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Shock Phase Transformation and Release Properties of Aluminum Nitride

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Resume: Aluminum nitride is characterized by a non-recoverable volume phase transformation from the wurtzite (hexagonal) to the rocksalt (cubic) structure that commences about 22 GPa, accompanied by a volume strain of about 20%. A study of the manifestation of this large volume change under shock loading was made with four uniaxial strain impact experiments. Particle velocity histories of the shock and release states were obtained corresponding to impact pressures of 40 GPa, including the evolution of the shock and release waves with propagation distance. Stress - strain features characterizing aluminum nitride, including the phase transformation, were determined with numerical analysis techniques.

Abstract: Aluminum nitride is characterized by a non-recoverable volume phase transformation from the wurtzite (hexagonal) to the rocksalt (cubic) structure that commences about 22 GPa, accompanied by a volume strain of about 20%. A study of the manifestation of this large volume change under shock loading was made with four uniaxial strain impact experiments. Particle velocity histories of the shock and release states were obtained corresponding to impact pressures of 40 GPa, including the evolution of the shock and release waves with propagation distance. Stress - strain features characterizing aluminum nitride, including the phase transformation, were determined with numerical analysis techniques.

1. INTRODUCTION

Aluminum nitride is characterized by a non-recoverable volume phase transformation from the wurtzite (hexagonal) to the rocksalt (cubic) structure that commences under shock compression at about 22 GPa. The phase transformation has been reported by Vollstädt, *et al.* [1] in static high pressure experiments, and by Kondo, *et al.* [2] in shock wave experiments. In the latter study, the discrete data, obtained with pin and electromagnetic gage techniques, clearly indicated the presence of the phase transformation. The current study expands on that uniaxial data with time-resolved measurement of particle velocity histories of the shock and release states. Four experiments are reported, all with peak impact pressures of approximately 40 GPa, well above the phase transformation pressure. The evolution of the shock wave with propagation distance is addressed with variations of the target layer thickness. Details of the material response are obtained by numerical analysis techniques developed to examine the response of several other ceramics to shock loading [3]. The data obtained in the present study provide a basis for development of accurate computational models that can represent the response of this ceramic to high strain-rate loading in shock wave

propagation codes. Such models can then be used to examine the consequences of the large energy-absorbing aspect of the volumetric phase transformation of this ceramic during shock loading events. Useful data for aluminum nitride have also been provided in the longitudinal and lateral shock wave measurements of Brar, *et al.* [4], and shock compression studies of phase transformations by Nakamura and Mashimo [5]. Elastic properties and their pressure and temperature dependence have been investigated by Gerlich, *et al.* [6], while Van Camp, *et al.* [7] have explained equation of state issues in aluminum nitride as well as other binary nitrides.

2. MATERIALS

The aluminum nitride used in this study was produced by DOW Chemical Corporation. The nominal reference density of the aluminum nitride was 3262 kg/m^3 . Ultrasonic measurements on samples of this material indicate a longitudinal velocity of 10730 m/s and a shear velocity of 6320 m/s . Associated elastic properties derived from these velocities are a Poisson's ratio of $\nu = 0.234$, and bulk sound speed of 7866 m/s . The hot-pressed ceramic had a nominal porosity of 1% and a grain size of approximately $2 \mu\text{m}$.

Pure tantalum and tungsten impactors were used in order to obtain initial shock amplitudes well above the phase transition point of the aluminum nitride at the impact velocities attainable with the impact facility employed. The lithium fluoride used as the laser window material in the present impact experiments was optically clear [100] oriented single crystals [8].

3. EXPERIMENTS

Uniaxial-strain compressive shock and release waves were produced in the aluminum nitride with a single-stage powder-gun facility. The 89 mm diameter smooth-bore gun is capable of achieving a maximum impact velocity of about 2300 m/s . Three electrically shorting pins were used to measure the velocity of the projectile at impact. Four similar pins were mounted flush to the impact plane and used to monitor the planarity of impact. Projectile velocity could be measured with an accuracy of about 0.5% and the deviation from planarity of impact was typically about 10^{-3} radians.

The experimental target assemblies consisted of a disk of aluminum nitride backed by a lithium fluoride window (Figure 1). Particle velocity measurements were made at the interface between the aluminum nitride and lithium fluoride with a velocity interferometer, VISAR [9], having a time resolution of approximately 1 ns.

Figure 1. Experimental configuration of impactor and target assembly.

