CO2 Summit II: Technologies and Opportunities | Proceedings

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Nitrogen-Functionalized Porous Carbons for Enhanced CO_2 Capture

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Clean Energy Conversions Lab

cec-lab.stanford.edu

Mission Statement:

To design and understand trace metal and carbon dioxide transformation and/or capture on surfaces to prevent their release into the atmosphere.

Theory/Computation

- **DFT: Membranes**
- **GCMC: Solid Sorbents**
- **GIS: Source/Sink**

NSF EPRI ARL GCEP Global Climate & Energy Project STANFORD UNIVERSITY Chevron

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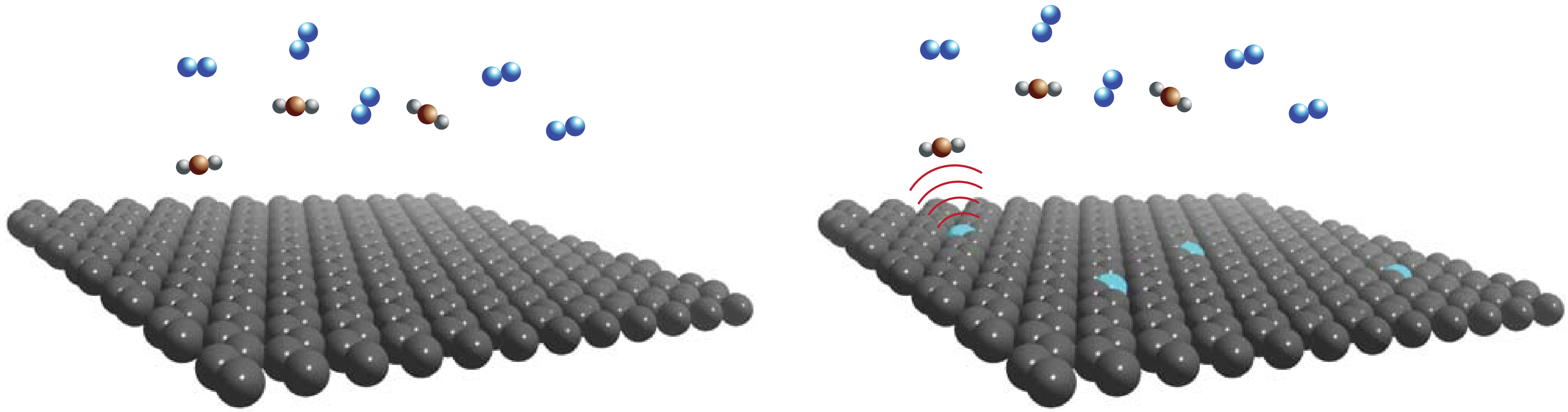
Surface Functionalization



$$\phi = \phi_D + \phi_R + \phi_P + \phi_\mu + \phi_Q + \phi_S$$

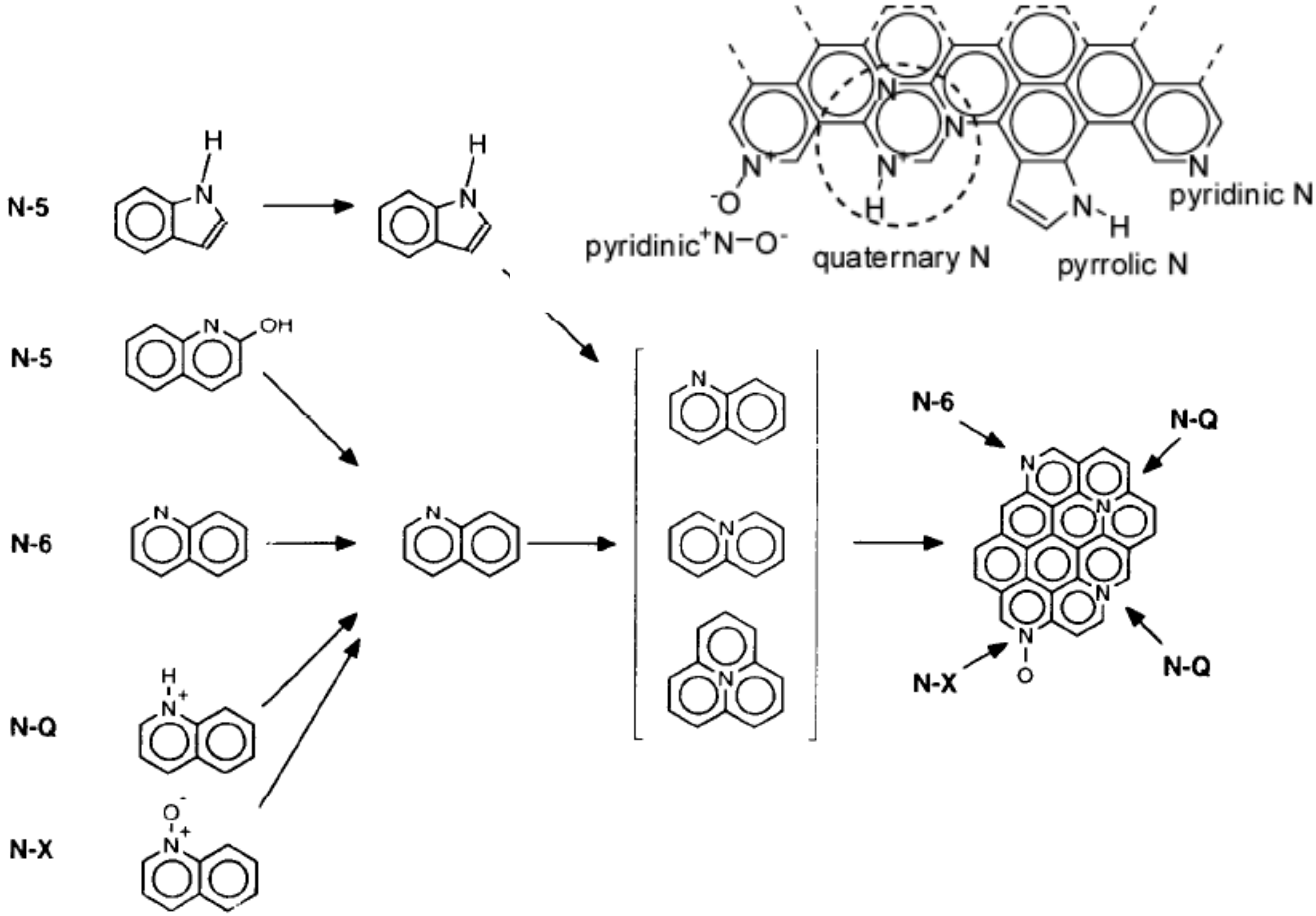
$$\phi_Q = \frac{1}{2} Q \frac{\delta E}{\delta r}$$

$$Q = \frac{1}{2} \int q(\rho, \theta)(3\cos^2\theta - 1)\rho^2 dV$$

Molecule	Kinetic diameter (nm)	Dipole moment (Debye)	Quadrupole moment (10^{-40} Coulomb·m^{2})	Polarizability (10^{-24} cm^3)
CO_2	0.330	0	–13.71, –10.0	2.64, 2,91, 3.02
N_2	0.364	0	–4.91	0.78, 1.74

Reproduced from Wilcox, Carbon Capture, Springer, 2012.

