



CONSOLIDATION AND HIGH STRAIN RATE MECHANICAL BEHAVIOR OF NANOCRYSTALLINE TANTALUM POWDER

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***Abstract** -- High ductility and strength exhibited in nanograined materials can potentially be exploited in explosively formed penetrator liner applications. Both coarse and nanocrystalline tantalum powder were consolidated by Plasma Pressure Compaction (P^2C) to study the effect of grain size on dynamic compression properties. The powders were consolidated rapidly with 1 minute of isothermal holding time to retain initial microstructure. The P^2C consolidated specimens were cut by electric discharge machining (EDM), polished for SEM characterization, and tested in dynamic compression using a Kolsky apparatus. The effect of grain size on yield stress and strain was investigated at various strain rates for a coarse grained and a nanograined specimen. Especially, the high strain rate response of nanocrystalline tantalum is discussed in this paper. ©1999 Acta Metallurgica Inc.*

INTRODUCTION

The high density and ductility of tantalum make it an attractive material for ballistic applications such as explosively formed penetrators (EFPs). The dynamic formability of an EFP depends on the physical and mechanical properties of the material, especially the initial crystallographic texture and the evolved deformation texture. To fabricate a tantalum EFP that meets dynamic formability requirements, processing-microstructure-property relationships must be established.

High strain rate deformation is the most critical performance feature for an EFP. Deformation studies at low strain rates have indicated that ductile behavior is observed in polycrystalline metals with fine grain structure including tantalum [1]. Therefore, it is important to understand the effect of grain size refinement on the flow and failure behavior for polycrystalline tantalum at high strain rates. In this study, the deformation behavior of two tantalum materials will be initially investigated especially with respect to their grain size at high loading rates.

EXPERIMENTAL PROCEDURE

Tantalum powder (-325 mesh) was obtained from Trinitech (Twinsburg, OH) and tantalum nanopowder (<100 nm) was procured from the Army Research Laboratory (Aberdeen, MD). Initial particle size was analyzed by the BET technique (Coulter, SA 3100). The powders were consolidated by Plasma Pressure Compaction, and density was measured using Archimedes' principle. Dynamic and quasistatic compression tests were performed on the consolidated samples at Michigan Technological University (Houghton, MI).

Plasma Pressure Compaction

Plasma Pressure Compaction (P²C) is a rapid consolidation technique that was developed by Materials Modification Inc, Fairfax, VA. In the P²C process, powders are simply poured into a graphite die without any additive and binder and compressed by graphite punches. At the beginning of the process, pulsed DC voltage is applied through the powder compacts to generate an interparticle plasma that activates the surface of the particles by removing the oxide and contaminant layers. Subsequently, DC voltage is applied to achieve rapid densification by resistance sintering under uniaxial pressure.

For tantalum consolidation, uniaxial pressure of 50 MPa was applied initially to the powder compact through graphite punches to establish a current path. DC pulses of 60 millisecond duration were applied at maximum amplitude of 1300 ampere, and the voltage varied between 0 to 5 V for total of 3 minutes to generate interparticle plasma which leads to surface activation. After the pulsed DC voltage application, constant DC voltage was applied to raise the sintering temperature rapidly at a rate of 500 °C/min. and held at 1750 °C and 2000 °C for less than 1 minute. The temperature was measured at the graphite die surface using an optical pyrometer.

Dynamic Compression Testing

In 1949, Kolsky developed a technique for the measurement of the properties of materials undergoing homogeneous deformation at fairly high strain rates [2]. The procedure has become known as the "Kolsky bar" or "split Hopkinson pressure bar (SHPB)". Since then, this technique has become the most widely used testing procedure for dynamic property measurement at strain rates between 10^2 to 10^4 s⁻¹.

In this study, cylindrical test specimens of diameter 2.5 mm and length 4.5 mm were cut from the consolidated samples using EDM. Room temperature, high strain rate experiments were performed using the SHPB apparatus to study compression behavior of tantalum at dynamic loading conditions.

RESULTS AND DISCUSSION

Plasma Pressure Compaction of Tantalum Powders

The particle size of tantalum powder was analyzed by using BET. The surface area of the particles was calculated by measuring the amount of gas adsorbed on the surface. The measured surface area can be used to estimate average particle size by assuming that particles

are spherical. Table 1 shows surface area and average particle size of the tantalum powders consolidated by P²C.

Table 1 BET analysis of the tantalum powders.

Powders	Gas	Surface Area	Average Particle Size
Coarse Tantalum	Nitrogen	0.024 m ² /g	15µm
Nano Tantalum	Nitrogen	5.772 m ² /g	63 nm

The tantalum powders with particle size 15µm and 63 nm were consolidated at 2000 °C and 1750 °C, respectively at the same pressure and isothermal holding time by Plasma Pressure Compaction (P²C). The consolidation conditions and resulting densities are shown in Table 2. The results show that tantalum powders can be consolidated to high density in a short cycle time in the P²C process.

Table 2. Effect of sintering parameters on density of consolidated samples.

