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Membrane Transport of *n*-butane by a Temperature Gradient

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Outline

- Motivation
- Method
- System
- Results
- Conclusion

Acknowledgment

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Motivation

- Study thermal influence on transport
- Transport properties in micro porous materials
- Transport properties across surfaces
- Coupling of heat and mass transport (NET)

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Model method

- Molecular dynamics simulations
  - Adsorption – Langmuir
  - Sampling in equilibrium
  - Deterministic
- Non-equilibrium Molecular dynamics simulations
  - Gradients in temperature and concentration
  - Steady-state transport

Newton's equations of motion

$$\mathbf{F}_i = -\frac{\partial V}{\partial \mathbf{r}_i}$$

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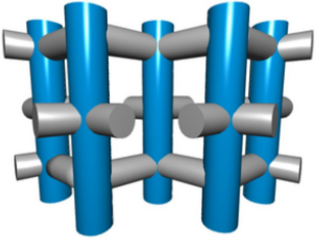
System

- Perfect zeolite: silicalite-1 ($[SiO_2]_{96}$)
- Zig-zag and straight channels (5.5 Å diam.)
- Channels interconnected
- All-Atom model – flexible grid



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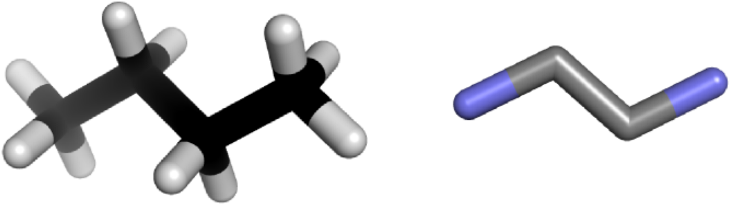
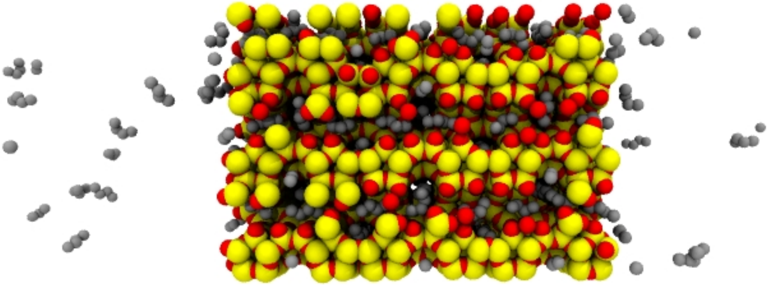
System cont'd

- *n*-butane
- United atoms model
- Stretching – Bending – Torsion



System cont'd

- Semi-infinite membrane (least image convention)
- Gas phases on both sides
- Different surface structures, and temperatures



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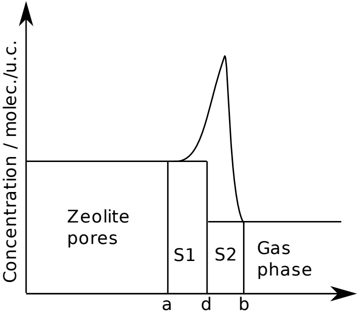
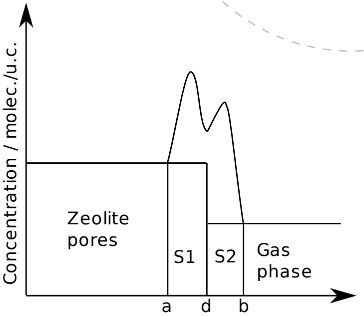
Surface


Concentration / molec./u.c.
Zeolite pores
S1
S2
Gas phase
a
d
b


Concentration / molec./u.c.
Zeolite pores
S1
S2
Gas phase
a
d
b

Interface transport equations

Describing the transport across the surface:

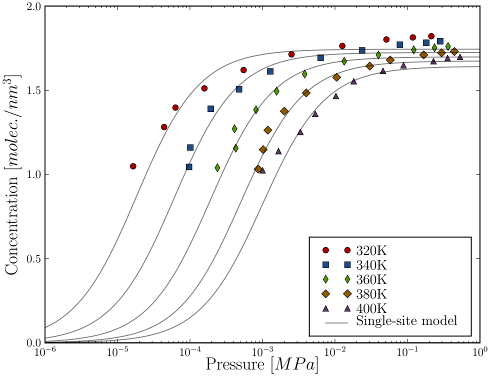
$$\Delta_{z,g} T = -\frac{1}{\lambda^s} \left(J'^g_q - q^{*s,r}_b J_b \right) \tag{1}$$

$$\frac{1}{T^z} \Delta_{z,g} \mu_{b,T} \left(T^z \right) = -\frac{q^{*s,r}_b}{T^z T^g} \Delta_{z,g} T - R^{s,r}_{bb} J_b \tag{2}$$

Derived from the entropy production at the interface.

