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July 11, 1979

This paper was prepared for submittal to the Proceedings of the Seventh AIRAPT International High Pressure-Conference, Le Crusot, France, July 30 to August 3, 1979.

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ELASTIC AND PLASTIC PROPERTIES OF URANIUM DIOXIDE FROM 5 TO 330 GPa*

W. H. Gust

University of California, Lawrence Livermore Laboratory
Livermore, CA 94550

ABSTRACT

We have measured the shock-compression parameters for UO₂ to 330 GPa. The Hugoniot elastic limit was found to be 5.7 GPa. Evidence for a shock-induced phase transition was observed at about 54 GPa.

INTRODUCTION

The study of the properties of solid and molten uranium dioxide (UO₂) is of considerable current interest in the field of liquid metal, fast-breeder-reactor excursion analyses. A fundamental requirement is the determination of reliable shock-compressed pressure-volume data for full-density fuel material. These data are essential for the extrapolations that permit calculations of material behavior during the transition from a dense solid to a molten liquid. A reliable equation of state that includes these phenomena provides the necessary link between the coupled hydrodynamic and neutronics codes.

PROCEDURE

Several shock-wave measurement techniques are capable of resolving multiple-wave shock structures up to about 50 GPa. Among these are the capacitor, manganin gage, inclined mirror, and inclined prism techniques. We used inclined prisms [1, 2] to about 50 GPa to resolve the two-wave shock structure related to the Hugoniot elastic limit (HEL) of UO₂. Experiments at plastic-wave velocities great enough to overdrive the elastic wave (50 to 150 GPa) were done with flash gap systems [3] (reverberations within the high velocity, explosively driven, flying plates used to obtain the high pressure made the use of inclined prisms inappropriate). Data from 150 to 330 GPa were obtained with a two-stage light-gas gun [4].

Use of inclined prisms to resolve multiple waves is not appropriate in experiments where compression is obtained through use of explosively driven flying plates. This is because reverberations within the explosively driven flyer may launch additional extraneous waves into the sample and may cause error. Hence in this pressure regime, the flash-gap technique [3], which is relatively insensitive to the flyer plate reverberations, was used. If the material transforms in this

*Work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore Laboratory under contract No. W-7405-Eng-48.
regime, the usual procedure is to identify the transition point through discontinuities in plots of shock-compression variables, e.g., shock velocity vs particle velocity.

In the experiments done on the gun, 1.5 mm-thick plates of Ta were impacted on 25.4-mm-diam by 3.2-mm-thick UO₂ samples. Compression parameters were determined through impedance matches that were centered on the Hugoniot for Ta [5], with Ta particle velocity equal to one-half the projectile velocity. Total uncertainty in the measurements is estimated to be less than 1.5%.

For multiple waves, stress and volume are given by

\[
\sigma_n = V^{-1}_{n-1} (U_{s_n} - U_{p_{n-1}}) (U_p - U_{p_{n-1}}) + \sigma_{n-1}
\]

and

\[
V_n = V^{-1}_{n-1} (U_s - U_{p}) / (U_{s_n} - U_{p_{n-1}}).
\]

The particle velocity for the elastic wave was obtained from the free-surface approximation, i.e., \( U_p = U_{51} \). For the experiments done with inclined prisms, \( U_p \) was determined from the free-surface approximation and from impedance matching.

MATERIALS

High-purity samples of uranium dioxide (UO₂) actually were used in these experiments. The uranium was obtained as a precipitate from a uranyl nitrate solution. The precipitate gel was calcined to UO₃, after which the UO₃ was reduced to UO with hydrogen at about 675 K. The UO₂ powder was then isostatically cold pressed at 100 MPa, followed by sintering at 2075 K for 2 h.

This process produced samples that were from 95 to 97% of theoretical density (10.86 Mg/m³). Spectrographic and chemical analyses on typical samples indicated about 80 other elements were present in very small amounts, i.e., a few parts per million or less. Total impurities present were less than 0.015 at.%.  

RESULTS

The results are displayed in Fig. 1. The Hugoniot elastic limit was found to be about 5.7 GPa. Linear relations between shock and particle velocities were found for two regimes. The low-pressure data can be described by \( U_s = 3.88 + 1.11 (U_p - 0.3) \) for \( 0.3 < U_p < 1.05 \) km/s. At higher pressure, the plot displays a discontinuity for \( 1.05 U_p < 1.8 \) km/s. The final segments are described by \( U_s = 5.30 + 1.28 (U_p - 1.8) \) for \( 1.8 < U_p < 4.0 \) km/s.