Powder Type	Temperature °C	Holding Time min	Pressure MPa	Density %
Ta (15µm)	2000	< 1	50	95
Ta (63 nm)	1750	< 1	50	95

Plasma Pressure Compaction is a novel consolidation technique capable of rapid densification by surface activation, particle welding, and plastic deformation mechanisms. The P²C consolidation of both coarse and nanocrystalline tantalum powders showed that the densification is enhanced as the particle size is reduced from 15µm to 63 nm. Nanocrystalline tantalum powder was consolidated to 95 % of the theoretical density at 250 °C below the sintering temperature required to achieve the same density with coarse tantalum powder. In both cases, the total cycle time required to consolidate tantalum powder was less than 10 minutes which is considerably shorter than conventional resistance sintering techniques.

The nanocrystalline particles have very high surface area which promotes densification. However, the greater intrinsic driving force associated with nanocrystalline particle size is not always available for sintering. Nanocrystalline particles have high cohesive force due to Van der Waals' attraction resulting in formation of agglomerates. Agglomerated particles resist compressive force by forming large interagglomerate voids which may not be broken up during the formation of the green body. In addition, nanocrystalline powders are so reactive that appreciable oxidation and adsorption of various gases occur upon exposure to the atmosphere. Surface oxides act as a diffusion barrier between particles and volatile contaminants cause outgassing during consolidation.

Despite such difficulties, P²C was able to rapidly consolidate nanocrystalline tantalum powder to high density. The successful densification of the nanopowder can be attributed to activating sintering mechanisms in the P²C process. In the initial stage of P²C, a pulsed DC voltage is applied directly through the powder compact. Electrical discharge takes at the interparticle gaps in the form of arc, plasma, and sparks which promote removal of oxides and other contaminants from the particle surfaces by generating significant heat and pressure [3]. Subsequently, external pressure and direct application of DC voltage accelerate the densification by inducing localized heating and plastic deformation at the interparticle contact areas.

Nevertheless, the SEM and EDAX results indicate that oxide phases are present in the nanocrystalline tantalum (Figure 1). Dense nanocrystalline tantalum without oxide phases may be produced by optimizing P²C consolidation parameters such as the pulsed DC voltage application (current, voltage, on-time, off-time, and baseline), constant DC voltage application (current density, voltage, and alternating polarity) heating rate, temperature, pressure, and isothermal holding time.

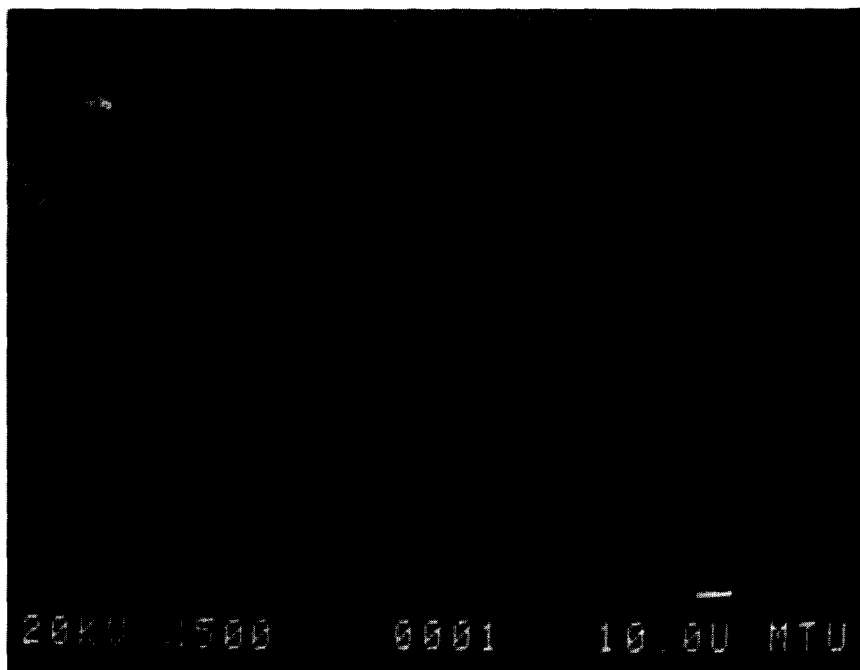


Figure 1 SEM picture of nanocrystalline tantalum consolidated by P²C. Dark areas indicate oxide phases

Dynamic Compression of Consolidated Tantalum Specimens

At high strain rates, both coarse tantalum and nano-tantalum exhibited ductile deformation behavior without significant brittleness/fracture. Stress-strain curves for the tantalum specimens are shown in Figures 2 and 3. Flow stress was essentially independent of strain rate, especially in the coarse tantalum specimen. This is in contrast to previous results wherein the flow stress of tantalum with grain size of $\sim 3 \mu\text{m}$ and its alloys has been observed to increase with increasing strain rate ($3000 - 10^3 \text{ s}^{-1}$) due to the high Peierl's stress in bcc metals [4]. Dynamic deformation testing should be conducted at a wider range of strain rate to confirm this result.

