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Irradiation-induced creep in metallic nanolaminates characterized by *In situ* TEM pillar nanocompression

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ABSTRACT

This work reports on irradiation-induced creep (IIC) measured on nanolaminate (Cu-W and Ni-Ag) and nanocrystalline alloys (Cu-W) at room temperature using a combination of heavy ion irradiation and nanopillar compression performed concurrently *in situ* in a transmission electron microscope. Appreciable IIC is observed in multilayers with 50 nm layer thicknesses at high stress, ≈½ the yield strength, but not in multilayers with only 5 nm layer thicknesses.

1. Background

Non-conservative fluxes of irradiation-induced point defects to non-recombinative sink sites underpins many of the mechanisms for irradiation-induced degradation of materials [1–3]. Mitigation strategies focus, accordingly, on introducing a high density of recombinative sink sites that are either kinetically stable under operating conditions or exist in a non-equilibrium steady-state under irradiation fluxes [4,5]. A common approach for implementing this strategy exploits the high density of interfaces associated with nanostructured materials [6,7]. Examples include oxide dispersion strengthened (ODS) alloys, self-organized nanostructured alloys, and nanolaminates [8–11]. Nanostructured alloys have indeed been shown to suppress irradiation induced defect evolution and promote defect recombination, and in many instances they also provide exceptional mechanical properties, making them attractive for application in extreme environments. Conversely, nanostructured materials are susceptible to thermal creep, owing to grain boundary and interfacial creep processes [12–14]. Limited work, however, has focused on creep in nanostructured materials, particularly in an irradiation environment. Mechanisms considered for irradiation-induced creep (IIC) have mostly involved dislocation interactions with irradiation induced point defects [15,16]. Since the dislocation content is inherently small in nanocrystalline metals, IIC has not been considered to be a significant issue for these materials. Recent IIC investigations on nanostructured Cu-W alloys, however, demonstrate that point defect fluxes to grain boundaries can drive creep deformation [17]. By this mechanism, fluxes of vacancies and interstitials are accommodated at the grain boundaries via atomic rearrangements similar to those in shear transformation zones [18]. The applied stress biases the direction of this atomic relaxation process, and over many such events, significant deformation takes place. In Cu-W thin films measured via *in situ* ion irradiation bulge testing, this mechanism was found to be active at stresses as low as 1 MPa and temperatures as low as room temperature, as interstitial and vacancy mobility are still appreciable in Cu at this temperature [17,19]. Radiation-induced stress relaxation measurements have similarly indicated that creep mechanisms are operative in several nanograined thin-film materials [20]. For grain sizes <9 nm, Mayr et al. [20] invoked an irradiation-induced coarsening mechanism to explain irradiation-induced stress relaxation. When the diameter of the thermal spike in a collision cascade is comparable to the grain size, lattice sites are not necessarily conserved and a creep mechanism similar to that in amorphous materials is suggested to be active. This mechanism, notably, does not require point defect migration. Irradiation induced stress relaxation was also observed in Pt films with an initial grain size of 9–12 nm over the range of 25–200 °C [21]. In this case, the large epitaxial stresses were thought to produce anisotropy in the point defect diffusivities by affecting the energy barrier for diffusional jumps, thereby biasing the fluxes of vacancies and interstitials [21]. These various

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experiments indeed demonstrate that IIC is active in nanograined polycrystals at low temperatures, and that the mechanisms are likely to differ significantly from those active in coarser grained materials. The range of possible creep mechanisms, however, remains to be fully explored in different regimes of grain size, temperature, particle flux, and stress.

Measuring IIC presents substantial experimental challenges [22,23]. Experiments performed in reactors are enormously expensive, and they require excessively long experimental times to reach doses on the order of a DPA (displacement per atom). Moreover, in-reactor experiments typically offer little opportunity to systematically vary the applied stresses and temperature. These limitations can be overcome using ion irradiation, but the penetration depths of ions are quite small for mechanical testing. Light ions, e.g., protons and He, have reasonably large penetration depths (microns), but they do not simulate the damage profile of fission neutrons, and owing to their small damage cross sections, they also require long exposures to reach damage levels beyond 1 DPA [23]. Such techniques, nevertheless, have been applied to characterize irradiation creep at low doses in ODS alloys with sample size on the order of 100 μm [24,25]. Heavy ions, on the other hand, simulate the damage profile of neutrons quite well, and they provide much higher DPA rates, but heavy ions provide uniform damage profiles only over very small length scales, 10's to 100's of nanometers [26]. This requires that sample sizes must be commensurate with the depth of relatively uniform damage. Mechanically testing samples smaller than a few hundred nanometers in diameter during ion beam irradiation introduces a number of experimental difficulties associated with beam heating, mechanical alignments, and overall achievement of precision testing. We refer the readers to reference [23] for a quantitative discussion of the practical experimental issues. In that work, creep was measured in metallic and oxide glasses for which the creep rates are quite high [22,23]. The lower creep rates associated with polycrystalline materials are far more challenging. In this work, we use *operando* high-energy, heavy-ion irradiation combined with *in situ* TEM nanocompression to study IIC. The *operando* characterization ensures that the samples and loading apparatus are well aligned and that deformation modes are consistent with expectations. Such *operando* TEM observation, also, enables direct verification of the strain values measured by the instrumented nanoindentation holder. Here we demonstrate the utility of this experimental approach for acquiring IIC rates from nanostructured alloys.

