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Project Title: Mimetic Synthesis Modeling of Amorphous Carbon Adsorbents

Principal Investigator: Dr. Christoffer Heath Turner
Department of Chemical and Biological Engineering
The University of Alabama

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Summary

Our goal is to develop mimetic synthesis approaches for modeling amorphous carbon materials. This is very different than most modeling studies in the past, which can be classified as 'reconstruction' techniques. These previous reconstruction techniques have simply focused on developing methods for reproducing as-synthesized experimental targets. The main limitation of this approach is that they rely heavily on experimental structural data (X-ray or neutron scattering) to reproduce an atomistic model for the carbon. Our simulation approach is intended to generate realistic atomistic models from minimal experimental data, and it is designed to mimic the actual experimental synthesis process, yielding more insight into the nuances of the microporous structural evolution of the carbon.

In this work, we have begun the development of a hybrid simulation technique, combining kinetic Monte Carlo (KMC) and molecular dynamics (MD) simulations to model the atomistic structure of amorphous carbons. There are several stages to our model development. The actual KMC code is being developed in-house and adapted to parallel computing architectures (via OpenMP). Also, while the MD simulations are being performed with a currently-available simulation package (LAMMPS), the interface between the KMC and the MD simulation cycles requires careful scripting and benchmarking for optimization. Finally, the underlying dataset for populating the KMC rate-based simulations is being constructed from the literature data and scaling relations.

Research Progress

So far, we have made progress in several aspects of our simulation development areas. First, the KMC code development is well underway. This is the core focus of our overall strategy. The KMC code has been designed to incorporate a large (100+) database of reactions that may be occurring during the experimental synthesis process. Although this may not be an exhaustive list, it is intended to capture the dominant reaction paths that are known to exist during synthesis. While almost all of these reactions are kinetically-limited, we also include the reverse reaction steps (to maintain detailed balance), which may play a role at certain conditions. The code can capture both nearest-neighbors, as well as second-nearest-neighbors, and this allows us to take into account the local coordination environment on the reaction kinetics. This is critical for realistically capturing the structural evolution over a range of different thermodynamic conditions. Although KMC codes do not typically scale well on parallel processors, we have made modest increases in performance by introducing OpenMP pointers in several of the more compute-intensive subroutines.

While the basic KMC framework is nearing completion, we are still in the process of populating the rate-based kinetic database. This step is the most elusive in the overall modeling hierarchy. Some of these rate parameters have been pulled from the literature, but there are always some unknown parameters that must be extrapolated or fit based on representative systems. We intend to complete this stage very shortly, so that we can quickly begin the experimental benchmarking steps.

Finally, we have made a lot of progress on developing scripts for automatically cycling between different simulation packages (KMC and LAMMPS). Making this transition can be relatively straight-forward if performed manually, but developing an automated scripting approach is much more challenging. Since these MD/KMC cycles must be performed many times, we have spent much effort developing robust scripting tools for handling this integration. Furthermore, these automated scripts can be applied to a lot of other modeling scenarios, outside of our current research focus.

Our overall modeling approach is very quickly coming together. Several different individual pieces are nearing completion within the next few weeks, so we expect to have an integrated and robust modeling tool ready for benchmarking within a couple of months. Once ready, we already have a comprehensive list of different experimental targets that we will be using for estimating the predictability of our hybrid KMC/MD model.

Impact on Career and Students

So far, the biggest impact on the career of the PI has been the opportunity to add a completely new research direction to our lab. We would have never otherwise been able to dedicate the time, energy, and resources to make meaningful progress in this area. We have been eager for many years to try out some of our ideas, so it is particularly rewarding to be able to finally see some fruits from our initial brainstorming and some unconventional modeling approaches. Although we have not yet been able to present our work, we excited about making several presentations in the upcoming year.

Currently, a post-doctoral student has joined the modeling effort. Although she has a general background in atomistic and molecular-level simulations, she is definitely broadening her skillset by contributing to the project. She will also have opportunities to personally present her work in the upcoming year. Within the near future, we are also planning to recruit an undergraduate student to help with certain aspects of the project. We are now at a stage of development where we have carved out some roles that would be appropriate for an undergraduate student researcher, so we expect an additional student to acquire some hands-on experience with our modeling tools.