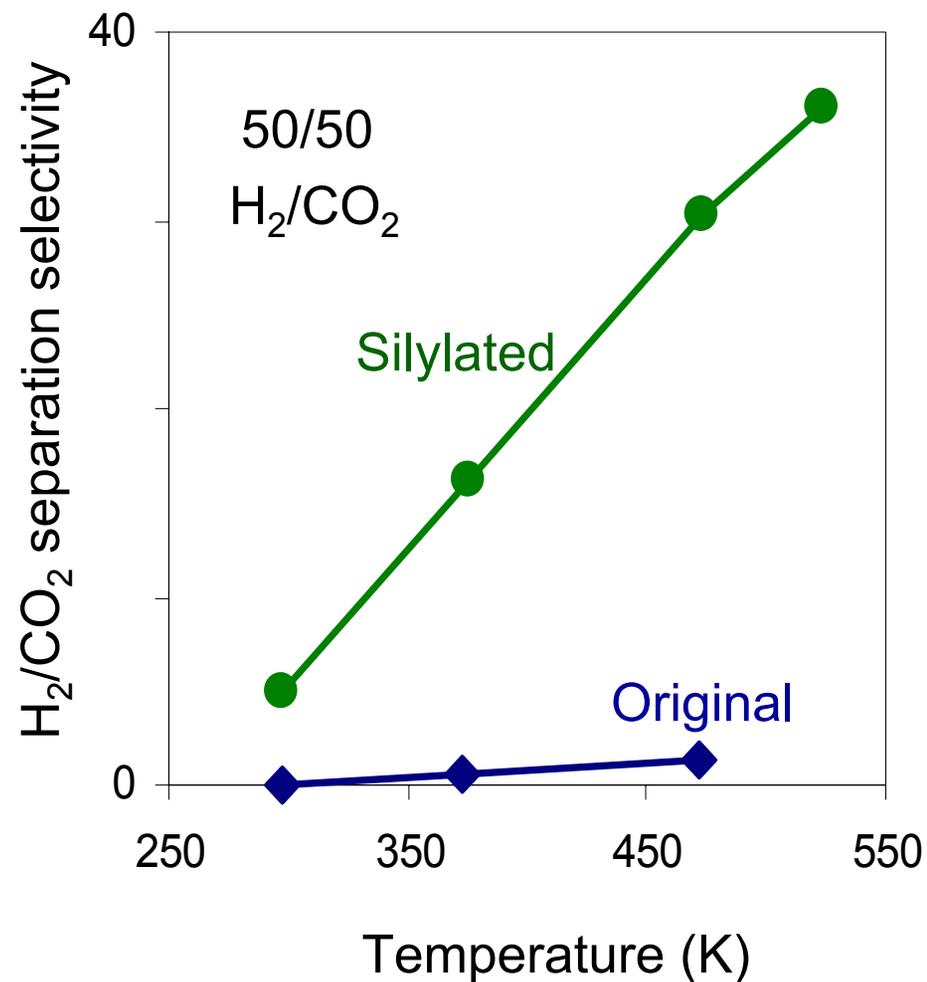
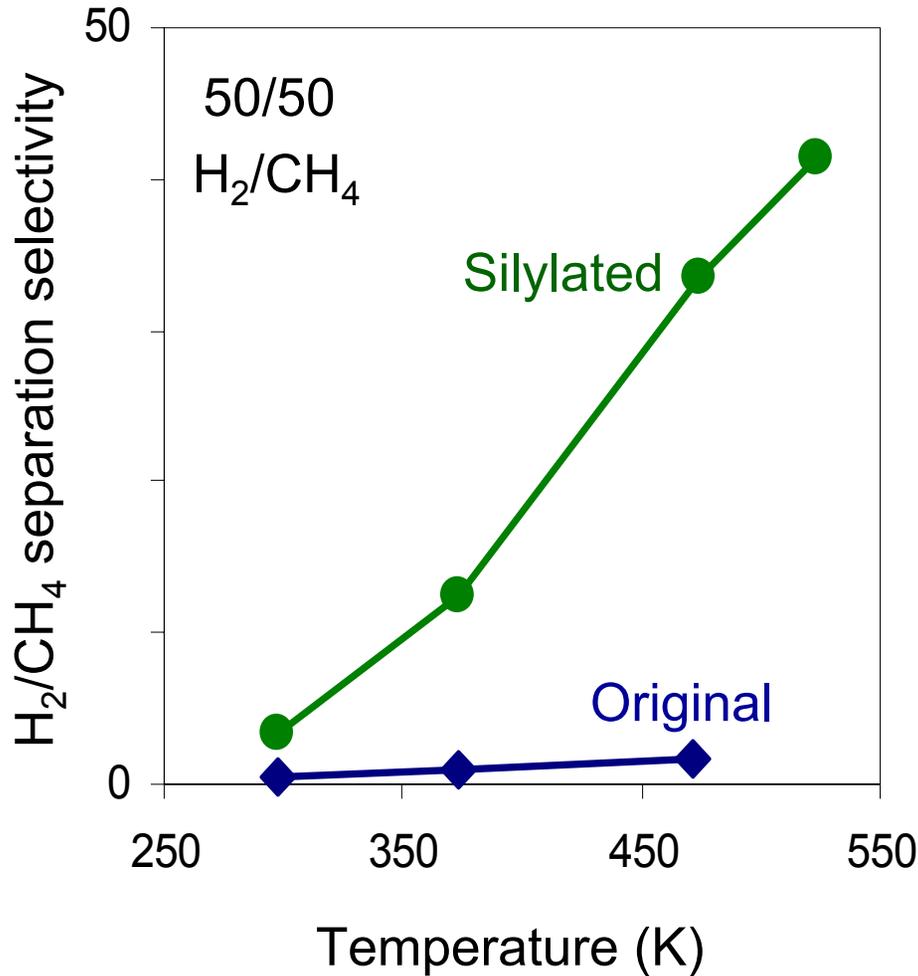
Methyl-diethoxy-silane fits into MFI pores