DISCUSSION

The results in Figs. 1 and 2 indicate that UO₂ exhibits a strong, relatively fast, elastic wave under shock compression. The amplitude of the Hugoniot elastic limit (5.7 GPa) was obtained from four excellent photographic records; all four displayed changes in slope that are a characteristic of two-wave shock structures (see Fig. 2, a typical example). The velocity of the elastic wave was in good agreement with our ultrasonically measured, longitudinal sound speed at 1 atm (5.26 vs 5.19 km/s). This measurement was made on samples used in the inclined-prism experiments. Samples used for the high-pressure gun experiments were about 1% less dense but had drastically different elastic properties (\( c_l \approx 4.71 \) km/s).
In the $U_{S1}$-$U_p$ plot (Fig. 1), the line segment that represents the low-pressure regime does not extrapolate to bulk sound speed at $U_p = 0$ km/s, which is an indication of irregular behavior at low pressure. Good agreement between $C_l$ and $U_{S1}$ indicates that third-order elastic effects are probably not responsible.

For normal materials in the low-pressure regime, the particle velocity obtained from an impedance match with the plastic wave is very nearly equal to or slightly less (1%) than half the free-surface velocity, i.e., $U_p = U_{S2}/2$. However, data for systems A to D indicate that $U_{S2}$ is 14 to 20% less than $U_p$, thus providing additional evidence of anomalous behavior in that regime.

At higher pressure, the plot displays a discontinuity for $1.1 < U_p < 1.8$ km/s ($\sigma = 52.5$ GPa). This is probably a manifestation of a phase transition. Such discontinuities are usually considered to be related to unusual material behavior, i.e., phase transitions, and represent a mixed-phase region. Uranium dioxide has the cubic fluorite crystallographic structure. It may be recalled that several substances with this structure transform to the orthorhombic PbCl$_2$ structure under static high pressure [6]. For example, this transition occurs between 2.7 and 11.0 GPa for BaF$_2$, CaF$_2$, SrF$_2$, and EuF$_2$ [7, 8].

For $2.1 < U_p < 2.3$ km/s, our data agree fairly well with those of Goplen [9]. In this regime, the elastic wave is overdriven so that flash gap results are valid. The discontinuities at 1.9 to 2.0 km/s probably mark the points where the phase transitional waves have been overdriven.

The final part is described by $U_p = 5.28 + 1.28 (U_p - 1.8)$ km/s for $1.8 < U_p < 4.0$ km/s. This portion does not extrapolate back to $C_p$, the bulk sound speed at $U_p = 0$, which provides additional evidence that at least one phase transition has occurred. In this regime, both the elastic and phase transitional waves have been overdriven so that the curve may be validly extrapolated to higher values.
Our low-pressure results shown in Fig. 1, do not agree well with those of Goplen [9] and of Allen et al. [10]. The reason apparently is that their results are erroneous, because they did not use measuring techniques capable of resolving strong multiple-wave shock structures.

Goplen's measurements were done with the flash gap technique. In the presence of a strong elastic wave, the flash gap measurements usually indicate values that are too low until the shock is of sufficient strength to overdrive the elastic wave.

Goplen's low-pressure results (0.6 < \(U_p\) < 2.0 km/s) apparently fall into this category. His results for 2.2 < \(U_p\) km/s, where the elastic wave is overdriven, agree with ours. His data, however, suffer from the fact that he was unable to obtain pressures high enough to completely overdrive the phase transitional wave, and his final results were obtained in a mixed phase regime. Hence his extrapolation based on his highest pressure data and on sound speed at zero pressure (\(U_s = 4.4 + 0.8 U_p\)) for crystal-density material is not valid (sound speed for the transformed material is not known).

The capacitance-technique experiments performed by Allan et al. [10] were not done at high enough pressure (4.2 GPa) to exceed the Hugoniot elastic limit and, therefore, did not display two wave-shock structures. Similar experiments with a final pressure twice as great should show it.

The electrical-shorting-pin technique used in their high explosively driven experiments was not capable of resolving two wave-shock structures. Their recorded data indicate that the shock velocity measured in each case was probably for the elastic wave. In no case does the recorded shock velocity exceed their ultrasonically measured longitudinal sound speed (5.147 km/s). The elastic wave was evidently strong enough to trigger the shorting pins. Needless to say, the particle velocity obtained from a single wave impedance match with the brass standard and the elastic wave is incorrect, because it does not account for the plastic wave.

REFERENCES

NOTATION
\[ CB = \text{bulk sound velocity} \]
\[ CL = \text{longitudinal sound velocity} \]
\[ CS = \text{shear sound velocity} \]
\[ n = \text{order of successive waves} \]
\[ U_p = \text{particle velocity} \]
\[ U_{fs} = \text{material free-surface velocity} \]
\[ U_s = \text{shock velocity} \]
\[ V = 1/p = \text{volume} \]
\[ \rho = \text{density} \]
\[ \sigma = \text{stress} \]
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