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Carbon Capture Simulation Initiative

Uncertainty quantification in chemistry sub-models

David S. Mebane,[†,*] K. Sham Bhat,[§] Joel D. Kress,[§] Lisa M. Moore,[§] Daniel J. Fauth,[*] McMahan L. Gray[*]

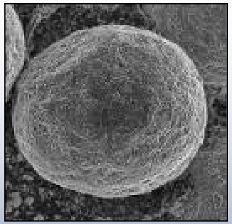
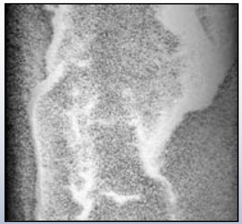
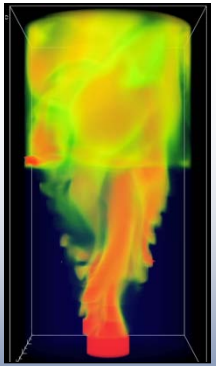
[*]National Energy Technology Laboratory

[§]Los Alamos National Laboratory

[†]ORISE Postdoctoral Fellow

U.S. DEPARTMENT OF ENERGY

Carbon Capture Simulation Initiative


Identify promising concepts → Reduce the time for design & troubleshooting → Quantify the technical risk, to enable reaching larger scales, earlier → Stabilize the cost during commercial deployment

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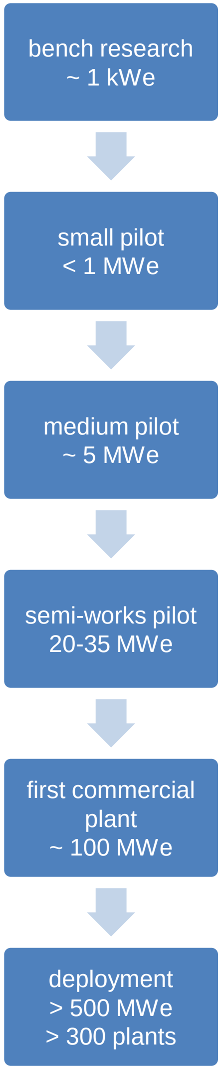
Carbon Capture Challenge

- The traditional pathway from discovery to commercialization of energy technologies can be quite long, i.e., ~ 2-3 decades
- President's plan requires that barriers to the widespread, safe, and cost-effective deployment of CCUS be overcome within 10 years
- To help realize the President's objectives, new approaches are needed for taking carbon capture concepts from lab to power plant, quickly, and at low cost and risk
- CCSI will accelerate the development of carbon capture technology, from discovery through deployment, with the help of science-based simulations



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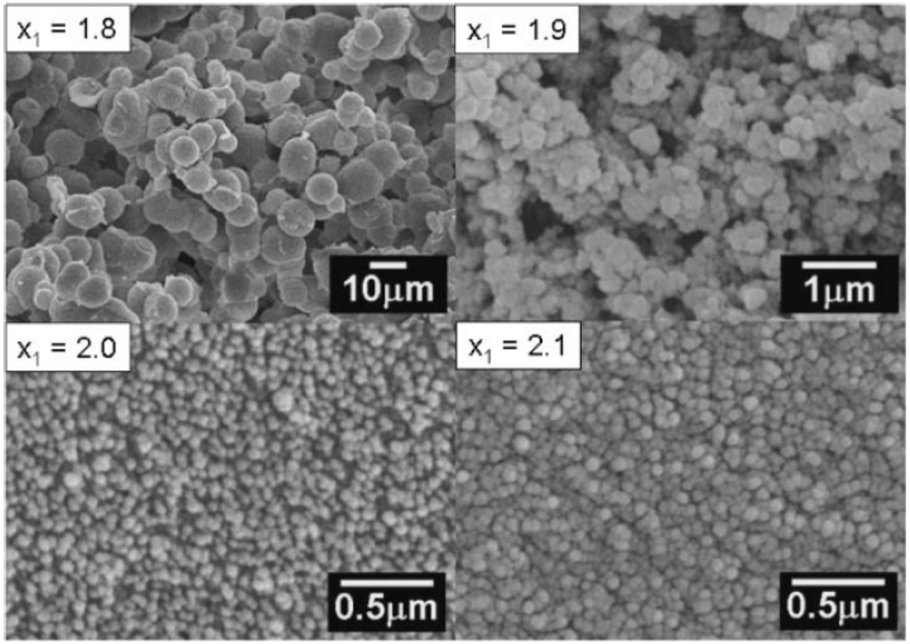
outline

- overview of high-fidelity sorbent modeling
- Bayesian calibration applied to sorbent equilibrium models