The impactors were either tantalum (backed by polyurethane foam) or tungsten (backed by polymethylmethacrylate, PMMA), accelerated with the powder gun to velocities of about 2200 m/s, providing shock amplitudes between 37 and 41 GPa. The thicknesses of all the tungsten and tantalum impactor plates were about 1.5 mm. The plate impactor thicknesses in these experiments were designed so that the complete shock and release profile could be measured at the recording interface.

The suite of target ceramic thicknesses led to variations in available time for both the development of the shock wave structure and the subsequent interaction of the release wave with the propagated shock wave. Table 1 lists the key parameters associated with these experiments.

Table 1: Uniaxial Strain Impact Experiments on Aluminum Nitride (Lithium Fluoride Windows)

Test No.	AlN Density (kg/m ³)	AlN Thickness (mm)	Impactor Materials	Impactor Velocity (m/s)	Impactor Thickness (mm)	Hugoniot Stress (GPa)
CE 44	3265	2.507	PMMA / Tungsten	2281	1.502	41
CE 43	3262	2.510	Foam / Tantalum	2207	1.528	37
CE 52	3258	4.183	Foam / Tantalum	2215	1.526	37
CE 37	3265	9.567	Foam / Tantalum	2239	1.508	38

The measured particle velocity profiles of all four experiments are shown in Figure 2. The arrival times of the initial, or “elastic”, waves correspond to the ultrasonic longitudinal transit time through the aluminum nitride targets. The two highest amplitude wave profiles (both with arrival time at 0.2 μ s) are for the tungsten (CE 44) and tantalum (CE 43) projectiles impacting 2.5 mm of aluminum nitride. The impactor thicknesses, impactor velocities, and aluminum nitride target thicknesses are of comparable magnitude for both these experiments (Table 1). The higher impedance of the tungsten, and its slightly larger impact velocity results in a larger amplitude particle velocity than for the corresponding tantalum case. This in turn leads to a much earlier arrival of the upper shock wave characterizing the shock-induced phase transformation. The initial break in the shock wave occurs at about 400 m/s, and represents onset of permanent deformation, or “yield”, as typically observed in many ceramics tested in this one-dimensional strain configuration [3],[10]. The second break which appears, at about 900 m/s, is believed to correspond to onset of the wurtzite (hexagonal) to the rocksalt (cubic) phase transformation, with a non-recoverable volumetric phase transformation. The release wave structure substantiates the non-recoverable aspect of this transition, as will be demonstrated by evaluation of the one-dimensional numerical calculations of the experimental wave profiles.

In three experiments, the thickness of the tantalum flyer plate was maintained at approximately 1.5 mm, and the impact velocity was fixed at about 2200 m/s (Table 1). Although for the 2.5 mm aluminum nitride target a wave with some time at the peak between shock and release is recorded, increasing the thickness of the aluminum nitride target effectively extends the propagation distance of both the release wave, which emanates from the trailing low impedance interface of the flyer, and the initial shock wave. Given sufficient propagation distance, the release wave will overtake the lower velocity phase transformation wave, and the interaction will attenuate the amplitude of the initial wave. The third profile in Figure 2 (4.2 mm aluminum nitride) shows the upper part of the shock wave just beginning to be eroded by the release wave at the recording interface. The 9.6 mm aluminum nitride target is of large enough thickness that almost complete attenuation of the upper shock wave has occurred by the time the rear surface of the ceramic target has been encountered.

There are two other noticeable trends in the particle velocity records for the tantalum impactor series in which the thickness of the aluminum nitride target increases from 2.5 mm to 9.6 mm: (1) the initial amplitude of the break in the first shock wave decays from about 400 m/s to 320 m/s, and (2) the amplitude of the second break, identified with the phase transformation, drops in amplitude from 900 m/s to 840 m/s.

Figure 2. Particle velocity records for tungsten impactor (1.5 mm, 2200 m/s) impacting 2.5 mm aluminum nitride target, and tantalum impactor (1.5 mm, 2200 m/s) impacting 2.5 mm, 4.2 mm, and 9.6 mm aluminum nitride targets.

4. COMPRESSION AND RELEASE PROPERTIES

The complex structure of the particle velocity history, particularly above the phase transformation point in both the shock and release waves, is a consequence of impedance differences at the impact and recording interfaces of the target. The tantalum and tungsten impactors have significantly larger impedances than the aluminum nitride targets, and the impedance of the lithium fluoride window is less than one-half that of aluminum nitride. As a consequence, when the release wave from the trailing interface of the impactor arrives at the impact interface, some unloading will occur. In the same way, at the aluminum nitride / lithium fluoride window interface, the reflected partial release wave will interact with the higher amplitude, slower shock of the phase transformation. The numerical techniques used here account for all these wave interactions, and clarify the original wave structure in the aluminum nitride targets.

