Computational Modeling to Aid in Data Analysis and Interpretation of Neutron Experiments: From Irradiated Silica to Oxidization of Schafarzikite Structures

Research Objectives

The over-arching goal of our project is to 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 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 in their research between experimental and theoretical techniques.

We are currently developing a modeling and analysis workbench called the Integrated Computational Environment-Modeling & Analysis for Neutrons, or ICE-MAN. This is an extension of the Eclipse ICE project[1]. 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 yield the average structure of the material (diffraction data) and the local structure via total scattering (the pair distribution function, or PDF). Molecular dynamics (MD) simulations can provide a trajectory for an atomistic model giving an ensemble of possible atomic configurations to compare to experiment. Sampled configurations from the trajectory could be used as inputs into reverse Monte Carlo modeling (RMC) to optimize the structures against the the diffraction and PDF data produced from the neutron scattering experiments. The MD trajectories could also then be used 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 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 the outlined projects that already have a need for computational approaches to solve complex structural data analysis and also multiple potential projects for the general neutron scattering community, specifically Users of the Spallation Neutron Source (SNS) instruments at Oak Ridge National Lab. We propose three projects that have already been studied via neutron scattering experiments and have a need for atomistic modeling to aid in the data analysis process. These include irradiated amorphous silica for nuclear radiation damage on materials, high-pressure amorphous silica for cheaper manufacturing of electronics, and oxygen insertion into shafarzikite-like structures for oxygen storage materials.

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 (SiO_2) before and after irradiation of high energy (tens of MeV or more) heavy ion bombardment, or swift heavy ions (SHIs). At these high energies, electronic stopping dominates over nuclear stopping, implying the SHI interacts with the electronic structure of the irradiated target material[2–4]. The SHI path deposits energy to the electronic structure which then dissipates the energy to the nuclei of the atoms via kinetic energy, causing a local heat spike in the material radially outward perpendicular to the SHI pathway. In Figure 1, we show models of quartz (crystalline silica) before and after a SHI bombardment event, looking directly down the SHI pathway through the material. In (a) and (b), we simply color the atoms by type where silicon atoms are yellow and the oxygen atoms are red . In (c) and (d), we color the atoms by their velocity, where the velocity scale increases going from red → yellow → white. The black circles in (b) and (d) show the amorphized region of the quartz due to the large deposition of kinetic energy in the material.

Previous studies have looked at the fine structure of SHI tracks in amorphous silica using non-equilibrium MD modeling and simulation techniques[5,6]. The non-equilibrium MD calculations were carried out using the simple approach of instantaneous heating of the atoms in the simulation cell to produce the SHI track path. This method has been motivated by the fact that most of the energy transfer from the electronic system to the atomic degrees of freedom occurs on the femtosecond timescale[3]. The timescale of ionic displacements is much greater, implying the transfer is immediate. These non-equilibrium MD simulations of an ion bombardment event, or cascade simulations, are sensitive to the system size where one does not want the radial dispersion of kinetic energy to reach the periodic boundary (in fact, one hopes the cascade dampens well before this boundary). This indicates a large system size is necessary. Also, multiple cascade simulations must be realized to produce an ensemble of events so averaging can be carried out for statistically meaningful results.

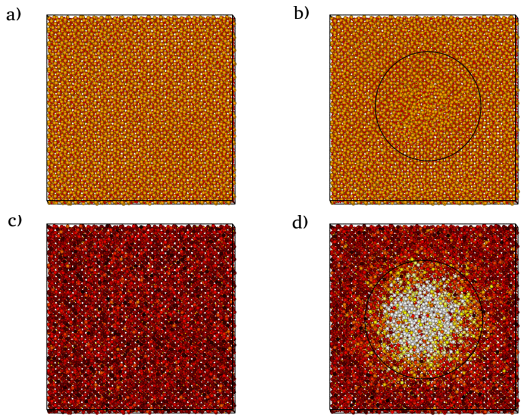

Figure 1: **Silica Before and After Swift Heavy Ion Bombardment** View down the SHI pathway. Before: (a) and (c) After: (b) and (d). In (a) and (b), color by type with silicon atoms as yellow and the oxygen atoms as red . In (c) and (d), atoms are colored by velocity (increasing from red → yellow → white). Black circles in (b) and (d) highlight amorphized region.