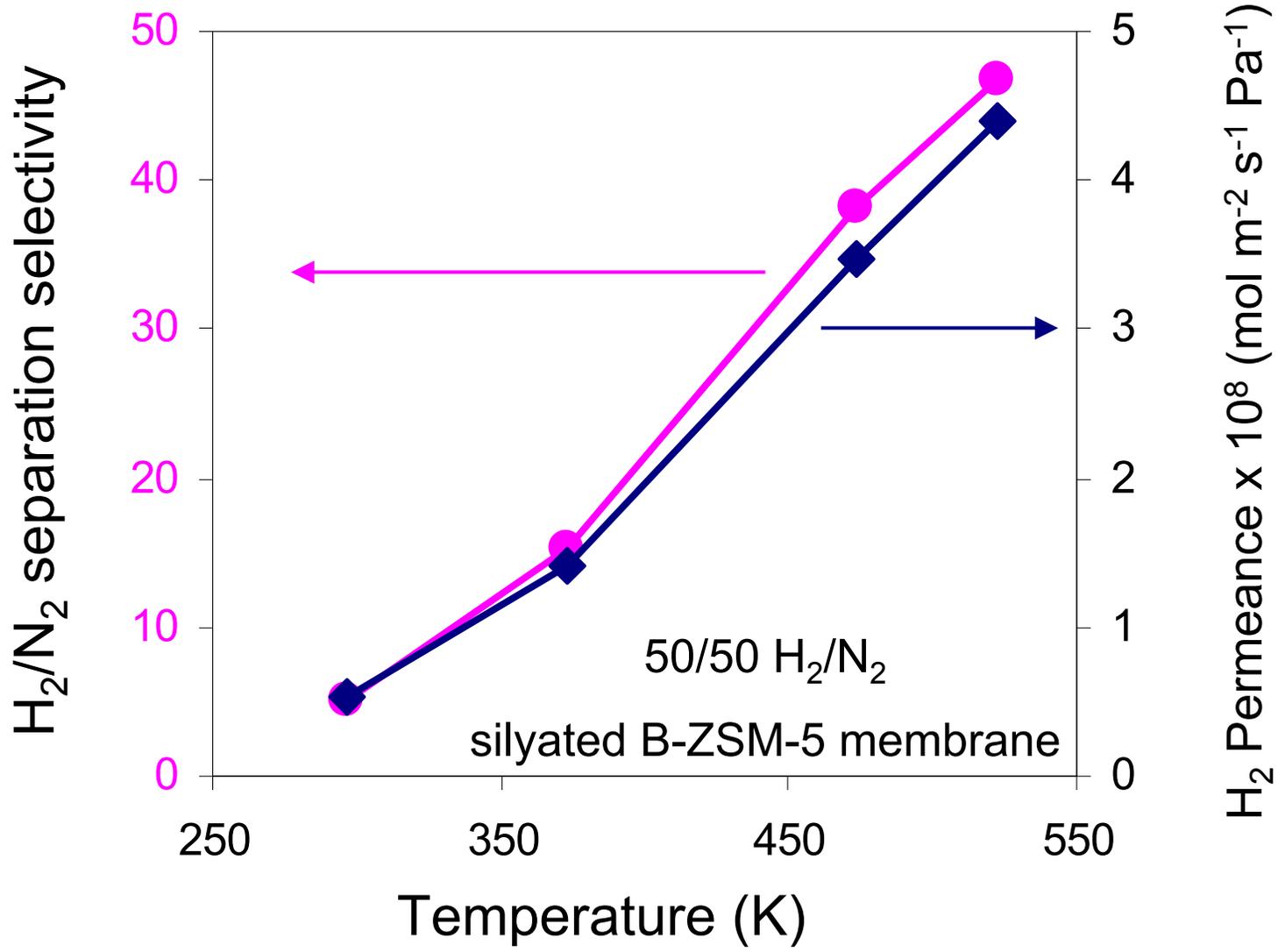
Silylation decreased H₂ permeance through a B-ZSM-5 membrane



