Bayesian Calibration of Models of SOFC Electrode Materials
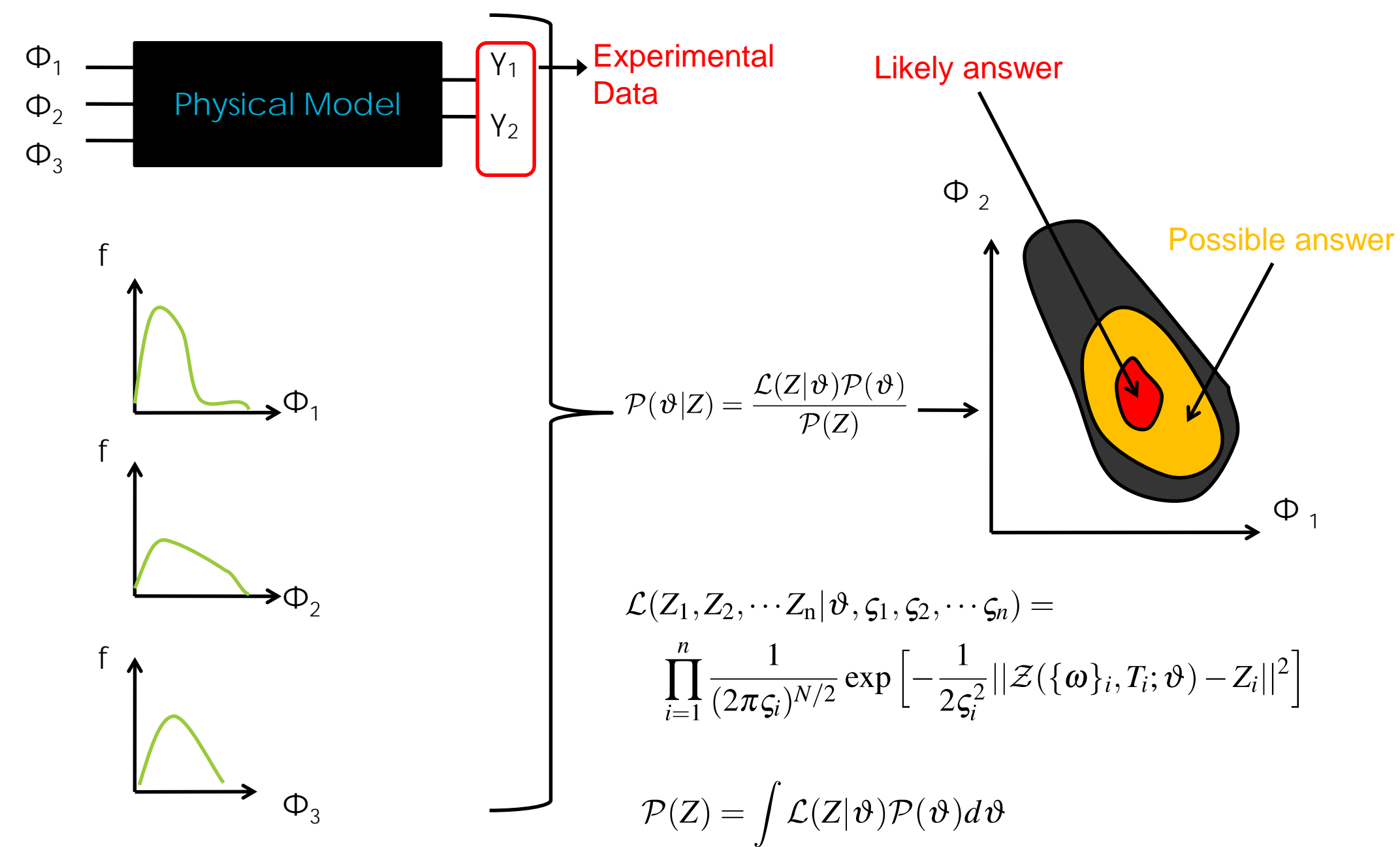
Giuseppe F. Brunello[1], Grigorios Panagakos[1], Jian Liu[1], Tobias M. Hunter[2], Harry O. Finklea[3] ,and David S. Mebane[4].

