Journal of Physics: Conference Series **500** (2014) 032003 doi:10.1088/1742-6596/500/3/032003

The influence of peak shock stress on the high pressure phase transformation in Zr

E K Cerreta, F L Addessio, C A Bronkhorst, D W Brown, J P Escobedo, S J Fensin, G T Gray III, T Lookman, P A Rigg and C P Trujillo

Los Alamos National Laboratory, Los Alamos, New Mexico, 87545, USA

E-mail: ecerreta@lanl.gov

Abstract. At high pressures zirconium is known to undergo a phase transformation from the hexagonal close packed (HCP) alpha phase to the simple hexagonal omega phase. Under conditions of shock loading, a significant volume fraction of high-pressure omega phase is retained upon release. However, the hysteresis in this transformation is not well represented by equilibrium phase diagrams and the multi-phase plasticity under shock conditions is not well understood. For these reasons, the influence of peak shock stress and temperature on the retention of omega phase in Zr has been explored. VISAR and PDV measurements along with post-mortem metallographic and neutron diffraction characterization of soft recovered specimens have been utilized to quantify the volume fraction of retained omega phase and qualitatively understand the kinetics of this transformation. In turn, soft recovered specimens with varying volume fractions of retained omega phase have been utilized to understand the contribution of omega and alpha phases to strength in shock loaded Zr.

1. Introduction

Under shock loading conditions, zirconium has been observed to undergo a phase transformation from hexagonal close packed (HCP) α-phase to simple hexagonal ω-phase [1, 2]. This phase transformation displays a hysteretic behavior [3, 4]. Specifically, upon shock-loading zirconium to stresses in excess of the phase transformation stress (7 GPa) and then unloading to ambient pressure, soft-recovered specimens have been observed to retain as much as 40% omega phase [5]. Many studies performed to examine this behavior have primarily focused on determining the pressure for the phase transformation, crystallography of the transformation, subsequent mechanical properties, and electronic structure calculations [6-10]. While the post-mortem nature of the retained ω-phase has been characterized, there are a number of unaddressed questions with regard to this high-pressure phase transformation. For example, few studies have focused on the role of dynamic drive condition on the hysteresis of the phase transition.

Currently, material models material models do not represent the physics of the relationship between microstructure and loading conditions well enough to enable predictive representation of non-equilibrium, extreme event processes such as phase transformation under dynamic loading conditions. Traditional polycrystalline models have been based on heterogeneous response at the multi-crystal length scale but have relied on continuum single crystal theories, which incorporate plastic deformation processes like dislocation slip and twinning in a highly homogenized way. A robust model for high-pressure phase transformations would account for the coupled physical processes of

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
Published under licence by IOP Publishing Ltd

plasticity and solid-solid phase transformations in materials. To do this, the hysteresis and meta-stability of the α- to ω-phase transition needs to be characterized in a way that determines microstructure and allows for the development of models that are not based on the concept of an "equilibrium" phase boundary. As such, the phase diagram under dynamic conditions, with hysteretic windows, must be mapped and microstructure evolution due to phase change, deformation twinning, and plasticity understood.

To address this, a study has been performed that examines the role of dynamic drive condition on the α- to ω-phase transformation in high-purity Zr. Plate impact experiments were conducted to peak stresses of 8 and 10.5 GPa at room temperature and at 200 °C to examine the role of peak stress and temperature on the transformation. *In situ* particle velocity measurements were made and all specimens were soft recovered for post-mortem analysis. Quantitative measurements of the volume fraction of omega phase retained upon unloading and the texture evolution of the α-phase were performed via neutron and x-ray diffraction. Subsequent properties of the shock-loaded specimens were examined via quasi-static reload experiments.

The results of this study indicate that the α to ω high-pressure phase transformation is particularly sensitive to small changes in dynamic drive condition. Additionally the subsequent strength of the material due to both plastic and phase transformation processes is altered as a function of this drive condition. These data yield insight toward an advanced understanding of the dynamic rather than equilibrium phase diagram for Zr and in this way lend important constraints for the development of phase aware strength and damage models.

