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Carbon Capture Simulation Initiative

Uncertainty quantification in chemistry sub-models

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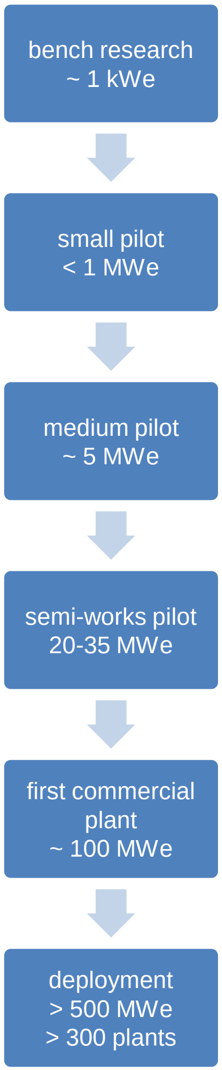
U.S. DEPARTMENT OF ENERGY

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 CCS 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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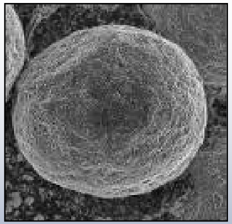 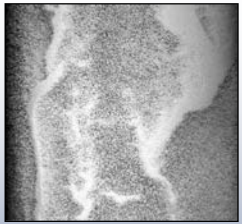 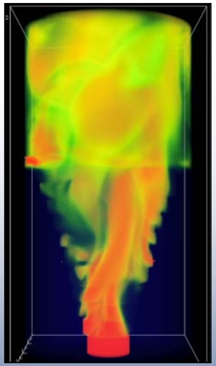   

Identify promising concepts → Reduce the time for design & troubleshooting → Quantify the technical risk, to enable reaching larger scales, earlier → Stabilize the cost during commercial deployment

National Labs	Academia	Industry
Berkeley Lab	Carnegie Mellon	Fluor
Pacific Northwest National Laboratory	Princeton University	ADA Environmental Solutions
NETL	West Virginia University	B&W the babcock & wilcox company
Los Alamos National Laboratory EST. 1943	Boston University	GE
Lawrence Livermore National Laboratory	Institute for Clean and Secure Energy The University of Utah	EPRI Electric Power Research Institute
		Air Products
		Burns & McDonnell Since 1898
		Duke Energy
		URS
		Alstom
		Southern Company
		Southern California Edison An Edison International Company San Onofre Nuclear Generating Station
		Boeing
		Chevron
		ExxonMobil
		WorleyParsons resources & energy
		Eastman

