Journal of Nuclear Materials 453 (2014) 1–7

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Defects interaction processes in deformed high purity polycrystalline molybdenum at elevated temperatures

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ARTICLE INFO

Article history:
Received 11 April 2014
Accepted 16 June 2014
Available online 23 June 2014

ABSTRACT

Mechanical spectroscopy (damping and elastic modulus as a function of temperature) and transmission electron microscopy studies have been performed in high purity polycrystalline molybdenum plastically deformed to different values of tensile and torsion strain. Mechanical spectroscopy measurements were performed from room temperature up to 1285 K. A relaxation peak in polycrystalline molybdenum related to the movement of dislocations into lower energy configurations near grain boundaries has been discovered to appear around 1170 K. The activation energy of the peak is 4.2 eV ± 0.5 eV. This relaxation phenomenon involves the interaction between vacancies and mobile dislocations near the grain boundaries. It should be highlighted that this relaxation process is controlled by the arrangement of vacancies and dislocations which occur at temperature below 1070 K.

1. Introduction

A broad range of alloys including austenitic stainless steels, ferritic–martensitic steels, refractory metals and titanium alloys have been investigated in extensive international materials testing programmes in order to identify potential candidates for the so-called first-wall region and other parts of a controlled thermonuclear reactor [1,2]. In particular, molybdenum has a high melting point, a high specific heat, good corrosion and creep resistance and strength at high temperatures. In addition, it has a relatively low thermal neutron cross section. These qualities make molybdenum attractive for the use in the nuclear industry [3]. Nuclear materials are exposed to irradiation and the same time to external stresses; therefore it is of great importance to understand the mechanisms of interaction between the defects into the material, in order to predict the long-time behaviour of these materials. Several works have been reported in the past 50 years about the mechanical properties in molybdenum and its associated recovery stages [4–14].

The temperature range around one third of the melting temperature (0.3 Tm, 865 K), usually related to stage V of recovery, is particularly interesting in molybdenum due to the strong influence on the mechanical properties of both the pure metal and technological molybdenum-based alloys. In fact, in non-irradiated and irradiated molybdenum at annealing temperatures within stage V (temperatures higher than 850–900 K), the yield stress and the ultimate tensile strength begin to decrease [9,11,15].

Mechanical spectroscopy (MS), referred to as the internal friction method in the early literature, offers unique opportunities to study the mechanical energy losses due to the dislocation arrangement produced by deformation and its interaction with point defects [16].

The internal friction in molybdenum single crystals has been extensively discussed in the 50–1300 K temperature range, see for instance Benoit [17], Seeger [18], Zelada et al. [19–21] and Lambri et al. [22]. In particular, for temperatures higher than stage IV (600 K to 850–900 K) we reported that single crystalline deformed molybdenum exhibits two damping peaks. The physical mechanism which controls the damping peak appearing at around 700–800 K (so called the LTP) was related with the dragging of jogs by the dislocation under movement assisted by vacancy diffusion [19,20,22]. The damping peak which appears at high temperatures of around 950–1000 K (so called the HTP) was controlled by the formation and diffusion of vacancies assisted by the dislocation movement [20]. In addition, in plastically deformed and electron plus neutron irradiated high purity single crystalline molybdenum,

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http://dx.doi.org/10.1016/j.jnucmat.2014.06.027
0022-3115/

oriented for single slip, the intermediate temperature damping peak (so called the ITP) was detected at around 900 K. It was related to the interaction of dislocations lines with both prismatic loops and tangles of dislocation [21].

Nevertheless, little work is done on the internal friction of polycrystalline molybdenum above room temperature. Consequently, our aim is to study the mechanical energy losses in plastically deformed polycrystalline molybdenum from room temperature up to near 45% of the melting temperature (0.45 Tm). In addition, as it will be shown through the paper, the present results shed light on the physical driving force controlling the behaviour of the yield stress, ultimate tensile stress and ductile to brittle transition temperature in neutron irradiated molybdenum within the temperature range of stages IV and V of recovery during post-irradiation annealing.

2. Experimental

The samples used in this work were prepared from a single batch of “Amax Specialty Metals Corporation” low carbon arc cast 3/8” diameter rods in A.E.R.E, Harwell, UK. The specifications of this starting material were: C 0.004%, O_2 0.0004%, H_2 0.0001%, N_2 0.0001%, Fe 0.002%, Ni 0.001% and Si 0.002%. Samples were decarburized for 48 h by heating them, using an R.F. induction furnace, to 1873 K in a partial pressure 8×10^{-3} Pa of oxygen. The oxygen atmosphere was maintained by continuously bleeding spectroscopically pure oxygen through a leak valve. The carbon removal was monitored by the amounts of CO and CO_2 in the residual vacuum by using a mass spectrometer. After the decarburization process the samples were annealed at 2073 K for 24 h in a vacuum better than 10^{-5} Pa to remove the oxygen.