2. Experimental techniques

This investigation was performed on high-purity, crystal bar grade Zr. Table 1 gives the chemistry for this material and processing details are provided elsewhere [5]. To investigate the influence of peak shock stress on ω-phase formation, wave profile and shock recovery experiments were conducted. All shock recovery experiments were performed on an 80 mm-single-stage launcher utilizing a shock assembly consisting solely of Zr. Room temperature (RT) wave profile experiments were conducted on a 40 mm single-stage launcher and the wave profiles were measured with a velocity interferometer system for any reflector (VISAR) built at Los Alamos [11]. The precision of the wave velocity measurements is estimated to be approximately 1% in particle velocity. Photomultiplier circuits were utilized that had a 1 ns rise time. These experiments were backed with a poly-methyl methacrylate (PMMA) window. All elevated temperature wave profile experiments were performed on the 80 mm single stage gas launcher and wave profiles were measured using a photon Doppler velocimetry (PDV) diagnostic. No window was utilized in these experiments.

Table 1: Chemistry of the Zr in wt% PPM.

Material	O	C	N	Al	V	Fe
High Purity Zr	<50	22	<20	<20	<50	<50

Neutron and x-ray diffraction were performed to characterize the as-annealed and shocked loaded materials. Neutron diffraction experiments were performed on the High Intensity Powder Diffraction (HIPD) instrument at the Los Alamos Neutron Scattering Center (LANSCE). X-ray diffraction experiments were performed on the 1ID-C beamline at the Advanced Photon Source (APS) at Argonne National Laboratory. Bulk specimens were examined without special preparation because the penetration is large enough to sample the entire specimen.

Post-shock mechanical properties were examined through quasi-static compression testing. Right-regular cylinders, 5 mm in height and 5 mm in diameter were sectioned from soft recovered, shock-loaded specimens. The loading axes of the compression specimens were aligned with the shock

direction of the recovered materials. Quasi-static tests were performed at room temperature and a strain rate of 10^{-3}/s to strains of approximately 15%.

3. Results and discussion

3.1. Wave profiles

The room temperature shock-wave-profile experiments conducted on Zr were performed to quantify the α- to ω-phase transition as a function of peak shock stress. Experiments were performed to peak shock stresses of 8 and 10.5 GPa using methods described elsewhere [12]. The shock-wave profiles measured using VISAR are given in figure 1a. From this figure, it can be seen that a pronounced Hugoniot Elastic Limit (HEL) exists for both experiments. Additionally, the onset of the phase transformation is highlighted by black arrows in the figure. From the U_p data at these points the stress for transformation was calculated and is given in table 2. Stress for the transformation increases with peak shock stress. The rise time to the peak stress after the phase transformation differs as a function of peak stress. A quicker rise time to peak stress is observed in the 10.5 GPa experiment. This result is consistent with the idea that, for high purity Zr, the kinetics of the phase transformation are influenced by the loading conditions. While this has not previously been reported in Zr, studies on Fe have reported similar findings [13]. Finally, the lack of a rarefaction shock in both cases suggests that reverse phase transformation may not be appreciable within these specimens. Instead, upon recovery of the specimen, some meta-stable ω-phase should be retained.

