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Carbon Capture Simulation Initiative

CFD model validation of a small scale carbon capture unit

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National Energy Technology Laboratory

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AIChE Annual meeting, November 2014, Atlanta, GA.

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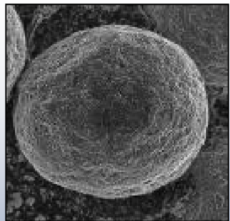

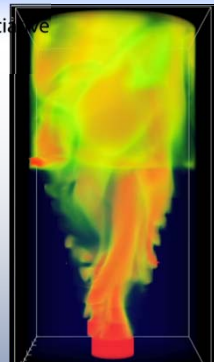

Rapidly synthesize optimized processes to identify promising concepts → **Better understand internal behavior to reduce time for troubleshooting** → **Quantify sources and effects of uncertainty to guide testing & reach larger scales faster** → **Stabilize the cost during commercial deployment**

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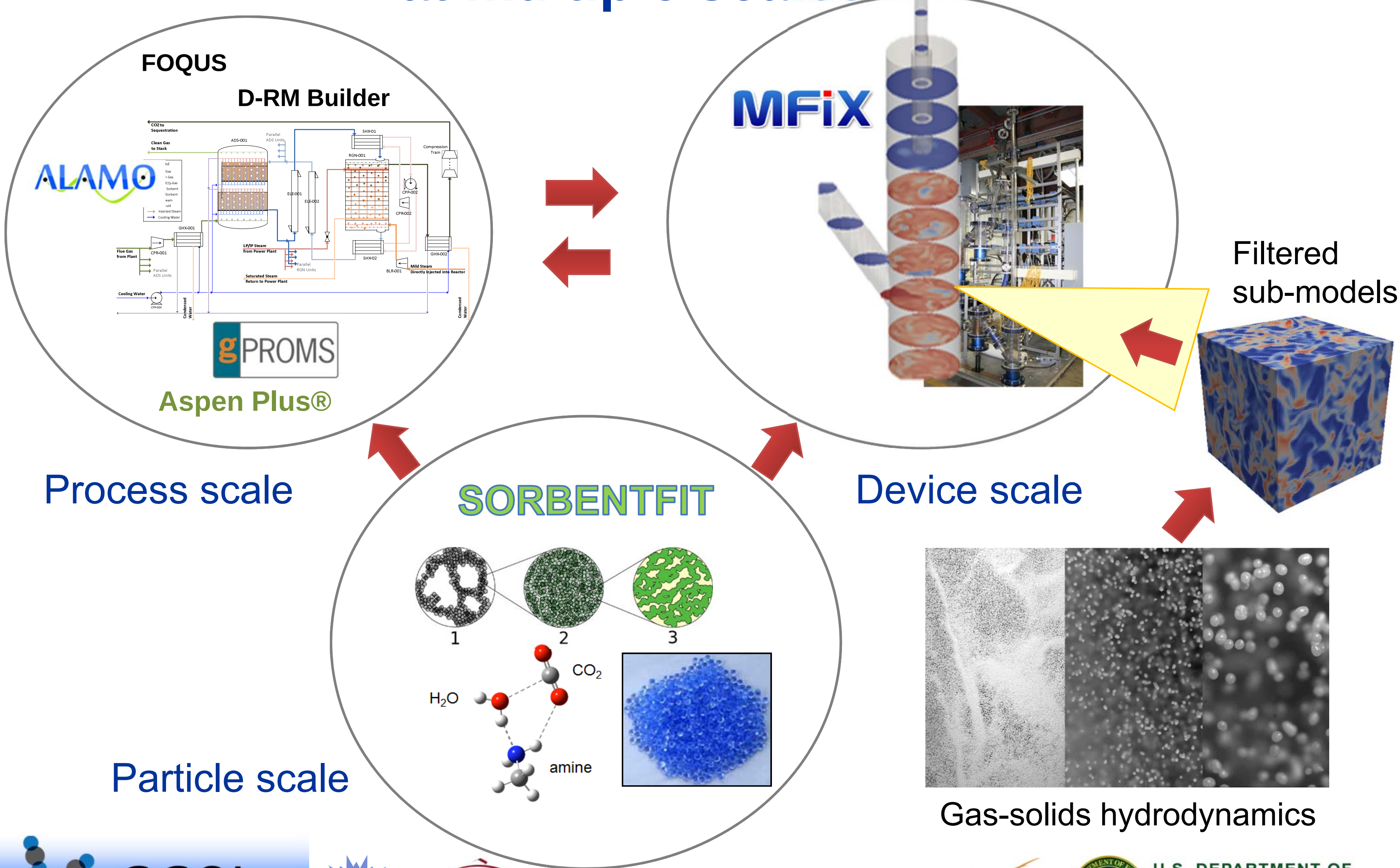
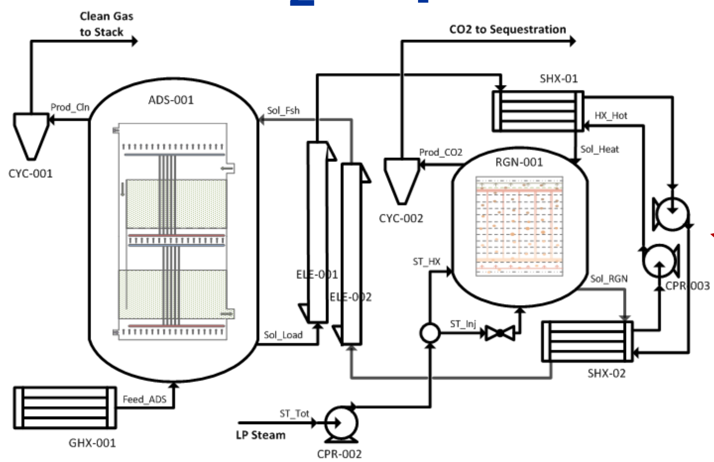
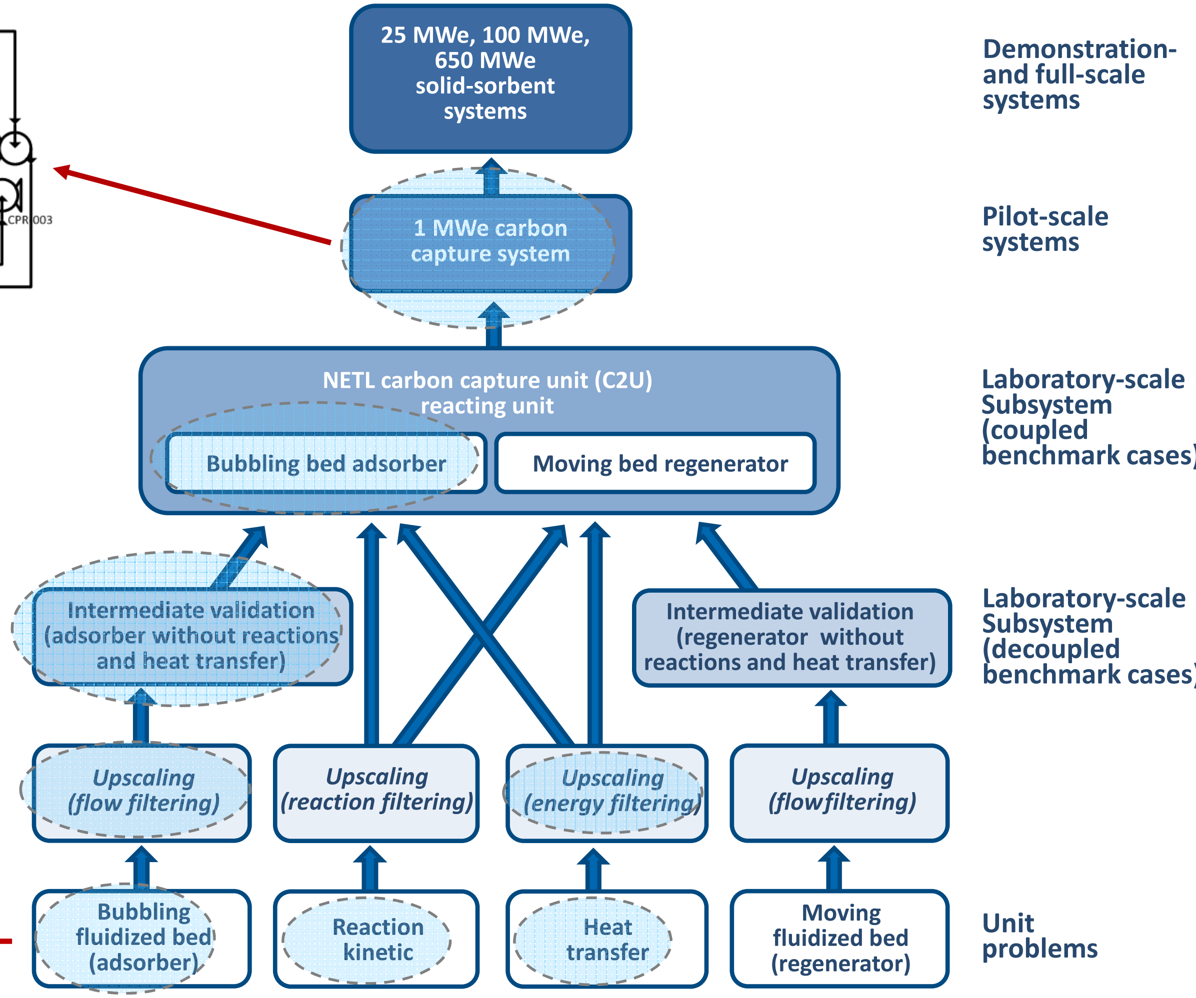
D.C. Miller et al., "Carbon Capture Simulation Initiative: A Case Study in Multiscale Modeling and New Challenges," *Annu. Rev. Chem. Biomol. Eng.*, 2014. 5:301–23