Tantalum normally exhibits significant strain hardening which is a function of the strain rate and pre-straining (cold work). Strain hardening is also found to be strongly dependent on texture and strain rate. However, strain hardening behavior was not observed in the P²C consolidated tantalum specimens at high strain rates. Moreover, nano-tantalum tends

to soften at high strain rates whereas coarse tantalum maintains its flow stress. It has been shown that prestraining reduces the hardening rate and limits the tensile failure strain [5]. Thus, plastic deformation occurring during the P²C process may have contributed to the dynamic compression behavior.

The compressive true stress-true strain response of tantalum consolidated from 15 μ m powder was found to be dependent on the applied strain rate (Figure 2). The strain increased with increasing strain rate while flow stress remained relatively constant. True strain between 0.11-0.18 was observed at a flow stress of 900-1000 MPa when compressed at strain rates between 1000-2500/s. The results correspond well with previous studies on dynamic compression properties of tantalum [5]. However, the lack of strain hardening observed in this P²C consolidated tantalum may be an advantage for high deformation rate applications.

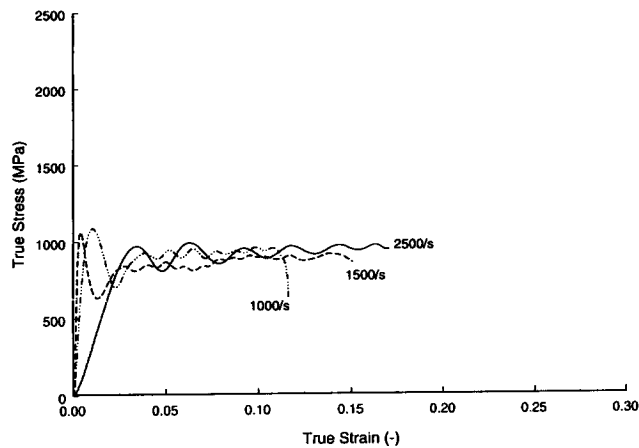


Figure 2 True stress-true strain curve of tantalum consolidated from coarse powder.

Ductility and flow stress increased dramatically as initial particle size is reduced below 100 nm. As shown in Figure 3, true strain and flow stress increased to 0.7 and 2400 MPa upon impact at strain rates of 9000/s and 12000/s. To the authors' knowledge, such a large strain at high flow stress has not been observed in randomly-oriented polycrystalline tantalum. The increase in flow strength of tantalum can be explained as follows. (1) grain boundary strengthening due to decrease in grain size as predicted by the Hall-Petch relation, and (2) presence of oxide phase detected in the nano-tantalum specimens.

Tantalum's properties, such as yield behavior, are highly sensitive to impurity and oxygen contents [4]. Typically, presence of oxide phase in the tantalum may increase the flow strength, but with a dramatic reduction in ductility because oxides are usually hard and brittle fracture initiating phases. In contrast, a significant increase in ductility has been observed in the rapidly consolidated nanocrystalline tantalum specimen despite a large amount of oxide phases. It is speculated that ductility of nano-tantalum is strongly influenced by its ultrafine grain size rather than the oxide phases present in the specimen. Even greater ductility may be achieved if the P²C consolidation parameters are optimized to remove most

of the oxygen in the initial particles. Further study must be conducted to understand the deformation mechanisms of the P²C consolidated nanocrystalline specimen.

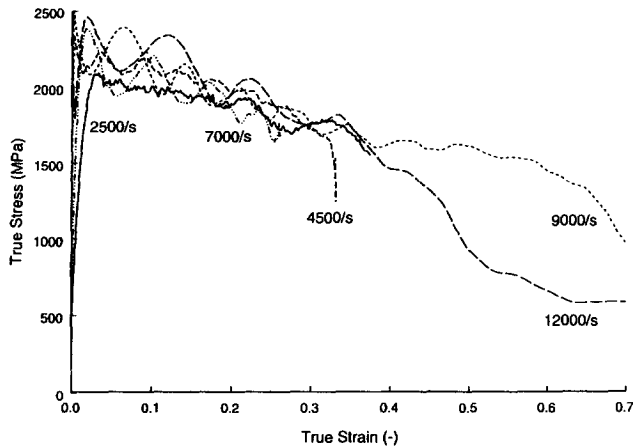


Figure 3 True stress-true strain curve for tantalum consolidated from nanocrystalline powder.

SUMMARY

- Nanocrystalline tantalum powders were consolidated rapidly to high density by P²C at a temperature 250 °C lower than coarse tantalum powder due to large intrinsic driving force associated with fine particles.
- Increased flow stress was not observed with increasing strain rate as is common in BCC metals with a high Peierl's stress.
- P²C consolidated tantalum is suitable for high strain deformation applications because it does not exhibit strain hardening behavior.
- Flow stress and ductility were increased dramatically in the nanocrystalline tantalum with an early onset of softening despite the significant amount of oxide phase.
- Further investigation is required to understand the deformation mechanisms at high strain rates.

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