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Carbon Capture Simulation Initiative

Uncertainty Quantification of VLE Models for an MEA System

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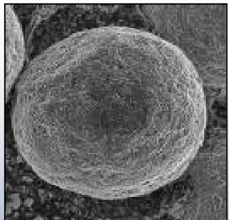
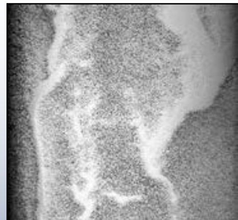
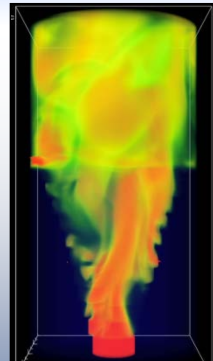





Identify promising concepts → **Reduce the time for design & troubleshooting** → **Quantify the technical risk, to enable reaching larger scales, earlier** → **Stabilize the cost during commercial deployment**

National Labs

- Berkeley Lab
- Pacific Northwest National Laboratory
- NETL
- Los Alamos National Laboratory EST. 1943
- Lawrence Livermore National Laboratory

Academia

- Carnegie Mellon
- Princeton University
- West Virginia University
- Boston University
- The University of Texas at Austin
- Institute for Clean and Secure Energy, The University of Utah

Industry

- ADA Environmental Solutions
- ALSTOM
- B&W the babcock & wilcox company
- GE
- EPRI Electric Power Research Institute
- Southern California Edison, An Edison International Company, San Onofre Nuclear Generating Station
- FLUOR
- Southern Company
- Phillips 66
- AEP American Electric Power
- Chevron
- URS
- ExxonMobil
- Air Products
- Eastman
- ANSYS
- aspentech
- invensys
- PSE

Outline

- Research Objectives and Motivation
- Overall Methodology
- Phoenix Model and e-NRTL Background
- Preliminary Results
  - VLE
  - Heat capacity
  - Heat of absorption
- Future Work

Research Motivation

- Develop robust algorithm for uncertainty quantification of CO_2 based carbon capture system
- Starting point: “Gold Standard” MEA model
  - 30% aqueous MEA solution is industry standard
- Deterministic models of system have been considered
  - “Phoenix Model” (Rochelle Group at UT-Austin) used as baseline in this work
- Methodology has been implied to standalone physical property models (e.g. viscosity, density, surface tension)
  - Thermodynamic framework considered to be most essential physical property

Deterministic and Stochastic Modeling

Deterministic Modeling

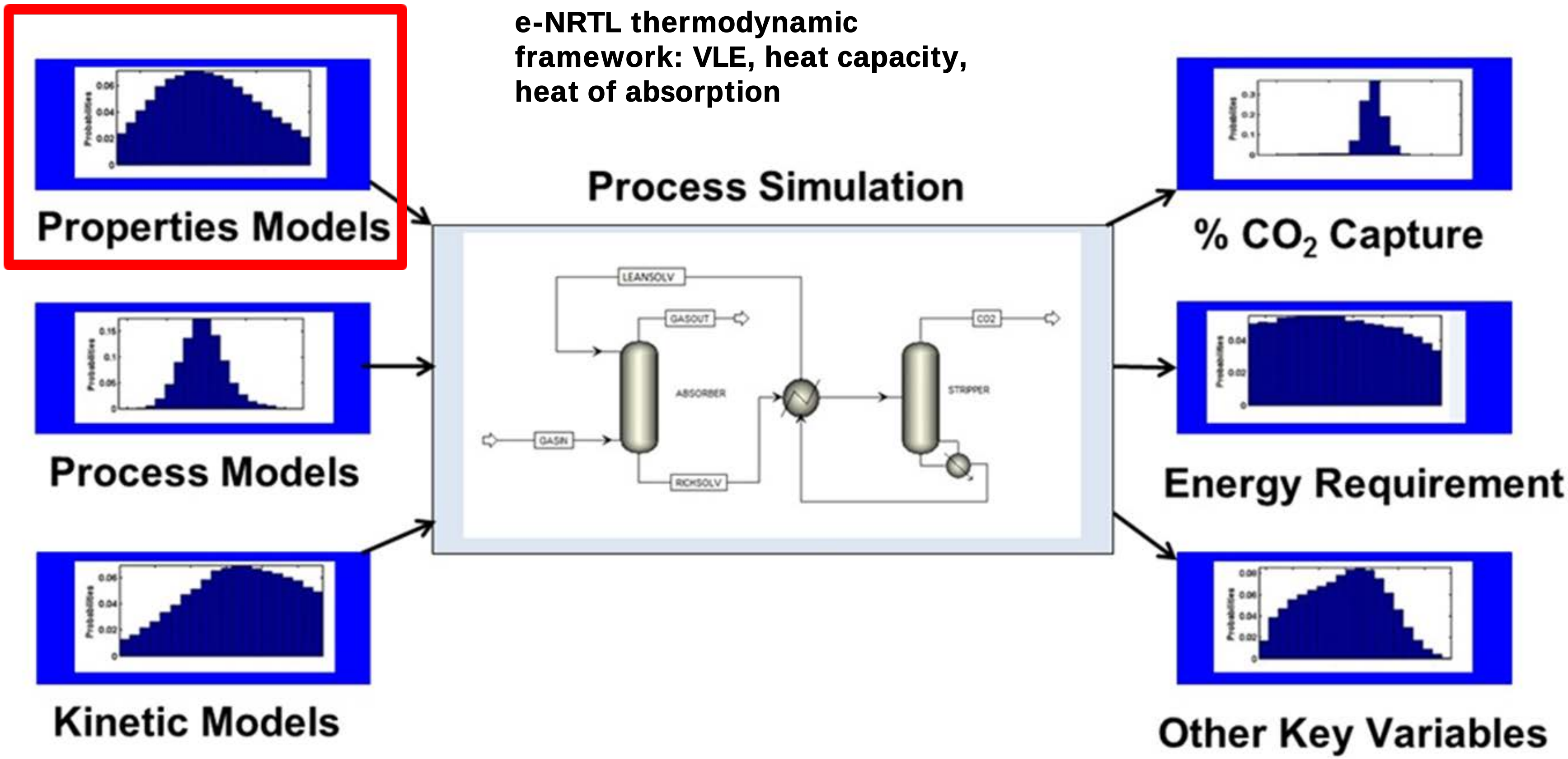
- Single value of
  - Predictor variables
  - Model parameters
  - Output variables
- Parameters calibrated from experiments
  - Best fit methods