[Inzoli, I., Kjelstrup, S., Bedeaux, D. and Simon, J.-M. *Microporous and Mesoporous Materials*, 125 (2009) 112–125.]

Equilibrium results



Langmuir adsorption

$$c_{pores} = \frac{c_{sat} \cdot K \cdot p}{1 + K \cdot p}$$

$$\Delta H^0_{ads} = -54 \pm 1\mathrm{kJ/mol}$$

$$c_{sat} \approx 1.75\mathrm{molec./nm}^3$$

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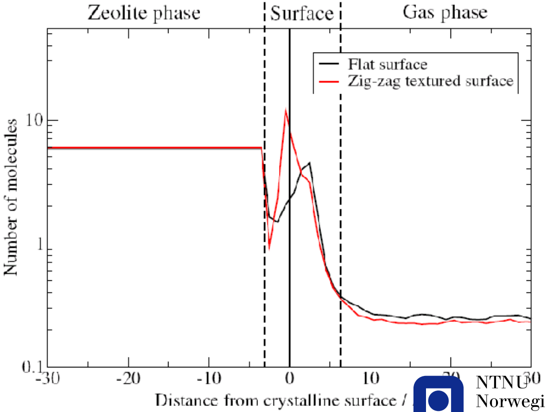
Surface excess

- Surface is rate-limiting for both heat and mass transfer (negative surface excess)



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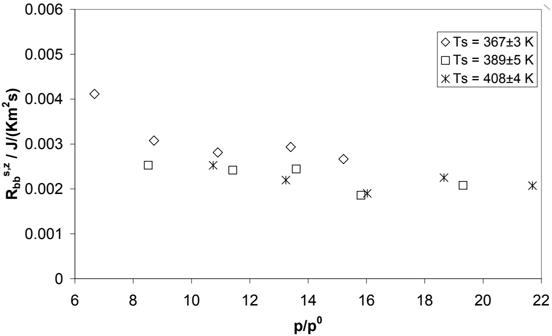
Surface resistance to mass transport

- Not very dependent on gas pressure.



$$\frac{1}{T^z}\Delta_{z,g}\mu_{b,T}\left(T^z\right) = -\frac{q_b^{*s,r}}{T^z T^g}\Delta_{z,g}T - R_{bb}^{s,r} J_b$$

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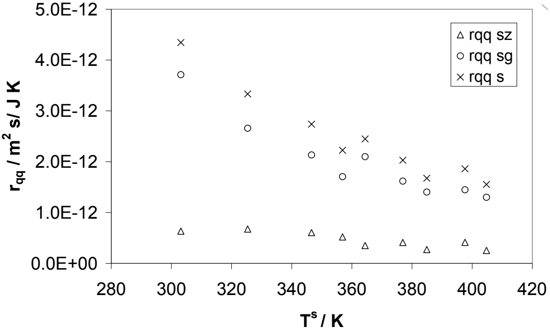
Surface resistance to heat transport

- Gas side dominates the resistance.



$$\Delta_{z,g}T = -\frac{1}{\lambda^s}\left(J_q'^g - q_b^{*s,r}J_b\right)$$

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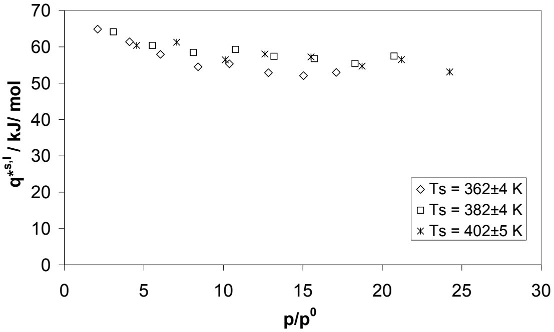
Coupling

- Large values for Heat of transfer, $q_b^{*s,r}$
- Significantly larger than for ΔH^0_{ads}



$$\frac{1}{T^z}\Delta_{z,g}\mu_{b,T}\left(T^z\right) = -\frac{q_b^{*s,r}}{T^zT^g}\Delta_{z,g}T - R_{bb}^{s,r}J_b$$

$$\Delta_{z,g}T = -\frac{1}{\lambda^s}\left(J_q^{\prime g} - q_b^{*s,r}J_b\right)$$

Conclusion

- A membrane surface can well be rate-limiting to transport of heat and mass.
- Proper descriptions of heat and mass transport must take into account the coupling between the two.

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Thank you for your attention!

Questions?

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