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Neutron irradiation embrittlement of polycrystalline and single crystalline molybdenum

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Abstract

Neutron irradiation-induced ductile-brittle transition behaviour of carburized molybdenum polycrystals and single crystals has been studied. These specimens were irradiated at 673, 873 and 1073 K to (7.9–9.8) $\times 10^{23}$ n/m^2 ($E \geqslant 1$ MeV). The results were analyzed as the difference in the behaviour between polycrystals and single crystals, and the effects of irradiation temperature. Three-point bend tests showed that the neutron irradiation-induced shift of ductile-brittle transition temperature (DBTT) at each irradiation temperature was much larger in single crystal specimens than in polycrystal ones. The carburization treatment was found to be more effective in polycrystals rather than in single crystals. This suggests that in polycrystals the grain boundary interface is strengthened due to carbon addition. At higher irradiation temperatures, this shift of DBTT decreased in both the specimens. It is characteristic in post-irradiation fracture behaviour than in single crystal specimens the cracks initiated from the island grains on the specimen surface.

1. Introduction

It is well known that molybdenum and molybdenum alloys possess favourable characteristics in that they offer, for example, high thermal conductivity and yield strength, and low sputtering yields and thermal expansion. They have been, hence, proposed as candidate structural materials for plasma facing component application. However, it has been often pointed out that neutron irradiation-induced embrittlement, i.e. the increase of ductile-brittle transition temperature (DBTT) under the conditions of practical use for these materials is of major concern from the viewpoint of fusion reactor safety [1–3]. This is one of the problems common to body-centred cubic metals and their alloys. In particular, molybdenum and its alloys exhibit a large increase of DBTT due to irradiation because of the weakness of grain boundaries, thereby drastically degrading material properties [4–9]. This is the most important issue to be solved from the practical viewpoint as well as from the fundamental viewpoint.

In the present work, post-irradiation bend properties of carburized molybdenum polycrystals and single crystals were examined through three-point bend tests in order to investigate the difference between polycrystals and single crystals, and the effects of irradiation temperature on the shift of DBTTs.

2. Experimental procedures

The chemical compositions of specimens used are listed in Table 1. Molybdenum single crystals were produced by the secondary recrystallization method. The crystallographic orientation was determined from the Laue backscattering method. The axial orientation of specimens was approximately in the $\langle 0\ 0\ 1 \rangle$ direction. These specimens were carburized at 1773 K for 1200 s [10]. Molybdenum polycrystals were made of sintered molybdenum containing a small amount of oxygen as

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Table 1
Chemical compositions of specimens (wt. ppm)

	O	N	C	Al	Ca	Cr	Cu	Fe	Mg	Mn	Ni	Pb	Si	Sn	Na	K
Polycrystals	15	<3	30	4	2	14	<3	30	0.8	<3	22	<3	20	5	<5	<5
Single crystals	10	<3	30	5	15	23	5	40	15	<3	25	3	20	7	<5	<5

the starting material. The heat treatment for carburization was made at the same condition as for single crystal specimens. The concentration of carbon after the treatment was 30 wt.ppm. The specimen size for the bend test was $1 \times 5 \times 25$ mm^3. The samples installed in helium-filled capsules were irradiated to levels of (7.9–9.8) $\times 10^{23}$ n/m^2 ($E > 1$ MeV) at about 673, 873 and 1073 K in the Japan Research Rector-2 (JRR-2). The corresponding total dose was in the range of 0.07–0.09 dpa [11].

Three-point bending tests were performed at a cross-head speed of 1.7×10^{-5} m/s at temperatures ranging from 133 to 513 K. After these tests, fractography of the specimens were done using scanning electron microscopy.

3. Results and discussion

Fig. 1 shows the relation between bend angle and test temperature on the specimens irradiated at 673, 873 and

