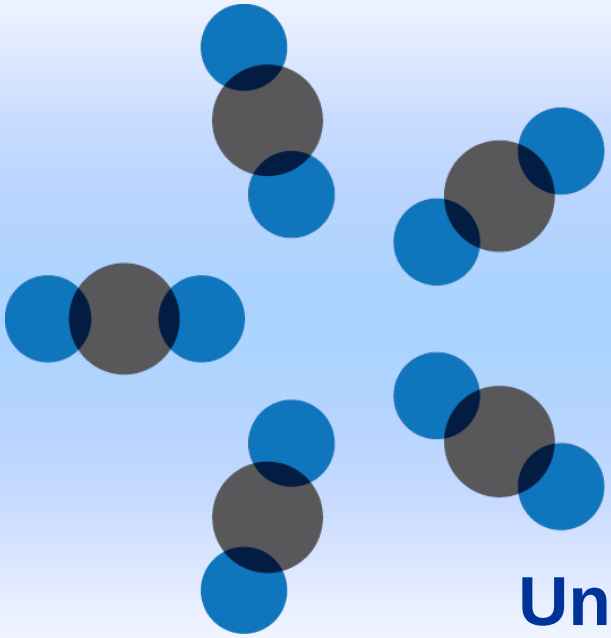

CCSI™

Carbon Capture Simulation Initiative

# Uncertainty Quantification for Carbon Capture Simulation

**J. Wendelberger, K. Sham Bhat, L. Moore, C. Storlie**
**Los Alamos National Laboratory**

**D. Mebane, National Energy Technology Laboratory**

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NETL BERKELEY LAB Lawrence Livermore National Laboratory Los Alamos NATIONAL LABORATORY EST.1943 Pacific Northwest NATIONAL LABORATORY U.S. DEPARTMENT OF ENERGY

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# Abstract

The Carbon Capture Simulation Initiative (CCSI) is developing technology to accelerate the process of making carbon capture from coal-fired power plants reliable and affordable by using simulation to reduce the need for physical testing for scaling up to commercial scale. Uncertainty quantification (UQ) capability is critical to simulation-based analysis of carbon capture systems for understanding and managing the complexity and economic impact of incorporation of carbon capture systems in current and future commercial operations. These uncertainty quantification tools include input sensitivity analysis, calibration of inputs, construction of surrogate models, and propagation of uncertainty. This talk will illustrate the use of selected UQ concepts in early stage development and evaluation of requirements for effective simulation of a solid sorbent process for carbon capture.

