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Correlation between the microstructure studied by X-ray line profile analysis and the strength of high-pressure-torsion processed Nb and Ta

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Abstract

Niobium and tantalum are two body centered cubic metals with very different elastic anisotropy. The $A_z = 2 \times c_{44}/(c_{11}-c_{12})$ constant for Nb and Ta is 0.51 and 1.58, respectively. The submicron grain-size state of the two refractory metals was produced by the method of high-pressure torsion with different pressure values of 2 and 4 GPa for Nb, and 4 and 8 GPa for Ta, and two different deformations of 0.25 and 1.5 rotations, respectively, with equivalent strains of up to ~40. The dislocation density and the grain size were determined by high-resolution diffraction peak-profile analysis. The beam size on the specimen surface was 0.2×1 mm, allowing the sub-structure along the radius of the specimen to be characterized. The strength of the two metals was correlated with the dislocation density and the grain size. It is found that, though the grain size is well below 100 nm, the role of dislocations in the flow stress of these two metals is significantly greater than that of the grain size.

Keywords: Tantalum; Niobium; High-pressure torsion; Dislocations; Grain size

1. Introduction

Niobium is used for superconducting radio frequency cavities in particle accelerators and in special boiler or pressure vessels [1,2]. Tantalum is a biocompatible, high-melting-point metal with surprisingly good ductility [3]. The elastic anisotropy of two body centered cubic (bcc) metals is very different. The Zener constant, $A_z = 2c_{44}/(c_{11}-c_{12})$, in the case of Ta is larger than unity ($A_z(\text{Ta}) = 1.58$), similar to most of the pure metals, whereas for Nb, $A_z(\text{Nb}) = 0.51$ [4,5]. Work hardening of the two metals was studied extensively in terms of the dislocation structure [6–11] and the grain size [9,11–16]. The flow stress σ of both Nb and Ta was investigated by several authors as a function of the grain size in terms of the Hall–Petch relation [9,11,12,15,17–19]:

$$\sigma = \sigma_0 + \frac{K}{\sqrt{d}} \tag{1}$$

where σ_0 is the elastic limit, K is the Hall–Petch constant [20,21], and d is the average grain size (or sub-grain size). The Hall–Petch constants determined by different authors in Nb and Ta are listed in Table 1. The values of K vary between ~4 and 20 for Nb and between ~7 and 22 for Ta. The range of the grain size corresponding to the different K values is also very different, it varies from the 100 μm range in Nb [12] down to the 20 nm range in Ta [15]. The dislocation structures are also very different in the different states of the materials where the Hall–Petch constants are different. Conrad and Jung [23] and Chinh and coworkers [24] have shown that, in different grain-size ranges, the grain-size relation works better with different exponents.

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Table 1
Hall–Petch constants K in Nb and Ta determined by different authors; all values are given in $GPa\sqrt{nm}$ units.

Nb	Ta
3.83 [17]	7.15 [9]
12.5–16[b] [18]	17.6 [14]
20 [12]	22.17[a] [15]
3.8 (Present work[c])	13 (Present work[c])

[a] This value is obtained from the first two data-points in Fig. 3 in Ref. [15]. The data-points were published in Refs. [11,22].
[b] The smaller and larger values correspond to the lower and upper yield points [18].
[c] These values are obtained by evaluation type (ii) of Eq. (8a) with $\alpha = 0$, i.e. ignoring the contribution of dislocations to the flow stress; see also Table 4.

It is claimed that, when the grain size d_{GS} is larger than ~1 μm, the grain-size relation works well with the usual −0.5 exponent. In the smallest grain-size regime, however, it is suggested that the exponent of −1 is more appropriate. The exponent of −1 was also observed and suggested by Dubravina et al. [25] in strongly deformed, submicron-grain-size Cu. In Ref. [25], the substructure was characterized by X-ray line profile analysis in terms of dislocation densities and crystallite size. Here the "crystallite-size" was obtained from the X-ray column-length distribution assuming log-normal size distribution [26,27]. One must distinguish between grain size determined by transmission or scanning electron microscopy (TEM or SEM), d_{TEM} or d_{SEM}, and that obtained by X-ray line profile analysis. It was shown in Ref. [28] that, when d_{TEM} or d_{SEM} is in the range of 100 nm or smaller, the X-ray crystallite size coincides with these values. More about the evaluation of X-ray crystallite size is given in Section 3. The size dependence of grain-size contribution to the flow stress is discussed in more detail in Section 4.3.