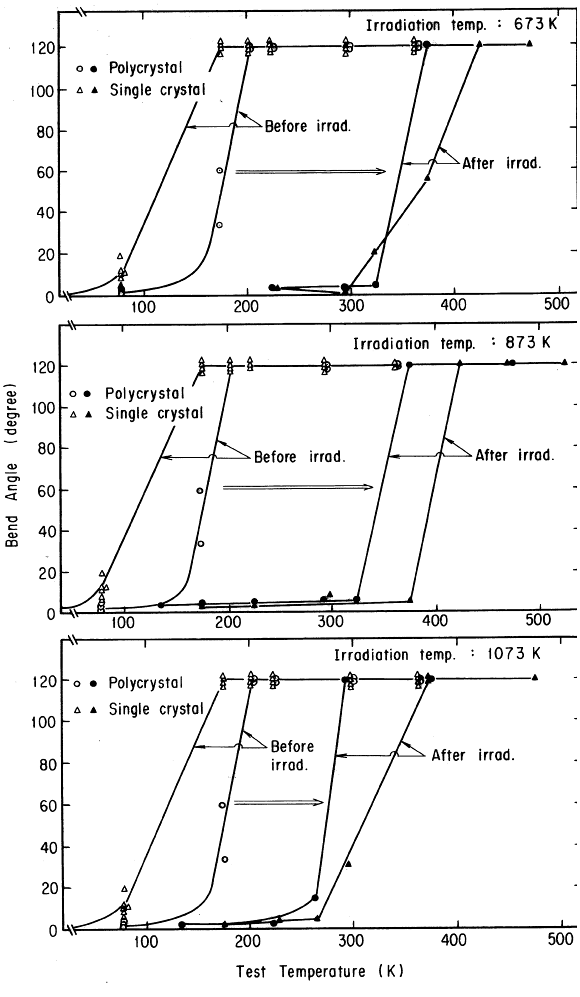

Fig. 1. Bend angle versus test temperature for polycrystal and single crystal specimens irradiated at 673, 873 and 1073 K.

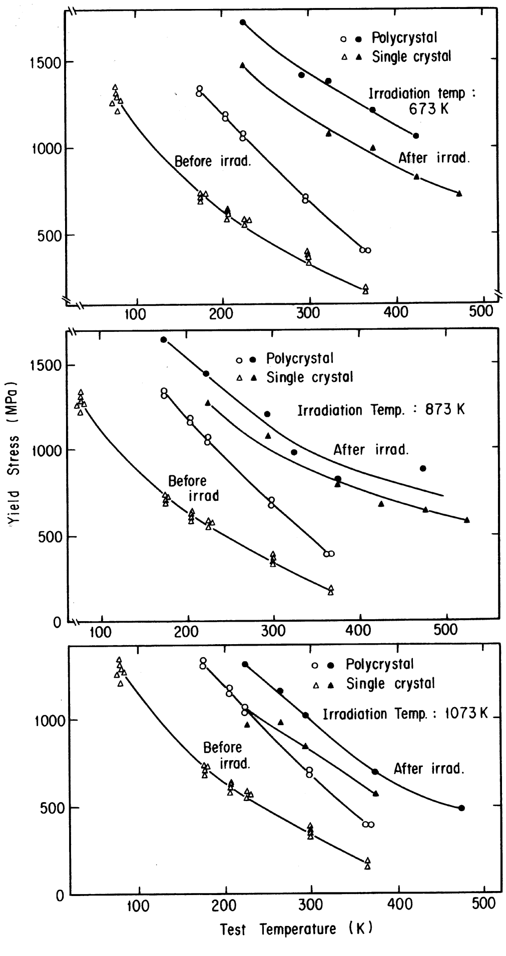

Fig. 2. Yield stress versus test temperature for polycrystal and single crystal specimens irradiated at 673, 873 and 1073 K.

1073 K. Before irradiation, molybdenum single crystals were lower at DBTT than molybdenum polycrystals. When irradiation temperature was at 673 K, DBTTs of both the specimens after irradiation were shifted to much higher temperatures. It was found that the relation between molybdenum single crystals and polycrystals before irradiation was reversed; after irradiation, DBTT in single crystals was higher than that in polycrystals. When irradiation temperature was at 873 K, DBTTs were remarkably raised by irradiation in both single crystals and polycrystals, as at the lower irradiation temperature. In addition, after irradiation, DBTT of single crystals, was higher than that of polycrystals, while the results before irradiation offered an opposite trend. When the irradiation temperature was at 1073 K, it was found that the irradiation-induced shifts of DBTTs in single crystals and polycrystals were not as large as the two cases mentioned above. The relation between yield stress and test temperature on the specimens irradiated at 673, 873 and 1073 K is presented in Fig. 2. The yield stress was determined from the load–deflection curves obtained by bend property measurements. It was calculated from the load corresponding to the uppe-yield point or 0.2% off-set on their curves, as is the case with the tensile test. From these results, yield stress before irradiation in the entire temperature range tested was, as a matter of course, much higher in polycrystals than in single crystals. After irradiation, yield stress was markedly raised in the entire temperature range tested, and was higher in polycrystals than in single crystals. Concerning the difference in the behaviour between single crystals and polycrystals, this fact was not in agreement with the results of the shift of DBTT due to irradiation. On the other hand, at higher irradiation temperatures, irradiation hardening had a tendency to decrease with increasing irradiation temperature.