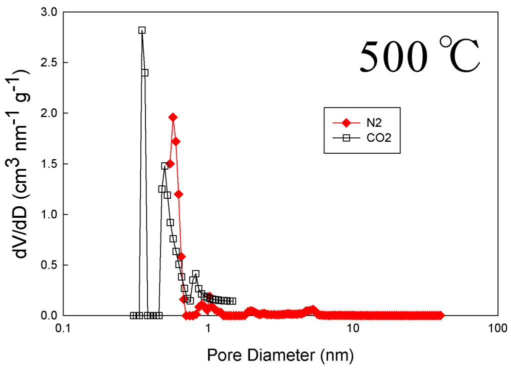
N-functionalization

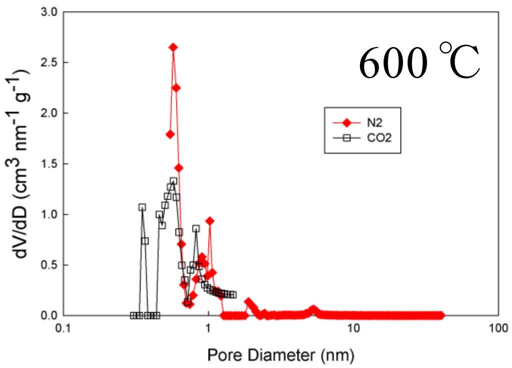


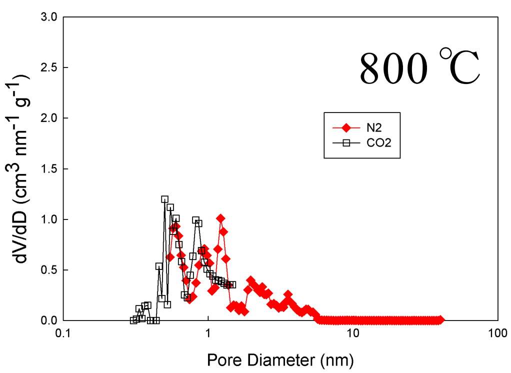
Carbon, 1995, 33(11), pp 1641-1653

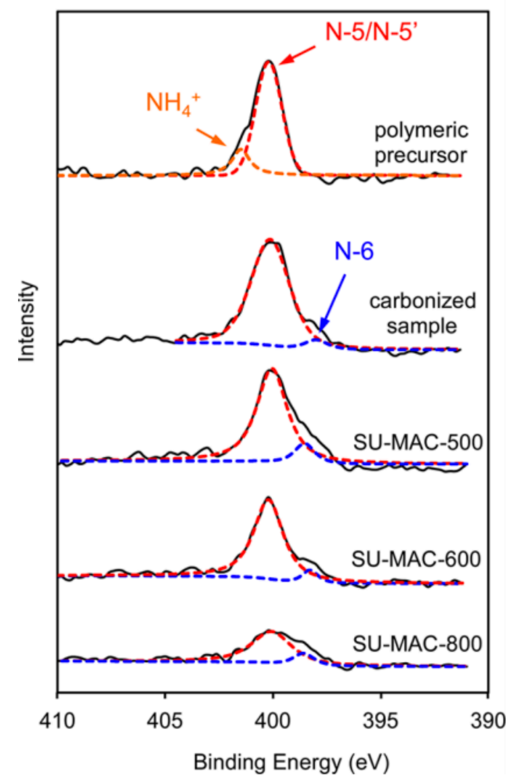
Carbonization and Activation Treatments

Carbonization = 350 ℃; Activation = 500, 600, 800 ℃







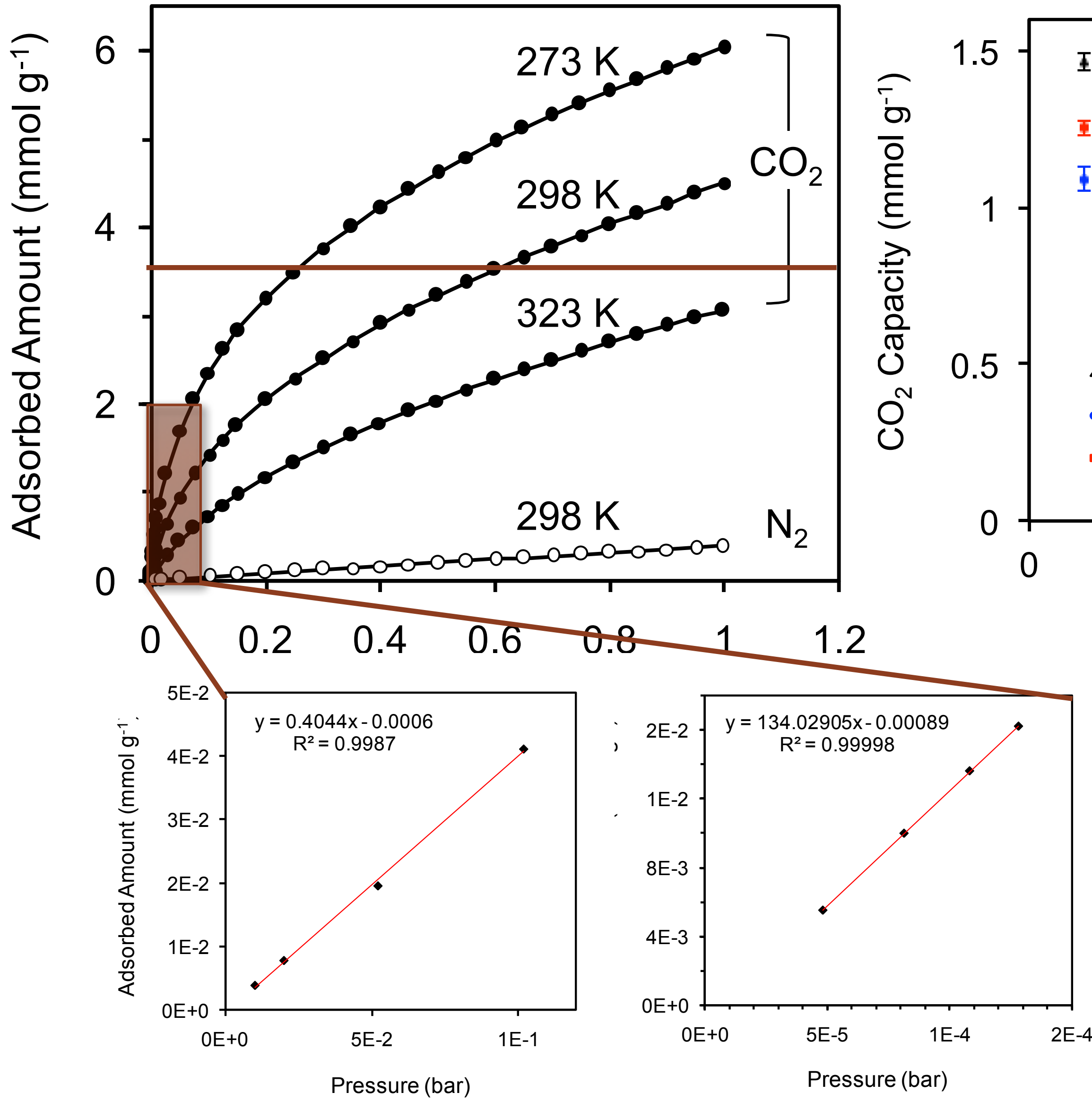


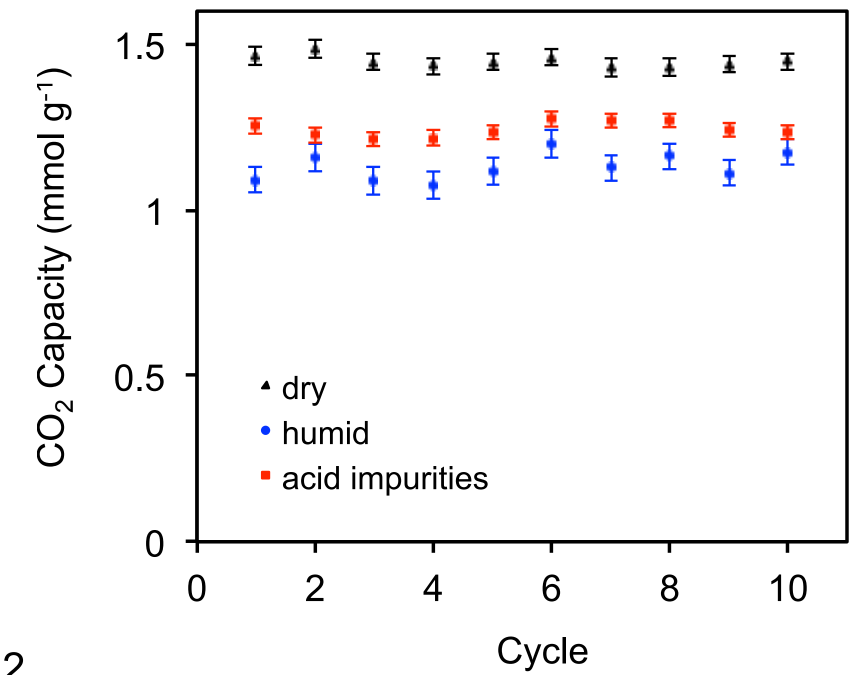
Sample	Carb. temp	Act. Temp	C content	N content	N-5/N5'	N-6
