SHOCK-INDUCED DEFORMATION OF TUNGSTEN POWDER

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ABSTRACT

An explosive formulation containing fine tungsten particles mixed with HMX was detonated. Samples of the tungsten powder were recovered and investigated to characterize the structural effects of detonation. Broadened X-ray diffraction profiles obtained from the shocked tungsten powder were analyzed employing double order and single order Fourier analysis methods and the Williamson-Hall Integral Breadth procedure. The average coherent diffracting domain size was determined to be 35.4 nm and the root mean square strain was of the order of 1.5x10⁻³. Strain results obtained in this study were less than those previously reported for filed tungsten, indicating polygonization occurred during the experiment. The information gathered furthers the understanding of the deformation processes associated with the detonation events.

INTRODUCTION

The description of the dislocation structure of cold worked metals from the X-ray Debye-Scherrer spectrum has been of interest since Williamson and Smallman provided their correlations between X-ray structural parameters and dislocation density [1]. Included in their work was the defect characterization of tungsten both in the cold work state produced by filing and in the partially annealed state. In the filed condition, the tungsten data indicated dislocation pileups were present; however, after annealing at 1150°C for one hour the dislocation structure polygonized. The isotropic elastic constants of tungsten provide a great advantage in investigating cold worked powders by X-ray line broadening techniques. Deconvolution of the particle size and strain contributions to broadening does not require double order data from an (hkl). The objective of this work was to characterize the defect structure generated in powder tungsten that was subjected to an explosive shock during the detonation event.

EXPERIMENTAL

Spherodized tungsten powder obtained from M.C. Stark (99% purity) was analyzed with a Coulter LS130 particle size analyzer via Fraunhofer laser scattering. The volume and number distribution functions shown in Figures 1 and 2, give average particle sizes of 37.0 µm and 2.23 µm, respectively. The tungsten powder was mixed into a cast-cure HMX formulation that exhibited a detonation (C-J) pressure of 24.3 GPa and a detonation velocity of 4.80 m/sec.

Detonation testing was performed in an open-air configuration with a boosted, end-initiated 5.08 mm (2 inch) diameter charge. A polypropylene witness plate collected the detonation products and tungsten powder. The plate was positioned sufficiently far from the charge so that the impact velocity of the powder would be minimized, preventing additional deformation of the powder. Post-test examination of the plate found the tungsten powder to be caked on, and
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imbedded in the polypropylene. Gentle scrapping released a coarse powdered material. Seven
samples were collected at variable distances from the explosive event and individually washed
with hexane in a soxhlet apparatus for 24 hours to remove organic residue and polypropylene.
Additionally, a quantity of as-received powder was annealed for one hour at 1250° C under 10⁻⁵
torr of vacuum. This powder was employed to measure the instrumental broadening.

![Figure 1: Cumulative volume particle size distribution.](image1)

![Figure 2: Cumulative number particle size distribution.](image2)

All 0-20 diffraction patterns were collected using a Philips X’Pert Pro MPD system, copper
radiation, focusing optics, and a graphite diffracted-beam monochromator. The divergent and
anti-scatter slits were set to 0.5° with a 0.20 mm receiving slit. Data was collected from 30.00 to
140.00 degrees (2θ) with a 0.01° step size and a measurement time of 4.0 sec/step. Samples
were sprinkled onto a silicon zero-background plate (lightly coated with vacuum grease), and
placed into a sample spinner stage.

**DATA ANALYSIS METHODS**

The coherent diffracting particle size and root mean square microstrains were determined for
each sample of deformed tungsten and the as received powder using three analyses methods;
integral breadths, double-order Fourier, and single-order Fourier analyses. The double-order
Fourier analysis followed the Warren-Averbach method [2] of deconvoluting strain and
crystallite size portions of the Stokes-corrected Fourier coefficients [3]:

\[
\ln A_L(h_\theta) = -\frac{2\pi^2 L^2}{a_\theta^2} \left\langle e_L^2 \right\rangle h_\theta^2 + \ln A_L^D
\]

(1)

Here \( a_\theta \) is the lattice parameter and \( h_\theta \) is defined to be the sum of the square of the Miller
indices, \( (h^2 + k^2 + l^2) \). The parameter \( a_\theta \) is expressed with units of [nm]. The slope of the plot
of \( \ln(A_L) \) versus \( h_\theta^2 \) at a fixed value of \( L \) [nm] gives \( \left\langle e_L^2 \right\rangle \) [nm/nm] and the intercepts give the
values of the particle size coefficients. The initial slope of the size coefficients, \( A_L^s \), as a
function of \( L \), renormalized to unity to account for the hook effect, gives a value for the average
crystallite size. Fortuitously, the elastic constants of tungsten are isotropic, thereby allowing
calculations to be performed with the traditional (110)(220) double order pair or the (110) and
(321) peaks, as the (220) reflection exhibited low signal to noise ratio in the collected data.
These analyses were carried out using numeric algorithms developed by De Angelis and
Schwartz [4].