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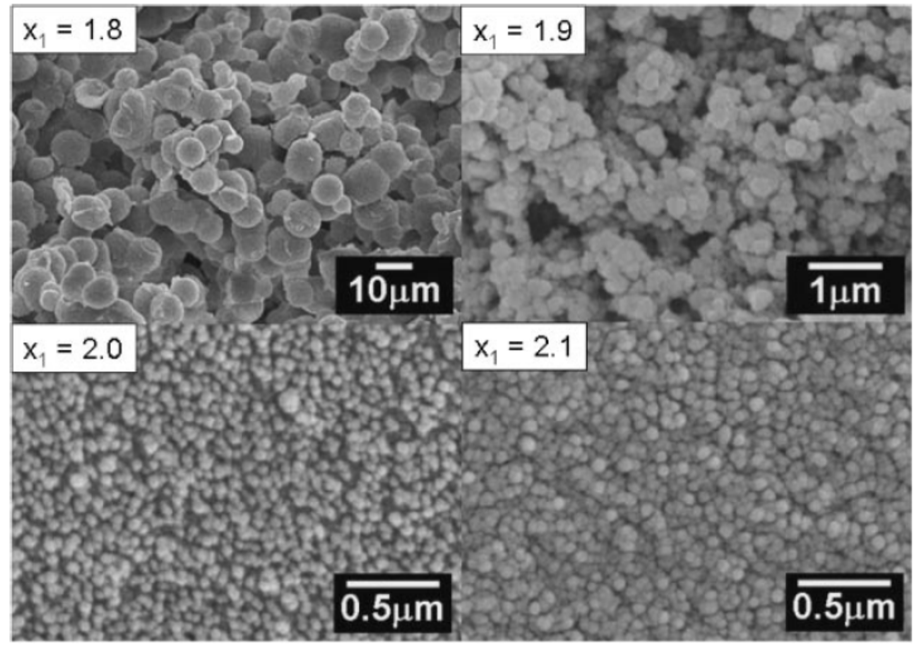
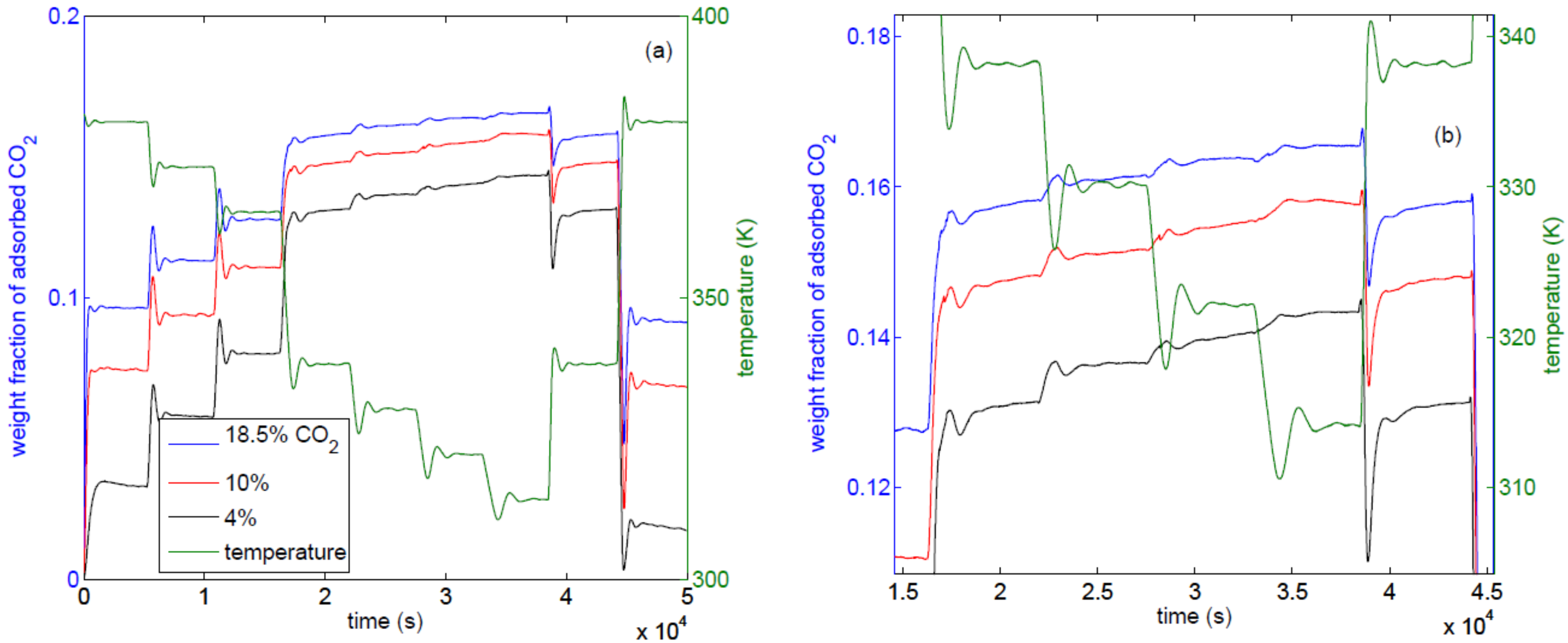
the sorbent

- 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



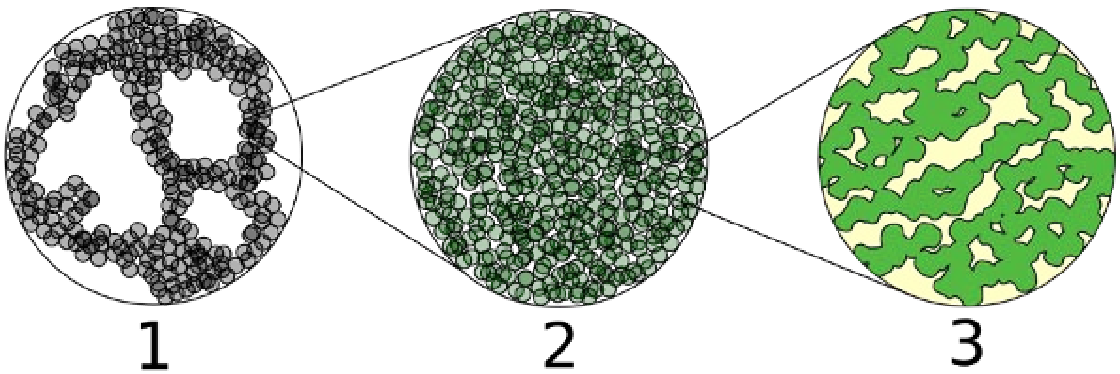
(a)-(b) Sorbent NETL-196C, ~44.1 wt-% PEI, Dry atmosphere. Sorbent synthesis: McMahan Gray, NETL; Sorbent characterization: Daniel Fauth, NETL.

