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Path Dependency of Phase Transformations

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Abstract. At high pressures titanium and zirconium are 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, the high-pressure omega-phase can be retained upon release. It has been shown that temperature, peak shock stress, and texture can influence the transformation. Moreover, under these same loading conditions, plastic processes of slip and twinning are also affected by similar differences in the loading path. To understand this path dependency and it role on microstructural evolution, in-situ velocimetry measurements along with post-mortem metallographic and neutron diffraction characterization of soft recovered specimens have been utilized to qualitatively understand the kinetics of transformation, quantify volume fraction of retained omega-phase and characterize the shocked alpha and omega-phases. Together the work described here can be utilized to understand phase and structure for these metals and lend insight into the partitioning of plastic processes between phases far from equilibrium conditions. Experiments discussed here, reveal that the transformation from alpha to omega phase is more difficult in Ti than in Zr metals. This result is discussed in terms of both differences in shear modulus and free space within the lattice for these systems.

INTRODUCTION

A number of applications subject materials to the extreme environment of high stress and high strain rate loading. For some of these cases, including the automotive, aerospace, and defense applications, catastrophic failure is highly undesirable and therefore, it is of interest to not only predict such failures, but also design materials that can mitigate them. Critical to such a capability is the understanding of material strength under these dynamic loads [1-3]. Currently, models for strength have limited applicability and this is believed to be related to the fact that in many of these models, microstructure of the material, and particularly its evolution during dynamic loading, is not well represented [4]. Specifically, mesoscale models are commonly used to represent the deformation and microstructural evolution due to the coupled processes of elasticity and plasticity due to slip and twinning. At high rates of loading these models also need to consider other dissipative processes like phase transformation. Further, the studies that have examined microstructural evolution during dynamic loading have focused on isotropic, randomly textured materials, rather than highly anisotropic, textured materials typical for many engineering applications. Data regarding the coupling of physical processes of elasticity, plastic slip, plastic twinning and phase transformation as well as the anisotropy of these responses during dynamic loading, are required to advance dislocation based plasticity theory in multiphase materials and to validate mesoscale models.

For this reason, here we investigate the structural evolution during dynamic loading for high purity, highly textured Zr and Ti. The mechanical response and structural evolution of these metals is well understood for quasi-static, uni-axial stress loading conditions [5-8]. As such it has been observed that the mechanical response in rolled plate material displays high yield stress and a concave down work hardening response when loaded in compression in a direction perpendicular to the rolling plane. This loading axis is sometimes referred to as the through thickness

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(TT) direction. When material is loaded in a direction within the rolling plane, frequently referred to as the in-plane (IP) direction, a relatively lower yield stress and a parabolic work hardening response is observed. This difference has been attributed to the rolling texture of the plate and its effects on the activation of slip and twinning, with respect to loading direction.

In addition to the role of slip and twinning in accommodating plasticity during deformation, it is also known that under high stresses, a martensitic, shear-driven, phase transformation from the hexagonal closed packed (HCP) alpha phase (stable at ambient conditions) to the simple hexagonal omega phase can take place [9]. The stress for this volumetric collapse is sensitive to the kinetics of loading. Additionally, the omega phase can be retained meta-stably at ambient pressures after unloading, thus suggesting that the equilibrium pressure-temperature phase diagram for Zr and Ti is inadequate to describe phase evolution during dynamic loading [10-12]. Previous studies that have focused on data for equation of state for these materials, have also shown that the phase transformation is sensitive to impurity concentrations within the metal [13]. The meta-stability of the high-pressure phase upon unloading, lends itself to post mortem metallographic examination and property measurements [14]. These studies have focused on the orientation relationship between the alpha and omega phases as an opportunity to postulate pathway for transformation [15-17]. Additionally, a few studies have taken material that was shock loaded to stresses above the transformation, characterized the volume fraction of retained omega phase, and then quasi-statically reloaded the material. These studies have shown that the shock loaded, two-phase Ti and Zr materials display an enhanced hardening over as-annealed material, upon reload [18]. Note, there is little data on the strength of this two phase material under dynamic conditions.

