DOE/NA-0066

NNSA National Nuclear Security Administration

Office of Research, Development, Test, and Evaluation

Stockpile Stewardship Quarterly

VOLUME 7 | NUMBER 4 | DECEMBER 2017

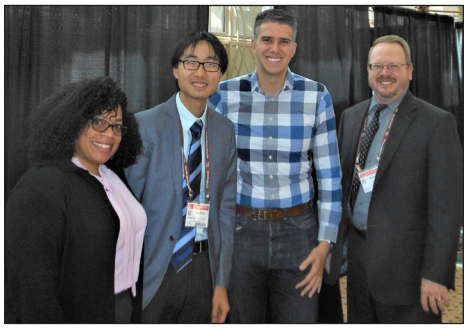
Message from the Assistant Deputy Administrator for Research, Development, Test, and Evaluation, Dr. Kathleen Alexander

In stockpile stewardship, one of the most widely studied phenomena is the equation of state (EOS). An EOS is a mathematical relationship that describes the state of matter (i.e., gas, liquid, or solid) using the material properties of temperature, volume, pressure, and internal energy. The EOS characterizes the properties of a state of matter under a given set of physical conditions. It is of interest for the metals and non-metals of stockpile stewardship application and is critical to understanding material behavior. Each of our weapon laboratories has extensive EOS activities. This issue of the *Stockpile Stewardship Quarterly* (SSQ) highlights representative EOS activities at our national laboratories, the Nevada National Security Site, and in the Laboratory Directed Research and Development (LDRD) program as well as efforts to continue our pipeline through the Research, Development, Test, and Evaluation academic alliances programs.

We begin with an article that discusses how Sandia National Laboratories (Sandia) uses pulsed power to carry out EOS experiments necessary to model and understand nuclear weapon performance. Sandia's Z pulsed power facility is providing capabilities that are unique for measuring the EOS properties of materials under dynamic compression. At Lawrence Livermore National Laboratory, Gbar pressures on the National Ignition Facility (NIF) are reaching unexplored regimes of density and pressure. NIF experiments on EOS allow us to address long-standing programmatic questions and meet new challenges associated with our national security mission. Los Alamos National Laboratory (LANL) takes a different focus, looking at the EOS and shock-driven decomposition of 'soft' materials. LANL uses a two-stage gas gun to collect shock data of polymers to incorporate in their EOS.

The Joint Actinide Shock Physics Experimental Research facility two-stage gas gun at Nevada National Security Site (NNSS) is used for both hazardous and non-hazardous material studies for input to EOS tables. Many EOS experiments at NNSS involve dynamic materials and associated diagnostic developments pertaining to stockpile materials.

Some of our innovative EOS work originated from the LDRD program projects at Sandia. Two Sandia projects are pushing the envelope to deliver EOSs that provide data used to determine phase transitions accurately under extreme conditions. We close out this issue by showcasing the Office of Inertial Confinement Fusion efforts to prepare tomorrow's stockpile stewardship leaders through the academic alliances programs. The High-Energy-Density Sciences Summer School aimed to inspire the 114 young scientists in attendance to become the next generation of leaders in stockpile stewardship.



Hong Sio, NNSA-supported senior Physics Department PhD student at the Massachusetts Institute of Technology (second from left) found time to strike a pose with (left to right) Dr. Njema Frazier and Dr. Bryan Sims (Research, Development, Test, and Evaluation) and Mark Visosky (TechSource, Inc.) at the 59th Annual Meeting of the American Physical Society Division of Plasma Physics in October 2017. Hong designed and implemented a new and unique diagnostic that can measure, simultaneously, multiple-nuclear-burn histories occurring during an ICF implosion. His innovative diagnostic work at NIF and OMEGA led to his invited talk by APS. Congratulations, Hong!

Your efforts in 2017 were greatly appreciated. Happy new year.

Inside

Comments

The *Stockpile Stewardship Quarterly* is produced by the National Nuclear Security Administration (NNSA) Office of Research, Development, Test, and Evaluation. Questions and comments regarding this publication should be directed to Terri Stone at terri.stone@nnsa.doe.gov.
Technical Editor: Dr. Joseph Kindel | Publication Editor: Millicent Mischo

Equation of State Research at Sandia National Laboratories *by T.R. Mattsson, D.G. Flicker, S. Root, and L. Shulenburger (Sandia National Laboratories)*

An EOS is a mathematical relationship that describes the state of matter (i.e., gas, liquid, or solid) using the material properties of temperature, volume, pressure, and internal energy. The EOS characterizes the properties of a state of matter under a given set of physical conditions. For example, the EOS determines how hot a material gets when subjected to a strong shock wave. High-fidelity EOSs are, therefore, necessary to model and understand a broad range of systems, including the structure of Earth and giant planets, high energy density (HED) and inertial confinement fusion (ICF) experiments on the Z machine at Sandia National Laboratories (Sandia) or the National Ignition Facility (NIF) at Lawrence Livermore National Laboratory (LLNL), and nuclear weapon performance. It is difficult to overstate the importance of using high-fidelity material models when performing multi-physics hydrodynamic simulations for stockpile stewardship applications, a critical activity at all NNSA national laboratories.

Under extreme conditions in temperature, density, and pressure, the EOS for solids, liquids, gases, and dense plasmas are far more complex than textbook EOS models like ideal gas or van der Waals. Many different experimental techniques, therefore, exist and new methods are developed to create and diagnose extreme conditions of matter. An example of a new exciting development for Z is the pre-compression capability described in the article "Compressing Materials Under Extreme Conditions" on page 11. By compressing the sample, otherwise immiscible fluids can be studied as a homogenous liquid mixture and a range of additional thermodynamic conditions can be accessed since the loading path is shifted compared to experiments at normal pressure.

Sandia has a long history of research in EOS, a history that was recently documented in the book "Impactful Times: Memories of 60 Years of Shock Wave Research at Sandia National Laboratories" by Jim Asay, Lalit Chhabildas, Jeff Lawrence, and Mary-Ann Sweeney.[1] The book presents six decades of development of experimental, material modeling, and hydrodynamics code technologies. At present, EOS properties are measured at multiple Sandia facilities: the Shock Thermodynamic Applied Research (STAR) facility holds several powerful gas guns/launchers and a range of diagnostics; the Dynamic Integrated Compression Experimental (DICE) facility with a small launcher and the 2.6-MA pulsed power machine VELOCE; the first next-generation pulsed power platform THOR described in an accompanying article "New Flexibility in Dynamic Materials Research Via Pulsed Power" on page 10 and finally the powerful Z machine. In addition to the experimental capabilities, our dynamic material properties program has a distinguished record of collaborative efforts between experiments and corresponding *ab initio* calculations, principally those with density functional theory (DFT). Theory is used in direct simulation of experimental conditions achieved on Z and other platforms, and these calculations have been critical to developing high-fidelity physics models for the design and simulation of Z experiments. Use of DFT is now routine for calculating EOSs and electrical and thermal conductivities.