anhydrous model

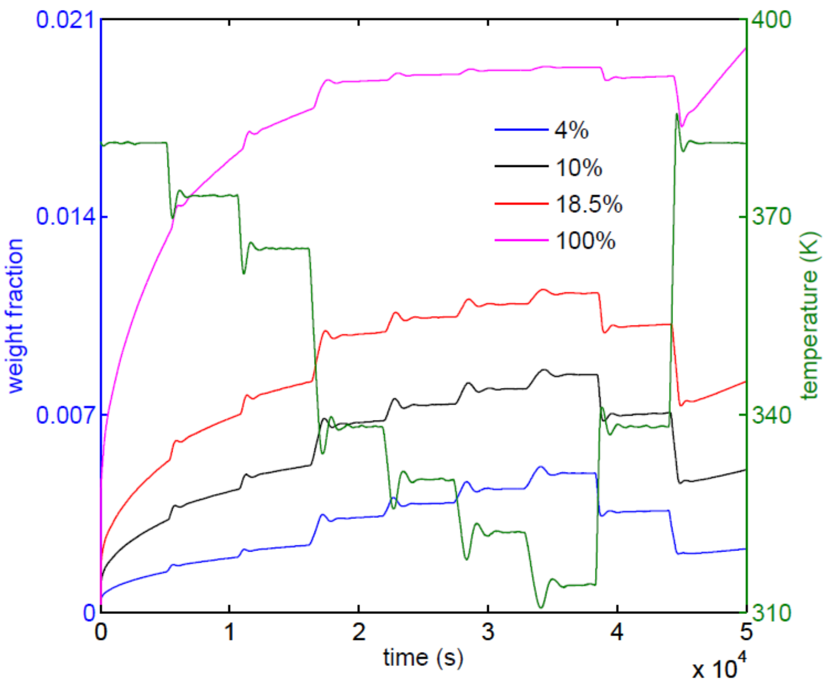
- two-step formation of carbamic acid:

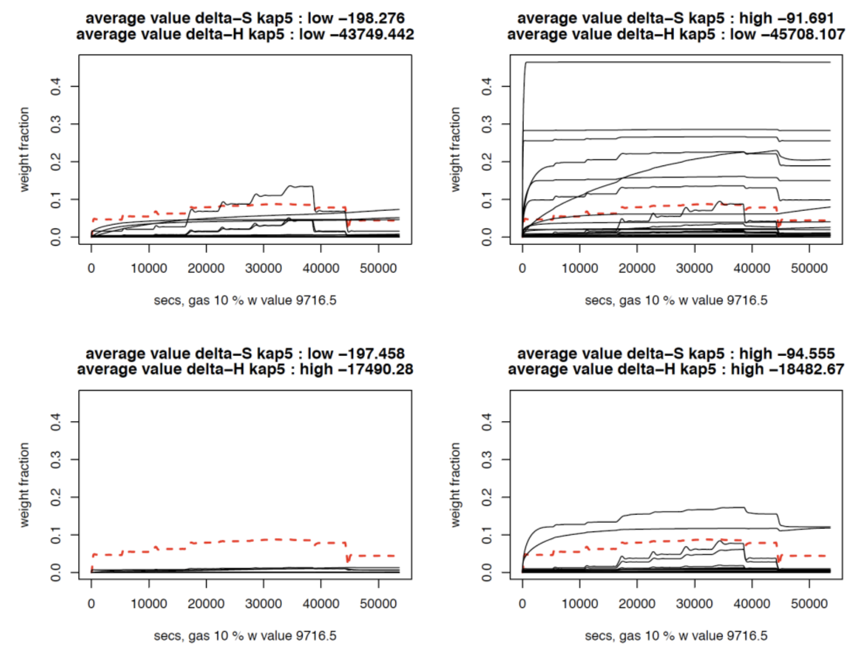
$$\mathrm{R_2NH + CO_2(g) \rightleftharpoons R_2NH^+ - CO_2^-}$$
$$\mathrm{R_2NH^+ - CO_2^- + R_2NH \rightleftharpoons R_2NCOOH : R_2NH}$$

