SAND2017-7665D

Exceptional service in the national interest

Sandia National Laboratories

Bayesian Regression of Thermodynamic Models of Redox Active Materials

Katherine Johnston
California Institute of Technology

Purpose:

Finding a suitable functional redox material is a critical challenge to achieving scalable, economically viable technologies for storing concentrated solar energy in the form of a defected oxide. Demonstrating effectiveness for thermal storage or solar fuel is largely accomplished by using a thermodynamic model derived from experimental data. The purpose of this project is to test the accuracy of our regression model on representative data sets.

Approach:

- Three data sets: Ceria, Panlener, and Goldyreva
  - Ceria and Panlener: from a material used for solar fuels by splitting water and carbon dioxide
  - Goldyreva: from a material for thermal storage
- The model has 8 parameters
- Use Bayesian inference and Markov Chain Monte Carlo to estimate the parameters to fit the data
- Evaluate the model's fit and other factors
- Increase fit by adjusting inputs and initial values

What is Bayesian Inference and MCMC?

- Bayesian Inference
  - Method for determining model parameters by calibrating against a set of data points
- Bayes formula: $p(\theta|D,M) \propto p(D|\theta,M)p(\theta|M)$
  - $p(\theta|D,M)$ is the posterior distribution; $p(D|\theta,M)$ is the likelihood; $p(\theta|M)$ is the prior
- Maximize the posterior to find the ideal parameters

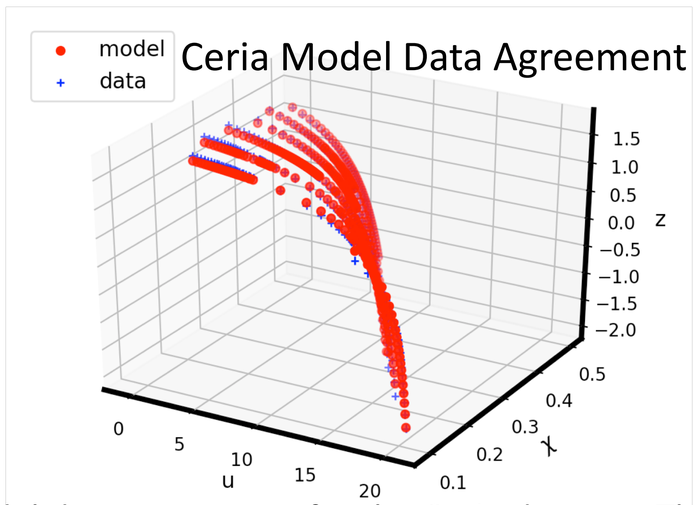
- Markov Chain Monte Carlo is a method to sample the posterior distribution

1. Start at a given point and find the posterior probability
2. Sample a second point from a proposal distribution and find the posterior probability.
3. Calculate α as the posterior of the new divided by the old posterior: $\alpha = \frac{p(new|D)}{p(old|D)}$.
   - If $\alpha > 1$, the new point is more likely and accept the new point.
   - If $\alpha < 1$, accept the new point at a probability of α
4. Continue for the desired number of samples

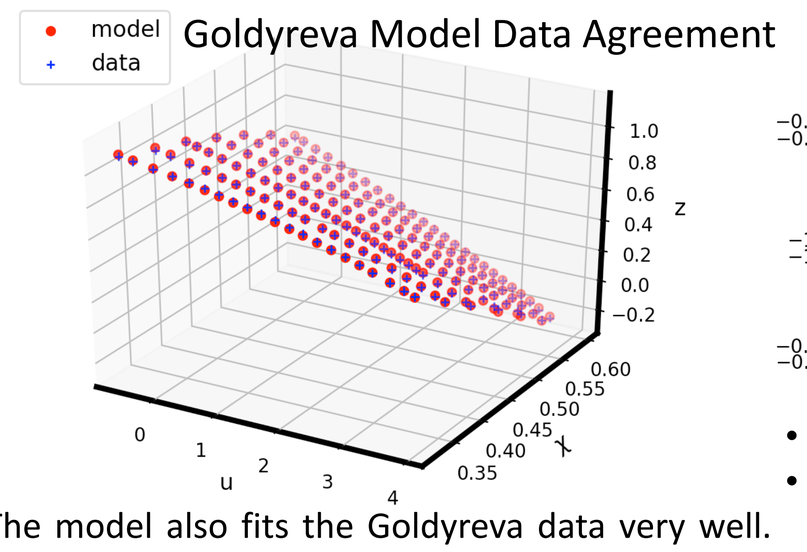
- Explores all areas of a given space, focusing on more likely areas with larger posterior probabilities

Results:

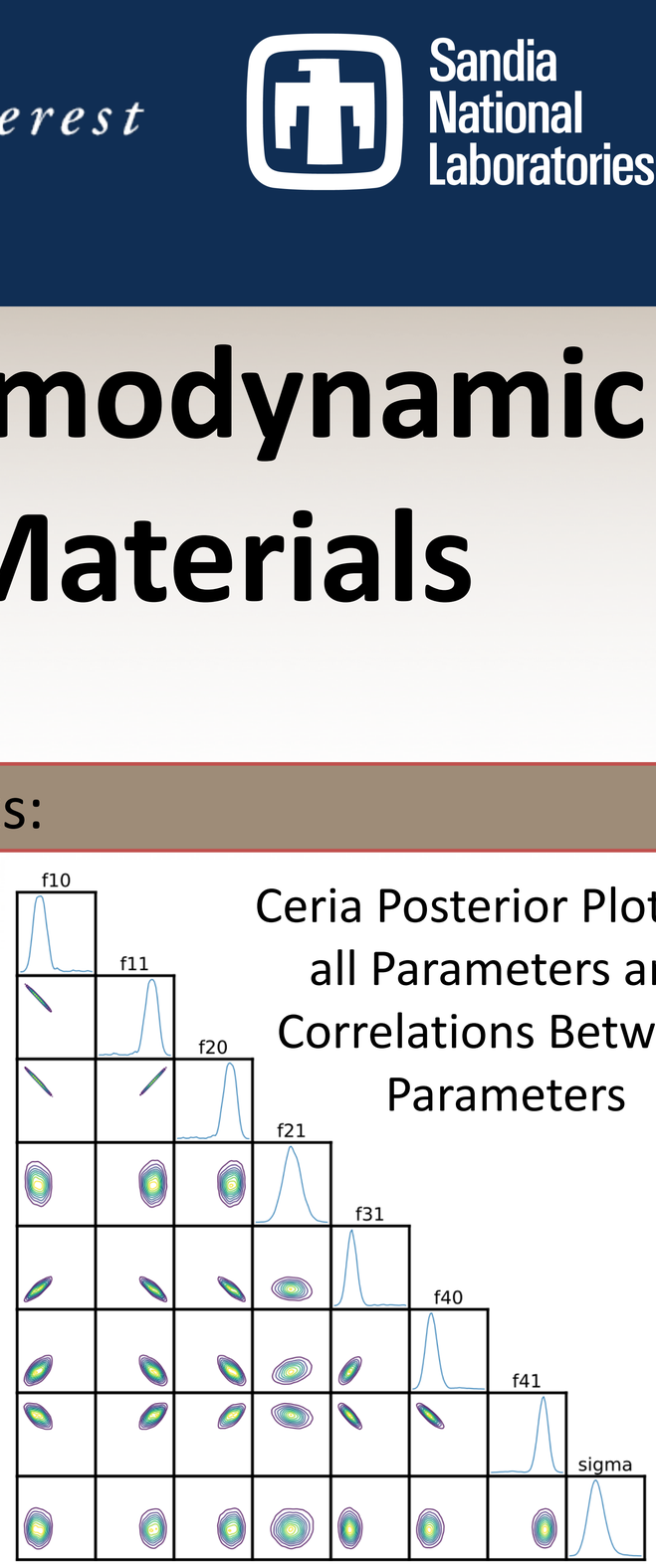
While using MCMC, the model is fitting all three data sets. Different inputs and initial values were used for each data set and had to be manipulated to get the model to fit the data. Because different data sets were tested, this model can be applied to different materials and uses.



Model data agreement for the Ceria data set. The model (red) fits the data (blue) pretty well. Both Ceria and Panlener data sets are of CeO_2 and used for solar fuels by splitting water and carbon dioxide.

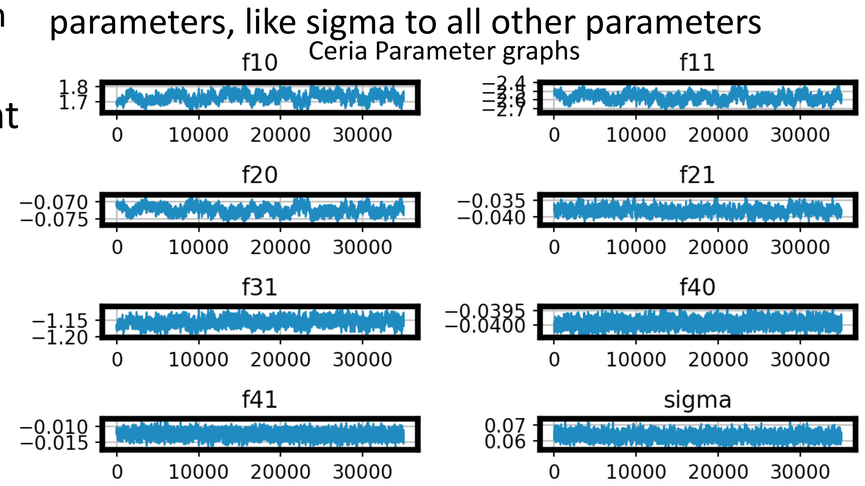


The model also fits the Goldyreva data very well. This data set is for a material involved in thermal storage.



Ceria Posterior Plots for all Parameters and Correlations Between Parameters

- Displays the marginalized posterior plots for each parameter and the correlations between parameters
- Straighter lines between the parameters indicate correlation, like f10 and f11
- The more circular the more independent the parameters, like sigma to all other parameters



- Example of the MCMC chains for the Ceria data
- Taken after a burn in and with a stride because MCMC takes many steps to find the interesting part of the space and takes a few steps to move causing repeated points Notice how the parameters jump around a lot. This is because MCMC is exploring everywhere around the peak

Further Work:

- Fit the data with different models and use Bayes factors to compare the models.
  - Use more/less parameters and compare the outcomes
    - Determine if all parameters are necessary
- Make this problem into a tutorial for the Uncertainty Quantification Toolkit (UQTk).

Acknowledgments:

Thank you Ellen Stechel and Anthony McDaniel for the model and work on the problem. Thank you Bert Debusschere for mentoring. Funding was provided by the DOE Workforce Development for Teachers and Scientists (WDTS) program as well as the DOE Office of Energy Efficiency and Renewable Energy (EERE)

Goldyreva, et al. *J. of Alloys and Compounds*, *638*, 44–49, 2015.
McDaniel, *Current Opinion in Green and Sustainable Chemistry*, 4, 37-43. 2017
Panlener, R. J., et al. *J. of Physics and Chemistry of Solids*, *36*(11), 1213–1222, 1975.
Zinkevich, et al. *Solid State Ionics* 177, 989-1001, 2006.