The Z machine is a multi-purpose high energy density (HED) facility that provides capabilities that are unique in the world for measuring the EOS properties of materials under dynamic compression:

- Flexible pulse shaping that enables adjustable loading rates for ramp compression, shock-ramp compression, and very high pressure (10-20 Mbar or 1,000-2,000 GPa) shock experiments

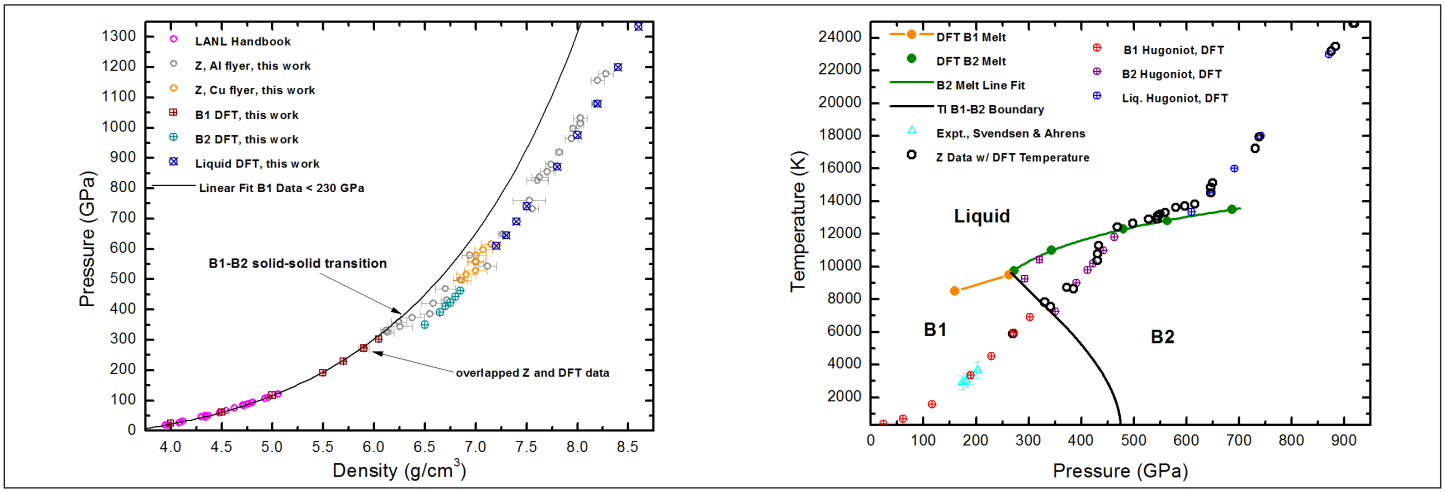


Figure 1. Shock compression of MgO. Left: Shock pressure as a function of density from first principles theory, experiments on Z, and data from the shock handbook.[3] The excellent agreement is strong support for the theoretical work.[2] Right: pressure/temperature phase diagram of MgO showing first principles results, direct measured temperatures,[4] and experimental data from Z in pressure with the temperature inferred from calculations at the same shock pressure. The behavior as the Hugoniot crosses the phase boundaries is particularly important. Notice the significant co-existance between solid B2 and liquid during melting. By combining first-principles theory with experimental data we learn more about the behavior of materials at extreme conditions.

- Adequate sample sizes for extreme pressure experiments to investigate the effects of microstructure on material response and/or understand the response of bulk heterogeneous materials
- Pressures and/or time scales that cannot be attained on gas guns or in laser-driven compression experiments
- Capability for hazardous materials like beryllium, uranium, and plutonium

An example of recent research combining theory and experimental efforts, where the joint analysis led to important insights, is the investigation of multiple phase transitions in MgO.[2] Figure 1 shows the phase diagram of MgO, calculated from DFT and quantum Monte Carlo (QMC) combined with data from Z. QMC is an advanced method for calculating properties of materials based on the electronic structure of the material. The dramatic increase in computational capabilities has made QMC a viable method for calculating properties of real materials not only model systems. Over the last few years, we did a comprehensive study of the high-pressure properties of MgO using experiments, DFT, and QMC methods. The work spanned a wide pressure range covering the B1 solid phase, the B2 solid phase, and the liquid from 270 to 1,200 GPa. The high-precision data constrain the Hugoniot at multi-Mbar pressures, and the DFT and QMC results further elucidate the phase boundaries along the Hugoniot (the Hugoniot is the thermodynamic location of end states in pressure and density for successively stronger shocks from a given initial state). We find that the B1-B2 solid-solid transition is sharp and driven by volume collapse while the B2-melt transition is gradual and is characterized by a large entropy change. The results show that DFT and QMC methods can also be used to study complex materials–a conclusion of importance to planetary science because the results place a lower bound on impact velocities for complete melting in MgO-dominated asteroids and planetesimals (minute planets formed early in the evolution of a planetary system). This constraint on the data and phase diagram provides a solid basis for the development of equations of state for the complex minerals relevant for planetary collision and evolution studies.

While the importance of having high-fidelity EOS models is well established, multi-physics simulations of complex dynamic materials and HED experiments on Z, NIF, and OMEGA also depend on accurate models for transport: electrical and thermal conductivity, opacity, and viscosity. In fact, the EOS and transport properties are closely linked; they change together at phase transitions. The transient character of dynamic experiments makes the measurement of these properties particularly challenging and first-principles based theory play a key role in improving our models and understanding of physical processes. Figure 2 shows the electrical conductivity of beryllium superimposed on a phase-diagram. A focus of our work going forward is to develop EOS and transport models jointly. The next generation EOS and transport model will for example share the underlying phase boundaries and be built from the same theoretical foundation – and allow for systematic uncertainty quantification of multi-physics simulations.

Even after 60 years of important discoveries, the field of EOS and other material models remains vibrant with new experimental and theoretical methods being invented and applied. These advances support stockpile, basic science and industrial applications. The precision of today's experiments in reaching new conditions, and diagnosing the states better than ever before, combined with the fidelity of state-of-the-art computer simulations promises new discoveries in the important field of EOSs.

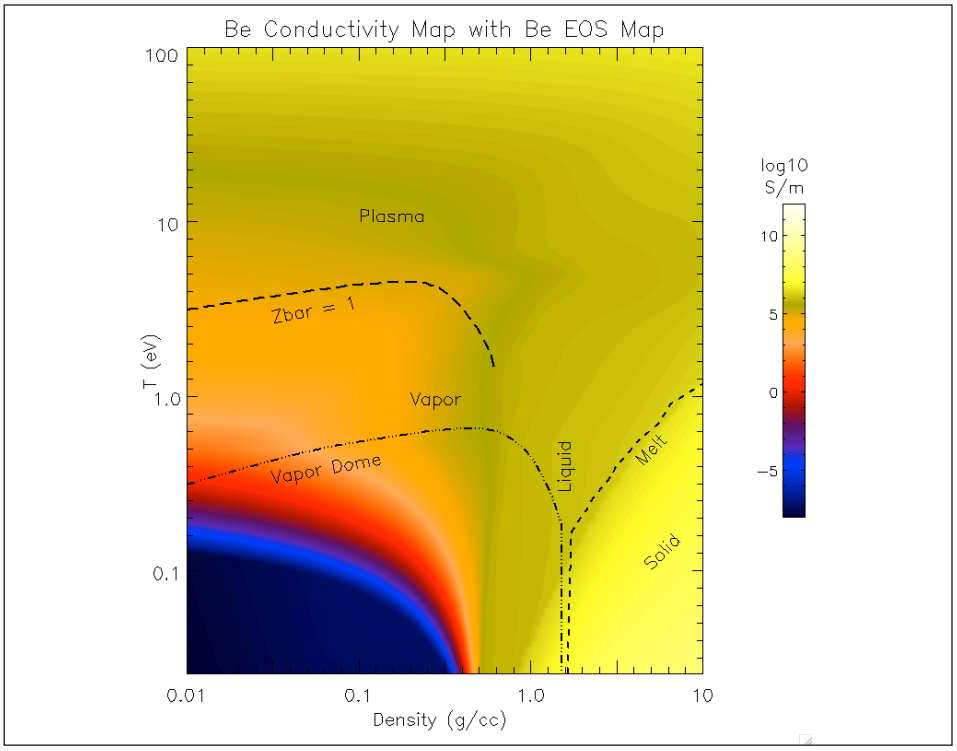


Figure 2. Electrical conductivity for beryllium in a color overlay of the equation of state. Changes in phase are correlated with changes in conductivity. The two properties are intrinsically linked and the behavior in simulations is therefore closely coupled.

References

[1]Impactful Times–Memories of 60 Years of Shock Wave Research at Sandia National Laboratories, J.R. Asay, L.C. Chhabildas, R.J. Lawrence, and M.A. Sweeney, Shock Wave and High Pressure Phenomena, Springer 10.1007/978-3-319-33347-2.

[2]Seth Root, Luke Shulenburger, Raymond W. Lemke, Daniel H. Dolan, Thomas R. Mattsson, and Michael P. Desjarlais, Phys. Rev. Lett. 115, 198501 (2015).

[3]LASL Shock Hugoniot Data (LANL Handbook), Ed. S.P. Marsh, University of California Press, Berkeley (1980).