Stochastic Modeling

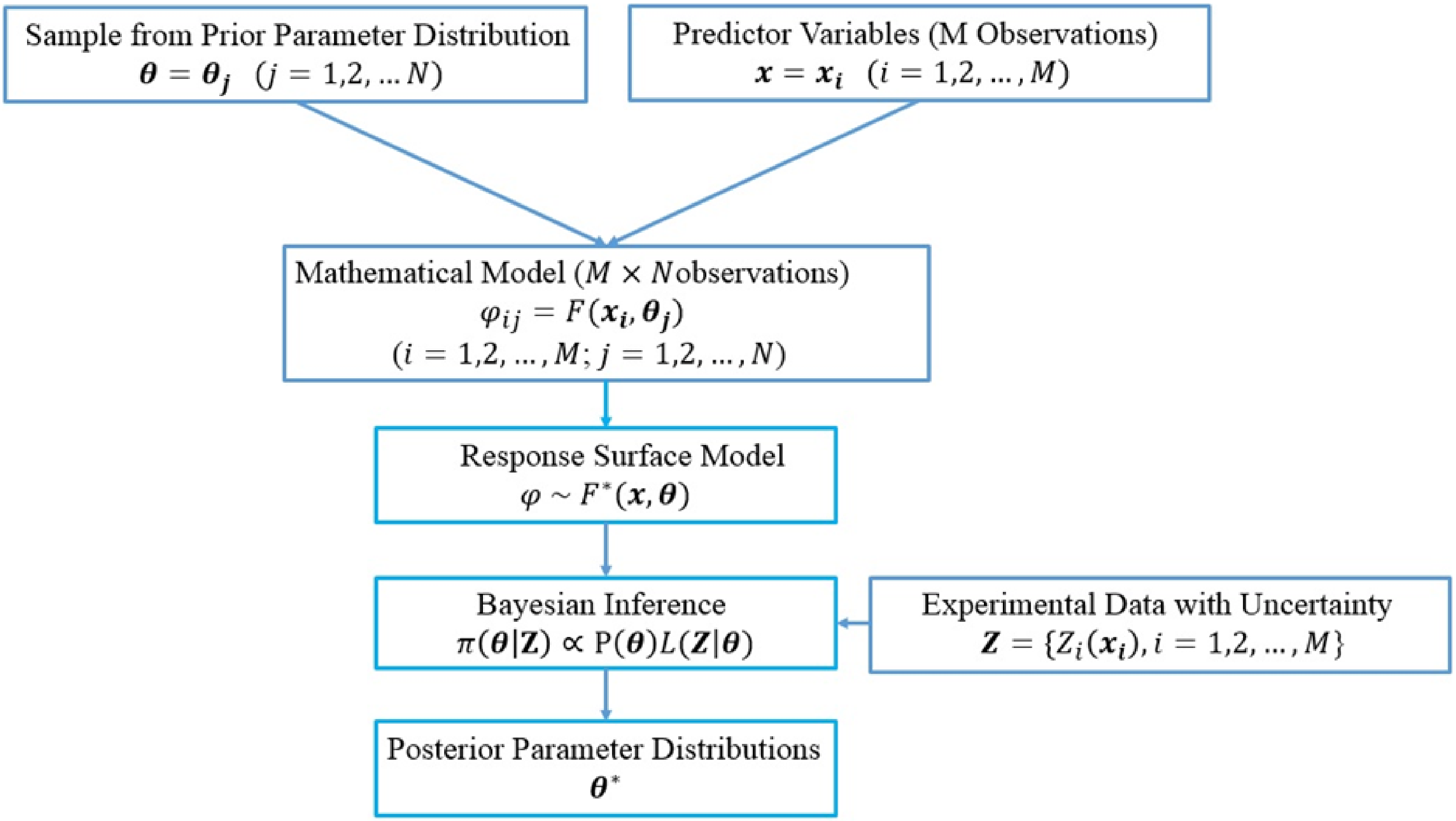
- Model inputs and outputs are probability distributions
- Rationale
  - Variability of measurements (input uncertainty)
  - Physical properties
    - Experimental data uncertainty
    - Model uncertainty