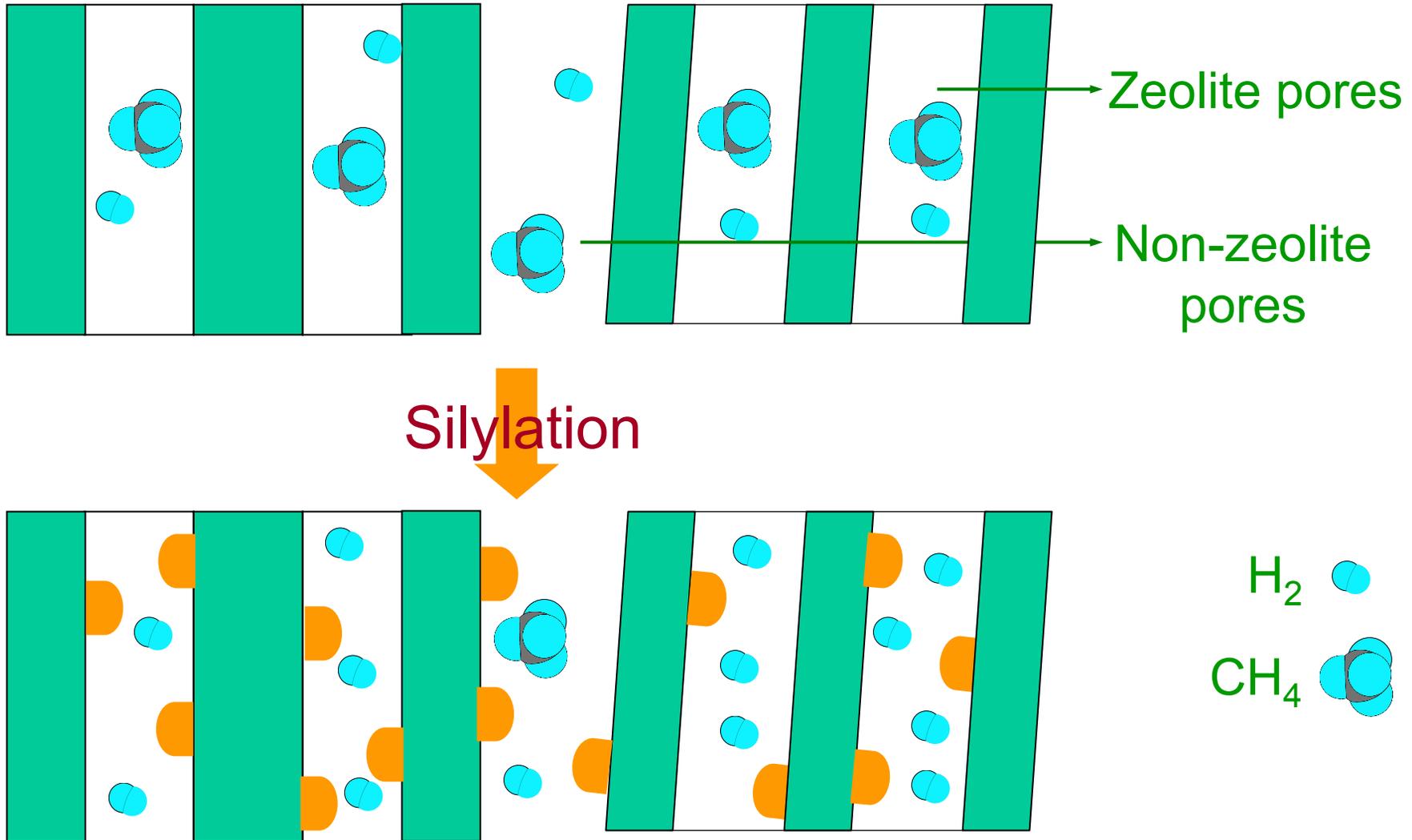
Silylation increased H₂ separation selectivity of a B-ZSM-5 membrane



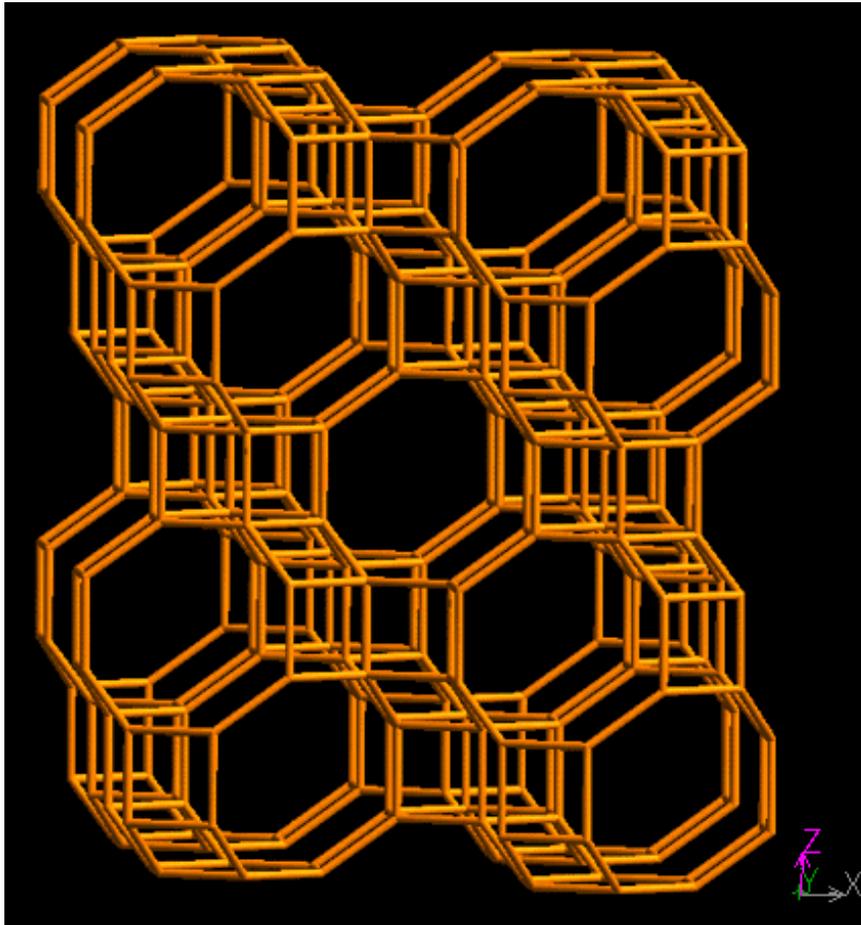
H₂ permeance and selectivity also ↑ with ↑ temperature for H₂/N₂ mixture