[4]B. Svendsen and T.J. Ahrens, Geophys. J. R. Astron. Soc. 91, 667 (1987). ●

Assessing Extreme Equation of State Conditions on the National Ignition Facility

by D.E. Fratanduono, D. Swift, A.E. Lazicki, and H.D. Whitley on behalf of the National Ignition Facility High Energy Density Equation of State Team (Lawrence Livermore National Laboratory)

Introduction

An understanding of the structure and composition of the gas giants (e.g., Jupiter and Saturn), terrestrial exoplanets, and stars is intimately tied to our understanding of the equation of state (EOS) of the elements and compounds that make up these astronomical bodies. An EOS is a mathematical relationship that describes the state of matter (i.e., gas, liquid, or solid) using the material properties of temperature, volume, pressure, and internal energy. The EOS characterizes the properties of a state of matter under a given set of physical conditions. In addition to constraining the composition of planets and stars, accurate EOS models are important for describing astrophysical impacts (and for designing ways to steer asteroids for planetary defense) and for modeling the performance and predicting the effects of projectiles and explosives.

Radiation hydrodynamic simulations for high energy density physics applications require EOS models for materials across a wide range of temperatures and densities, starting from the room-temperature solid phase and extending to densities that are present at the cores of planets and temperatures that are commensurate with the surface of the sun. EOS models for materials within the high energy density regime (pressures > 1 Mbar) were historically based solely on theoretical calculations and largely unconstrained by experimental data in the relevant regime. With the development of the National Ignition Facility (NIF), EOS conditions previously inaccessible within the laboratory are now being probed by the shock gigabar (Gbar) platform and the isentropic ramp compression (Ramp) platform (see Figure 1). The Gbar platform extends the range of accessible shock compression data to pressures in excess of ~1 Gbar (1 Gbar = 1,000 Mbar, ~ 1 billion atmospheres), making it possible to probe plasma states at unprecedented temperatures. In contrast, the ramp compression platform accesses solid states along the material isentrope to unprecedented densities. These platforms are providing critical data to ensure that EOS theory and models accurately capture the behavior of materials in previously unexplored regimes.

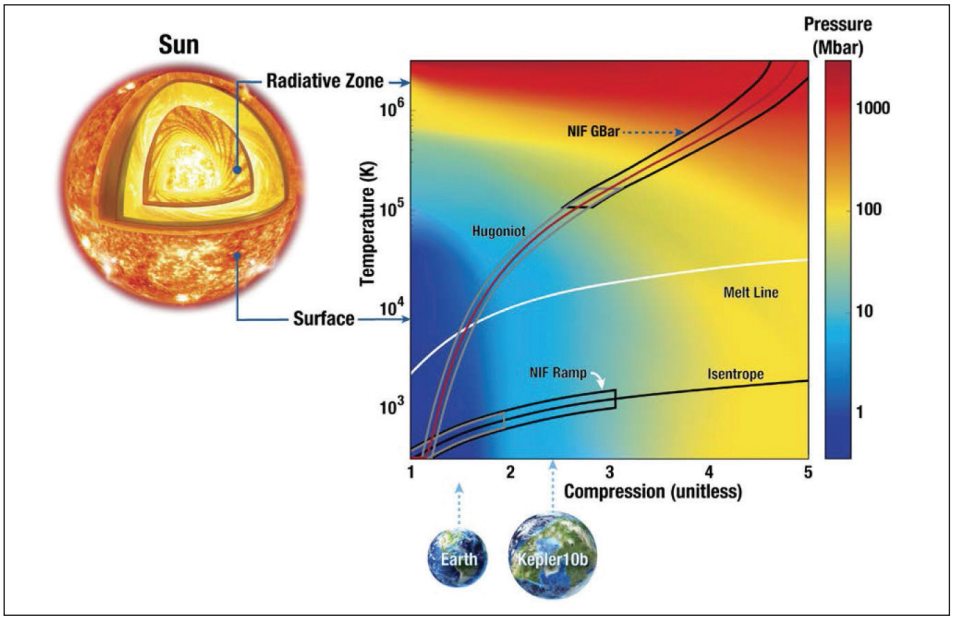


Figure 1. The equation of state of iron. The NIF GBar platform accesses unparalleled conditions (black box encompassing red line) along the Hugoniot (red line) order of magnitude higher in pressure than other platforms (grey box encompassing red line) and well into the plasma phase. These conditions are similar to the interiors of stars. In contrast, the NIF ramp platform (black box encompassing black line) probes high-compression at low temperatures where the materials remain solid along the principal isentrope (black line). The NIF ramp compression platform reaches pressure states higher than any other platform (grey box encompassing black line). These conditions are applicable to the interiors of giant terrestrial exoplanets.

Accessing Extreme States of Hot Matter on the NIF: The Gbar Shock Platform

The Gbar platform is designed to measure the EOS of plasmas at pressures and temperatures up to several hundred million atmospheres and several million degrees, which are in the regime of brown dwarf stars and the cores of massive exoplanets and are particularly relevant for modeling fusion-energy experiments. At these conditions, excitation of tightly bound electrons is predicted to have a dramatic effect on plasma compressibility. The detailed structure of the Hugoniot (the series of shock states) in this atomic ionization regime is closely tied to the precise quantum-mechanical state of the material. Consequently, EOS models have historically differed significantly in this regime, depending on the approximations used to describe this detailed physics, and there has been a complete lack of experimental data to test these approximations.

Matter can be brought to these high-pressure and -temperature states, and its properties measured, by subjecting it to a shock wave. The new NIF Gbar platform successfully uses laser ablation to achieve Gbar pressures by launching a shock wave into a solid sphere. As the shock wave converges toward the center of the sphere, there is a massive pressure amplification. EOS states in the sample are measured using x-ray radiography by imaging a cross-section of the sphere onto a streak camera which continuously records the shock waves converging with time until shock coalescence (see Figure 2). This allows a range of states to be sampled on a single experiment.[1,2] The radiograph yields both shock velocity and density so the measurement is absolute; it does not rely on knowledge of any other material EOS. The radiography method currently limits the measurement to materials of

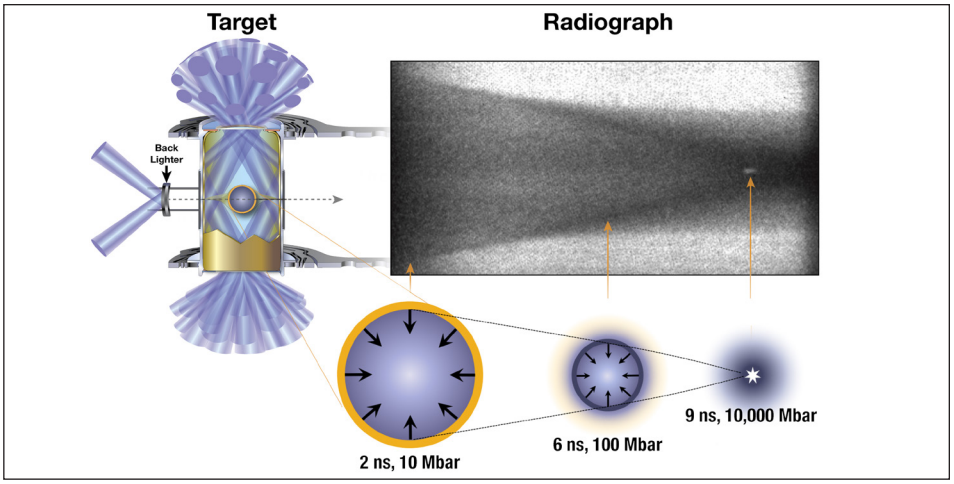


Figure 2. The NIF Gbar shock platform uses a hohlraum to drive a solid sphere and a backlighter to image the imploding sphere onto a streak camera through narrow slits in the hohlraum. The streak camera radiograph records the position of the converging shock front from the time when it passes from an ablator layer into the sphere until coalescence, where x-ray self-emission from the hot spot is evident.