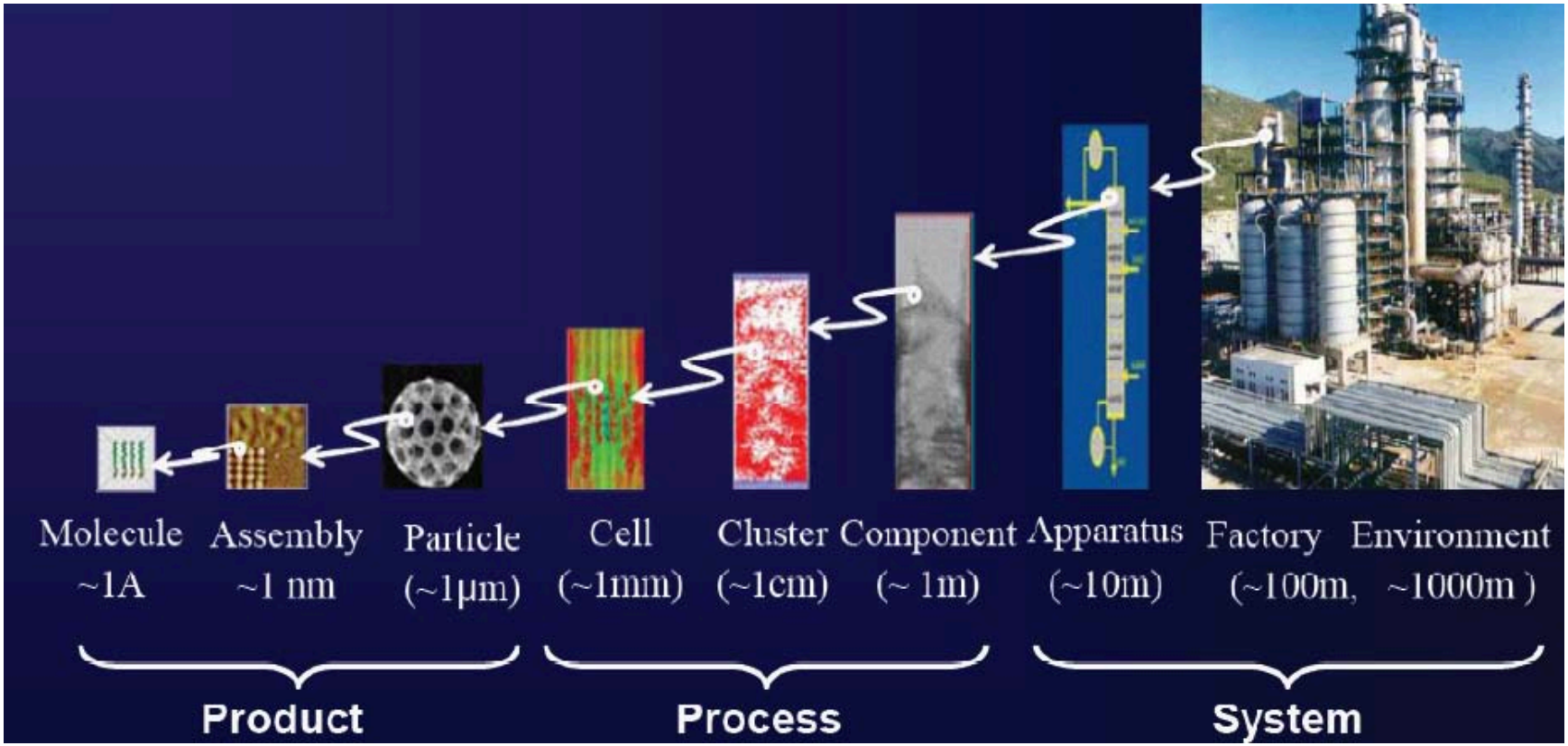
# Overview

- Carbon Capture Simulation Initiative
- Uncertainty Quantification
- Model Development for Solid Sorbents
- Sensitivity Analysis
- Parameter Estimation and Prediction Uncertainties
- Experiment Planning
- Continuing Efforts

# Carbon Capture Simulation Initiative

- **Carbon Capture and Storage (CCS) technology goal**: convert fossil fuels into reliable low carbon energy supply, maintaining relative affordability for consumers.
- Scale-up of existing CCS technology to large power plants may be easy but result in unanticipated inefficiencies and large cost increases.
- Developing, proofing and implementing new technology may take decades, requiring testing at increasingly larger scales.
- **Carbon Capture Simulation Initiative (CCSI) goal:** develop simulation-based toolset for new carbon capture technologies, scalable to commercial level without physical testing, include uncertainty quantification (UQ) tools to aid in interpreting simulation results.

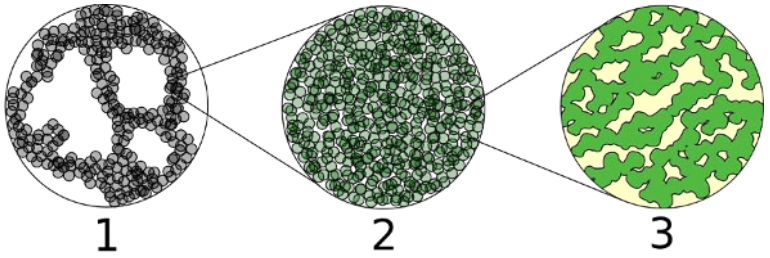
# Carbon Capture and Storage Technology



# Uncertainty Quantification

- **Uncertainty Quantification (UQ)** broadly includes methods and tools to identify and quantify uncertainty at all levels of a system and to incorporate that uncertainty in full system performance analyses.
- UQ capability is critical to simulation-based analysis of CCS technology due to complexity, high cost of implementation of candidate systems and need to understand and manage economic impact of incorporation of CCS technology in current industry operations.
- We demonstrate use of a few UQ concepts in early stage development and evaluation of requirements for effective simulation of a **solid sorbent process** for carbon capture in collaboration with David Mebane (NETL), code developer.