We intend to carry out similar simulations of comparable and larger size (500k to 1M

atom simulations) to those of previous studies that can be used to elucidate the structural changes observed in neutron scattering experiments of the average and local structure of different polymorphs of silica from a range of ion bombardment energies (up to GeV energies). Atomistic configurations from these MD trajectories can then be used to carry out RMC modeling to optimize the structure against neutron total scattering data, collected at the Nanoscale Ordered Materials Diffractometer (NOMAD) at the SNS. The results of this study can help understand the fundamental degradation of silica materials undergoing radiation damage[7,8] and also to future work to manipulate nanoclusters within solid silica materials using ion bombardment[9].

Using the instantaneous kinetic energy deposition technique to simulate the SHI bombardment event is good for a first order approximation. An extension of this project would be to incorporate recent, more realistic modeling[7,8,10–12] of the electronic heat conduction to the atomic degrees of freedom. In these models, the solution of the electronic heat conduction is embedded into the MD simulations via including an electronic continuum used to solve the heat equation for modeling the energy transport between the atomic and electronic subsystems. The two-temperature model formalism of including the electron-phonon coupling has been previously used in modeling and simulating SHI bombardment of alpha-quartz, laser ablation, and shock simulations[10]. For the ICE-MAN project, this extension would allow us to bridge a quantum-to-classical multiscale-modeling barrier. An important input parameter into the two-temperature model is the electronic specific heat. Recently, it has been shown that the electronic temperature dependence of the electronic specific heat can have an effect on the morphology of the SHI track that is formed[11,12]. To produce this relation, one calculates the difference in internal energy with change in the electronic temperature using finite-temperature density functional theory (DFT). Previous work of others have used the popular Quantum ESPRESSO[13,14] and the VASP[15–17] softwares to carry out these calculations. Thus, we hope to use this project as a proof-of-concept workflow for ICE-MAN to: begin with quantum calculations, use outputs of these calculations as inputs into non-equilibrium MD simulations, and then use the atomic configurations of outputs from these simulations as inputs into RMC modeling to optimize the atomic structures against neutron scattering data. The SHI workflow will not be a single-use case. Neutron scattering experiments have already been discussed for 2017 to look at other irradiated nuclear waste glass materials before and after irradiation.

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

The second project is determining 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[18–20]. Similar to our first project, we will be modeling systems based on silicon that have been probed via neutron scattering carried out under high-pressure, being compressed in diamond anvil cells or being indented using diamond tips. These experiments were carried out at the Spallation Neutron and Pressure Diffractometer (SNAP)[21] at the SNS. Upon releasing the pressure load, these samples do not return to their initial state but stay in a different, metastable

state. Based on either indentation or compression, improved electronic and photovoltaic properties have been observed in these materials. Also, high-pressure processing methods from this study could provide cheaper manufacturing methods for electronics[22].

We will determine atomistic models that best fit neutron scattering experiments probing the average and local atomic structure via diffraction and pair distribution functions from experiments on both SNAP and NOMAD, along with vibrational density of states from vibrational spectroscopy carried out on the VISION neutron vibrational spectrometer instrument[23] at the SNS. To create models of a-Ge and a-Si in agreement with a large range of 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 (WWW) method[24] to produce amorphized structures of a-Ge and a-Si via the Atomic Simulation Environment (ASE)[25,26] from initially crystalline structures. Using this technique, we get amorphous structures that retain their four-fold coordination and bond-angle deviations in qualitative agreement with experiments[24]. Equilibrium MD simulations can be used to further relax structures and non-equilibrium MD simulations can be used to replicate different experimental pressure loading and thermal annealing cycles. Atomic configurations from the MD trajectories are inputs into reverse RMC modeling to optimize the structure against the experimental data. 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 and faster 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 ($FeSb_2O_4$) 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[27].