CCSI toolset enables the integration of models at multiple scales



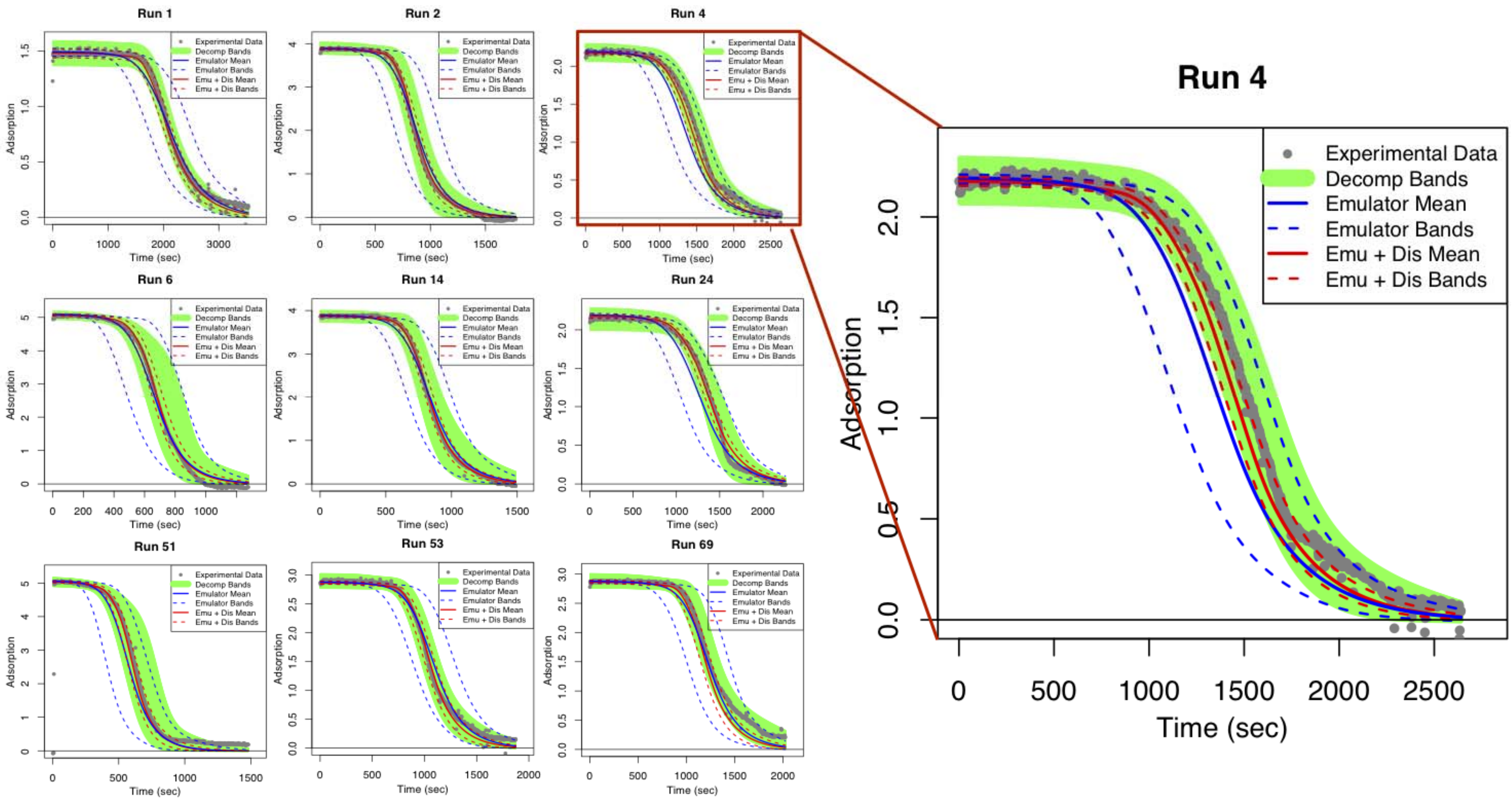
Building predictive confidence for device-scale CO_2 capture with multiphase CFD models





