特集 — 衝撃超高圧力研究の新展開 —

Use of the Z Accelerator for Condensed Matter Studies
at Multi-Mbar Pressures

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The ability to study material response during isentropic compression has been a grand challenge of the scientific community for several decades. However, development of precision techniques for producing isentropic compression at high pressures has been limited. The revolutionary advance for using planar magnetic loading on the Z accelerator accelerated this goal by enabling quasi-isentropic studies on macroscopically sized materials to over 5 Mbar. In addition, the accelerator is easily configured to launch flyer plates to velocities more than four times higher than possible with conventional launchers, thus allowing shock compression studies in the laboratory to pressures exceeding 20 Mbar.

[Z accelerator, high-pressure, equation of state, shock and isentropic compression]

1. Introduction

The properties of matter at extreme pressures and temperatures have been of interest to the scientific community for many decades, both from the standpoint of understanding the fundamental properties of matter at these states, as well as application to planetary physics. P.W. Bridgman[1] pioneered development of hydrostatic pressure compression for studying physical and mechanical properties of materials at pressures of several GPa (1 GPa = 10 kbar). His prolific work provided a solid basis for studying the properties of a wide variety of materials at high pressures and temperatures, including studies of equations of state (EOS), phase transformations, optical and mechanical properties. These studies were conducted for quasi-hydrostatic loading, so that kinetic properties were not generally studied. The time factor is important in many physical processes, such as phase transitions, since nucleation and growth of new phases, generally occur on nanosecond time scales. Understanding temporal effects is crucial to identifying fundamental physical mechanisms and developing material models.

The limiting pressures of a few GPa achievable with hydrostatic compression was overcome in the 1940's

through development of shock wave techniques for producing controlled pressures of several Mbar[2-6] under dynamic loading. In the standard shock wave experiment, a planar compression is applied over the surface of a flat specimen, producing a planar shock wave that propagates through the material, compressing it to high pressure. Initially, plane wave explosive lenses[2, 3] were used to produce high-pressure shock waves, although a variety of other methods became available, as illustrated in Fig. 1. These include smooth bore high velocity guns to launch flyer plates[4], laser deposition[7], and magnetically-driven flyer plates for higher plate velocities and hence higher impact pressures[8]. Shock wave methods have been extremely successful for accurately determining dynamic properties at high pressures, resulting in an extensive number of experiments that have been conducted with this method. In parallel with development of shock compression techniques, a variety of diagnostic methods were developed for probing material response on time scales ranging from pico- to microseconds (10^{-12} - 10^{-6} s), which has clarified the role of time in describing phase transition kinetics and other time-dependent processes. Several review articles summarize previous studies[4-6].

In a planar shock wave, two properties are typically

measured - the particle and wave velocities[4]. The conservation equations of momentum, mass and energy then allow determination of the pressure, density, specific energy and particle velocity produced by the shock [2-6, 9], as illustrated in Fig. 1. A single shock wave experiment starting from ambient conditions produces a series of high-pressure data points referred to as the principal Hugoniot for the material[4], that have been obtained for a large variety of materials[2-6].

The Hugoniot curve is important in developing an EOS for a material, but is limited to a single curve on the EOS surface and therefore does not provide a complete representation of material response. In addition, the highly irreversible nature of shock compression produces high temperatures at pressures of several Mbar. Measurements of temperature in the shock state would be useful for developing a complete $P(V,T)$ EOS, but presently these are not sufficiently accurate to constrain EOS theories. To provide additional information for developing a complete EOS, it is necessary to supplement Hugoniot data with other high pressure response, such as isothermal compression. Since the temperature remains at ambient values, comparing response of isothermal compression to shock loading provides a determination of thermal contributions. A major limitation of hydrostatic compression, however, is that the peak pressures attainable are presently limited to 2-3 Mbar with Diamond Anvil Cell (DAC) techniques.

The need to more completely understand material response at ultra-high pressures motivated development of shockless loading techniques to extend the pressures achievable for dynamic off-Hugoniot loading paths.

Fig. 1. Experimental methods used to produce planar shock loading of materials at high pressures.