Under quasi-static conditions, the microstructure resulting from the alpha to omega transformation and defect evolution to accommodate plastic deformation have been shown to contribute directly to the strength of Ti and Zr. As such, microstructural evolution due to dynamic loading should also lead an evolving strength of the material at high stresses. To develop a physics based model of material strength under dynamic loading, these mechanisms and their effect on microstructural evolution have been studied. As part of this study, the role the plate impact loading on the transformation stress and the retention of omega phase upon unloading has been investigated and is discussed here. Differences in the observed responses between Ti and Zr are rationalized in terms of free space within the respective HCP unit cells.

EXPERIMENTAL

The materials chosen for this study were high purity Ti and Zr polycrystalline plates. In both cases the initial crystal bar material was upset, forged, rolled and annealed to produce highly textured, equaixed grain microstructures. Figure 1 shows the microstructure of Ti and the strong alignment of the basal pole texture with the normal of the plane of the rolled plate material. Figure 2 shows the same for the Zr material used in this study. Chemistry for the Ti and Zr materials are given in Tables 1 and 2, respectively. All of the materials were fully annealed in vacuum prior to testing.

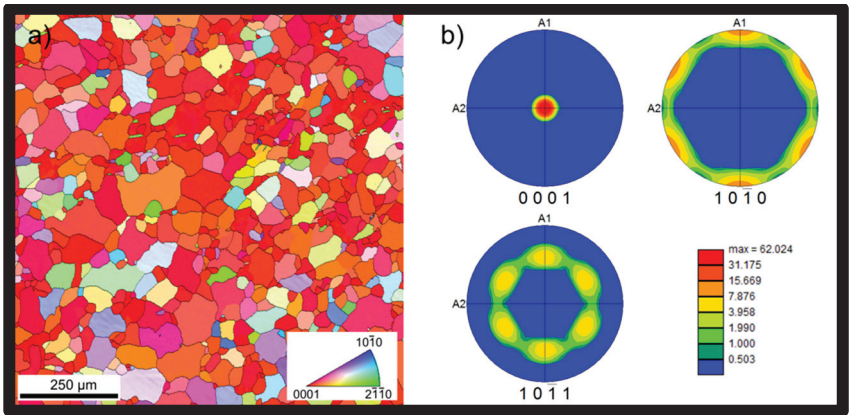

FIGURE 1. (a) Electron backscattered diffraction image and (b) texture of the as-annealed Ti plate microstructure prior to shock loading, as viewed from the normal of the plane of the plate.

As-annealed materials and tested materials were prepared via standard metallographic techniques to allow for examination of the microstructure and texture prior to dynamic testing. Metallographic specimens were examined optically as well as with electron back scattered diffraction (EBSD) in a scanning electron microscope.

Samples of titanium and zirconium were shock-loaded via gas-gun launched plate-impact experiments. The peak stresses obtained were 15 GPa in the Ti samples and 8 GPa and 10.5 GPa in the Zr samples. Shock-wave profiles were measured with a photon Doppler velocimetry (PDV) system, a non-contact interferometer can measure velocities in excess of 10^3 m/s. The precision of the wave velocity measurements is estimated to be approximately 1% in particle velocity. Stress-time histories for all specimens were calculated using Rankine-Hugoniot equations[19]. The PDV measured the rear free surface of the Ti samples, whereas the Zr samples were backed with a LiF window and as such the PDV measured the particle velocity at the Zr / LiF interface.

In all three cases discussed above, a second set of experiments were conducted to shock load and then soft-recover the specimens for post-mortem characterization. These tests were designed to load the materials in similar conditions to the first tests. The shock-load and recovery technique used here has been described in detail by Gray[20]. All samples were recovered with minimal residual strain and therefore cross sectioned for metallographic analysis. In all cases material was also reserved for bulk neutron and electron back scattered diffraction analysis at the Los Alamos Neutron Scattering Center (LANSCE) and within the Electron Microscopy Laboratory (EML) at Los Alamos National Laboratory. Grain orientation maps were obtained via EBSD. EBSD scans, with a 0.1μm step size, were collected using an FEI XL-30 scanning electron microscope equipped with the TSL/OIM EBSD system using an accelerating voltage of 20 kV. Data processing utilized the built-in tools in the TSL/OIM Data Analysis software package. More details regarding these data processing tools, can be found elsewhere [21]. Each scan was cleaned by removing all points with a confidence index of less than 0.05. These measurements as well as the neutron diffraction measurements were used to determine volume fractions and textures of individual phases present in the recovered materials.

