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Departing from the mutual exclusiveness of strength and ductility in nanocrystalline metals with vacancy induced plasticity

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ABSTRACT

Strength and ductility are mutually exclusive in metallic materials. To break this relationship, we start with nanocrystalline Zirconium with very high strength and low ductility. We then ion irradiate the specimens to introduce vacancies, which promote diffusional plasticity without reducing strength. Mechanical tests inside the Transmission Electron Microscope reveal about 300% increase in plastic strain after self ion-irradiation. Molecular dynamics simulation showed that 4.3% increase in vacancies near the grain boundaries can result in about 60% increase in plastic strain. Both experimental and computational results support our hypothesis that vacancies may enhance plasticity through higher atomic diffusivity at the grain boundaries.

Strength of metals typically increases with decreasing grain size due to reduced number and mobility of dislocations [1]. Unfortunately, this also usually means lower ductility and toughness. The mutual exclusiveness of strength and toughness is well-documented in the literature [2,3]. The progress in nanocrystalline materials with very high strength has inspired researchers to further engineer the microstructure for high ductility without losing strength [4,5]. Earlier studies explored bi- (or multi-) modal grain distribution, where the smaller and larger grain sizes contribute to the strength and ductility respectively [6]. However, the exact distributions of the grain size, grain shape, and spatial locations depend on many processing parameters and may be difficult to control. Exploiting deformation twins through cryo-milling is another effective approach [7]. An innovative variation of this approach involved gradient hierarchical nano-twinned structure [8,9]. Dispersions of nanoparticles and nano-precipitates have been studied on engineering alloys [10,11], where the hard precipitates initiate, drag, and pin dislocations to reduce dynamic recovery. The result is a significant dislocation storage required for compatible plastic strains, allowing a high strain-hardening rate with larger uniform strains while elevating strength. High strength and toughness can also be achieved through using transformation-induced plasticity [12,13] and twinning-induced plasticity. They are operative in ultrafine grains albeit at relatively high flow stresses. A more recent approach exploited decreased phase stability in high-entropy alloys [14], which exploits meta-stability of phases to break the classical strength-ductility relationship. The state of the art in the literature is conclusion that microstructural heterogeneity is a unifying design philosophy tying seemingly different approaches toward high strength and ductility [3,15].

In this study, we propose a new concept that exploits point defects to enhance plasticity, while the strength is dictated by the nanocrystalline grain size. The fundamental concept is to start from a high strength, low ductility condition and then inject defects in form of vacancies to promote diffusional plasticity. Thus, our study starts with a nanocrystalline form of a pure metal with grain size 10–15 nm. The literature suggests that this grain size is very close to the critical or crossover size (d_c) between the diffusion and dislocation dominated plasticity [16]. In other words, this grain size corresponds to the highest strength and lowest ductility. We then increase the plastic deformation without decreasing the strength by irradiating the specimens. The excess vacancies due to irradiation are expected to induce high grain boundary (GB) mobility [17–19], and creation of dislocations [20,21]. This corresponds to the final state, where small grain size impedes dislocations for high strength, yet the excess vacancies promote the diffusional plasticity at the same time. Vacancies increase the grain boundary energy but reduce the sliding energy [22]. They can be thought of as virtual lubricants for grain boundary mediated deformation mechanisms such as sliding, rotation and growth [23,24]. The literature suggests enhanced grain boundary mobility can *increase toughness* by mitigating the localized

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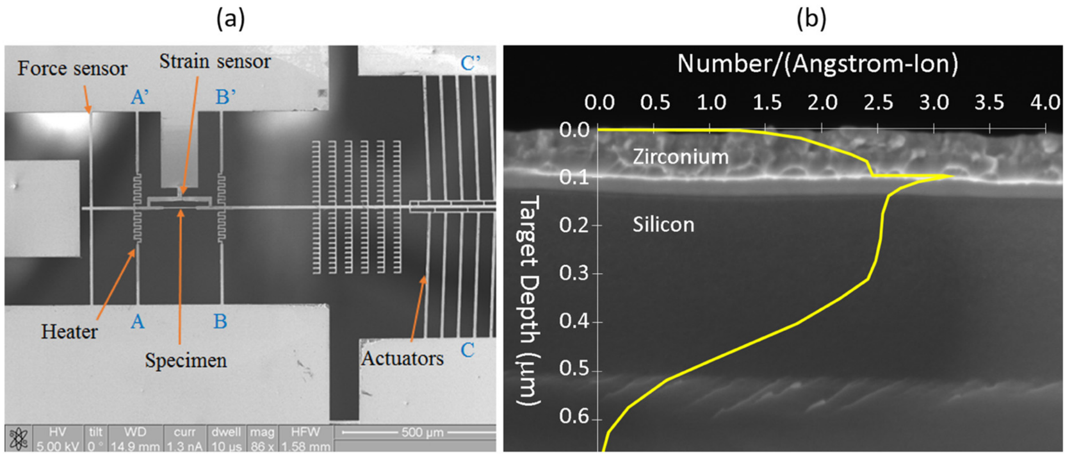

Fig. 1. (a) MEMS-based in-situ TEM tensile testing setup (b) SRIM simulation results on the ion irradiation damage profile.

stresses developed at the grain boundaries [25], thereby preventing crack nucleation and growth. The implication is that enhanced diffusional plasticity can be achieved in the region known to be dislocation dominated. Thus, we hypothesize that the extrinsic vacancies in a nanocrystalline metal can induce co-existence of dislocation dominated strength and diffusion dominated plasticity. This is the proposed fundamental mechanism behind the departure from the mutual exclusiveness of strength and ductility.

