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SHOCK WAVE EXPERIMENTS TO EXAMINE THE MULTIPHASE PROPERTIES OF CERIUM

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Abstract. There is a scientific need to obtain new data to constrain and refine next generation multi-phase equation-of-state (EOS) for metals. Experiments are needed to locate phase boundaries, determine transition kinetic times, and to obtain EOS and Hugoniot data for relevant phases. The objectives of the current work was to examine the multiphase properties for cerium including the dynamic melt boundary and the low-pressure solid-solid phase transition through the critical point. These objectives were addressed by performing plate impact experiments that used multiple experimental configurations including front-surface impact experiments to directly measure transition kinetics, multislug experiments that used the overtake method to measure sound speeds at pressure, and preheat experiments to map out phase boundaries. Preliminary data and analysis obtained for cerium will be presented.

Keywords: multiphase, equation of state, preheat
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INTRODUCTION

There is a scientific need to acquire new data to develop and constrain next generation, multiphase equations-of-state (EOS) models for metals. Experiments are needed to locate phase boundaries, to obtain EOS and strength data on the relevant pure phases, and information on transition kinetics. Because traditional shock wave experiments provide information on-Hugoniot, additional dynamic loading methods are required to access other regions of the phase diagram including shock-ramp, ramp, and preheat methods for example.

In this paper, some preliminary data are presented to illustrate current efforts to examine the multiphase diagram for cerium. Cerium was chosen for this work because it exhibits a complex phase diagram in a moderate stress regime (less than 250 kbar) with multiple phases at zero pressure and even more at higher pressures[1, 2, 3]. It also exhibits an anomalous melt boundary[4] and a low-pressure, solid-solid phase that ends in a critical point[5, 6, 7].

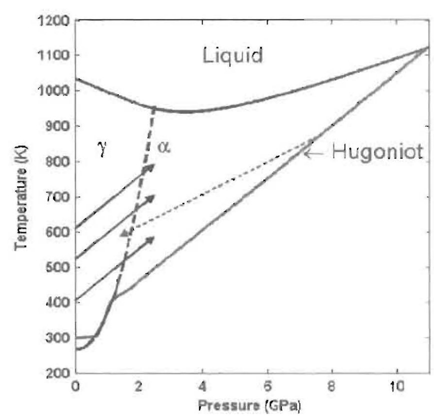

FIGURE 1. Schematic of the phase diagram for cerium showing the low-pressure phases. Also shown is the Hugoniot, possible paths achieved using the preheat experimental configuration (solid arrows) and the front-surface impact experiments (dashed arrow)

The large-volume collapse associated with this low-pressure transition is hypothesized to lead to a low-pressure melting transition[5, 6, 7]. The dynamic response of cerium was initially examined by Carter[8] where explosive flyer plate experiments were performed to obtain high-pressure Hugoniot data up to approximately one Mbar. Shock velocity data exhibited multiple features observed as kinks in the shock velocity at approximately 10 and 48 GPa and these were interpreted as a low-pressure solid phase transition and a higher pressure melt, respectively. Later work by Zhernokletov[9] obtained both Hugoniot and sound speed data and although the Hugoniot data were in good agreement with Carter, sound speed data did not show a melt transition at the higher pressures. Theoretically, significant progress has been made to develop multiphase diagram based on static diamond anvil cell data[10].

A schematic of the cerium phase diagram is shown in Fig. 1. The anomalous melt boundary and the low-pressure $\gamma - \alpha$ phase boundary are indicated. Several loading paths are shown to describe the three types of experiments performed in this work including: (1) standard shock wave experiment to obtain Hugoniot and sound speed data using the overtake method[11], (2) front-surface impact experiments to examine the $\gamma - \alpha$ phase during release (dashed arrow), and (3) preheat experiment to examine the $\gamma - \alpha$ boundary during compression (solid arrow).

MULTI-SLUG EXPERIMENTS

To obtain Hugoniot data and sound speed data that span the melt transition, multislug experiments were performed that used the overtake method[11] on a large-bore powder gun[12]. The experimental configuration is shown in Fig. 2 and consisted of an impactor backed by foam, a baseplate (same material as impactor), and a Ce-LiF sample assembly. Multiple VISAR and PDV probes were fielded to measure the shock velocity through the four Ce samples (1.5 to 5 mm thick) along with the particle velocity history at the Ce-LiF interface. In all experiments, the standard shorting pin method or PDV[13] were used to obtain accurate projectile velocity data at impact. Hugoniot and sound speed data were also obtained in the front-surface impact experiments discussed in the next section.

