Sound speed measurements in zirconium using the front surface impact technique

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Abstract. We have performed a series of experiments impacting zirconium samples of varying purity level directly onto lithium fluoride (LiF) windows to determine both the Hugoniot and sound speed as a function of stress up to 70 GPa. This front surface impact (FSI) geometry is useful for determining sound speed in shock-compression experiments because wave interactions are mostly eliminated and multiple sample thicknesses are not needed in each experiment. The experimental results show two kinks in the sound speed which correlate well with the location of the $\alpha \rightarrow \omega$ and $\omega \rightarrow \beta$ transitions, respectively. A rarefaction shock also forms in the release wave in experiments conducted at 31 GPa giving further evidence that this phase transition is being observed.

1. Introduction

Kinks in $U_S - u_p$ data have been attributed to the presence of shock-induced solid-solid phase transitions for several metals including iron, tin, titanium, and zirconium (see data reproduced in Ref. [1]). In some cases, these transitions lead to a splitting of the initial shock wave into multiple waves, which has been observed using velocimetry techniques [2, 3]. However, the observation of multi-wave structure due to a shock-induced phase transformation may not be possible and kinks in the $U_S - u_p$ curve can be subtle if the volume change between the parent and daughter phases is small. This has been shown to be the case for the solid-liquid transition in copper [4], for example. In this case and others [5, 6], the determination of the longitudinal sound speed as a function of shock stress or particle velocity has been used to locate the intersection of the melt boundary with the Hugoniot.

Zirconium has three solid phases, $\alpha$ (hcp), $\omega$ (hex-3), and $\beta$ (bcc), which can all be accessed through shock compression experiments. In this work, we measure the longitudinal sound speed as a function of shock stress in zirconium through all three phases to determine if solid-solid phase boundaries can also be located using this technique.

2. Experimental Method

Zirconium samples were directly impacted onto lithium fluoride (LiF) windows as shown in Fig. 1 and the particle velocity at the sample/window interface was measured using VISAR [7] and/or PDV [8]. The two materials investigated here are the two materials identified as ZrI and ZrII in previous work [3], and the impurities of each material are given in that reference with the ZrII material being the least pure of the materials studied.
Zirconium samples were accelerated using both single and two-stage gas and powder driven guns at Los Alamos National Laboratory to velocities ranging from 0.2 to greater than 4.5 km/s and produced stresses from 1.8 to nearly 69 GPa spanning all three solid phases of zirconium. A thin coating (~400 nm) of aluminum was vapor deposited on the impact surface of the LiF window to obtain a reflective surface for the velocimetry measurements. Previous studies using the Front Surface Impact (FSI) geometry – also referred to as reverse impact or reverse ballistic experiments – have used a buffer [9] or foil [10] glued onto the impact side of the window to preserve the integrity of the reflective surface of the window. However, this can introduce unwanted effects such as the development of multi-wave structure from the elastic precursor of the buffer material or ring-up of the glue bond between the buffer and window. We found that polishing the impact surface of the zirconium impactor was quite effective in preserving the integrity of the mirror after impact and allowed us to measure the true impact state between the sample and window.

3. Experimental Results

Eleven FSI experiments were performed on each material. Figure 2 shows the wave profiles obtained using either VISAR or PDV for a representative sample of the total set of experiments. The values of particle velocity, \( u_p \), shock velocity, \( U_S \), stress, \( P \), and density, \( \rho \), in the shocked zirconium samples were determined from the impedance matching solution between the window with known Hugoniot and the impactor. The impedance matching solution is easily derived from the \( P - u_p \) diagram shown in Fig. 3(a). The Hugoniot for the window (at rest) is represented by the blue line while the Hugoniot for the impactor traveling to the right (as in Fig. 1) at a velocity, \( u_d \), is represented by the red line. At impact, both the stress, \( P^w \), and particle velocity, \( u^w_p \), are the same on either side of the sample/window interface. Thus, given the measured value of \( u^w_p \) using VISAR or PDV, the stress at the impactor/window interface is obtained directly from the LiF Hugoniot [11]:

\[
P^w = \rho^w_0(C^w + S^w u^w_p)u^w_p,
\]

where \( \rho^w_0 = 2.64 \text{ g/cm}^3 \), \( C^w = 5.148 \text{ km/s} \), and \( S^w = 1.353 \).

The general Hugoniot jump condition for conservation of momentum gives the following relationship in Lagrangian coordinates [12]:

\[
P_i - P_{i-1} = \rho_0 U_{S_{i}}(u_{pi} - u_{pi-1})
\]

where \( P_i \), \( U_{S_{i}} \), and \( u_{pi} \) are the stress, shock velocity, and particle velocity in the \( i \)th shocked state and \( \rho_0 \) is the initial density of the material. From this equation, it can be shown that the
Figure 2. Wave profiles from selected experiments on (a) Zr\textsubscript{I} and (b) Zr\textsubscript{II}. The longitudinal sound speed for each experiment is determined using the time of arrival difference between the leading edge of the rarefaction wave (\(t_3\)) and the shock (\(t_0\)).