[1]US Department of Energy, National Energy Technology Laboratory, Pittsburgh, PA / Morgantown, WV; [2] Vienna University of Technology Institute of Chemical Technologies and Analytics Research Division Electrochemistry Getreidemarkt; [3] Department of Chemistry, West Virginia University [4] Department of Mechanical and Aerospace Engineering, West Virginia University
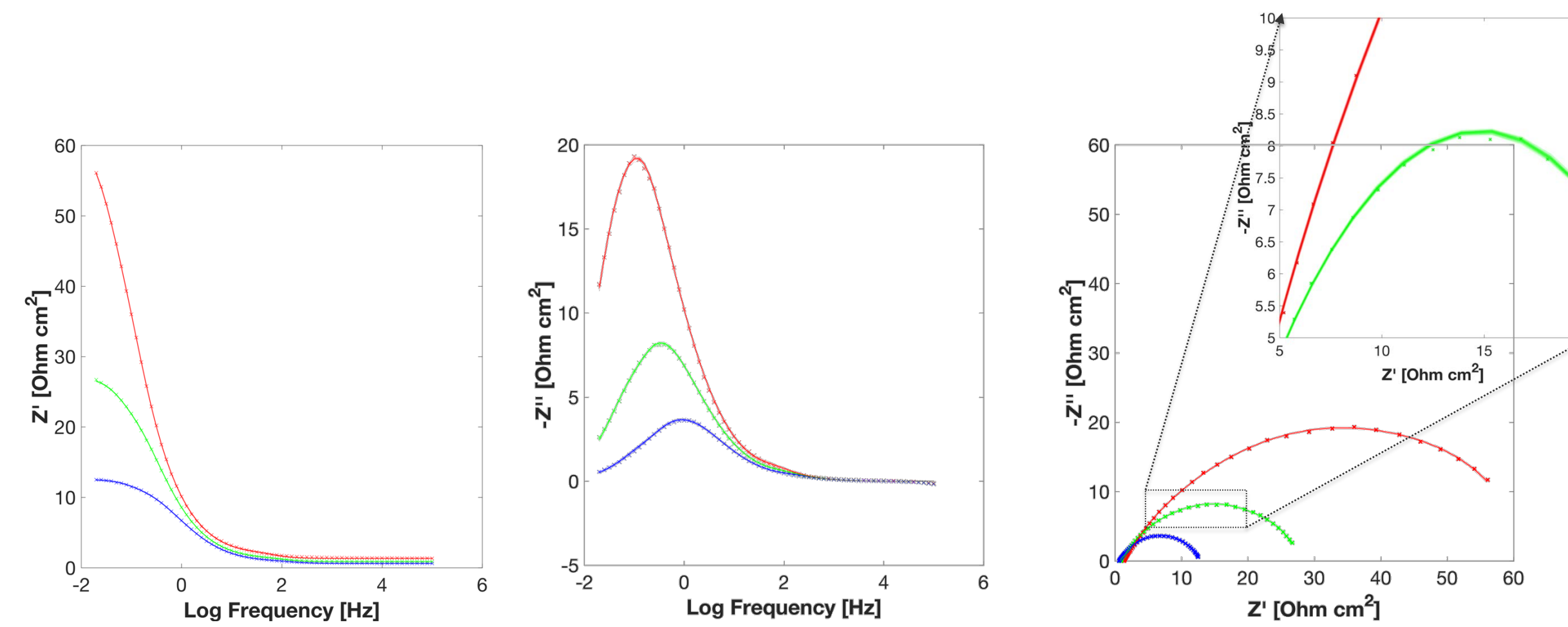
Research & Innovation Center

NATIONAL ENERGY TECHNOLOGY LABORATORY

Introduction

$$\mathcal{P}(\vartheta|Z) = \frac{\mathcal{L}(Z|\vartheta)\mathcal{P}(\vartheta)}{\mathcal{P}(Z)}$$

$$\mathcal{L}(Z_1, Z_2, \cdots Z_n|\vartheta, \varsigma_1, \varsigma_2, \cdots \varsigma_n) =$$
$$\prod_{i=1}^{n} \frac{1}{(2\pi\varsigma_i)^{N/2}} \exp\left[-\frac{1}{2\varsigma_i^2}||\mathcal{Z}(\{\omega\}_i, T_i; \vartheta) - Z_i||^2\right]$$

$$\mathcal{P}(Z) = \int \mathcal{L}(Z|\vartheta)\mathcal{P}(\vartheta)d\vartheta$$

Bayes' Theorem is used to update the prior probability of a model's parameters from experimental evidence to obtain the posterior probability. This is typically done using Markov Chain Monte Carlo since integrating the P(Z) term above is, usually, intractable.

