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ISENTROPIC COMPRESSION EXPERIMENTS ON THE Z ACCELERATOR

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Abstract. In many technological and scientific applications it is desirable to accurately determine the off-Hugoniot isentropic response of materials at ultra-high pressures. Although unloading isentropes can be determined from unloading profiles, experimental techniques for measuring compressive isentropic response have been extremely limited. A brief summary is presented of the various techniques that have been developed for isentropically compressing materials to high pressure. A new technique is also discussed, which shows considerable promise for performing isentropic compression experiments with smooth ramp waves to multi-Mbar pressures. This approach uses the high current densities produced with fast pulsed power accelerators to create continuous magnetic loading to a few hundred kbar in the present study over time intervals of 100 ns. Application of the differential equations of motion to the resulting ramp waves determines pressure-volume states continuously along the isentrope. The method has been successfully applied on copper and iron; these results determine the isentropic compression curve in copper and evaluation of the kinetic properties of the alpha-epsilon phase transition in iron.

INTRODUCTION

The high-pressure equation-of-state (EOS) of materials is normally determined experimentally using shock loading techniques [1]. In these methods, flat cylindrical specimens are subjected to planar shock loading for time durations of a few hundred nanoseconds to several microseconds and kinematic properties of the steady shock waves produced, such as the shock velocity and particle velocity behind the shock, are measured. These data can then be used with the equations of motion to determine the pressure-volume-energy response of the material along a path referred to as the Hugoniot, which is the locus of end states produced by steady shock compression. Shock wave techniques have been extremely useful for generating high pressure EOS properties, but the Hugoniot curve is not sufficient to determine a complete pressure-volume-temperature surface. In this regard, several scientific and technological applications require specification of thermodynamic response along thermodynamic paths that do not lie on the Hugoniot. [2]

In this paper, a brief review is provided of previous experimental approaches for measuring the high-pressure isentropic response of materials.

A new capability for producing isentropic compression with fast pulsed power techniques is then discussed, with specific examples of recent isentropic loading data on copper and iron to about 300 kbar that illustrate feasibility of this technique. Prospects for producing isentropic compression to multi-Mbar techniques are also discussed.

BACKGROUND

The relationship between the isotherm, typically measured with hydrostatic pressure vessels, the Hugoniot produced by shock loading, and the room temperature isentrope is shown in Fig. 1. The isentrope, which lies between the isotherm and the Hugoniot, represents the response obtained for continuous, adiabatic, and reversible compression, whereas the Hugoniot is the locus of end states achieved by single shock compression. The Hugoniot represents an adiabatic compression process by steady shock waves, but is a highly

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irreversible process, in contrast to the isentrope. The isentrope and the Hugoniot are second-order tangent at the initial volume state [3].

It is extremely difficult to produce states of isentropic compression with static methods. However, it is possible to approach the isentropic loading response of solids and liquids with dynamic techniques. Dynamic methods can produce shockless, adiabatic loading, but the process is not reversible because of elastic-plastic and dissipative viscoplastic processes involving dynamic viscosity that invariably arise. For this reason, ramp or continuos loading is often referred to as quasi-isentropic.

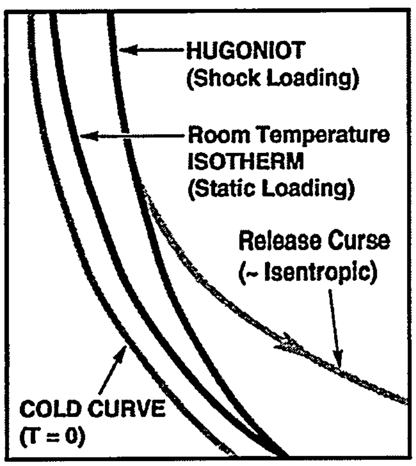


FIGURE 1. Relationship between the room temperature isotherm, isentrope, and Hugoniot.

Flat-plate projectile impact techniques provide a way to produce precision loading of solids and liquids to high pressure. In these techniques, a flat plate is launched to high velocity and impacts a flat disk, as shown in Fig. 2a. At high pressures, a planar shock is produced directly upon impact with the pressure amplitude determined by the impact velocity. Each experiment produces a unique state of pressure, volume and internal energy, which can be determined from the conservation relations. Projectile impact techniques have been previously used to study a large number of materials to shock pressures of several Mbar however, they have not been generally used for producing continuous pressure loading.

Barker and Hollenbach [4] were the first to produce well-controlled isentropic loading of solids using projectile impact technologies. The first measurements were made with a ramp wave generator, fused silica, which has a longitudinal stress-uniaxial strain loading path to approximately 30 kbar that exhibits negative curvature. The negative curvature of the loading path results in an unstable condition for shock loading, thereby generating linear ramp waves rather than shock waves to 30 kbar. The risetime of the ramp wave can be controlled by the thicknesses of the fused silica buffer used, since the foot of the ramp travels with the ambient sound velocity and the top part travels with the sound velocity at that pressure. The output ramp wave from the fused silica generator can thus be used to produce initial ramp loading in a test sample by placing the

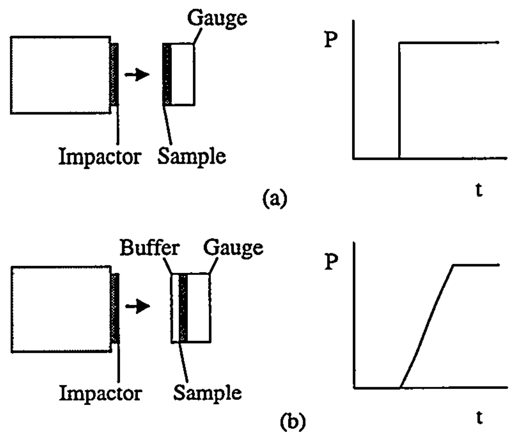


FIGURE 2. (a) Typical impact condition for shock loading of samples; (b) use of fused silica buffer for producing shockless loading of samples.

sample on the back side of the fused silica buffer, as shown in Figure 2b. In addition to EOS measurements, ramp loading can be used for a variety of other applications, such as studying the dependence of mechanical properties on loading rate [5].

Fused silica ramp generators are limited to pressures of about 30 kbar. However, other materials have been used to generate ramp waves to higher pressure. The principal requirement is that the curvature of the loading path be concave downward so that stable shock waves cannot be supported. Some ceramic materials have this characteristic, due to the open nature of the crystal structure. Asay and Chhabildas [6] used a commercially available ceramic to produce shockless loading in aluminum to pressures of about 200 kbar. Ramp wave profiles were obtained on several different sample thicknesses of aluminum. If rate-dependent effects are minimal, the resulting particle velocity profiles can be analyzed as simple compression waves [7]. In this case, the stress-volume loading curves can be determined from the differential equations:

$$d\sigma = \rho_0 \, c_l(u) \, du, \tag{1}$$

$$dV = V_0\, du/c_l(u), \qquad (2)$$
$$dE = 1/2\, \sigma(V)\, dV, \qquad (3)$$

where σ is the longitudinal stress component, ρ_0 is initial density (1/V), $c_l(u)$ is the Lagrangian wave velocity a given particle velocity, u, in the wave profile, and E is specific energy along the quasi-isentropic loading path. The resulting pressure-volume states produced in the experiments are shown in Fig. 3.

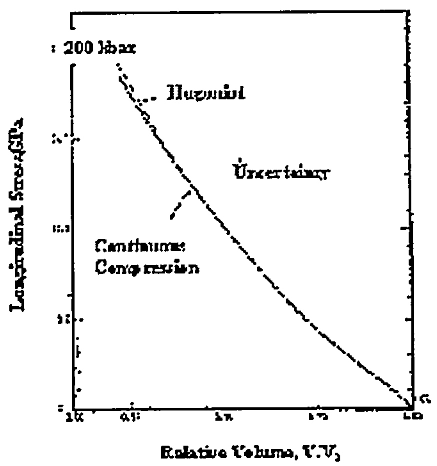


FIGURE 3. Free surface velocities obtained on aluminum samples continuously loaded to 200 kbar.

Since the Lagrangian wave velocity will increase with pressure in most materials because of non-linear response, the isentropic compression waves will steepen ultimately into steady shock waves. The error bar of 2% given in the figure is typical of the accuracy that can be achieved with this technique.

Barker [8] developed a method for producing quasi-isentropic loading of materials to very high pressures. In this technique, the constant density impactor shown in Fig. 2a is replaced with a graded density impactor. Barker used particle sedimentation techniques to produce an impactor with a very low density, approximately equal to that of a plastic, at the impact surface, followed by a gradual increase to a final density characteristic of a high impedance material such as tungsten. Through careful control of the density profile, he was able to produce impact conditions that comprised of an initial small shock followed by ramp loading to high pressure.

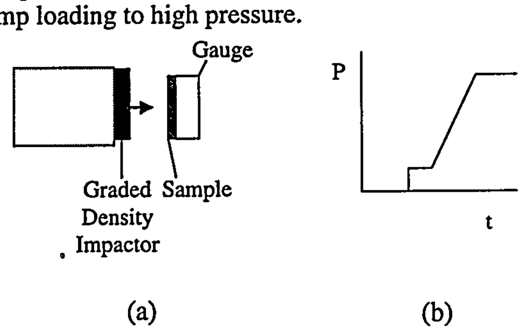


FIGURE 4. Use of graded density impactors for producing shockless loading of specimens. (a) impact configuration, (b) typical input stress profile.

Although this approach was successfully used in a few applications, the technology for making graded density impactors with particle sedimentation techniques proved to be a formidable task, so that this technique has not gained widespread acceptance. Chhabildas et al. [9] developed a layered-plate technique for introducing a series of small shocks into samples in a nearly continuous fashion, that approximated the graded density approach achieved in the sedimentation techniques. This technique provided much better control over density variations so that initial input conditions could be closely replicated in experiments on different sample thicknesses.

Z ACCELERATOR

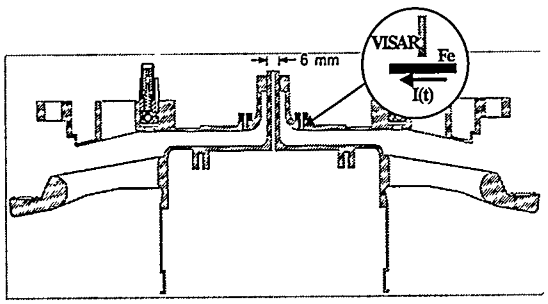
Pulsed energy sources offer another approach to producing shockless loading of specimens. Hawke et al. [10] used explosively driven pulsed power techniques to cylindrically compress liquid deuterium to high pressures in hopes of achieving metallization of this material; metallization has been predicted to occur at isentropic pressures as low as 2 Mbar. However, this technique is not easily capable of producing continuous measurements of pressure and volume under isentropic loading conditions. A new technique has been recently demonstrated on the Z Accelerator at Sandia which has considerable promise for producing isentropic compression in a variety of materials. The Z Accelerator is a low inductance pulsed power generator capable of capacitively storing 11.6 MJ of electrical energy to produce up to 20 MA of electrical current. The capability has been used for a variety of applications, including acceleration of electrons and ions for inertial confinement fusion applications and generation of intense x-ray environments through Z-pinch driven plasma implosions [11]. In the present applications, it is used in the current generation mode, which can be used to magnetically load specimens to high pressure.

The technique for magnetically loading specimens is illustrated in Fig. 5. Two configurations have been demonstrated in experiments to demonstrate feasibility of using this technique for isentropic compression experiments. In the geometry shown, the sample is located in an annular geometry for a direct-current short between the anode and cathode. Magnetic field is produced in the region between the anode and cathode as the current increases to its peak value. A typical current profile is a S-shaped curve peaking at about

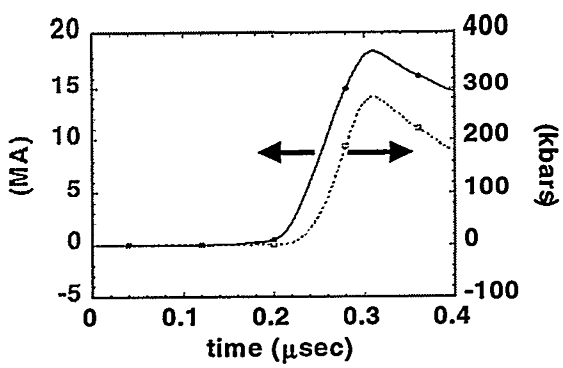
20 MA over a risetime of approximately 100 ns and falling off after that. Magnetic pressure is produced during this period through the following relation:

$$P(t) = (1/2\mu_0)\, B^2 (t), \\ = 1/2\mu_0 J^2 (t), \qquad (4)$$

where μ_0 is the permeability of free space, J is current density, and B is magnetic field.



(a)



(b)

FIGURE 5. Schematic for producing isentropic compression with the Z Accelerator. (a) technique used to load iron samples to 300 kbar; (b) input current and pressure applied to the sample.

The first Isentropic Compression Experiments (ICE) wave experiments were performed with the Z Accelerator to establish the feasibility of detecting polymorphic phase transitions. Two experiments were performed on iron samples 3 mm in diameter and with nominal thicknesses of 0.5 mm and 0.8 mm. The iron used in these experiments had a purity of 99.8%, an average grain size of 150 μm and a density of 7.85 g/cm ^{3}. The iron samples used were from the same stock as those studied by Barker and Hollenbach [12]. The samples were located in the annular anode plate at a radius of 1.385 cm from the center of the anode, as shown in Fig. 5a. The current applied to the front surface of both iron specimens ramped up to about 19.3 MA over a time interval of about 100 ns. For this configuration, the current density is not constant across the loading surface of the sample, but varies with radius as I/2πr, where I is the applied current. This induces a non-planar pressure gradient that was estimated to be as high as 7% from the center to a radius that could influence motion at the center. Although this variation is not acceptable for accurate EOS measurements, it is low enough perturbation to demonstrate the sensitivity of the technique for detecting phase transitions for evaluating kinetic effects.

Figure 6 illustrates resulting wave profile obtained on the free surface of 0.5 and 0.8-mm thick samples of iron. The iron samples were polished to a mirror finish and dual 200-μm fiber optic cables were used to couple VISAR signals to each sample. The current risetime was measured to be 100 ns. The free surface velocity profile at 0.5 mm thickness shows an elastic precursor followed by the onset of the α - ε phase transition at a velocity of about 0.6 km/s. A plateau is observed at a larger velocity signifying the onset of a two-wave structure characteristic of shock-induced phase transformations. The profile for the thicker sample also shows the elastic wave, followed by a well defined two-wave structure.

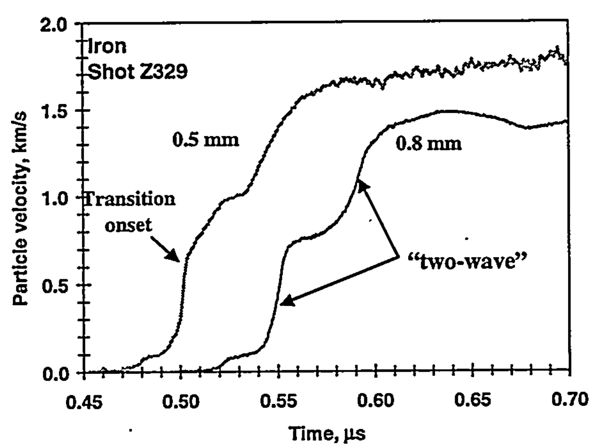


FIGURE 6. VISAR profiles obtained on 0.5 and 0.8 mm thick iron samples.

The experimental results were modeled with a rate-dependent model for the α - ε phase transition, which can be cast in the familiar Maxwellian relation [13].

These results demonstrate the importance of phase transition kinetics in the α - ε phase transition and the ability of ICE wave experiments to detect and quantify the transition rates. The average transition rate required to model the present experimental results is about 50 μs^{-1}, which is compatible with previously observed transition times under shock compression [12].

To obtain approximate 1-D loading, another set of experiments was performed in which thin copper disks were placed on the cylindrical feed of the anode, rather than the geometry as shown in Fig. 5a. The configuration is nearly cylindrical, except for the interface of the flat specimens with the cylindrically symmetric anode. The specimens were located at a radius of 20 mm with respect to the center of the anode. The location of the flat specimen in the cylindrical anode introduces a small variation in pressure loading across the 3-mm diameter face of the sample on the order of 1%. The copper samples used were OFHC with a density of 8.93 g/cm^3 and thicknesses of 0.6 and 0.9 mm. They were polished to a mirror finish for the two VISARS, which were coupled to the samples with dual 200-μm fiber optics.

The resulting wave profiles can be analyzed to provide an estimate the loading path in the sample followed by the ramp waves. Lagrangian wave profile techniques have been developed to analyze arbitrary ramp waves [7] with rate dependent properties. Generally, these analysis require a minimum of three independent profiles. Since this was not possible in the present experiments, Eq. (1) and (2) were used to obtain an approximate loading curve. To perform this calculation, it was assumed that the in-situ particle velocity at any point in the wave was one half of the measured free surface velocity. Computer simulations of the experiments indicated that this was a good approximation. The Lagrangian wave velocity corresponding to the selected particle velocity was obtained from the difference in transit time between the two samples. This approach does not account for the perturbation to the waves near the free surface and will lead to errors in the measured wave velocities at a give particle velocity. However, for the low peak pressures obtained in the present experiments, computer simulations showed that the error is less than 0.5% in pressure and less than 0.1% in specific volume. This level of error is consistent with conclusions reached by Barker for using the free surface approximation to estimate the in-situ particle velocity in iron [12].

For the approximations discussed above, it was possible to estimate the stress-volume loading path over the free surface velocity range to 0.4 levels. The results are shown in Fig. 7. Also shown is an estimate of the isentrope in copper [14]. The figure illustrates that the experimental data are slightly higher than the theoretical predictions, by a maximum of about 4% at 40 kbar. Part of this difference could be due to the known strong work hardening of copper under shock compression [15] and also from previous observations [9] that the compressive strength of metals under isentropic compression is usually larger than that for shock compression to comparable pressures. Preliminary numerical simulations of the experiment indicate that inclusion of the work hardening data from Chhabildas [15] should result in closer agreement between experiment and theory.

It was also possible to estimate the spall strength in copper from the decrease in current, resulting in a pullback in particle velocity of about 0.07 km/s. This corresponds to a spall strength of approximately 17 kbar, which is compatible with reported values .

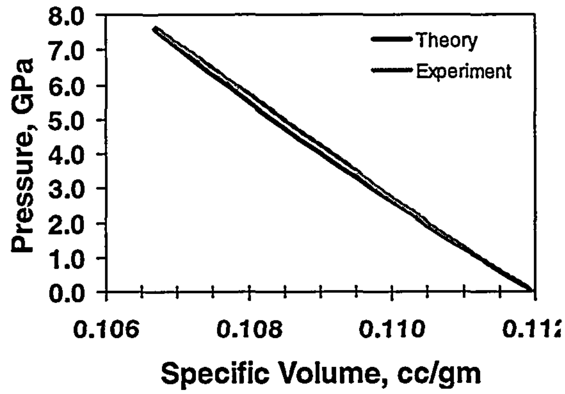


FIGURE 7. Stress-volume curve in copper resulting from the ICE wave profiles

The peak pressures achieved to date have been limited to about 300 kbar. This occurred because these experiments were add-ons to performance shots on the accelerator. However, it should be possible to design ICE wave experiments on the accelerator which would produce planar loading of up to eight specimens simultaneously to peak pressures of 1.6 Mbar. These experiments are planned for the near future.

CONCLUSIONS

This paper provides a brief review of experimental techniques for producing dynamic isentropic compression of samples to pressures of several hundred GPa. Traditional gun launch techniques include use of buffer plates, such as fused silica, that exhibit negative curvature to their stress-strain response and graded-density impactors. Graded-density impactors have been used to study isentropic compression of specimens to pressures exceeding 2 Mbar on high-impedance materials. A recent development includes the use of the Sandia Z Accelerator to produce magnetic compression in planar specimens to pressures of a few hundred kbar over time scales of 100 ns. These techniques have been successfully applied to isentropic compression of iron to 300 kbar and copper to 130 kbar. The iron results indicate that it is possible to study the polymorphic phase change that occurs at 130 kbar and also the kinetic properties of the transformation. The copper results indicate that with further improvements in progress it should be possible to measure continuous isentropic compression curves in materials of interest to pressures exceeding 1 Mbar.

The Z accelerator is limited to peak currents of about 20 MA. By reconfiguring the anode-cathode geometry it should be possible to obtain constant current density and thus driving pressure to about 3 Mbar. The next generation accelerator referred to as ZX, which is being proposed will have the capability to generate currents to 50 MA and resulting peak pressures to 15 Mbar.

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REFERENCES

1. Asay, J.R, and G.I. Kerley, *Int'l J. Impact Eng'g*, **5** 69-99 (1987).
2. Lindl, J., *Physics of Plasmas*, Nov. (1995).
3. Duvall, G.E., unpublished, 1968.
4. Barker, L.M. and R.E. Hollenbach, *J. Appl. Phys.* **41**, 4208-4226 (1970).
5. Barker, L.M., Bull. Amer. Phys. Soc. 20, 1498 (1974).
6. Asay, J.R. and L.C. Chhabildas, HIGH PRESSURE SCIENCE & TECHNOLOGY, ed. by B. Vodar and Ph. Marteau, Pergamon Press, Vol. 2, 958-965 (1980).
7. Aidun, J.B. and Y.M. Gupta, *J. Appl. Phys.* **69**, 6998-7014 (1991).
8. Barker, L.M., SHOCK WAVES IN CONDENSED MATTER - 1983, J.R. Asay, R.A. Graham, G.K. Straub, editors, Elsevier Sci. Publ, B.V., 217-224 (1984).
9. Chhabildas, L.C and L.M. Barker, SHOCK WAVES IN CONDENSED MATTER – 1987, S.C. Sschmidt, N.C. Holmes, editors, Elsevier Sci. Publ, B.V. 111-114 (1988).
10. Hawke, R. D.E. Duerre, J.G. Huebel, J.G. Klapper, D.J. Steinberg and R.N. Keeler, *J. Appl. Phys.* **43**, 2734-2741 (1972).
11. Matzen, M.K., *Phys. Plasmas.* 4 (5), 1519- 1527 (1996).
12. Barker, L.M. and R.E. Hollenbach, , *J. Appl. Phys.* **45**, 4872-4887 (1974).
13. Asay, J.R., C.A. Hall, K.G. Holland, M.A. Bernard, W.A. Stygar, R.B. Spielman, S.E. Rosenthal, D.H. McDaniel, D.B. Hayes, "Isentropic Compression on Iron with the Z Accelerator", this proceedings.
14. Hayes, D.B., private communication (1999).
15. Chhabildas, L.C., and J.R. Asay, HIGH PRESSURE IN RESEARCH AND INDUSTRY, ed. By C. M. Backman, T. Johannisson and L. Tegner, Vol. 1, 183-187 (1982).