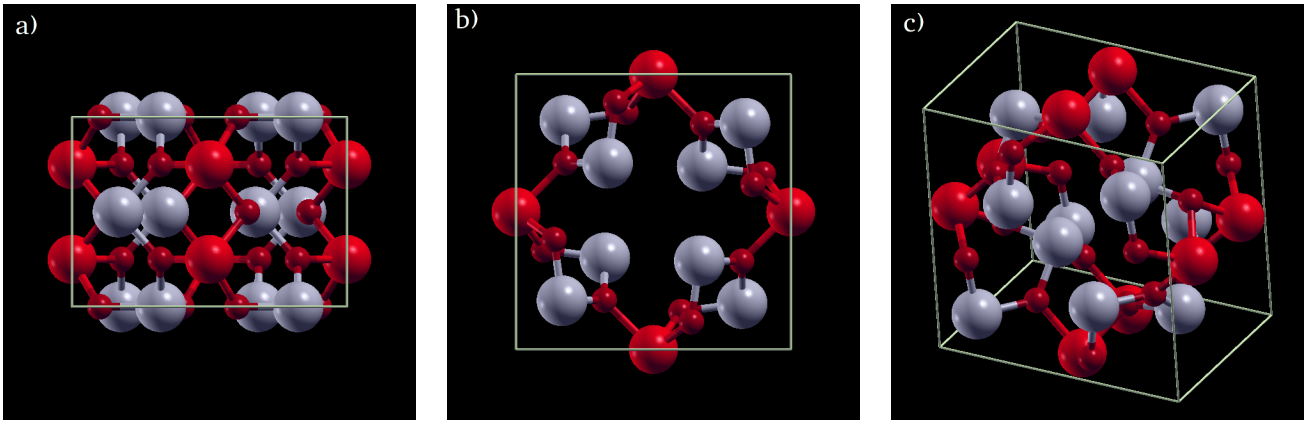

Figure 2: **Schafarzikite** Fe atoms: red O atoms: dark red Sb atoms: gray . Showing the orientations for (a) (100) (b) (001) and (c) (111).

In Figure , we show the shafarzikite structure along different axes. The Fe atoms are show as red , the O atoms are show as dark red and the Sb atoms are shown as gray . 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[27]. For this project, we plan to analyze the neutron scattering data first 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 nudged elastic band[28,29] calculations using density functional theory to determine the minimum energy pathway during the oxidation process. This would allow one to know the energetic barrier at the transition state. Results from this study would help in clarifying the proposed defect cluster produced by the low temperature oxidation reaction and help in proposing new chemistry of schafarzikite-like materials to optimize oxygen storage.

Computational Methodology (applications/codes)

LAMMPS

The main engine for the MD simulations will be the Large-Scale Atomistic/Molecular Massively Parallel Simulator (LAMMPS) software[30,31]. This code has been shown to be highly-scalable for a variety of machine architectures: IBM BG/L, Cray XT3, Cray XT5, and Intel clusters with GPUs (Comet)[32]. For the MD simulations, LAMMPS is used due to its extremely parallel platform, large and active developer base, and large range of features and MD techniques. This last point is a main reason we will LAMMPS over other codes that have shown better performance using GPUs (i.e. HOOMD-Blue[33,34]). Specifically, the electronic-phonon coupling capabilities are already present in LAMMPS that will be used for the cascade simulations.

DFT Codes

Currently, VISION[23] has its own dedicated computer cluster, Virtual Experiments in Spectroscopy with neutrons (VirtuES), for carrying out computer modeling as an integral part of the neutron data analysis and interpretation of the spectra (discussed more in the Additional Comments). This cluster has a variety of DFT codes installed for carrying out electronic structure calculations to determine vibrational density of states spectra. VISION Users have a suite of codes to choose from: VASP[15–17], Quantum ESPRESSO[13,14], CASTEP[35?] , ABINIT[36–38], CP2K[39?] , and RMG-DFT[40,41]. Also, the O'Climax software (the new version of aCLIMAX[42]) is currently being developed by the VISION team as software that can generate calculated incoherent INS spectra from the output of these listed DFT codes.

A major thrust of the ICE-MAN project is to have an interface to the O'Climax code within the first year of the project. This implies that ICE-MAN will also interface with

these DFT codes to create the workflow between these codes and eventually feed in to the structural modeling.