The present work investigated the hardening of Nb and Ta by high-pressure-torsion (HPT), applying different pressure values. The sub-structure of the specimens was determined by the method of X-ray line profile analysis. Dedicated, high-angular resolution X-ray micro-diffraction was applied with the spatial resolution of ~0.2 × 1.5 mm. The dislocation density and arrangement, and the grain size were determined as functions of deformation and correlated with the flow stress measured by hardness tests. The mechanical properties are discussed in terms of the dislocation structure and the grain size by analyzing the data with the Taylor [29] and the Hall–Petch [19,20] equations.

2. Experimental

2.1. Samples

High-purity Nb and Ta discs 8 mm in diameter and ~0.5 mm thick were prepared by HPT. The HPT deformation was carried out applying different pressure values of 2 and 4 GPa in the case of the Nb, and 4 and 8 GPa in the case of the Ta specimens, with two different rotations of 0.25 and 1.5, respectively. The sample notations are given as two numbers: *a–b*, where *a* is the applied pressure in gigapascals, and *b* is the rotation, e.g. Nb-2-0.25 is the Nb specimen deformed by 0.25 rotation (i.e. $\pi/2$) with the application of 2 GPa pressure. Since the deformation changes systematically in the radial direction of the samples, both the X-ray and the mechanical tests were carried out at different positions, and all the data are presented as a function of the von Mises equivalent-deformation ε_{vM} at different radial distances, where

$$\varepsilon_{vM} = \frac{2\pi X r}{h\sqrt{3}} \tag{2}$$

where X is the number of rotations, r is the radius, and h is the thickness of the sample. The applied rotations provide up to $\sim\varepsilon_{vM} = 40$ in the edge regions of the samples deformed by 1.5 rotations.

2.2. X-ray diffraction experiments

X-ray diffraction measurements were carried out in a special high-resolution diffractometer dedicated to line profile analysis with a plane Ge (220) primary monochromator operated at the Cu Kα fine focus rotating copper anode (Nonius, FR-591) at 45 kV and 80 mA [30]. The distance between the source and the monochromator was 240 mm, and a slit of ~160 μm was placed before the monochromator, at a distance of 200 mm from the X-ray source. At this distance, the $K_{\alpha 1}$ and $K_{\alpha 2}$ components were received by the Ge (220) crystal at a large enough separation allowing for cutting off the $K_{\alpha 2}$ component by the 160 μm wide slit. The size of the Cu Kα1 beam was ~0.2 × 1.5 mm on the specimen surface. The scattered radiation was registered by three flat imaging plates (IP) with linear spatial resolution 50 μm. The IP were placed at distances of 196 mm from the specimen, covering the angular range between $2\theta = 30$ and 140°. The distances between the specimen and detector were selected such that the instrumental effect was always <10% of the physical broadening. The diffraction geometry is of parallel-beam type, and therefore the specimen does not have to be moved while the angular resolution is sufficiently good over the entire angular range of measurement. The diffraction patterns were obtained by integrating the intensity distributions along the corresponding Debye–Scherrer arcs on the IP. Only the central parts of the arcs were used for the integration where the curvature does not yet affect line broadening. The X-ray beam was positioned on the specimen surface using a low-depth-resolution microscope coupled to a TV screen. The specimens were placed at three positions in front of the beam, as shown schematically in Fig. 1. The measurements were done at the center, at half radius and close to the edge of the samples, denoted by $R0$, $R1/2$ and $R1$, respectively, where $R \cong 4$ mm. The gray disc and the green rectangles showing the specimen and the beam positions in Fig. 1 are in scale. Typical IP readouts of