114b An Integrated Approach to Catalyst Design: Model Guided High Throughput Experimentation

Prasenjeet Ghosh and Anantha Sundaram

Discovery and design of novel high-performance catalysts requires an understanding across many dimensions. Detailed knowledge of the catalyst material structure and synthesis along with its complex interaction with feed and operating process conditions need to be effectively exploited to generate novel catalyst leads. Traditional design approaches have focussed on only one or a select few of these dimensions ignoring the other important dimensions of the problem. The use of high-throughput experimentation (HTE) in catalysis provides an accelerated way to explore multiple dimensions simultaneously. However, HTE for catalyst discovery needs to overcome two important problems. First, the combinatorial size of the problem, including the variety of feed and operating conditions is prohibitively large to navigate even with large parallel experiments. Secondly not all aspects, vital to catalyst performance in a commercial setting may be duplicated in a HTE setting. These problems severely limit the extrapolation of leads from HTE onto the pilot-plant or commercial phase. The use of computational models in tandem with HTE offers an effective way to navigate this combinatorial space for new catalysts. We develop a new two-phase approach to guide high-throughput experimentation for catalyst discovery by design.

The first phase is a prediction phase, that involves model development to predict catalyst performance from typical catalyst design parameters while the second phase is a guidance phase that searches and locates optimal design regions and allocates the next set of experiments. The prediction phase is constructed to best utilize the data and knowledge available on the catalyst performance. Results from catalyst design runs are typically in the form of the concentration profiles of the different reaction components. In the absence of any knowledge of the underlying kinetics the prediction phase would employ black-box statistical models to predict yields directly from catalyst design parameters. When a kinetic description is available, a correlative model would be applied only to predict the kinetic parameters as a function of catalyst characteristics. A rigorous reaction model would then generate the component concentration profiles. The guidance-phase of the catalyst design framework searches and locates optimal design regions using the predictive models. These regions are augmented by selectively choosing unexplored regions of the design space. The combined set balances focus on optimality with exploration and the next set of HTE is constructed based on this. The results from the HTE experimentation are then used to refine the prediction models, which are then used to pursue better solutions in the guidance phase.

We demonstrate these ideas using a simple, but typical series-parallel reaction scheme of four components, where the trade-off between conversion and selectivity is the objective to be optimized for catalyst performance. The two-phase approach is tested using data simulated on the above reaction system. The catalyst formulations are characterized by different design variables such as the amount of active mesoporous material, its pore-size, amount of metals dispersed on it, and the characteristics of the support. Two different prediction phase models are examined in this paper:- (i) Black-Box neural network models correlating catalyst design variables directly to product compositions and (ii) Grey-box models that use a combination of neural-networks and a kinetic model to connect catalyst design variables to predict product compositions. Stochastic search algorithms are used to search the design space using these prediction models and locate promising regions. Around these promising regions, a D-optimality based experimental design procedure is used to construct the next set of experiments. The results from these new experiments serve as a feedback to refine the black or grey-box models and the procedure is repeated until new catalyst designs are found. We show that depending on the nature of the prediction models (black or grey box), novel specialized or robust catalyst designs can be constructed to work under specific or widely different feed and operating conditions. We show that the iterative

refinement approach can converge to novel high-performance designs starting from a limited model of catalyst performance.