- three modes of mass transport:
  - gas phase bulk
  - gas phase Knudsen
  - solid state (zwitterion-mediated hopping)



anhydrous model

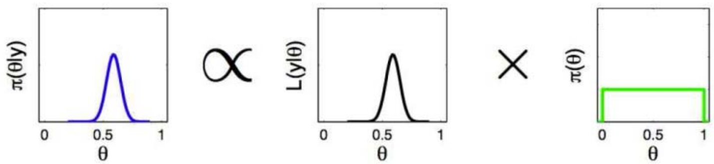




(left) sample calculated output of the sorbent model showing diffusion effects (right) sensitivity analysis highlighting the importance of zwitterion stability to sorbent working capacity

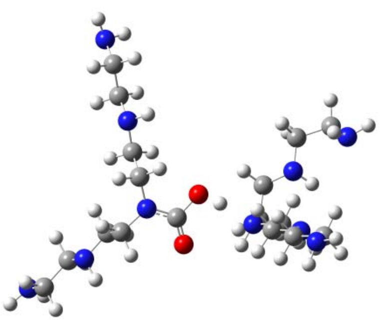
Bayesian methods in parameter estimation

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

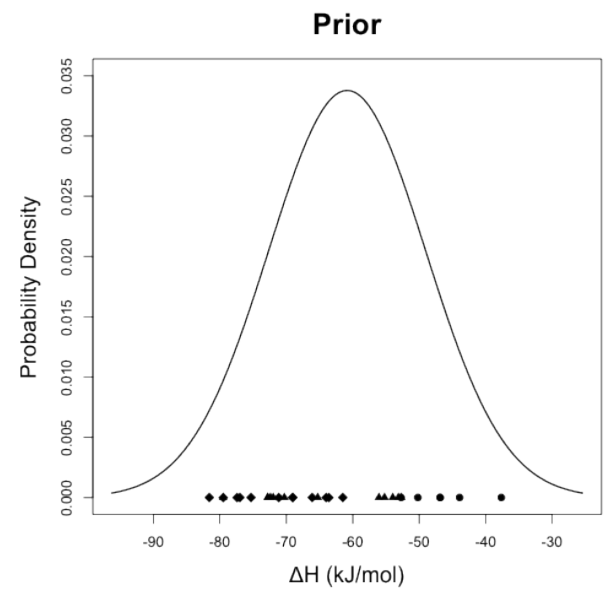


$$\pi(\theta|y) \propto L(y|\theta) \times \pi(\theta)$$

- 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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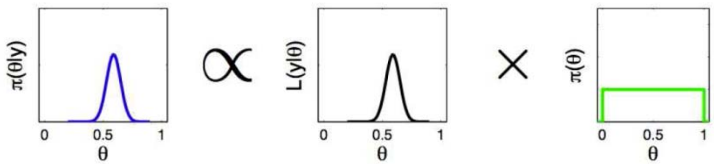
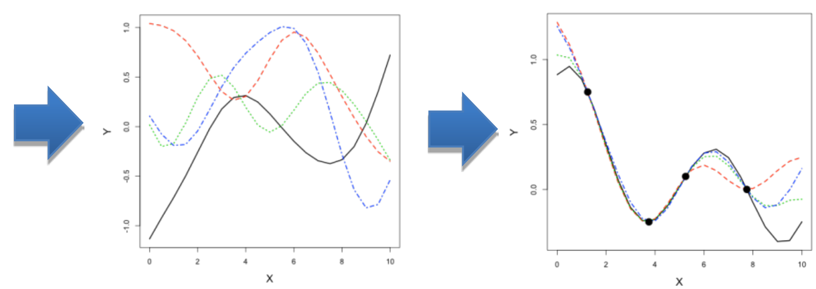


$$\pi(\theta|y) \propto L(y|\theta) \times \pi(\theta)$$

- 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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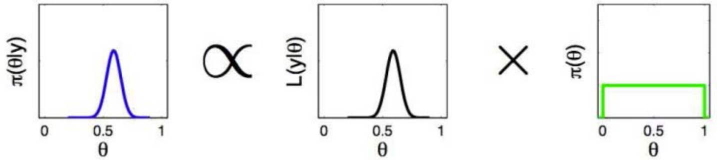

$$\pi(\theta|y) \propto L(y|\theta) \times \pi(\theta)$$

- 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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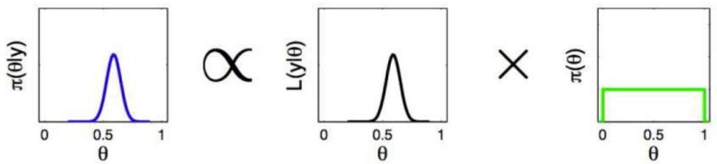

