Development of Stimuli Responsive Metal-Organic Frameworks for Energy Efficient Post-Combustion CO_2 Capture

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Summary

The goal of this work is to develop innovative metal-organic framework-based molecular sieves whose adsorption and desorption properties can be finely tuned for energy-efficient post-combustion CO_2 capture.

Motivation

- Coal-fired power plants are the single largest anthropogenic CO_2 emission sources domestically and globally.
- Post-combustion CO_2 capture can be retrofitted to existing plants (in contrast to oxy-combustion or pre-combustion capture technologies).
- DOE/NETL goal : **90% CO2 capture at less than 35% increase in the cost of electricity**
- Finding a new sorbent is important for this goal.

Key Advantages of Proposed Materials and Membranes:

1) ***Metal-Organic Frameworks*** – ***Constructed of metal containing nodes and organic linkers with high surface area and chemical functionalizability.***[1]

2) ***High CO_2 / N_2 selectivity*** – unlike conventional physical sorbents, the adsorption properties of MAMS can be tuned specifically for CO_2 (i.e. adjusting the size of its mesh by slightly changing temperature). This unique tunability enables unprecedented CO_2 / N_2 selectivity.

3) ***High CO_2 loading*** – MAMS materials are highly porous MOFs with high surface area, thereby exhibiting high CO_2 loading. Tuning the length of organic ligands can control the pore / cavity size thereby the CO_2 uptake.

4) ***Efficient regeneration*** – slight increase in temperature (e.g. ΔT regeneration ~ 10 C) will release CO_2 by opening up the gates.

5) ***Membranes with high CO_2 / N_2 selectivity*** – CO_2 / N_2 selectivity in the range of 200 will be possible due to the precise control of mesh size.

6) ***Novel membrane modules*** – by stacking a series of MAMS membranes, each of which can be continuously tuned so that the module can be used to continuously separate H_2O, CO_2, and SO_2 / N_2 (2.65 Å, 3.3 Å, and 3.6 Å / 3.64 Å, respectively) (see Figure 5).

References

[1] Yaghi, O.M.et al, *Nature*, **2003**, *423*, 705-714

[2] Zhou, H.-C. et al, *J. Am. Chem. Soc.* **2009**, *131*, 6445-6451.

[3] Li, J.-R.; Zhou, H.-C. Nature Chem. **2010,** *in press*.

Mesh-Adjustable Molecular Sieves (MAMS)

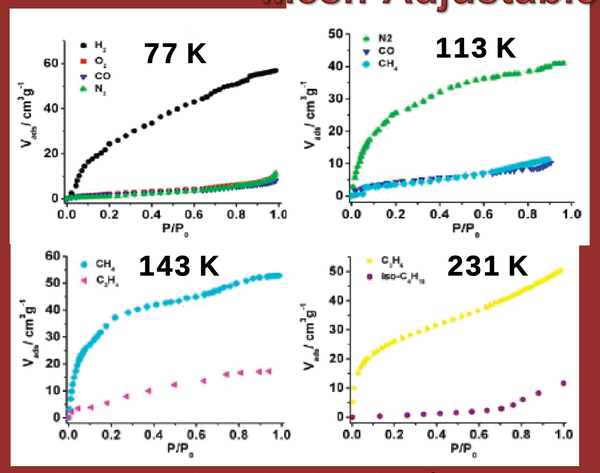

Figure 1. MAMS gas adsorption isotherms[2]

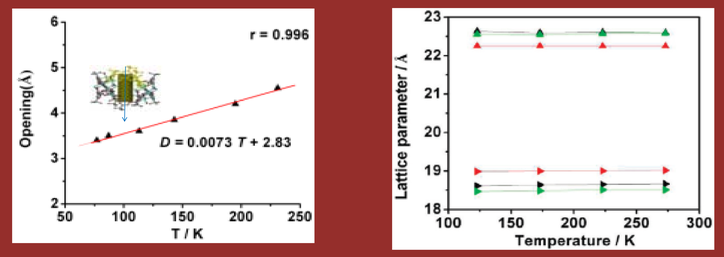

Figure 2. (***Left***) Plot of measured gate opening size (Å) with temperature. Data shows a linear correlation.[2] (***Right***) Plot of MAMS-2 (black), MAMS-3 (red), and MAMS-4 (green) lattice parameters with temperature.[2]