# Solid Sorbent Models




- Sorbent consists of mesoporous silica backbone embedded with amine PEI, which adsorbs $CO_2$ thru chemical reaction with temperature impacting effectiveness of adsorption.

# Development of Solid Sorbent Models

- 0th generation model- a pure **ideal equilibrium model**, simplified to assume fast kinetics with equilibrium achieved in test runs: 5 parameters.
- 1st generation model- **lumped kinetic model**, base model that includes ideal thermodynamics, kinetic and basic water effects. Currently used in process/CFD models.
- 2nd generation model- **NETL 32D**, basic transport w/ ideal thermodynamics and basic water effects (not included in current version).
- 3rd generation model- includes non-ideal thermodynamics, and site-competitive water adsorption effects.

# Solid Sorbent Model- Equations

$$R_2NH + CO_2 \xleftrightarrow{\kappa_5} R_2NH^+ : CO_2^-$$

$$R_2NH + R_2NH^+ : CO_2^- \xleftrightarrow{\kappa_6} R_2NH_2^+ + R_2NCO_2^-$$

$$\kappa = \kappa_5 \kappa_6, \ \kappa_6 = k_6 k_{-6}$$

$$\kappa = \exp(-\Delta G/(RT))$$

# Ideal Equilibrium Model (0th Generation)

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

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

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

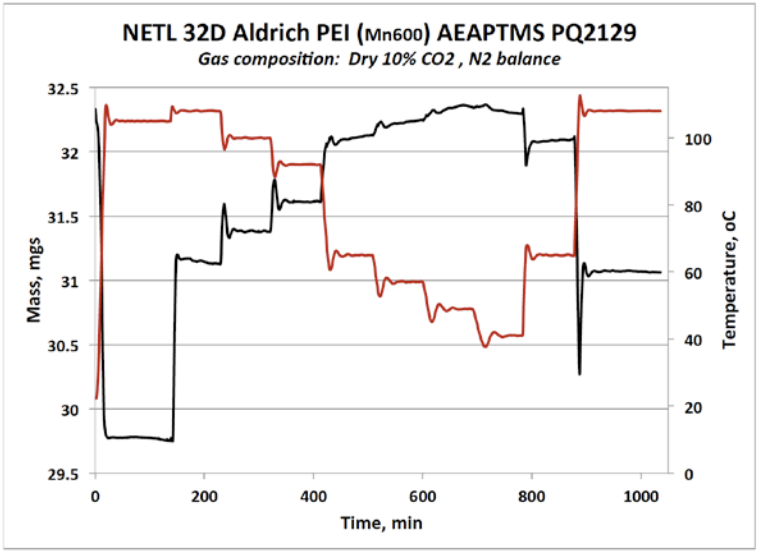
| Parameter | Description | Lower Bound | Upper Bound | Unit |
|---|---|---|---|---|
| $m_s$ | Sorbent weight per active amine site | 0.1886 | 0.48 | kg/mol |
| $\Delta H_5$ | Change in enthalpy for first reaction | -40000 | 0 | J |
| $\Delta S_5$ | Change in entropy for first reaction | -150 | 0 | J/mol-K |
| $\Delta H_6$ | Change in enthalpy for second reaction | -100000 | 0 | J |
| $\Delta S_6$ | Change in entropy for second reaction | -150 | 0 | J/mol-K |

# Uncertainty Quantification

- **Immediate objectives:**
  - Input sensitivity and uncertainty: identify appropriate input ranges and impacts on simulation code results: demo for NETL 32D with 12 inputs.
  - Input calibration: estimate 'best' simulation input values for parameters that determine equilibrium constants consistent with physical experimental results: demo for ideal equilibrium model, 5 inputs.
  - Plan experiments to obtain physical data.
  - Forwarding-looking goal: quantify distributions that capture uncertainty associated with model parameters. This will be used in turn for next higher level system uncertainty quantification.

