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Interaction processes between vacancies and dislocations in molybdenum in the temperature range around 0.3 of the melting temperature

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ABSTRACT

Mechanical spectroscopy, electrical resistivity and transmission electron microscopy studies have been performed on pre-strained neutron irradiated single crystalline molybdenum in order to check the interaction processes between vacancies and dislocations in the temperature range between room temperature and 1273 K. The anelastic relaxation in molybdenum which appears between 800 K and 1273 K has been separated in two different physical mechanisms depending on the temperature of appearance of the relaxation peak. The physical mechanism which controls the damping peak appearing at around 800 K was related with the dragging of jogs by the dislocation under movement assisted by vacancy diffusion. The damping peak which appears at higher temperatures of about 1000 K was more consistent with the formation and diffusion of vacancies assisted by the dislocation movement.

1. Introduction

The production of electricity from controlled nuclear fusion represents one of the major scientific and technical challenges of the 21st century. Over the past 50 years, the main emphasis of fusion research has been the study of plasmas, the development of plasma theory and pursuit of the technology of magnetic plasma containment. However, over the past three decades, there has been an increasing emphasis on the identification and solution of problems associated with the structural materials, in particular those that will be situated closest to the plasma. The so-called first-wall region of a controlled thermonuclear reactor. 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 candidate materials for reactor applications [1].

In particular, molybdenum has a high melting point, a high specific heat, good corrosion, 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 [2].

Nuclear materials are exposed to external stresses and at the same time to irradiation, because of this, it is of great importance to understand the mechanisms of interaction between the defects produced, in order to predict the long-time behaviour of these materials. Several works have been reported in the past 50 years about the radiation damage behaviour in molybdenum and also about its associate recovery stages [3–13].

The temperature range around 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 both in the pure metal and in technological molybdenum-based alloys. In fact, neutron irradiation in molybdenum at temperatures below 1000 K followed by annealing at temperatures higher than 473 K leads to an increase in the yield stress, the ultimate tensile strength (UTS) and the ductile–brittle transition temperature (DBTT) [8,14]. At annealing temperatures within stage V (>900 K), the yield stress and the UTS begin to decrease.

Mechanical spectroscopy (MS), referred to as the internal friction method (IF) in the early literature, offers unique opportunities to study the mechanical losses due to the interaction, for instance, between dislocation and point defects produced during neutron irradiation in nuclear materials, as molybdenum [15]. The interaction between dislocation and point defects studied by internal

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friction in molybdenum has been extensively discussed in the 50–450 K temperature range, see for instance Benoit [16] and Seeger [17]. But very little work is done on the internal friction of molybdenum due to the interaction between dislocation and intrinsic point defects above room temperature. Our aim is to study the mechanical losses due to the interaction between dislocations and point defects from room temperature up to near one-third of the melting temperature (0.3 Tm) in deformed and irradiated molybdenum single crystals.

In the previous works [18,19], we found an intense relaxation peak at high temperature for single crystalline samples previously deformed at room temperature. The damping intensity of the peak depends on the degree of plastic deformation and once the peak appeared, after annealing at temperatures above that for vacancy diffusion, the damping values were independent of the amplitude of oscillation. The peak temperature of this relaxation increases with the prior annealing temperature of the sample. However, once the peak stabilises, it has an activation energy of 1.6 eV and 2.7–2.8 eV for peak temperatures of about 800 K and 1000 K, respectively. The activation energy of the relaxation is independent of the crystal orientation.

It has been shown that dislocations are responsible for the relaxation, but also intrinsic point defects as vacancies can be involved. Nevertheless, some open questions remain as: which are the defects involved in the relaxation peak and which mechanism produces these relaxations or energy losses?

In the present work we have studied, using MS, electrical resistivity (ER) measurements and transmission electron microscopy (TEM), the effect of room temperature neutron irradiation on pre-strained samples, with the aim of answering to the above questions and to contribute to the knowledge of the mechanisms, which produce mechanical losses in irradiated and cold-worked molybdenum.