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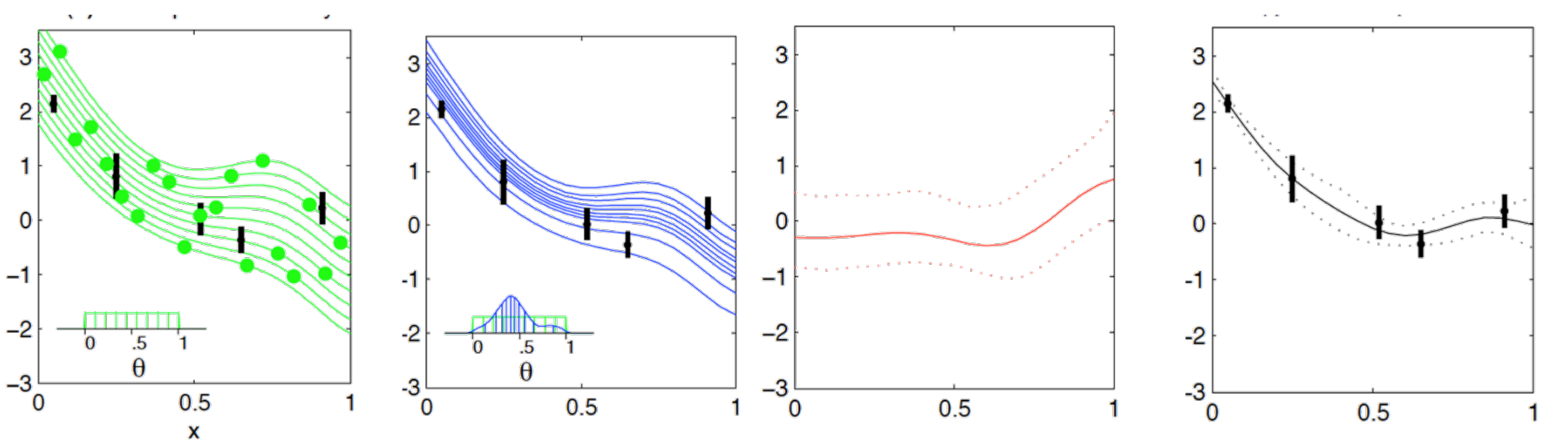
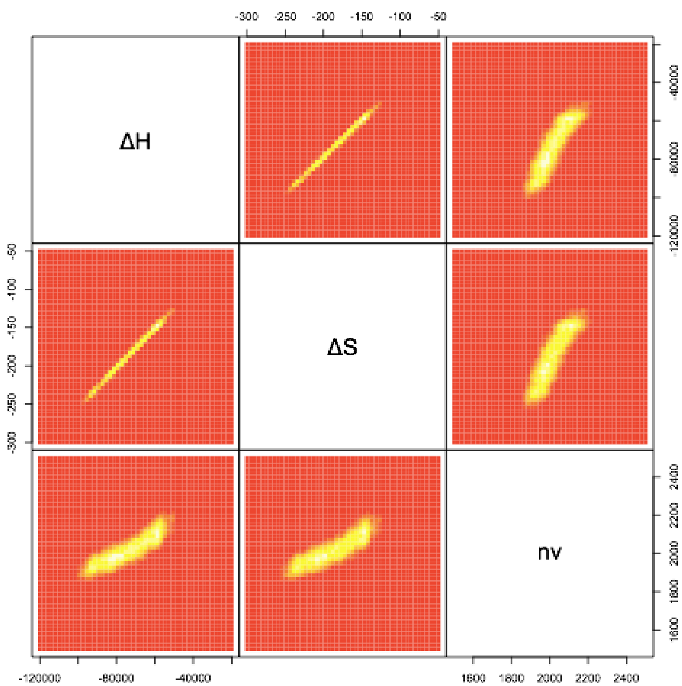