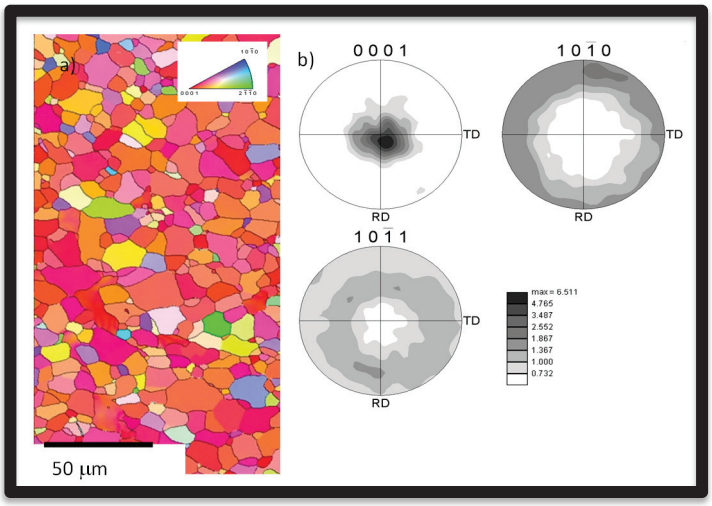

FIGURE 2. (a) Electron backscattered diffraction image and (b) texture of the as-annealed Zr plate microstructure prior to shock loading, as viewed from the normal of the plane of the plate.

TABLE 1. Chemistry of the as-received titanium plate in parts per million. Balance is titanium

O	Fe	Al	Ni	Cr	Mg	Si	Cu	Mn	V
<80	10.7	3.6	3.2	2.6	2.5	1.6	1.5	1.2	1.1

TABLE 2. Chemistry of the as-received zirconium plate in parts per million. Balance is zirconium

O	Hf	Fe	Al	V	N	C
<50	35	<50	<50	<20	<20	22

RESULTS

The free surface velocity histories for the plate impacted titanium and zirconium samples are given in Figs. 3 and 4, respectively. For all tests, a three-wave structure is observed. A clearly distinguishable elastic wave is followed by two plastic waves indicating the onset of the alpha to omega phase transformation. In the Ti data and for the Zr specimen tested to 10.5GPa, the transition in plastic, two-wave structure was sharper than in the Zr specimen tested to 8GPa, where the transformation appears to be gradual. From stress-time calculations, the stress for transition can be obtained. For the Ti specimen examined in this study, the transition stress was found to be 10.1 GPa. For the Zr, it was observed that the transition stress was loading path dependent the transition occurred at 5.8 GPa for the specimen impacted at 658m/s and at 6.4 GPa for the specimen impacted 834m/s [13,22].

Metallographic analysis of the shock loaded specimens revealed that in all cases, meta-stable omega phase was retained upon release. Additionally, alpha phase material was also found in all recovered specimens. Volume fractions of the metastable omega phase found in each of the recovered specimens, as measured by neutron diffraction for Zr and electron back-scattered diffraction for Ti, are given in Table 3.

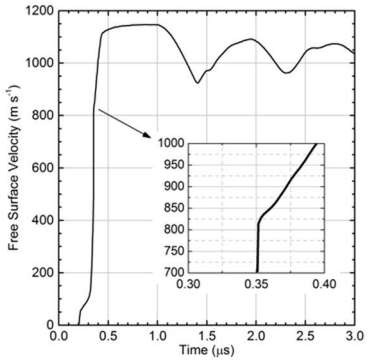

FIGURE 3. Free surface velocity data for plate impact experiment performed on Ti [23].

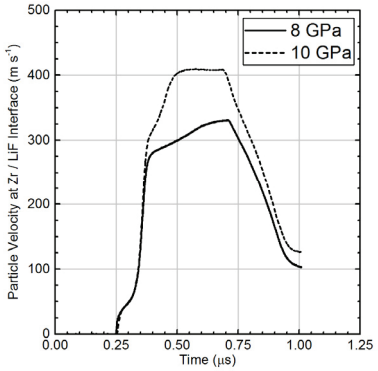

FIGURE 4. Shock-wave profile at the sample/LiF window interface for plate impact experiments performed on Zr.

