ANNEALING STUDIES OF PURE AND ALLOYED TANTALUM
EMPLOYING ROCKING CURVES

David W. Richards¹, Michael P. Kramer¹, Joel W. House¹
and Robert J. De Angelis²

¹ Air Force Research Laboratory, Eglin AFB, Florida, 32542
² University of Florida/GERC, Shalimar, Florida, 32579

Abstract

To control the mechanical response of pure and alloyed tantalum requires tailoring the microstructure
by carefully controlling thermo-mechanical processing. In this investigation X-ray rocking curves were
employed to characterize the microstructure and further the understanding of the mechanisms of
annealing. Plate material produced from the alloyed tantalum demonstrated annealing stages 300ºC
higher than the pure tantalum. Activation energies determined from the rocking curve data were one-
half to one third of the self-diffusion activation energy.

Introduction

Controlling texture and grain structure in unalloyed tantalum has been difficult [1,2]. Sources of the
inhomogeneous structure are ingot chemistry, ingot grain size and mechanical processing. Introduction
of high levels of plastic strain by rolling and forging prior to annealing only partially homogenize the
grain structure. Heterogeneous grain structures in components generate local variations in mechanical
properties and are potential sources for unanticipated failures.

The annealing response of forged tantalum, reported by O’Brien et al. [1], demonstrated that
significant differences in grain size, size distribution, and morphology existed in microstructures
obtained after the annealing of forged tantalum. In a recent study the effects of an intermediate
annealing cycle (before forging) on texture and the microstructure formation in forged and annealed
tantalum plates were quantified [3]. The current investigation examines the benefits of X-ray rocking
curves in studying the processes of annealing in tantalum and tantalum 2.5% tungsten.

Material and Thermo-Mechanical Processing

Chemical composition of the pure tantalum studied is listed in Table I. The material was purchased
from H.C. Starck Inc., Newton, MA. The composition of the tantalum 2.5% tungsten alloy was the
same except for the additional tungsten. Two 165 mm (6.5 inch) diameter ingots were produced at
Starck utilizing a process consisting of vacuum arc remelting of a dual electron beam processed
electrode ingots.

Table I. Composition of the tantalum. (PPM by weight)

<table>
<thead>
<tr>
<th>O</th>
<th>N</th>
<th>C</th>
<th>H</th>
<th>Fe</th>
<th>Ni</th>
<th>Cr</th>
<th>Cu</th>
<th>Si</th>
<th>Ti</th>
<th>Mo</th>
<th>W</th>
<th>Nb</th>
</tr>
</thead>
<tbody>
<tr>
<td>72</td>
<td>11</td>
<td>13</td>
<td>1</td>
<td>24</td>
<td>29</td>
<td>11</td>
<td>1</td>
<td>8</td>
<td>5</td>
<td>12</td>
<td>117</td>
<td>43</td>
</tr>
</tbody>
</table>

The cast ingots were turned and cold swaged to 63 mm (2.5 inch) diameter bars. The swaged bars
were sectioned into 86 mm (3.4 inch) long billets. The billets were upset forged in multiple steps with
the application of lubrication between each step to 228 mm (9-inch) diameter plates by 6.35 mm (0.25
inch) thick.
This document was presented at the Denver X-ray Conference (DXC) on Applications of X-ray Analysis.

Sponsored by the International Centre for Diffraction Data (ICDD).

This document is provided by ICDD in cooperation with the authors and presenters of the DXC for the express purpose of educating the scientific community.

All copyrights for the document are retained by ICDD.

Usage is restricted for the purposes of education and scientific research.

DXC Website — www.dxcicdd.com

ICDD Website — www.icdd.com

ISSN 1097-0002
The post-forge annealing study was performed on samples from two plates, one of each composition. Employing a water-jet cutter, twenty specimens approximately two by two centimeters were sectioned from each plate. These specimens were cut from a circle of material centered on a 64 mm (2.5 inch) radius of both plates.

Hardness measurements were made on the surface of the plate. X-ray measurements and optical observations were made on the mid-plane of specimens that were annealed for one hour in vacuum at intervals of 50°C at temperatures between 750 and 1300°C for tantalum and 900 and 1500°C for tantalum 2.5% tungsten. The specimens were wrapped in tantalum foil and annealed in a furnace under a vacuum of $<10^{-5}$ torr.

**Experimental**

After heat treatment, hardness data were collected from the annealed specimens of tantalum and tantalum 2.5% tungsten. Rockwell B hardness numbers as a function of annealing temperature are shown in Fig. 1. Rapid softening of pure tantalum set in at 900°C and annealing was complete at 1150°C. For the alloyed tantalum softening started at temperatures above 1050°C, was most rapid in the 1150°C temperature range and was complete at 1350°C. The hardness increase observed in specimens annealed at temperatures above 1350°C indicated oxygen contamination occurred at these higher temperatures.

The annealed specimens were sectioned on their mid-plane using a diamond saw. The split specimens were mounted and metallographically polished using the technique described by Kelly et al. [4]. All of the X-ray data was collected from the mid-plane surface of the annealed specimens employing copper K$_\alpha$ radiation. The X-ray data were collected on a Siemens diffractometer outfitted with a split cradle and a graphite diffracted beam monochromator. The X-ray beam geometry on the specimen at sixty degrees two theta was one centimeter by one millimeter.

