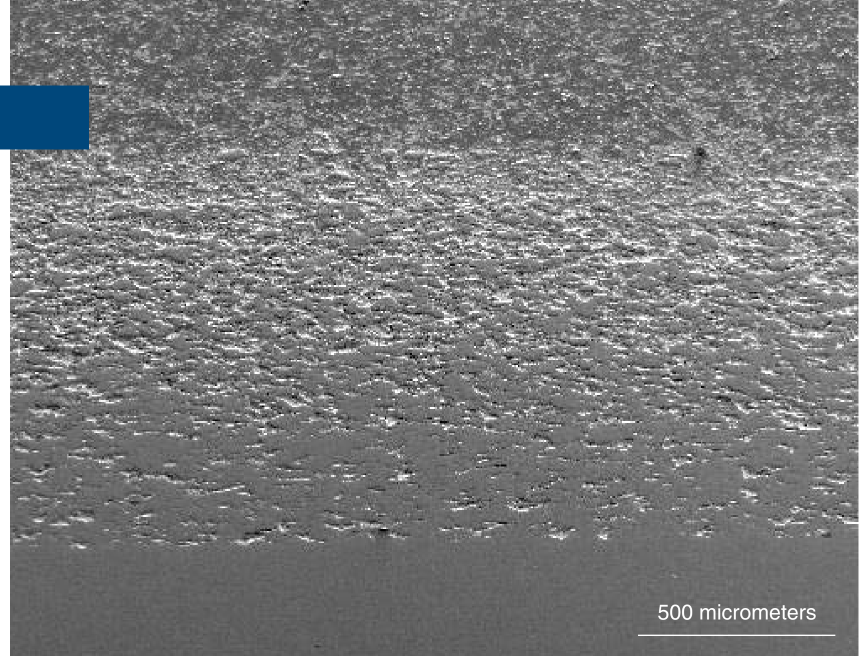
New Routes to High Temperatures and Pressures



A micrograph of a functionally graded material impactor in cross section reveals its constituent layers (magnified). The image depicts a smooth transition in density from one layer to the next between each of the five layers shown.

FOR several decades, Lawrence Livermore physicists have been among the scientific leaders investigating the properties of material under high temperatures and pressures. Such experiments help them fully understand how material behavior changes at the extreme conditions that occur in a nuclear detonation or a planet's interior. For example, in 1996, Livermore researchers used one of the Laboratory's two-stage light-gas guns to shock-compress hydrogen to its metallic state, a form of the element believed to be prevalent in Jupiter. (See *S&TR*, September 1996, pp. 12–18.)

Using various experimental platforms such as gas guns, lasers, magnetic accelerators, and diamond anvil cells, they can conduct experiments as close as possible to the desired pressures and temperatures and extrapolate the results to the conditions they want to investigate. A problem with these techniques is that researchers cannot examine the full range of material behavior as conditions change. Each platform yields slightly different but complementary information about the material being studied.

For example, typical shock-wave experiments on gas guns or lasers measure the Hugoniot, a particular relationship of pressure to density for a single shocked state. Lasers and magnetic accelerators can be used to study the principal quasi-isentropes, which represent the pressure–density relationship of materials at nearly constant entropy (a measure of the system's randomness or of irreversible work). In contrast, diamond anvil cells provide insight into material behavior on an isotherm or isobar, the pressure–density behavior at constant temperatures or pressures. (See *S&TR*, December 2004, pp. 4–11.)

All of these techniques provide valuable information, but each one is limited to a specific thermodynamic path: a Hugoniot, a quasi-isentrope, or an isotherm. Realistic materials and conditions do not occur along only one thermodynamic path but within a range of conditions. Extrapolating data between these constraints introduces uncertainties into the computer models used to simulate extreme environments. For instance, the temperature at a given pressure on the Hugoniot curve is higher than the temperature at the same pressure on a quasi-isentrope. In addition, the strain rates, or rates of compression, produced in shock experiments can vary as much as 10 orders of magnitude from those produced in quasi-static diamond anvil cell experiments.

To address this problem, physicists Jeff Nguyen, Daniel Orlikowski, and Neil Holmes have developed an experimental approach to study the exact conditions at which materials exist. Their technique uses multilayered functionally graded material (FGM) impactors (also called graded density impactors) in gas-gun experiments to simulate conditions that were previously inaccessible in a precisely controlled laboratory environment. Impactors are the projectiles a gas gun launches toward its target. With FGM impactors, data from a set of continually varying experiments can be used to create models of material behavior, say, along a path to a planet's core. Scientists no longer need to compare extrapolated data from several experiments conducted at disparate pressures and temperatures. The new impactors also increase design flexibility so that experiments are not constrained to one thermodynamic path.

Building Multilayered Impactors

More than 20 years ago, gas-gun experiments designed to achieve quasi-isentropic compression used impactors composed of several layers of different materials with increasing density. Experiments with these impactors created a modest shock, followed by a series of small steplike pressure increases.