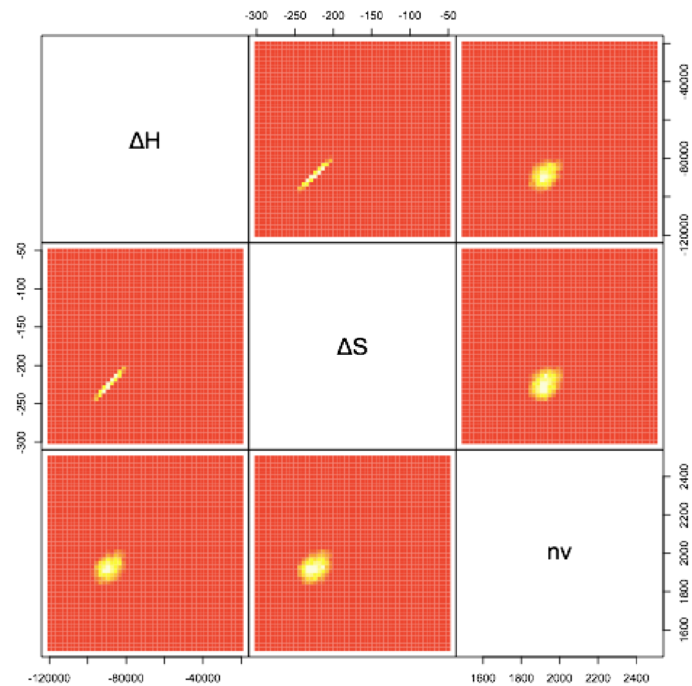
Bayesian methods in parameter estimation

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

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



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



Bayesian methods in parameter estimation

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

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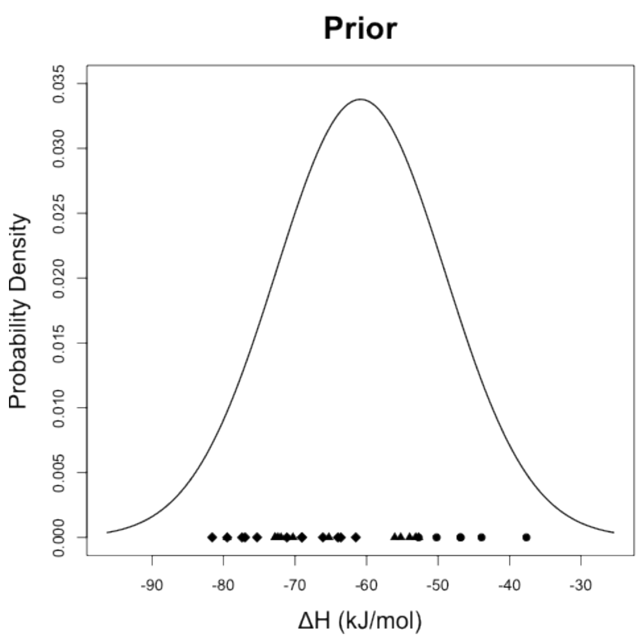
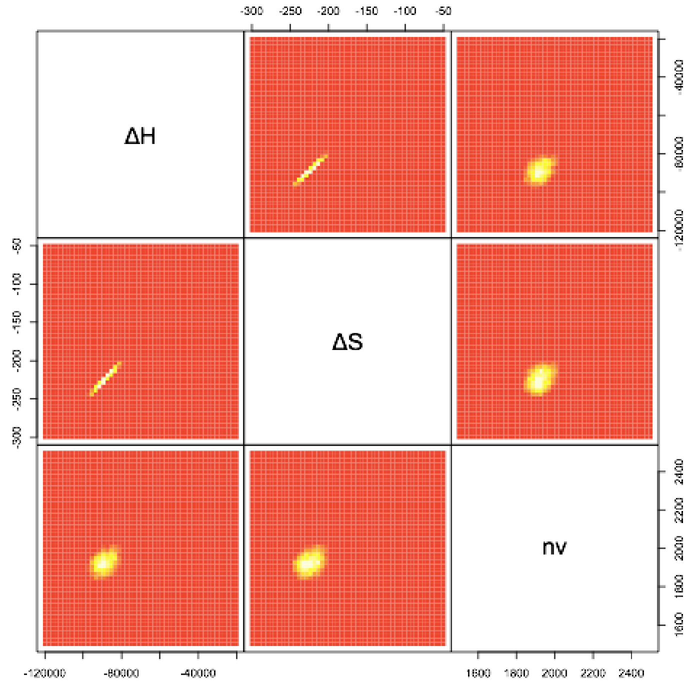
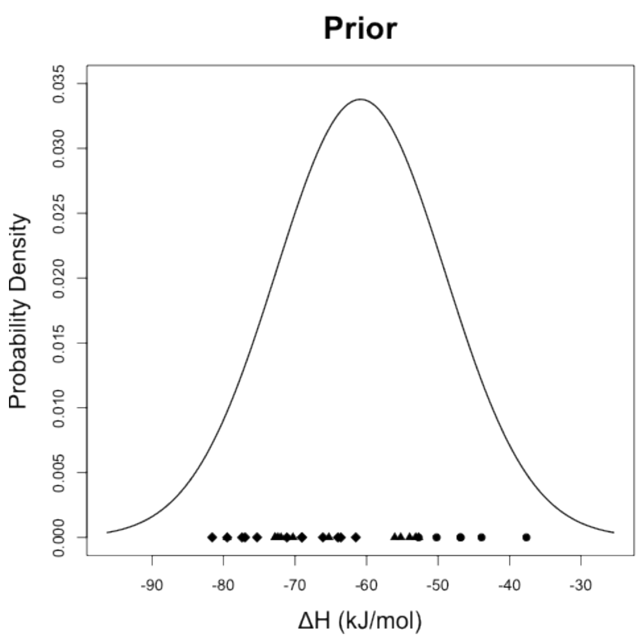