	deg C	deg C	wt%	wt%	wt%	wt%
350-500	350	500	56.8	5.77	5.01	0.76
350-600	350	600	57.0	4.0	3.69	0.31
350-800	350	800	65.4	3.19	2.66	0.54

To, He, et al. *J. Am. Chem. Soc.*, **2016**, *138* (3),

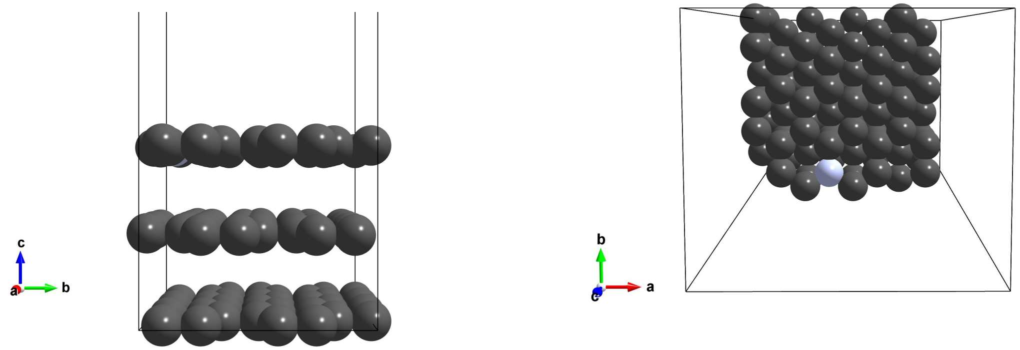
CO_2 Performance

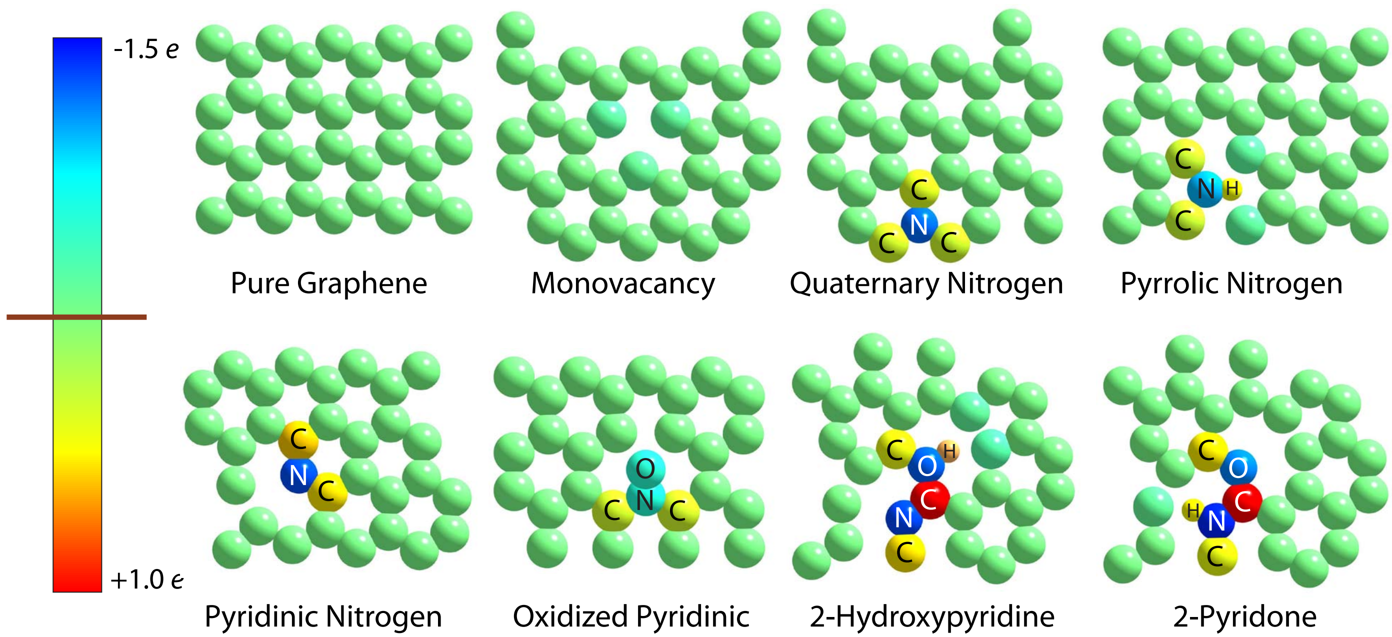
To, He, et al. *J. Am. Chem. Soc.*, **2016**, *138* (3),



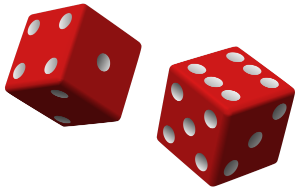


Geometric Optimization and Surface Charge (Bader)