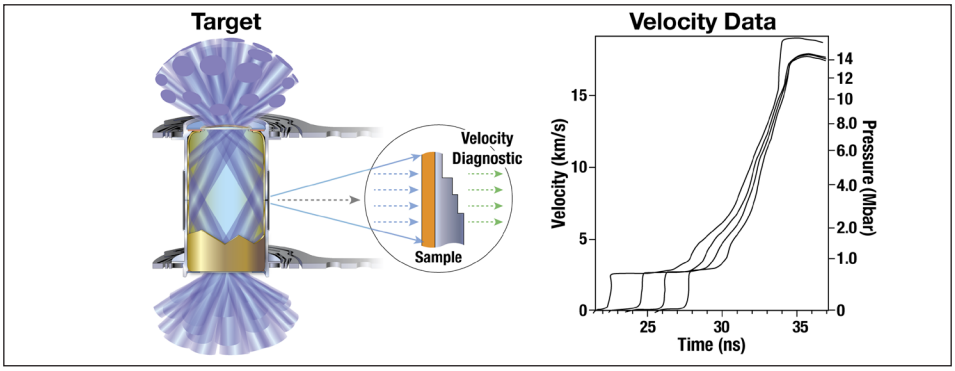


Figure 3. The NIF Ramp platform utilizes a hohlraum target with a stepped sample mounted on the side. In these experiments, a velocity diagnostic measures compression wave steepening as a function of material thickness. By measuring the wave steepening for different sample thicknesses, we determine the material sound speed and ultimately the sample pressure and density response.

low atomic number. Experiments on CH, diamond, and boron have probed EOS to 600-800 Mbar pressures, nearly an order of magnitude higher than other facilities or experimental platforms are capable of achieving.

Accessing High-Density and Low-Temperate Matter on the NIF: The NIF Ramp Platform

The NIF Ramp platform is designed to measure the EOS of solids to ultra-high densities using isentropic compression.[3] A schematic of the platform is shown in Figure 3. Isentropic compression can be understood as the material response to an infinite number of weak shock waves. Using this technique, the entropy increase in the sample is minimized such that high-pressure, low-temperature (below melting) conditions are accessed.

With the high laser power available at NIF, these experiments are reaching compression states predicted in the interiors of massive extrasolar planets—a regime where little or no experimental data existed previously. In the absence of data, EOS models are currently benchmarked to theoretical calculations, which are increasingly predicting that material properties can be unexpectedly complex at compression states where the most tightly bound electrons on the atoms begin to interact. Experimental data at these extremes is therefore of great interest for developing more accurate material models.

The isentropic compression technique requires meticulously careful sample preparation and tuning of the compression rate to ensure that strong shock waves do not form within the sample. Due to its exquisite pulse-shaping abilities and high laser power, the NIF is able to access compression states nearly an order of magnitude greater than other platforms can achieve. This measurement also relies on extremely accurate target fabrication. The small scale of the targets requires that the flatness of the part be better than 50 nm. If the target were scaled to the size of a football field, this required flatness would be similar to demanding that the height of all the blades of grass in the field differ by less than the width of a pencil lead. Through significant advancements in target manufacturing, this incredible accuracy is now routinely achieved. Thanks to this combination of excellent NIF laser and diagnostic capabilities and target fabrication, we have constrained the EOS of a variety of materials to pressure conditions 2-5 times greater than that achieved at other facilities. We are now able to measure unparalleled high-pressure and low-temperature EOS conditions on the NIF, enabling us to validate material models in regions of phase space that were previously inaccessible.

Conclusions

With the Gbar and Ramp platforms developed on the NIF, we are collecting experimental data far into unexplored regimes of density and pressure which will provide a critical constraint for EOS models in regimes of direct relevance for understanding extrasolar terrestrial planets and stars. When applied to materials of interest for stockpile stewardship, these techniques will allow us to address long-standing programmatic questions and meet new challenges associated with our national security mission. The techniques also allow us to answer important basic science questions and improve our overall understanding of materials, leading to the development of models with better predictive capability.

References

[1]A.L. Kritcher et al., High Energy Density Phys. 10, 27–34 (2014).

[2]J. Nilsen et al., High Energy Density Phys. 21, 20-26 (2016).

[3]R.F. Smith et al., Nature 511(7509): 330-333 (2014). ●

Equation of State and Shock-Driven Decomposition of 'Soft' Materials *by Joshua D. Coe and Dana Dattelbaum (Los Alamos National Laboratory)*

Equation of state (EOS) efforts at National Nuclear Security Administration (NNSA) national laboratories tend to focus heavily on metals, and rightly so given their obvious primacy in nuclear weapons. Our focus here, however, is on the EOS of 'soft' matter such as polymers and their derived foams, which present a number of challenges distinct from those of other material classes. This brief description will cover only one aspect of polymer EOS modeling: treatment of shock-driven decomposition. These interesting (and sometimes neglected) materials exhibit a number of other challenging features—glass transitions, complex thermal behavior, response that is both viscous and elastic—each warranting additional discussions of their own.

Polymers are pervasive in weapon systems, filling a wide variety of roles including structural support, maintenance of engineering tolerances, and impact mitigation. Some stockpile-relevant examples are provided in Figure 1a, where moving from left to right are images of epoxy, polysulfone (PS), high-density polyethylene (HDPE), and polyurethane (PU). All of these materials decompose chemically and irreversibly upon application of sufficient heat and/or compression,[1] a feature shared with high explosives (HE). All are also polymeric in character, built up from long chain molecules or precursors of the type shown below each photograph.

Because many interesting scenarios in stockpile stewardship involve shock waves, one of the most valuable pieces of information on a material is its shock Hugoniot. The Hugoniot is a collection of states obtained by varying the strength of a single applied shock, and is readily calculated from an EOS. Each state on the Hugoniot represents a discrete experiment initiated from a common origin; if the origin is standard temperature and pressure, the locus is known as the *principal* Hugoniot. Planar shocks with precisely defined input conditions often are generated using smooth bore light gas guns of the type shown in Figure 1b. Gaseous helium compressed up to 15,000 pounds per square inch in the breech (just out of view in the foreground)

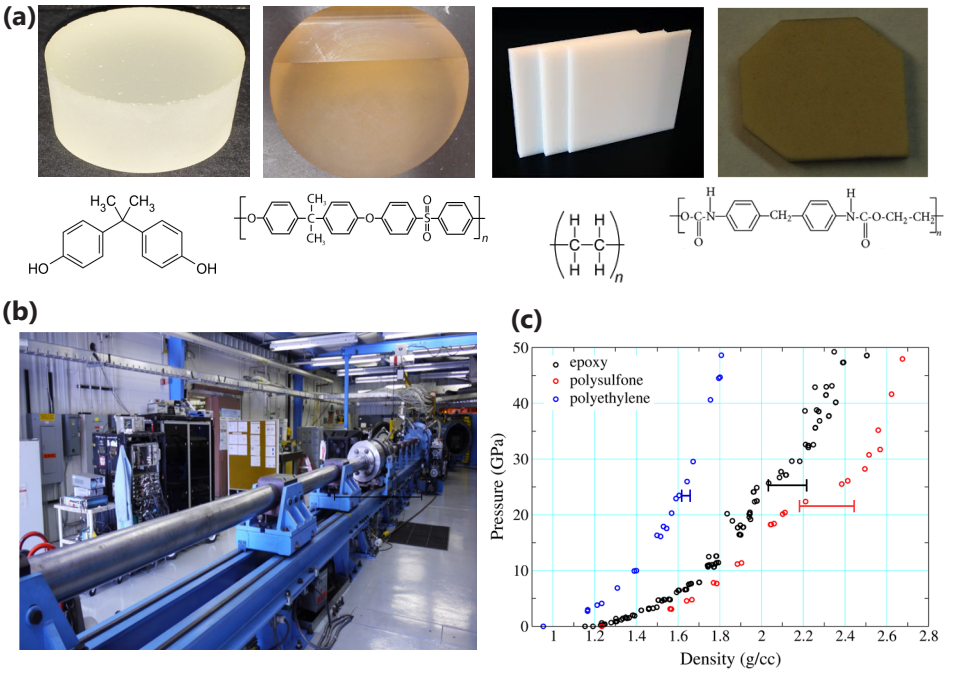


Figure 1. (a) Photographs of some polymers relevant to the stockpile: (left to right) epoxy, polysulfone, polyethylene, and polyurethane. Chemical structure of the repeat unit or (in the case of epoxy) a precursor of the bulk samples is shown below each photograph. (b) LANL's two-stage light gas gun, used to collect shock data of the sort depicted in part c and Figure 2. (c) Principal Hugoniot data for three of the polymers depicted in parts a and b. The interval bars attached to each dataset indicate approximate degree of volume collapse due to shock-driven chemical decomposition.

is used to launch projectiles at velocities up to 3.6 km/s into the target chamber visible at the back.