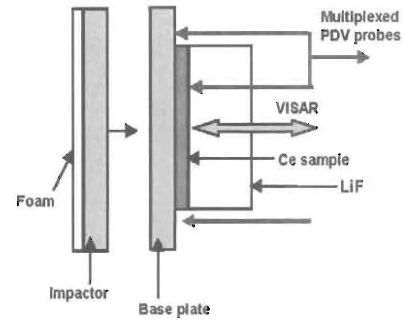

FIGURE 2. Experimental configuration for transmission experiments which consists of an impactor (backed by foam), a baseplate, and a cerium-LiF assembly. Multiple optical probes are used to measure the shock velocity, the shock wave tilt, and wave profile data.

To minimize the number digitizer channels used in these experiments, some of the probes (typically one baseplate free-surface probe and one Ce-LiF probe) were multiplexed together using 50-50 AC photonics splitters. Examples of PDV data obtained for one of the multi-slug Ce experiments are shown in Fig. 3 where the particle velocity (km/s) is plotted versus time (μs). Multiple shock events were observed including the shock arrival at the free-surface of the base plate, shock arrival at the Ce-LiF interface, and shock arrival at the free-surface of the LiF window. The wave profile for the Ce-LiF interface shows a shock-jump to a steady state followed by a release where the peak state time duration is observed to decrease with sample thickness. The PDV data were used to determine the shock velocity (2.415 km/s) through the samples and the standard impedance matching method (using the known impactor shock response) was used to determine the particle velocity (0.812 km/s) in the sample. These data provided the Hugoniot state within the material.

Because of the difficulty in determining the release arrival time from the PDV data, VISAR data were also obtained for each sample (1-ns resolution). Examples of the VISAR data are shown in Fig. 3 where the particle velocity (km/s) is plotted versus time (μs). The data are similar to the PDV data shown in Fig. 3 with a sharp shock jump observed followed by a steady state then the release which was observed to overtake the shock front with distance. The peak

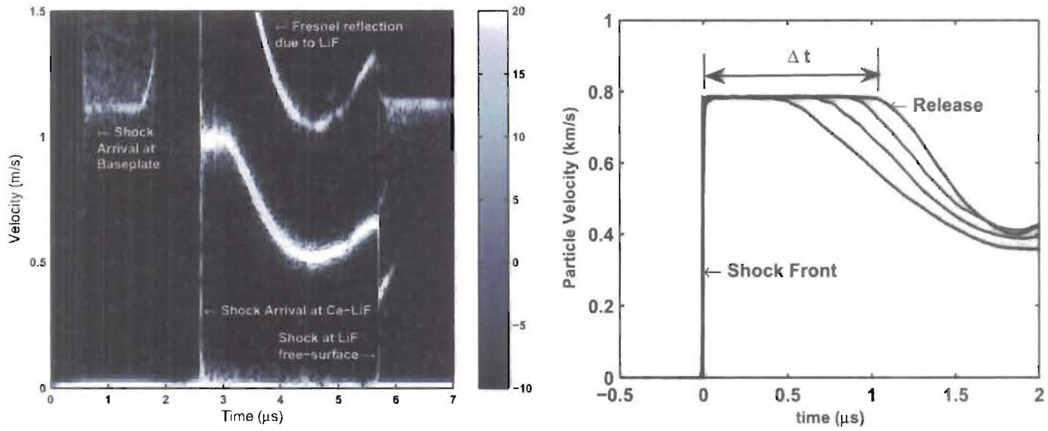

FIGURE 3. PDV and VISAR data obtained for a multislug Ce experiment which consisted of a copper impactor (3.908-mm thick) and base plate (3.915-mm thick) backed by Ce samples and LiF windows. The measured projectile velocity was 1.119 km/s. (left) PDV data from a 4.883-mm sample. Multiple shock events are observed including shock arrival at the baseplate, at the Ce-LiF interface, and at the back of the LiF window. (Right) VISAR data obtained from the four Ce samples with thicknesses of 1.523, 2.504, 3.325, and 4.883 mm.

state time duration for each sample was then plotted versus the corresponding sample thickness ,as shown in Fig. 4, to determine the overtake distance X_{max} where the time duration is equal to zero. To estimate the uncertainty in the X_{max} a Monte-Carlo method was used to calculate all possible least-square fits to the data assuming uncertainty values of approximately ± 2.5 μm and ± 5 ns for the thickness and time duration, respectively. The intercept values for each line was binned into a histogram the mean was taken as the value for X_{max} and the uncertainty equal to the one-sigma width of the distribution (see Fig. 4 inset) The sound speed is then related to the shock velocity U_s (obtained from PDV data) and the overtake distance through the equation[9],

$$C = X_{max} \left[t_{sw} - tr + \frac{X_{max}}{U_s} \right]^{-1} \qquad (1)$$

where t_{sw} and t_r are the shock wave arrival time and the release arrival time at the Ce sample. These values were obtained from a wave propagation code that incorporated models for the shock response of the baseplate and impactor materials and the measured impact velocity. For the data shown in Fig. 3 and Fig 4, the values for t_{sw} and t_r were 0.8208 and 2.308 μs, respectively. Using Eq. 1, the sound speed at pressure was determined to be 4.713 km/s. Additional data were obtained and analysis is currently underway to determine the solid and liquid Hugoniots and the melt transition stress.

