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ScienceDirect
Journal of Nuclear Materials 376 (2008) 11–28
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Low temperature neutron irradiation effects on microstructure and tensile properties of molybdenum

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Received 5 June 2007; accepted 12 December 2007

Abstract

Polycrystalline molybdenum was irradiated in the hydraulic tube facility at the High Flux Isotope Reactor to doses ranging from 7.2×10^{-5} to 0.28 dpa at ~80 °C. As-irradiated microstructure was characterized by room-temperature electrical resistivity measurements, transmission electron microscopy (TEM) and positron annihilation spectroscopy (PAS). Tensile tests were carried out between −50 and 100 °C over the strain rate range 1×10^{-5} to 1×10^{-2} s^{-1}. Fractography was performed by scanning electron microscopy (SEM), and the deformation microstructure was examined by TEM after tensile testing. Irradiation-induced defects became visible by TEM at ~0.001 dpa. Both their density and mean size increased with increasing dose. Submicroscopic three-dimensional cavities were detected by PAS even at ~0.0001 dpa. The cavity density increased with increasing dose, while their mean size and size distribution was relatively insensitive to neutron dose. It is suggested that the formation of visible dislocation loops was predominantly a nucleation and growth process, while in-cascade vacancy clustering may be significant in Mo. Neutron irradiation reduced the temperature and strain rate dependence of the yield stress, leading to radiation softening in Mo at lower doses. Irradiation had practically no influence on the magnitude and the temperature and strain rate dependence of the plastic instability stress.

1. Introduction

Molybdenum is of great interest for high temperature applications in advanced fission and fusion reactor systems because of its high melting point, excellent high temperature strength, good thermal conductivity, resistance to irradiation-induced swelling and corrosion resistance in liquid metal coolants [1]. However, Mo, like other body-centered cubic (bcc) metals, is susceptible to low temperatures embrittlement and suffers an increase in ductile-brittle transition temperature (DBTT) after neutron exposure [2–11]. The improvement in low temperature ductility of neutron-irradiated Mo is of great importance for its applications in advanced nuclear systems.

Low temperature embrittlement is associated with irradiation hardening that is controlled by the interactions of mobile dislocations and irradiation-induced defects. Whether or not Mo can be engineered to resist irradiation-induced low temperature embrittlement is dependent on the formation process of sessile defect clusters in Mo. It is known that two formation processes are often competing during irradiation, namely in-cascade clustering and diffusive nucleation and growth. If the formation of sessile defect clusters is dominated by a nucleation and growth process such as in bcc Fe [12–16], solute additions may have a strong effect on the formation of defect clusters and radiation hardening. The resistance to irradiation embrittlement may, therefore, be improved by metallurgical approaches. On the other hand, if large, sessile defect clusters originate from displacement cascades, e.g. in bcc W [12–14,17], the low temperature irradiation embrittlement is essentially an inherent problem, and may not be overcome.

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0022-3115/$ - see front matter © 2008 Published by Elsevier B.V.
doi:10.1016/j.jnucmat.2007.12.001

Defect formation and accumulation in irradiated materials is influenced largely by crystalline structure and atomic mass. Molecular dynamic (MD) simulations and heavy-ion irradiation experiments have clearly showed that displacement cascades are more compact and production efficiency of visible defect clusters is greater in fcc Cu than in bcc Fe [12,13,18–21]. The difference is primarily due to the more compact lattice structure of fcc metals vs. the more open lattice structure of bcc metals. The mass difference between metals is important in the defect production and accumulation as well. The displacement cascades were found to be more compact, and the energy threshold for subcascade formation is higher in a high-mass metal than in a medium-mass metals, such as bcc Mo vs. bcc Fe [20,21]. These differences in displacement cascades have significant implications in in-cascade clustering and defect cluster evolution.