Immiscible alloy nanolaminates have been intensively studied for possible application in extreme particle irradiation environments because they are thermally stable and promote defect recombination at their interfaces [11,27,28]. The creep response of such materials in radiation environments, however, is largely unknown. Specifically, it is unclear whether the accommodation mechanisms for point defects at heterophase interfaces in these materials are similar to those at grain boundaries. Nanolaminate materials are anticipated to be high strength structural alloys for next generation reactors, however their irradiation creep response is completely unknown experimentally. This work compares room temperature irradiation creep in Cu-W nanolaminates and Ni-Ag nanolaminates, as well as to dilute Cu-W alloys. We specifically consider the high stress regime relevant to extreme environments that could be encountered in future reactor environments. We select these alloys are models since, interstitials are highly mobile in all of these metals. Vacancies, though, are immobile in W and Ni at room temperature, quite sluggish in Cu, but reasonably mobile in Ag (vacancy diffusivity in Ag, ≈ 10 nm^2-s^{-1}, is about a factor of 5 higher than in Cu at 300 K) [29]. Comparing irradiation creep in the two systems and two morphologies thus enables us to assess the underlying creep mechanisms.

2. Experimental methods

Multilayer nanolaminate samples of Cu-W and Ni-Ag were grown on pre-tilted Si pillars following a method described in detail elsewhere [30]. Pre-tilt angles near 45° were used such that if defect fluxes to the interface were to drive creep, the maximum resolved shear stress would be applied at these interfaces. Cu(99.99% Lesker) - W(99.95% Lesker) multilayers with individual layer thicknesses, h, of nominally 50 nm and 5 nm were grown via magnetron sputtering in Ar in a system with a base pressure of $\approx 10^{-8}$ torr. Ag (99.99% Lesker) - Ni (99.99% Lesker) multilayers with $h = 50$ nm and $Cu_{99}W_1$ alloy films were also grown in the by magnetron sputtering. Pillars for nanocompression were fashioned into their final geometry using annular milling in a focused ion beam (FIB, FEI DB238). The 250–550 nm pillar diameters far exceed the grain size (around 50 nm) of the Cu and W and therefore surfaces should not be a significant defect sink. The literature suggests that the bulk yield strengths of Cu-W and Ni-Ag multilayers of comparable thickness are ≈5.5 GPa and ≈1.17 GPa, respectively [31,32]. Their interfacial shear strengths are reported to be ≈ 0.55 GPa and 0.53 GPa, respectively [33]. Nano-compression tests, imaged *in situ,* were performed using a Hysitron PI-95 Picoindentor in a 200 kV JEOL 2100 I^{3}TEM [34]. Values of strain reported by the instrumentation were verified through imaging. Results obtained under a constant loading rate were much more reliable than those under constant load, as the latter was more prone to sample drift. The load control experiments were performed with a loading rate of 25 MPa s^{-1} up to ~750 MPa, after which the loading rate was varied between 0.63 and 33 MPa s^{-1}. The attached 6 MV EN Tandem Van de Graaff-Pelletron accelerator was used as the source of 3 MeV Cu^{3+}; it delivered between 30 and 170 nA within a spot size of approximately 4 mm in diameter, resulting in average damage rates in Cu between 2×10^{-3} to 1×10^{-2} DPA s^{-1}. Ni-Ag samples were irradiated at 30 nA producing a damage rate in Ag of 2×10^{-3} DPA s^{-1}.

3. Results

We first performed stress-strain measurements on 50 nm Cu-W multilayers in the absence of irradiation in load controlled and displacement controlled modes and after irradiation to 100 DPA in load controlled mode. Fig. 1a shows the response of these samples. Interfacial shearing introduces a discontinuity in the curve, indicated by the red arrow, and is the primary source of plastic deformation. Pre-irradiation clearly enhances the interfacial shear strength of Cu-W, similar to previous reports for Cu-Nb [30]. The increased interfacial shear strength of Cu-Nb saturated at <1 dpa, which explains why the lower shear strength is not observed in the pristine sample tested during in-situ irradiation. No observable bulk plasticity occurs in any of the Cu-W samples loaded to 1.5 GPA, which is consistent with the anticipated high bulk yield strength of such nanolaminate alloys and the expectation of negligible thermal creep at room temperature. These load control tests were performed at 0.63 MPa s^{-1} and the noise in the curve is associated primarily with drift in the instrumentation over the time scale of the experiment (10^3 s). The *in situ* imaging is thus particularly valuable for distinguishing between noise associated with long loading times and actual strain in the sample and observing critical events such as interfacial sliding. Low rates of loading (0.63 MPa s^{-1}) were used in this experiment in order to be commensurate with the lowest loading rates utilized for the IIC experiments to follow. The displacement controlled experiment was performed at 1 nm s^{-1}, and it yields similar results. Fig. 1b depicts the stress-strain curves of 50 nm Cu-W multilayers, first irradiated *in situ* at approximately 1.4×10^{-2} DPA s^{-1}, and then