2. Experimental

Twelve single crystals have been used in this work. They were prepared from zone refined single crystal rods of molybdenum in A.E.R.E., Harwell. The residual resistivity of the samples was about 8000, tungsten being the main residual impurity. Samples with the ⟨1 1 0⟩ and ⟨1 4 9⟩ crystallographic tensile axis have been selected to favour the deformation by multiple and single slip, respectively.

The samples used in the mechanical spectroscopy studies were sheets of 20 mm length, 0.2 mm thickness and 2 mm width. In contrast, the samples employed in the electrical resistivity measurements were single-crystal rods of 5 mm diameter and 50 mm length.

Samples were annealed and then deformed in extension at a constant speed of 0.03 cm/min, followed by torsion at room temperature. After the plastic deformation process the samples were irradiated, at room temperature, with neutrons. The status of the samples used is shown in Table 1.

Single crystals, after plastic deformation and mechanical spectroscopy tests, were checked by means of X-rays and metallographical study. Results indicated that the single crystalline state was not changed by the plastic deformation or annealing to the work temperatures.

In the mechanical spectroscopy, MS, measurements, damping (Q^{-1} or internal friction) and natural frequency were measured in an inverted torsion pendulum for free-decaying vibrations, under high vacuum of about 5×10^{-5} Pa; see Ref. [20] for a description of the experimental setup. The maximum strain on the surface of the sample was 5×10^{-5}. 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.

Table 1
Status of the used samples

Sample	Orientation	Elongation (%)	Torsion (%)	Irradiation time (h)
a/sheet	110	3	1	0
b/sheet	110	3	1	10
				10 re-irradiated*
c/sheet	110	3	1	20
d/sheet	149	5	1	0
e/sheet	149	5	1	20
				10 re-irradiated*
f/rod	110	3	1	10
g/rod	149	3	1	10

* Irradiated again after the mechanical spectroscopy measurements, see explanation in the text.

The electrical resistivity, ER, measurements were performed at room temperature by the eddy current decay technique [21]. ER values were measured both before irradiation and after irradiation as a function of the annealing temperature. Annealing treatments were performed by heating the sample at a rate of 1 K/min under pure argon at normal pressure followed by cooling into the furnace, under the same protective atmosphere.

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 with an energy dispersive X-ray spectrometer EDAX DX-4, operated at 200 kV.

Low flux neutron irradiation was performed at room temperature, at the Siemens SUR 100 nuclear reactor, RA-4, of the National University of Rosario – National Atomic Energy Commission of Argentina. The RA-4 was operated at 0.7 W. Samples were positioned in the horizontal channel, which passes through the reactor core inside of a cylinder of poly-methyl-methacrylate (PMMA) of 250 mm length and 25 mm diameter, with a wall and a bases thickness of 2 mm and 20 mm, respectively. The fluxes of thermal, epithermal and fast neutrons at the position where the samples were placed were 5.7×10^{11} n/m^2 s, 8.1×10^{9} n/m^2 s and 5.0×10^{11} n/m^2 s, respectively. The energy of the thermal neutrons was about 0.025 eV. Epithermal and fast neutrons had averaged energies of 50 KeV and 0.8 MeV, respectively. Taking into account the previous reported works [13,22], an estimation of the irradiation dose in dpa (displacement per atom) less than 1×10^{-5} could be done.

Considering that the maximum energy transfer, E, for a neutron of mass m and kinetic energy, E_o, emitting a lattice atom of molybdenum of mass M is [23,24]

$$E = \frac{4\,\mathrm{mM}}{(m+M)^2} E_o \tag{1}$$

we have calculated the number of displacements through the Kinchin–Pease model for hard sphere collisions [23], such that

$$\text{Number of displacements} = \frac{E}{2E_d} \tag{2}$$

where E is the transferred energy (56 KeV, according to Eq. (1)) to the primary knock atom corresponding to fast neutron damage (with $E_o = 0.8$ MeV) and E_d is the displacement threshold energy for molybdenum ($E_d = 37$ eV [25]). A value of 800 displacements was obtained. It is convenient to mention that the number of displacements calculated from the Kinchin–Pease model is a maximum limit attained for a temperature of 0 K, where recombination of defects does not occur. In addition, around 600 displacements are obtained if Eq. (2) is multiplied by 0.8, corresponding to the modified Kinchin–Pease model which takes into