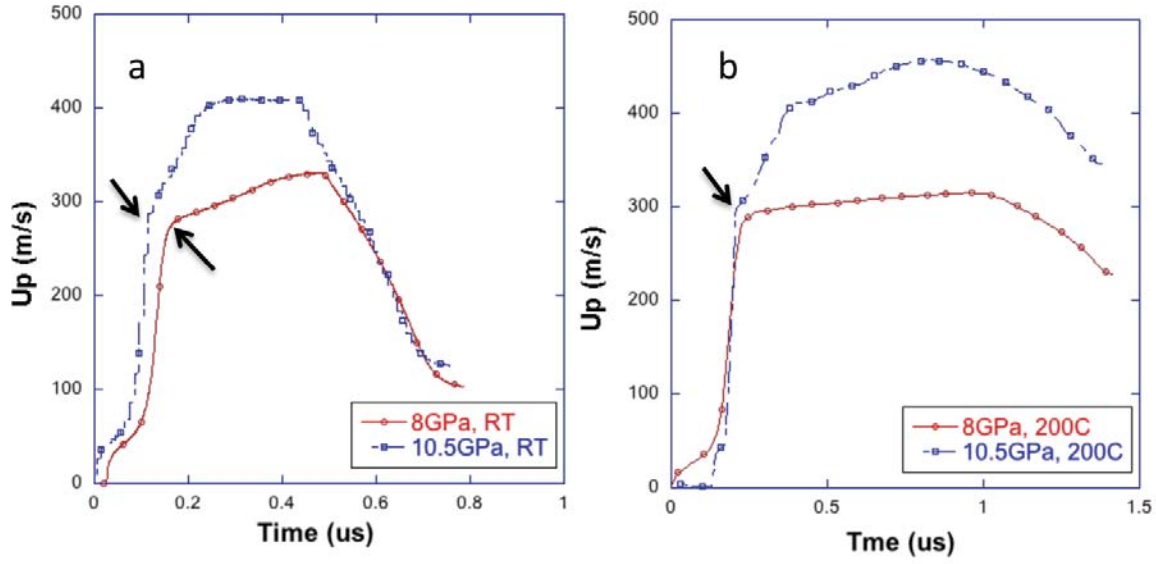

Figure 1. (a) RT VISAR wave profiles on windowed Zr specimens shocked to peak stresses of 8 and 10.5 GPa, (b) 200 °C PDV wave profiles on Zr specimens shocked to peak stresses of 8 and 10.5 GPa with no window and (c) RT wave profiles at 8 GPa on windowed and non-windowed specimens measured with VISAR and PDV, respectively.

The 200 °C shock-wave-profile experiments conducted on Zr were performed to identify the α- to ω-phase transition at elevated temperature. Again, experiments were performed to peak shock stresses of 8 and 10.5 GPa. The shock-wave profiles measured using PDV are given in figure 1b. From this figure, it can be seen that a pronounced Hugoniot Elastic Limit (HEL) exists only for the lower stress experiment and that while there is a clear signature of the α to ω-phase transition for the 8 GPa experiment, this is not the case for the 10.5 GPa experiment.

It is well recognized that HCP metals, and Zr in particular, display enhanced slip activity under elevated temperature testing at conventional stresses and strain rates [14, 15]. This enhanced slip, if still present under these dynamic loading conditions, would be expected to be an important energy dissipation mechanism. The fact that enhanced slip exists under the 200 °C loading condition is supported by the lack of a well-defined HEL for the higher stress experiment. Instead, an almost immediate transition to plastic behavior is observed there. Such energy dissipation is believed to be important to the relief of the local stress concentrations that may drive shear processes like twinning and phase transformation [16]. From the RT experiments, figure 1a, it can be seen that the transformation is on the cusp of occurring for the 8 GPa drive condition. Therefore taken together, it suggests that the enhanced slip at 200 °C is enough to mitigate the stresses for the phase transformation under this drive condition.

3.2 Quantification of the retained ω-phase

Shock recovery experiments with experimental details given in table 2 were performed. These specimens were examined for quantification of the volume fraction of the retained metastable, high-pressure phase as a function of dynamic drive condition. Neutron and x-ray diffraction techniques revealed the bulk volume fraction of retained ω-phase and these values are reported in table 2. It can be seen from these data that the hysteresis of the transition depends significantly on the initial temperature and peak stress of the plate impact experiment. This correlates well with the steep transition in particle velocity slope after the onset of transition in the 10.5 GPa, RT experiment as compared to the 8 GPa RT experiment, figure 1a, as well as the lack of an indication for transformation in the 8 GPa, 200 °C experiment, figure 1b.

Table 2: Percent retained ω-phase as a function of dynamic drive condition.

Sample thickness (mm)	Impactor			Peak stress (GPa)	Phase transition stress (GPa)	Temperature (°C)	Retained omega (%)
	Material	Thickness (mm)	Velocity (m/s)				
5.0	Zr	2.5	834	10.0	6.4	25	80
5.0	Zr	2.5	658	8.5	5.8	25	65
5.0	Zr	2.5	825	10.0	6.6	200	10
5.0	Zr	2.5	643	8.5	none	200	~0

3.3 Quasi-static reload properties as a function of retained ω-phase

The results from quasi-static compression testing of each of the microstructures given in table 2 are shown figure 2 and plotted along with the mechanical response of Zr loaded along the through-thickness direction in the as-annealed state. Enhanced hardening is observed in all shock loaded specimens as compared to the as-annealed material. The 80% retained ω-phase microstructure and the ~0% retained ω-phase microstructure display the highest and lowest enhanced hardening due to shock loading, respectively. This difference in subsequent materials properties is likely the result of the relative contribution of ω-phase volume fraction to deformation and the relative resistance of the ω-phase as compared to the α-phase to plastic processes [17]. The enhanced hardening behavior of the 65% and 10% retained ω-phase microstructures display intermediate states of enhanced hardening response due to shock loading but this response does not directly correlate with the volume fraction of retained ω-phase. This suggests that the role of stored plastic work within the α-phase is also

important to the subsequent mechanical response of shock loaded Zr just as has been observed previously in FCC metals that do not undergo phase transformation [4, 18].