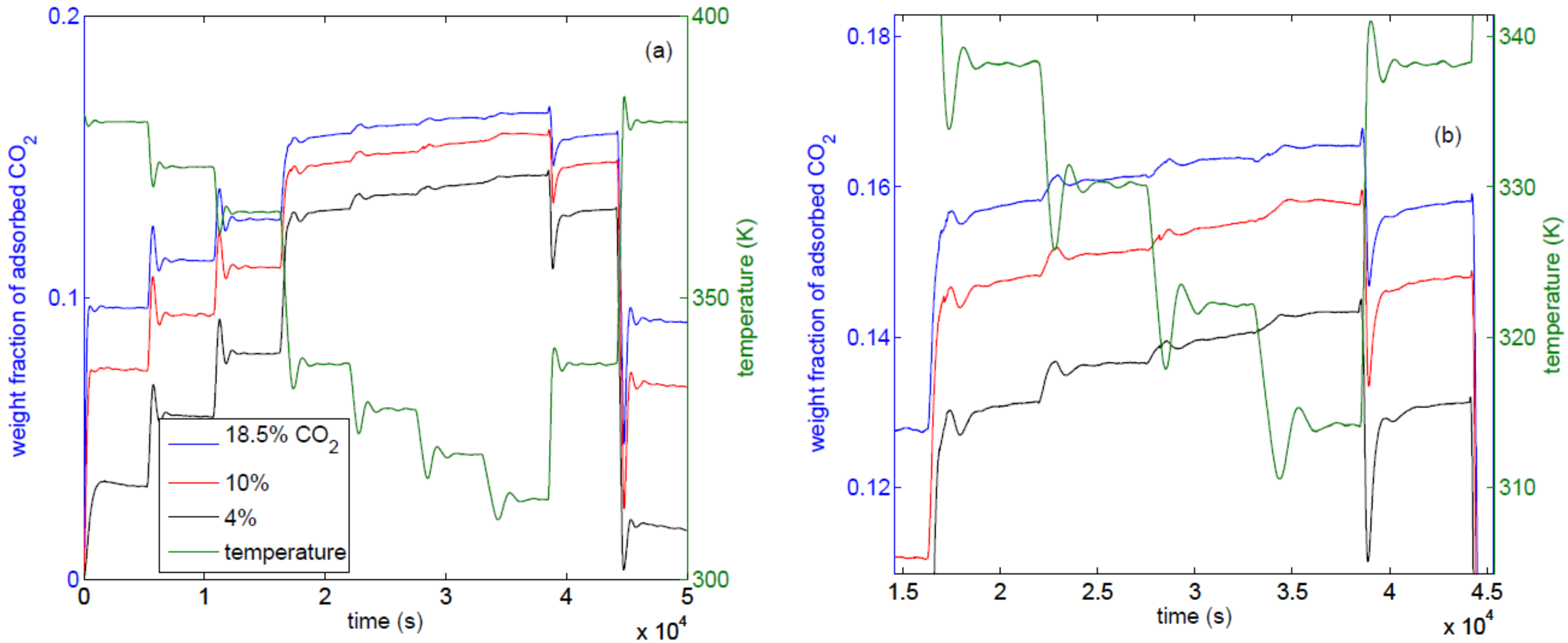
the sorbent

- mesoporous silica forms the substrate
- substrate particles agglomerates of micron-sized mesoporous particles
- mesopores impregnated with an active material, such as polyethyleneimine (PEI)



K Kajihara, et al., Bull Chem Soc Jpn, 82 (2009) 1470.

the sorbent: dry TGA behavior



(a)-(b) Sorbent NETL-196C, ~44.1 wt-% PEI, Dry atmosphere. Sorbent synthesis: McMahan Gray, NETL; Sorbent characterization: Daniel Fauth, NETL.

anhydrous model

- two-step formation of carbamic acid:

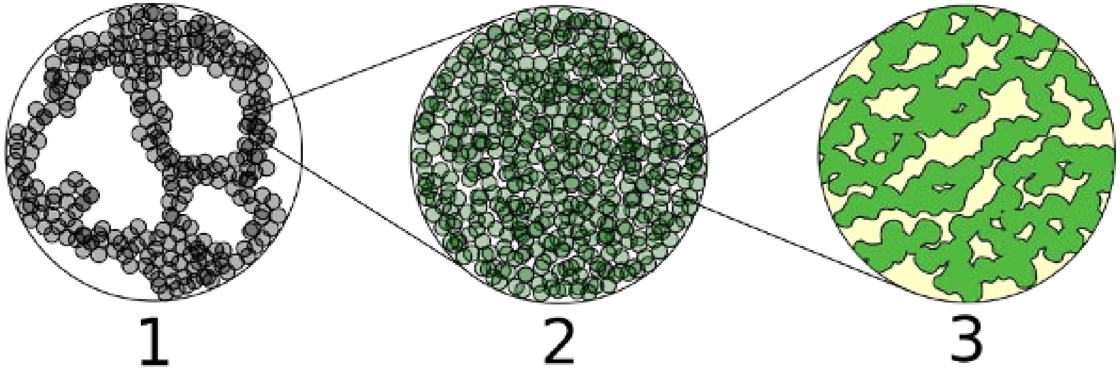
$$R_2NH + CO_2(g) \rightleftharpoons R_2NH^+ - CO_2^-$$
$$R_2NH^+ - CO_2^- + R_2NH \rightleftharpoons R_2NCOOH : R_2NH$$

- three modes of mass transport:

  gas phase bulk

  gas phase Knudsen

  solid state (zwitterion-mediated hopping)