A formidable challenge in the proposed approach is the generation of extrinsic vacancies in the specimens. Mechanical stress or high temperature create vacancies, which are not stable. We suggest that this can be achieved through (a) controlled irradiation and (b) passing electrical current through the material. During irradiation, knock-on of the energetic particles and subsequent cascade collisions cause displacement of atoms from their equilibrium positions. This generates point defects (vacancies and interstitial atoms). Defects of opposite nature recombine, while the same types clusters and eventually agglomerate to loops due to their surface energy. Current flow also generates vacancies by momentum transfer of high-energy electrons at the grain boundaries. In this study, we use self-ion irradiation because it is difficult to control the vacancy generated by current flow.

We developed an experimental procedure to perform uniaxial tensile tests on self-ion irradiated specimens in-situ inside a Transmission Electron Microscope (TEM). The setup involves a micro-electro-mechanical (MEMS) device with actuators and displacement sensors integrated with the 99.99% pure and nominally 100 nm thick zirconium specimen. Fig. 1a shows the experimental setup. Device fabrication and tensile testing details are given elsewhere [26]. Before making the specimen freestanding, the MEMS devices were ion irradiated with an 800 keV Zr^+ beam using the 6 MV HVE Tandem accelerator at Sandia's Ion Beam Lab. The doses achieved were varied from 3×10^{10} ions/cm^2 to 3.26×10^{14} ions/cm^2 (from 1.26^{-04} to 1.37 displacement per atom). Fig. 1b shows the Stopping and Range of Ions in Matter (SRIM) simulations on damage profile [27]. The specimens were then made freestanding for tensile testing.

Since it is extremely difficult to visualize or quantify the individual or cluster of vacancies, we studied the fundamental mechanisms using molecular dynamics (MD) simulation. Even though the time and length-scales cannot be directly compared, MD simulation is able to capture fundamental mechanics involving vacancies, dislocations and grain boundaries. We used the Embedded Atom Method (EAM) potential [28], which can accurately predict the mechanical properties of zirconium thin film [29]. The nanocrystalline zirconium model with average grain size of 10 nm was built with Voronoi tessellation. The dimension of the simulation cell is 20 nm × 20 nm × 5 nm and periodic boundary conditions were applied along all directions. In our present study, we orient grains at different angles where 0° angle lies along [0001] direction corresponds to film normal (Z-axis). The model is checked for any overlapping of atoms at the grain boundaries. To build the vacancy enriched zirconium thin films we introduced vacancy randomly within ~3 nm of grain boundaries. Energy minimization is carried out using conjugate-gradient (CG) method followed by NPT dynamics for several thousand steps in LAMMPS. During our simulation, we maintained periodic boundary conditions in all directions. Verlet algorithm is employed for time integration considering a time step of 0.5 fs during the NPT dynamics. Initially we equilibrated the simulation cell at 300 K under NPT dynamics using Noose-Hoover thermostat for 50 ps. Then we heat up the system to 800 K at a ramp rate of 0.01 K/fs. We hold the sample at 800 K for 100 ps then we slowly cool down the sample to 300 K at a cooling rate of 0.01 K/fs to relax the grains. Later, we equilibrated the sample at 300 K for 50 ps to obtain internal stress-free grains. In our present study, all the tensile testing was performed at 300 K at a strain rate of $5 \times 10^8 s^{-1}$ under NPT dynamics.

Uniaxial tensile testing was performed on each of the self-ion irradiated specimens. We observed gradual increase in the fracture strain as function of the dose or displacement per atom. Fig. 2 shows the test results for two extreme cases, i.e. specimens with no and the highest radiation dose. It shows TEM bright field (BF) images, selected area electron diffraction (SAED) and stress-strain response of pre- and post-irradiated samples respectively. The corresponding electron diffraction patterns are shown in supplementary Fig. S1. We notice that irradiated sample can sustain higher tensile strain compared to as-deposited sample. This phenomenon can be attributed to our hypothesis that extrinsic vacancies can induce higher atomic mobility through its interaction with the grain boundaries. Another source of plasticity could be the dislocations created in the originally dislocation-starved grain interior during irradiation. The literature contains evidence of similar irradiation enhanced plasticity in bulk metallic glasses [30]. Fig. 2 also shows noticeable grain coarsening due to irradiation. This may contribute to the enhancement of ductility by accommodating more dislocations. However, the coarsened grain size in the irradiated specimen is still small enough to store statistically significant dislocation density. Therefore, the ductility enhancement contribution from grain coarsening could be insignificant compared to the vacancy effects in an irradiated sample. This is further supported by the strong role played by the vacancies in our computational model.

To understand the mechanics behind the increased ductility, we simulate the tensile response of the pristine and irradiated zirconium samples. This is shown in Fig. 3. We notice that irradiated sample can sustain higher tensile strain prior to failure compared to pristine zirconium thin film. Plastic yielding starts at 7.4% (marked by green vertical dotted line in Fig. 3a) for pristine sample whereas irradiated sample can