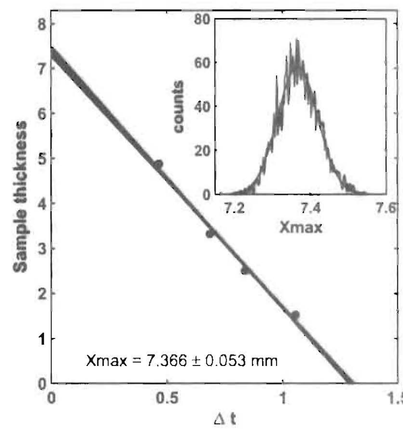

FIGURE 4. Plot of sample thickness versus peak state time duration to determine the overtake distance X_{max}s. The measured values for Δt were determined to be 1.0544, 0.8348, 0.6832, and 0.4644 μs from the VISAR data. The corresponding thickness values were shown in the caption of Fig. 3.

FRONT-SURFACE IMPACT EXPERIMENTS

Front-surface impact experiments provide a simpler method for examining phase transitions while providing needed Hugoniot data because the shock wave is directed into the impactor away from the measurements interface[14]. The experimental configuration consists of Ce impactor (sample) backed by foam impacting a LiF optical window front-plated with aluminum. PDV and VISAR were used to obtain the particle velocity history at the impact surface. This configuration was used to obtain both Hugoniot data and to examine the $\gamma-\alpha$ transition during release.

Two examples of VISAR data obtained for front-surface impact on Ce are shown in Fig. 5 where the particle velocity is plotted versus time. The front-surface impact data show a sharp jump to a steady state followed by release. During release, a rarefaction shock was observed likely due to the reverse transition from the α to the γ-phase. These data compare well to simulated wave profiles obtained from a multiphase EOS for Ce that includes the low-pressure solid-solid phase transition[15]. The peak particle velocity u_p^m obtained from the velocimetry data coupled with the longitudinal stress P_x determined from the known Hugoniot for the window material and the measured projectile velocity V_p, the shock velocity U_s is given by the Rankine-Hugoniot jump conditions[16]. In addition, sound speeds at pressure can be obtained using the quation

$$C = \left[\frac{\Delta t}{L} - \frac{1}{U_S} \right]^{-1} \quad (2)$$

where L is the sample thickness and Δt is the duration of the peak state obtained from the velocimetry data. The sound speed values obtained in this way have large uncertainties mostly because of the larger uncertainty in the calculated shock velocity.

PRE-HEATED TRANSMISSION EXPERIMENTS

A pre-heat target capability was developed for transmission shock wave experiments. This experimental configuration was used to examine the γ-α boundary upon compression and to obtain elevated temperature Hugoniot data. The preheat experimental configuration is shown in Fig. 6 (left) and consisted of an aluminum impactor impacting an aluminum base plate backed by the Ce-LiF assembly. The base plate was integrated into the heater design (typically 1-2 mm in thickness) to provide an oven-like environment so that the sample temperature was as uniform as possible. A 400 watt resistive ring heater was used to heat the target to temperatures up to approximately 300 degrees celsius and a standard omega thermocouple placed in the aluminum wall near the sample was used to measure the initial temperature of the sample. An important requirement for the heater system was the ability to uniformly heat the sample to the desired temperature. To confirm this, a thermal imaging camera was used to monitor the target/sample while it was heated to the maximum expected temperature (262 C). For all temperatures, the sample temperature was observed to be uniform within approximately 1 degree C or less. Both configurations used VISAR and PDV to obtain the particle velocity history at the Ce-LiF interface and either the standard shorting pins or PDV were used to measure the projectile velocity at impact.

