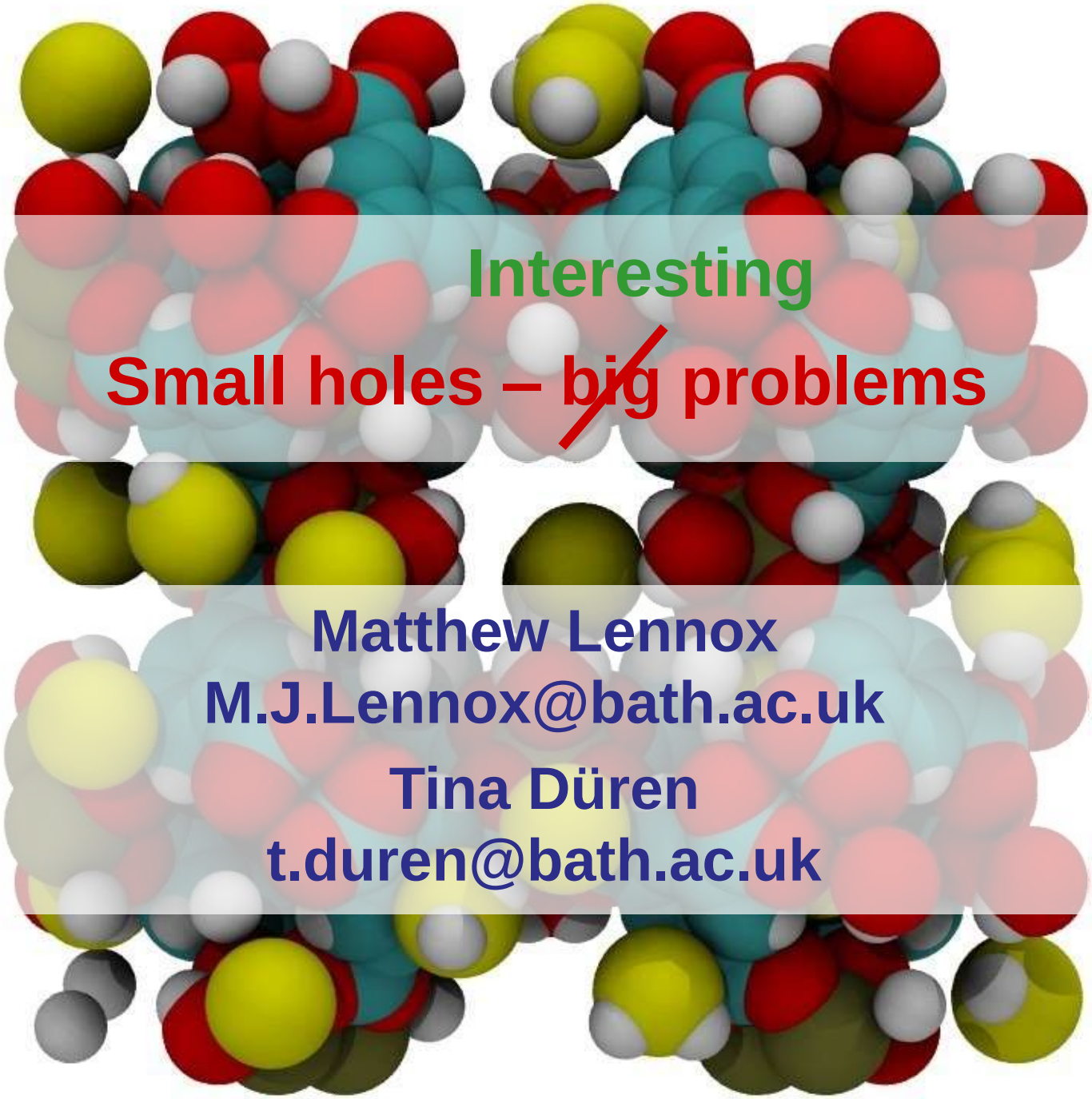

Interesting

Small holes – ~~big~~ problems

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What is adsorption?

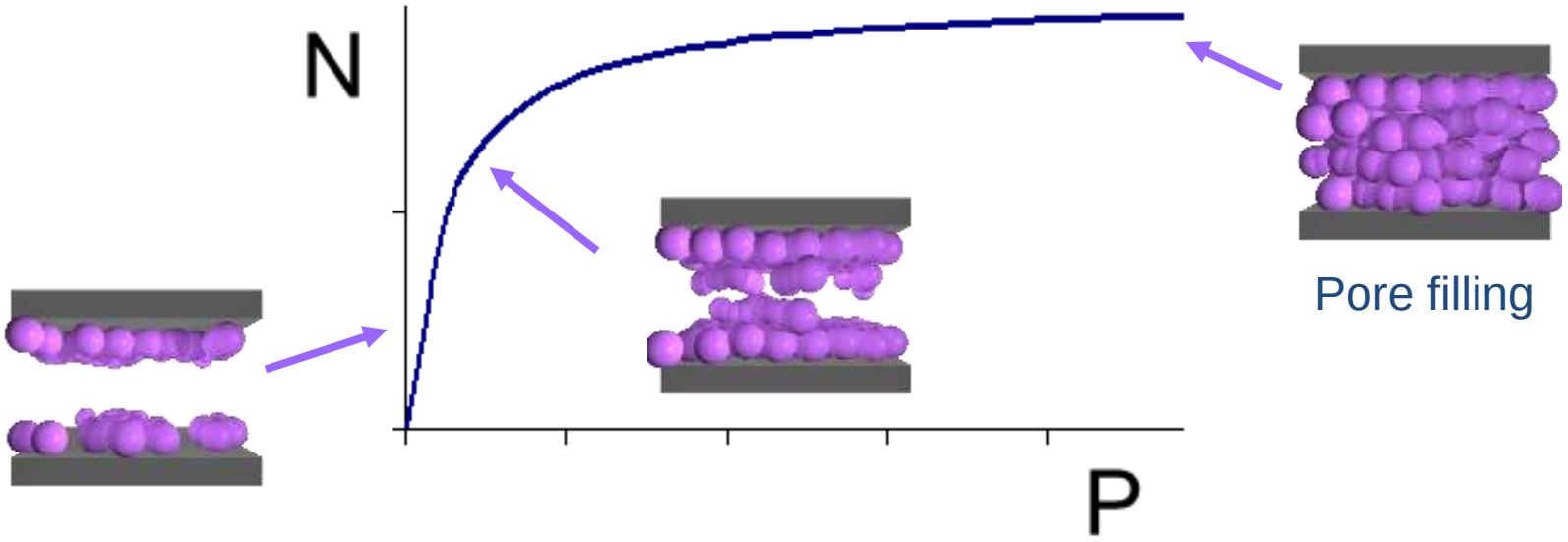
Adsorption is the adhesion of atoms, ions, or molecules from a gas, liquid, or dissolved solid to a surface.