Overall Approach

Focus of this work:
e-NRTL thermodynamic framework: VLE, heat capacity, heat of absorption



Stochastic Modeling Methodology



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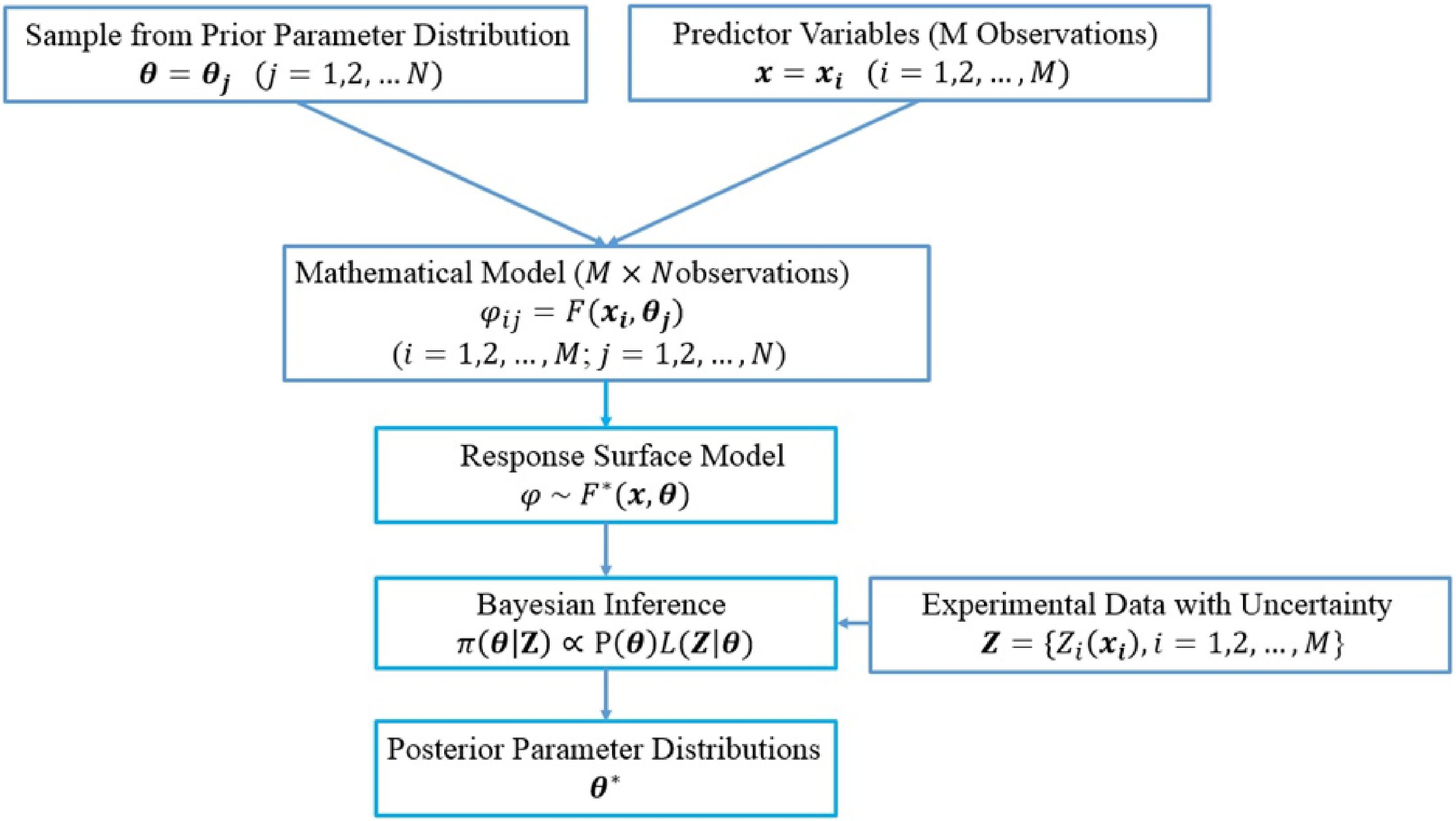
Response Surface Analysis

- Computationally inexpensive surrogate models
- Method
  - Multivariate Adaptive Regression Splines (MARS)
- Procedure
  - Generate input sample
  - Collect output from model simulation
  - Select a response surface scheme and perform fitting
  - Validate the response surface