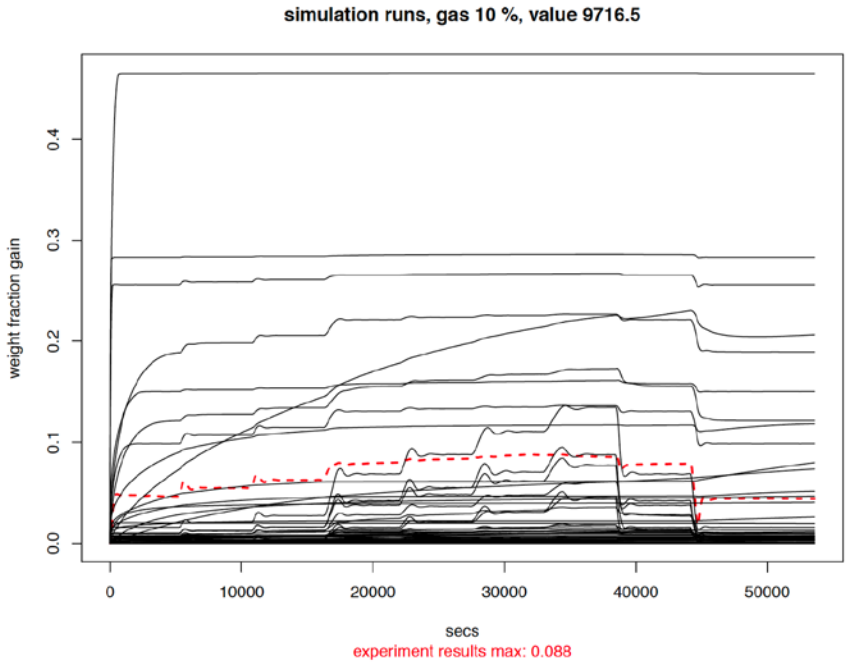
# Solid Sorbent Experimental/Simulation Data

Experimental Data- TGA




NETL experiment with dry $CO_2$ sorbent, thermogravimetric analysis (TGA) weight differential measured over time/temperature-dependent profile (in red), at specific gas composition (determined by the partial pressure).

Simulations:
TGA curve or equilibrium weight, NETL experimental results in red.

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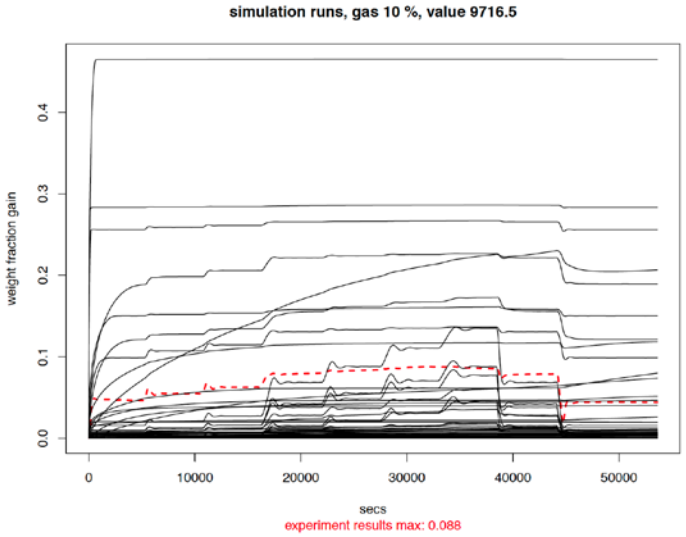
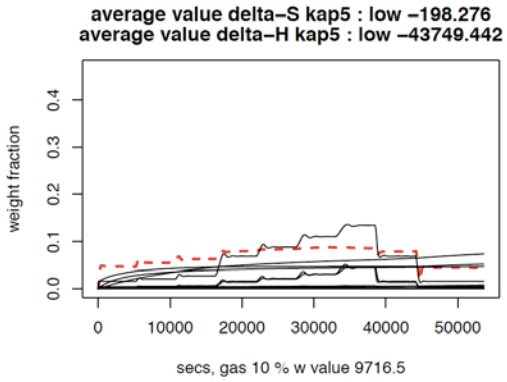
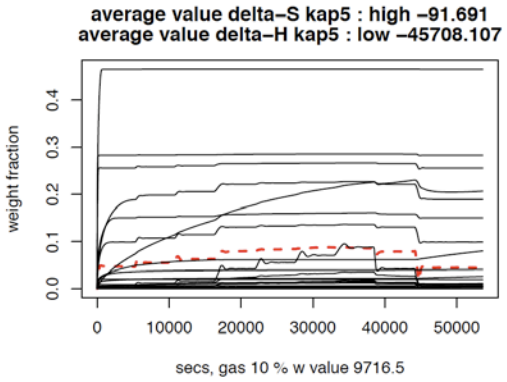
# Sensitivity Study for NETL 32D, Solid Sorbent Model