The DFT jobs will be kept to a minimum on the Comet resource due to VirtuES being available for these calculations and scaling is harder to achieve with DFT codes compared to empirical potential calculations/simulations. Yet, the VirtuES resource is for Users of the VISION beamline and can become saturated with work outside the scope of the projects listed in this proposal. Comet would serve as a supplementary machine for majority of the quantum calculation work proposed here.

RMCProfile

The majority of fitting to neutron scattering data will be carried out using the RMCProfile[43,44] software to perform reverse Monte Carlo optimization modeling. This software is able to fit many data types simultaneously (Neutron and X-ray total scattering, Bragg diffraction profiles, EXAFS, and single crystal diffuse scattering) and use a range of constraints to produce atomic models that are consistent with all the available data. The fitting will be carried out in a ”perfectly parallel” (or ”embarrassingly parallel”) manner where an array of fits will be carried out simultaneously on serial processors with no communication between the processes. This makes the Open Science Grid XSEDE resource optimal to carry out the fitting optimization. The ICE-MAN project is an extension of the ICE project[1], which is already able to handle the launching and monitoring of local and remote jobs, visualizing and analyzing data, and managing data transfers. Thus, we will work towards automating a seamless transition from taking data located on an HPC resource generated by atomistic modeling and simulation and transferring it to the High Throughput Computing (HTC) resource where it will automatically be launched for fitting optimization, preserving HPC service units for jobs that can make use of the resource. This project would provide an impetus to push parallelization of the RMCProfile software to handle the large atomistic configurations generated and make use of the HPC resources, reducing optimization timescales.

ICE-MAN

Developing ICE-MAN on XSEDE resources would open up the machines that the software is available on and diversify the architecture development. Users at neutron scattering facilities who already have access to XSEDE resources or collaborate with other research teams that do could have ICE-MAN as a common platform to bring together their atomistic modeling data with their neutron experiment data.

Comet

Comet is the ideal resource for our research for the following reasons:

1. It provides a heterogeneous research platform that is capable of providing domain-specific, ideal architectures. The compute nodes are ideal for our large-scale atomistic simulations and the large memory nodes are ideal for our quantum calculation work. The GPU nodes are available to tackle very large problem sizes that make use of massive parallelization efficiently.

2. We are targeting the Oak Ridge National Laboratory Leadership Computing Facility resources (i.e. Titan) to launch ICE-MAN. However, due to all nodes containing a GPU on Titan, we can only use the machine to its full potential for a subset of our project. Using Comet as the first target machine allows us access to the resources we will eventually use, broaden the scope of machines that ICE-MAN can utilize, and be accessible to a larger part of the research community.

3. Members of the team already have experience using Comet and have had publications as a direct result of the allocations awarded on the machine.

4. The Data Oasis Lustre parallel file system ensures plenty of scalable storage available for the jobs run on the machine.

Open Science Grid

The Open Science Grid would be an optimal resource to use for launching multiple arrays of RMC jobs in a high throughput manner for large spanning of the phase space. The ICE-MAN project could take advantage on its already-underlying remote job launching capabilities to transfer work to the appropriate computational resource.

Performance and Scaling

Classical/empirical potential MD simulations scale much better than their electronic structure counterparts due the parallel strategy being simpler and more straightforward. Thus, the larger jobs using the most amount of nodes in this project are the MD calculations. We focus below on the scaling and performance of the silica simulations using LAMMPS on Comet (CPU only simulations).

In Figures 3 and 4, we show the benchmark calculations on Comet for both bulk quartz simulations and two-temperature cascade simulations, respectively. In these figures, we show the strong scaling (speedup and parallel efficiency for a fixed problem size), performance (MD steps per second) and weak scaling (fixed number of atoms per node) for each system. For the simulation setup, the system size consists of 648k atoms (Strong scaling: problem size, Weak scaling: atoms / node), we used a Tersoff empirical potential[45,46], and the simulations were for 100 MD steps with a 1 femtosecond timestep.