Silylation partially blocked MFI zeolite pores

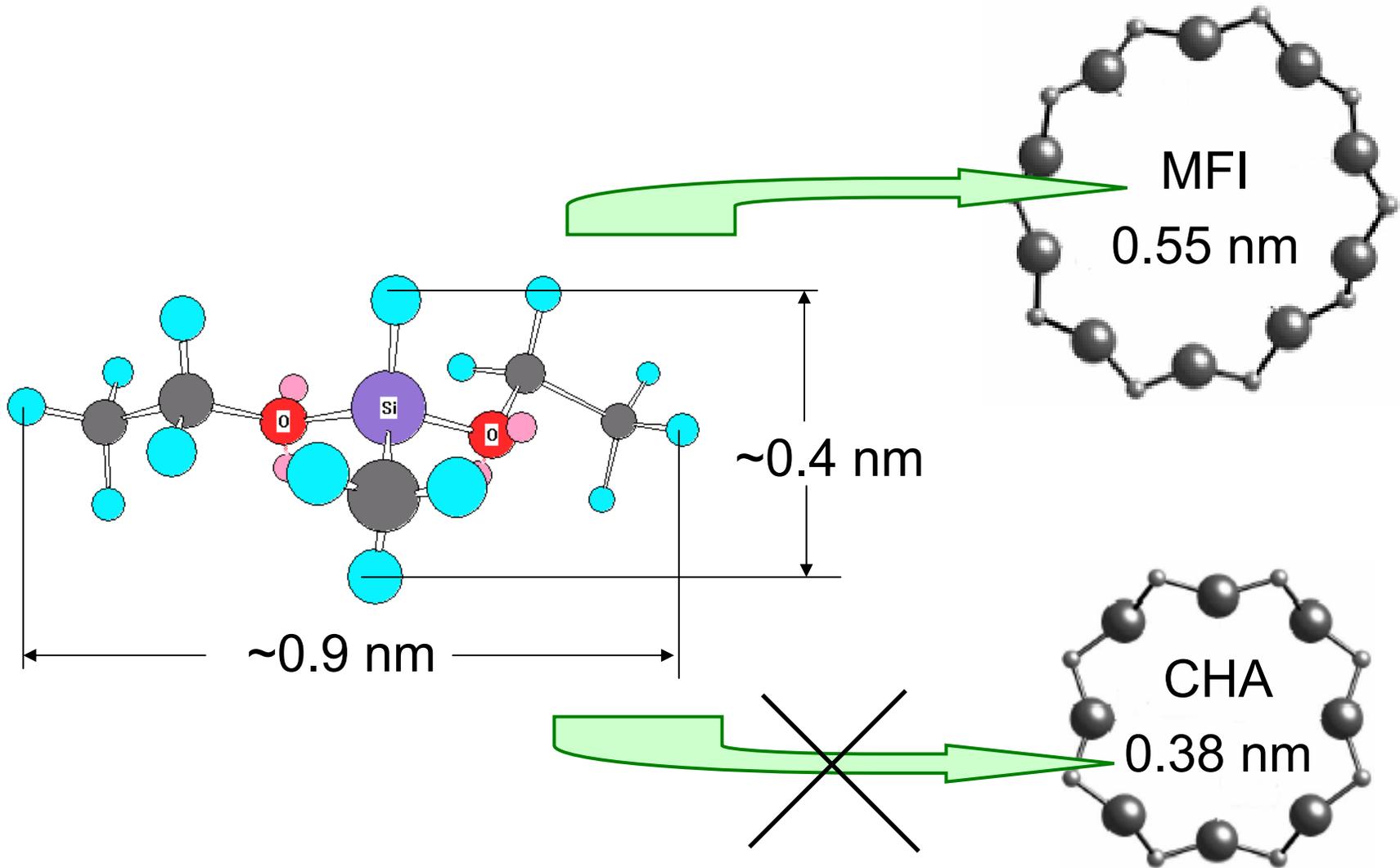


CHA zeolite membranes have smaller pores

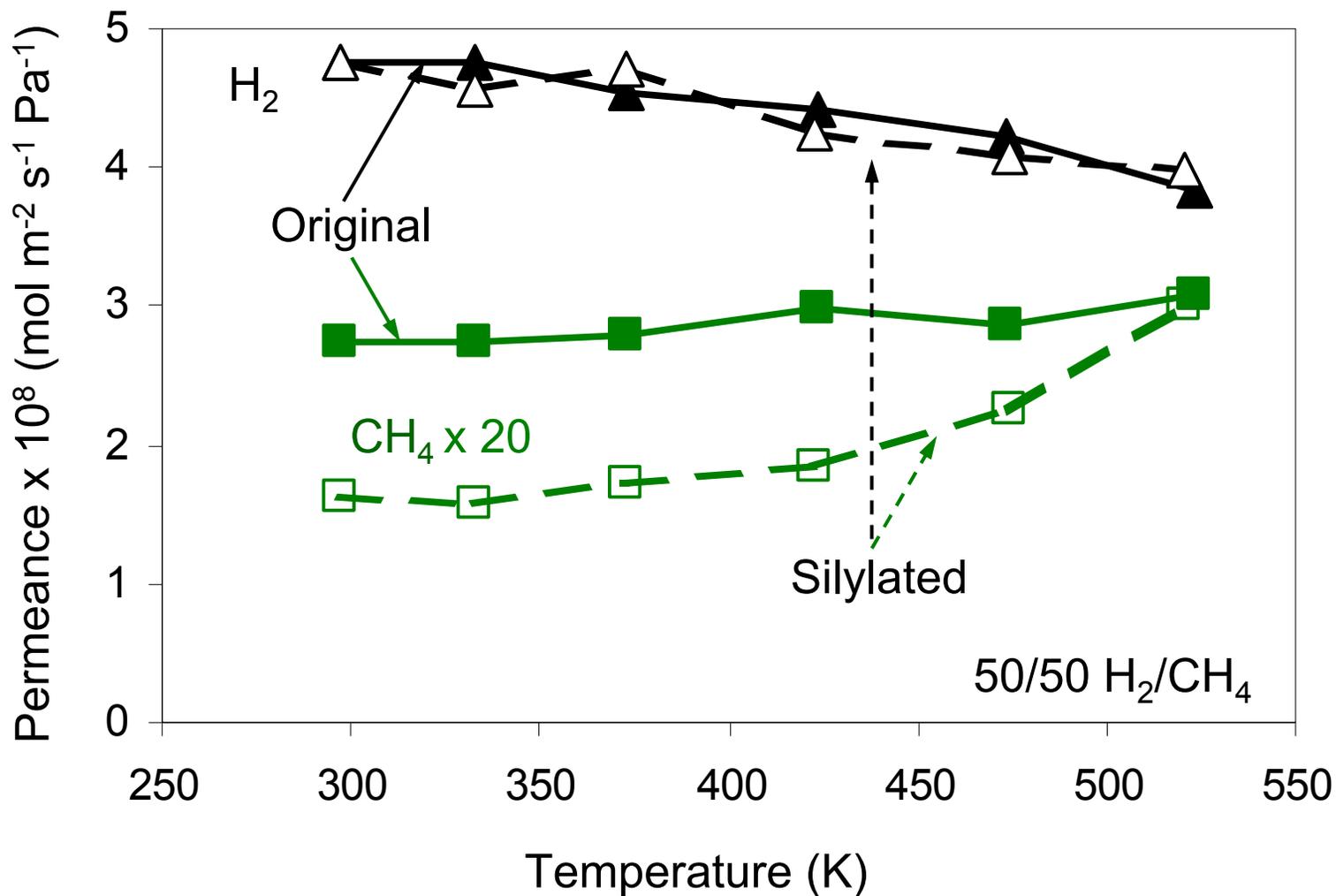


- CHA (e.g., SAPO-34)
 - 8-membered ring