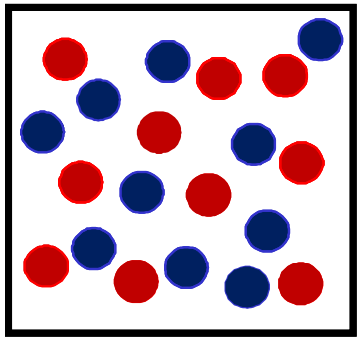
Adsorption
Surface process

Absorption
Bulk process

http://teaching.ust.hk/~ceng510/notes/CENG511/Lecture5.pdf

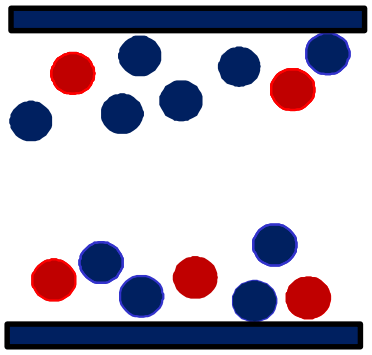
Adsorption isotherms & selectivities





y: mole fraction in bulk

$y_{red} = 0.5$



x: mole fraction in pore

$x_{red} = 2/3$

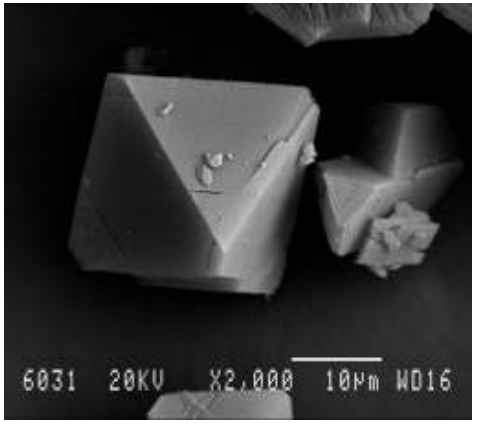
$$\text{Selectivity} = \frac{x_1/x_2}{y_1/y_2}$$

$S_{blue\ /\ red} = 2$

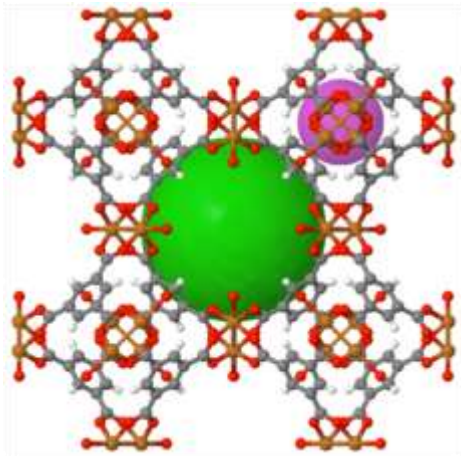
Length scales : from adsorption columns to individual molecules in the solid

m

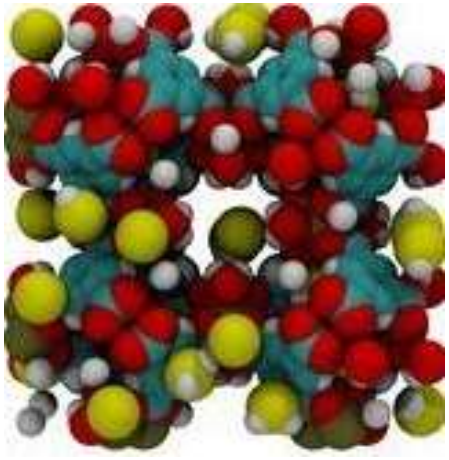
mm



µm



nm = 10^{-9} m



CO_2 & SO_2 in CuBTC

Metal-organic frameworks – Building blocks allow tailoring for practical applications

