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Machine Learning for Reducing Parametric and Model Form Uncertainty of Predictive, Complex Chemical-Reaction Models

Dionisios G. Vlachos[1]*

1 *Department of Chemical and Biomolecular Engineering, Catalysis Center for Energy Innovation, Delaware Energy Institute, University of Delaware, Newark, DE 19716-3110, USA*
**Corresponding author: vlachos@udel.edu*

Highlights

- Process modeling incurs major uncertainty and experimental data fusion is essential.
- Thermodynamic properties are equally, if not more, important than kinetic ones.
- Prediction of materials discovery requires microstructure information.
- Machine learning and *in situ* spectroscopy can close the materials gap.

1. Introduction

Sustainability of chemical process requires more energy-efficient processes, utilization of renewable energy, such as solar and wind, to drive reactions and separations, better catalysts to improve activity and selectivity and thus to reduce separation cost and energy demand, new more efficient technologies, and our ability to tap into underutilized and renewable resources, such as offshore and stranded gas, biogas, food waste, and biomass. The distributed nature of many underutilized and renewable resources and the low energy density begs for distributed manufacturing, which can be achieved with modular systems and process intensification, such as plants on wheels. The design of such systems needs much more intimate process integration with high fidelity models. A cross cutting need in all of these systems is the need for better materials, whether catalysts, adsorbents, battery materials, or electrocatalysts, to improve performance, catalyst stability, and robustness and reduce cost. Multiscale modeling can be an important pillar toward meeting this dual – process and materials discovery – goal.

Over the past two decades, multiscale modeling has advanced tremendously, and several algorithms currently exist [1, 2]. Yet, our ability to apply first principles modeling to process design and materials discovery is seriously limited due to multiple challenges. In this talk, we will outline these challenges and introduce computational methods to overcome of them. Specifically, we will discuss how to handle complex reaction networks with first principles accuracy but at a very low computational cost [1, 3], how to estimate and reduce errors in multiscale models [4], how to determine the active site of a catalyst [5], and how to predict novel combinations of active sites to drive activity and selectivity. The concepts of small data, correlations in energies and entropies, correlative uncertainty quantification, machine learning for catalysis, and atomistic optimization for improved activity and stability, will be discussed. These concepts will be illustrated with examples focusing on ammonia decomposition chemistry and electrocatalysis in alkaline media focusing on the oxygen reduction reaction (ORR).

2. Methods

We will introduce multiple methods. First the group additivity method for catalytic surfaces using semi-supervised machine learning that picks up graphs (descriptors) automatically and reduces modeling error [6]. In combination with the extended linear scaling relations, it provides a comprehensive semi-empirical methodology for parameter estimation of complex reaction networks. Hierarchical refinement allows to introduce coverage effects and improve the accuracy of the important parameters, estimated by semi-empirical methods, to first-principles' level of accuracy.[1, 3] The same framework can be used for high throughput computing to screen materials. We will introduced the correlative global uncertainty quantification method to propagate errors in process and materials modeling.[4]

The graph-theoretical kinetic Monte Carlo (gt-KMC) method [7] allows one to resolve catalysts with atomic resolution and account for microstructure effects, and by doing this to overcome a crucial model form uncertainty. The recent acceleration wrapper provides automation and significant acceleration.[8] We will introduce various machine learning algorithms to create surrogate models for various tasks. We will formulate the catalyst activity in terms of the microenvironment of the active site and introduce optimization methods (simulated annealing and genetic algorithms) to predict optimal catalyst microstructure.

3. Results and discussion

It will be shown that model uncertainty is significant in process modeling and that experimental data fusion into multiscale models is essential. On the other hand, prediction of materials incurs very low error. Comparison of computational to experimental data demonstrates that a main uncertainty arises from the lack of knowledge and predictive ability of catalyst microstructure. We reveal correlations of vibrational frequencies [9] and discuss how *in situ* spectroscopy and machine learning can be integrated to provide the actual catalyst structure and close the materials gap. Finally, we will show that machine learning can be used with statistical mechanics to develop surrogate models that capture efficiently the active site microenvironment. Equipped with these methods, optimal catalyst prediction can be accomplished. It is shown that defect engineering can improve performance by at least one order of magnitude.

4. Conclusions

This talk discusses the grand challenges in multiscale modeling of complex chemical kinetic networks and provides a perspective on model uncertainty and model-error reduction. Both parametric and form uncertainty will be shown to be important, with the latter being critical and related to the microstructure and the active site of a catalyst. It will be shown that operando spectroscopy along with machine learning provide a link to address the model form uncertainty. Furthermore, it will also be shown that energetic models that account for the microenvironment of the active site are ideal to predict optimal facets and defect density of a catalyst.

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Keywords
Multiscale modeling; uncertainty; catalyst; microstructure.