Stochastic Modeling Methodology



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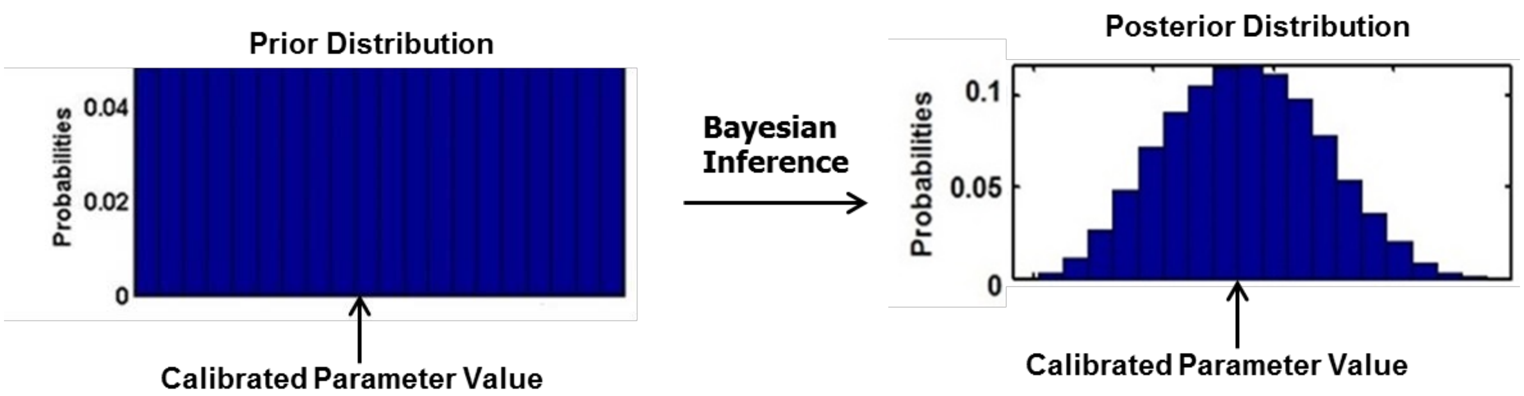
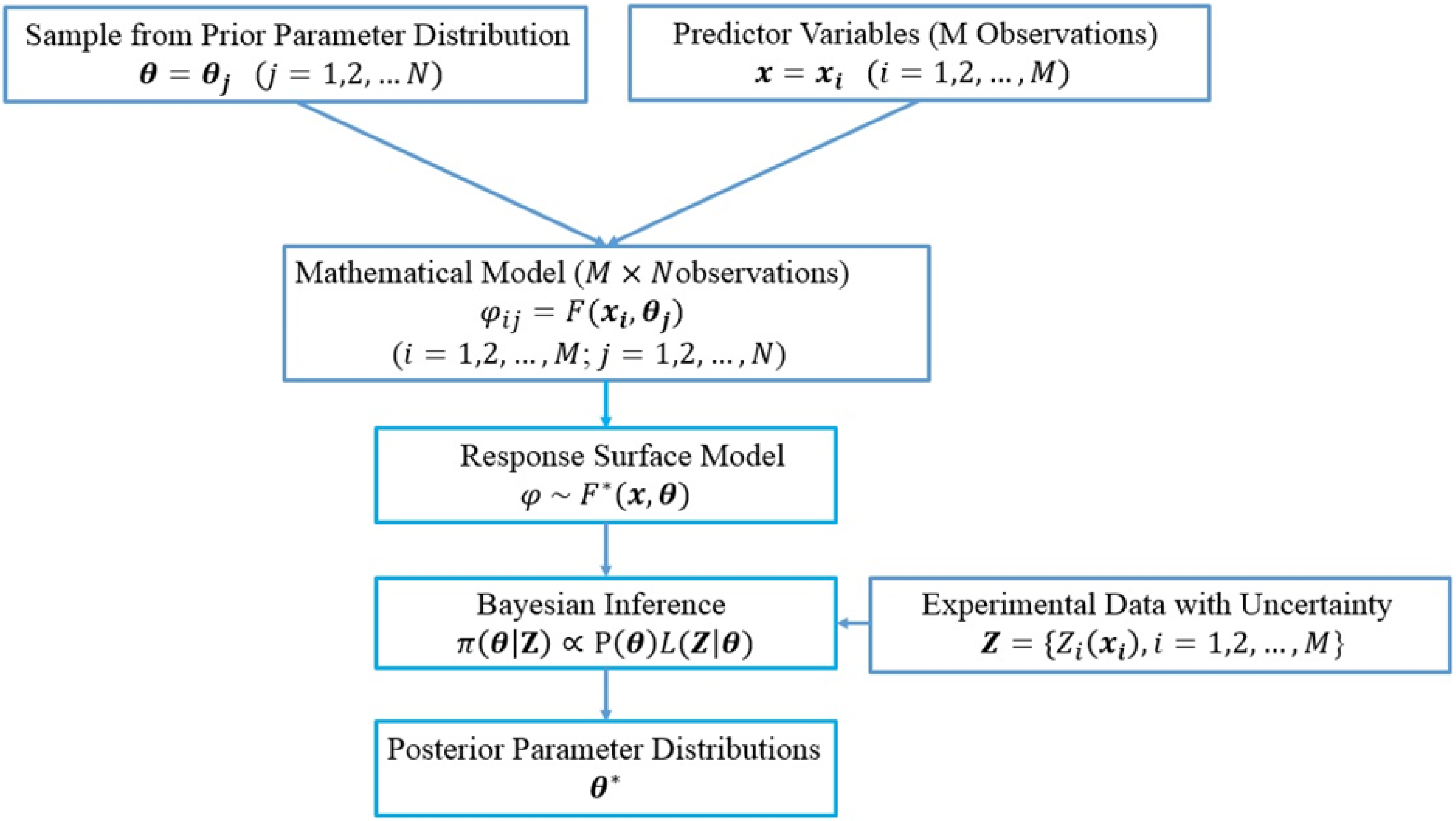
Bayesian Inference