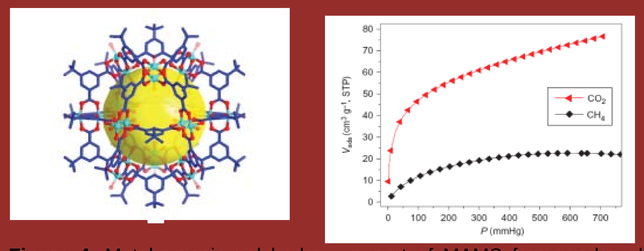

Figure 4. Metal-organic polyhedra – a part of MAMS frameworks which exhibits selective adsorption.[3]

Stimuli Responsive Sorbents

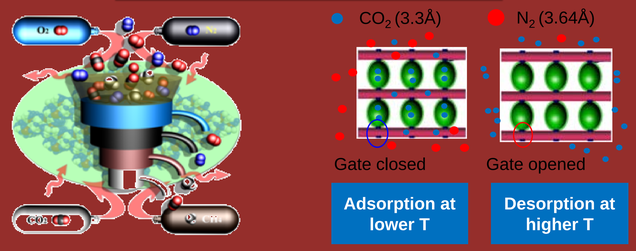

Figure 3. Conceptual illustrations of MAMS applications as stimuli-responsive sorbents.

Simulation of MAMS

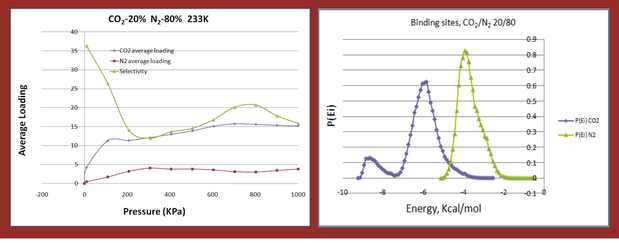

Figure 6. Preliminary GCMC results of adsorption isotherms, CO_2/N_2 selectivity, and binding energies of a 20/80 mixture of CO_2 and N_2 in MAMS-2.

Stimuli Responsive Membranes

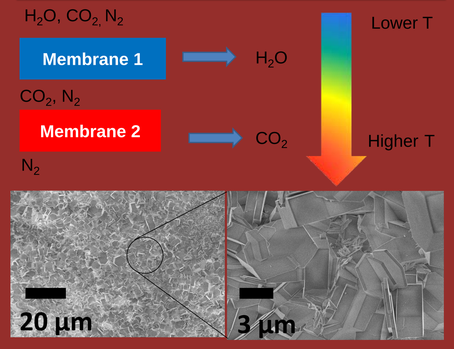

Figure 5. MAMS membranes conceptual illustration and preliminary results.

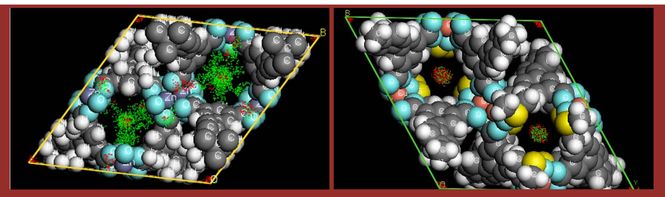

Figure 7. Preliminary GCMC simulations of a 20/80 mixture of CO_2 and N_2 at 233K and 1000 kPa on MAMS-2 (**left**) and MAMS-5 (**right**). Small dots represent the adsorbate molecules: red (CO_2) and green (N_2).