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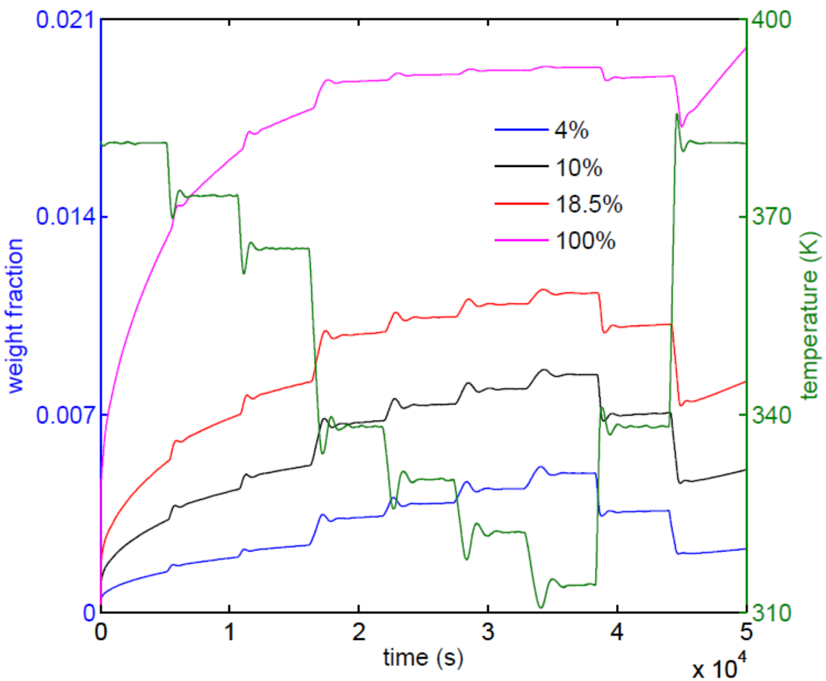
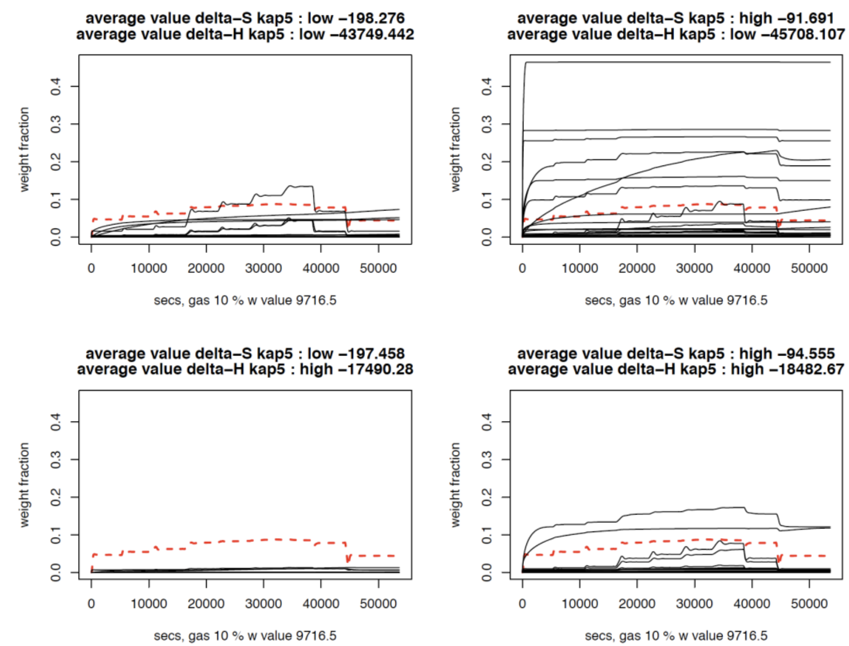
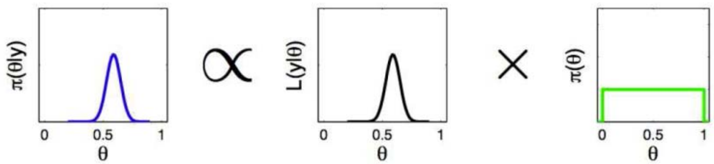




(left) sample calculated output of the sorbent model showing diffusion effects (right) sensitivity analysis highlighting the importance of zwitterion stability to sorbent working capacity

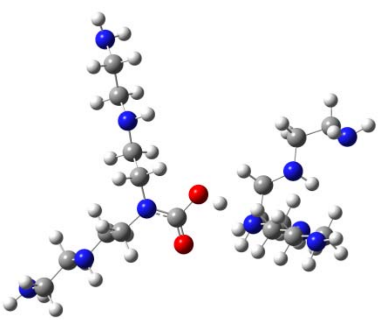
Bayesian methods in parameter estimation

- Bayes' theorem enables the incorporation of prior information in model-based parameter estimates.

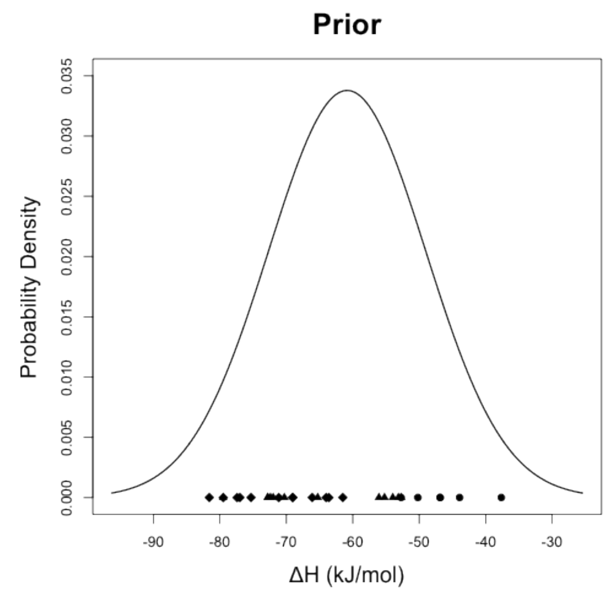


$$\pi(\theta|y) \propto L(y|\theta) \times \pi(\theta)$$

- If model parameters relate to physical quantities, prior information is available through *ab initio* calculations.



rxn	B3LYP	PBE	MP
1	-52.72	-76.36	-62.76
2	-46.86	-70.29	-62.97
3	-50.21	-72.8	-62.76
4	-46.86	-70.71	-64.43
5	-37.66	-69.04	-62.76
6	-43.93	-68.41	-72.38



DS Mebane, KS Bhat, JD Kress, et al., in prep.

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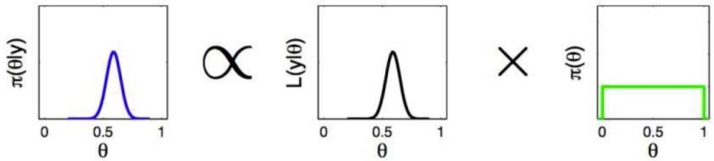
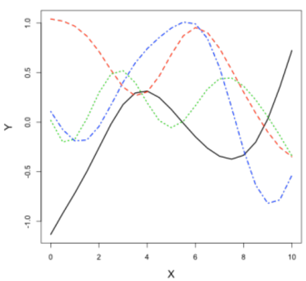
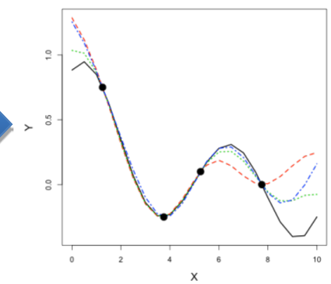


$$\pi(\theta|y) \propto L(y|\theta) \times \pi(\theta)$$

- The error in the form of the model must also be accounted for.
- A Gaussian process generates a stochastic set of curves adhering to certain general properties.