+

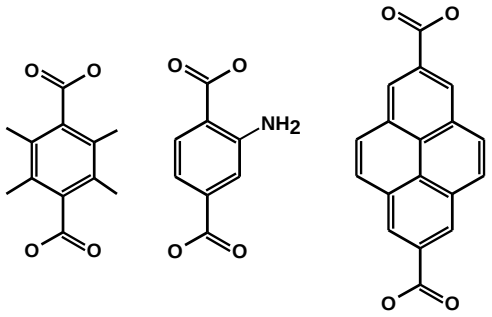
Zn

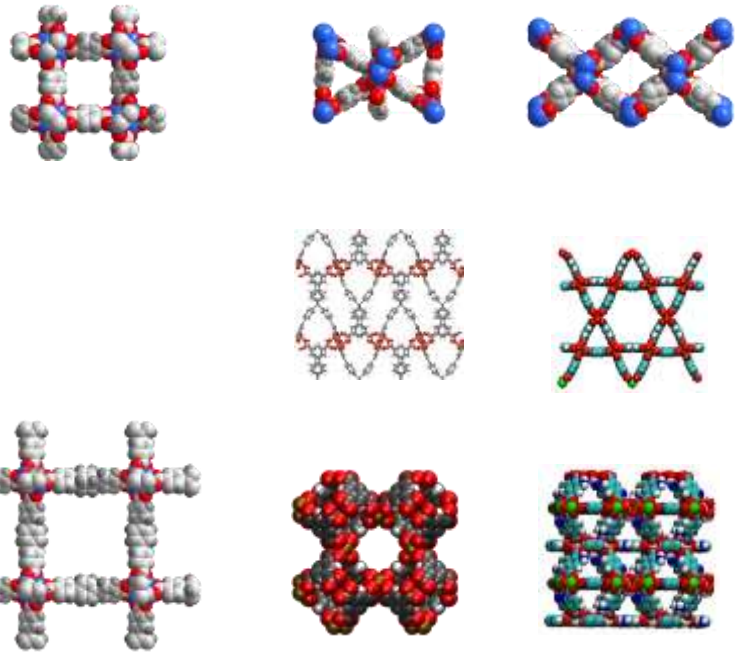
V

Cu

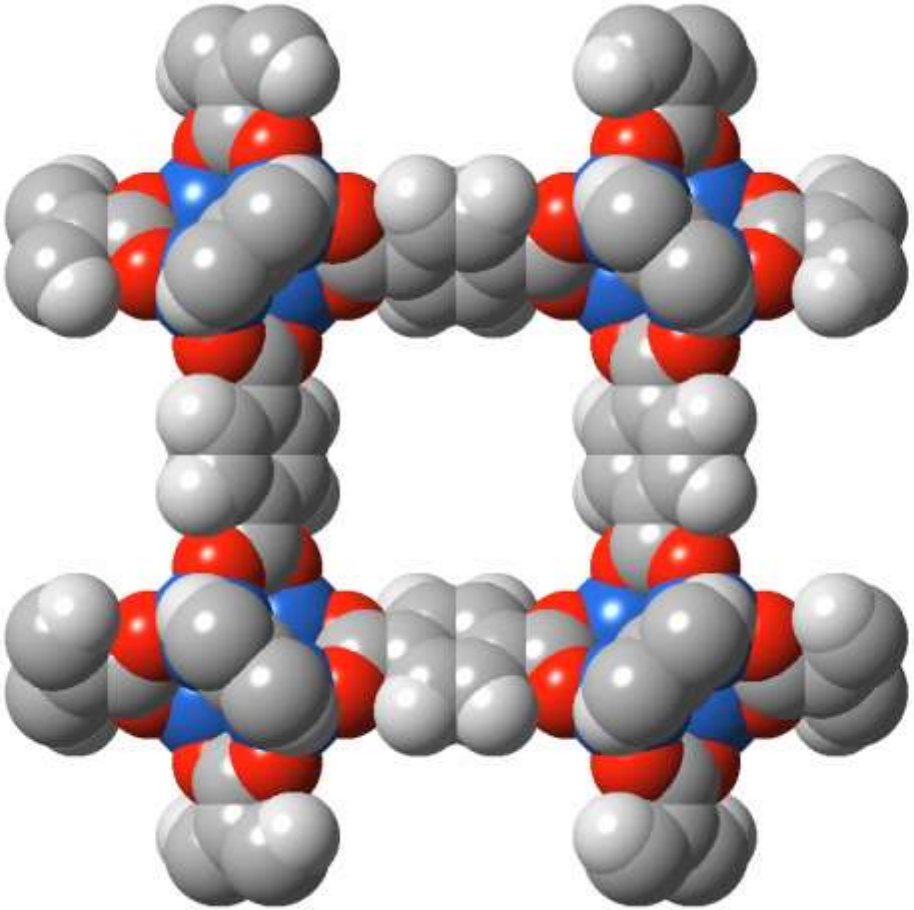
Zr

+



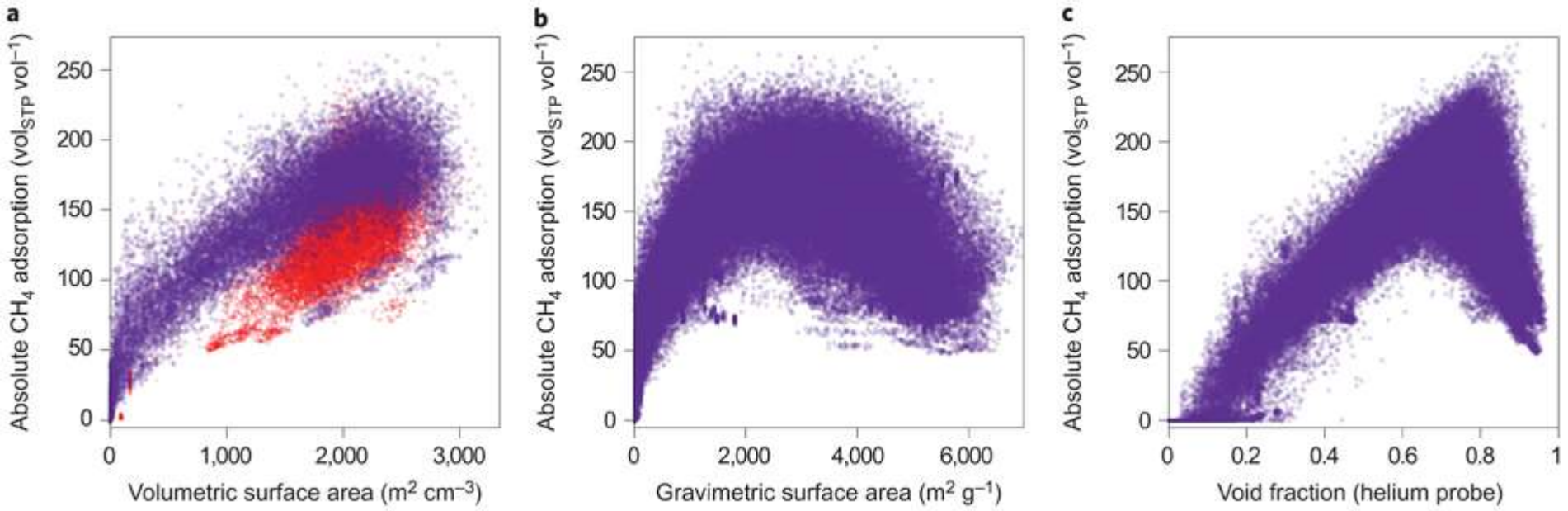


What is it like inside IRMOF-1?



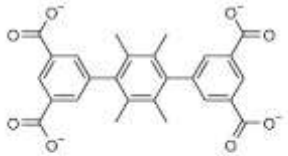
Characterisation, screening, finding applications

Computational screening of 137,953 hypothetical MOFs for methane storage

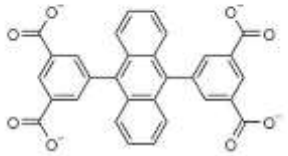


- Screening allows finding promising