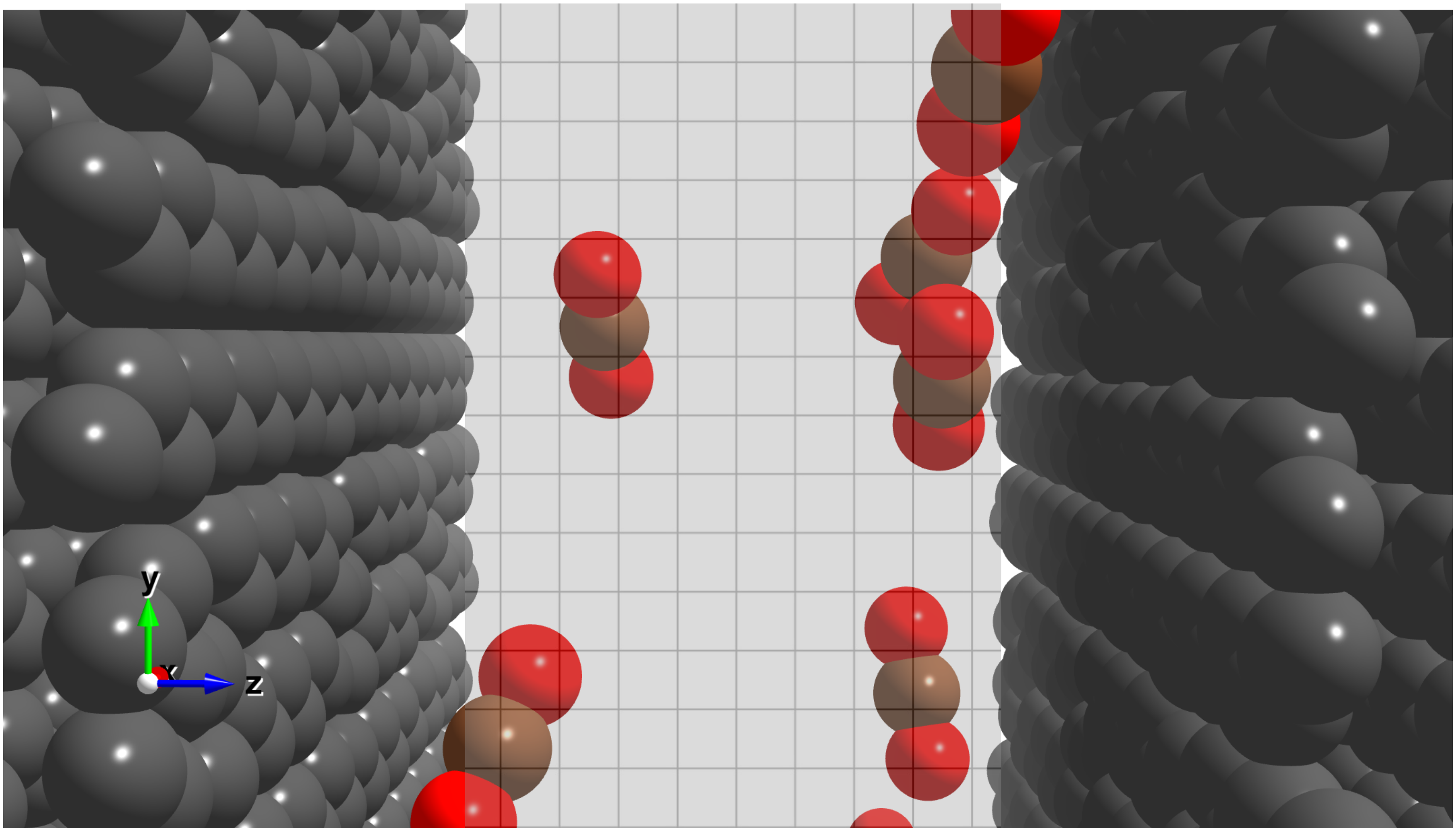
*VASP, 750ev, 6x6x1, PBE



GCMC Molecular Simulation

$$E = E_{LJ} + E_{coul}$$

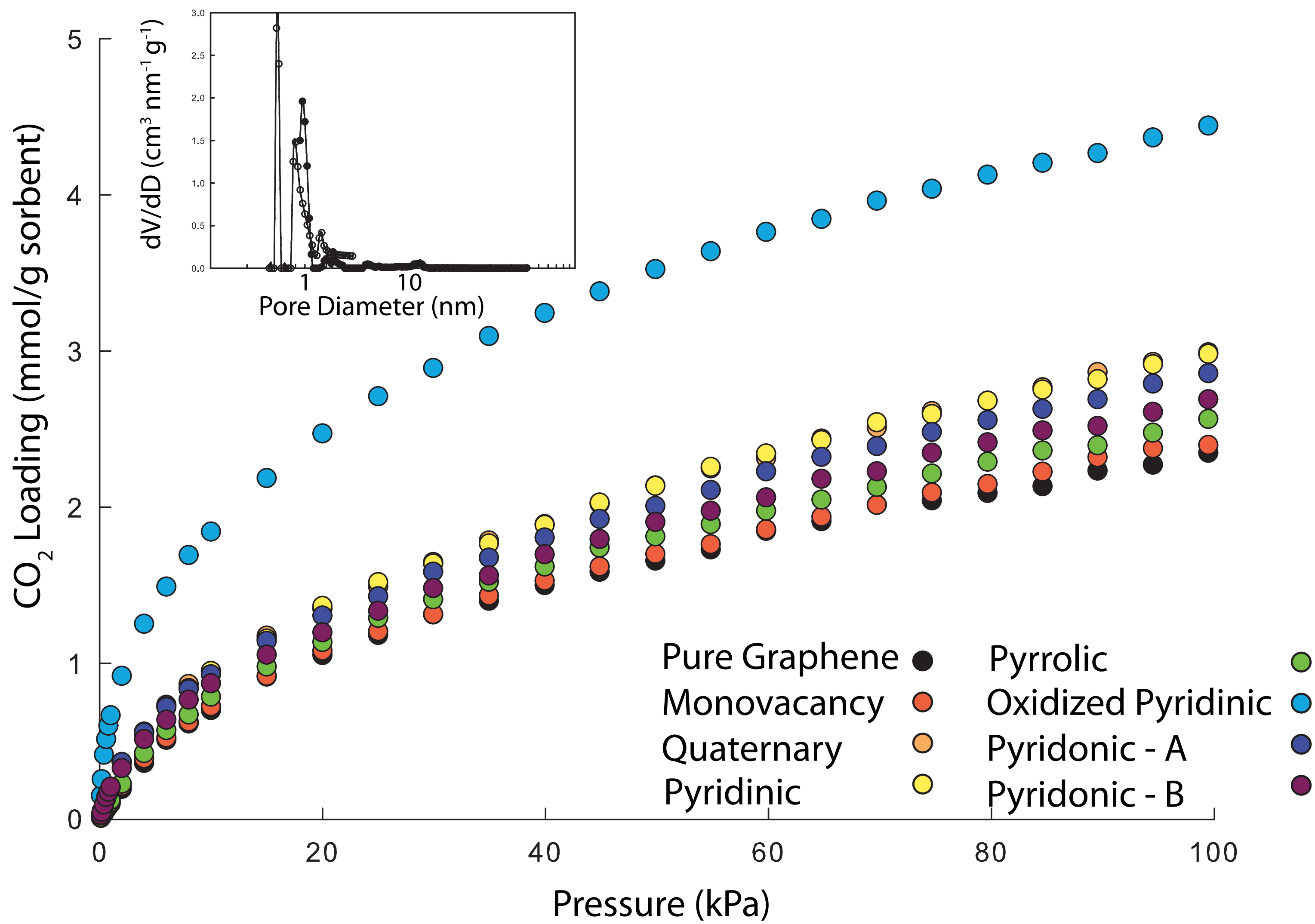
$[0,1]$ $e^{-\beta \Delta E}$ μ, V, T



Part I

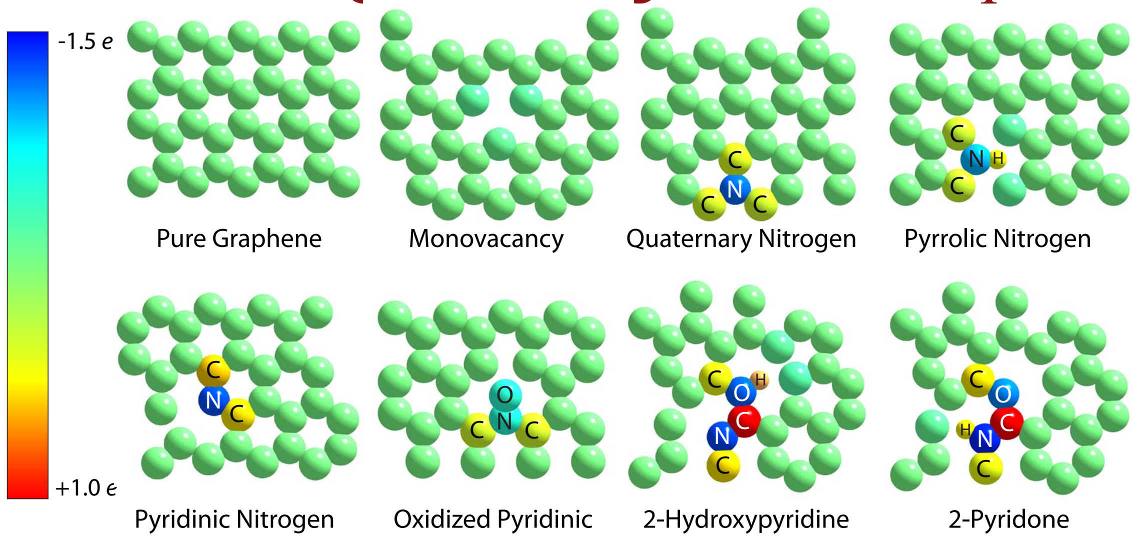
N-Functionalized Carbon Sorbents for Post-Combustion Capture