Ryan E.M., Montgomery C., Storlie C., Wendelberger J. CCSI validation and uncertainty quantification hierarchy for CFD models. CCSI Tech. Rep. Ser., 2012.

"The emulator prediction bands are within observation error in all cases"*



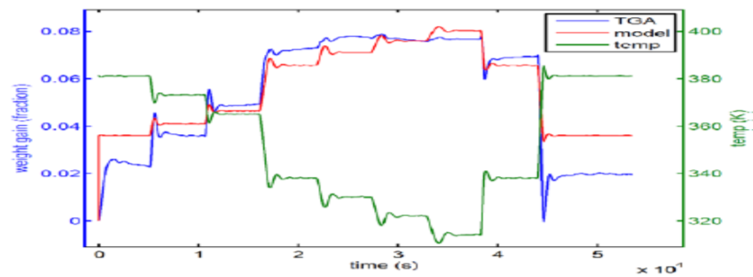
* K. Lai, Z. Xu, W. Pan, L. Shadle, C. Storlie, J. Dietiker, T. Li, S. Dartevelle, X. Sun, "Hierarchical Calibration and Validation of High-fidelity CFD Models with C2U Experiments ," CCSI Milestone Report, 2014.

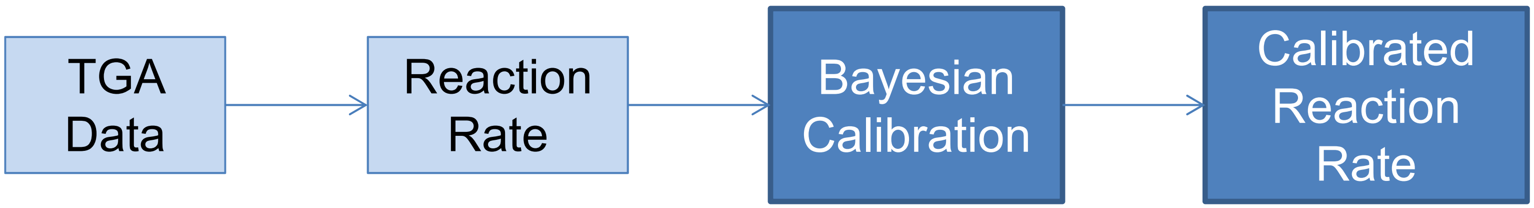
Is the reaction kinetics derived from TGA adequate?

- **CCSI sorbent kinetics (Bhat et al., 2012)**

$$2R_2NH + CO_2(gas) \rightleftharpoons R_2NCO_2^- + R_2NH_2^+$$
$$R_2NH + H_2O(phys) + CO_2(gas) \rightleftharpoons HCO_3^- + R_2NH_2^+$$
$$H_2O(gas) \rightleftharpoons H_2O(phys)$$

$$k_i = A_i T exp\left(\frac{-E_i}{RT}\right) \quad K_i = exp\left(\frac{\Delta S_i}{R}\right) exp\left(\frac{\Delta H_i}{RT}\right)/P$$

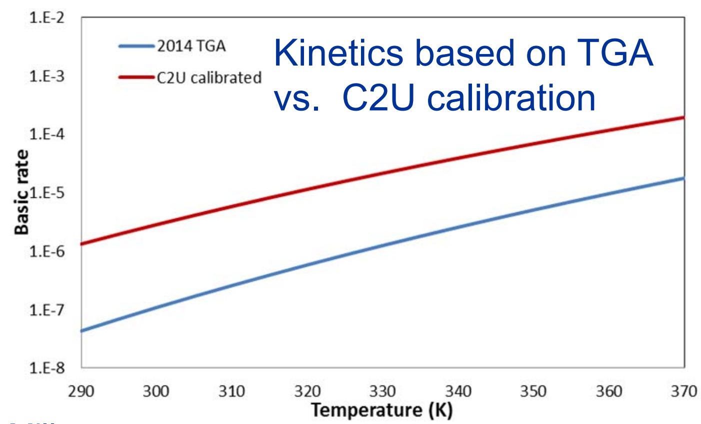




- The calibrated reaction rate parameters differed considerably from the parameter values obtained from TGA data