  - Structures (NOTT-107 better than PCN-14)
  - Functional groups (methyl, ethyl, propyl)

NOTT-107



PCN-14

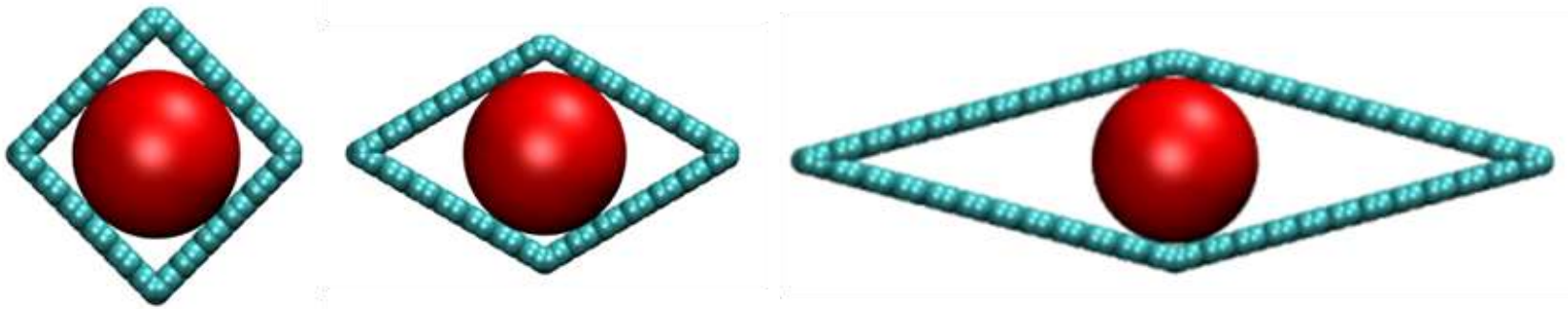


- But contain no information about how (if) structures can be synthesised.

Work from Northwestern University : C.E. Willmer et al Nature Chemistry 4, 83–89 (2012)

Geometry: size is not enough

- Pores often described by largest sphere diameter:

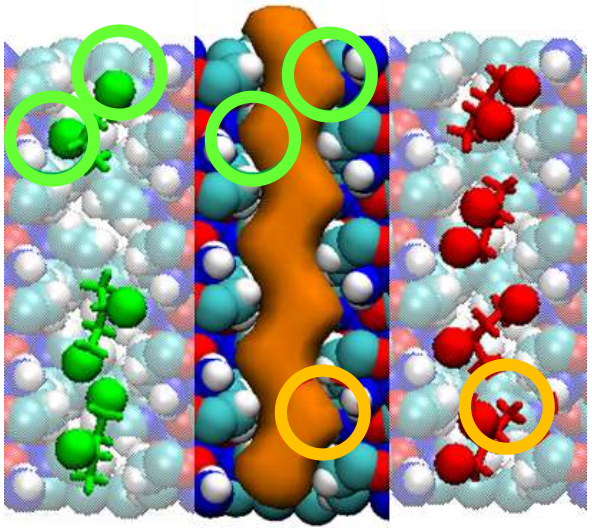


- Same ‘size’ but very different performance!