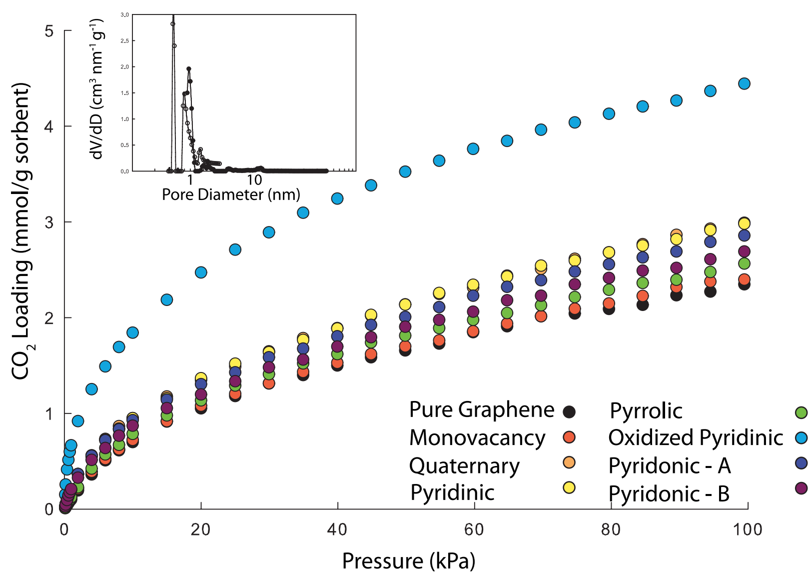
Functional Effect

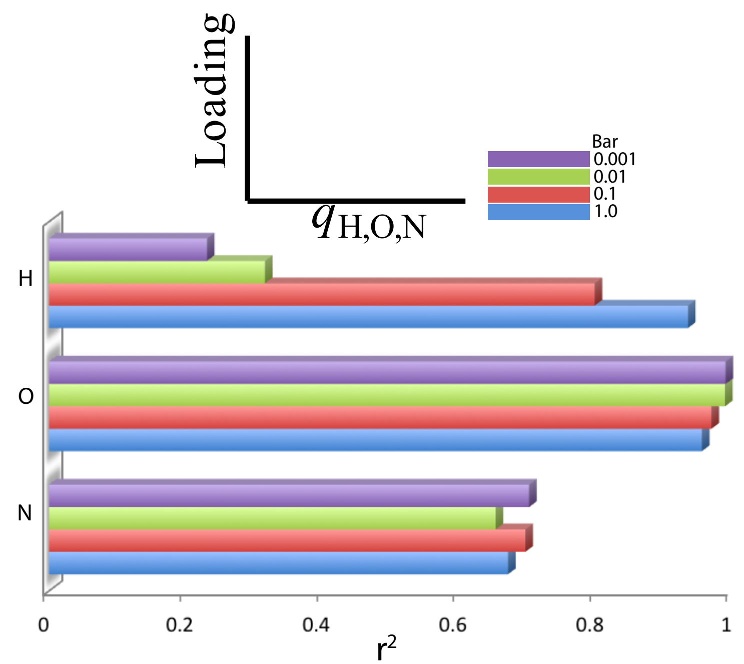


Psarras et al., *manuscript in preparation*

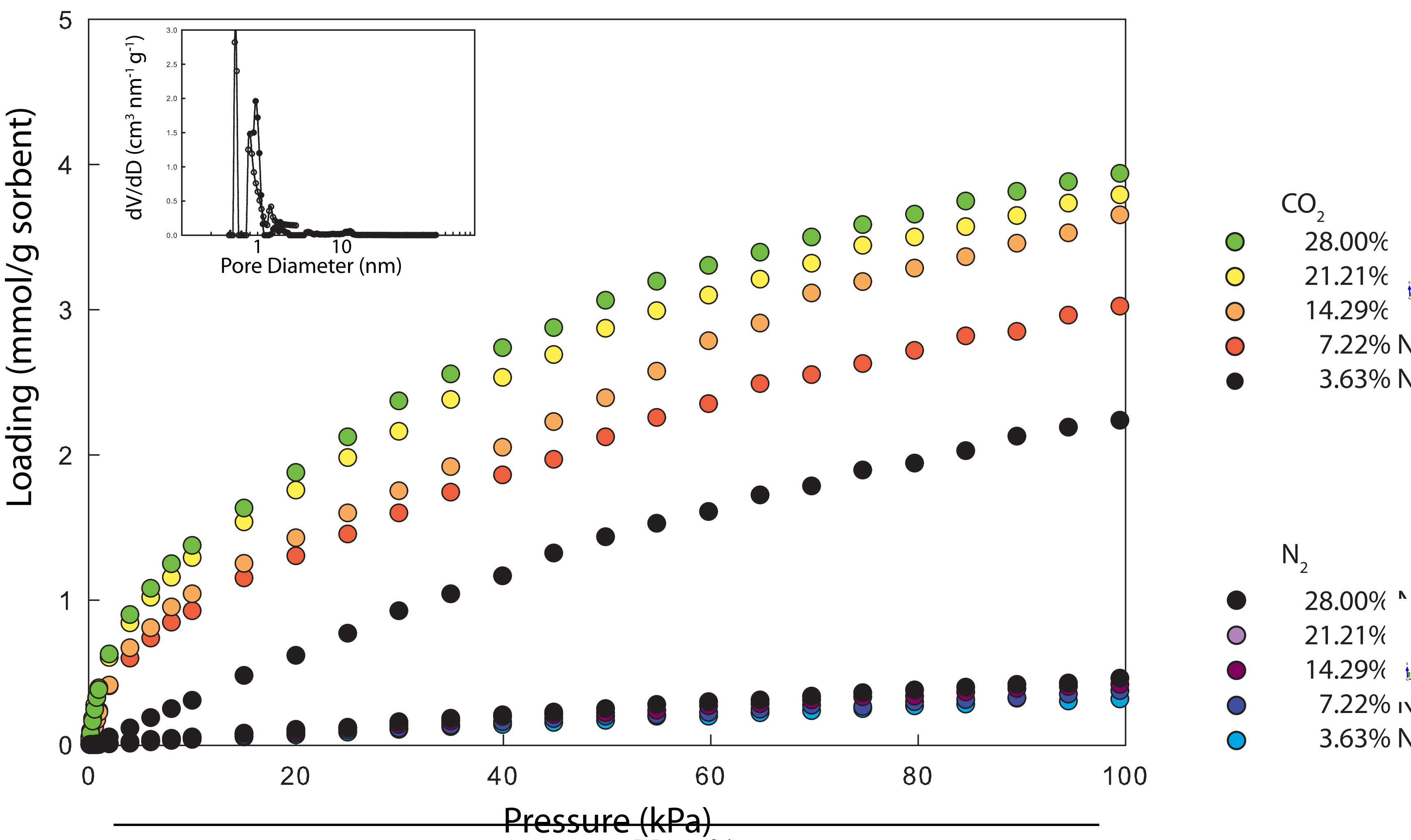
Charge/Loading Relationship

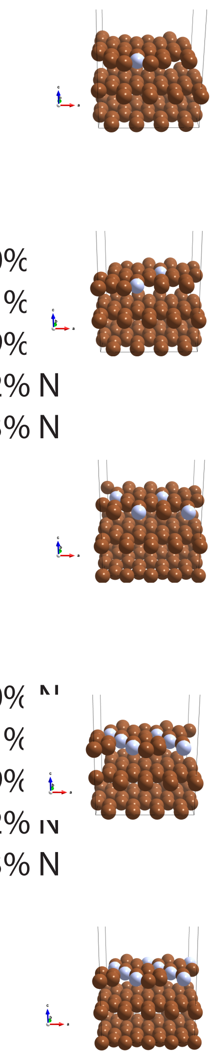






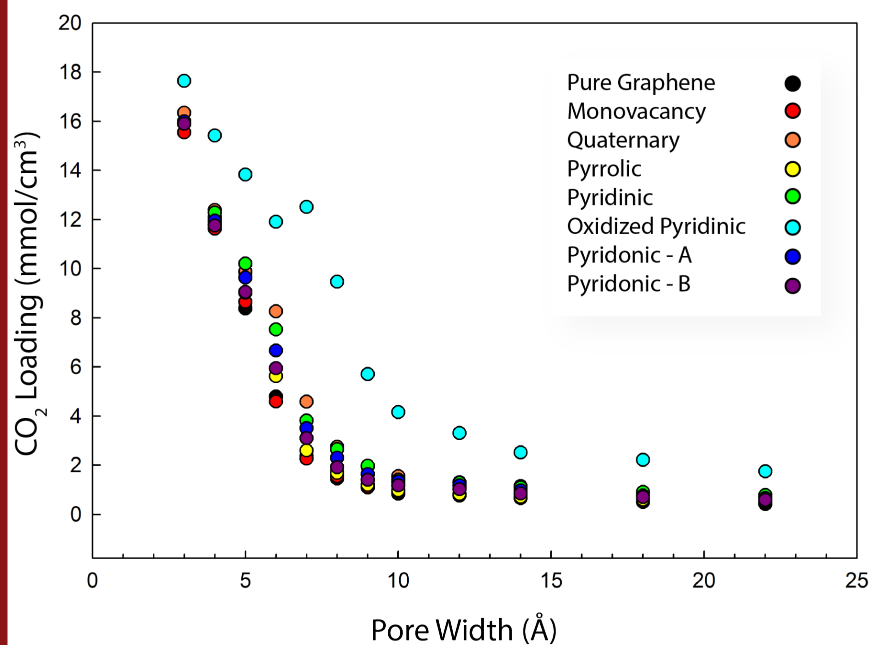