- Understand the cause of discrepancy
  - Inherent limitation of rate derived from TGA?
  - Predicted bed hydrodynamics is not correct?

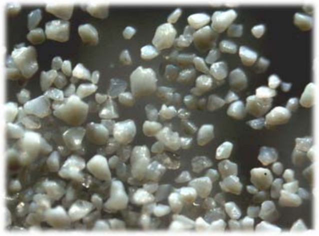
- Simplify the hydrodynamics
  - Remove the heat transfer coil
  - Run fixed to bubbling bed tests



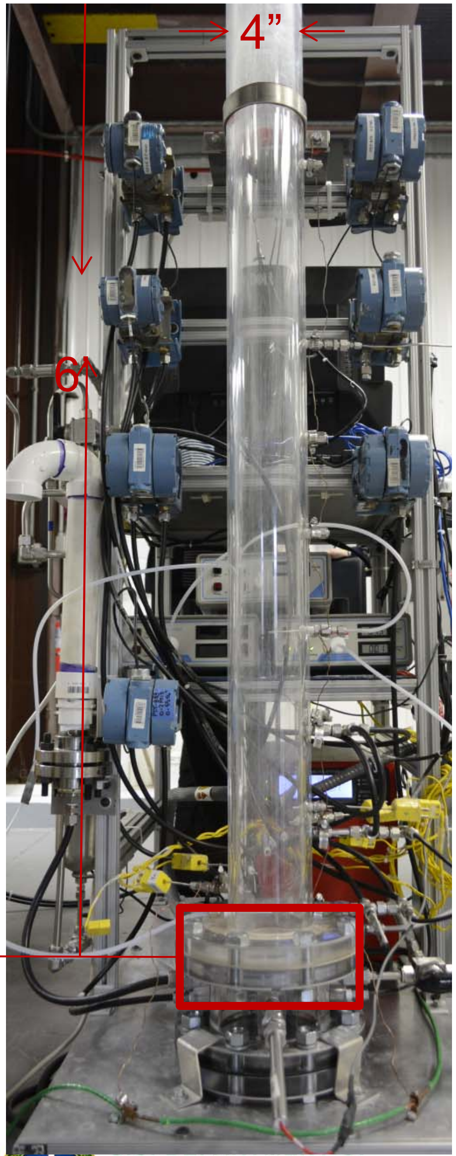
K.S. Bhat, D.S. Mebane, H. Kim, J. Eslick, J.R. Wendelberger, D.C. Miller, LANL Tech. Rep. LA-UR-12-21855, 2012.

Mini-C2U: well controlled experiments

- **Materials**
  - NETL 32D (100μm, 0.48 g/cc)
  - ADA-ES sorbent
- **Cold flow in bubbling beds**
  - Pressure drops, visual observation
  - Experimental runs
    - Static bed heights (4, 6, 8 in)
    - Gas flow: 1, 3, 5, 7 u_{mf}
    - 7 repeats for most conditions
- **Reacting flow in fixed and bubbling beds**
  - Pressure drops, temperature, breakthrough curve
  - Experimental runs
    - Various superficial gas velocity, bed height
    - Different CO_2 concentrations



Sorbent 32D



Carefully designed distributor ensures uniform flow in fixed bed.

