Dislocations and point defects in neutron irradiated single crystalline Mo at elevated temperatures

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Abstract. At temperatures of about 0.3 \(T_m\), the mobility of dislocations out of thermodynamical equilibrium is controlled by the agglomerates of defects reducing their mobility. Annealing at \(T > 973\) K is needed for dissolving the agglomerates, allowing the dislocations start their movement. In this work we have related the results of mechanical spectroscopy, transmission electron microscopy, electrical resistivity, differential thermal analysis and small angle neutron scattering, as obtained in deformed and neutron irradiated Mo single crystals.

1. Introduction

Nuclear materials are exposed to external stresses and at the same time to irradiation and temperature. Because of that, it is of great importance to understand the behaviour of the defects produced as a function of temperature, in order to predict the long time behaviour of these materials. In the present work the interaction processes between dislocations and point defects, from room temperature (RT) up to 30\% of the melting temperature, \(i.e. \) 0.3 \(T_m\) are studied. We have related the results of mechanical spectroscopy (MS, internal friction or damping measurements), transmission electron microscopy (TEM), electrical resistivity (ER), differential thermal analysis (DTA) and small angle neutron scattering (SANS), as obtained in deformed and neutron irradiated molybdenum single crystals.

2. Experimental

Fifteen single crystals were prepared from zone refined single-crystal rods of Mo in A.E.R.E., Harwell, UK. Samples with the \((110)\) and \((149)\) crystallographic tensile axis have been selected to favour deformation by multiple and single slip, respectively; their orientations are shown in the inset of Fig. 1. The \((110)\) single crystals were stretched 3\% at RT and \((149)\) samples were stretched 5\% at RT. For the \((149)\) samples, this is enough to achieve the condition of single slip, as revealed by TEM studies and the fact that similar deformation produced single slip before \([1, 2]\). After plastic deformation samples were irradiated with neutrons (dose < \(10^{-5}\) dpa) at RT, at the nuclear reactor RA-4, UNR-CNEA, Argentina \([1]\). The details for MS, ER, DTA, TEM and SANS (D11, ILL, Grenoble, France) measurements are given elsewhere \([1, 3]\).
3. Results

During the first heating (Fig. 1), after the RT deformation, the sample showed an increasing background with temperature. Nevertheless, on cooling a well developed damping peak is present (peak A), which moves to a slightly higher $T$ and increases in intensity when heating to 973 K, but then stabilised at $\approx 800$ K (peak B). When the maximum annealing temperature, during the thermal cycles (heating run plus its cooling run), is increased to 1250 K, the peak moves to about 1000 K (peak C). Further runs to 1250 K decrease the intensity of the peak, which disappears if the sample keeps vibrating for long time at 1250 K.

Damping spectra for deformed and then irradiated (10 h) $\langle 110 \rangle$ sample show an increasing background with $T$ during the first heating, but the damping peak (peak D) appears on cooling down (Fig. 2). In this case the peak appears at lower $T$ and has smaller intensity than the one for non-irradiated samples. Annealing up to 973 K, produce a slight increase both in the peak temperature and peak height. Successive thermal cycles up to 1073 K stabilise the peak at around 800 K (peak E), being the relaxation temperature very similar to the non-irradiated samples although the peak is less intense. Further increase in the annealing temperature up to 1250 K leads to a strong shift in the peak and to a decrease in the peak height, in a similar way than for the non-irradiated sample (peak F). The behaviour of the damping spectra for the different runs for a sample $\langle 110 \rangle$ deformed and irradiated 20 h, is similar to the exhibited by the sample irradiated during 10 hours, although in this case the peak presents a final lower intensity due mainly to the decrease in the damping background (G and H spectra in Fig. 2). Deformed $\langle 149 \rangle$ samples present a damping peak whose intensity results smaller than in $\langle 110 \rangle$ samples. In addition, the irradiated $\langle 149 \rangle$ sample (during 20 hours) presents similar behaviour to the non-irradiated one [1].

In all irradiated samples, the damping peak practically disappears when the sample is vibrated for a few hours at 1250 K. A new re-irradiation at RT during 10 hours and subsequent annealing at 973 K for both $\langle 110 \rangle$ and $\langle 149 \rangle$ samples, restore the peak during the cooling run. A similar behavior is observed in the samples that were only re-deformed. The promotion of similar damping spectra after re-deformation and after re-irradiation could indicate that

Figure 1. Damping spectra for $\langle 110 \rangle$ deformed samples. Inset: Stereographic projection showing the orientation of the used single-crystals.

Figure 2. Damping spectra for $\langle 110 \rangle$ deformed plus neutron irradiated samples. Inset: TEM micrographs for deformed samples: (a) $\langle 149 \rangle$, (b) $\langle 110 \rangle$.  

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the relaxations peaks would develop in samples which are firstly irradiated and subsequently plastically deformed. However, it should be expected some change in the shape of the peak due to a different configuration of defects during the development of the relaxation process.