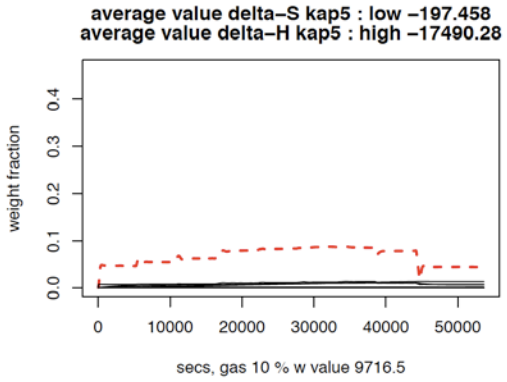
- Goal: Evaluation of code function and sensitivity with respect to 12 parameters.
- Initial experiment design 128 runs, varying 12 inputs prescribed by an orthogonal array based Latin Hypercube sample (OA-based, LHS), achieving balanced representation of multiple factors at high and low levels.
- Sensitivity analysis based on identifying trends of simple scalar calculated from TGA curves (mean weight fraction) with individual or pairs of parameters.

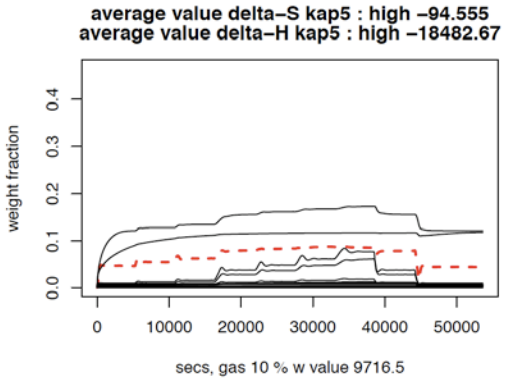
# EVALUATE CODE FUNCTION AND SENSITIVITY




Initial simulation experiment: 12 variables, 128 runs using an orthogonal array based Latin Hypercube Sample.

Code Function: Identify appropriate input ranges and impacts on code results.

Sensitivity analysis: Identify trends of a simple scalar metric calculated from TGA curves, mean weight fraction, with individual or pairs of parameters.

**Look for settings where weight increases: $\Delta H_{k5}$ and $\Delta S_{k5}$.**

# Ideal Equilibrium Model

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

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

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- 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.