TABLE 3. Volume fraction of retained omega phase in recovered tests

Material	Impact velocity (m/s)	Stress for Transformation (GPa)	Peak Stress (GPa)	Volume fraction retained ω (%)
Ti	822	10.1	15	65
Zr	658	5.8	8	65
Zr	834	6.4	10.5	80

Detailed metallographic analysis, via EBSD, also revealed the textures of the individual phases of the shock loaded specimens. These are given in Figs. 5 and 6, for Ti and Zr, respectively. It is important to note that the textures provided in Figs. 1 and 2 are the result of sampling multiple grains within the undeformed polycrystalline materials. Whereas, the textures provided in Figs. 5 and 6 are provided from single grains within the shock deformed materials. In the case of the Ti (Fig. 5), it appears that all six variants of the omega phase are present within the shock loaded structure, although not in equal fractions. Additionally, the alpha phase texture indicates that a significant fraction, perhaps all of the alpha phase present within the sample, possesses a texture that correlates well with plastic deformation occurring in the original, alpha phase material. In contrast to this, the single grain of Zr sampled, for the texture given in Fig. 6, displays an omega texture, in which only five variants of the high-pressure phase are present within the loaded and recovered specimen. Upon sampling other grains, it was apparent that all six variants of the omega phase were nucleated within the shock loaded polycrystal, although they were not nucleated in equal fractions in all grains. This observed variant selection difference is believed to be caused by the small orientation differences between neighboring grains with respect to the loading condition within the polycrystalline sample. Within this same grain of Zr that is analyzed in Fig 6b, Fig. 6a reveals that the alpha phase texture is completely reoriented from its original, undeformed state. This reorientation is not consistent with plastic processes like slip and twinning. Instead this material is believed to be the product of the high-pressure omega phase reverting to alpha phase upon release to ambient pressure. Since the paths for forward and reverse transformation are not uniquely known for the alpha-omega transformation in this material or expected to be the same, this cannot be validated definitively at this point in time. Future, in-situ, diffraction experiments are planned to interrogate this point. Finally, it should be noted that the above suggests that the Ti did not fully transform to the high-pressure phase during loading, but that the Zr, examined in this study, did.

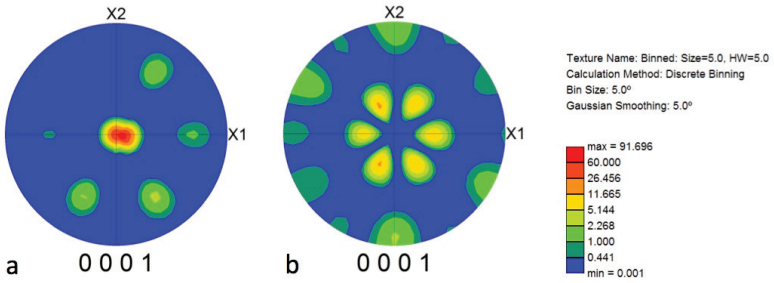

FIGURE 5. EBSD (0001) texture of the alpha and omega phases present in the Ti specimen shock loaded to a peak stress of 15GPa: (a) shock loaded alpha phase and (b) shock loaded omega phase.

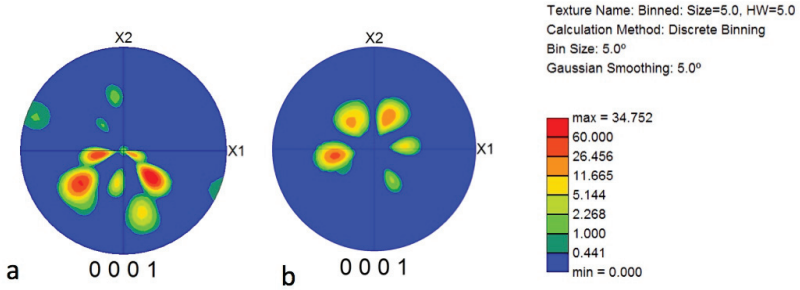

FIGURE 6. EBSD (0001) texture of the alpha and omega phases present in the Zr specimen shock loaded to a peak stress of 8GPa: (a) shock loaded alpha phase, and (b) shock loaded omega phase.