$$\Sigma(i', j'; \boldsymbol{\xi}) = \eta \exp\left[-\frac{(\zeta_{i'} - \zeta_{j'})^2}{\phi^2}\right]$$





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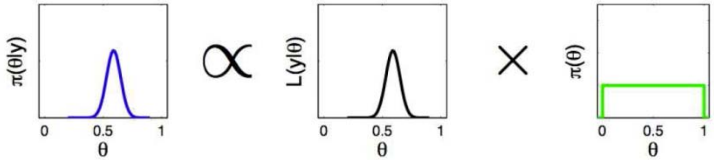

$$\pi(\theta|y) \propto L(y|\theta) \times \pi(\theta)$$

- The error in the form of the model must also be accounted for.
- A Gaussian process generates a stochastic set of curves adhering to certain general properties.

$$\mathbf{Y} = \mathbf{Z}(\boldsymbol{\theta}, \boldsymbol{\zeta}) + \boldsymbol{\delta}(\boldsymbol{\xi}, \boldsymbol{\zeta}) + \boldsymbol{\epsilon}(\psi)$$

$$\mathbf{Y} \sim N\left[\mathbf{Z}(\boldsymbol{\theta}, \boldsymbol{\zeta}), \Sigma(\boldsymbol{\xi}) + \psi\mathbf{I}\right] = \mathcal{L}(\mathbf{Y}|\boldsymbol{\theta}, \boldsymbol{\xi}, \psi)$$

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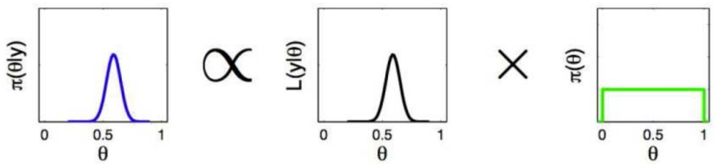

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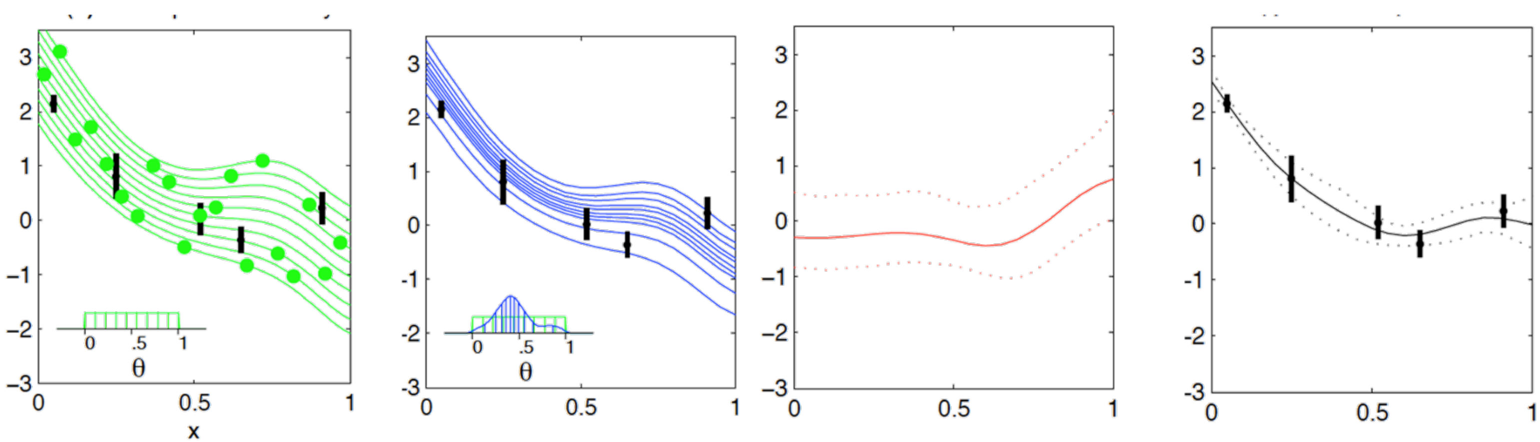

MC Kennedy and A O'Hagan, J Royal Stat Soc B, 63 (2001) 425.

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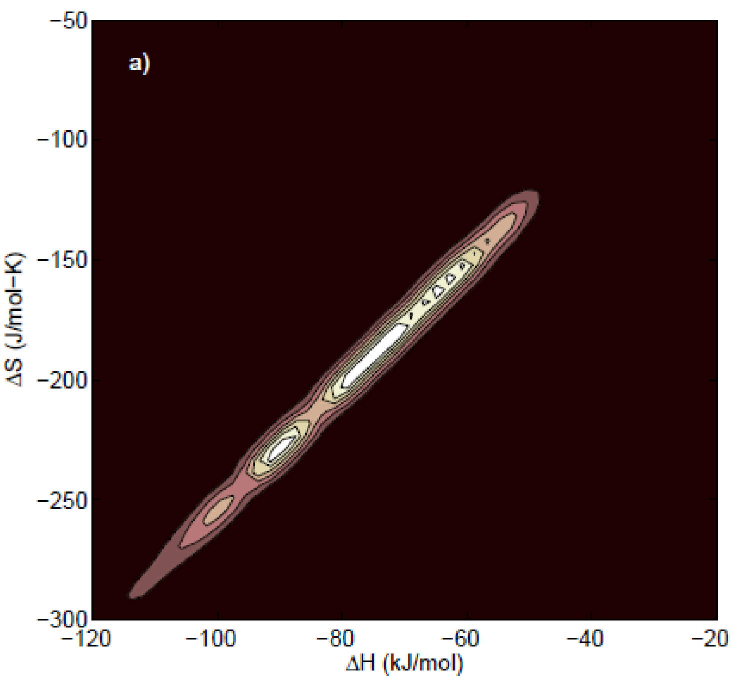
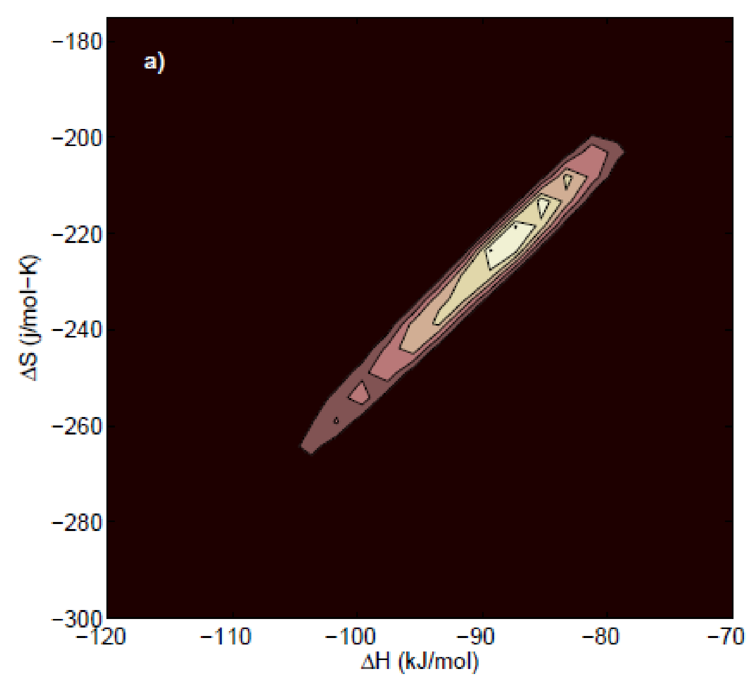
Bayesian methods in parameter estimation