Data derived from gun experiments frequently are presented as in Figure 1c, illustrating the principal Hugoniots of HDPE, epoxy, and PS in the pressure-density plane. Pressures are expressed in gigapascal (1 GPa ≈ 10,000 atmospheres), densities in gram/cubic centimeter (the density of water at standard temperature and pressure is very close to 1 g/cc). Common to all three curves—although quite muted in HDPE—is an abrupt change in density (sometimes referred to as the "cusp"[2-3]) at around 25 GPa in pressure, indicated approximately by the interval bars included with each data set. This feature is caused by rapid decomposition of the initial materials shown in 1a into complex mixtures of molecular and solid constituents such as CO_2, H_2O, and carbon particles of nanometer dimension.[4] The collapse in volumes shown in 1c can be correlated (at least roughly) with the molecular structures of 1a. Chemically simple polymer chains such as those of HDPE pack more efficiently into a solid matrix than do complex chains such as those of PS. When the long-range order of the matrix is destroyed by shock-driven chemical reaction, the excess volume due to inefficient packing is first "freed up," then rapidly "squeezed out." In the case of HDPE, on the other hand, there is very little volume to free, and so the difference in density above and below the cusp is so small as to be barely discernible. We know from recovery experiments, however, that it does irreversibly decompose.[5]

The manner in which shock-driven decomposition of polymers is incorporated into their EOS has hydrodynamic implications analogous to those of shock-driven transitions in other material types. Panel a of Figure 2 depicts the shock Hugoniot of epoxy drawn from a variety of sources, as well as three distinct approaches to modeling the transition. The red line (no-reaction) simply ignores it, passing

smoothly from the bulk polymer to the reaction products without any change in the underlying description. The blue line attempts to capture both reactants and products, but as a single material—again, with no change in the underlying description upon "reaction". The structure evident in the shock data is simply built into the EOS without regard to its physical origin. The black line represents reaction products only, and is based on thermochemical modeling specifically tailored to complex mixtures in chemical equilibrium. The curve extends down only so far in pressure because—as a material distinct from what it was initially—its Hugoniot doesn't exist below some threshold for shock-driven decomposition of the original polymer. Because this EOS applies only to material shocked in excess of the cusp, its use in a hydrodynamic simulation must be paired with that of a separate EOS for material below it (the reactants) and some mechanism for transitioning from one to the other where appropriate. This fact is represented in the figure by the black arrow drawn from the reactant-product EOS below the cusp to the products-only EOS above it, and is standard practice in HE detonation modeling.

Now consider a scenario as in Figure 2b, where the polymer is driven above the threshold for reaction such that the final shocked state sits at ~30 GPa (a reasonable value for shocks imparted by HE detonation waves) on its product Hugoniot, as indicated by the large dots. Assuming inviscid, compressible flow, the material will then release along isentropes given by the dashed lines. For reactants and products modeled as distinct materials (black lines), this isentrope will be that of the products only and is therefore smooth. When reactants and products are treated as the same material, the structure built into the Hugoniot is mirrored in the isentrope; the material "back-reacts" upon release and what was originally a single wave will split into two. This is equivalent to assuming that the transition is completely reversible, making the reaction threshold perfectly analogous to phase transition lines in an equilibrium phase diagram. This may be correct for melting in a metal, but is highly suspect when covalent chemical bonds have been rearranged. One would never think of adopting such an approach in the case of HE detonation—despite strong similarities in the underlying chemistry—simply because the whole event is so undeniably irreversible.

Polymers of fixed chemical composition but widely differing density can be generated through the process of foaming, whereby the density is decreased through introduction of voids. Foaming has profound effects on the form of the principal Hugoniot, as reflected in the results for PU shown in Figure 2c. As the initial density is reduced (i.e., the initial porosity increased), the Hugoniot flattens at very low pressure but then steepens rapidly. Within the scatter of the data (error bars have been suppressed for clarity, and are highly suspect anyway), the Hugoniot becomes vertical by the time porosity has been increased to 50 percent, and further increases actually yield shock densities that decrease with increasing shock pressure. This highly counterintuitive feature, known as "anomalous compression," is the result of tremendous heating upon removal of free volume. This feature has a natural corollary in that the high temperatures generated in porous materials drive down the threshold for reaction, as shown in Figure 2d in terms of both pressure (left ordinate) and particle (or mass) velocity (right ordinate); 75 percent porous PU decomposes at a shock pressure more than an order of magnitude lower than at full density.[6]

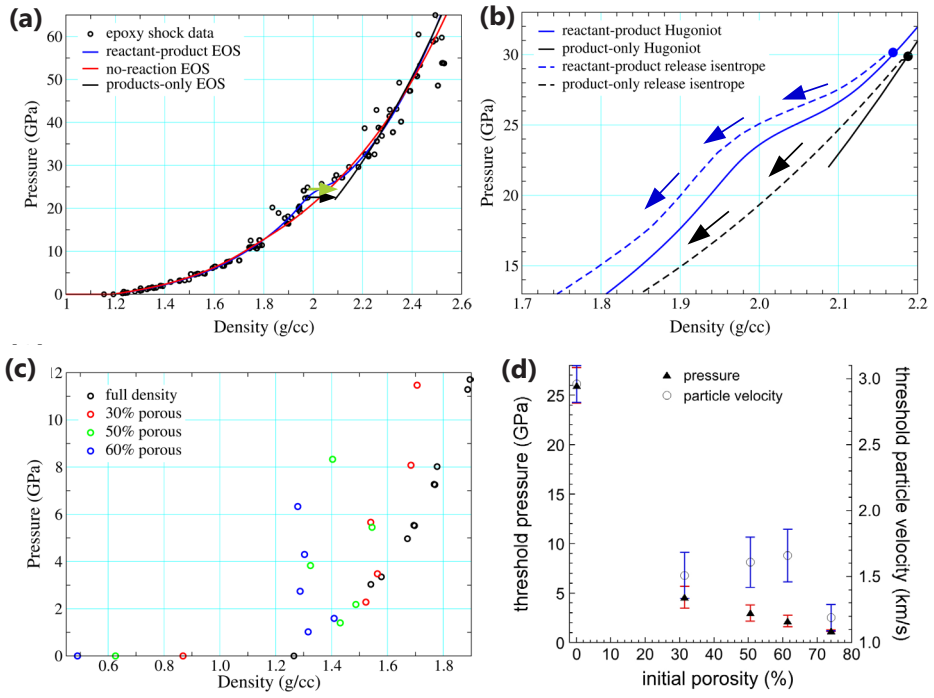


Figure 2. (a) Principal Hugoniot data for epoxy (points), and three different approaches to EOS modeling (see text for a full description). (b) Hugoniots predicted by two of the modeling approaches in part A, along with release isentropes initiated from the large dots at ~30 GPa. Arrows indicate the path of evolution away from the Hugoniot. (c) Principal Hugoniots of polyurethane at four different initial densities. Note the dramatic steepening of the curve as porosity increases. (d) Thresholds for shock-driven decomposition of polyurethane, as extracted from the curves shown in part c. Note the dramatic reduction in threshold pressure and mass velocity as initial porosity increases.

References

[1]W. Carter and S. Marsh, "Hugoniot Equation of State for Polymers," LA-13006-MS, Los Alamos National Laboratory (1995).

[2]D.M. Dattelbaum and S.A. Sheffield, "Shock-Induced Chemical Reactions in Simple Organic Molecules," AIP Conf. Proc. 1426, 627-632 (2012).