*Presented at **National Lab Day on the Hill***

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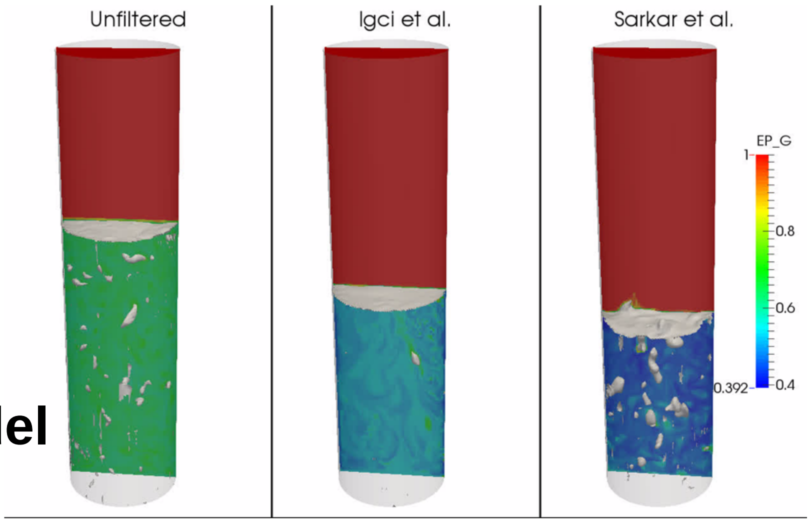
First, get the hydrodynamics right

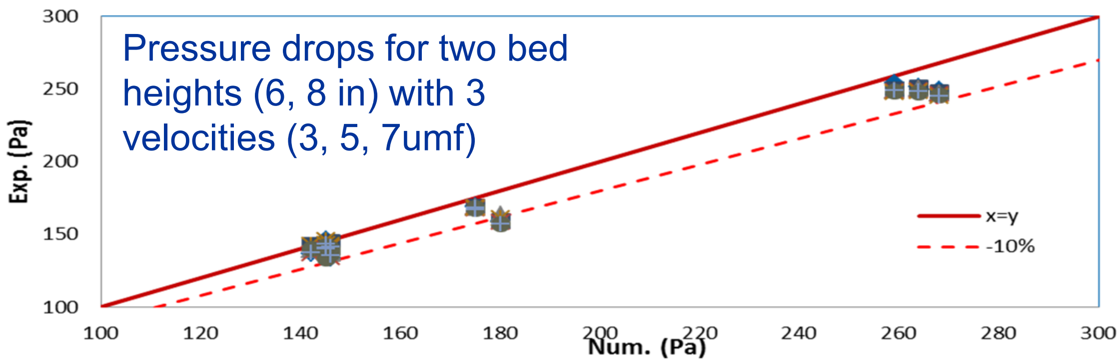
- **Tested three models: Unfiltered model and the filtered models of Igci et al. (2008) and Sarkar et al. (2014)**

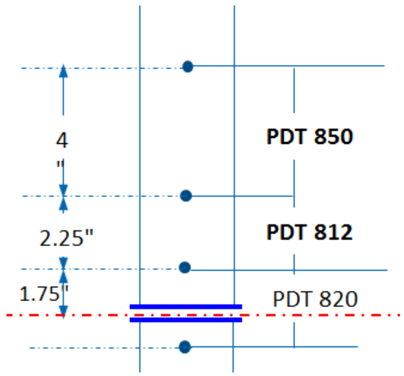
Model	Unfiltered	Igci et al.	Sarkar et al.	Exp.
Bed height (cm)	26	19.8	18	19



- **Selected Sarkar et al. (2014) filtered model**
  - Based on 3D periodic domain simulations
  - Filtered expressions based on two markers (ε_s, V_g-V_s)
  - Predicted bubbling behavior, qualitatively the best