Analysis of the data was made with a one-dimensional Lagrangian finite-difference wave propagation code, WONDY [11]. The aluminum nitride material response was characterized with parameterized sound velocity and strain linear segments, with separate loading and unloading curves defined so that both shock and release properties of the ceramic could be represented [3]. Linear variations in sound speed with strain result in a quadratic dependence of the local material modulus on strain. Particular care was required to monitor the current amplitude and rate of loading so that the correct unloading and reloading curves were used for the local material response. The local material modulus was calculated from the sound speed based on the local strain and the appropriate sound speed segment, and determined the increment in axial stress at each time step in the calculation. Ultrasonic data were used to determine the initial loading moduli. The parameters of the sound speed curves for the inelastic response were determined iteratively, tailored to match the particle velocity history for each experiment. By taking account of the interactions of the waves at the lithium fluoride / aluminum nitride and flyer / aluminum nitride interfaces, this approach provided the capability to examine the ceramic response interior to the samples, and in particular, to determine the effect of the volume phase transformation on the transmitted wave structure.

The tantalum and tungsten impactors were modeled with an anisotropic strain hardening model in

WONDY [11]. Shock and release data for each of these materials in this regime of pressure verified the parameters used to characterize these materials. Data for the tungsten were obtained from Chhabildas, *et al.* [12] and for the tantalum from Chhabildas and Asay [13]. The release wave that originates at the trailing interface of the impactor is transmitted to the target through the impact interface. It is of particular importance that the initial structure of this transmitted release into the aluminum nitride sample be as accurate as possible. Otherwise, modeling errors in the impactors will corrupt the analysis of the particle velocity data by requiring compensating errors in the model constructed for the aluminum nitride.

Numerical analysis of the experiment in which a 2.5 mm target was impacted with a 1.5 mm tantalum projectile (Experiment CE 43) resulted in the fit to the particle velocity data shown in Figure 3. Nearly all of the features of the data are captured by this analysis technique; iterations on the sound speed - strain function focused primarily on the first microsecond of the record in order to reproduce the shock and release response of the aluminum nitride in as much detail as possible. The fit to the particle velocity data in which a 2.5 mm aluminum nitride target was impacted by 1.5 mm tungsten projectile (Experiment CE 44) is shown in Figure 4. The sound speed - strain functions used to fit these two experimental profiles differ only slightly at large strains, in order to accommodate the increased velocity of the upper shock at the higher impact pressure. The unloading curves are identical for these two cases.

Figure 3. Numerical fit to interface particle velocity data for tantalum impact on 2.5 mm aluminum nitride (CE 43)

Figure 4. Numerical fit to interface particle velocity data for tungsten impact on 2.5 mm aluminum nitride (CE 44)

The appearance of a transmitted stress wave in an aluminum nitride target is recorded in Figure 5. The three-wave structure is much more well-defined in this representation than in the particle velocity history at the target / window interface, because no interface wave interactions have occurred yet. The structure in the particle velocity data in both Figure 3 and Figure 4, beginning at 0.35 μ s and ending at about 0.5 μ s in the particle velocity interval 900 m/s to 1250 m/s, is all due to wave interactions at the target / window interface, and is not present in the transmitted wave prior to encountering the interface. The three primary waves are identified in Figure 5 as the initial elastic wave, the deformation (or "plastic") wave, and the phase transformation wave.

The stress - strain response that corresponds to this transmitted wave profile is shown in Figure 6. The first (elastic) wave has an amplitude of approximately 10 GPa, Figure 5 and Figure 6, at which point a break in the curve indicates the onset of yield. This amplitude agrees well with 9-10 GPa reported by Nakamura and Mashimo [5], and 9.4 GPa reported by Brar, *et al.* [4]. The next break in the wave occurs at 22 GPa, which compares well with 21 ± 1 GPa shock transition pressure reported by Kondo, *et al.* [2], and

19 GPa reported by Nakamura and Mashimo [5]. The total axial strain at the beginning of the phase transformation is about 7.5%. The maximum amplitude of the shock wave in this case corresponds to about 28% strain, with the phase transformation accounting for about 20% strain between the onset and the maximum load. This volumetric compression agrees well with the value of 22% obtained by Kondo, et al. [2] from shock wave experiments. These results also compare favorably with static measurements, in which a transition pressure of 16.5 GPa and volume strain of 21% were reported by Vollstädt, *et al.* [1].