While the fundamentals of defect production and evolution in medium-mass metals, such as fcc Cu, Ni and bcc Fe are well established, the understanding of defect production in high-mass metals such as fcc Pd and bcc Mo and W is still limited. Recent MD computer simulations of atomic displacement cascades in Mo [20] have provided new insights into the primary radiation damage in Mo, and have identified the differences in in-cascade clustering behavior between medium-mass bcc Fe and high-mass bcc Mo. Validation of these simulation results requires experimental evidence from well-designed mechanistic neutron irradiation experiments such as low temperature, low dose neutron irradiation. One of the primary interests of this study is to provide direct measurements of defect clusters and resultant hardening to validate in-cascade clustering results obtained from MD simulations.

The interaction between irradiation-induced defect clusters and glide dislocations is the key to understand the hardening and embrittlement in irradiated Mo. Irradiation hardening not only depends on the nature and size of defect clusters effective as barriers to dislocation motions, but also can be strongly affected by the strain rate and test temperature [22–30]. In contrast to fcc crystals, a large temperature and strain rate dependence of yielding occurs in bcc metals even in unirradiated conditions. The yield stress increases rapidly with decreasing temperature and increasing strain rate in unirradiated Mo [31]. A good understanding of the radiation hardening mechanism in bcc Mo requires detailed studies of neutron irradiation effects on the temperature and strain rate dependence of the flow stress. Temperature and strain rate-insensitive irradiation hardening implies that irradiation does not affect the rate-controlling deformation mechanism operative in the unirradiated condition, i.e. athermal hardening resulted from long-range barriers, while temperature and strain rate-sensitive irradiation hardening indicates that irradiation-induced obstacles can be overcome by dislocations with thermal assistance, i.e. thermal hardening from short-range barriers [23]. The study on the dependence of the true stress at the ultimate tensile strength, so-called plastic instability stress (PIS) on the temperature and the strain rate is lacking.

The increase in yield stress is often accompanied by a loss of strain hardening capacity and tensile ductility, and can also lead to embrittlement by shifting the DBTT to a higher temperature. Plastic deformation tends to occur in an inhomogeneous manner and to be localized in dislocation channels after irradiation. Premature plastic instability occurs at yield once the yield strength reaches the level of the PIS of an irradiated material [32–35]. Wechsler [36] suggested that the low temperature irradiation embrittlement is likely associated with changes in plastic properties, particularly with inhomogeneous plastic deformation rather than changes in inherent fracture processes. Crack formation from coarse slip steps and the intersection of coarse slip bands was suggested to cause 'channel fracture' [37,38]. Cracks may also initiate at grain boundaries due to local stress concentration caused by dislocation pile-up in a defect-free channel [5]. Direct correlation between dislocation channeling and premature plastic instability and correlation between dislocation channeling and radiation embrittlement, however, have not been confirmed by experiments.

The experiments described in this paper were designed to understand the role of in-cascade clustering in the formation of sessile defect clusters and the nature of interactions between irradiation-induced defects and moving dislocations. The ultimate purpose is to determine whether or not the resistance to low temperature embrittlement of irradiated Mo can be improved by metallurgical approaches. Neutron irradiation experiments were performed at seven low doses over a range of 7.2×10^{-5} to 0.28 dpa at reactor ambient temperature (~80 °C) on pure Mo. The formation of sessile defect clusters was investigated by characterizing as-irradiated microstructure by electrical resistivity, transmission electron microscopy (TEM), and positron annihilation spectroscopy (PAS). Irradiation hardening mechanisms in Mo were understood by examining the dose, temperature and strain rate dependence of the yield stress from tensile properties measurements between −50 and 100 °C at strain rates over the range of 1×10^{-5} to $1 \times 10^{-2}\ s^{-1}$. Deformation microstructure of irradiated specimens was examined after tensile testing. Fracture surfaces of broken tensile specimens were observed by scanning electron microscopy (SEM). The effects of neutron irradiation on strain hardening, plastic instability and fracture in Mo was described, and deformation and fracture mechanisms in neutron-irradiated Mo were discussed.

2. Experimental procedure

The material examined was low-carbon arc-cast (LCAC) Mo with purity >99.95% and interstitial impurity contents of 140 wppm C, 6 wppm O and 8 wppm N. Subsize SS-3 sheet tensile specimens with gauge dimensions of $7.62 \times 1.52 \times 0.50$ mm and rectangular coupon speci-