 - Pore size ~ 0.38 nm
 - Separate H_2 from CH_4
- Silylation on CHA
 - Eliminate defects
 - Better H_2 separation

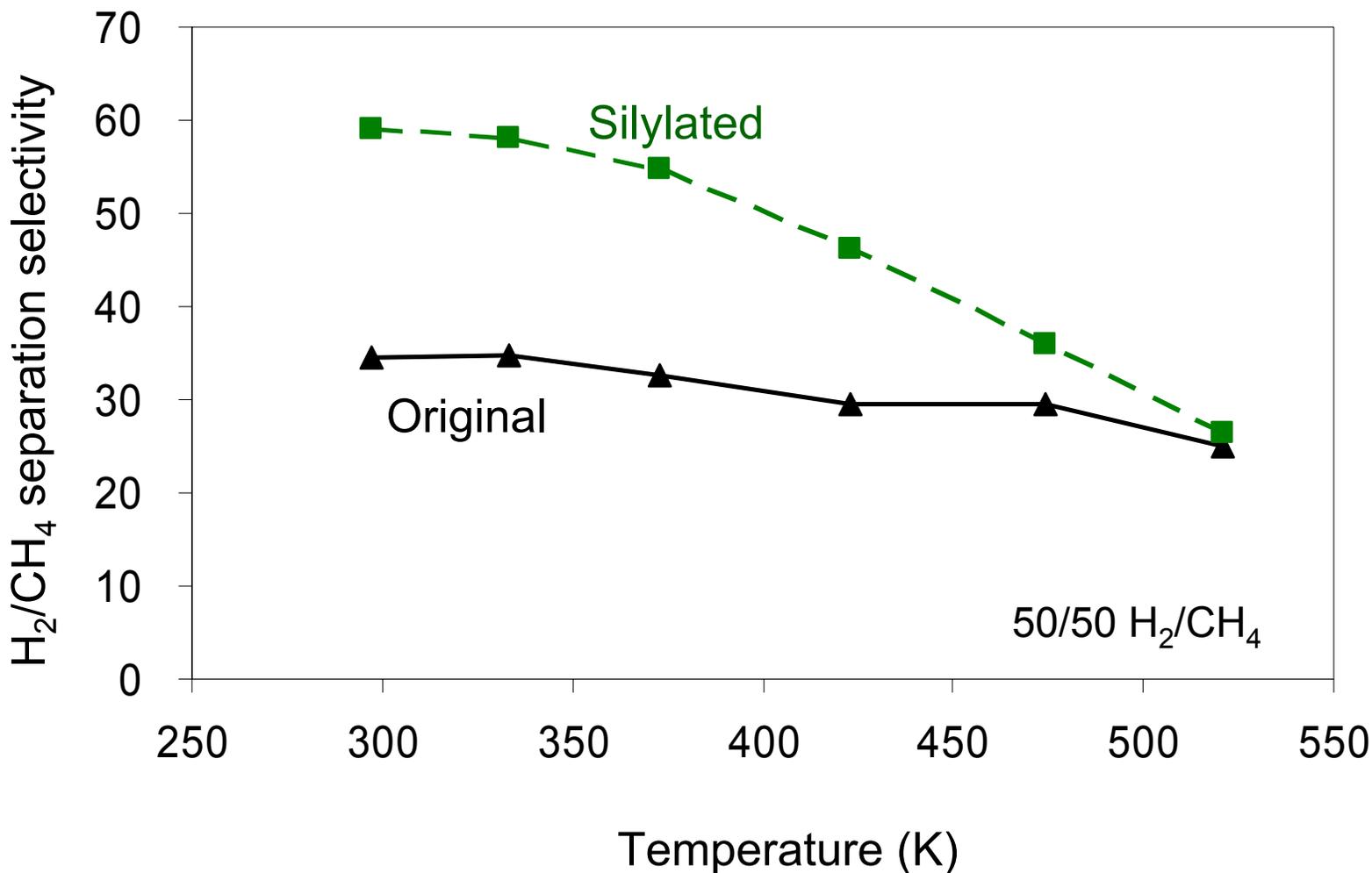
Methyl-diethoxy-silane not fit into CHA pores