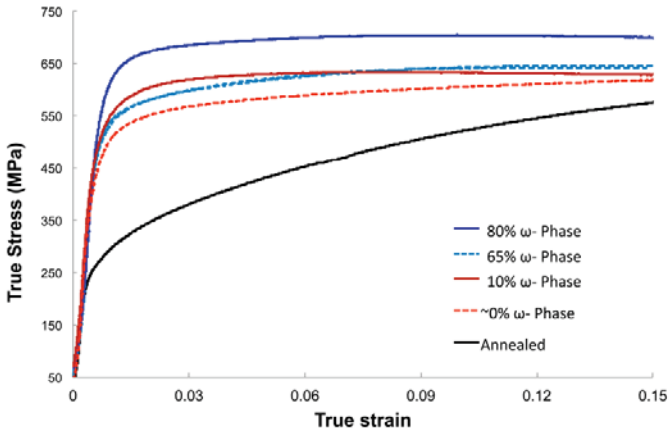

Figure 2. The stress-strain response in RT, quasi-static, compression of the shock recovered Zr as compared to that of as-annealed Zr.

4. Summary

The current study was designed to gain an understanding of the role of dynamic drive condition of the high-pressure ω-phase transformation in Zr and then the influence of this transformation on subsequent material properties. Small differences in the peak stresses and temperatures during dynamic loading were found to affect the kinetics of transformation as well as the activation of important deformation mechanisms, like slip, which can modify the shear stresses necessary for transformation. This then resulted in recovered specimens with differing volume fractions of retained, high-pressure ω-phase and evolved plasticity. The resulting microstructures displayed enhanced hardening over the as-annealed Zr. However, this enhanced hardening did not directly scale with the volume fraction of retained ω-phase, except at the extremes of the volume fraction values for the retained ω-phase.

Acknowledgements

LANS operates Los Alamos National Laboratory, LLC, for the NNSA of the U.S. Department of Energy under Contract No. DE-AC52-06NA25396. Campaign 2 and LANL LDRD programs have supported this work. The authors wish to thank M.F. Lopez for the mechanical testing as well as R.T. Olson, C. Greeff, and J.F. Bingert for thoughtful contributions.

References

[1] Greeff C, Trinkle D and Albers R 2001 *J. Appl. Phys.* **90** 2221-6
[2] Sikka S K, Vohra Y K and Chidambaram R 1982 *Prog. Mater. Sci.* **27** 245-311
[3] Jamieson, J C 1963 *Science* **140** 72-73
[4] Gray G T 1992 *Shock Experiments in Metals and Ceramics* (NY:Marcel Dekker Inc.)
[5] Cerreta E, Gray G T, Hixson R, Rigg P A and Brown D W 2005 *Acta Mater.* **53** 1751-8
[6] Rigg P A, Greef C W, Knudson M D, Hayes D, Hixson R and Gray G T 2003 *AIP Conf. Proc.* **706** 209-12
[7] Rabinkin A, Talianker M and Botstein O 1981 *Acta Metall. Mater.* **29** 691-698
[8] Cerreta E, Gray G T, Henrie B, Brown D W, Hixson R and Rigg P A 2003 *AIP Conf. Proc.* **706** 541-4

[9] Hennig R G, Trinkle D R, Bouchet J, Srinivasan S G, Albers R C and Wilkins J W 2005 *Nature Mater*. 129-134
[10] Rigg P A, Greef C W, Knudson M D, Gray III G and Hixson R S 2009 *J. Appl. Phys*. **106** 123531-1-9
[11] Hemsing W 1979 *Rev. Sci. Instrum*. **50** 73-78
[12] Gray G T 1993 *Influence of Shock Wave Deformation on Structure Property Behavior of Materials* (New York:Springer-Verlag)
[13] Smith R F, Eggert J H, Swift D C, Wang J, Duffy T S, Braun D G, Rudd R E, Reisman D B, Davis J P, Knudson M D and Collins G W 2013 *J. Appl. Phys*. **114** 223507-1-11
[14] Jain A and Agnew S R 2007 *Mater. Sci. Eng. A-Struct* **462** 29-36
[15] Addessio L B, Cerreta E and Gray G T 2005 *Metall. Mater. Trans*. A **36A** 2893-2903
[16] Zaretsky E 1995 *Acta Metall. Mater*. **43** 193-198
[17] Zhang S, Zhang X, Zhu Y, Zhang S, Qi L and Liu R 2012 *Comp. Mater. Sci*. **61** 42-49
[18] Gray G T and Vecchio K 1995 *Metall. Mater. Trans*. A **26A** 2555-2563