# Calibration

$$Z = Y(\theta^*) + \delta + \varepsilon$$

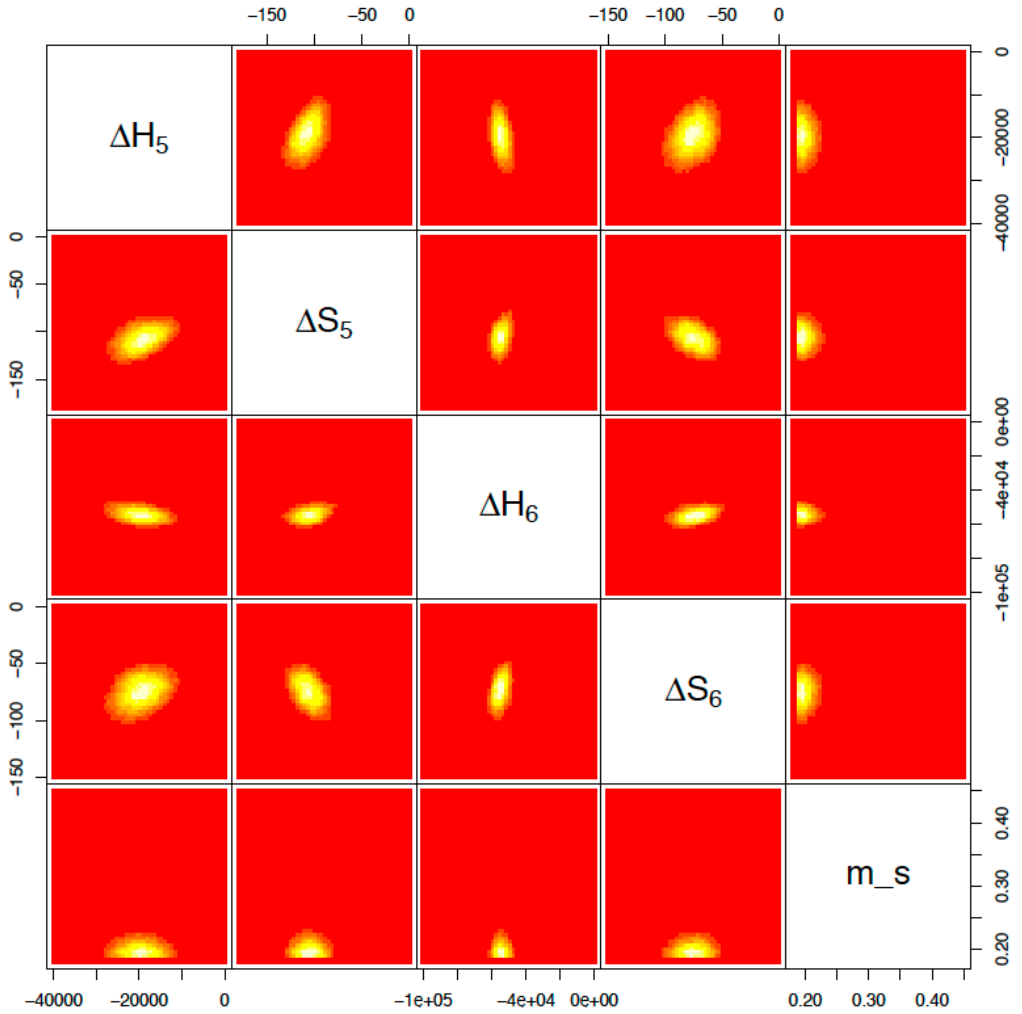
- Model discrepancy δ is a zero-mean Gaussian Process.

$$\delta \sim N(\mathbf{0}, \Sigma) \qquad \varepsilon \sim \mathrm{N}(\mu, \varphi\mathbf{I})$$

$$\Sigma_{ij} = \kappa \exp\left( -\frac{T_i - T_j}{\phi_T} - \frac{p_i - p_j}{\phi_p} \right)$$

