Computational Modeling to Aid in Analysis and Interpretation of Multi-Modal Neutron Experiments

Research Objectives

Significance of

The over-arching goal of our project is create a streamlined workflow for experimental data analysis and interpretation needs of the neutron scattering community using atomistic modeling and simulation. Neutron scattering experiments require users to model and interpret data at the atomic/molecular level. With numerous software applications and a large array of different file formats with each, scientists tend to use a limited (and sometimes dated) subset of software tools to tackle data analysis from neutron experiments. This creates a barrier to use other methods or cutting-edge atomistic modeling softwares in their research that could help in bridging the gap between experiment and theory.

We are currently developing a modeling and analysis workbench called Integrated Computational Environment-Modeling & Analysis for Neutrons, or ICE-MAN. We hope to create a seamless transition from both different types of neutron scattering experimental data and computer modeling and simulation techniques to tackle multi-modal data analysis. An example of a workflow would be the study of a disordered material. First, neutron scattering experiments could yeild the average structure of the material, providing Bragg diffraction data, and the local structure via total scattering, providing the pair distribution function. Molecular dynamics (MD) simulations can provide a trajectory for an atomistic model of the material in time giving an ensemble of possible atomic configurations to compare to experiment. These sampled configurations from the trajectory could be converted and refined with reverse Monte Carlo modeling (RMC) to determine which atomic configurations produce the best fit to both the Bragg diffraction pattern and pair distribution functions produced from the neutron scattering experiments. The MD trajectories could also then feed into software to calculate the inelastic neutron scattering spectrum of the system. This data could feed back into the RMC modeling as a constraint. At present this process would be exceedingly time consuming, involve expertise in at multiple techniques, and most likely present a barrier that would seldom be overcome by general users.

We are requesting XSEDE's HPC resources to carry out this workflow on projects that are already ready to be fed into the pipeline and also for potential projects for the general neutron scattering community, specifically Users of the Spallation Neutron Source instruments.

Proposed Research

Project 1: Investigate structural modifications of irradiated silica for nuclear materials

The first project would be the study of the local structure changes in amorphous silica (SiO2) due to radiation damage. Previous studies have looked at the fine structure of ion tracks (narrow trails of permanent damage along penetrating heavy ion pathways) in thin film amorphous silica using small angle x-ray scattering (SAXS) measurements combine with non-equilibrium MD modeling and simulation techniques. These studies revealed that these ion tracks formed a shell around the path of penetration through the sample which consisted of a core lower in density and a shell high in density. The non-equlibrium MD calculations were carried out using an 30k atom system where the ion track was produced by instantaneous deposition of kinetic energy to the atoms in the simulation cell. More recently, simulation sizes of 600k atoms have also been carried out to compare to SAXS measurements as well. We intend to carry out similar simulations of comparable size that can be used to elucidate the structural changes observed in neutron scattering experiments of the average and local structure of these polymorphs of silica. Atomistic configurations from the end of this trajectory can then be fed into the RMC modeling to optimize the structure against the experimental data. The results of this study can help understand the fundamental degradation of silica materials exposed radiation damage and also to future work to manipulate nanoclusters within solid silica materials.

Project 2: Modeling of amorphous germanium and silicon to determine high-pressure phase transformations

The second project we would like to undertake is trying to determine different structural changes of high-pressure amorphous germanium (a-Ge) and silicon (a-Si) from different forms of compression or indentation loads that result in new materials for industry applications. Similar to our first project, we will be modeling systems based on silicon that have been probed via neutron scattering studies to determine structure changes while under high-pressure, being compressed in diamond anvil cells or being indented using diamond tips. Under compression, these samples become metallic and do not return to their initial state upon releasing the pressure. Based on either indentation or compression, different structures are observed with improved electronic and photovoltaic properties with cheaper manufacturing methods.

Our modeling and simulation effort will attempt to find atomistic models that best fit neutron scattering experiments probing the average and local atomic structure via diffraction and pair distribution functions along with vibrational density of states from vibrational spectroscopy. To create models of a-Ge and a-Si in agreement with all experimental data is still a current research challenge, and to date no such model exists. We have developed a structural relaxation program based on the Wooten-Winer-Weaire method to produce realistic random-network models of a-Ge and a-Si via the Atomic Simulation Environment (ASE). From this program, we have initial amorphous structures that retain their four-fold coordination and bond-angle deviations in qualitative agreement with experiment. We use these structures to then perform a range of equilibrium or non-equilibrium MD simulations to best replicate different pressure loading and thermal annealing cycles.

We feed atomic configurations from the MD trajectories into reverse RMC modeling to optimize the structure against the experimental data taken throughout thermal and pressure load cycles and the send this back to the MD simulations for further refinement and progess in the cycle. Experimentally constrained methods have been pursued in recent studies with success using very similar Monte Carlo relaxation methods. These include pure Monte Carlo using just the experimental data and hybrid approaches that add an emperical potential as an additional constraint. Our hope that using ICE-MAN's unique capability of lowering the barrier for data cross-over from the MD simulations to the RMC optimization to fit to experimental data will allow a larger search in the phase space and accelerate the convergence of a best fit to a variety of experimental data with strong emphasis on neutron data for these materials.

Project 3: Modeling oxygen insertion in one-dimensional channels of shafarzikite-like structures

Recent study of schafarzikite-like (FeSb2O4) materials have been shown to store relatively large amounts of oxygen (3.5% mass) within the structure at low temperatures (350 C) without decomposition, leading to new possibilities as potential oxygen storage materials. Specifically, oxygen insertion into cobalt and lead doped derivatives of shafarzikite show promise in applications of electro-catalysis due to their unique 1-D cation channels with high peroxide anion mobility and potential for high, directed electronic conductivity. We plan to analysis the neutron scattering data with RMC modeling to clarify the structures that occur before and after oxidization and also variants of the material with different doping agents. Structures generated from the RMC modeling can then be used to construct nudge elastic band calculations using density functional theory to determine transition state energy barriers during the oxidation process. Results from this study would help in clarifying the proposed defect cluster produced by the low temperature oxidation reaction.

Propose the research projects that we can answer.

3) Sankar's project

Computational Methodology (applications/codes)

LAMMPS

The main engine for the MD simulations will be the Large-Scale Atomistic/Molecular Massively Parallel Simulator (LAMMPS) software. This code is highly-scalable for a variety of machine architectures: IBM BG/L, Cray XT3, Cray XT5, and Intel clusters with GPUs (Comet). It can take advantage of GPU and Intel MIC accelerators with good performance scaling. For documentation on general benchmarking of LAMMPS, the following website contains several benchmark problems for a variety of machines: http://lammps.sandia.gov/bench.html.

Computational Research Plan

This is the plan

Table	Project	Machine	Program	Service-Units	Storage (GB)
1	Irradiated SiO2	Comet/Oasis	LAMMPS	300,000	500
2	High-pressure a-Ge/a-Si	Comet/Oasis	LAMMPS	300,000	500
3	Oxygen in FeSb2O4	Comet/Oasis	QE	100,000	500
4	RMC modeling	OSG	RMCProfile	25,000	100

Table 1: Summary of requested service units for projects

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Justification for Service Units (SUs) Requested

Additional Comments