(left) prior distribution for adsorption enthalpy, and (right) posterior distribution



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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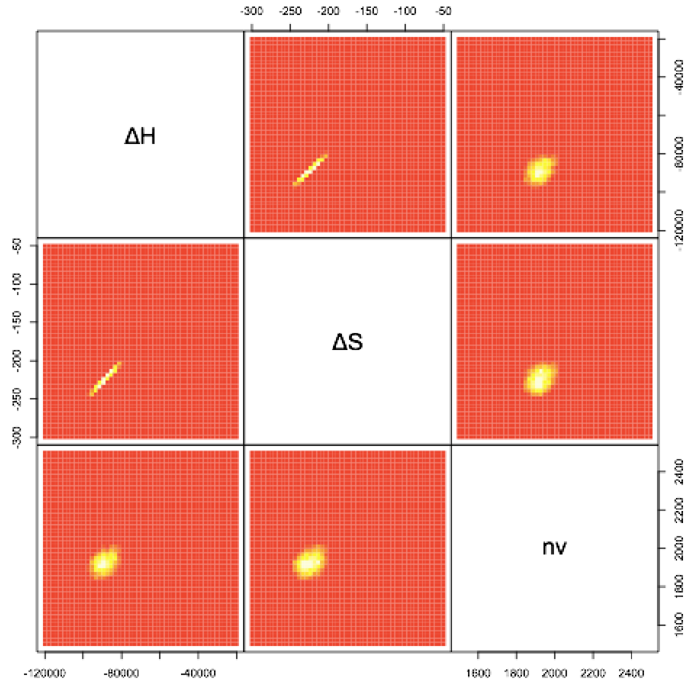
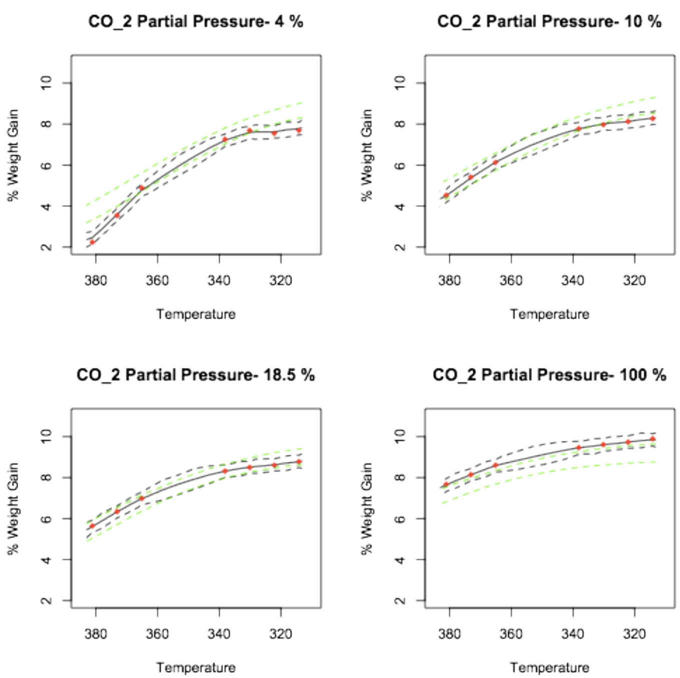