- Bayesian inference seeks to update prior beliefs of parameter uncertainties in view of data
  - Idea: scan intelligently the prior parameter uncertainty space to identify values that match well with available data
  - Algorithm: Markov Chain Monte Carlo (MCMC) method using Gibbs sampling



Stochastic Modeling Methodology



Phoenix Model Overview

- Developed by Rochelle group at University of Texas-Austin[1]
- Liquid phase modeled by e-NRTL model
- Vapor phase modeled by Redlich-Kwong equation of state
- Simplified electrolyte speciation[2]

$$2MEA + CO_2 \leftrightarrow MEA^+ + MEACOO^-$$

$$MEA + CO_2 + H_2O \leftrightarrow MEA^+ + HCO_3^-$$

- Highly non-ideal solution

1. Jorge Mario Plaza, Ph.D. Dissertation, UT Austin, May 2012
2. Marcus Hilliard, Ph.D. Dissertation, UT Austin, May 2008

Phoenix Model Overview

- 41 individual parameters regressed
  - Standard Gibbs Free Energy/ Enthalpy of Formation for Electrolytes
  - Ideal Gas and Electrolyte Component Heat Capacity Parameters
  - Henry's Constant Parameters
  - Molecule-Molecule Binary Parameters
  - Electrolyte-Molecule Pair Parameters
  - Electrolyte-Electrolyte Pair Parameters
- Data types considered
  - Vapor liquid equilibrium
  - Heat capacity
  - Heat of absorption

Thermodynamic Relationships

Equilibrium condition for system $\hat{\varphi}_i y_i P = \gamma_i^* x_i H_i$

$$ln(\gamma_i) = \frac{1}{RT}\frac{\partial(nG^{ex})}{\partial n_i}\bigg|_{T,P,n_{j\neq i}} \qquad G^{ex} = G^{ex}_{PDH} + G^{ex}_{Born} + G^{ex}_{LC}$$

Interaction parameters embedded in local contribution of excess Gibbs energy

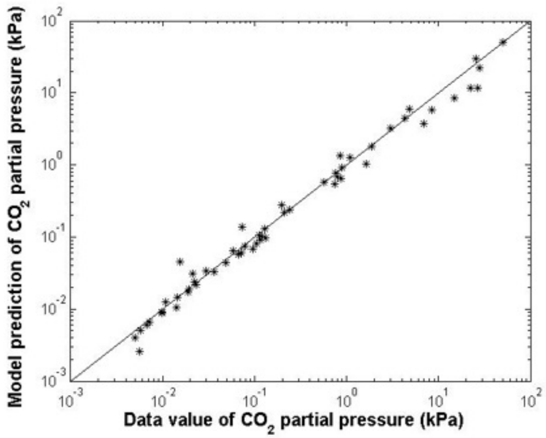
$$\Delta H_{abs} = R\frac{\partial \ln(\hat{f}_{CO_2})}{\partial\,(1/T)}\bigg|_{P,x_i}$$

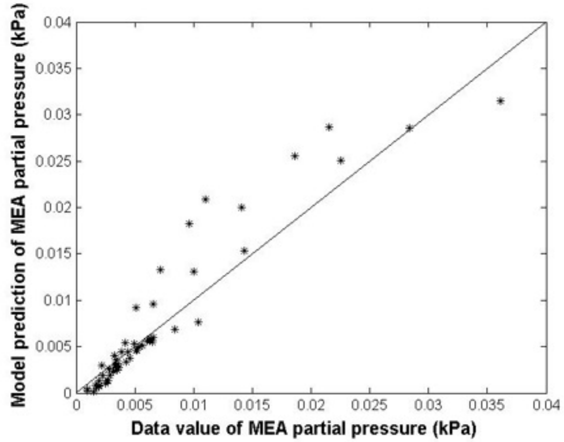
$$\frac{-H^{ex}}{T^2} = \frac{\partial(G^{ex}/T)}{\partial T}\bigg|_{P,x_i}$$