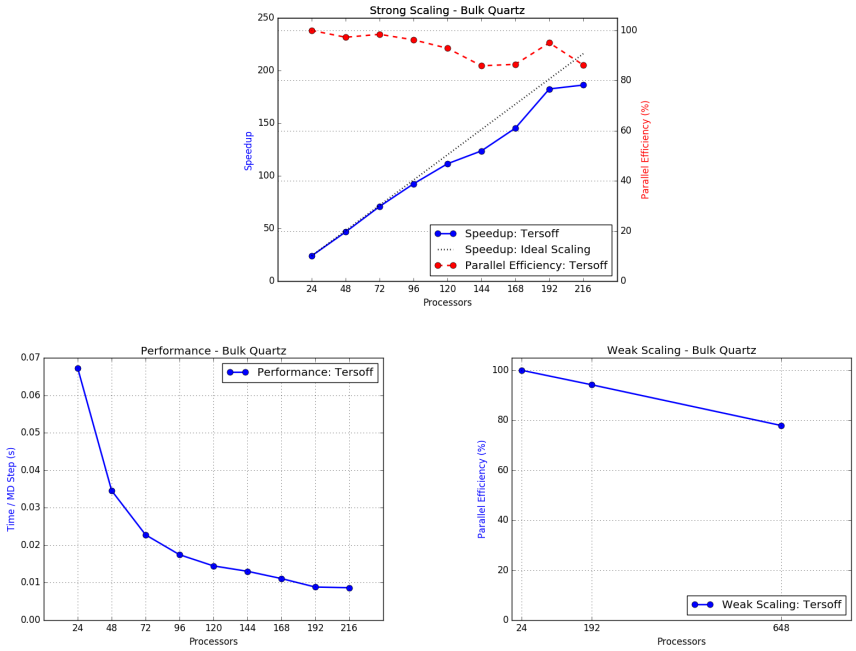

Figure 3: **Performance and Scaling of LAMMPS for Quartz Simulations.** Simulation consists of 640k atoms, Tersoff Potential, Δt = 1 fs, for 100 MD timesteps.

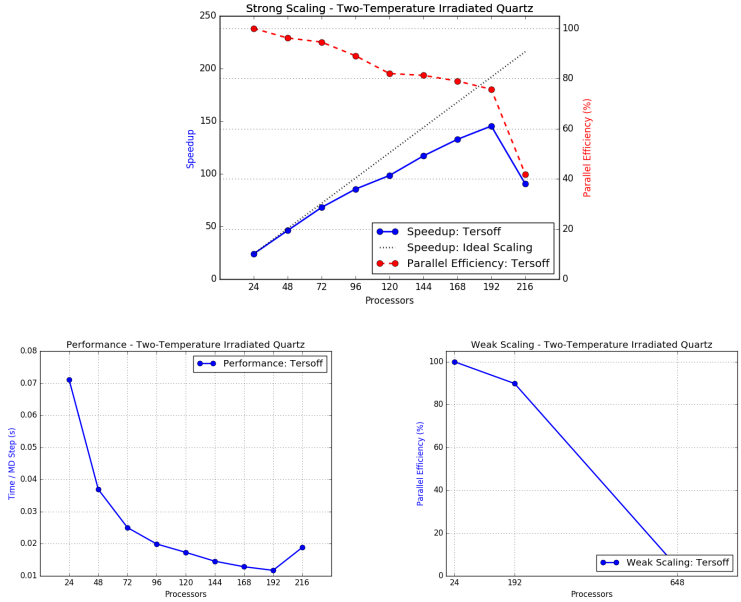

Figure 4: **Performance and Scaling of LAMMPS for Irradiated Quartz via Two-Temperature Modeling Simulations.** Simulation consists of 640k atoms, Tersoff Potential, Δt = 1 fs, for 100 MD timesteps.

Computational Research Plan

Irradiated Amorphous Silica by Swift Heavy Ions Swift heavy ion cascade simulations of amorphous silica will be carried out to produce atomistic configurations that can be directly compared and optimized to neutron scattering experiments via RMCProfile. This material will be first of many irradiated nuclear waste glass materials we will study via neutron scattering experiments in the upcoming year. ICE-MAN will be developed and optimized for the automated workflow between these modeling techniques: non-equilibrium MD simulations to RMC fitting of experimental data. Realistic modeling of the electron-phonon coupling via two-temperature modeling will also be explored. This will require electronic structure calculations to determine the temperature-dependent electronic specific heat, which will be an input into the SHI cascade simulations. The simulation trajectories will again be inputs into RMC modeling and fitted to neutron experimental data.

