PHYSICS-BASED AND DATA-DRIVEN IN-SILICO METHODS FOR THE INVERSE DESIGN OF NANO AND POLYMERIC MATERIALS

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Abstract

The rational design of novel high-performance materials is imperative to sustainably addressing the unprecedented challenges such as climate change, increasing energy demands, and pollution that are associated with anthropogenic activities. Design of novel materials has traditionally relied on the forward design process, a trial-and-error based approach where candidates are systematically synthesized, characterized, and finally benchmarked for its targeted application. The sequential forward design process is conceptually straightforward and has driven many major scientific discoveries over the years; however, this empirical methodology is inefficient as it is both time-consuming and resource-intensive due to the vast size of the chemical space. Despite advances in automated high-throughput experimental techniques that improve design space exploration rates by multiple orders-of-magnitude, this naïve brute-force approach has motivated development of inverse design approaches to guide rational design. Inverse design involves starting with a target material property and searching for designs that meet the specified criteria. Intelligent and efficient navigation of high-dimensional design spaces is required to address the intrinsic challenge that plagues inverse design problems as they often are ill-posed (or weakly conditioned) and may not have unique solutions.

In this thesis, we focus on developing novel physics-based and data-driven in-silico methodologies to solve inverse design problems for engineering novel materials (catalysts and polymeric materials) to drive high-throughput experimentation. We combine first-principles calculations with modern computing and statistical techniques such as Bayesian Inference, Bayesian Optimization, and machine learning to address two separate thrusts: (1) in-silico driven

spectroscopic analysis of material structure and (2) efficient inverse design of high-performance materials.

In Chapter 2, we combine data-based approaches with physics-driven surrogate models to develop a high-throughput methodology to predict catalyst structure directly from adsorbate infrared spectroscopy for supported subnanometer clusters. In Chapter 3, we present a novel plasma-driven methodology for synthesizing shape-controlled nano-catalysts and develop a computational methodology for identifying and monitoring structure during the synthesis procedure directly from adsorbate infrared spectroscopy and X-ray photoelectron spectroscopy. In Chapter 4, we study the fluxional nature of supported catalysts due to temperature from X-ray spectroscopy and elucidate structural changes using computational methods for structure-dependent chemistries. In Chapter 5, we present a novel molecular dynamics driven methodology called Rapid Analysis of Polymer Structure and Inverse Design strategY (RAPSIDY) to drive high-throughput screening of block copolymers stability with multiscale structures and accelerate the process over traditional computational methods by 2-orders-of-magnitude. In Chapter 6, we present a new version to RAPSIDY, RAPSIDY 2.0, to drive inverse design of high-performance block copolymers that exhibit desired macroscale material properties (e.g., tensile strength and thermal conductivity) using a combination of molecular dynamics and Bayesian optimization-driven active learning. Finally, in Chapter 7, the conclusions of the works presented in this dissertation are summarized and future research directions are discussed.