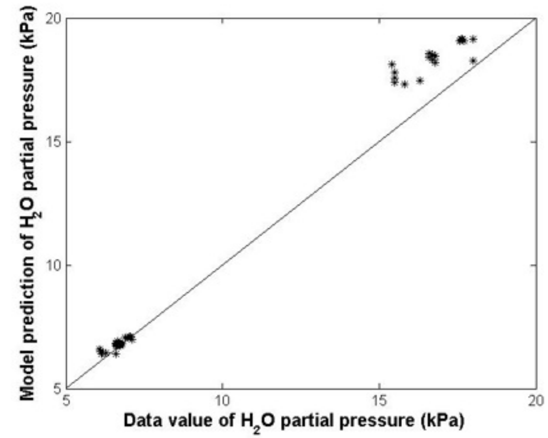
Heat capacity parameters used in enthalpy calculation

Need to regress data types simultaneously to maintain thermodynamic consistency

VLE Example: Phoenix Model Comparison







Data Predictor Variable Ranges

3.5 and 7 m MEA solutions

Loading = 0.1-0.6 mol CO2/ mol MEA

Temperature = 40°C and 60°C

Data from: Marcus Hilliard, Ph.D. Dissertation, UT Austin, May 2008

	Average	Standard Deviation
P_{CO_2}	22.46 %	29.43 %
P_{MEA}	31.05 %	25.69 %
P_{H_2O}	4.80 %	4.37 %

VLE Example, Continued

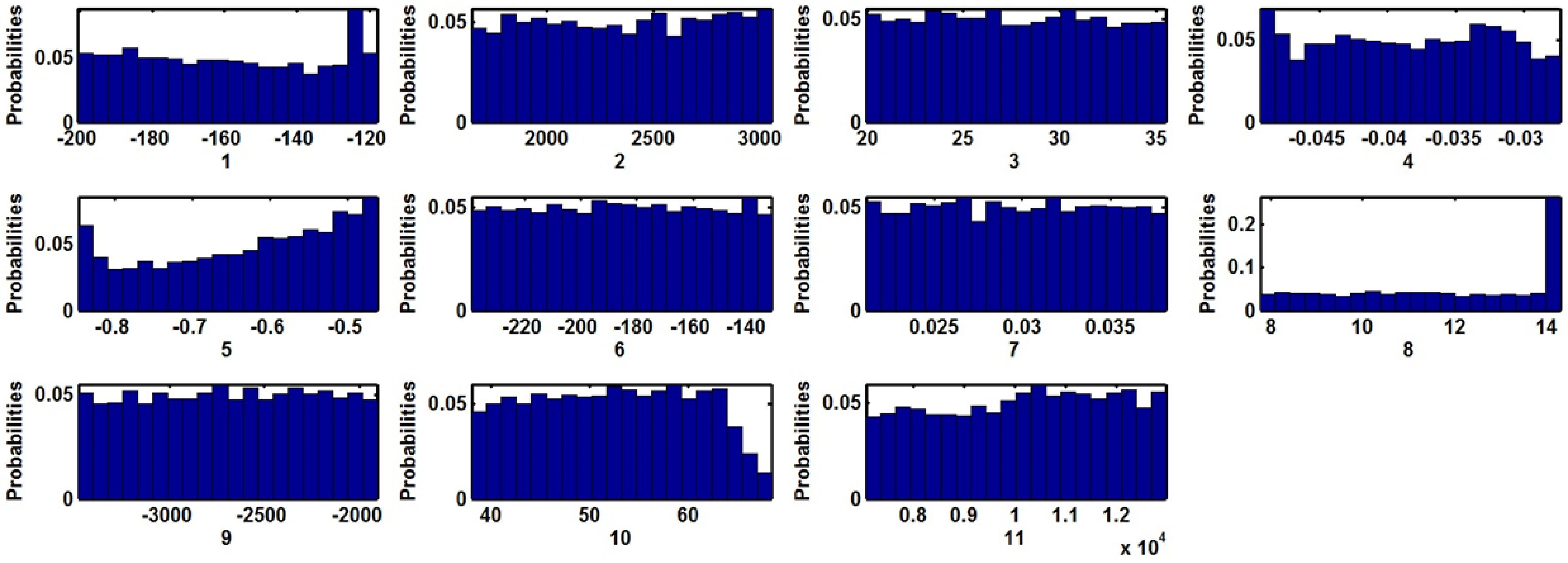
- Performed Bayesian inference with prior distributions of ±30% Phoenix model values for 11 molecule-molecule binary interaction parameters

$$\tau_{mm'} = A_{mm'} + \frac{B_{mm'}}{T} + E_{mm'} \ln(T) + F_{mm'} T$$

Equation embedded in local contribution of excess Gibbs free energy equation

Parameter Number	Parameter Identity
1	A_{H_2O-MEA}
2	B_{H_2O-MEA}
3	E_{H_2O-MEA}
4	F_{H_2O-MEA}
5	A_{MEA-H_2O}
6	B_{MEA-H_2O}
7	E_{MEA-H_2O}
8	A_{CO_2-MEA}
9	B_{CO_2-MEA}
10	A_{MEA-CO_2}
11	B_{MEA-CO_2}

