Equation of State Developments in T-1

T-1 Equation of State (EOS) Team

The Equation of State (EOS) and Mechanics of Materials Group (T-1) at Los Alamos National Laboratory conducts research in EOS to support the nuclear and conventional defense communities, civilian research communities, and commercial applications. The group maintains the SESAME EOS and Materials Properties Library.

SESAME Library Update

In February we updated the unclassified SESAME library to include approximately eight new EOS, while the classified library will include five new EOS. The update will also include additional cold + nuclear sub-tables where possible and a large number of edits to the comment tables. We have also produced more than ten additional EOS for special applications.

Multiphase and Multicomponent Equations of State

We have completed an ASC Level-2 milestone by delivering an improved capability for modeling multiphase materials, and we have also improved our models for plutonium.

(a) New techniques have been developed for using Density Functional Theory (DFT) to determine EOS properties, and these techniques have been applied to phases of plutonium.

(b) New models and algorithms for handling multiphase materials have been incorporated into the EOS-production code OpenSesame.

(c) A new hydrocode interface capability giving users access to phase data has been put into the code EOSPAC.

Combining theory with experimental data, parameters have been determined for eight phases of plutonium, leading to new predictions concerning the high-pressure phase diagram. Anomalous properties of liquid plutonium have proved difficult to model consistently, and improving the liquid EOS will be a high priority in the coming months, along with verifying the hydrocode implementation of the models.

We continue to develop a new EOS for carbon dioxide as part of the expansion of EOS capabilities within T-1 to multiphase and multicomponent systems. The proximity of carbon to oxygen in the periodic table allows us to use a simple average atom approach. The final EOS will be finished soon.

Beryllium is a very important technological material, specifically in aerospace and nuclear engineering, because of its light weight, high melting point, and large bulk and shear moduli. However, its phase diagram has remained virtually unknown over decades. In particular, its melting curve has never been measured beyond 6 GPa, and numerous empirical models have not converged on beryllium melting temperatures at high pressures. Using the Vienna *Ab Initio* Simulation Package (VASP), we calculated the melt curve to 10 Mbar using *ab initio* MD solid-liquid coexistence simulations. We studied three (bcc, hcp, and fcc) solid phases of beryllium using computational cells of different sizes. The three melting curves, each for a given solid phase, are essentially identical and form a melting curve of beryllium, which is in excellent agreement with the one predicted by the most recent SESAME Be EOS—SESAME 2024.

Density Functional Theory

To complement experimental data and modeling in boosting the reliability of EOS, our DFT calculations continue to provide data, in particular cold curves and thermal contributions from the electrons and phonons. The reliability of these results is supported by collaborations with researchers of the High Pressure Science and Engineering Center at the University of Nevada, Las Vegas, whose measured phonon densities of states of tin under pressure agree well with results from our calculations.

Vibration Transit Theory
More progress has been made on the Vibration-Transit (V-T) theory of liquid dynamics, both in the development of the theory and its application to actual systems. Through extensive quenching studies in simulated sodium, we have found further support for the main postulate of the theory, the equivalence of the atomic configurations (called random valleys) in a monatomic liquid. Explicit expressions within the theory have been derived for the vibrational contributions to the density and current autocorrelation functions. These quantities are directly related to the spectra measured by x-ray or neutron scattering. We have gained insight into the physics of these processes by studying the multimode expansions of the autocorrelation functions, and we have also studied the microscopic nature of low-temperature transits. In collaboration with the University of Trento, V-T theory is being applied to water.

We are developing a set of new tools based upon the tessellation of real space into Voronoi polyhedra (Wigner-Seitz cell-like constructions) as part of the ongoing elucidation of V-T theory. Within this theory, liquid state properties are derived from the random valleys and transitions between them. An efficient, general characterization of these random valleys is crucial for both the verification and use of V-T theory. By applying several localized Shannon entropies as well as other statistical tools to the geometry and topology of each atom's Voronoi polyhedron, we are able to effectively probe the local structure of the system to detect various types of localized order or symmetry in real space. Our suite of Voronoi-based tools will thus aid in the identification and classification of random valleys, simplifying the general application of V-T theory.

We have succeeded in combining V-T theory with DFT methods to accurately predict liquid state properties of sodium at ambient pressure, such as the density of the liquid at melt, its bulk modulus, entropy, and internal energy. This approach is as accurate but computationally significantly cheaper than a Carr-Parinello or Born-Oppenheimer molecular dynamics (MD) simulation. We will submit these results for publication.

OpenSesame
We have improved the capabilities of the EOS-production code OpenSesame in the following ways:

(a) We have greatly simplified the build process by using standard GNU utilities and support a variety of platform/compiler combinations, expanding the list as users require.
(b) We have implemented an issue-tracking system that will allow users to report issues and follow the progress of their resolution.
(c) We have moved the source code to a subversion server, which allows users to participate in the code development process to whatever degree is desired.
(d) We have updated the OpenSesame home page, https://keiki.lanl.gov/opensesame, to allow users to download source code directly.

Our goal this year is to release a stable version 1.0 and then begin work on two longer-term projects:

(a) producing a new Java-based graphical user interface, which will ultimately absorb the entire code, to provide superior portability and even further simplify code installation, and
(b) setting up a build farm that will build and verify the most recent version of the code, incorporating the previously developed validation suite.

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