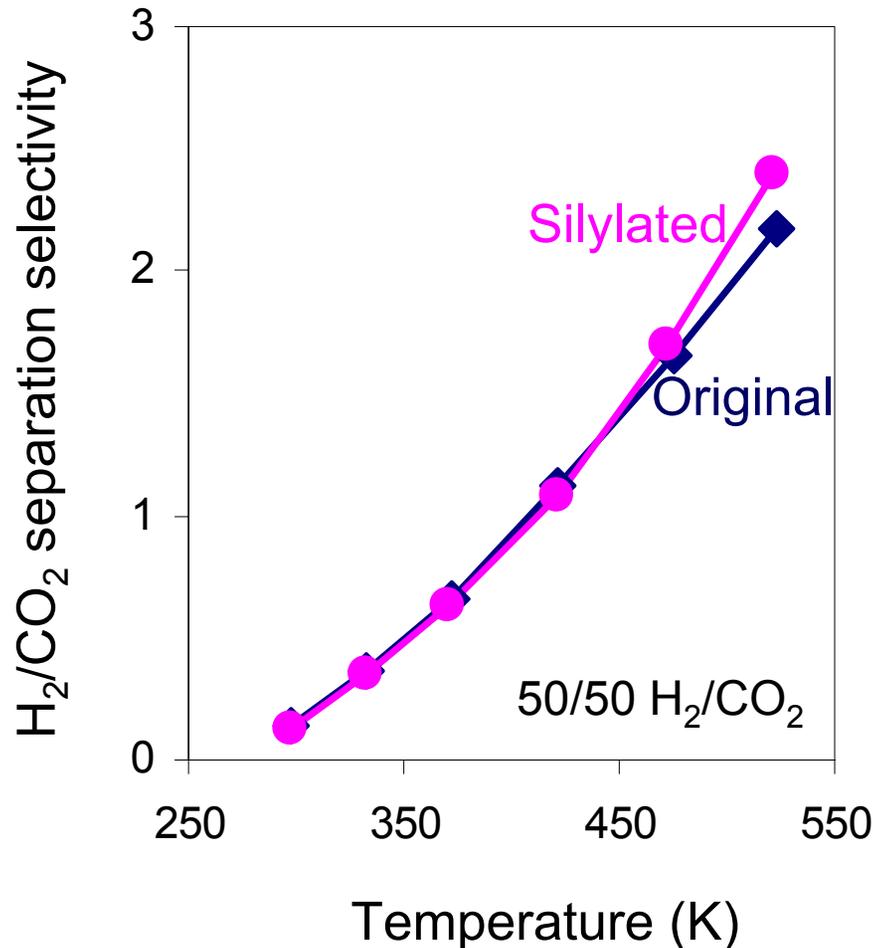
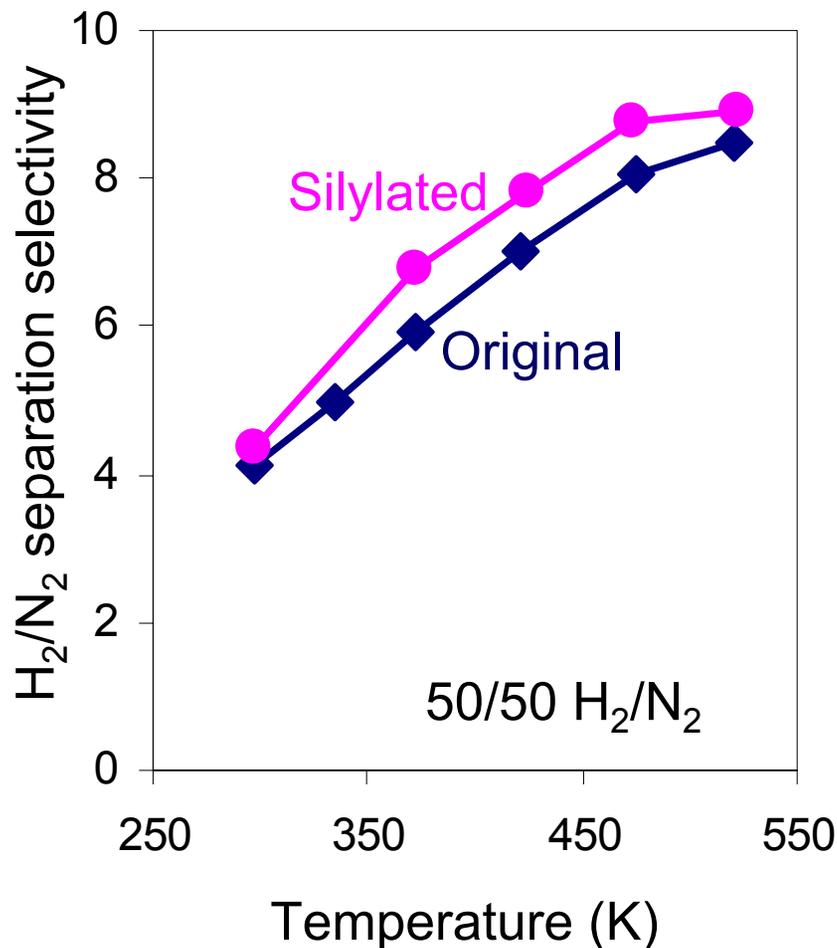
Silylation decreased CH_4 but not H_2 permeance through a SAPO-34 membrane