[3]D.M. Dattelbaum et al., "Shockwave Response of Two Carbon Fiber-Polymer Composites to 50 GPa," J. Appl. Phys. 116, 194308 (2014).

[4]F.H. Ree, "Systematics of High-Pressure and High-Temperature Behavior of Hydrocarbons," J. Chem. Phys. 70, 974-983 (1979).

[5]C.E. Morris et al., "Shock Induced Dissociation of Polyethylene," Shock Compression of Condensed Matter–1989, 687-690 (1990).

[6]D.M. Dattelbaum et al., "Reactive, Anomalous Compression in Shocked Polyurethane Foams," J. Appl. Phys. 115, 17490 (2014). ●

Dynamic Materials Research at the Nevada National Security Site

by William D. Turley, Robert S. Hixson, David L. Esquibel, Thomas E. Graves, Trenton Otteson, Drew D. Martinson, Robert J. Scharff, Sarah A. Thomas, Lynn R. Veeser, Michael Grover, Brandon M. La Lone, Gerald D. Stevens, Ruben J. Valencia; Jeffery G. Cates, Edward P. Daykin, Michelle C. Hawkins, Russell A. Howe, and Melissa K. Matthes (National Security Technologies, LLC, now Mission Support and Test Services, LLC)

Understanding how materials respond under high pressure, temperature, stress, and strain conditions is a critical element of the Stockpile Stewardship Program (SSP). In collaboration with national laboratory staff, Nevada National Security Site (NNSS) develops new diagnostic techniques and instruments to measure such properties. In addition, NNSS researchers develop new experimental techniques and work with their laboratory partners to conduct focused science experiments to confirm that these methods work. Within the SSP and the National Nuclear Security Administration's (NNSA's) Site-Directed Research and Development (SDRD) program, NNSS scientists are constantly exploring new diagnostics and dynamic measurement techniques for improving the accuracy of future and existing datasets.

NNSA uses such fundamental data to constrain and develop new physics models for multiphase equations of state (EOSs), better understand constitutive properties (such as compressive and tensile strength), and investigate other aspects of how materials respond under extreme conditions.

To enable new experimental methods and diagnostics tools, there are a number of cost-effective shock dynamic materials research facilities. These facilities also support researchers across the nuclear weapons enterprise and serve as training platforms for early-career science, technology, engineering, and math (STEM) workers in dynamic materials science and general experimental physics.

The Joint Actinide Shock Physics Experimental Research (JASPER) two-stage gas gun facility located at the Nevada National Security Site is currently being used for both actinide (i.e., plutonium [Pu]) and non-actinide material studies (see Figure 1). The nuclear weapons enterprise is most interested in the plutonium data to test and validate theories of hazardous material (Pu) response for input to EOS tables. Although the JASPER gas gun has primarily been used to map single-shock (Hugoniot) data, scientists have begun using it to explore regions off of the Hugoniot at different ranges of pressure, temperature, and time scales. Utilizing the data captured at JASPER will help scientists from the nuclear enterprise to assess and certify the nuclear stockpile and life extension programs.

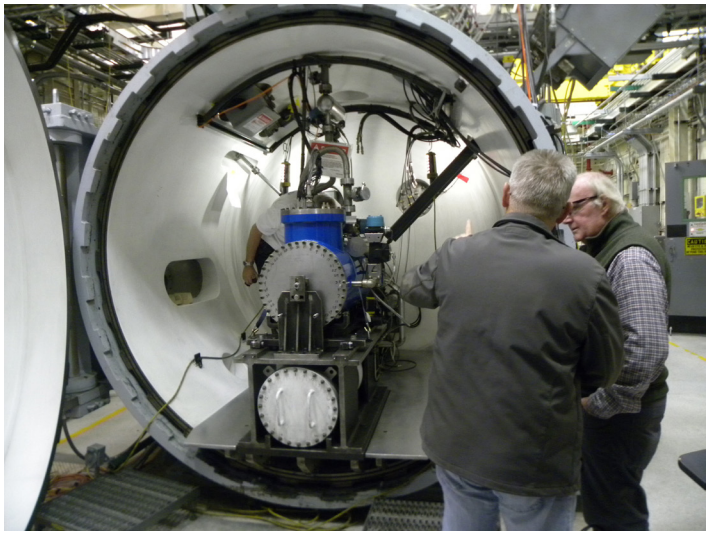


Figure 1. Neil Holmes (LLNL Chief Scientist) and Sam Williams (LLNL Experiment Operations Manager) discuss the status of an upcoming JASPER experiment while diagnostic and support staff finalize activities to secure the primary diagnostics and the Secondary Confinement Chamber.

Figure 2. NNSS's North Las Vegas launcher facility used for diagnostics development, staff training, and selected scientific studies. Left to right: Jeff Cates, Cameron Hawkins, Drew Martinson, Melissa Matthes, Russel Howe, Tom Graves, Rob Hixson, Sarah Thomas, and Ed Daykin.

Other NNSS facilities enable nonhazardous material studies and diagnostic development experiments. These include the single-stage gas gun in North Las Vegas, Nevada (see Figure 2) and the shock-physics diagnostic development research facility in Santa Barbara, California, which has a variety of platforms, including a single-stage gas gun, propellant-driven guns, and a small-scale explosive facility. These dynamic compression facilities are heavily used by both NNSS and national laboratory scientists for material response experiments.

Advanced Diagnostics

As the national laboratories request more complex and sophisticated measurements with higher and higher fidelity, NNSS personnel find themselves at the leading edge of continuous improvement in advanced diagnostics. Partnered with national laboratories, the NNSS actively fields and develops a diverse set of diagnostics systems that support the national objectives in dynamic materials studies. These activities support the transition of new diagnostic methods to the following platforms: JASPER, the Dual Axis Radiographic Hydrodynamic Test facility at Los Alamos National Laboratory (LANL), National Ignition Facility at Lawrence Livermore National Laboratory (LLNL), Dynamic Integrated Compression Experiment at

Sandia National Laboratories (Sandia), Dynamic Compression Sector at Argonne National Laboratory, Stanford Synchrotron Radiation Lightsource, and other facilities to help national laboratories meet their goals of improved characterization of stockpile-critical materials.

Phase change, temperature, and ejecta measurements are a high priority in shock physics-related science. Optical velocimetry, reflectometry, emissivity, x-ray diffraction, soft x-ray radiography, and pyrometry are all used in the study of these phenomena. The diagnostics developed at NNSS are expected to be used on dynamic compression experiments and contribute to a better understanding of how material properties react at different pressures, temperatures, and time scales.

Refining Physics Codes

At the most fundamental level, shock wave speed (U_s) and internal material velocity (u_p) measurements (together with conservation equations used within the Rankine-Hugoniot relations) are used to determine the energy, density, and pressure in dynamic compression experiments. By systematically determining these two velocity values over a range of stress conditions, one can construct a material EOS. Subtle changes in a shocked material's velocity, temperature, or optical reflectance can indicate the location of high-pressure phase boundaries (i.e., solid, liquid, and gas phases). Fundamental properties are used to construct a material EOS for each phase, and then phases are connected at phase boundaries to construct a multiphase EOS. NNSS has teamed with the laboratories to develop velocimetric techniques and phase-diagnostics for applications in one-dimensional or even more complex hydrodynamic experiments. In addition to EOS measurements, these techniques also deliver crucial insight into properties such as the compressive and tensile strength of materials.

The results of a recent study measuring shock behavior of iron and various steel alloys are shown in Figure 3. Note that the wave speeds of the different alloys are the same within the uncertainties, but as uncertainties are driven down, systematic differences may be revealed. Understanding such subtle differences can help test existing dynamic materials models and support the development and refinement of new physics codes.

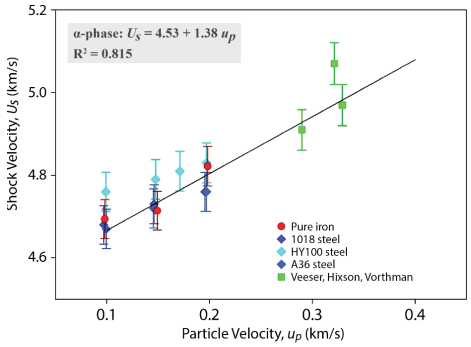


Figure 3. Shock velocity versus particle velocity shock Hugoniot of α-phase iron and steel alloy samples.