Amorphous Ge/Si via High-Pressure a-Ge and a-Si will be modeled using varying compression and nanoindentation MD simulations to replicate experimental preparation methods. Using RMCProfile, atomic configurations from MD simulations will be used to optimize and fit against neutron scattering experiments. These optimized structures can be fed back into the MD simulations to overcome barriers not accessible in the temporal limits of MD or for virtual experiments to help direct future material processing methods. ICE-MAN will be developed to reduce the barrier in the forward and backward direction of data transfer between the MD and RMCProfile modeling.

Oxygen Insertion in 1-D Cation Channel Materials RMCProfile will be used to clarify atomic structures of schafarzikite-like, 1-D cation channel materials before and after oxidation based on available neutron scattering data. The final structures can then be linked via electronic structure nudge elastic band calculations to determine minimum energy pathways for oxygen insertion into the lattice structure. ICE-MAN will again handle launching and data transfer workflow between the RMCProfile and quantum modeling calculations.

Justification for Service Units (SUs) Requested

For the irradiated amorphous silica by swift heavy ion project using the thermal spike via instantaneous deposition of kinetic energy, we plan to carry out upwards of 100 cascade simulations. We will use 8 nodes (192 processors) per simulation with a 1 femtosecond timestep with a total simulation time of 1 nanosecond (equilibration and production combined). Thus, given that it will take 0.009 seconds per MD step, we will require 48,000 service units. The two-temperature model for the electron-phonon coupling simulations will also be upwards of 100 cascade simulations. We will use 5 nodes (120 processors) per simulation with the same timestep and simulation time. With 0.017 seconds per MD step, we will require 57,000 service units. Also, we request 50,000 service units for carrying out DFT calculations to determine electronic specific heat for input into the MD

simulations. The Quantum ESPRESSO code is currently planned to be used for these calculations. Thus, the total time will be 155,000 service units for the silica project. However, we already have another proposed nuclear waste glass material that will be studied via neutron total scattering. This experiment could be completed as soon as April 2017. Thus, we request an additional 155,000 service units to also support this work, bringing the total requested service units to **310,000** for this part of the overall project.

For the amorphous silica under high-pressure, we request 40,000 service units to carry out the parallel structure optimization jobs for producing the initial amorphous silicon and germanium configuration. We also plan to carry out upwards of 100 simulations under different pressure conditions (compression and nanoindentation). We will use 8 nodes per simulation with a 1 femtosecond timestep with a total simulation time of 20 nanosecond (cycling of equilibration and production for continual pressure loading). Thus, given that it will take 0.009 seconds per MD step, we will require 600,000 service units for this part of the project. This will bring the total requested service units to **640,000** for this part of the overall project.

We request **100,000** service units for carrying out DFT calculations for the schafarzikite project. With poor scaling, we plan to use no more than 8 nodes per job. The Quantum ESPRESSO code is currently planned to be used for the schafarzikite calculations. These calculations will be shared between the VirtuES machine and Comet.

We request **50,000** service units for running RMCProfile optimization modeling on the Open Science Grid. These calculations will be launched as arrays of serial jobs.

Table 1 summarizes the justification for the requested resources to begin our project.

Table	Project	Machine	Program	Service-Units	Storage (GB)
1	Irradiated SiO2	Comet/Oasis	LAMMPS/QE	310,00	500
2	High-pressure a-Ge/a-Si	Comet/Oasis	LAMMPS	640,000	500
3	Oxygen in FeSb2O4	Comet/Oasis	QE	100,000	100
4	RMC modeling	OSG	RMCProfile	50,000	100
	Total	Comet		1,100,000	
		Oasis			1,100
		OSG		50,000	100
	Grand Total			**1,100,000**	**1,200**

Table 1: Summary of requested service units for projects

Additional Comments

VirtuES is available but is dedicated to VISION Users at the SNS. The machine consists of 50 nodes with each node consisting of two 16-core Intel Xeon E5-2698 v3 running at 2.30GHz.

MTM has currently just finished using a initial startup allocation for 100,000 total SUs from a previous project on the two XSEDE HPC resources at the San Diego Supercomputer Center: Comet and Gordon (50,000 SUs each)

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