The spectra showed in Figures 1 and 2 are composed by a damping peak at ≈ 800 K and other peak at ≈ 1000 K. The physical mechanism controlling the peak at low-\( T \) was related to the dragging of jogs assisted by vacancy diffusion during the dislocation motion. In contrast, the peak at high-\( T \) was related to movement of dislocations controlled by the formation and diffusion of vacancies assisted by the dislocation movement [1, 3]. The damping response in the MS tests is in agreement with that exhibited by the ER, DTA (Fig. 3) and SANS studies. Indeed, the increase in the ER values from 300 K up to 600 K in both \( \langle 110 \rangle \) and \( \langle 149 \rangle \) samples, coincides with the stage III in irradiated samples, which was related both to the movement of vacancies and to the appearance of internal stresses [1, 3]. In fact, the appearance of five stages of recovery has been proposed for molybdenum depending of their temperature range. They can be defined as stage I, below 120 K; stage II, from 120 K to 330 – 350 K; stage III, from 330 – 350 K to 600 K; stage IV, from 600 K to 850 – 900 K and stage V, for temperatures higher than 850 – 900 K [2, 3, 4, 5, 6, 7]. Stages I and II are attributed to migration of interstitials, meanwhile stage III is attributed to the movement of vacancies. Stages IV and V are attributed to the decrease in the concentration of vacancies out of thermodynamic equilibrium, the dissolution of defects-agglomerates and the recovery of the dislocation structure.

Figure 3. The ER and DTA thermogram in irradiated samples during 10 hours: ● \( \langle 110 \rangle \), \( \Delta \langle 149 \rangle \). Empty symbols: prior to irradiation. ——: \( \langle 110 \rangle \) irradiated sample.

Figure 4. Lower and left axis: Radius of the agglomerates of vacancies for a 10 hours irradiated \( \langle 110 \rangle \) sample. Upper and right axis: \( I \) vs \( q \) curves.

4. Discussion
The micrographs for a deformed sample of type \( \langle 149 \rangle \) and \( \langle 110 \rangle \) after the MS tests (inset Fig. 2) and an analysis of constructive interference, allowed us to relate the \( \langle 110 \rangle \) plane with the sliding one and the Burgers vectors with the \( \langle 111 \rangle \) direction. After irradiation, dislocations are pinned by vacancy-type points defects producing a decrease in the damping background (Figs. 1 and 2). In addition, samples \( \langle 149 \rangle \) are less sensitive to irradiation than \( \langle 110 \rangle \), being the effect in the latter the reduction of the damping peak and the shift to lower temperatures. In contrast, in samples oriented for single slip the irradiation after deformation does not produce clear changes. The difference in the MS for irradiated \( \langle 110 \rangle \) and \( \langle 149 \rangle \) samples can be related to different
dislocation arrangement in each type of deformed single crystal. Indeed, samples oriented for multiple slip have more active slip systems and present higher density of dislocation, determined by counting the dislocation lines, more jogged and shorter dislocations than the ⟨149⟩ samples (see the inset of Fig. 2).

The temperature intervals where DTA reactions appear are in reasonable agreement with the salient features of the ER curve. The first endothermic peak at about 650 K can be related to the maximum in ER which corresponds to the stage III of recovery. The exothermic reaction can be related to the decrease in the ER values from 600 K and with the development of the stages IV and V of recovery, where the movement of vacancies towards the dislocations takes place, decreasing the structural free energy. Therefore, the appearance of internal stresses in stage III, due to the reorganization of defects out of thermodynamic equilibrium, leads to the locking of dislocations during the first run-up in temperature in the MS test. It gives rise to the damping background, which is amplitude dependent [1, 3], without the appearance of the damping peak (curves A and D, during the first heating run in Figs. 1 and 2). These vacancies during the first run-up in temperature in the MS test migrate to the dislocations and produce the appearance of the damping peak in the cool down run, after annealing at $T > 973$ K. This corresponds to temperatures where the ER curves start to decrease and to the exothermic reaction in DTA.

In stage III for irradiated samples the movement of vacancies out of thermodynamic equilibrium was verified by means of SANS studies, where the evolution of the arrangement of vacancies as a function of $T$ was determined. At $T \approx 550$ K the radius $R$ of the (spherical) agglomerates of vacancies [3] achieves the largest value (Fig. 4), in agreement with the ER, DTA and MS results. In fact, increasing the sample temperature for values above those for the maximum in the ER curve ($\approx 600$ K), the size of agglomerates of vacancies decreases. Moreover, we also have verified that both: (a) for $T > 850$ K, the dissolution of agglomerates take place and (b) for $T > 920$ K the concentration of vacancies out of thermodynamic equilibrium decreases markedly, as revealed by the decrease in the scattering intensity curves (Fig. 4). These facts lead to the decrease in internal stresses of the microstructure giving place, after a heating to 973 K, to the appearance of the above mentioned damping peak during the cooling.

5. Conclusions

The mobility of dislocations after plastic deformation and/or irradiation is controlled by the agglomerates of defects which reduce their mobility. The starting of the mobility of vacancies out of thermodynamic equilibrium was found at temperatures within the stage III of recovery, and at about 550 K, the vacancies agglomerates achieve the largest size. After annealing at $T > 973$ K, both the defects agglomerates dissolve and the concentration of vacancies out of thermodynamic equilibrium decreases markedly, starting the dislocations movement.

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