MFIX Simulations

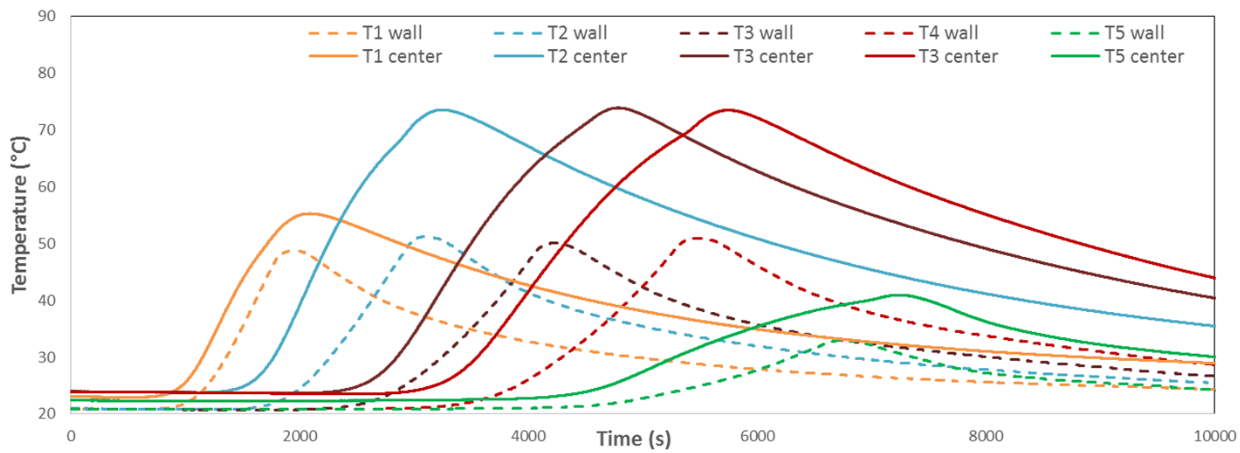




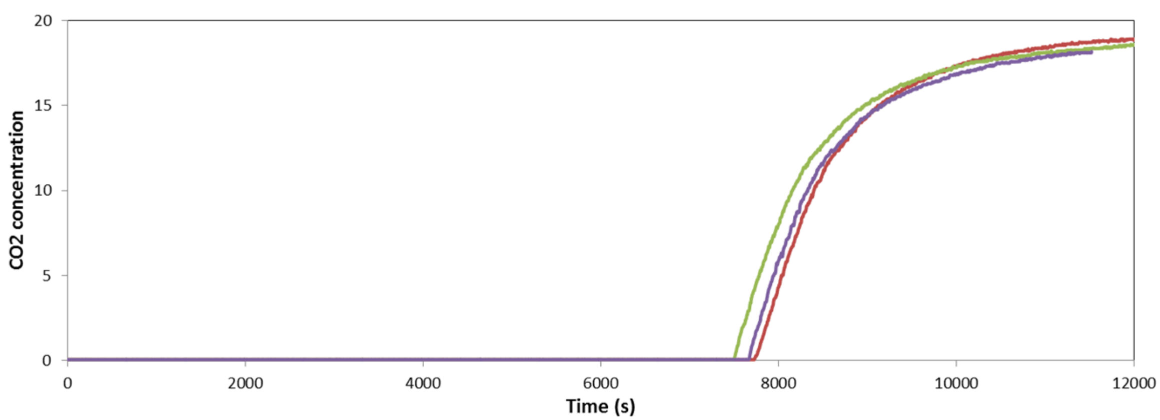
Igci, Y., Andrews, A. T., Sundaresan, S., Pannala, S. and O'Brien, T. AIChE J., 54, 1431–1448, 2008.
Sarkar, A., Sun, X. and Sundaresan, S., AIChE Annual Meeting, Paper 407b, 2014.

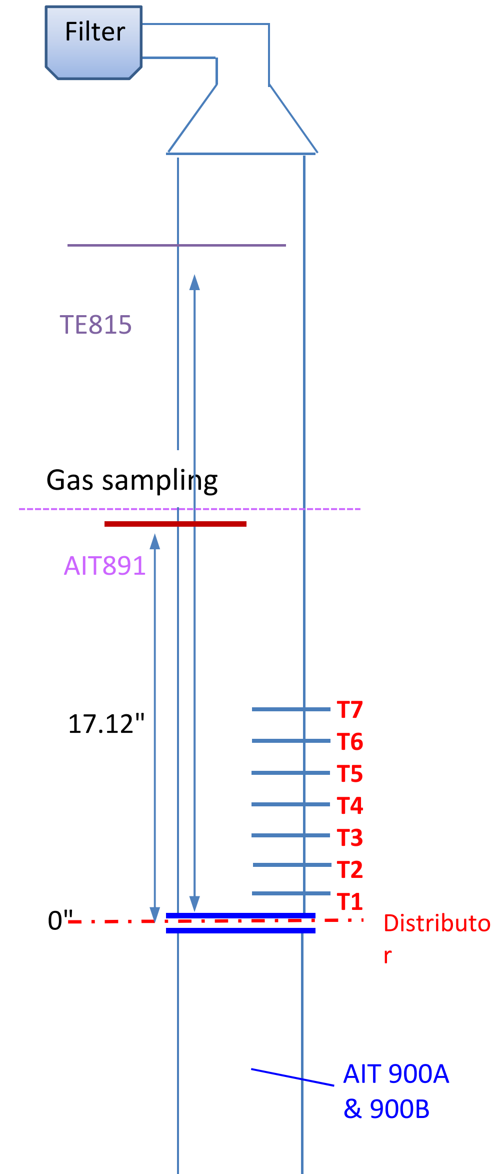
CO_2 adsorption in a fixed bed

- **Operating conditions**
  - 0.8 u_{mf}, N2:CO2 = 0.8:0.2, 6" bed height
- **Center and wall temperatures**