To use Bayesian Calibration a Likelihood function is required. This is a statistical model of the process.

Dissociative adsorption:
$$\frac{1}{2}O_2(g) + Mn_B^x + V_{ad}^x \longleftrightarrow O'_{ad} + Mn_B^\cdot$$

Incorporation:
$$O'_{ad} + V_O^{\cdot\cdot} + Mn_B^x \longleftrightarrow O_O^x + Mn_B^\cdot + V_{ad}^x$$

Vacancy Transfer:
$$O_O^x + V_{O,YSZ}^{\cdot\cdot} \longleftrightarrow O_{O,YSZ}^x + V_O^{\cdot\cdot}$$

Triple Phase Boundary:
$$O'_{ad} + V_{O,YSZ}^{\cdot\cdot} + Mn_{Mn}^x \longleftrightarrow V_{ad}^x + O_{O,YSZ}^x + Mn_{Mn}^\cdot$$

Schottky:
$$nil \longleftrightarrow 3V_O^{\cdot\cdot} + V_{La}''' + V_{Mn}'''$$


Results

As a result of the Bayesian Calibration, coverage of the experimental data was obtained. Note that there are 200 superimposed line in the figures above.
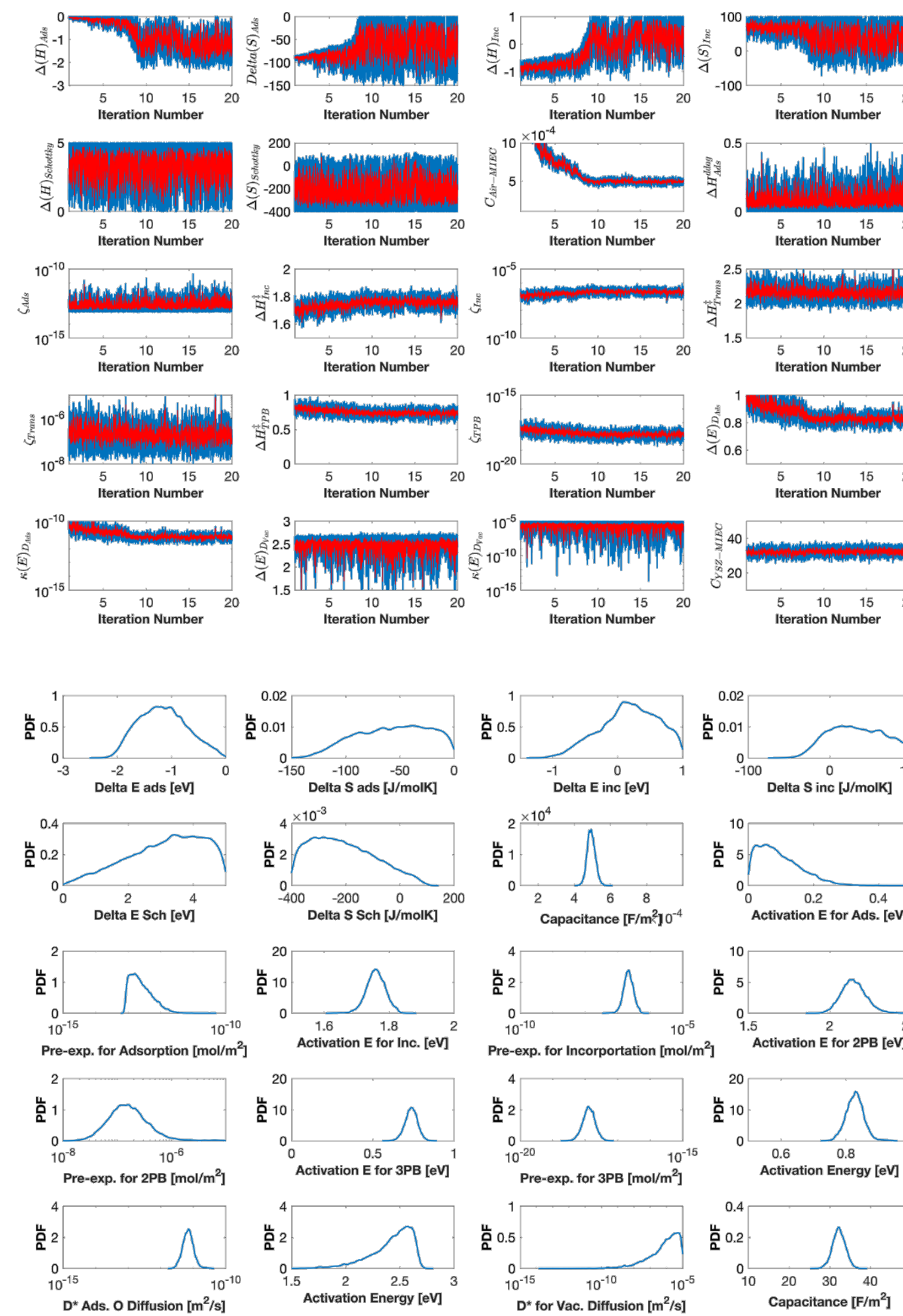