$$2R_2NH + CO_2(g) \rightleftharpoons R_2NCOOH : R_2NH$$

$$\kappa = \frac{x^2}{(1-2x)^2 p} = \exp\left(\frac{\Delta S}{R}\right)\exp\left(\frac{-\Delta H}{RT}\right)/P \qquad w = M n_v x/\rho$$



posterior distributions (left) without and (right) with informative priors



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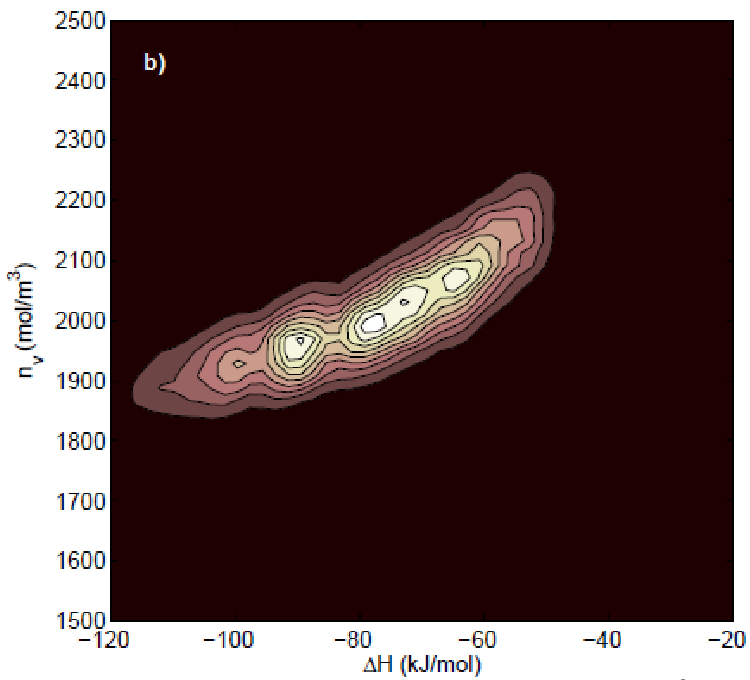

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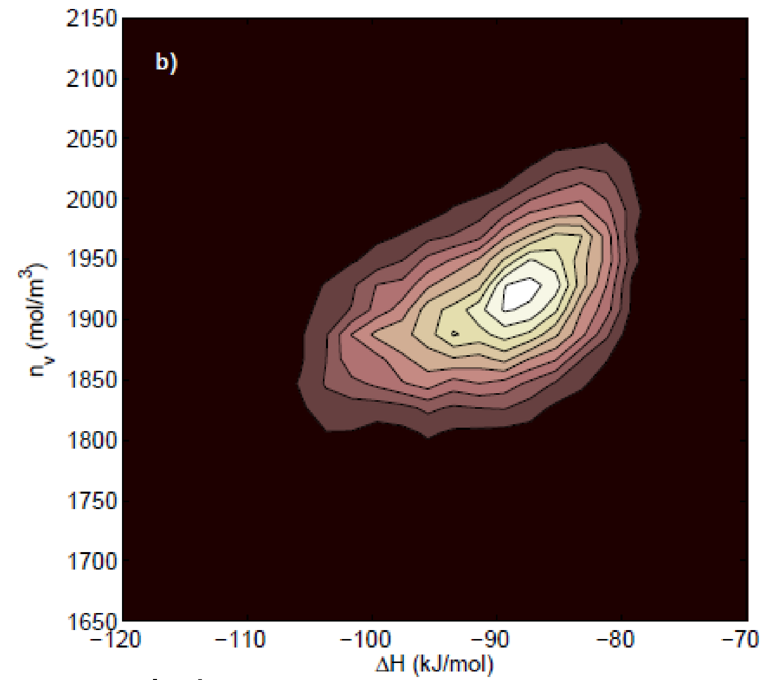