VLE Example UQ Results



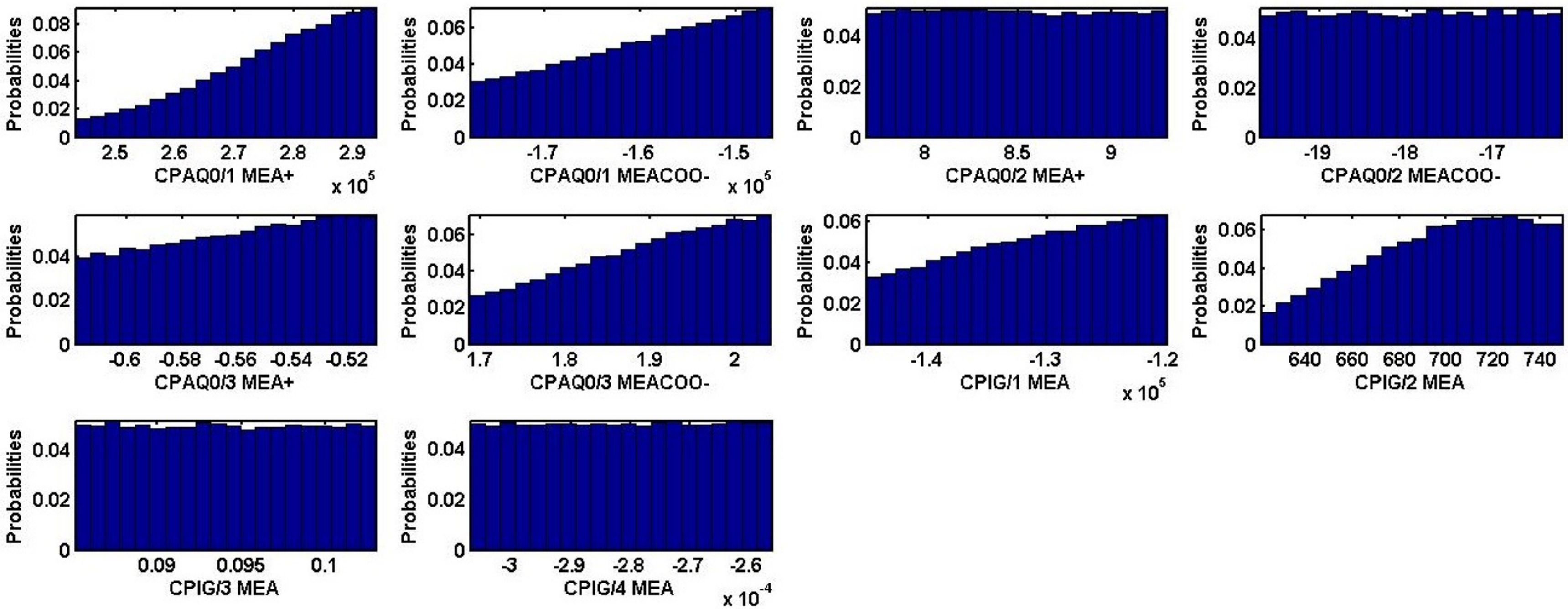
Example: Heat Capacity

- Performed Bayesian inference with prior distributions of ±10% of parameters
  - 3 parameters for CPAQ0 model of MEA+
  - 3 parameters for CPAQ0 model of MEACOO-
  - 4 parameters for CPIG model of MEA
- Used data from Hilliard and Weiland
- Heat capacity polynomial forms:

$$C_{p,i}^{ig} = C_{1i} + C_{2i}T + C_{3i}T^2 + C_{4i}T^3 + C_{5i}T^4 + C_{6i}T^5$$

$$C_{p,k}^{\infty} = C_{1i} + C_{2i}T + C_{3i}T^2 + \frac{C_{4i}}{T} + \frac{C_{5i}}{T^2} + \frac{C_{6i}}{\sqrt{T}}$$