After one million steps the parameters reach their equilibrium values. By sampling the the next one million steps, we reconstruct the posterior distribution. Note, the $C_{LSM-YSZ}$ is higher than expected, indicating that the model did not resolve C_{chem}.
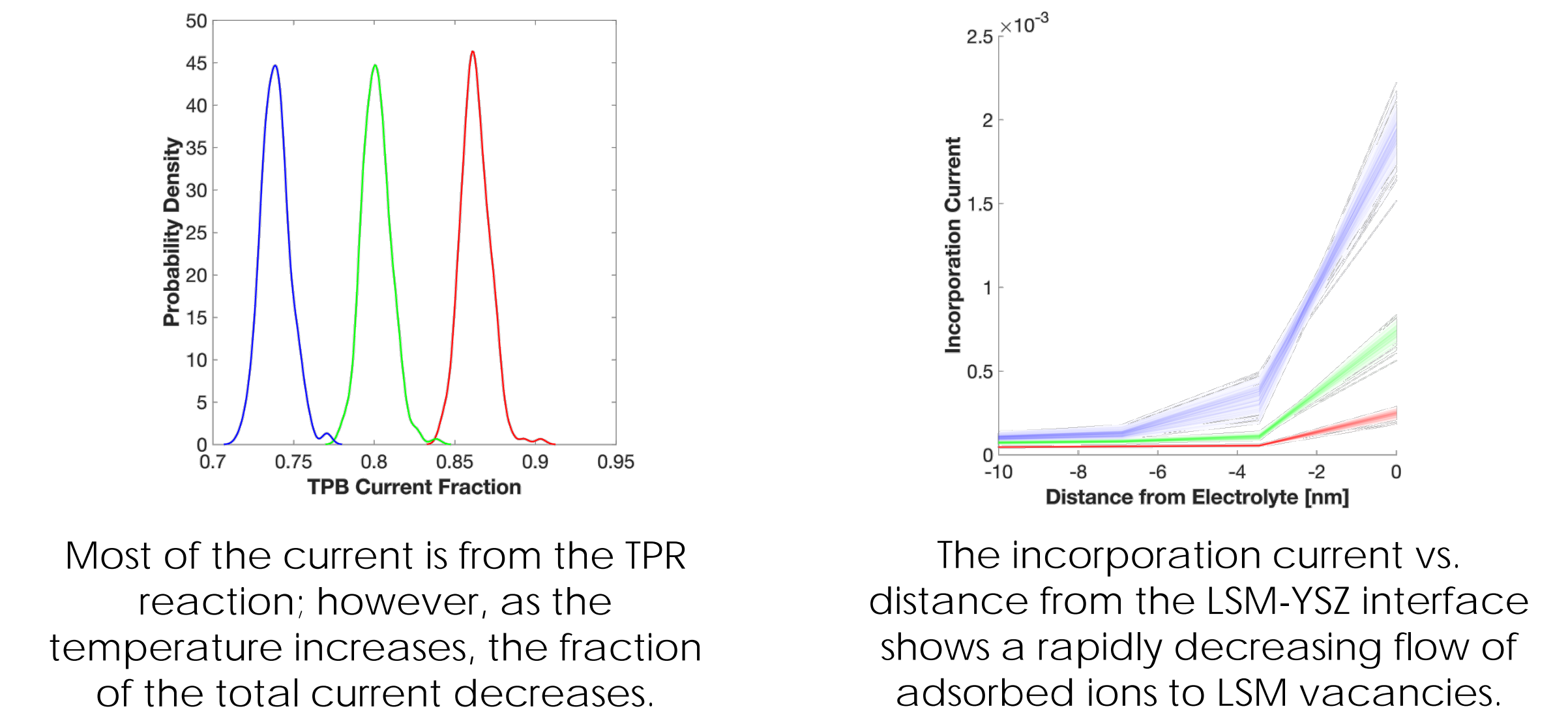
Most of the current is from the TPR reaction; however, as the temperature increases, the fraction of the total current decreases.