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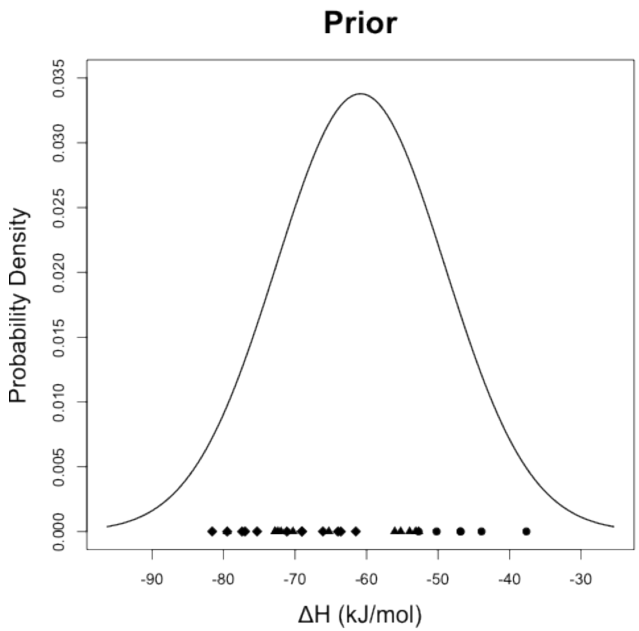
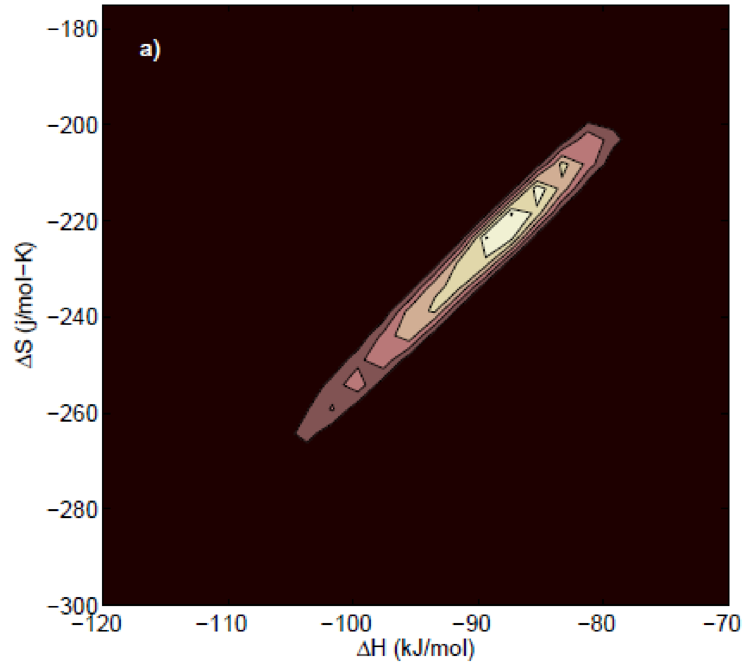


(left) prior distribution for adsorption enthalpy, and (right) posterior distribution



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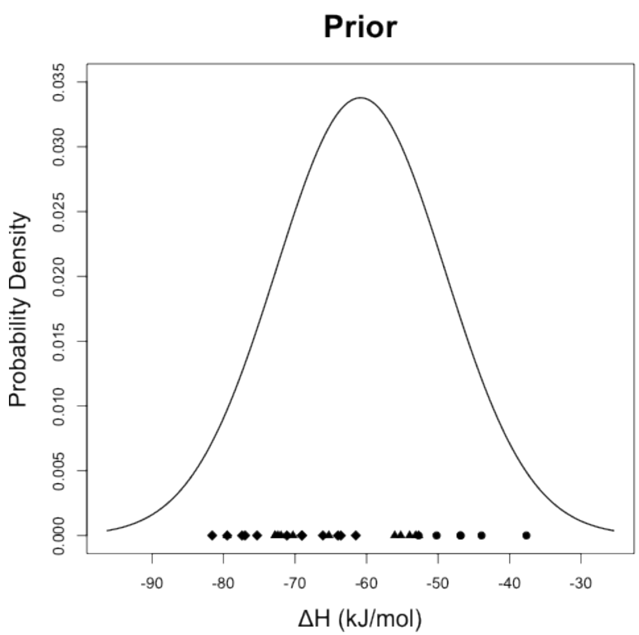
Bayesian methods in parameter estimation

reaction	B3LYP	PBE	PBE0	MP2	MP3
CO_2+2MMA→P-COOH:P	-52.72	-71.13	-81.59	-52.72	-72.8
CO_2+MMA+DMA→S-COOH:P	-46.86	-63.60	-76.99	-53.97	-71.96
CO_2+MMA+DMA→P-COOH:S	-50.21	-66.11	-79.50	-53.14	-72.38
CO_2+2DMA→S-COOH:S	-46.86	-64.02	-77.40	-56.07	-72.80
CO_2+DETA+EDA→P-COOH:S	-37.66	-69.04	-69.04	-55.23	-70.29
CO_2+DETA+EDA→S-COOH:P	-43.93	-61.50	-75.31	-65.27	-79.50



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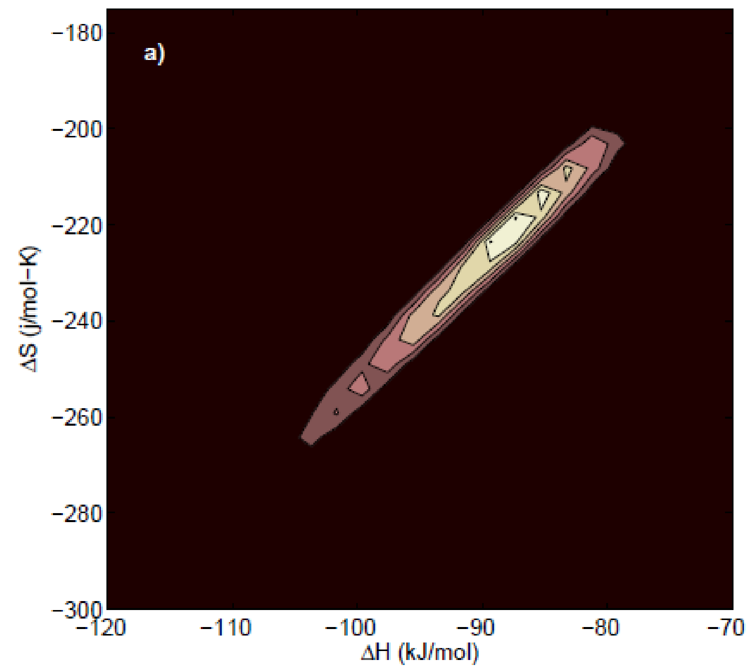