N-Coverage Effect

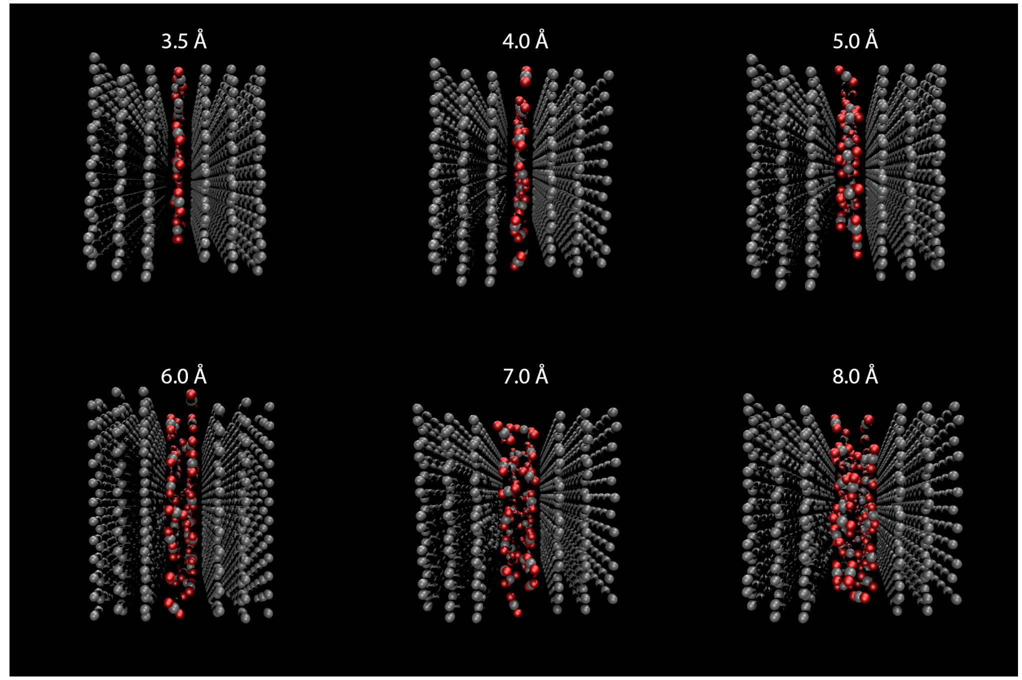


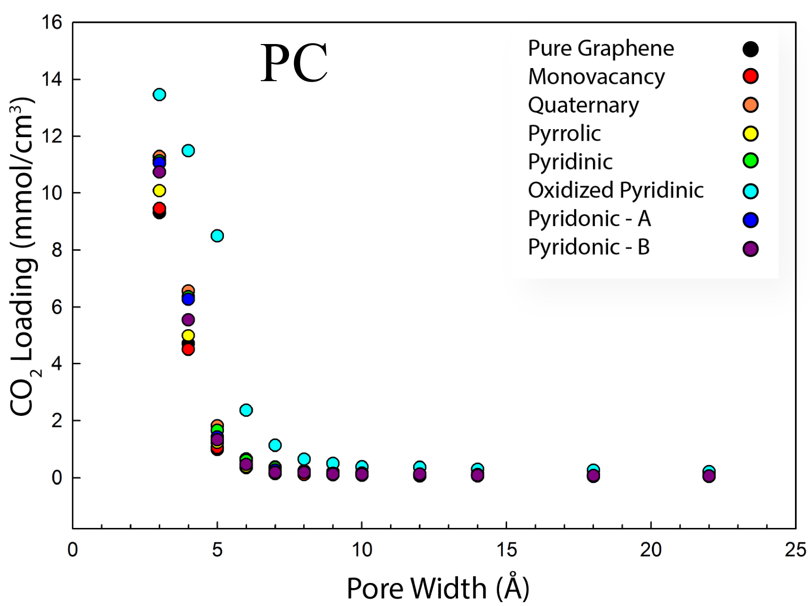


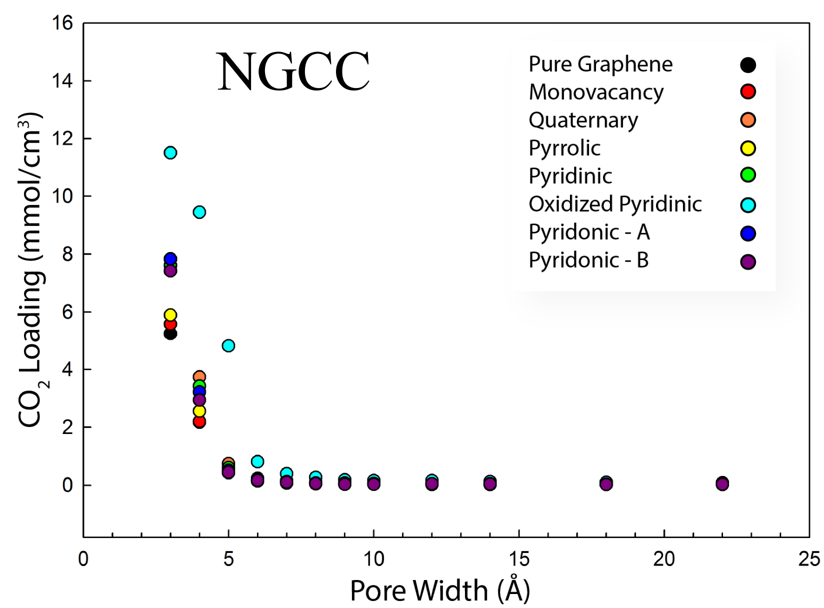
	N wt %				
	3.63	7.22	14.29	21.21	28.00
CO_2	0.1740	0.2213	0.2181	0.3772	0.3808
N_2	0.0016	0.0031	0.0054	0.0055	0.0057
Selectivity	108.8	71.4	40.4	68.6	66.8