The incorporation current vs. distance from the LSM-YSZ interface shows a rapidly decreasing flow of adsorbed ions to LSM vacancies.
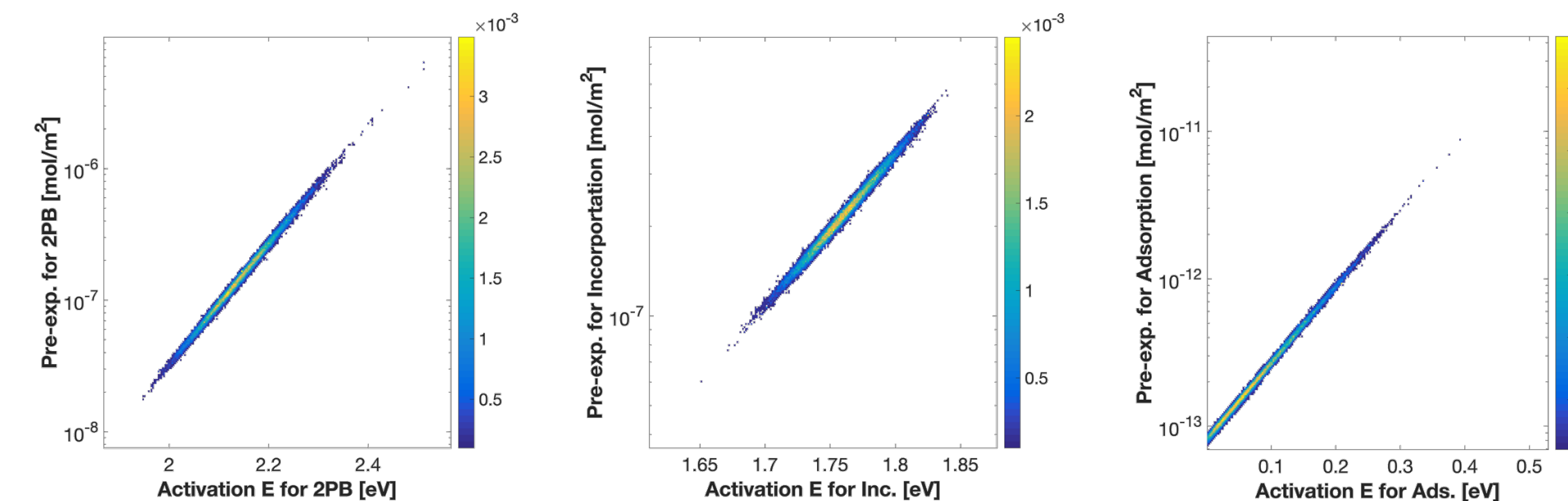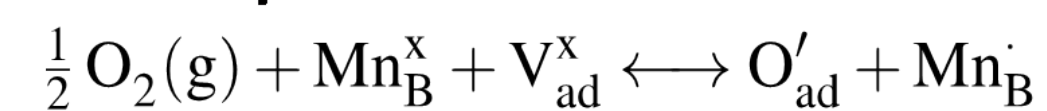

Posterior distribution of the 2PB reaction's activation energy and pre-exponential factor
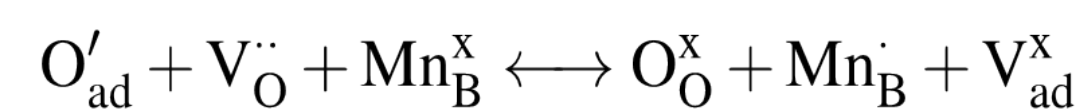
Posterior distribution of the incorporation reaction's activation energy and pre-exponential factor