The fracture surfaces of the specimens after bend-testing were examined using scanning electron microscope. In the specimens which were bend-tested at 293 K

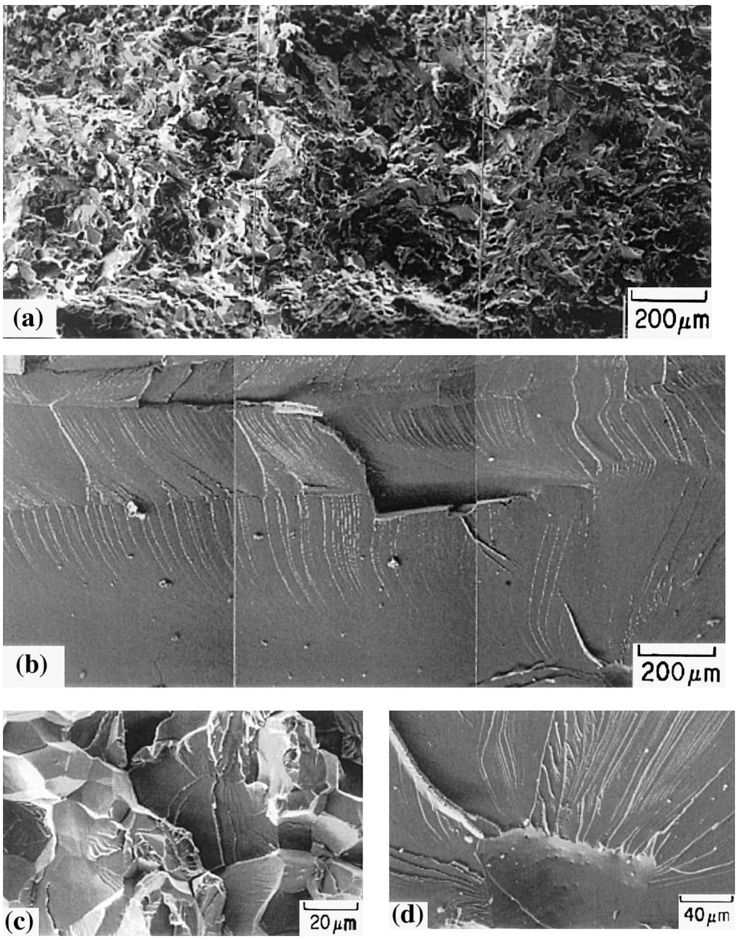

Fig. 3. SEM micrographs of polycrystal (a,c) and single crystal (b,d) specimens tested at 293 K after irradiation at 673 K.

after irradiation at 673 K, polycrystal specimens exhibited a mixed mode consisting of intergranular and cleavage fracture, as shown in Fig. 3. In contrast, the single crystal specimens showed a mode of typical cleavage fracture accompanied by many river patters. It is clear from this figure that cracks initiated from the island grains at the specimen surface. In the specimens which were bend-tested at 263 K after irradiation at 1073 K, polycrystal specimens showed almost all intergranular fracture mode while cleavage fracture also was partially observed as shown in Fig. 4. In single crystal specimens, cleavage fracture occurred and the cracks initiated from the island grains, as was the case with the specimens irradiated at 673 K.

Fig. 5 shows the plots of the shift of DBTT versus irradiation hardening for polycrystals and single crystals. Compared with the data for the same irradiation temperatures, single crystals showed larger irradiation hardening than polycrystals. Consequently, the shift of DBTT was much greater in single crystals than in polycrystals. This shift is closely related to irradiation hardening. Therefore, it is concluded from the viewpoint of reducing irradiation embrittlement that the carburization treatment was demonstrated to be much more effective for polycrystals rather than for single crystals. This also suggests that in polycrystals the grain boundary interface was strengthened due to carbon addition [10–13]. As for the effects of irradiation temperature, both molybdenum single crystals and polycrystals possessed less irradiation hardening at higher temperatures than at lower temperatures. This behaviour is consistent with the tensile property data which was measured on the specimens prepared from the same materials [14]. Therefore, the higher the irradiation temperature, the smaller is the shift of DBTTs.