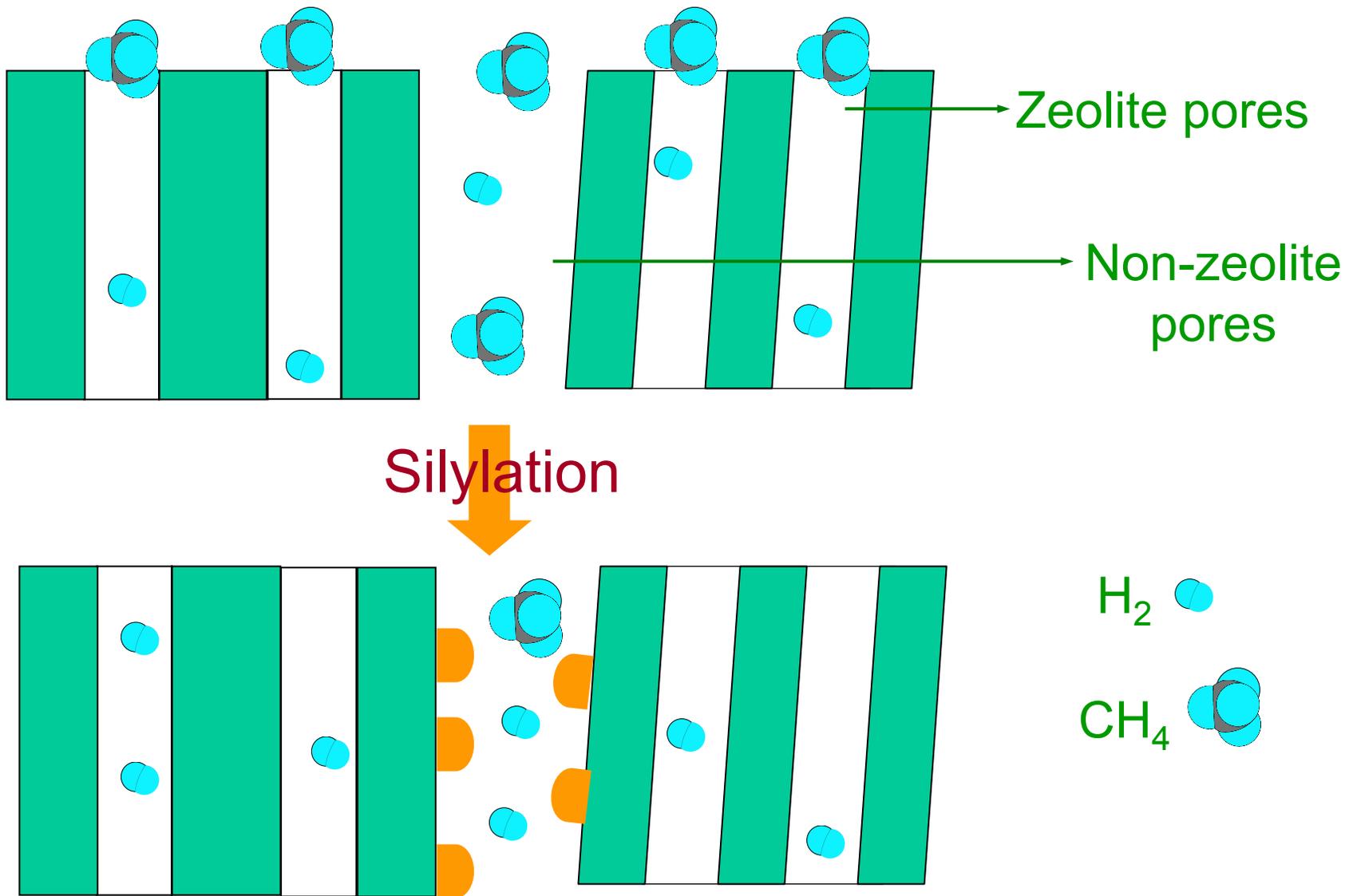
Silylation increased H₂/CH₄ separation selectivity of a SAPO-34 membrane



Silylation not change H_2/N_2 and H_2/CO_2 separation selectivity of a SAPO-34 membrane