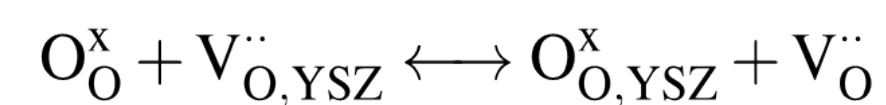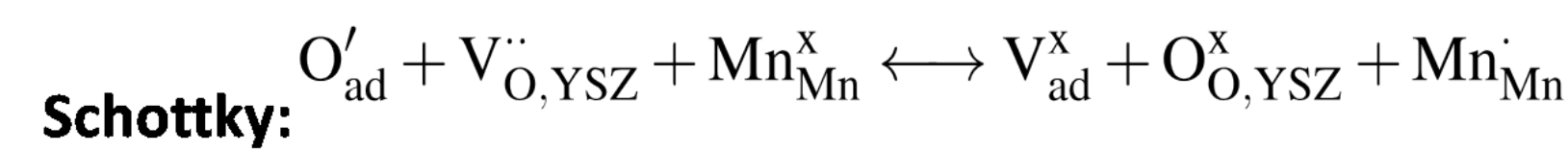Posterior distribution of the adsorption reaction's activation energy and pre-exponential factor

On Going

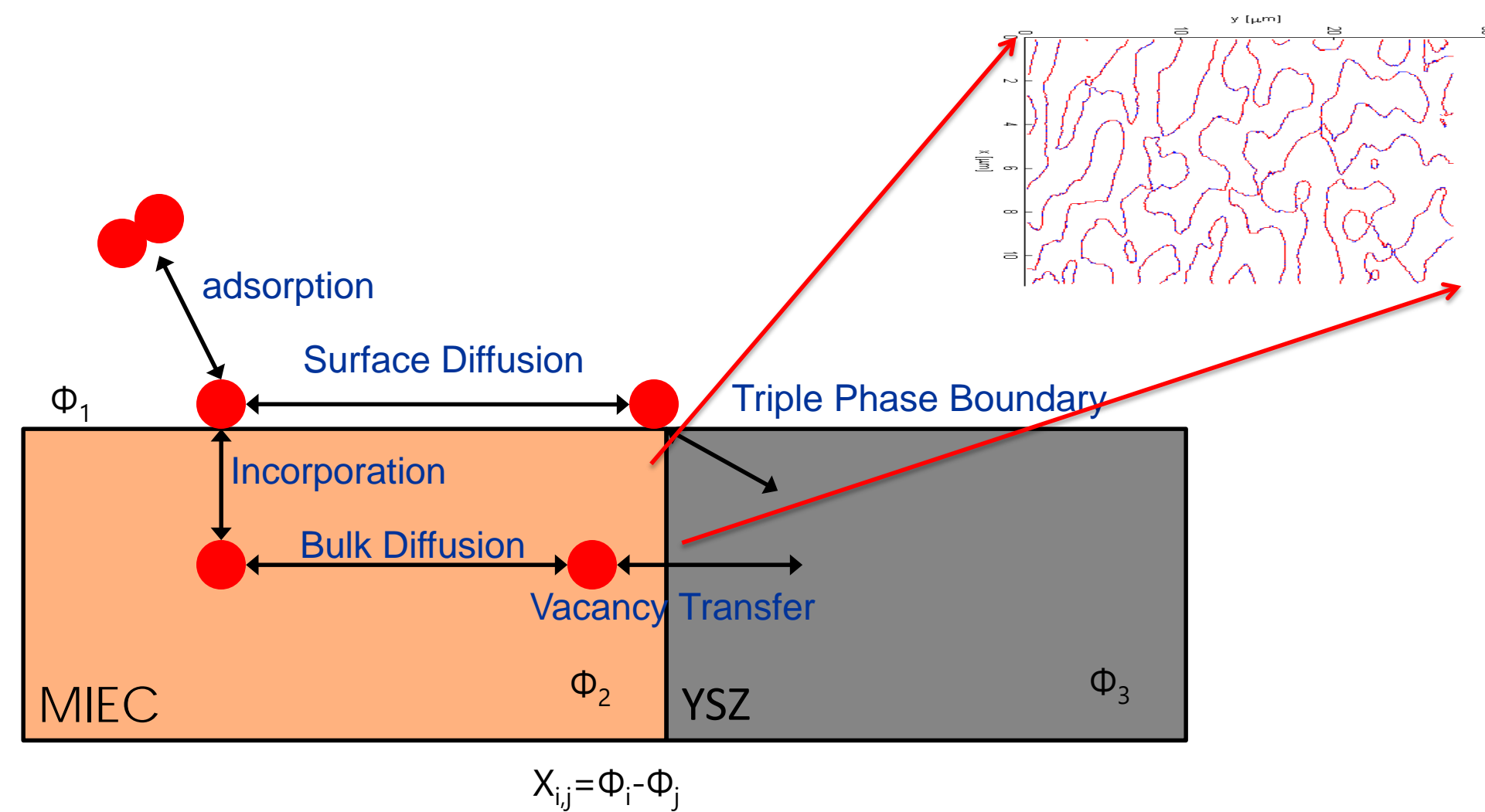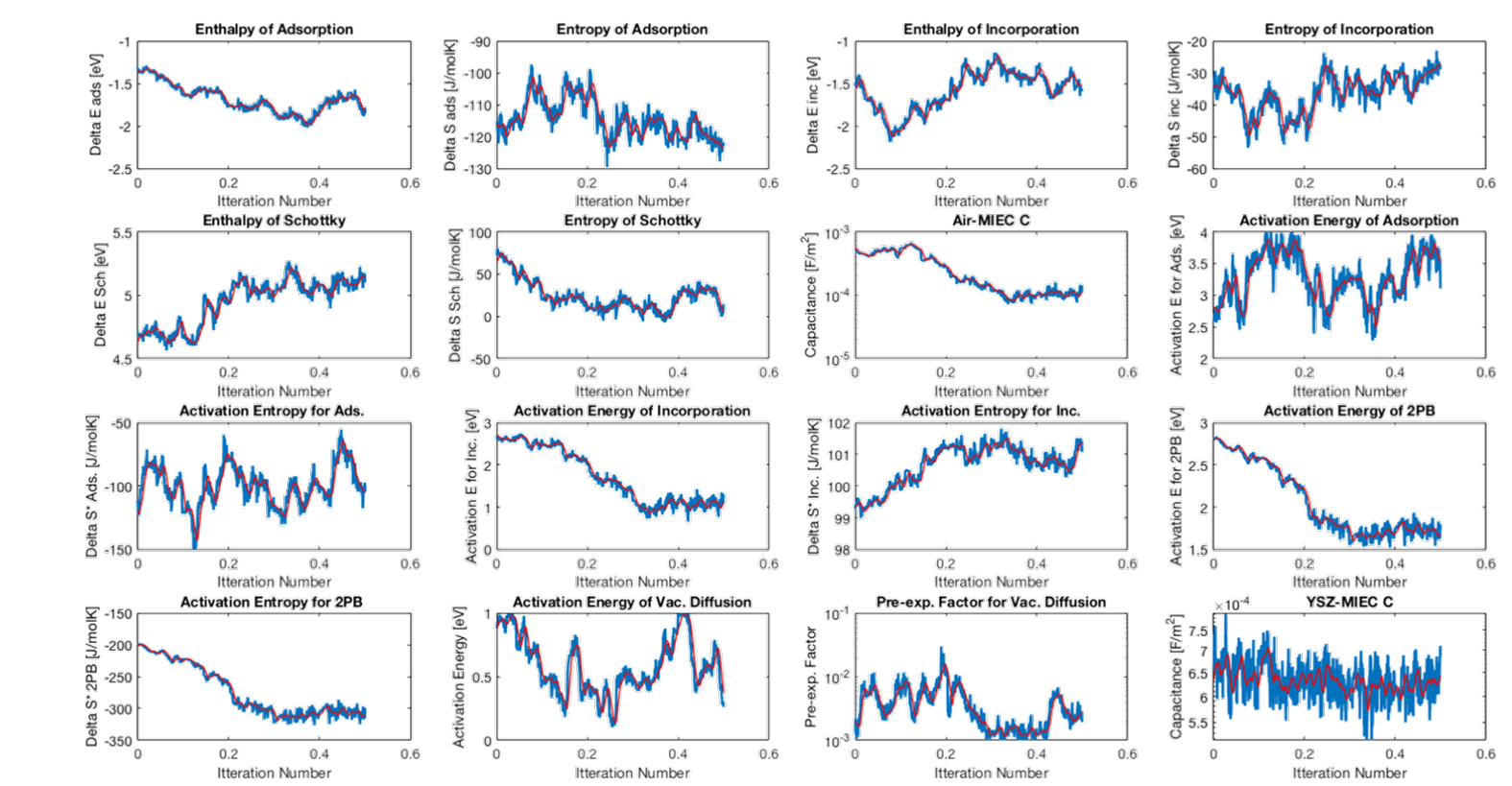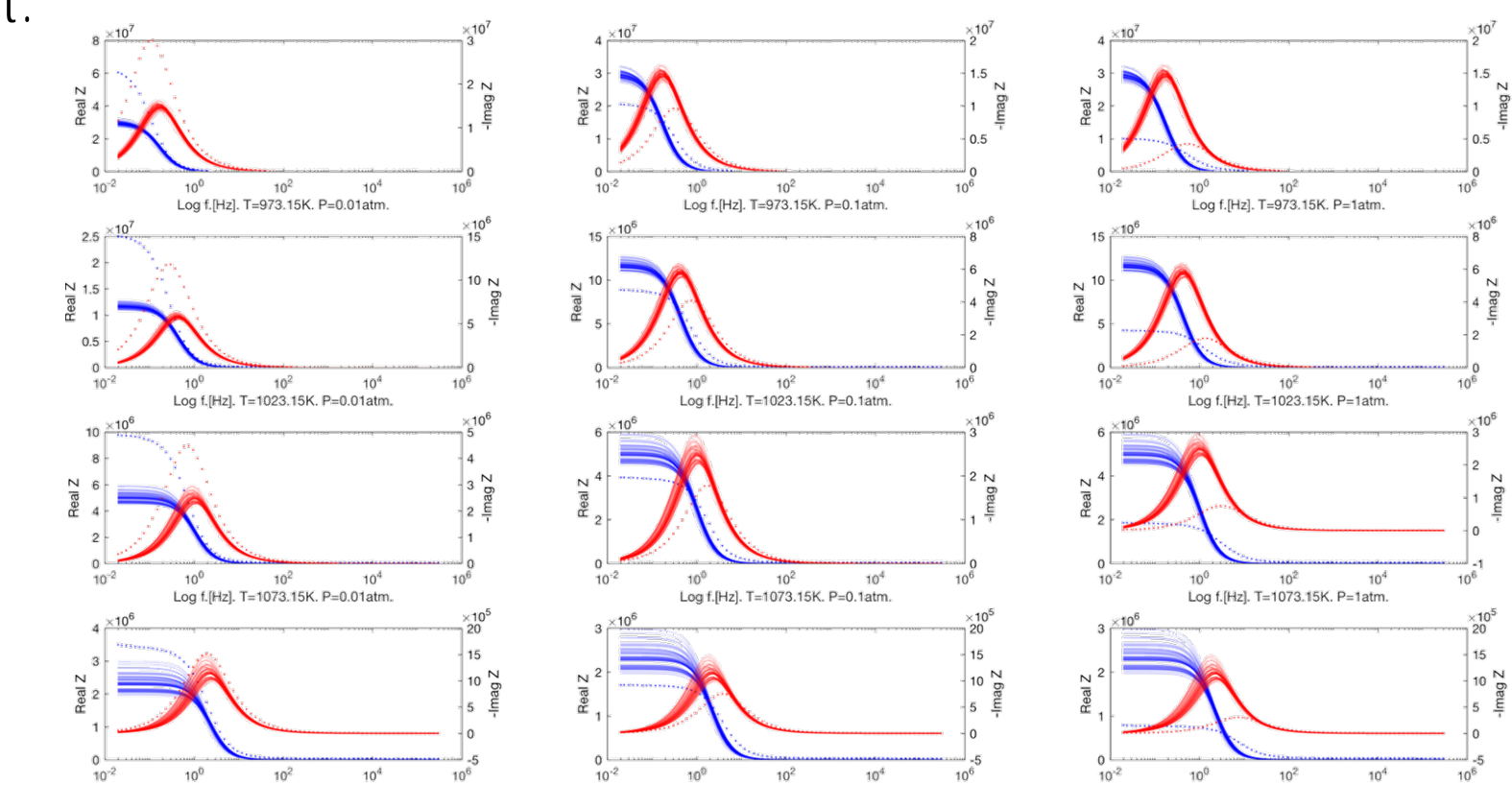We are currently investigating a thin film system at different temperatures and pressure using a modified version of the porous model. Currently, the simulations have not reached equilibrium and the pressure dependence is not yet correct.

Science & Engineering To Power Our Future