The solid red curve in Fig. 3(a) can be represented by the equation,

\[ P - P_1 = \rho_0 U_S(u_d - u_{p1} - u_p), \]  

(3)

where \(P_1\) and \(u_{p1}\) are the stress and particle velocity in the initial shocked state (at the Hugoniot Elastic Limit, for example) and (in the figure) \(u_{p1}^* = u_d - u_{p1}\) where \(u_d\) is the projectile velocity. Since the point \((u_{p1}^*, P^w)\) is known from the experiment and window Hugoniot (Eq. 1), we can now obtain an expression for the shock velocity of the impactor in the final state:

\[ U_S = \frac{P^w - P_1}{\rho_0(u_d - u_{p1} - u_{p1}^*)}. \]  

(4)

Determination of the longitudinal sound speed is done with help of Fig. 3(b) where the times \(t_0\) and \(t_3\) are the arrival times of the shock and rarefaction waves, respectively, as determined from the experimental velocimetry record (see Fig. 2(b)). Again while it is not observable in the FSI experiment, we must know whether multiple waves exist in the impactor and the properties of the initial state of the material to perform the calculation correctly. From Fig. 3(b), The longitudinal sound speed in Lagrangian coordinates is given by,

\[ C_L = \frac{h - h_1}{(t_3 - t_0) - \frac{(h - h_1)}{U_S}} \]  

(5)

where

\[ h_1 = h \frac{C_{L1}(U_{S1} - U_S)}{U_{S1}(C_{L1} + U_S)} \]  

(6)

is the transit distance of the rarefaction through the initial state of the material, \(h\) is the impactor thickness, \(U_{S1}\) and \(U_S\) are the shock velocities in the initial and final states, respectively, and
Figure 3. (a) Graphical representation of the impedance matching solution to the Front Surface Impact experiment. The blue line represents the window Hugoniot which is at rest before impact. The red line represents the impactor Hugoniot with an initial velocity of \( u_d \). The dashed section represents the Hugoniot of the initial state of the material while the solid section represents the Hugoniot of the final state of the material with the transition between the two states occurring at \((u_{p1}, P_1)\). This is representative of a material with an elastic-plastic or structural phase transition. Note that \( u_{p1}^* = u_d - u_{p1} \). (b) Graphical representation of the propagation of shock and rarefaction waves through the sample. The blue and green solid lines represent the first and second shocks, respectively, while the red line represents the rarefaction traveling through the first (dashed) and second (solid) shocked states. For simplicity, only the leading edge of the rarefaction is shown.

\( C_{L1} \) is the longitudinal sound speed in the initial state of the material. In order to calculate \( U_S \) and \( C_L \) from the FSI experiment, we must first determine \( P_1 \), \( u_{p1} \), \( U_{S1} \), and \( C_{L1} \) by other means. These quantities were determined by re-analyzing experiments performed previously [3] and assuming that \( C_{L1} = U_{S1} \) since \( C_{L1} \) could not be determined independently.

The Eulerian shock velocities and sound speeds determined from all FSI experiments performed are shown against the particle velocity in Fig. 4, where the conversion from the Lagrangian to Eulerian frame was done in the usual way:

\[
V^E = \frac{\rho_0}{\rho} V^L, \\
\tag{7}
\]

where \( V \) represents either the shock velocity or sound speed, \( \rho_0 \) is the initial density, \( \rho \) is the density in front of the wave, and the superscripts, \( E \) and \( L \), represent the Eulerian and Lagrangian quantities, respectively. One standard deviation error bars were calculated via Monte Carlo error propagation [13]. Also shown is a linear fit to the \( U_S - u_p \) data (solid blue line) with 95% confidence bands (dashed blue lines) which capture most of the data reasonably well and kinks due to phase transitions are not obvious. However, based on the size of the error bars obtained here, this technique is less accurate than the more direct methods typically used to determine shock velocity in plate impact experiments [14].

In sharp contrast, the sound speed data show obvious breaks at two locations for each data set as indicated in the figure. The first break appears at different particle velocities (and, therefore,
stresses) for the two different materials. This is consistent with observations seen previously [3] that the transition stress of the \( \alpha \) to \( \omega \) phase depends on material purity. The second break in the sound speed is a very abrupt drop at a particle velocity near 1 km/s. We are attributing this to the transition from the \( \omega \) to the \( \beta \) phase. Further evidence that this transition is taking place can be seen in Fig. 2. The green traces in this figure are for experiments performed at a stress of 31 GPa and both show a definite overshoot and subsequent relaxation at impact as well as the formation of a shock in the rarefaction wave. The first feature may be an indication that the kinetics of this transformation are quite slow, while the second feature indicates that a reversion from the \( \beta \) to the \( \omega \) phase is occurring upon release of the stress at the impactor/window interface. As before [3], no rarefaction shock was observed for experiments performed just above the \( \alpha \to \omega \) transition indicating that reversion of the \( \omega \) phase may not occur on these time scales.

4. Conclusions
We have performed experiments on zirconium samples in the FSI geometry and have determined the shock velocity and sound speed as a function of particle velocity for all experiments. This technique minimizes the wave interactions occurring at the point of measurement and can therefore allow the observation of phenomena that may be obscured by wave interactions otherwise. However, for materials that are known to have an unsteady shock condition (i.e. a single shock splits into multiple shocks due to elastic-plastic deformation and/or a phase transition), knowledge of the properties of these initial states (which are unobservable in this geometry) are needed to calculate the shock velocity and sound speed. Regardless of the technique used, determination of the sound speed as a function of particle velocity (or stress) has proven quite effective in locating intersection of the melt boundary with the Hugoniot (see, for example, the work in Refs. [4–6]). Here, we have also demonstrated that it can be a powerful way to determine the location of solid-solid phase boundaries as well.

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