UNCERTAINTY QUANTIFICATION IN THE CARBON CAPTURE SIMULATION INITIATIVE

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Carbon Capture Simulation Initiative

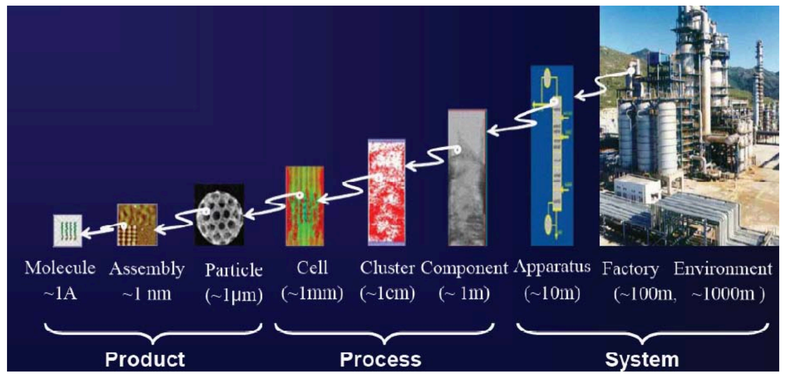
The Carbon Capture Simulation Initiative (CCSI), is a Department of Energy Office of Fossil Energy effort involving 5 national laboratories in collaboration with the coal-fired power plant industry to investigate carbon capture technology. CCSI is developing technology to accelerate the process of making carbon capture reliable and affordable.

Uncertainty Quantification (UQ):

UQ capability is critical for simulation based analysis due to the complexity and cost of implementation of candidate systems in industry operations. UQ includes methods and tools for identification and propagation of uncertainty at all levels of a system. This poster illustrates UQmethods being developed for effective simulation of a solid sorbent process for carbon capture.

Preliminary objectives:

- Input sensitivity and uncertainty: identify appropriate input ranges and impacts on simulation code results; demo for more complex model, NETL 32D with 12 inputs.
- Input calibration: estimate "best" simulation input values for parameters that determine equilibrium constants consistent with physical experimental results..
- Goal is to quantify distributions that capture uncertainty associated with these parameters, which is useful for UQ efforts for higher level systems.



Solid Sorbent Models

Sorbent consists of mesoporous silica backbone embedded with amine PEI, which adsorbs CO2 thru chemical reaction with temperature impacting effectiveness of adsorption.

0th generation model: a pure **ideal equilibrium model** is simplified to exclude kinetic parameters, assuming fast kinetics with equilibrium achieved in test runs.

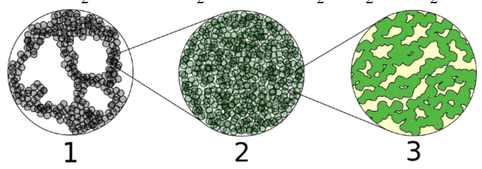
1st generation: **lumped kinetic model**, base model w/ kinetic, thermodynamics, and water effects used in process/CFD models.

2nd generation: **NETL 32D,** includes transport effects.

3rd generation: includes non-ideal thermodynamics and site-competitive water adsorption effects.

$$R_2NH + CO_2 \xleftrightarrow{K_5} R_2NH^+ : CO_2^-$$

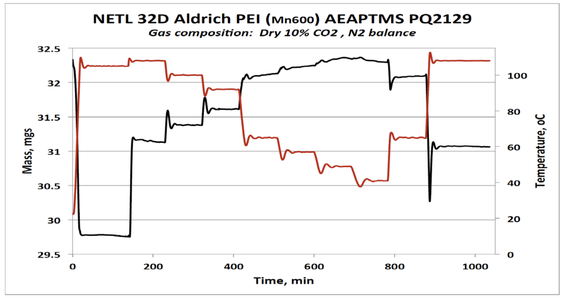
$$R_2NH + R_2NH^+ : CO_2^- \xleftrightarrow{K_6} R_2NH_2^+ + R_2NCO_2^-$$



Physical Data

Thermogravimetric analysis (TGA) response data is weight (or % increase) of sorbent measured every second.

NETL conducted TGA experiment, results pictured below with temperature profile (red) including 7 temperatures and 8 transitions over time. The same profile was used in model runs.