Several methods have been investigated for over thirty years to develop these methods. During shockless loading, a smooth compression is produced adiabatically so that for an inviscid fluid the compression process is essentially isentropic. This allows dynamic response to be evaluated for temperature states substantially removed from the Hugoniot, providing additional information about the EOS. Although shockless fluid compression is nearly isentropic, the response of solids is usually described as quasi-isentropic because of irreversible effects produced by plastic work and visco-plastic compression.

The earliest technique for producing quasi-isentropic response involved use of ramp-wave generators such as fused silica, which exhibit anomalous stress-density response over a portion of their dynamic response[10], resulting in conversion of an input shock wave input to a planar ramp wave output on the back side. A sample mounted on this surface thus experiences smooth quasi-isentropic compression. Peak pressures of about 3 GPa can be obtained with this method. Higher shockless pressures have been obtained through the use of graded-density impactors, illustrated in Fig. 2, which use a distribution of material densities in a plate impactor to produce a low initial shock, followed by smooth compression to peak stress. Barker[11] first developed this technique using particle sedimentation techniques to produce the desired density gradient. The method has been referred to as ICE, for Isentropic Compression Experiment, because of the near-isentropic compression response obtained. The graded density impact method has been improved upon recently through use of vapor plating methods to achieve the desired density distribu-

Fig. 2. Techniques used to produce shock and shockless loading of materials at high pressures.

tion and to obtain more precise control of loading and unloading conditions[12]. Both of these methods are limited by small initial shocks produced by the initial impact and by reproducibility issues. Magnetic compression[13], which will be discussed in a following section, and laser plasma compression[7, 14] have also been used to produce shockless loading.

Fig. 3a illustrates schematically the pressure-volume dependence obtained for isothermal, isentropic, and shock compression of a fluid. The Hugoniot lies above both of these curves at a given volume because of increased shock temperature; however, it should be noted that the Hugoniot is second order tangent to the isentrope at ambient conditions, so the pressure differences are small at low pressures[9]. The specific temperature states obtained in aluminum[15] for these different loading conditions are shown in Fig. 3b. Shock heating induces the onset of melting for shock pressures of about 120 GPa (1.2 Mbar), with complete melting for shock pressures above 1.6 Mbar. Because temperature increases are minimal, use of isentropic or isothermal compression provides unique opportunities to study the solid phase to several Mbar; in fact, the highest pressure achievable with either method are theoretically unlimited if the melt boundary slope remains below that for isentropic or isothermal compression.

2. Z Accelerator

For most accurate data, quasi-isentropic compression requires smooth, shockless loading on samples large enough to accurately measure the wave velocity at all

Fig. 3. Relationship of shock and shockless loading, referred to as quasi-isentropic loading. (a) Schematic pressure-volume response for shock and isentropic loading of a fluid. (b) Temperature-pressure diagram for shock and isentropic loading of aluminum.

points in the evolving ramp wave that propagates through the sample. In 1998, a magnetic loading technique was developed which has been revolutionary in producing accurate measurements of quasi-isentropic compression on macroscopically sized samples[13]. The method uses an intense plane of current to produce a uniform magnetic pressure on the surface of a planar sample. To achieve Mbar pressures in sub-microsecond times, very high current densities are necessary, typically several mega amperes per centimeter. The Z accelerator at Sandia National Laboratories provides the required time-dependent current pulses. A picture of the accelerator is shown in Fig. 4a. A circular array of thirty six capacitors is used to store a total electrical charge of about 11 MJ. Upon discharge of the accelerator, the charge is released through a series of fast acting switches, resulting in the transmission of an electrical current pulse of about 20 Ma in magnitude and 100-300 ns in duration applied to a target configuration containing multiple specimens. To achieve planar loading, the current is applied across parallel sections shaped in the form of a square anode and cathode, as shown in Fig. 4b; this configuration produces a current density, J , of several MA/cm on both surfaces. The resulting magnetic field is the product of the permeability, μ_0 , and current density, $\mu_0 J$. The magnetic pressure electrode gap is $(1/2 \mu_0) B^2$ [8, 16-18] and increases smoothly during the pulse risetime. The magnetic pressure is converted to hydrodynamic pressure in the electrode and consequently propagated as a smoothly increasing ramp wave through the electrode and then into a specimen placed on its back surface. Generalized Lagrangian analysis methods have been developed to analyze the evolution of ramp waves in order to deduce the continuous stress-volume states[4, 19, 20, 21]. An important point is that the evolution properties of the ramp waves must be measured before the ramp forms into a shock wave, which will produce high temperature states in the compressed state, as discussed earlier. This is achieved by tailoring the shape of the input pressure wave[8], which significantly extends the sample thickness required for shock formation and results in typical sample thicknesses on the order of 1 mm or larger.

The use of the Z accelerator to obtain the first measurements of smoothly increasing quasi-isentropic compression in the Mbar pressure regime[19, 20] is an

Fig. 4. Magnetic loading technique for producing isentropic compression of materials to Mbar pressures. (a) Picture of the Z accelerator. (b) Sample configuration for planar magnetic loading. The cylindrical red/purple disks represent samples placed on an anode with a small gap to the cathode (purple). A typical current pulse used to produce magnetic field is shown.

exciting development that has been a goal of the high-pressure community for several decades. With input current/pressure tailoring, the technique has achieved peak pressures of over 4 Mbar, although higher pressures are anticipated with recent improvements to the accelerator[23]. Subsequently, laser loading methods have also achieved similar pressure levels for loading time scales of a few nanoseconds and sample dimensions of a few tens of microns, compared to the hundreds of nanoseconds and mm-sized samples possible with magnetic loading. In addition, the loading or strain rates achieved in the laser-induced ramp waves are large, typically on the order of $10^8/s$ compared to the rates of $10^6/s$ characteristic of magnetic loading. This has important consequences in examining time-dependent processes under dynamic loading, such as kinetics of phase transitions or time-dependent yielding effects.

It is also possible to use magnetic loading with the accelerator to launch flyer plates to ultra-high velocities unachievable with conventional gas gun methods. As example, it has been used to accelerate aluminum flyer plates of ~ 1 mm thickness to velocities of about 34 km/s[8, 22, 23], resulting in impact pressures up to 13 Mbar in low impedance materials such as aluminum

Fig. 5. Pressure-temperature-density EOS surface for aluminum, showing the regions accessible by a combination of shock compression, shock-unloading and quasi-isentropic compression.

and about 24 Mbar in high impedance materials such as tungsten. To date, shock compression states up to about 8 Mbar have been achieved in aluminum (23) and 13 Mbar in diamond (23). The flyer plate approach preserves the accuracies inherent to traditional gas gun impact techniques and also allows a large variety of experimental configurations, including determination of unloading curves, measurement of sound speeds, and

multiple loading. This flexibility in varying loading conditions allows study of a rich plethora of thermo-physical properties, including measurement of local sound speeds to identify phase transitions such as melting[24], strength properties[15], unloading isentropes[24], phase transitions[25] and a variety of other properties.

A representation of the EOS surface for aluminum accessible through a combination of shock compression followed by unloading, and also isentropic loading is represented in Fig. 5. For impact pressures exceeding 1.2 Mbar the Hugoniot enters the solid-liquid mixed phase region, resulting in compression of the liquid phase at higher pressures. Unloading from shock states in the fluid region produces isentropic decompression. For sufficiently high shock pressures of about 10 Mbar the unloading isentrope intersects the liquid-vapor mixed phase region, resulting in partial vaporization of the liquid phase. The unloading isentrope intersects the critical point for shock pressures of about 12 Mbar, resulting in complete gaseous response for unloading from higher shock pressures. This example illustrates that a combination of shock loading and unloading can be used to probe both high-temperature compressed and expanded regions of the EOS.

In contrast to shock compression, isentropic compression probes a substantially cooler region of the EOS surface, as also illustrated in Fig. 5, namely, the low temperature, high-pressure phase. Thus, through a combination of shock loading, followed by isentropic loading or vice versa it is possible to explore with dynamic com-

pression experiments the full EOS region lying between the isentrope and Hugoniot. Furthermore, by unloading from different Hugoniot states, it is possible to probe the complete hot, expanded surface between regions of condensed matter and regions of dense vapor and plasma states so that in principle, the entire surface can be probed. Presently, isentropic compression of aluminum has been achieved to about 2.4 Mbar[20] as shown in Fig. 6a, shock compression to about 8 Mbar[22, 23] shown in Fig. 6b, and also unloading response of aluminum to states below the critical point[24].

In addition to studies of the EOS surface in aluminum, the Z accelerator has been used for a variety of other ICE studies, including polymorphic phase transitions in zirconium to investigate the effects of impurities on phase transition kinetics[25]; temperature dependence of the α - ϵ phase transition kinetics in iron[26]; resolidification in tin[27, 31]; optical properties of sapphire[28]; and determination of strength properties during isentropic loading[15]. The magnetically driven flyer plate technique has been used to study the high-pressure EOS properties of liquid deuterium for shock loading to 1.8 Mbar[23, 29, 30], shock loading of aluminum to 8 Mbar[23], unloading of aluminum from a shock pressure of 5 Mbar[24], and shock wave studies of low

(a)

(b)

Fig. 6. Experimental data obtained on aluminum to several hundred GPa (several Mbar) using the Z accelerator. (a) Measured principal isentrope with expected error bounds. (b) Measured principal Hugoniot (light gray points) with expected errors.

density aerogel foams used for unloading experiments[23]. Work is also in progress to study the Hugoniot and unloading response of diamond and beryllium to pressures of about 13 Mbar and 6 Mbar, respectively, using layered composite copper-aluminum flyer plates[23]. These capabilities have allowed measurement of Hugoniot states with accuracies of less than 1% in shock and particle velocities, as well as estimates of material strength and sound speed measurements at these pressures[23]. The sound speed measurements have been useful for identifying high-pressure melt boundaries, as well as polymorphic transitions occurring at high pressure[23].

3. Future directions

The smoothly varying pressure loading histories and ultra-high velocities obtainable on the Z accelerator are an exciting new development in high-pressure experimental capabilities and have enabled high precision studies of material response at multi-Mbar pressures. Work is presently in progress to improve the performance of the accelerator, which will result in increased pressures at which isentropic compression studies can be conducted and increased flyer plate velocities, which will double accessible shock pressures. It is anticipated that isentropic pressures exceeding 6 Mbar and aluminum flyer plate velocities up to 50 km/s should be possible in the near future. With these capabilities, the ICE method will allow study of high-pressure phases of materials not previously detected at ultra-high pressures. With the increased flyer plate capability, shock pressures to about 25 Mbar will be achievable in medium-impedance materials, such as aluminum, and over 50 Mbar in high-impedance materials, such as tungsten. This will further extend high-precision material studies into thermodynamic regions presently inaccessible with conventional methods, which will validate existing theoretical models, such as the Debye and Thomas-Fermi EOS theories and provide critical data for developing new theories in EOS regions where present theories are not applicable. For the hot, expanded phase of the EOS, routine studies of dense gases and plasmas near critical points will also be possible, which is an area of intense interest although data about material response in this regime are limited[32]. In

addition, because of the ability to measure physical properties such as sound speeds and mechanical properties such as material strength in very high pressure solid phases, as demonstrated by the recent work on diamond and beryllium[23], new opportunities will be available for determining thermophysical properties needed for developing equations of state based on first-principles.

An important recent development is the transfer of the pulsed power techniques developed for ICE experiments on the Z accelerator to compact pulsed power generators that will provide more access to the capability and result in more cost efficient experiments. This capability has now been demonstrated on several applications[15, 33], including measurements of dynamic yielding and optical properties during isentropic compression. In addition, soft recovery methods are being developed on the small pulser for subjecting materials to high pressure ramp loading, followed by soft recovery of the compacted material. This capability will also provide opportunities for producing advanced materials through high-pressure synthesis by allowing control of pressure and temperature states produced under dynamic compaction, which will be important for developing computer-designed materials.

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