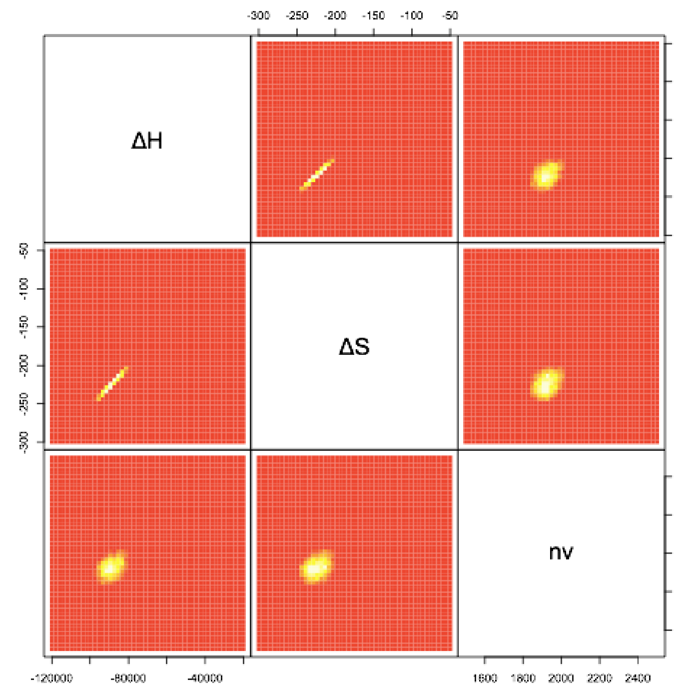
Bayesian methods in parameter estimation

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

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



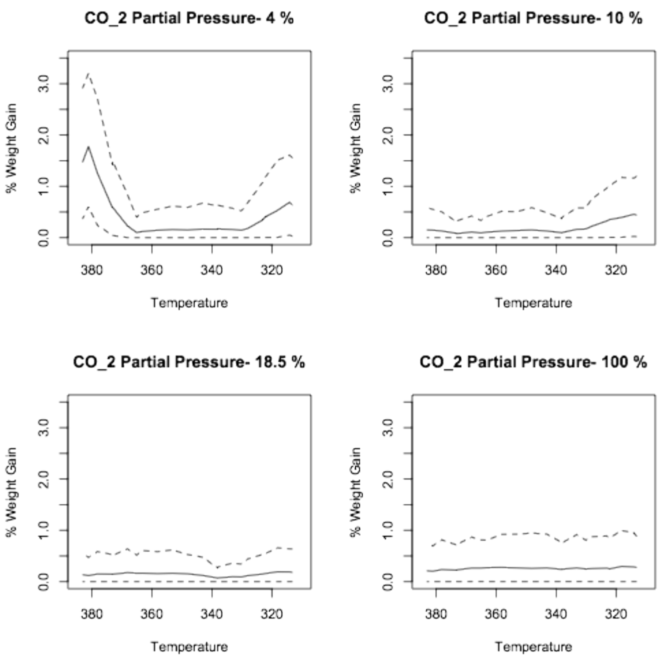
(left) conditioned model + discrepancy predictions with 95% bounds, and (right) posterior distribution



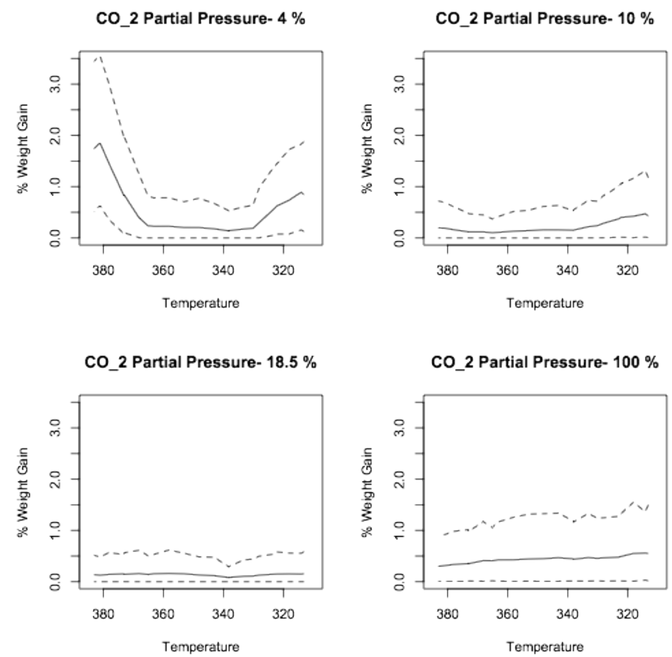
Bayesian methods in parameter estimation

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

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



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



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conclusions

- The stability of diffusive intermediates exercise primary control over the working capacity of mesoporous silica-supported, PEI-based CO_2 sorbents.
- 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.

acknowledgements

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- Greg Ball, NETL
- Andrew Lee, ORISE/NETL