Single order Fourier analysis methods, as proposed by Minot and Rondot [5], were also carried
out with numeric techniques. In this case, the size and strain contributions to equation (2) are
deconvoluted by developing the relationship of their contributions to the measured strain into a
second order polynomial equation.

\[
A_L = A_L^S \cdot A_L^D
\]

(2)
If $D$ is used to express the crystallite size and $d$ the diffraction plane spacing, both with units of [nm], $K$ [nm$^{-2}$] and $X$ [nm$^{-1}$] can be defined:

$$K = \frac{2\pi^2}{d^2} \quad (3)$$

$$X = \frac{1}{D} \quad (4)$$

As a result, the size and strain terms of equation (2) can now be expressed as:

$$A_L^S = (1 - LX) \quad (5)$$

$$A_L^p = (1 - KL^2 \langle \varepsilon_L^2 \rangle) \quad (6)$$

The strain function is then assumed to be [6]:

$$\langle \varepsilon_L^2 \rangle = \left( \frac{C \delta}{L} \right) \quad (7)$$

$\delta$ [nm] is used to represent the lattice parameter in Fourier intervals. From this, it can then be shown that the resulting polynomial takes the form:

$$A_L = 1 - L(X + CK\delta) + L^2(XCK\delta) \quad (8)$$

Values for $C$ and $X$ are arrived at by fitting to the Stokes corrected coefficients [7]. A series of constraints and restraints are then used to select the best solutions.

Integral breadth, by the traditional Williamson-Hall method, was carried out by first separating the $K_\alpha$ doublet via the method developed by Landel et al. [8] Instrumental broadening effects where then removed by the relationship [9]:

$$B(2\theta)_{corr} = B(2\theta)_{broadened} - \left[ \frac{B^2(2\theta)_{inst}}{B(2\theta)_{broadened}} \right] \quad (9)$$

A Gaussian shape was assumed for both the size and strain functions, thereby allowing a description of the size and strain contributions to the integral breath to take the form [10]:

$$B^2(S)_T = \left( 2\varepsilon \frac{1}{d} \right)^2 + \left( \frac{1}{D} \right)^2 \quad [\text{nm}^{-2}] \quad (10)$$

By plotting the square of the measured integral breadths, $B^2(S)_T$, versus the values of $S^2$ (also expressed as $1/d^2$ [nm$^{-2}$]), one arrives at the value for the average crystallite size by taking the intercept of the plotted function. The average strain is taken from the slope of the plotted curve, $\langle 2\varepsilon \rangle^2$. From this, the root mean square strain is found according to equation (11):

$$\varepsilon_{rms} = \sqrt{\varepsilon_{avg} \cdot \frac{1}{1.25}} \quad (11)$$
Dislocation densities are calculated from the crystallite size results with the assumption that the diffraction particle size is the average distance between single dislocations. The density of dislocations is given by:

\[ \rho_{\text{size}} = \left( \frac{1}{D^2} \right) \left[ \text{cm}^{-2} \right] \tag{12} \]

Additionally, strain values are used to find dislocations densities from the relationship [11,1]:

\[ \rho_{\text{strain}} = \frac{12\langle \varepsilon^2 \rangle}{b^2} \left[ \text{cm}^{-2} \right] \tag{13} \]

Here, \( b \) is the magnitude of a \( \frac{1}{2} <111> \) Burgers’ vector, which for tungsten takes the value 2.74x10^{-8} cm.

RESULTS AND DISCUSSION

Comparing the SEM images of the as-received and the post-shot materials in Figure 3 shows the experiment produced heavily deformed and agglomerated particles. The observed agglomeration was attributed to shock welding processes resulting from particle-particle impacts. These impacts could have occurred during the detonation event or after the particles had been accelerated, or both. On a qualitative basis, no correlation was observed between the morphologies of the agglomerates and their distance from the charge.

A typical X-ray scan showing the extent of line broadening observed from these materials is shown in Figure 4. Most samples were contaminated with SiO\(_2\) a native material present at the test site. Thus, the usable data range of several peaks was restricted due to inference from the SiO\(_2\) pattern. This can be seen around the (110) tungsten peak.