Note that as the aluminum nitride unloads from peak compression of about 40 GPa down to about 10 GPa, the unloading progresses nearly elastically, recovering about 6% strain, and does not follow the initial compression curve that included the phase transformation (Figure 6). This indicates that no reversal of the phase transformation on unloading to approximately 10 GPa has occurred in the elapsed time of this experimental configuration, and that the deformation (volume strain) accompanying the phase transformation is permanent. Were there a reversal of the phase transformation during unloading, a much more complex representation of the unloading curve would be required.

Figure 5. Transmitted wave profile in aluminum nitride from tantalum impact.

Figure 6. Stress-strain response of aluminum nitride.

The functional relationship of sound speed to strain determined for the aluminum nitride that engenders the accurate fit to the particle velocity data in Figure 3 is shown in Figure 7. The discrete nature of the sound speed fitting segments and the distinct loading and unloading curves are quite evident in this figure. The reference zero-strain velocity is the ultrasonic velocity, followed by a slight increase (non-linear elastic) to the yield point at about 2% strain. A sudden drop in sound speed to 8300 m/s occurs to represent the "plastic" deformation wave. At the onset of the phase transformation, there is another large decrease in sound speed to accommodate the volumetric compression of the phase transformation (Figure 7). The sound speed then gradually recovers with increasing strain as the phase transformation deformation limit is approached. The sound speed function between "onset" and "completion" determines the structure in the particle velocity histories at the recording interface above the "plastic" wave (0.35 - 0.5 μ s, Figure 3 and Figure 4). The separate unloading sound speed - strain function (Figure 7) accommodates the large change in wave velocities between shock and release states in the aluminum nitride. The parameters for this unloading curve were also derived iteratively to match the release waves in Figure 3 and Figure 4. In the calculation, when material begins to unload from compressive states, the local modulus evaluation abruptly changes from the loading curve to the unloading curve.

The shock and particle velocities associated with the transmitted wave profiles for both tantalum (2207 m/s, CE 43) and tungsten (2281 m/s, CE 44) 1.5 mm impactors onto aluminum nitride are plotted in

Figure 8. The shock velocity is calculated from the arrival time of an increment of the stress wave at a chosen monitoring position in the aluminum nitride, and the distance from the impact interface to the monitored point. The displaced position of this monitoring point is included in the shock velocity calculation. Data points from Kondo, *et al.* [2] are compared with this data extracted from the analysis of the present experiments. We observe that there is a good agreement with the two data points at a particle velocity of 8.6 km/s, defining the shock velocity of the plastic deformation wave. Three other data points from Kondo, *et al.* [2], in the particle velocity range of these experiments, are also in excellent agreement with the shock velocities determined for the phase transformation wave. The data point near 175 m/s is in the elastic regime, albeit somewhat lower than that obtained in the material of the current study. (The reported longitudinal velocity in [2] is 10000 - 10400 m/s compared to the 10730 m/s for the material used here.)

Figure 7. Characterization of aluminum nitride with sound speed function of strain.

Figure 8. Shock velocity *versus* particle velocity response of aluminum nitride.

From the calculated change in shock velocity with particle velocity at the phase transformation wave, an effective coefficient, S , in the linear shock velocity - particle velocity relationship is determined to be about 4.8; this is much larger than the value of about 2.0 determined to be the observed upper limit for shock compression of the preponderance of single phase materials [14]. Kondo, *et al.* [2] report shock velocity *versus* particle velocity data to about 3000 m/s, and a value of S of at least 3.4 is suggested by their fit to that data. These large values of S indicate that there are significant differences in the nature of compressibilities between single and multiple phase materials.

5. CONCLUSIONS

The present experiments and analysis of aluminum nitride provide substantial evidence for a non-recoverable volumetric phase transformation under shock compression at a pressure of about 22 GPa. The fine time-resolution of the data permit accurate assessment of the initial permanent deformation, the onset of the phase transformation, the loading path through the transformation to final shocked states of 40 GPa, and the unloading behavior from these shocked states. Numerical analysis provides insight into the development of the three wave structure that forms from the initial shock at the impact interface, and is an effective analytical utility for establishing dynamic stress *versus* deformation behavior. The data are generally consistent with previously reported data for aluminum nitride. These data and analyses of deformation provide a solid basis for the construction and evaluation of material models for this material, particularly in the accounting of the large energy dissipation that accompanies the phase transformation.

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