Sensitivity Study for NETL 32D Solid Sorbent Model

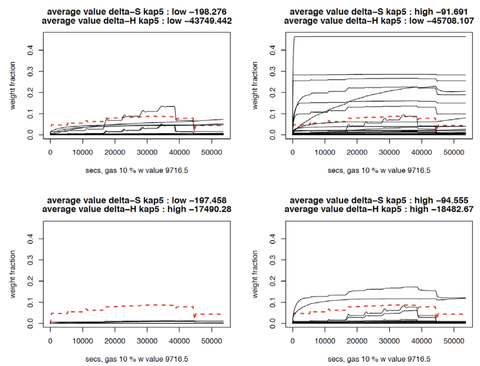
Parameter	Description	Low Value	High	Unit
$\log(K_0/\tau)$	Porosity and tortuousity of movement	-10.5	-8.0	
τ_3	Bulk-phase diffusion movement	1	10	
n_v	Amine site density	5000	25000	mol/m^3
f	Constant related to n_v	0.1	5	
ΔH_κ	Change in enthalpy for full reaction	-96485	-19297	J
ΔS_κ	Change in entropy for full reaction	-332.56	-41.57	J/mol-K
ΔH_{k5}	Change in enthalpy for first reaction	-57891	-4824.25	J
ΔS_{k5}	Change in entropy for first reaction	-249.42	-41.57	J/mol-K
ΔH_{k6}	Change in enthalpy for second reaction (forward)	9648.50	964850	J
$\log(\zeta_{k6})$	Change in entropy & add'l parameters	1	8	
$\Delta H\_\{\mu_h\}$	Change in enthalpy of jump barrier	19297	125431	J
$\log(\zeta\_\{\mu_h\})$	Change in entropy & add'l parameters	-11	-2	

Sensitivity analysis based on identifying trends mean weight fraction calculated **D, Solid Sorbent Model**

Goal: Evaluation of code function and sensitivity for 12 parameters.
• Initial experiment design 128 runs, varying 12 inputs prescribed by OA-based, LHS, achieving balanced representation of multiple factors .

Sensitivity Analysis

- Sensitivity analysis based on identifying trends mean weight fraction calculated from TGA curves, with individual or pairs of parameters.
- Nearly half of simulations did not complete (20) or have little signal.
- Sorbent kinetics seem to be controlled by number and mobility of zwitterions.
- Sensitivity analysis focused attention on two parameters with apparent main effects: ΔH_{k5} and ΔS_{k5}, illustrated graphically, right.



Calibration Study for Ideal Equilibrium Model

$$g = 33.9\frac{z+x}{m_s}$$

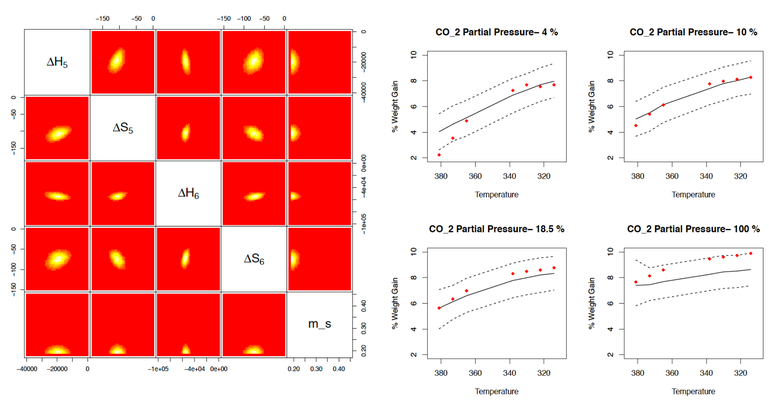
$$\kappa_5 = \frac{z}{(1-2x-z)p} = \frac{1}{P}\exp\left(\frac{\Delta S_5}{R}\right)\exp\left(-\frac{\Delta H_5}{RT}\right)$$

$$\kappa_6 = \frac{x^2}{(1-2x-z)z} = \exp\left(\frac{\Delta S_6}{R}\right)\exp\left(-\frac{\Delta H_6}{RT}\right)$$

Parameter	Description	Lower Bound	Upper Bound	Unit
m_s	Sorbent weight per active amine site	0.1886	0.48	kg/mol
ΔH_5	Change in enthalpy for first reaction	-40000	0	J
ΔS_5	Change in entropy for first reaction	-150	0	J/mol-K
ΔH_6	Change in enthalpy for second reaction	-100000	0	J
ΔS_6	Change in entropy for second reaction	-150	0	J/mol-K

Goal: Estimation of parameters (5) that determine equilibrium constants in an ideal equilibrium model, such that there is consistency between physical observation and simulation of *g*, equilibrium weight increase.

- Incorporate expert prior information about parameters.
- Bayesian statistical approach quantifies model discrepancy and observation error.



Parameter uncertainty

Color-scale bivariate posterior probability densities for pairs of parameters – shows most likely parameter values for simulation agreement with physical observations.

Predictions and Bounds

Physical observation in red, simulation model estimates and bounds (95%) for 4 cases of CO_2 pressure.

Continuing Efforts

- Estimate parameters for complex sorbent model, NETL 32D, accounting for time component with temperature transitions and gas pressure cases. May require construction of surrogate model (emulator).
- Thus far, focus has been on a baseline model. Future work will compare to a model with variable reaction enthalpies.
- Constrained Sampling Design
- Model Comparison Using Sensitivity Analysis on model discrepancy estimates.
- Study propagation of parameter uncertainty for upstream modeling effects.