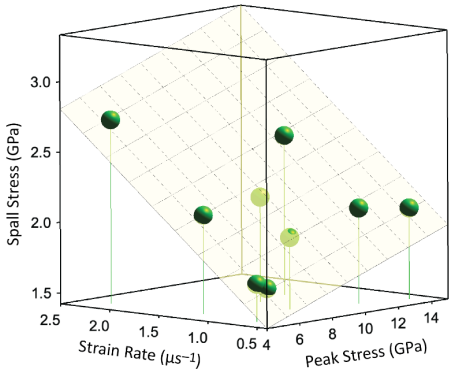


Figure 4. Spall stress as a function of peak stress and calculated tensile strain-rate for single-crystal [100] copper.

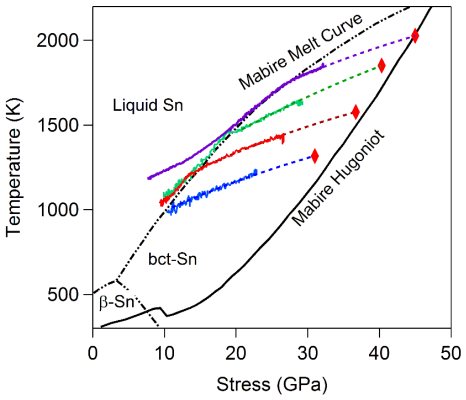


Figure 5. Calculated stress versus temperature phase diagram for tin. Dynamic temperature measurements, overlaid in color, show good agreement with a model by Mabire and Hereil.[3]

Polycrystalline and Single-Crystal Copper Studies

NNSS and LANL scientists recently performed dynamic experiments (funded by SDRD) that measured the tensile response of polycrystalline and single-crystal copper for three orientations: [100], [110], and [111].[1] This work was motivated by a desire to better understand how the dynamic spall damage process works in single-crystal copper when there are no grain boundaries to aid the nucleation of the damage process and to determine the role of crystalline anisotropy. The team demonstrated that the dynamic spall strength of copper depends upon crystal orientation, tensile strain rate, and peak compressive stress prior to the onset of tension. Results for the [100] orientation are shown in Figure 4.

Dynamic Temperature Measurements

Temperature (along with pressure, volume, and internal energy) is needed to construct a thermodynamically complete EOS. Although methods to measure stress and densities in dynamic experiments are developed, temperature is much harder to measure, and the shock physics community has struggled for decades to make credible dynamic temperature measurements. NNSS researchers, working closely with staff from LANL, LLNL, and Sandia, have developed optical pyrometric methods for obtaining dynamic temperature measurements. Using multichannel radiance and reflectance techniques, recent experiments at NNSS are returning temperature measurements with a few-percent uncertainty. Figure 5 shows results from a collaboration with Dr. Paul Asimow's shock physics group at Caltech.[2] The results of four dynamic experiments (shown in color) are overlaid on a calculated phase diagram for tin. The curves, which are constant-entropy release paths, move from right to left in the diagram and then turn downward upon intersection with the melt boundary. The inferred melt boundary is remarkably well matched to a calculated melt curve.[3]

Summary

Researchers at NNSS have strong collaborations with the national laboratories in their pursuit of a better understanding of dynamic materials properties, especially actinide materials. Data obtained from the latest dynamic materials experiments and diagnostic developments have been used to further understanding of the fundamental

properties used to construct EOSs for materials important for the stockpile (e.g., multiphase actinides); deliver crucial insight into properties such as the compressive and tensile strength of materials; improve the Nation's understanding of materials at extreme temperatures and pressures; and refine physics codes by testing dynamic materials models. As simulations become a more prominent aspect of understanding and managing the Nation's stockpile, dynamic materials research plays an ever increasingly important role in NNSA's stockpile stewardship mission.

References

[1]W.D. Turley et al., "Spall Response of Single-Crystal Copper," to be published 2017.

[2]B.M. La Lone et al., "High-Pressure Melt Curve of a Shock-Compressed Metal Measured Using Pyrometry and Reflectance Techniques," to be published 2017.

[3]C. Mabire and P.L. Hereil, "Shock Induced Polymorphic Transition and Melting of Tin," AIP Conference Proceedings 505, 93–96 (2000). ●

Laboratory Directed Research and Development Innovations for Stockpile Stewardship

by E.C. Bumstead, B.S. Stoltzfus and W.A. Stygar, and C.T. Seagle (Sandia National Laboratories)

Laboratory Directed Research and Development (LDRD) projects perform creative, innovative, leading edge research and development that is focused on challenging science and engineering problems that will have significant impact on the national laboratorys' national security missions. LDRD projects are generally too high risk for NNSA programs or other sponsored work, but when successful may lead to advancements in science or engineering, innovative technological solutions to mission problems, intellectual property, new capabilities, strategic partnerships, and new sponsored work.

A fundamental understanding of the science of nuclear weapons is imperative for the future of the stockpile. Achieving this future without the ability to conduct underground testing requires radically new capabilities in the laboratory and simulation environments.

An ambitious LDRD project by Brian Stoltzfus, William Stygar, and their colleagues developed, as discussed below, Thor's conceptual design for pulsed power. The initial building blocks are important foundational technology for the next-generation pulsed power accelerator. Chris Seagle's LDRD project faced challenges because of the difficulty in achieving required static pressures; results will produce data off-the-principal Hugoniot (the Hugoniot is the thermodynamic location of end states in pressure and density for successively stronger shocks from a given initial state). The initial pressure control capability for dynamic experiments can be applied to a wide variety of materials. These bold projects help push the envelope in delivering equations of state that provide real-world data used to determine phase transitions accurately under extreme conditions. Both LDRD-developed capabilities now have been absorbed into programs programs illustrating how these anticipatory investments enable success in our stockpile stewardship mission.

New Flexibility in Dynamic Materials Research Via Pulsed Power

by B.S. Stoltzfus and W.A. Stygar (Sandia National Laboratories)

Since the 1960s, Sandia National Laboratories' pulsed power devices have helped to assure the performance of every nuclear system in the stockpile. Sandia's next-generation technology and know-how continue to maintain Sandia's role as the world's leader in pulsed-power accelerator physics.

One recently completed Laboratory Directed Research and Development (LDRD) project created the foundation of next-generation pulsed-power technology. In pulsed power and dynamic materials physics experiments, electric currents are used to generate magnetic fields. These are used to drive acoustic pressure waves for dynamic materials physics experiments. This technique is unique in its ability to conduct tailored shockless experiments at high pressures, and other material-physics experiments driven by a wide variety of pressure-loading time histories.

Thor 48 is a pulsed-power accelerator consisting of 48 highly reliable, high-power-density "bricks." Bricks are small electrical storage units. The brick switches are triggered to launch 100-nanosecond pulses into long coaxial cables. The cables provide transit time isolation between the bricks, which allows Thor to act as an arbitrary waveform generator at megampere currents. The pulses are summed together at the load to generate a tailored magnetic-acoustic pressure wave.

Thor's design concept offers these advantages:

- Precision control and pulse shape tailoring
- Single-stage pulse compression
- Low-voltage switching
- Efficient delivery of energy from capacitors to the physics load
- Less damage from faults in the system

An overhead view of Thor 48 with six brick towers, six cable towers, and the load region. (Photo by Brian Stoltzfus)

- Modular design operation from Thor 8 to Thor 72
- Low experimental cost (~$1 k per shot) and high shot rate (~5 per day)
- Reconfigurable load/center section
- Engineered safety
- Economies of scale

Recent experimental results on Thor include completion of the assembly and commissioning of Thor 48. Experimental shots have been conducted at capacitor-charge voltages of as high as 80 kV. Peak currents reached as high as 2.16 MA; peak pressures as high as 250 kbar. A phase change in zirconium was observed at ~80 kbar. Strength experiments in copper were conducted at pressures as high as 100 kbar. The elastic-plastic transition in a calcium-fluoride crystal was observed as a function of strain rate.