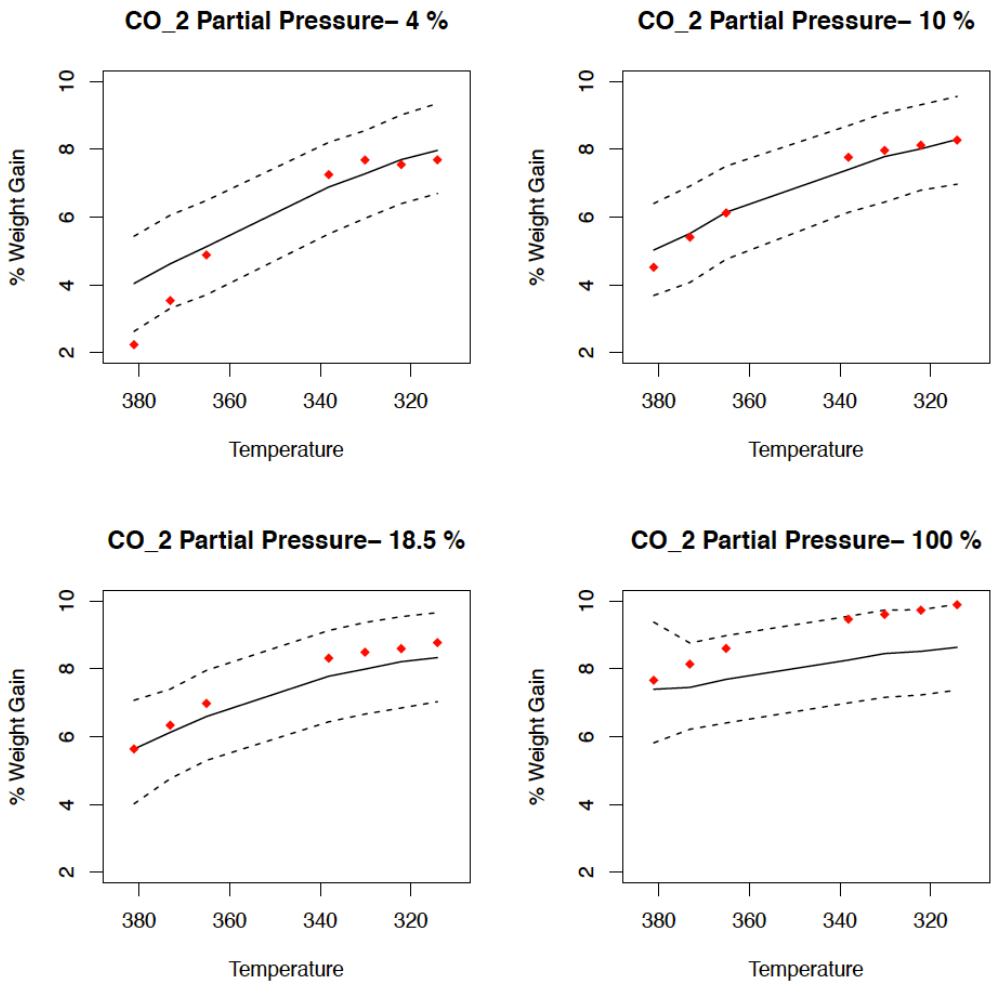
- Priors on $\theta^*$ based on scientific information. Tight priors for other parameters
- Known confounding between $\theta^*$ and δ.
- Posterior distribution of $f(\theta,\kappa,\phi,\phi,\psi|Z,Y)$ obtained using MCMC.

# Calibration-Ideal Equilibrium Model




- Bayesian approach incorporates expert prior information about parameters and quantifies uncertainty, including model and observation error.
- Color-scale bivariate posterior probability densities for pairs of parameters – shows most likely parameter values for simulation agreement with physical observations.

# Calibration-Ideal Equilibrium Model




- Physical observations in red.
- Simulation model predictions and bounds (95%) for 4 cases of $CO_2$ pressure.

# Experiment Planning

- Opportunity to obtain experimental data that will aid the process of model development and assessment.
- 8 factors of interest for initial set of runs. One factor is difficult to change.
- Developed a 2**(8-4) design split in half, with the last 2 columns completely confounded.
- Phase 1: Run 1st half of design, plus 3 center points (11 runs) to obtain preliminary information on effects of factors, possible nonlinearity, and error.
- Phase 2: Following initial analysis, option to run 2nd half of design, plus additional center point.

# Continuing Efforts

- Estimate parameters for other sorbent models that may include additional effects, such as water.
- May need to consider use of surrogate models (emulators) for more complex models .
- Design and analyze simulation and experimental studies to obtain additional data that can be incorporated into the modeling process.
- Examine propagation of uncertainty as results are scaled up and impact on uncertainty in higher level system.
- Consider validation of Computational Fluid Dynamics models in conjunction with uncertainty quantification.

# Thank You!