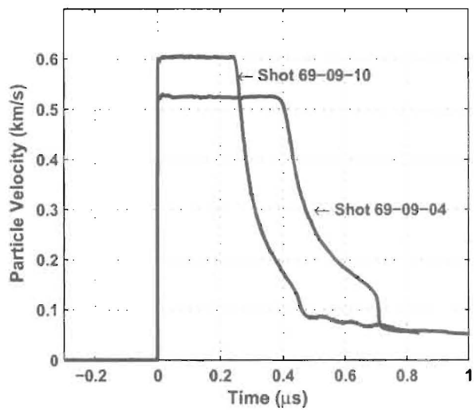

FIGURE 5. Wave profile data obtained from two front-surface impact experiments that show a shock to a steady state followed by release. A reversion shock is observed during release due to the transition from the α to the γ phase

Examples of VISAR data obtained for two of the pre-heated experiments are shown in Fig. 6 where the particle velocity is plotted versus time. The pre-heated data show a ramp-like behavior for the room-temperature experiment followed by a shock as the

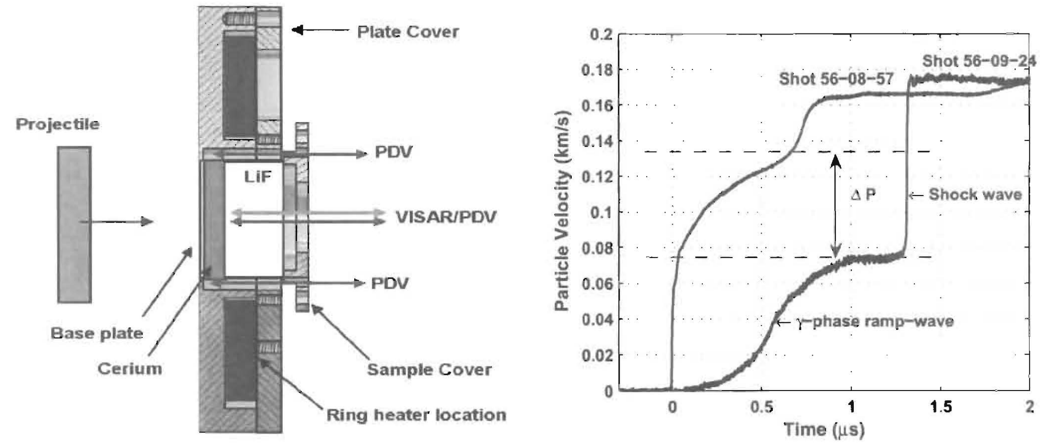

FIGURE 6. (Left) Experimental configurations for pre-heated transmission experiments. (Right) Two examples of velocimetry data obtained from for experiments at room temperature (Shot 56-09-24) and at a preheat temperature of 262 C (Shot 56-08-57).

stress exceeds the phase transition stress. The second wave profile obtained at a preheated temperature of 262 degrees Celsius shows a similar wave profile though the peak of the ramp wave occurs at significantly higher velocities indicating a higher transition stress for the higher temperature initial conditions. Additional data have been obtained and efforts are underway to analyze these data using the recently developed multiphase EOS.

To examine the effect of the preheat temperature on the window correction factors for VISAR and PDV, one experiment was performed (similar to the configuration in Fig. 6) which consisted of an aluminum impactor and baseplate backed by a LiF window. The initial temperature was set at 262 degrees Celsius and the measured projectile velocity was 0.345±0.005 km/s. The peak particle velocity was approximately 0.174±0.003 km/s comparable to the value predicted from the known shock response for aluminum and LiF[17, 18]. Thus, it is concluded that the window correction values for PDV and VISAR[13] do not change significantly with temperature up to 262 degrees Celsius. This is not a surprising result given that the window correction was determined up to shock stresses that exceed 20 GPa which would result in significant shock heating.

SUMMARY

Some preliminary experimental efforts to examine the dynamic multiphase properties of Ce have been presented. These efforts include multislug experiments on the large-bore powder gun[12] to obtain Hugoniot data and sound speeds at pressure. These experiments used PDV to measure the shock transit time through the Ce and to obtain wave profile data at the Ce-LiF interface. Additional experiments have been conducted to date and analysis of these data are underway to determine the solid and liquid Hugoniots for Ce and to determine the onset of the melt transition.

The phase boundary for the low-pressure, solid-solid transition ($\gamma - \alpha$) was examined for release and compression using front-surface impact and preheated experiments, respectively. Velocity data obtained from the front-surface impact experiments showed a reversion shock because of the reverse phase transition during release. This has been confirmed by comparing the velocity profiles to predictions obtained from the multiphase EOS[15]. To examine the phase boundary more directly during compression, a new heating capability was developed to preheat the samples prior to shock loading. The pre-heat data show a strong dependence of the transition stress on initial temperature and one experiment performed without the Ce sample provide some evidence that the window correction factors for PDV and VISAR do not change significantly with temperature. Additional experiments for a range of temperatures are needed to confirm this. Efforts are underway to use a multiphase equation-of-state for Ce to analyze both the front-surface impact experiments[15] to determine the location of the $\gamma - \alpha$ transition. Future work is underway to use the preheat configuration to obtain elevated temperature Hugoniot response in an attempt to examine the anomalous melt boundary.

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