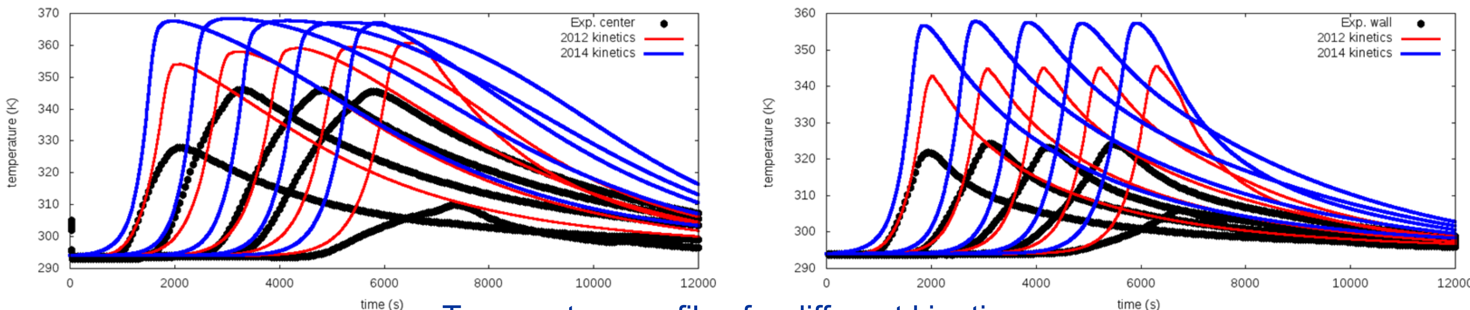
- **Break-through curve (3 repeats)**





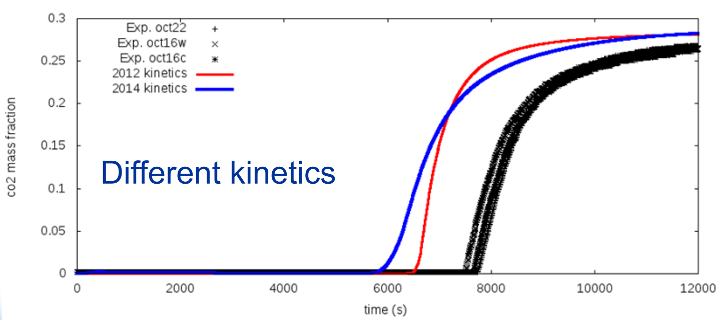
Results of fixed bed parametric study

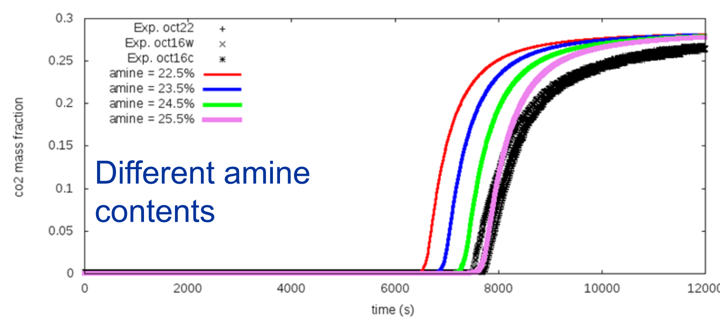
- **Temperature profile**
  - Pattern is captured, but the peak value is over predicted
  - Heat transfer to ambient is considered, which is considerable in the current setting
  - Thermal properties (Cp & k) and reaction rates affect the peak temperature



Temperature profiles for different kinetics

- **CO_2 breakthrough curve**
  - Breakthrough time is under predicted
  - Amine concentration and reaction rate affect the breakthrough time





Summary

- **Preliminary conclusions**
  - The hydrodynamics predicted with Sarkar et al. (2014) filtered model agrees well with experimental data
  - Need accurate measurements of Cp and k
  - Reaction kinetics need to be improved

- **Future plans**
  - Simulate experiments with only heat transfer for verification
  - Compare predicted temperature profiles and break through time with fixed bed data, and calibrate reaction rate parameters
  - Validate bubbling bed simulations with experimental data on CO2 breakthrough and temperatures at different locations as functions of time
  - Apply the CCSI validation hierarchy to ADA-ES process

Acknowledgements

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