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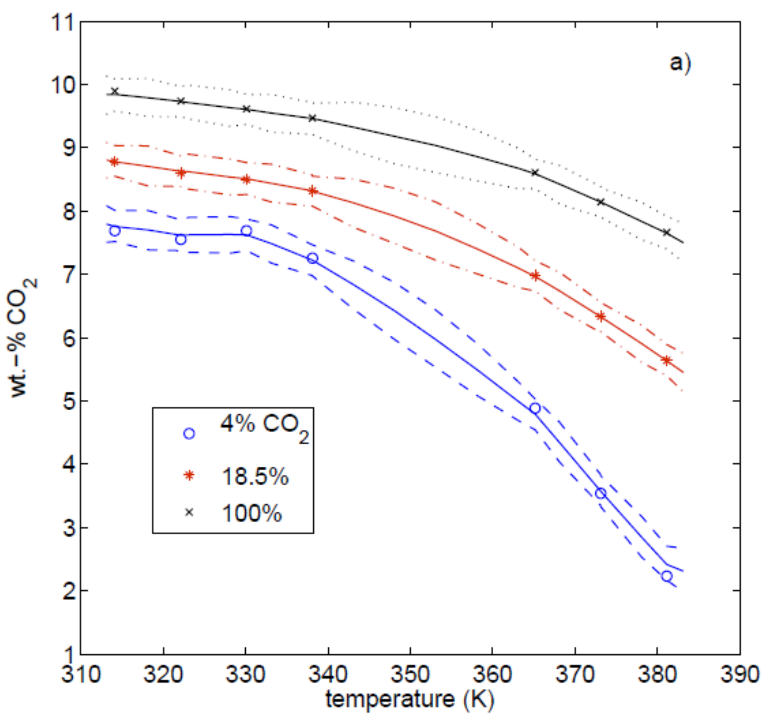
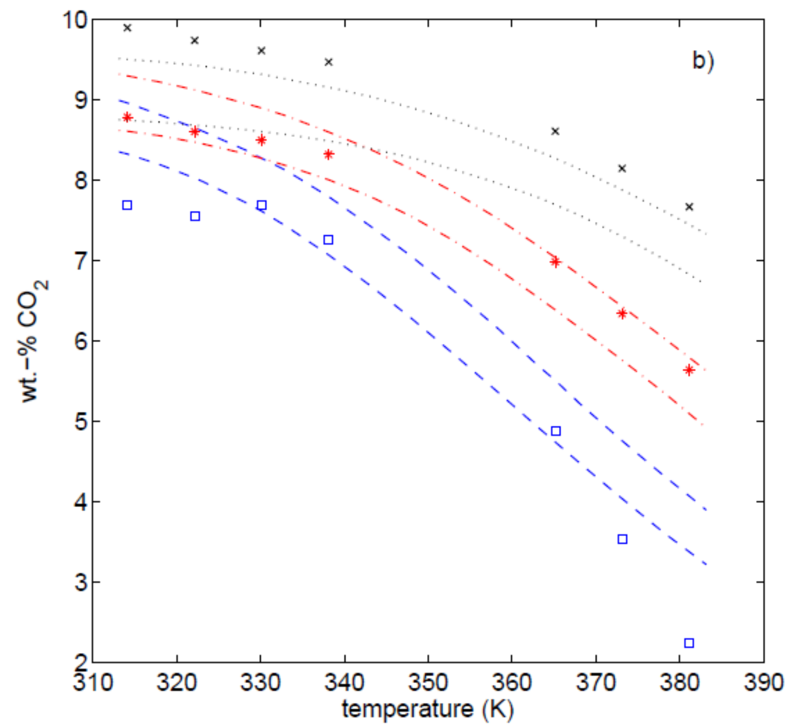


(left) conditioned model + discrepancy predictions, and (right) model predictions, with 95% confidence bounds



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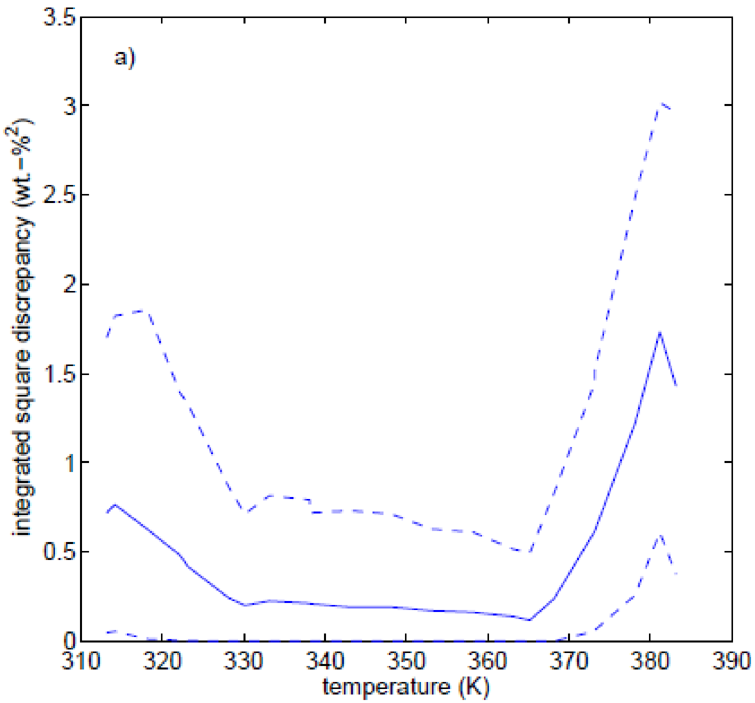
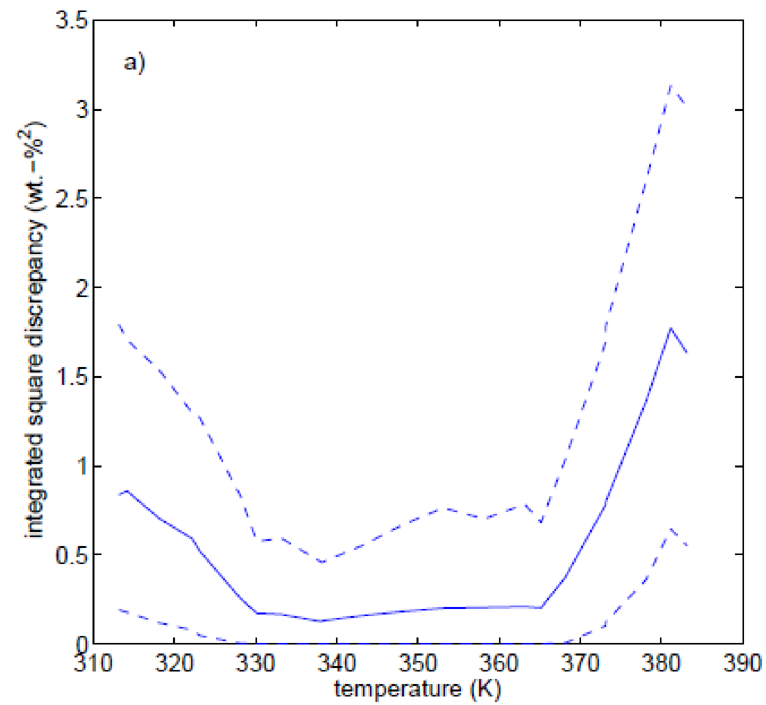