Pore Size Effect



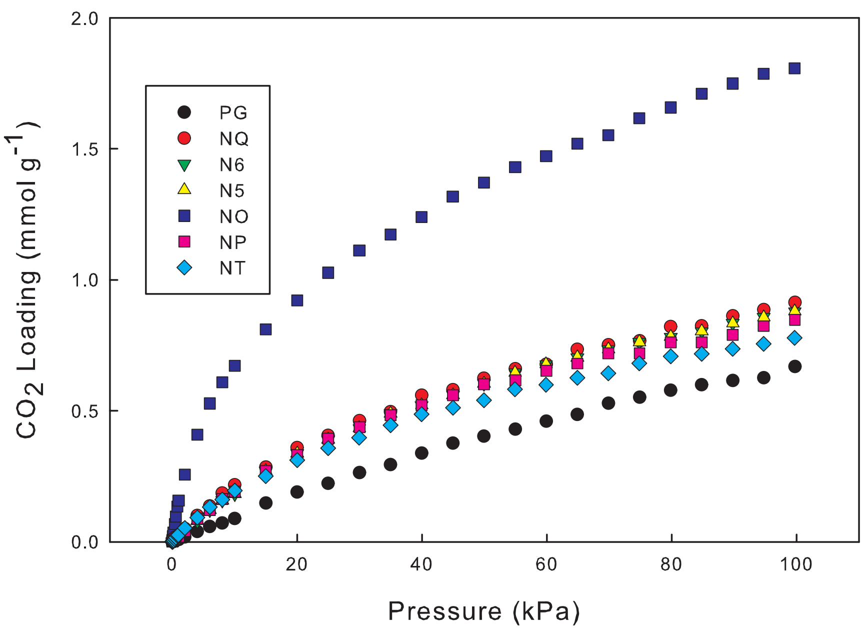


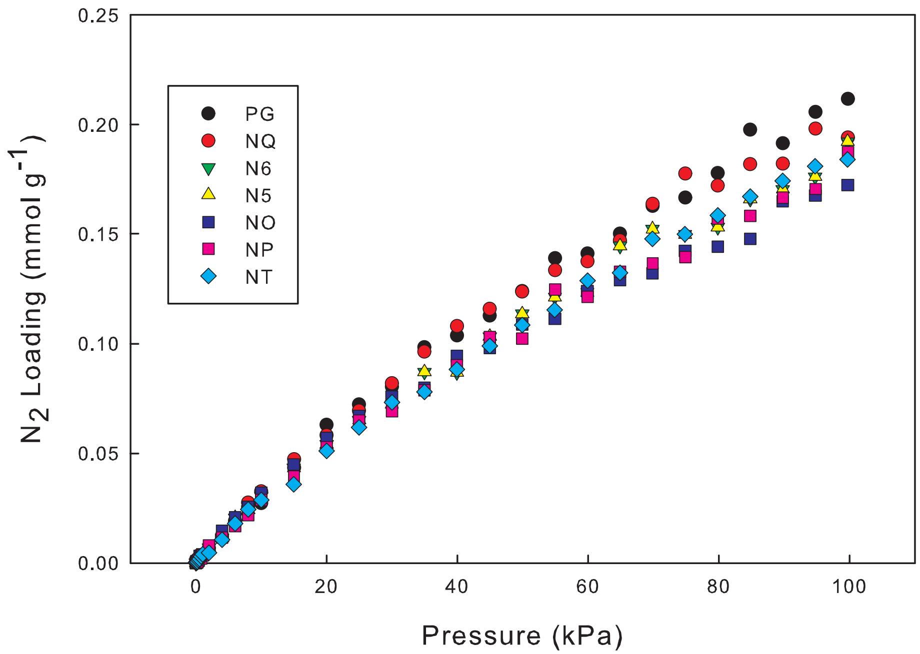


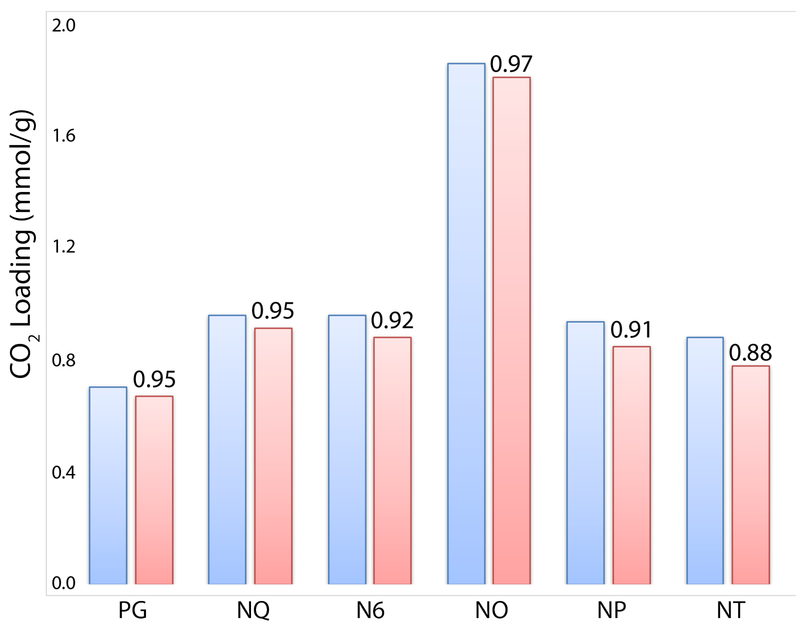


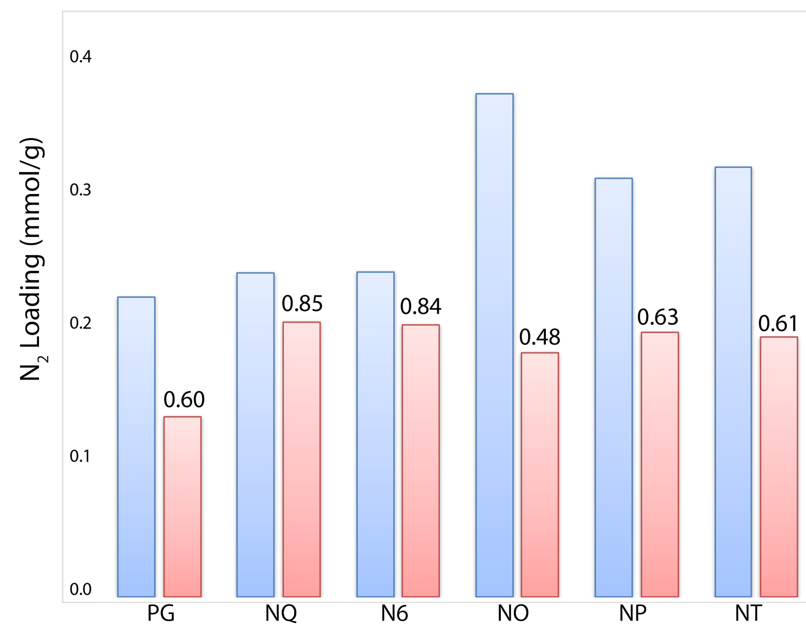
Binary Loading

10:90 CO_2:N_2

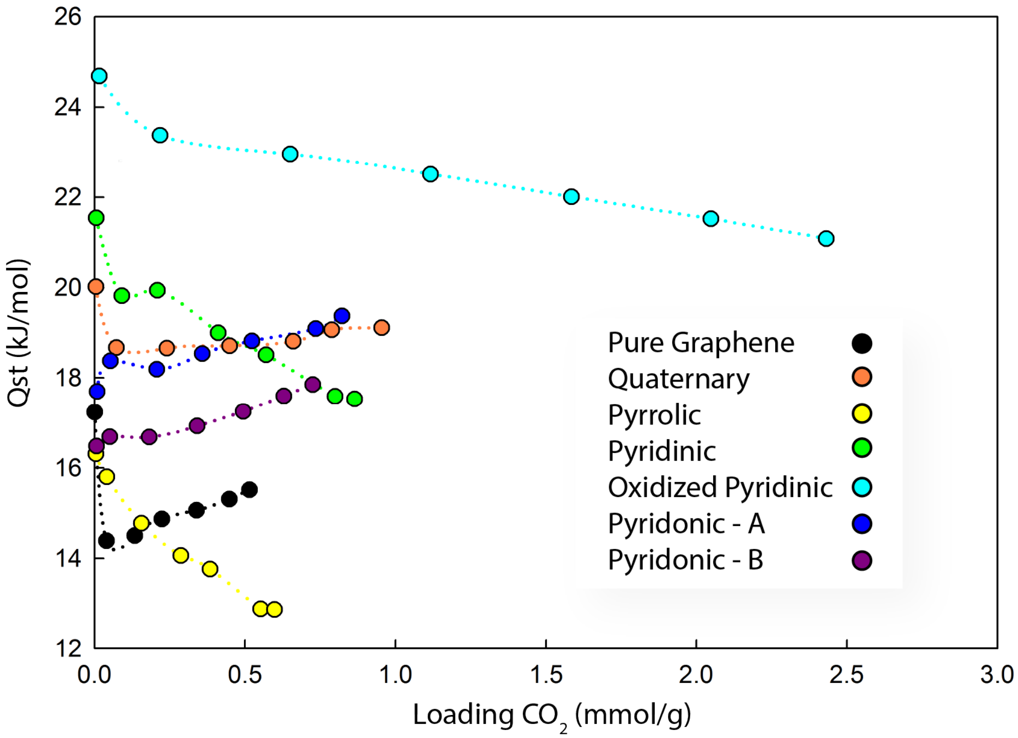


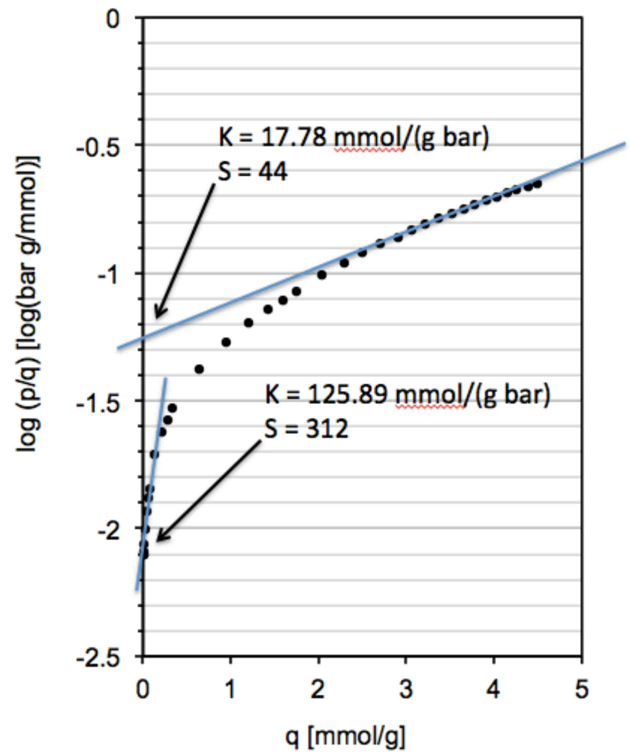






CO_2:N_2 Selectivity





			Pore Size (Å)					
	IAST	HL	3.5	4	5	6	7	8
PG	21.70	30.12	61.81	26.29	10.00	6.84	3.89	3.58
NQ	37.67	87.84	225.69	97.13	18.63	11.15	10.81	6.69
N6	37.56	78.11	212.97	101.00	14.23	10.61	9.21	6.92
N5	24.11	31.13	73.66	29.12	13.07	7.80	4.31	1.92
NO	**47.02**	**153.51**	251.93	199.63	100.93	41.65	21.45	28.52
NP	28.45	72.74	119.65	40.85	18.17	7.71	11.36	1.34
NT	26.05	91.00	129.86	43.91	18.23	7.82	9.77	12.17

Part II

N-Functionalized Carbon Sorbents for Natural-Gas Sweetening

Conclusions

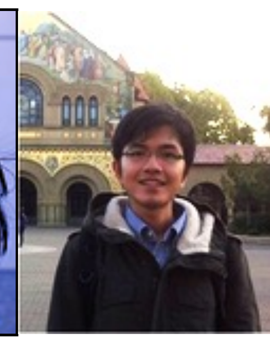
- Ultra-microporous volumes are crucial to the enhancement of CO_2 capacity and HL selectivity
- Oxidized pyridinic nitrogen was most influential to loading enhancement, followed by quaternary and pyridinic groups
- There appears to be an optimal coverage for N-moieties – increasing N coverage did not enhance CO_2 selectivity
- PC/NGCC PCC is particularly sensitive to ultra-microporous structure

Acknowledgements

Professors Jennifer Wilcox and Zhenan Bao

John To, Jiajun He, and the Stanford Center for Carbon Storage



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