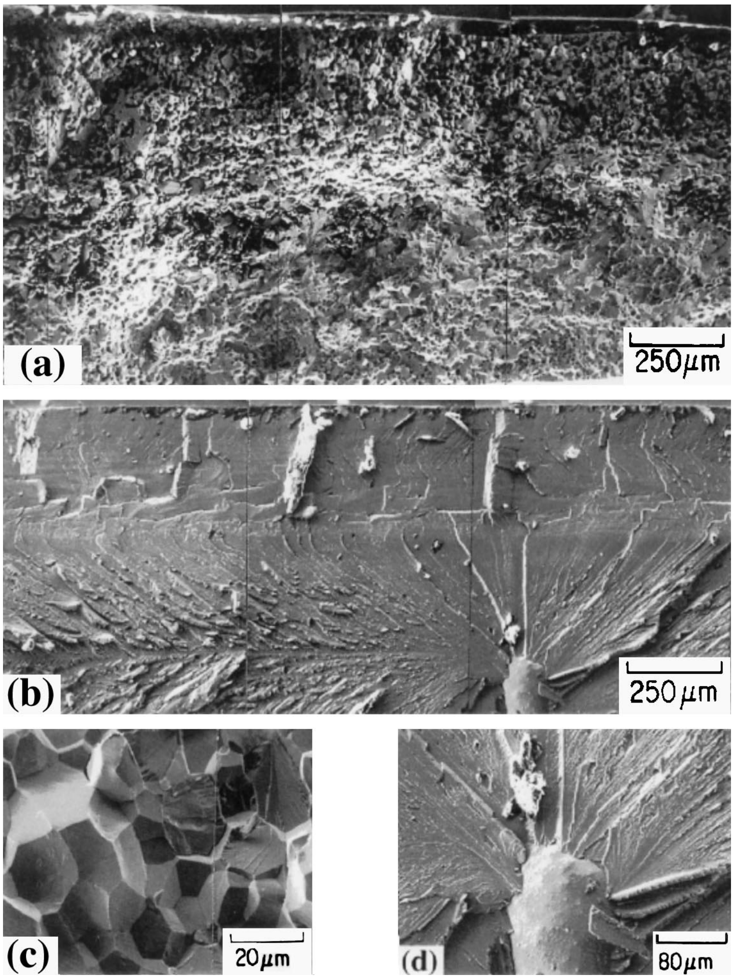

Fig. 4. SEM micrographs of polycrystal (a,c) and single crystal (b,d) specimens tested at 263 K after irradiation at 1073 K.

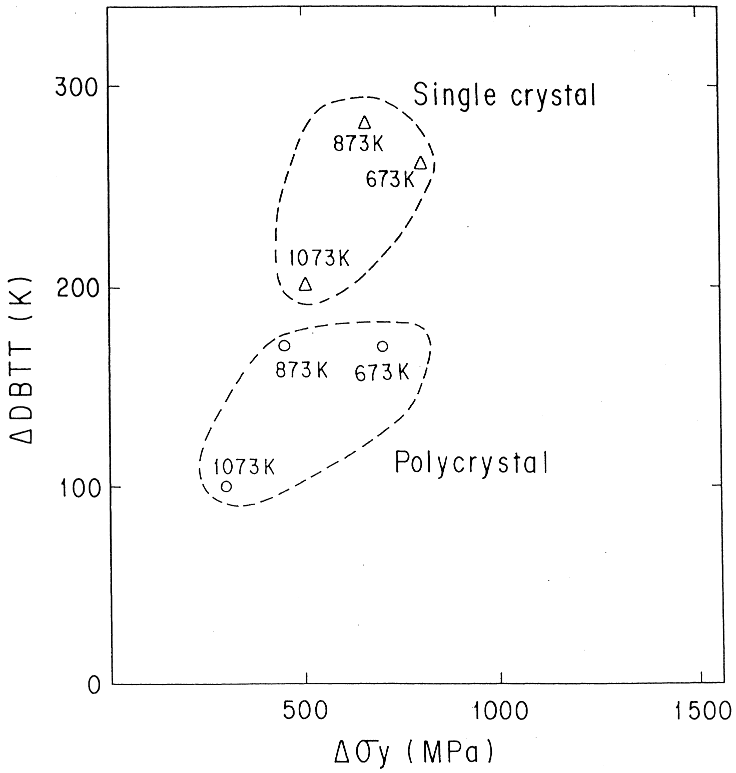

Fig. 5. Relation between the shift of DBTT and irradiation hardening in polycrystal and single crystal specimens irradiated at 673, 873 and 1073 K.

4. Conclusions

Post-irradiation bend properties of carburized molybdenum polycrystals and single crystals were examined, and the following conclusions were drawn:

1. The increase of DBTTs was much larger in single crystals than in polycrystals. This is closely related to irradiation hardening.
2. From the viewpoint of controlling irradiation embrittlement, the carburization treatment was found to be much more effective for polycrystals rather than for single crystals.
3. At higher irradiation temperatures, the shift of DBTTs decreased in both polycrystals and single crystals.

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