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Project Title: Mimetic Synthesis Modeling of Amorphous Carbon Adsorbents

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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 structural evolution of the carbon.

In this work, we have completed 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. The KMC code was developed in-house and combined with the MD simulation package LAMMPS through a scripting interface. These KMC-MD hybrid simulations are now being used to generate amorphous carbon structures and compare their structural details against currently-available modeling approaches and experiments. Furthermore, we have developed a number of different analysis techniques for automatically characterizing the properties of the generated carbon structures.

Research Progress

We have completed and performed benchmarks of our scripts for automatically cycling between different simulation packages (KMC and LAMMPS). These MD/KMC cycles must be performed many times, and we have spent significant effort developing robust scripting tools for handling this integration. Computational efficiency also arose as a challenge. As a result, we have performed significant code optimizations, such as the development of linked-lists and refinement of data structures. At this point, we now have an efficient and automated process for generating our amorphous carbon structures. Our more recent efforts have been focused on designing quantitative structural analysis techniques, so that we can make rigorous comparisons of structures generated at different conditions and against the experimental data. For instance, in Figure 1 below, we illustrate comparisons of our model against previous modeling efforts and experimental benchmark data. This shows the carbon-carbon radial distribution function of the sample, which is one of the common metrics used to evaluate the performance of the model. We are able to capture the positions of many of the g(r) peaks and the heights are very similar, as well.

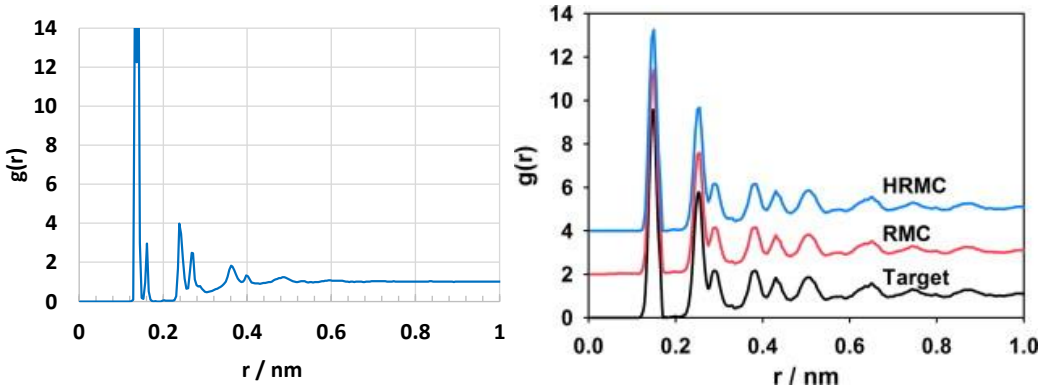

Figure 1. Comparison of our KMC-MD generated amorphous carbon structure (left) against literature data (right).[1] The g(r) represents the carbon-carbon radial distribution function.

In addition to some of the standard comparisons, we have also developed custom codes and algorithms for performing some deeper analysis. For instance, we are calculating properties like pore volume, pore size distribution, surface area, etc. These properties can be directly compared against the experimental structures in order to provide an even more rigorous analysis of our model quality. In Figure 2 and 3 below, we illustrate our pore volume analysis and our bond order analysis, respectively.

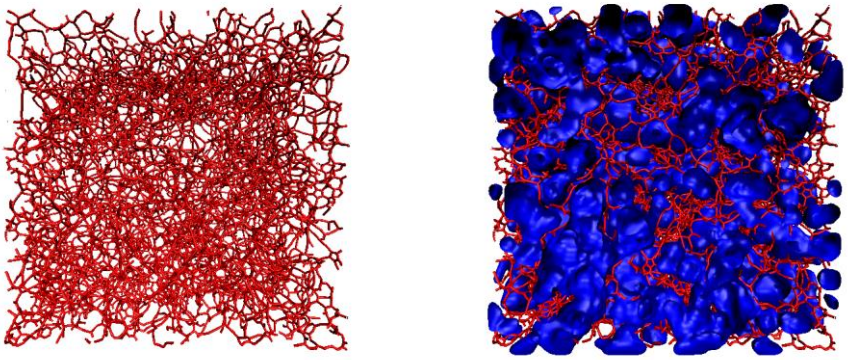

Figure 2. Atomistic structure of our KMC-MD carbon model (left), as well as an illustration of the pore volume available within the structure (right). The carbon atoms and bonds are shown in red, while the void volume is shown in blue.

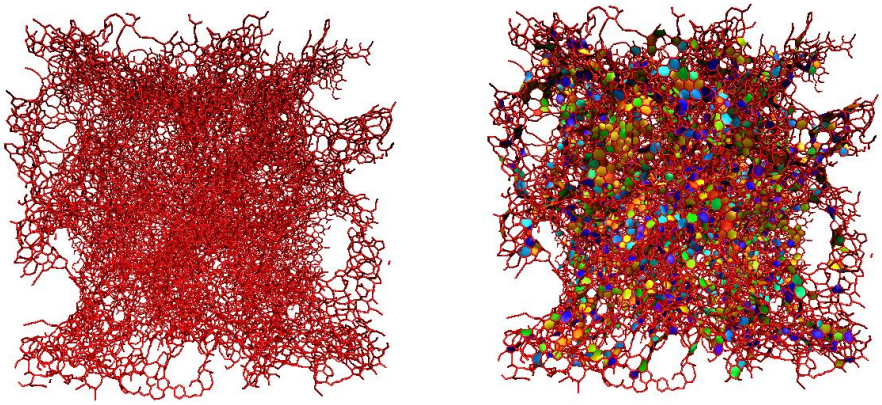

Figure 3. Atomistic structure of our KMC-MD carbon model (left), as well as an illustration of the local carbon-carbon bond order within the structure (right). Different colors are used to designate carbon rings with different numbers of carbon atoms present.

Impact on Career and Students

At this point, we have already been able to generate a great deal of data with regard to our modeling approach, and we have presented our results to a focused international audience. The response to our work and ensuing discussions have been particularly beneficial to the advancement of our efforts and have significantly raised the visibility of our work.

During the course of the past year, a post-doctoral student has been contributing to the modeling effort, and she has recently been able to obtain a full-time position in industry. Her success on this project was particularly helpful for her employment opportunities. More recently, we have added a new Ph.D. student to this project, and she has quickly become familiarized with the background and goals of our simulation efforts. I anticipate that this research project will be an excellent educational experience for her to develop her analytical and programming skills. Furthermore, we expect to generate several publications over the next year, which will provide important training opportunities for her writing, analysis, and scholarship.

(1) Palmer, J. C.; Gubbins, K. E. Atomistic models for disordered nanoporous carbons using reactive force fields. *Microporous and Mesoporous Materials* **2012**, *154*, 24-37.