(left) normalized discrepancy for uniform priors with 95% bounds, and (right) normalized discrepancy for informative priors, 4% CO_2



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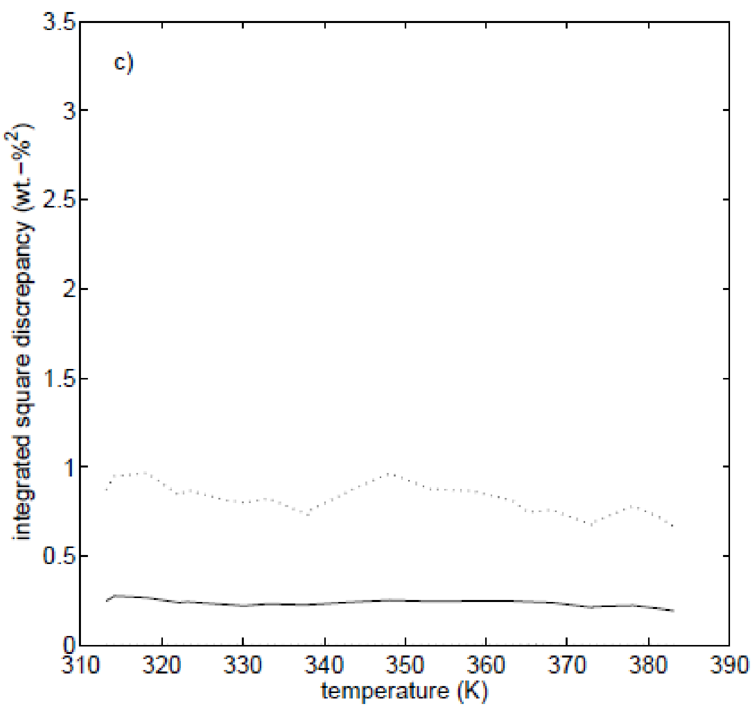
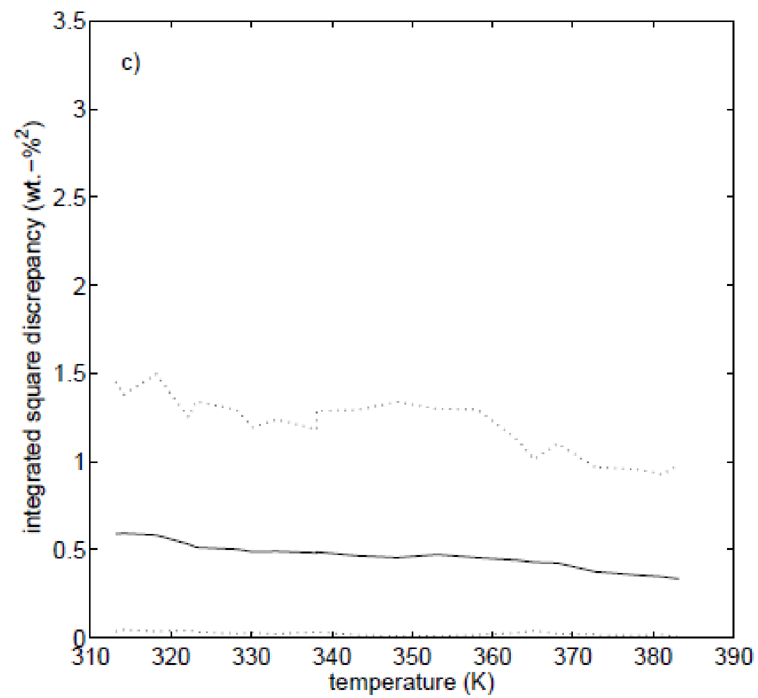


(left) normalized discrepancy for uniform priors with 95% bounds, and (right) normalized discrepancy for informative priors, 100% CO_2



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conclusions / future work

- *Ab initio* calculations can be used in along with a valid model form discrepancy in a Bayesian framework to influence the experimental calibration of engineering-useful models of complex chemical systems.
- A conditioned model form discrepancy enables the direct use of experimental information in scale-up through a model-plus-discrepancy approach, providing penalties for interpolation and extrapolation that become smaller as models improve.
- Work is underway to demonstrate the effects of model form discrepancy in upscaling of a simple kinetic model for CO_2 capture through to the process scale.

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