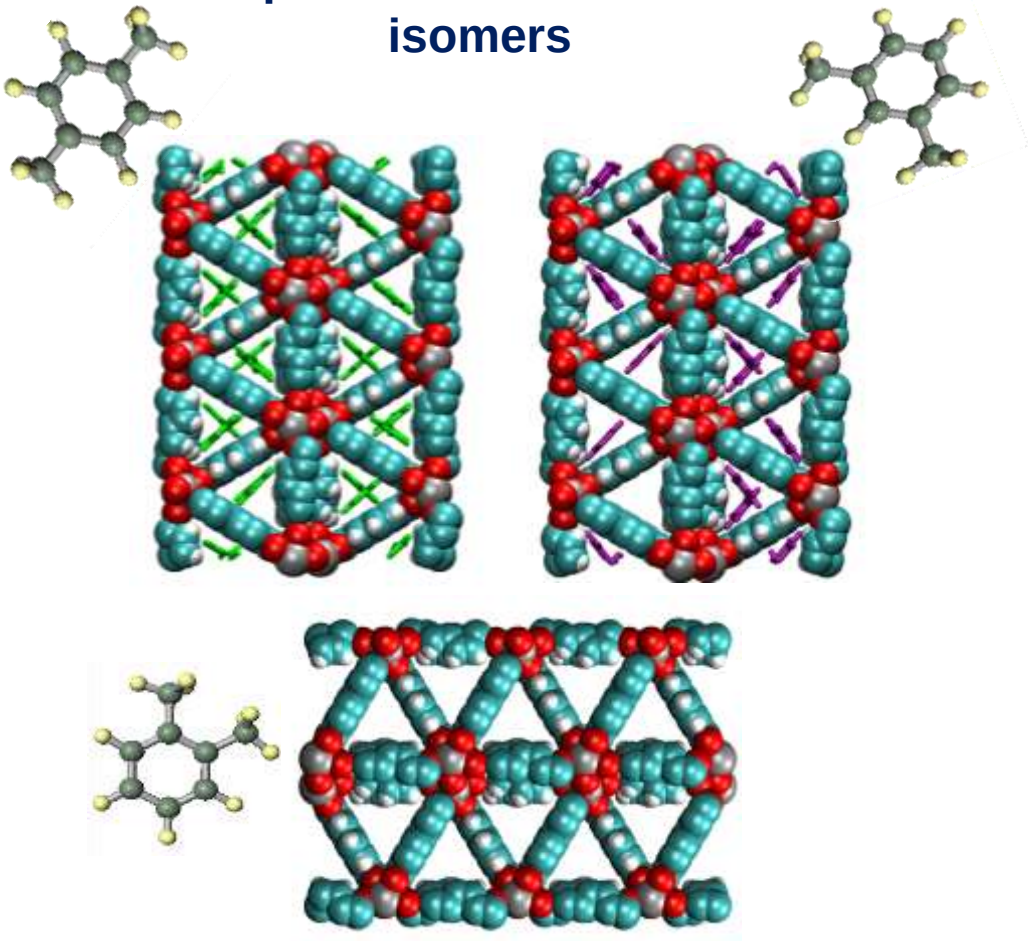
Can we quantify/characterise by shape – even for complex pore geometries?

Shape: important for (some) separations

Chromatographic separation of chiral molecules

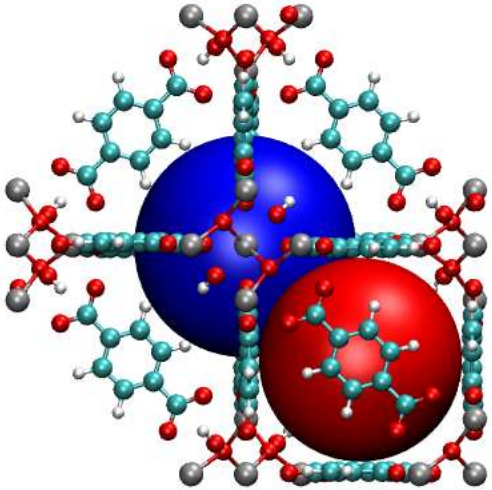


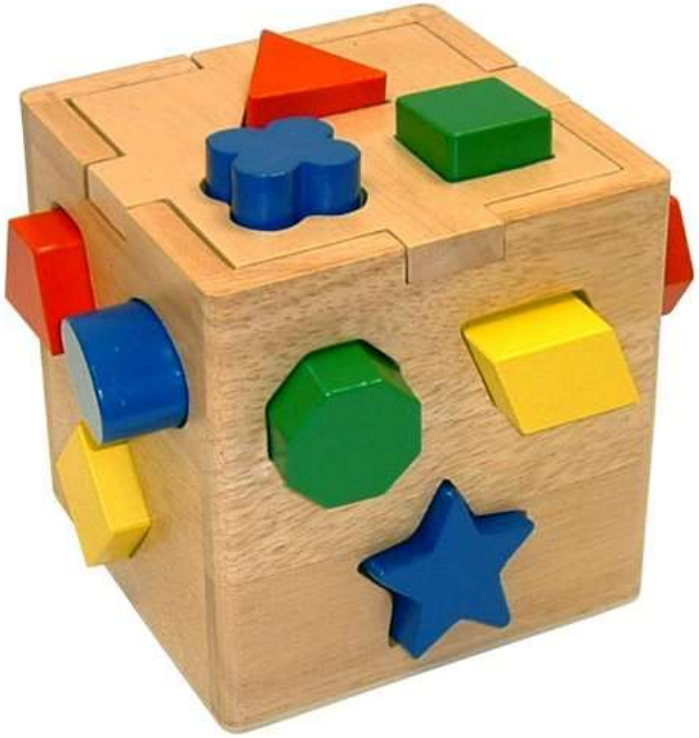
Separation of non-chiral isomers

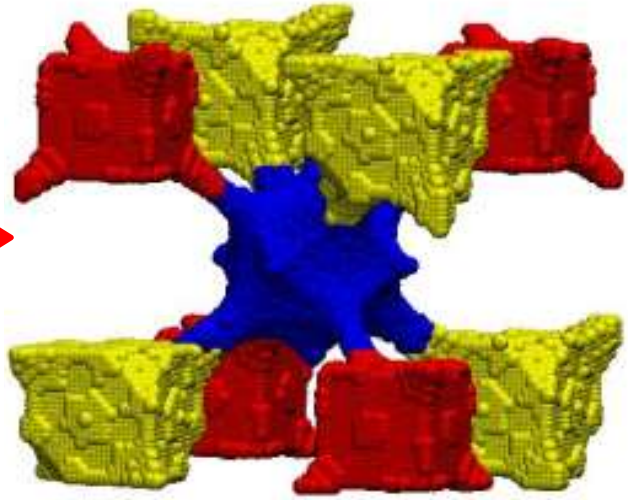


Is there a clever way of finding “similarity”?