The diffracting particle size and strain determinations as a function of distance from the detonated charge from all methods employed are shown in Figures 5 and 6. Strain values reported here for all three Fourier analyses methods were taken at an \( L \) value of 15.0 nm. In the case of both crystallite size and root mean square strain, no clear relationship to distance from the detonation event was observed. The lack of variation as a function of distance both in the XRD results and the SEM images gave reason to treat the size and strain values as fluctuating around a single average value, within some limit of variation introduced by experimental procedures and such factors as signal-to-noise ratios and the above mentioned sample.
contamination. The average values and standard deviations for diffracting particle size and strain for each method are reported in Table 1. For the sake of comparison, results for the as-received powder are also included. This sample was found to be sufficiently close to an annealed state that only the single-order Fourier analysis method yielded numerically valid crystallite sizes when performed on the (200) reflection (or any other lower \( d \)-spacing plane).

![Figure 4: Typical X-ray scan of deformed and annealed powders.](image)

![Figure 5: Size versus distance results](image)

![Figure 6: Strain versus distance results](image)

Table 1. Analysis results on shocked tungsten and as-received powder.

<table>
<thead>
<tr>
<th></th>
<th>Size (nm)</th>
<th>Std Dev (%)</th>
<th>Strain</th>
<th>Std Dev (%)</th>
<th>Size-Calculated Dislocation ( \rho(1/cm^2) )</th>
<th>Strain-Calculated Dislocation ( \rho(1/cm^2) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>W-A (110) (220)</td>
<td>36.7</td>
<td>17.1</td>
<td>1.54E-03</td>
<td>7.7</td>
<td>8.03E+10</td>
<td>3.82E+10</td>
</tr>
<tr>
<td>W-A (110) (321)</td>
<td>35.4</td>
<td>13.8</td>
<td>1.44E-03</td>
<td>6.1</td>
<td>8.41E+10</td>
<td>3.31E+10</td>
</tr>
<tr>
<td>M-R (110)</td>
<td>28.8</td>
<td>16.7</td>
<td>1.54E-03</td>
<td>25.8</td>
<td>1.32E+11</td>
<td>4.01E+10</td>
</tr>
<tr>
<td>W-H</td>
<td>45.2</td>
<td>20.8</td>
<td>1.79E-03</td>
<td>7.3</td>
<td>5.44E+10</td>
<td>5.15E+10</td>
</tr>
<tr>
<td>As-Received M-R (200)</td>
<td>1939.6</td>
<td>--</td>
<td>1.14E-06</td>
<td>--</td>
<td>2.66E+07</td>
<td>2.08E+04</td>
</tr>
</tbody>
</table>
Dislocation densities calculated from strain results were found to be consistently less than those from crystallite size values. This disparity was attributed to the occurrence of polygonization and recovery at elevated temperatures during the detonation and subsequent events, decreasing the dislocation strain fields. Further support for recovery was found in comparison with previously reported work, found in Table 2. In each investigation of heavily cold-worked tungsten, greater strain values were reported, while the crystallite sizes were nearly identical to those obtained in this work.

Table 2: Data comparison for deformed tungsten.

<table>
<thead>
<tr>
<th></th>
<th>Size (nm)</th>
<th>Strain</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Current study</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(shocked tungsten)</td>
<td>Double Order Fourier (110)(321)</td>
<td>35.4</td>
</tr>
<tr>
<td></td>
<td>Single Order Fourier</td>
<td>28.8</td>
</tr>
<tr>
<td></td>
<td>Integral Breath, Gaussian</td>
<td>45.2</td>
</tr>
<tr>
<td><strong>Halder and Wagner</strong></td>
<td>Double Order Fourier</td>
<td>21.0</td>
</tr>
<tr>
<td>(filed tungsten)</td>
<td>Variance</td>
<td>14.0</td>
</tr>
<tr>
<td></td>
<td>Integral Breath, Cauchy/Gaussian</td>
<td>43.0</td>
</tr>
<tr>
<td><strong>Wagner and Aqua</strong></td>
<td>Double Order Fourier</td>
<td>22.0</td>
</tr>
<tr>
<td>(filed tungsten)</td>
<td>Integral Breath, Gaussian</td>
<td>40.0</td>
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<tr>
<td></td>
<td>Integral Breath, Cauchy</td>
<td>85.0</td>
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<tr>
<td></td>
<td>Integral Breath, Gaussian (Parabolic)</td>
<td>37.5</td>
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<tr>
<td><strong>Williamson and Hall</strong></td>
<td>Integral Breath, Cauchy</td>
<td>20.0</td>
</tr>
</tbody>
</table>

CONCLUSIONS

The detonation of an explosive tungsten particle formulation was found to produce complex deformation of the tungsten powder. The powders were agglomerated during the event into lamellar composite particles, due to large plastic deformation and shock welding. As a result of this agglomeration, no variations in deformation were measured as a function of the flight distance from the detonation event. The defect structure was not measured in the state of maximum deformation due to polygonization and recovery processes resulting from the elevated temperatures at the detonation front and the rapid severe plastic deformation.

REFERENCES