Nguyen's team solved the problems of the initial shock and step-wise pressure increases with a rudimentary powder technique. The FGM impactors designed by Nguyen's team are discs with as many as 100 or more layers, each less than 30 micrometers thick (roughly equivalent to half the thickness of a sheet of paper). As the number of layers increases and the thickness of any particular layer decreases, the behavior of the observed pressure increases changes from steplike to continuous.

Livermore technician Eamon Loughnane built the first impactors by hand to demonstrate the flexibility of the FGM technique. In early experiments with these prototypes, density ranged from 0.1 to more than 10 grams per cubic centimeter. With the tape-casting technique developed by Laboratory engineer Peter Martin, the level of reproducibility and planarity improves to within 1 to 2 percent.

Now, impactors that create nearly continuous pressure profiles can be rapidly fabricated using advanced powder-processing techniques such as tape casting. In this process, individual layers are prepared from powdered metals such as copper, magnesium, and tungsten. The metal powders are mixed with an organic solvent, plasticizers, and binders to form a slurry, which is then cast onto a Mylar film. After drying, the tapes are smooth and flexible.

Tapes with different densities can be commercially prepared in advance, and a tape's composition can be tailored to provide specific properties. To fabricate an FGM impactor, technicians punch circular discs from different rolls of tape and stack them in the precise order determined for an experiment. The stacked tapes are laminated together, heated to remove the organic plasticizers and binders, and hot pressed to increase the density of the metal powders.

By carefully selecting the different layers for an FGM impactor, an experimenter can design the density profile for each experiment. For example, a series of impactor layers with increasing density imparts a compressive force, while a series of layers with decreasing density creates a controlled release of pressure. An abrupt increase in density from one layer to the next can be used to generate a shock wave. An FGM impactor made of up to 100 different tapes provides an unprecedented level of control of the temperature and pressure conditions. Thus, researchers can combine a powerful shock, quasi-isentropic compression, controlled pressure release, and periods of continuous pressure, all in one experiment. The currently achievable densities with the tape-casting technique range from 1.7 to 9 grams per cubic centimeter. Nguyen's team is working to extend this capability for a density range from 0.1 to about 15 grams per cubic centimeter.

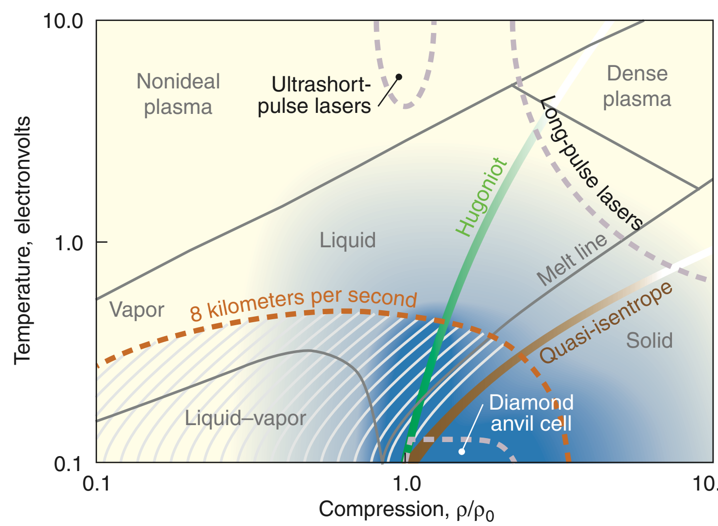
Experiments Validate the Approach

The Laboratory Directed Research and Development Program initially funded the FGM impactor research, which was proposed by Nguyen, Holmes, and physicist Fred Streitz. Orlikowski and Martin later joined the team along with physicist Reed Patterson and engineer Ryan Krone.

In the past six years, Nguyen's team has conducted more than 125 experiments using the FGM impactors on Livermore's light-gas guns. The researchers have analyzed the performance of the FGM impactor, obtained the equation of state (EOS) of tantalum and aluminum, and probed the strength of solid aluminum and the strain-rate effects of twinning (a form of deposition) in copper. They also have examined the liquid-to-solid phase transitions in bismuth and water and made novel materials.

In the gas-gun experiments, an FGM impactor, 3 to 8 millimeters thick and about 3 centimeters in diameter, hits its target at velocities of 1 to 8 kilometers per second. Solid targets are typically disks with thicknesses up to 5 millimeters, attached to a 10-millimeter-thick single crystal of transparent lithium fluoride. Measurements are usually made of the particle velocity of the material undergoing compression, which is recorded at the target–lithium fluoride interface by a diagnostic system called VISAR (velocity interferometer system for any reflector). In addition, an ellipsometer, which measures the polarization of light reflected from the target, can be used to provide information on the changing structure of a solid metal under pressure. Recovered samples can also be analyzed by such techniques as scanning electron microscopy and Vickers hardness testing.

Experimental observations are compared with simulations developed by Orlikowski using Livermore's CALE code. According to Orlikowski, the simulations have accurately modeled the experimental results. Models also guide researchers in designing the gas-gun experiments and choosing the impactor layers.