The average grain size was 50 μm. No appreciable change in grain size could be detected after the heat treatments during the performed measurements in the present work. The samples were sheets of 20 mm length, 0.4 mm thickness and 2.5 mm width.

Samples were deformed in tensile at a constant speed of 0.03 cm/min, followed by 1.5% torsion at room temperature.

Damping, Q^{-1} (or internal friction), and natural frequency were measured in an inverted torsion pendulum, under a vacuum of about 10^{-5} Pa. The maximum strain on the surface of the sample was 5×10^{-5}. The measurement frequency was around 1 Hz except in the determination of the frequency dependence of the peak temperature. The heating and cooling rates employed in the tests were of 1 K/min. A heating and its corresponding cooling run will be called hereafter a thermal cycle. There was no hold time once the maximum temperature had been achieved, during the thermal cycle.

During the run -up and -down in temperature, Q^{-1} was calculated from the slope of the straight line which results from the least squares fitting of the natural logarithm of all the decaying amplitudes versus time, such that

$$\ln(A_n) = \ln(A_0) - \pi Q^{-1} n \qquad (1)$$

where A_n is the area of the nth decaying oscillation, A_0 is the initial area of the starting decaying oscillation and n is the period number. For all these measurements the same initial and end values of the decaying amplitudes were used for eliminating some possible distortion for the appearance of amplitude dependent damping effects [23].

Amplitude dependent damping (ADD), i.e. damping as a function of the maximum strain on the sample, ε_0, was calculated from Eq. (2) [19,23,24]

$$Q^{-1}(\varepsilon_0) = -\frac{1}{\pi}\frac{d(\ln(A_n))}{dn} \qquad (2)$$

The decaying of the oscillations were performed at constant temperature ($T \pm 0.5$ K). Polynomials were fitted to the curve of the decaying areas of the torsional vibrations as a function of the period number by means of Chi-square fitting. Subsequently the Eq. (2) was applied. Polynomials of degree higher than 1 indicate that Q^{-1} is a function of ε_0, leading to the appearance of ADD effects, as it can be inferred easily. This procedure allows obtaining the damping as a function of the maximum strain (ε_0) from free decaying oscillations [19,23,24]. The degree of fitted polynomials was smaller than 3.

The strength of ADD effects is measured through S parameter, such that [19,23,24].

$$S = \frac{\Delta Q^{-1}}{\Delta \varepsilon_0} \qquad (3)$$

For transmission electron microscopy (TEM) examinations, thin foils were prepared with the double jet technique using 12% H_2SO_4 in methyl alcohol. Observations were carried out in a Phillips CM200 transmission electron microscope operated at 200 kV.

3. Results

3.1. Mechanical spectroscopy

Fig. 1 shows the damping spectra measured from 800 K up to 1285 K, for a polycrystalline sample deformed 1% in tensile and 1.5% in torsion at room temperature. Full symbols are for heating runs and the corresponding empties ones, are for cooling. In the first run up to 1240 K (full rhombus in the figure) the samples showed an increasing background with temperature. Nevertheless, on cooling a small damping hump around 1170 K appears. For the next heating run a step like damping is seen at this temperature (squares in the figure). During the following heating run (full circles) a damping peak develops which is clearly observed at 1170 K, but during cooling the peak disappears. In the following runs, for thermal cycles from room temperature up to 1285 K the peak stabilizes at around 1170 K (for the last spectrum, triangles, only the heating run is shown), but it is always absent during the cooling part of the cycles. The HTP appearing in single crystals cannot be clearly observed, although a hump around 950 K is present in the spectra which presents the damping peak at 1170 K, see full circles and triangles in Fig. 1.

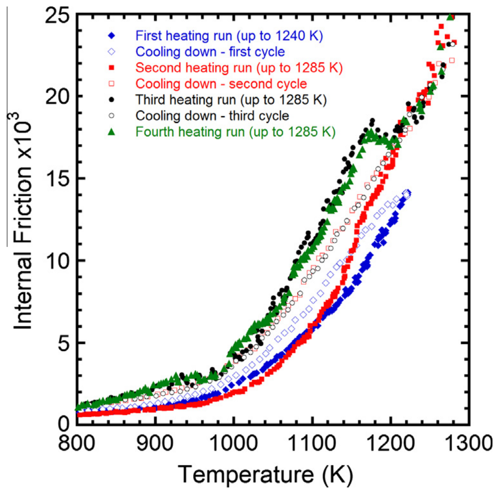

Fig. 1. Damping spectra measured during successive thermal cycles for a polycrystalline molybdenum sample deformed 1% in tensile and 1.5% in torsion at room temperature. Full symbols: heating part of thermal cycle. Empty symbols: cooling part of thermal cycle.