A new MOF – what can we do with it?



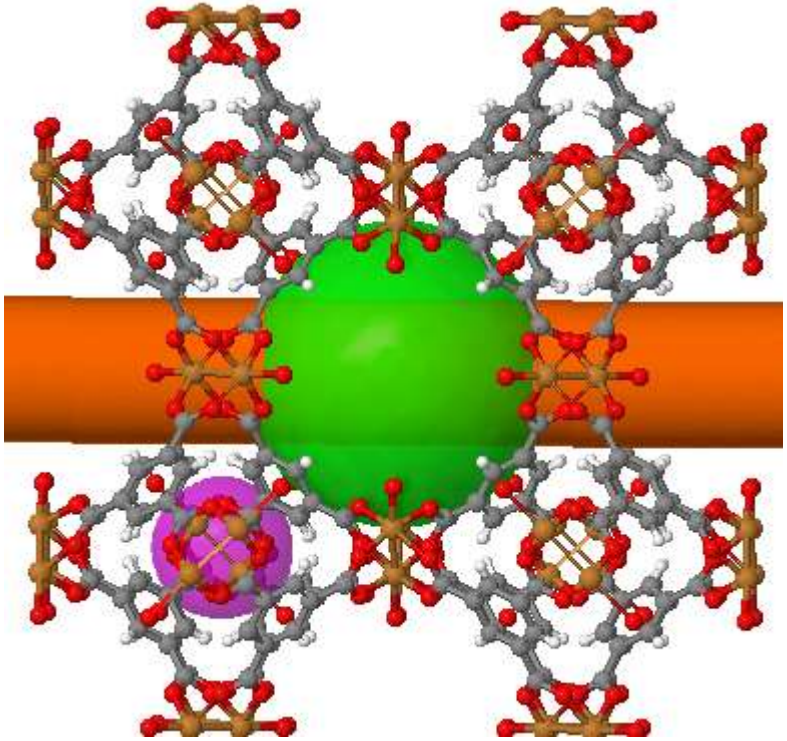




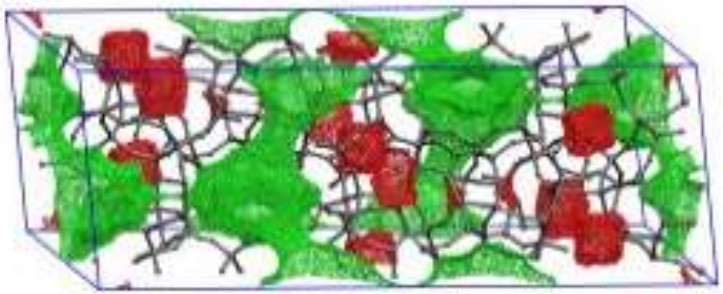
?

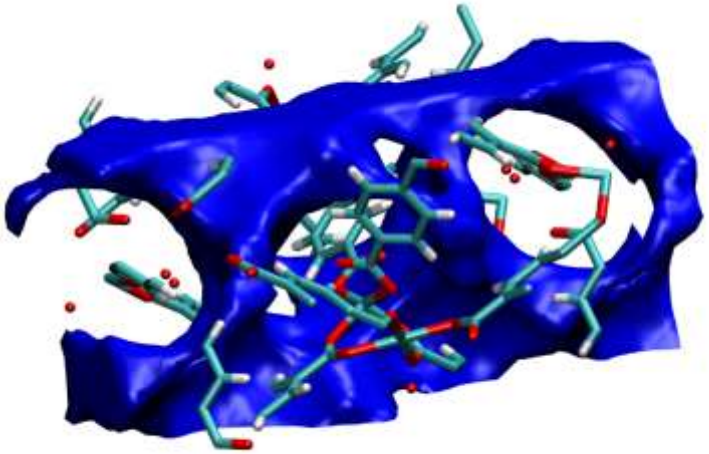
Challenge: Integrating complex geometry into screening

Simple sphere and channel representation



Simple shapes don't help with many MOFs





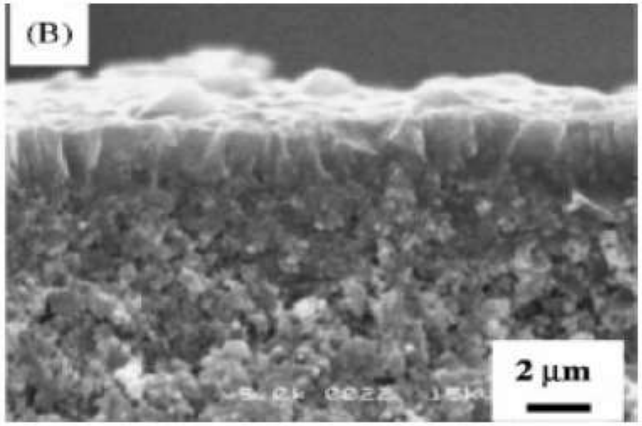
Useful things to look at

- http://www.chemtube3d.com/solidstate/MOF-home.html
  Images to play around with on the web
- http://helios.princeton.edu/mofomics/
  Numbers & images
  (java)
- http://www.maciejharanczyk.info/Zeopp/
  Numbers & images
  (Linux, Windows, Mac)
- http://www.homepages.ed.ac.uk/lsarkiso/Research.html
  Numbers
  (Linux, Windows)