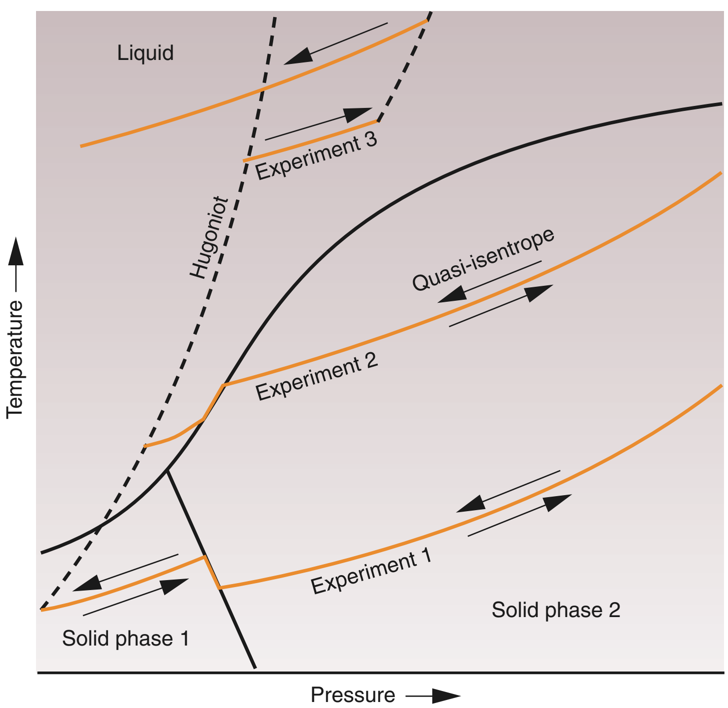


Experiments using long- and ultrashort-pulse lasers, diamond anvil cells, and gas guns access different regions of high temperature and pressure, or compression, where materials can change phase. Gas-gun experiments using Livermore's functionally graded material impactors can record measurements in continuous areas (hatched) that are not accessible with traditional techniques. Blue indicates the level of understanding from high (dark) to low (light).

A New Class of Experiments

With the greater flexibility offered by today's FGM impactors, researchers can design experiments that address precise questions about material behavior. For planetary studies, they can re-create the exact conditions of a planetary isentrope, which is hotter than a principal isentrope, by shocking the sample to a temperature-elevated state before the sample is quasi-isentropically compressed. To study the boundary between material phases, they are no longer constrained to data on the Hugoniot, isentrope, or isotherm. Instead, they can look at the exact temperatures and pressures of the phase transitions.

The team's experiments are helping scientists better understand the dynamic response of materials, including the kinetics of phase transitions occurring at high temperatures and pressures in nonequilibrium situations. Phase diagrams produced by FGM impactors can then be compared with those obtained using traditional methods. In one series of experiments, Nguyen's team quasi-isentropically compressed liquid bismuth and liquid water, separately converting both to solids. A simple shock on a liquid bismuth sample imparts an overly significant amount of energy (more than quasi-isentropic compression) to the sample, which prohibits solidification. Similar experiments with water produced a phase transition from liquid water to ice at elevated temperatures normally associated with the liquid phase.



Functionally graded material impactors allow researchers to examine material behavior within a range of extreme pressures and temperatures. In hypothetical experiment 1, a solid in one phase is quasi-isentropically compressed into a solid of a different phase, or crystal structure, and released back into the original phase. In experiment 2, a solid is shocked to its liquid phase and then quasi-isentropically compressed into a solid phase. In experiment 3, a solid is shocked into a liquid and then isentropically compressed while remaining a liquid. It is then shocked again and released to a lower pressure state.

The team is also using the FGM impactor to measure the principal quasi-isentrope for an EOS study. Another set of impactors will be used to create strain rates that fall between the capabilities of the traditional experimental techniques. These impactors are tailored to the compression (strain) rates obtained in magnetic accelerator and laser experiments. Data from the different techniques will be compared to examine strain-rate effects.

In addition, Nguyen's team is evaluating a metal's strength through a sequence of shock, compression, and then controlled release. Data from these experiments can be used to improve the fidelity of material strength models and thus help scientists better understand how metals respond to extreme conditions. The team is also collaborating with physicist James McNaney to examine how strain rate affects high-pressure twinning in copper. A research project with physicist Eduardo Bringa is studying superhard materials.

The experimental approach is being extended to the Joint Actinide Shock Physics Experimental Research (JASPER) Facility at the Nevada Test Site. Experiments with the JASPER gas gun will improve scientific interpretations of EOS measurements and predictions of phase behavior and material strength. In fact, Nguyen and his colleagues received a Defense and Nuclear Technologies Directorate award for their work in support of JASPER experiments. "We are accessing new regions of phase space by designing virtually any combination of shock, release, quasi-isentropic compression, and static pressure," says Nguyen. "We're controlling experiments to a degree never thought possible."

—Arnie Heller

Key Words: diamond anvil cell, equation of state (EOS), functionally graded material (FGM) impactor, graded density, Hugoniot curve, Joint Actinide Shock Physics Experimental Research (JASPER) Facility, quasi-isentropic compression, two-stage light-gas gun.

For further information contact Jeff Nguyen (925) 423-6838 (nguyen29@llnl.gov).