DISCUSSION

The alpha to omega phase transformation in both Ti and Zr is considered to be a shear driven transformation. While numerous paths for the transformation have been postulated [10,15,16,24], most involve a relatively large, in magnitude, shearing of atoms followed by a smaller shuffling of atoms to complete the transformation. The transformation has been observed to be affected by factors that also affect other shear driven processes, like twinning. In particular, the stress for transformation has been observed to be highly sensitive to interstitial oxygen content within the material [11,12]. Since oxygen atoms in solution are too large to sit within the tetrahedral sites of the alpha phase Ti or Zr unit cell, they sit in the octahedral sites of the lattice. As such, it was postulated that during the transformation, the necessary free space within the lattice to accommodate the shearing required for the volumetric collapse that occurs during transformation is partially occupied by the interstitial oxygen atoms. Thus, making the transformation more difficult with increasing impurity content.

Here and elsewhere [11,12,25,26], it has been observed that the transformation stress for the alpha to omega phase is higher in Ti than in Zr. It was also suggested, based on the texture analysis of the recovered specimens, that full transformation to the high-pressure phase and some reversion from the high-pressure phase back to the ambient pressure alpha phase occurred in Zr. This was inferred because there is no alpha phase found in its original undeformed orientation, the (0001) orientation, or in an orientation consistent with twinning of that original orientation in the alpha phase. However, the textures of the deformed alpha and omega phases in the shock loaded Ti do not appear to be consistent with full transformation or significant reversion from the high-pressure, omega phase. This is because alpha phase Ti with a texture consistent with slip and twin deformation was readily found in the shock deformed material. Taken together, these observations indicate that the transformation to omega phase is harder to drive in Ti than in Zr. It is interesting to note that this correlates well with the fact that there is 37% more free space in the alpha phase Zr unit cell as compared to the alpha phase Ti unit cell. This was calculated based on a hard sphere approximation.

Additionally, the difference in reversion from the high-pressure omega phase back to alpha phase differs between the two materials: little to no reversion is observed in the Ti experiments performed in this study while texture of the alpha phase present in shock loaded and recovered Zr, appears to be consistent with a reverted alpha phase. This maybe the result of two considerations. The first is that the drive conditions for the Ti experiments lead to a faster rate of release than the drive conditions for the Zr experiments. Kinetics for the reverse transformation (omega back to alpha) may not be favorable for this faster rate of release. A second consideration is the relative differences in free space within the ambient pressure phase lattice and its effect on shear processes discussed above. If shear processes related to reversion to alpha phase are more difficult to drive in the Ti than in Zr because of less free space in the alpha phase Ti lattice as compared to the Zr, this might also influence kinetics and driving force for reversion processes.

CONCLUSIONS

Experiments have been performed to interrogate the microstructural evolution during plate impact loading of high purity zirconium and titanium metals. These experiments were designed to examine the contribution of plasticity and phase transformation to evolving microstructure during shock loading as a way to inform strength models. Through these experiments, it was observed that even in the narrow range of loading conditions pursued under plate impact loading, that the alpha to omega phase transformation stress is highly sensitive to drive condition. Additionally, as has been previously observed, the phase transformation stress was significantly higher in titanium specimens than in zirconium specimens. Finally, metallography of recovered specimens revealed that the shock loaded microstructures of Ti and Zr differed. All specimens had a two phase, alpha-omega microstructure. However, the texture analysis of recovered Zr specimens revealed an alpha phase texture more consistent with alpha phase that had reverted from high pressure omega phase back to ambient pressure alpha phase upon release. The Ti specimens displayed an alpha phase texture that was consist with plastically deformed alpha phase material rather than alpha phase that had transformed to omega phase under high stress and reverted back to alpha phase upon release. These differences have been rationalized in terms of relative differences in free space in the ambient pressure, alpha phase lattice. Based on a hard sphere approximation, the alpha phase Zr lattice as 37% more free space than the Ti lattice. As such, the alpha phase Zr lattice may have more free space available to accommodate shear processes thought to be the mechanisms for both the forward and reverse alpha-omega phase transformation.

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