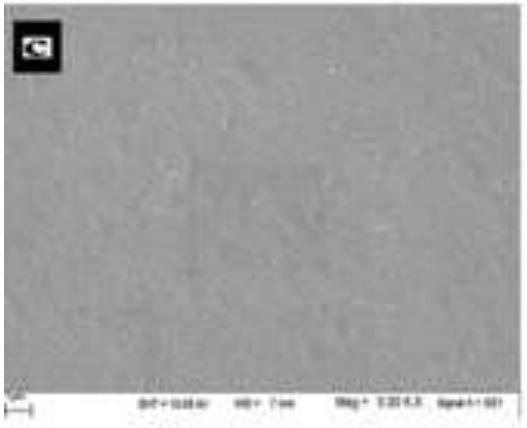
Multiscale modelling: connecting atomistic and continuum models

Mixed Matrix Membranes

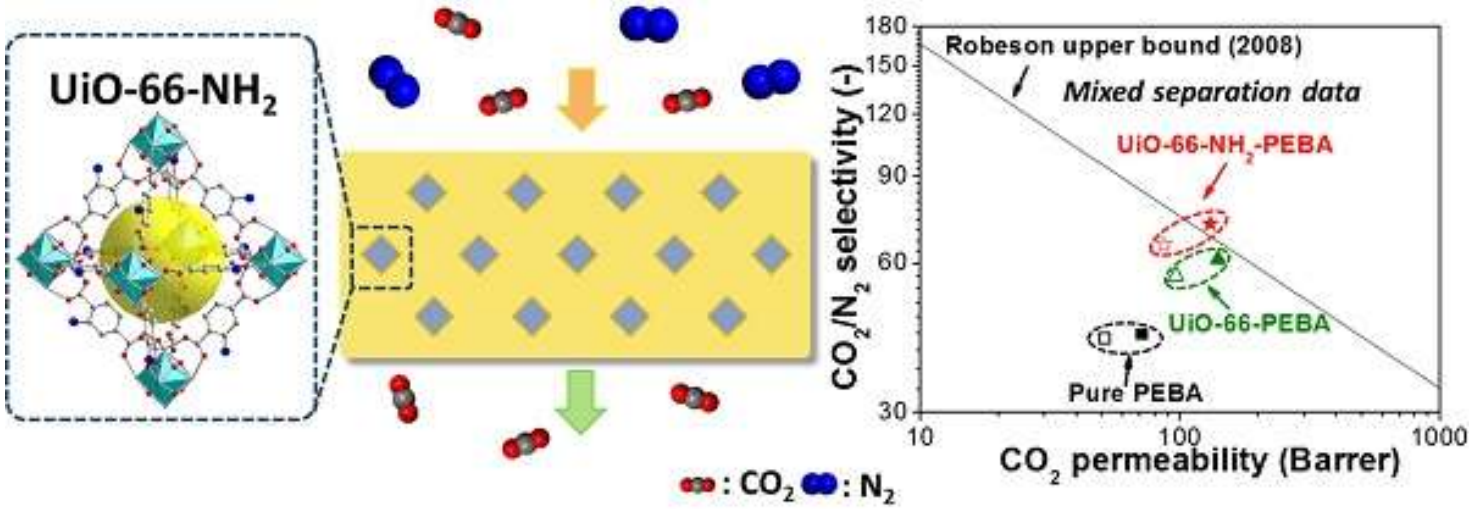
MOF membrane



Polymer membrane

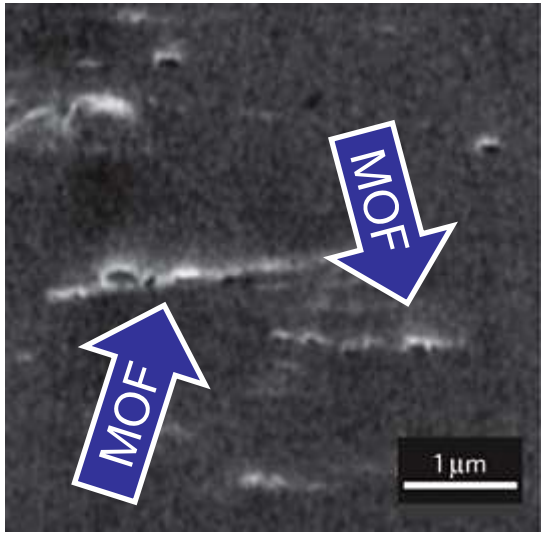


The best of both worlds: mixed matrix membranes

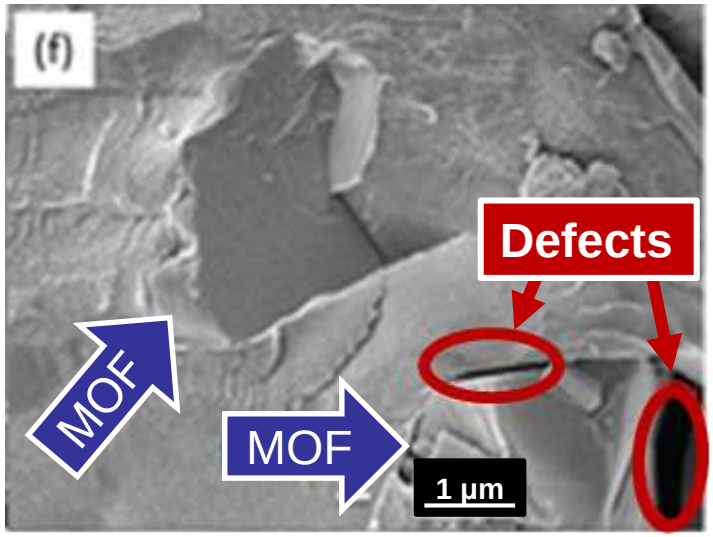


Mixed Matrix Membranes

- Detailed atomistic description for the MOF
- Bulk models for polymers
- But no good way of connecting the two
- Interfaces? Defects?