Silylation only blocked non-zeolite pores of of CHA membranes



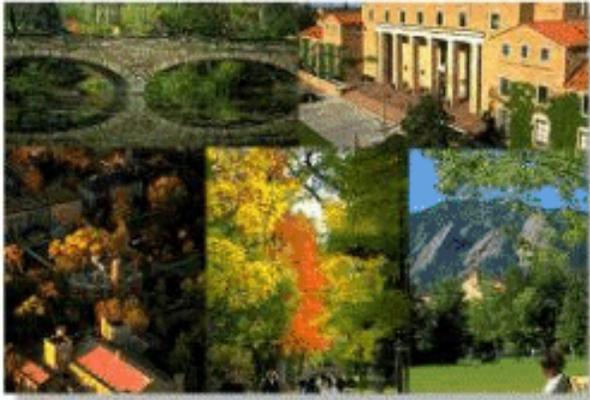
Silylation on zeolite membranes

- Partially blocked MFI membrane pores
 - H₂ selectivity increased
 - H₂ permeance decreased
 - H₂ permeance and selectivities increased with temperature
- Blocked non CHA pores in SAPO-34 membranes
 - H₂ permeance not affected
 - H₂/CH₄ selectivity increased
 - H₂/CO₂, H₂/N₂ selectivities unchanged

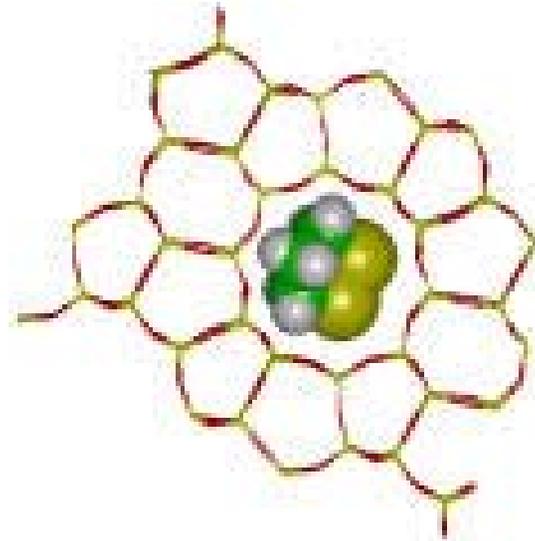
Future work on MFI membranes

- Separate at higher temperatures
 - expect higher selectivities and permeances
 - H₂/CO separation
- Use more realistic conditions
 - H₂/CO₂/CO/CH₄/H₂O mixture
 - Higher pressure
- Increase H₂ flux in silyated membranes
 - Thinner initial membranes (seeding)
 - Masking to silylate only part of membrane
- Modify silyation for improved separations
 - Different compounds, conditions

Questions and Comments?

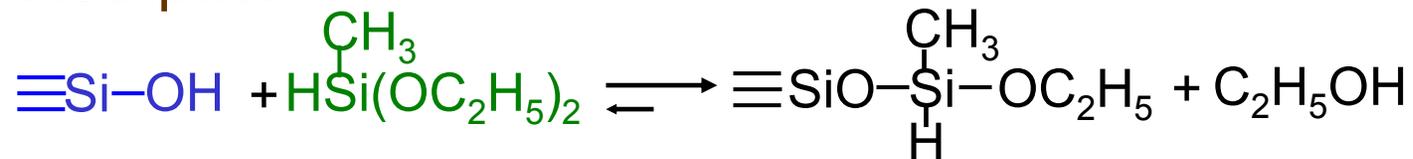


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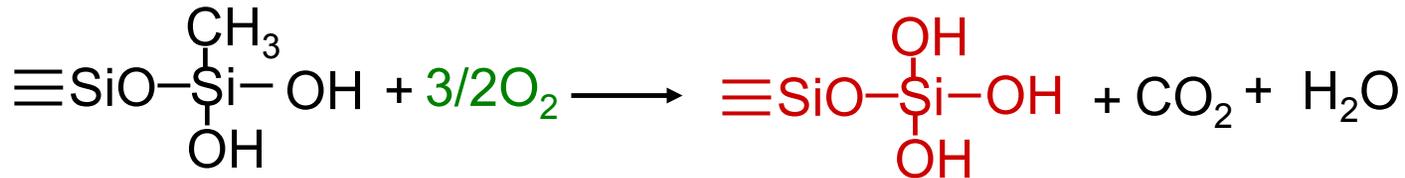
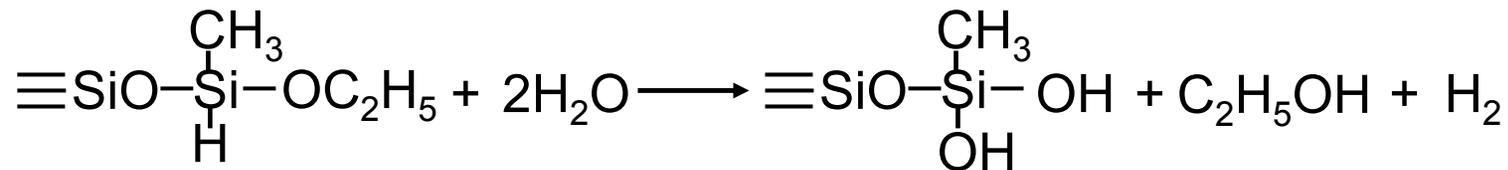


Zeolite membranes were silylated

Chemisorption:



Oxidation:



Elimination reaction:

