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PRECISE ULTRAHIGH-PRESSURE EXPERIMENTS

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A nuclear-explosive-generated shock has been used to obtain Hugoniot data at pressures corresponding to threefold compressions. The shock passed through a 12-mm-thick by 180-mm-diam molybdenum base plate and into 13 samples; 80 electrical-contact pins were used to determine shock velocities with uncertainties of 1.4 to 2.6%. The measured molybdenum shock velocity of 30.60 km/s corresponds to a pressure of 6.43 TPa, based on its SESAME equation of state. Impedance-matching analyses provided Hugoniot data in pressure (TPa)-particle velocity (km/s) coordinates (P,u) for the following materials: Al(2.93,27.57), 19%-porosity Mo(4.65,19.99), C(2.64,28.38), LiH(1.225,35.52), Cu(6.06,21.15), Fe(5.71,21.73), and crystalline quartz(2.75,26.76). These results indicate the need for improved theoretical calculations for some materials.

1. INTRODUCTION

For many years, dynamic methods have been used in conventional laboratory experiments[1] to obtain precise equation-of-state (EOS) data at pressures up to several hundred GPa (1 GPa=10 kbar $\approx 10^8$ atm). At these pressures, however, state-of-the-art theoretical predictions based on different models[2-6] often show rather small differences but may exhibit large variations at much higher pressures. Since the 1960's, nuclear-explosive-driven shocks have been used for experiments at ultrahigh pressures. Early Soviet work[7] stimulated the development of improved techniques, and in two rather recent experiments (referred to as I[8] and II[9]), we used the impedance-matching technique[1,9] to obtain Hugoniot data for 1 sample in I and 13 samples in II.

In these two experiments, the radiation energy from the explosion was used to produce shock pressures corresponding to nearly threefold compressions. The results indicated the need for improved theoretical calculations for some materials and stimulated interest in additional ultrahigh-pressure measurements, as an extension of this work, we fielded a similar nuclear explosive-driven experiment about a year ago.

2. EXPERIMENTAL SETUP AND TECHNIQUE

The configuration for the EOS experiment was nearly identical to that used in II[9]. The radiation energy from the explosion flowed by an indirect route to the position of the EOS package located ~ 3.5 m away, and extensive shielding was used to reduce the neutron and γ -ray backgrounds. In the package, 13 samples were positioned atop a 180-mm-diam molybdenum base plate as shown in Figure 1. The six outer stacks of 10-mm-thick samples contained the materials indicated in the figure and were arranged around a central 12-mm-thick sample of

uranium (^{235}U). A quartz gauge provided a measure of the energy deposition by background radiations, which were expected to heat the fissionable material by <100 K and the other samples by <10 K.

The small circles in Figure 1 show the x-y distribution of the 80 electrical-contact pins[9] used to determine shock arrival times. The pins were positioned so that rarefactions originating at edges would not perturb the shock front prior to its arrival at a given pin. Each pin produced a sharp pulse upon shock arrival at its base, and five such pin signals were multiplexed onto each of 16 uphole cables;

Figure 1: Schematic indicating the arrangement of the sample materials atop the molybdenum base plate and the positions of the pins.

different time constants provided a unique signature for each pulse. All signals were recorded as photographs of oscilloscope traces at sweep rates of ~ 100 ns/cm. A series of common timing (CT) recordings were made to relate the time on one trace to that on another; the CT uncertainties ranged from 0.5 to 2.5 ns.

3. EXPERIMENTAL RESULTS AND DATA ANALYSIS

All 80 pins produced very high quality signals from which the relative closure times were determined, with an overall uncertainty for each time of ~ 3 ns. The signal from the quartz gauge indicated a temperature rise of ~ 65 K for the fissioning samples -- in very good agreement with Monte Carlo predictions. In preliminary analyses, the interior pins were found to close before the corresponding pins located at larger radii, indicating that the shock front was not flat; however, these analyses indicated that shock-decay effects were minimal and that none of the pins behaved erratically.

The closure times t and pin coordinates (x, y, z) were then used as input variables in a least-squares fitting program to determine the shape and position of the shock front as a function of time. Different analytic functions for $t = t(x, y, z)$ were used to express the closure times in terms of the coordinates, and the coefficients and associated uncertainties resulting from the fits were used to determine the shape of the front and the shock velocities. Fits using different functional forms and subsets of pins indicated that the shock velocity varied by less than 1% radially across a small sample and that the radius of curvature was ~ 2 m.

To check for deviations from axial symmetry, terms linear in x and y were added to the fitting function, and the results of these fits implied an upper limit of 1.5 mrad on the tilt angle for the central axis of the shock front. As an additional check, the function for a tilted plane was independently fitted to the pin data for each sample stack and for the central sample. The coefficients from these fits indicated that the shock front at each sample position was tangent to an axially symmetric, spherically shaped front with a radius of curvature of ~ 2 m. The effect of this curvature is illustrated in Figure 2, which shows plots of the closure times as a function of the z coordinates for the pins in the copper and iron samples. The crosses indicate the experimental results, and the dashed lines are drawn to guide the eye. The curvature coefficient determined from a global fit was used to calculate the closure delay of each pin, and the corrected times are shown as open circles in the figure. The solid curves show the result of least-squares fits made using a function without an r dependence for the shock velocity. The scatter of the crosses is obviously greater than for the circles. Any effects associated with a decrease in the shock velocity are not evident.

To check for changes in velocity as the shock propagated, a term in z^2 was added to the fitting function. Several sets of fits were made with different constraints imposed on the velocity-change coefficient (δ_1) to reduce the number of free parameters (which nearly double when a separate z^2 is used for each sample). The best fits were obtained using a function in which the fractional change in the shock velocity was assumed to be inversely proportional to the velocity so that the correction was larger for a slower shock.

The least-squares fits using this function gave lower values of chi square than any others and indicated that the vertical decreases in shock velocities across the small samples were ~ 2.5 –6%. For the molybdenum standard, the results were consistent with a constant shock velocity; for the other samples, the uncertainties in the velocity decreases were 30 to 70%. These large uncertainties resulted in artificially large error bars for the derived shock velocities. Therefore, an additional series of fits were performed in which the δ_1 's were held fixed at various values in the range determined from the unconstrained fits, and the smallest value of chi squared was used to select the best fit. These fits implied a value of chi squared near 1.0 and indicated that the fitting function provided an accurate representation of the shockfront position vs time. The statistical errors from the best fits were combined with variations resulting from different functional forms to assign overall experimental errors of 1.4–2.6% to the derived shock velocities. These measured values and associated errors are summarized in Table I.

4. IMPEDANCE-MATCHING RESULTS

A graphical analysis based on the impedance-matching technique [9] and the measured interface shock velocities was used to determine a Hugoniot point for each lower layer sample.

Figure 2: Plots of closure time vs the vertical z -coordinate for the pins in the copper and iron samples; the solid curves are least-squares fits.

TABLE I. Comparison of Measured and Calculated Results

Material ^b	Density (g/cm ³)	Shock Velocity (km/s)		Difference(%) ^b		Hugoniot Points ^a	
		Measured	Error (%)	Old ^c	New ^c	Pressure (TPa)	Particle Velocity (km/s)
Mo Std	10.22	30.60	1.4	-----	-----	6.434(3.15)	20.57(1.75)
Mo	10.22	29.49	1.4	-----	-----	5.918(3.17)	19.64(1.78)
Al	2.700	39.41	1.6	-0.79	-0.63	2.933(3.26)	27.57(2.18)
Mo	8.240	28.21	1.7	2.62	2.23	4.647(3.59)	19.99(3.30)
						4.676(3.58)*	20.12(3.28)*
C	2.235	41.68	1.7	0.78	-----	2.644(3.31)	38.38(3.31)
LiH	0.700	49.27	2.6	0.66	-----	1.225(4.38)	35.52(2.96)
Cu	8.935	32.10	1.4	-2.64	-0.89	6.065(3.12)	21.15(2.62)
					1.12	-----	-----
Fe	7.85	33.47	1.4	-3.82	0.51	5.709(3.14)	21.73(2.60)
Quartz	2.645	38.87	2.4	3.23	-0.43	2.752(3.96)	26.76(2.77)
						2.894(3.85)*	28.16(2.56)*

^aBased on measured shock velocities and improved EOSs for standards (see text); percent errors in parentheses. Values flagged with an asterisk are based on original SESAME EOSs for the standards.

^b $100 \times (\Delta_{cal}/\Delta_{exp} - 1)$.

^cCalculated results using the original SESAME library (Old) and improved theories (New).

relative to the molybdenum standard and for each upper layer sample relative to the adjacent lower level material. Each graph consisted of plots in the pressure -- particle-velocity (P-u) plane of the experimentally determined Rayleigh line ($P = \rho_0 \Delta u$) for the upper material and the calculated release isentrope (RI) or reflected shock (RS) Hugoniot for the lower standard material, whose initial state was defined by its measured interface shock velocity. Similar plots using shock velocities that differed by one standard deviation for both materials defined a region of uncertainty in this plane and provided error bars for the Hugoniot point. The position of the calculated Hugoniot of the upper material on this graph provided a comparison with the measured point and gave a direct check on the consistency of the theoretical treatments used for the two EOSs.

The derived P-u Hugoniot points are given for the indicated materials in the last two columns of Table I, with percent errors shown in parentheses. The calculated shock velocities based on theoretical EOSs are compared with the measured values in columns 5 and 6. These comparisons are given as percent differences between the calculated (cal) and experimental (exp) results [$=100 \times (\Delta_{cal}/\Delta_{exp} - 1)$].

For the case in which a material served as a standard without being treated as an upper level sample, the Hugoniot data in Table I were derived from its theoretical EOS using the measured shock velocity and its associated uncertainty. The values given for the molybdenum (two cases) were obtained in this manner using the SESAME EOS(2) for material no. 2981, and the results reflect the P-u errors corresponding to the uncertainty in Δ .

Details of the analysis technique are illustrated for the copper sample and for the iron/quartz

stack in Figures 3 and 4, respectively. Each plot is on an expanded scale and shows only the region of interest around the measured point.

In Figure 3, the molybdenum RI ($\Delta = 30.60$ km/s) intersects the copper Rayleigh line ($\Delta = 32.10$ km/s) to define a Hugoniot point (circled) for copper. The region bounded by the long-dashed curves shows the uncertainty for this measured point. Calculated Hugoniots based on three different EOSs are also shown in the figure as heavy curves. The Hugoniot based on the original SESAME EOS (dashed, material no. 3330) barely passes through the error box. The other Hugoniots, shown as solid (material no. 3332) and as dot-dashed (material no. 3331) curves

Figure 3: Illustration of the impedance-matching analysis technique for the copper sample.

Figure 4: Illustration of the analysis technique for the iron(a) and quartz(b) samples.

are based on more recent improved theoretical treatments[3,4] and give much better agreement with experiment. The large dot indicates the predicted point based on the SESAME EOS for material no. 3332.

Figure 4 shows a similar comparison for the iron and the quartz samples. In (a), the calculated Hugoniot for iron (dashed) based on the original SESAME EOS (material no. 2140) lies outside the region of uncertainty. The heavy solid curve shows the calculated Hugoniot based on an improved theoretical treatment[5] and is in very good agreement with the experimental point (circled). The derived Hugoniot point for quartz based on the improved iron EOS is circled in (b). The SESAME based Hugoniot (heavy solid curve) for quartz (material no. 7380) is in good agreement with the experiment and results in the predicted point indicated by the large dot. A similar analysis based on the original SESAME EOS for iron (old) results in an experimental point at a considerably higher pressure (~2.9 TPa).

5. SUMMARY AND CONCLUSIONS

This experiment provides precise Hugoniot data at pressures nearly 10 times those reached using conventional laboratory techniques and at densities corresponding to threefold compressions. These data and those from similar earlier experiments[8,9] provide benchmarks for testing theoretical treatments in a pressure region where sizable differences can occur between predictions based on different models.

Results obtained using the SESAME EOS library[2] are in reasonably good agreement with the experimental results for many of the sample materials, especially when the EOS tables are based on improved[3,6] theoretical calculations. However, for the porous materials, predictions

of shock velocities are too high, implying that the Hugoniots for these materials need to be softer. Hopefully, these results will stimulate additional improved theoretical calculations and the development of more sophisticated EOS models that provide added insight into the physics at these extreme conditions.

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