In FY 2018, the Thor Team plans to assemble and commission Thor 64. Thor 64 is expected to reach currents as high as 3 MA, and 600-kbar pressures on an 8-mm-wide panel with a synchronous pulse. Among the materials proposed for testing are cerium, zirconium, gallium, tantalum, titanium, iron, and water.

Thor is an ideal platform for the development of advanced physics load and diagnostic concepts. The results of the LDRD project aided in the design of powerful next-generation pulsed power accelerators that play a key role in several Department of Energy missions. The accelerators continue to support the nuclear stockpile and military needs of the country. Pulsed power machines are used to drive material-physics experiments from which equations of state are derived. Results are used to certify that our nuclear weapons will operate as intended and assure that we understand and predict their behavior without underground testing.

The initial development of Thor's conceptual and physics designs was supported by Sandia's LDRD office. After completion of the LDRD project, further development via engineering design, fabrication, assembly, and commissioning of 1-, 8-, 16-, 24-, and 48-brick versions of Thor was supported by the National Nuclear Security Administration's Research, Development, Test, and Evaluation programs.

References

[1]B.S. Stoltzfus, W.A. Stygar et al., Creating the Foundation of Next-Generation Pulsed-Power-Accelerator Technology, Sandia National Laboratories Report SAND2016-9670 (September 2016).

[2]W.A. Stygar, D.B. Reisman, B.S. Stoltzfus et al., "Conceptual Design of a 10^{13}-W Pulsed-Power Accelerator for Megajoule-Class Dynamic-Material-Physics Experiments," PRAB 19, 070401 (July 2016).

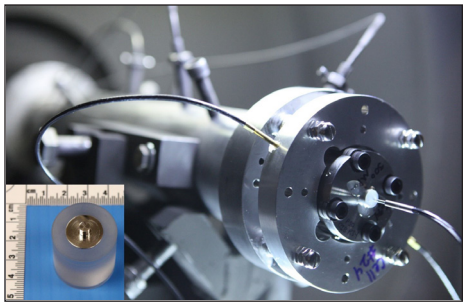
[3]D.B. Reisman, B.S. Stoltzfus, W.A. Stygar et al., "Pulsed Power Accelerator for Material Physics Experiments," PRSTAB 18, 090401 (September 2015).

Compressing Materials Under Extreme Conditions *by C.T. Seagle (Sandia National Laboratories)*

Hydrogen and helium are the most abundant elements in the universe and occur in a variety of extreme environments, both natural and man-made. Mixtures of hydrogen and helium exhibit an extraordinary degree of nonideality; the volume of the mixture is not the sum of individual components. The relative simplicity of these elements would suggest theoretical calculations of the properties of the mixture would be accurate; however, major discrepancies in predicted properties exist between different calculation methodologies and few experimental data are available to benchmark these calculations.

The difficulty associated with achieving controlled extreme states in H2-He mixtures arises from the inability to mix homogeneous fluids of the two at ambient pressure; like water and oil the fluids separate. High static pressures force hydrogen and helium to mix into a single fluid phase. In a recent Sandia National Laboratories' (Sandia) Laboratory Directed Research and Development-funded project, Chris Seagle led a group of scientists investigating the behaviors of materials at very high pressures and densities. A pre-compression capability was developed to study not only H2-He mixtures, but other super-critical fluids as well. The initial pressure of these samples is on the order of about 1000s atmospheres. Launching a shock wave into these mixed, pre-compressed samples results in compression of the mixture to ultrahigh density and moderate temperature. Achievement of the required static pressures on millimeter-scale samples required for gas-gun experiments was the primary challenge because of the high energy density associated with pre-compression. Despite this challenge, pre-compression cells have been successfully developed to support the required pressures on macroscopic samples. The pre-compression capability greatly expands Sandia's initial condition control for shock experiments to probe dynamically compressed states. The pre-compression cell was used in dynamic testing of super-critical helium up to dynamic stresses of 18 GPa, or 178,000 atmospheres. The initial density of these samples were about twice the

Chris Seagle filling a pre-compression target with a super-critical hydrogen-helium mixture at Sandia National Laboratories. (Photo by Nicole Cofer)



A pre-compressed hydrogen-helium target mounted on a two stage light gas gun in preparation for a dynamic compression experiment at Sandia National Laboratories. Inset – platinum impactor mounted on a projectile for the gas gun. The projectile was launched into the target at five kilometers per second. (Photo by Bernardo Farfan)

cryogenic density of liquid helium at 4 kelvin. Current research is focused on the hydrogen-helium mixture and increasing peak dynamic pressures.

In dynamic phenomena, such as explosions, materials are often subjected to complex loading paths. Traditional dynamic experiments designed to constrain material response often are limited to particular regions of phase space that do not always coincide with real-world material trajectories. This research led to a new capability at Sandia to access regions of phase space at higher compression relative to the shock adiabat by pre-compressing samples prior to shock loading. High accuracy equation-of-state (EOS) data in previously inaccessible regions can be obtained, providing a mechanism to benchmark theoretical calculations and EOS models in more relevant regimes.

The Department of Energy (DOE) and National Nuclear Security Administration (NNSA) have sought accurate equation of state and material property information in a wide region of phase space because of the varied loading paths encountered in inertial confinement fusion and weapons applications. The ability to pre-compress samples for dynamic studies opens up regions of phase space not previously accessible–higher compressions may be reached at much lower temperatures. The study of EOSs of mixtures, particularly hydrogen mixtures, benefits a broad range of missions of DOE and NNSA. These studies enhance predictive capability by providing benchmarks for theoretical studies and hydrodynamic simulations.

Reference

[1]Seagle et al., "High Precision Hugoniot Measurements on Statically Pre-Compressed Fluid Helium," J. Appl. Phys. 120, 125902 (2016). ●

Preparing Tomorrow's Stockpile Stewardship Leaders *by Farhat Beg (University of California, San Diego) and J. Tiberius Moran-Lopez (National Nuclear Security Administrator)*

The Office of Research, Development, Test, and Evaluation leads various initiatives supporting early-career professionals in high-energy-density sciences (HEDS) across the country; partners include academic centers, the national laboratories, and scientific institutes. One example accentuating these fruitful partnerships is the HEDS Summer School hosted by the High-Energy-Density Science Association in collaboration with the University of California Center for Frontiers in HEDS. Held on a biennial basis at the University of California San Diego (UCSD), 2017 marks the fourth time the Summer School was sponsored. The purpose of the program is twofold: first, it aims to inspire young scientists to pursue HEDS careers while having specialists deliver the fundamental knowledge and critical skills; second, it also offers early-career professionals the opportunity to network with principal academic and national laboratory scientists as they train to become the next generation of leaders in Stockpile Stewardship.

This year, the Summer School was extended to cover two weeks of lectures, simulation and modeling workshops, open forum discussions, and laboratory tours. The first component of the program consisted of lectures generally held in the mornings; HEDS fundamentals and multidisciplinary topics encompassed: materials science, atomic physics in plasmas and radiation spectroscopy, laser-driven shock hydrodynamics, kinetic vs. fluid effects, and magnetized plasma dynamics and instabilities. Afternoon workshops in computational modeling and experimental design supervised by leaders in academia, industry, and the national laboratories formed the second portion of the program; the workshops primarily focused on radiation hydrodynamics computational modeling, spectroscopic analyses, and particle-in-cell codes. Lastly, tours of UCSD and General Atomics were also scheduled to enhance participants' understanding of diagnostics, ICF target engineering for HED experiments, and future high-power laser technologies.

This year, 114 participants attended the Summer School, including 27 lecturers, 14 postdoctoral scholars, 58 graduate students, and 5 undergraduate students. Support from the National Nuclear Security Administration, the DOE Office of Fusion Energy Sciences, and University of California Lab Fees Program made this event possible. For additional information, please contact Dr. Farhat Beg (fbeg@eng.ucsd.edu) or Dr. J. Tiberius Moran-Lopez (tiberius.moran-lopez@nnsa.doe.gov). ●

Article Suggestions

Have a great idea for an article? Please send your idea to Terri Stone at terri.stone@nnsa.doe.gov.