Rocking curve data were collected in steps of 0.02 degrees and two second counting times. Texture data were collected in five degree steps of phi and chi to describe the {110}, {200} and {211} pole figures. The pole figure data was processed employing the analysis software package popLA [5] to calculate the inverse pole figure. Inverse pole figure data were analyzed using the techniques reported in [6,7] to give the fraction of grains in the plane of the plate that have normals with orientations within 12.5 degrees of the $<111>$, $<110>$ and $<100>$ directions. These quantitative texture results

![Fig. 1. Rockwell B Hardness of Tantalum and Tantalum 2.5% Tungsten Versus Annealing Temperature.](image1)

![Fig. 2. Densities of Planes within 12.5 Degrees of $<111>$ and $<100>$ in Tantalum Versus Annealing Temperature.](image2)
Fig. 3. Densities of Planes within 12.5 Degrees of <111> and <100> in Tantalum 2.5% Tungsten Versus Annealing Temperature.

from the pure tantalum and alloy plates were plotted versus annealing temperature and are shown in Figs. 2 and 3 respectively. Texture strengths of the <110> components were less than 0.01 times random and were not included in these figures.

During the initial stage of annealing of the pure tantalum (see Fig. 2) in the temperature range of 750°C to 900°C, where the softening rate is at its maximum, there is an increase in the {100} component. In this same temperature range the {111} texture strength decreases by about a factor of four. Annealing at temperatures of 900°C to 1100°C decreases the fraction of grains with {100} orientation and increased the {111} component more than a factor of four. At annealing temperatures above 1100°C preferred growth of the {111} grains occurs producing texture strengths greater than eight times random and the magnitudes of the {100} components are below 0.3 times random.

The texture changes demonstrated by the alloyed tantalum (see Fig. 3) during annealing were in general similar to those observed in pure tantalum. The strength of the <111> component increases at higher annealing temperatures and the <100> component decreases. However the magnitude of the changes in texture strength during annealing were lower in the alloyed material.

Fig. 5. (222) Rocking Curve of Tantalum After Annealing at 950°C.

Fig. 6. (222) Rocking Curve of Tantalum After Annealing at 1000°C.
Rocking curves of the (222) obtained from the pure tantalum in the “as forged” and after annealing at 850°C are shown in Fig. 4. The (222) rocking curves for the tantalum after annealing at 950°C and 1000°C are shown in Figs. 5 and 6. Intensity spikes were observed to form on the rocking curve obtained from the material annealed at 850°C. These spikes are from poligonized regions of the structure. These occurred in an annealing temperature range in which the hardness was just starting to decrease. After annealing at 950°C and 1000°C the spikes increased in intensity, as the scale of the ordinate in Figs. 5 and 6 indicate, signifying the formation of larger diffracting crystallites.

(222) Rocking curves from the tantalum 2.5% tungsten alloy in the “as forged” condition and after annealing at 1050°C are shown in Fig. 7. The (222) rocking curves for the alloyed material after annealing at 1150°C and 1500°C are shown in Figs. 8 and 9. Intensity spikes of about one-tenth of those at 1150°C were observed to form on the rocking curve obtained from the alloyed material annealed at 1050°C. These spikes indicated that poligonization was occurring in the alloyed material at a temperature 300°C higher than in the pure tantalum.

The averages of the intensity spikes present in the (222) rocking curves from the pure and alloyed tantalum were determined at each of the annealing temperature. The averaging was accomplished by subtracting the background intensity from a pattern, performing a peak search to identify seventy to one hundred peaks, summing the intensity values at the peak maximums and dividing by the number of peaks. Plots of the logarithms of the magnitude of the intensity spike averages versus the reciprocal of the annealing temperature are shown in Figs. 10 and 11. Activation energies for the mechanism controlling poligonization were calculated from the slopes of these plots. The activation energy was determined to be 29,200 calories per mole for pure tantalum and 55,800 calories per mole for tantalum 2.5% tungsten. The energy value for pure tantalum, about one-third of the activation energy of self-diffusion, is in the range of activation energy values for boundary diffusion [8]. The much greater activation energy for boundary migration observed for the tantalum 2.5% tungsten indicates the additional tungsten solute atoms reduce the mobility of the migrating boundaries significantly.

Fig. 7. (222) Rocking Curves of Tantalum 2.5% Tungsten in the “As Forged” Condition and After Annealing at 1050°C.

Fig. 8. (222) Rocking Curve of Tantalum 2.5% Tungsten After Annealing at 1150°C.
Discussion

There are two advantages in employing rocking curves to investigate the annealing behavior of tantalum and its alloys. The most apparent advantage was the appearance of intensity spikes in the rocking curve patterns. The lowest annealing temperature that was associated with the appearance of the intensity spikes defined the annealing temperature that produced significant recovery of the deformed substructure. Annealing at higher temperatures provided a method to study the crystallite size growth kinetics. The second and less obvious advantage of the rocking curve technique is the data it provides on the grain orientation relationships between the deformed structure and the recovered or recrystallized material. Nucleation of the recrystallized grains in both tantalum materials investigated took place within ten degrees of the center of the intensity distribution of the rocking curves. The magnitude of the intensity spikes in this angular range is proportional to the strength of the <111> component of texture. This is easily seen by comparing the intensities of the intensity scales on the plots of the rocking curves with the texture strengths of the <111> shown in Figs. 2 and 3.
Acknowledgements

The financial support from the United States Air Force for RJD under contract FO8635-98-D-0016 is acknowledged.

References