T. Rodenas et al, Nature Materials, 2014



R. Lin et al ACS Appl. Mater. Interfaces, 2014

Back-up slides

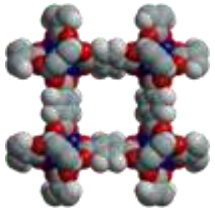
Molecular simulation

Input

e.g. model for fluids and solids
force fields to describe interactions

CH_4

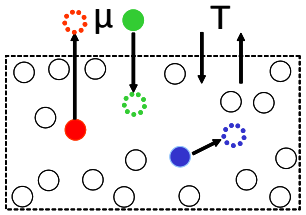
CO_2



IRMOF-1

Simulation methods based on statistical mechanics

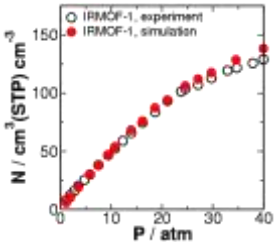
e.g. Monte Carlo
Molecular Dynamics



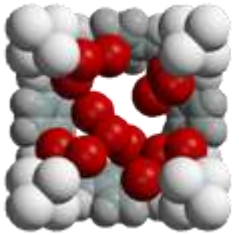
GCMC simulation

Output

e.g. adsorption isotherms
diffusion coefficients
detailed picture on molecular level

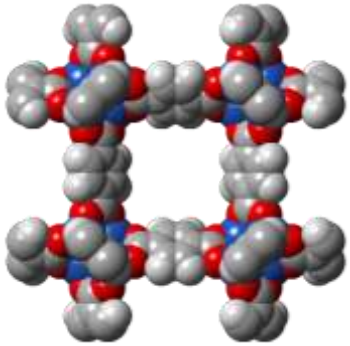


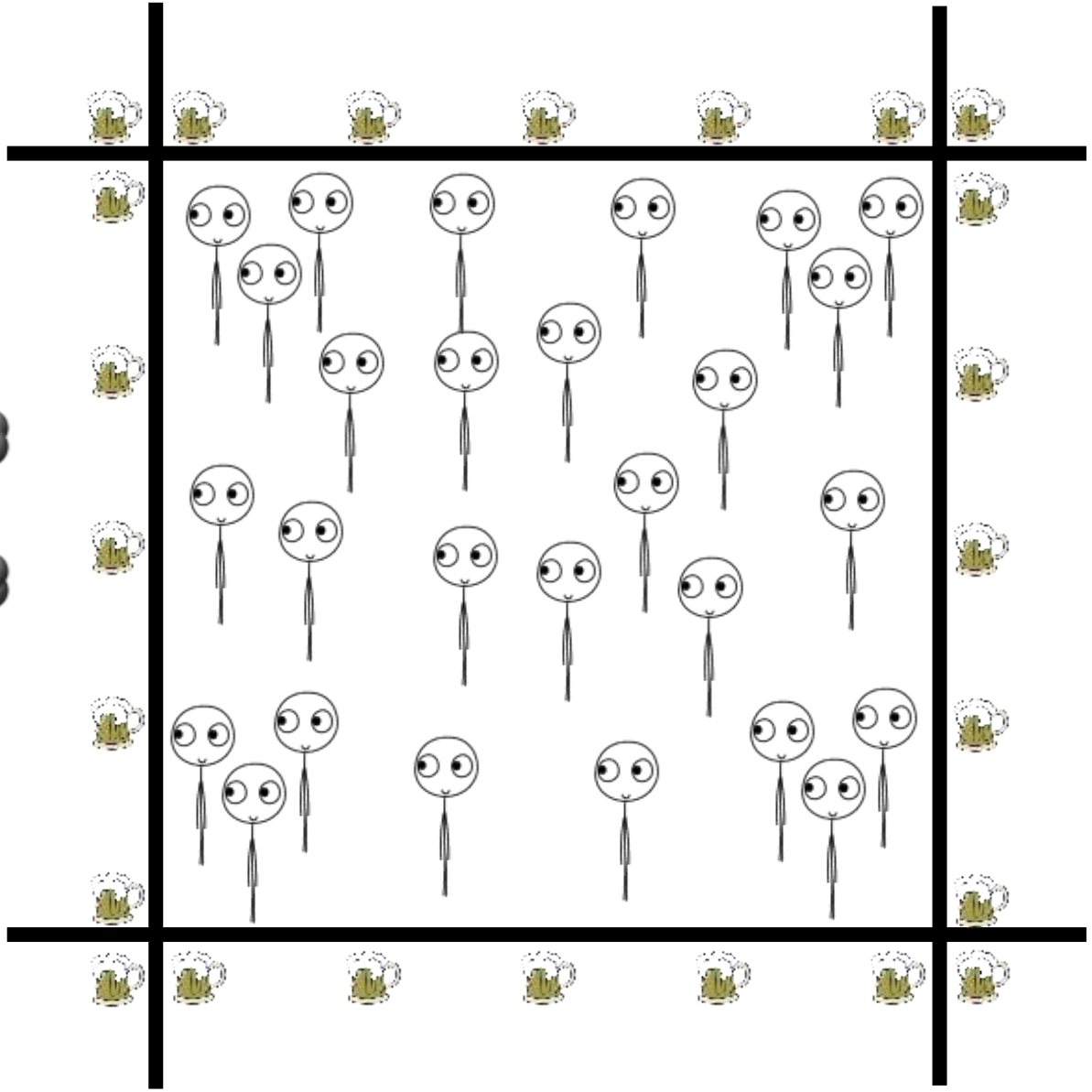
adsorption isotherm



snapshot

Guest molecules behave not unlike people in a bar





Low coverage, N ~ Q_{st}