Heat Capacity UQ Results



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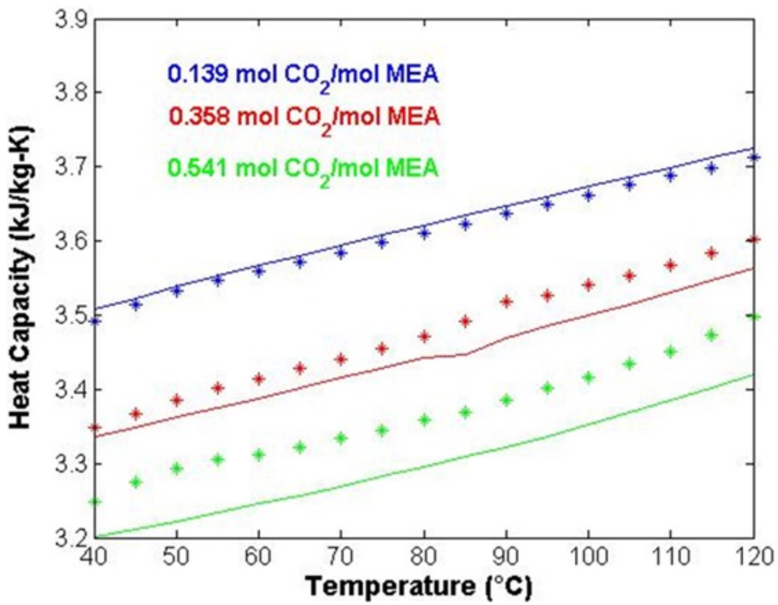
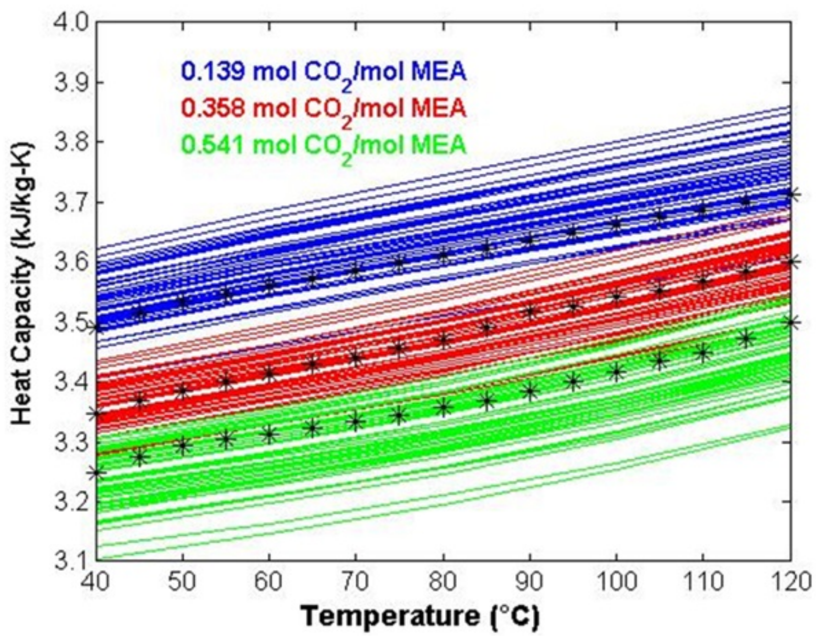
Heat Capacity: Deterministic and Stochastic Models

7 m aqueous MEA solutions

Phoenix Model



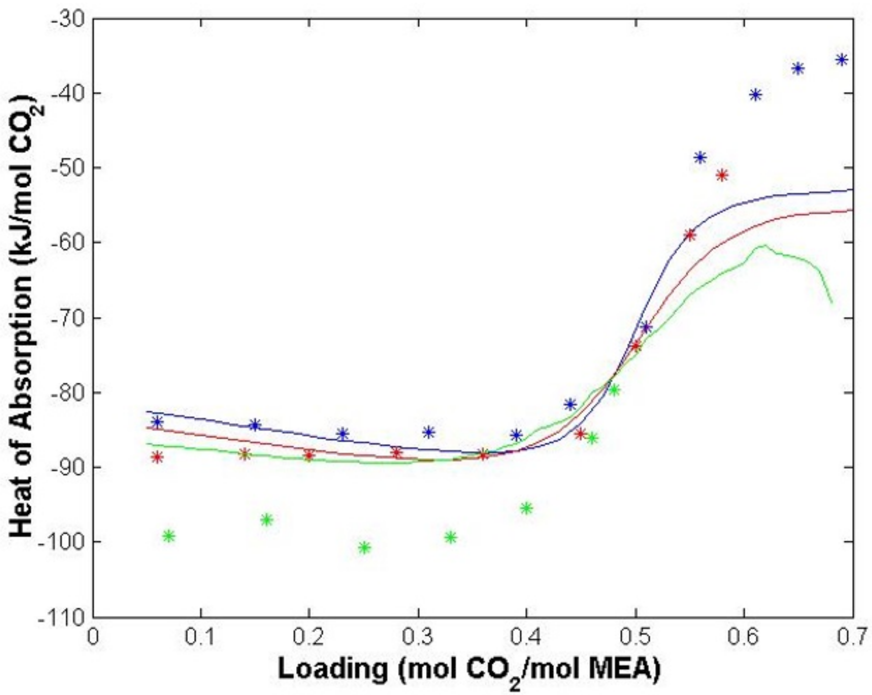
Posterior Distribution of Sample Size 50



Experimental data from: Marcus Hilliard, Ph.D. Dissertation, UT Austin, May 2008

Heat of Absorption

Phoenix Model Comparison (parameters not regressed to match data)



$$\Delta H_{abs} = R \left. \frac{\partial \ln(\hat{f}_{CO_2})}{\partial\,(1/T)} \right|_{P,x_i}$$

Data from Kim et al., GHGT-12

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Future Work

- Complete physical property models uncertainty quantification
  - e-NRTL thermodynamic framework: VLE, heat capacity, heat of absorption
  - Diffusivity
- Propagate all stochastic models (e.g. physical properties, kinetics, mass transfer and hydraulics) through process simulation
- Validation of overall stochastic model with process data
  - Steady state data from UT Austin pilot plant
  - Steady state and dynamic data from NCCC

Thank you!

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As part of the National Energy Technology Laboratory's Regional University Alliance (NETL-RUA), a collaborative initiative of the NETL, this technical effort was performed under the RES contract DE-FE0004000.

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