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A comparative study on the *in situ* helium irradiation behavior of tungsten: Coarse grain vs. nanocrystalline grain

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ABSTRACT

Increasing the density of sinks such as grain boundaries and interfaces for irradiation-induced defects and implanted ions has been demonstrated to be an effective way to improve the irradiation resistance of materials. To understand the effects of grain boundaries on the degradation mechanism of nanostructured materials, nanocrystalline tungsten was fabricated by high pressure torsion (HPT-W). Morphological changes of HPT-W and coarse grain tungsten (CG-W) during helium ion irradiation were evaluated *in situ* in a helium ion microscopy. It has been shown that the degradation mechanisms of CG-W and HPT-W are remarkably different. Blister occurs on the surface of CG-W when the irradiation dose increases up to 5.0×10^{21} m^{-2}, and orientation dependence of blistering has been observed. However, no blister is formed on the surface of HPT-W even when the irradiation dose increases up to 1.0×10^{23} m^{-2}. Instead, crack formation along grain boundaries is the major degradation mechanism during helium irradiation of HPT-W, supporting a different irradiation degradation mechanism. This explains the unprecedented irradiation tolerance of HPT-W in terms of blistering. Molecular dynamics results also show that grain boundaries and helium clusters play an important role during the propagation of a crack. The zigzag crack planes are attributed to the coalescence and growth of helium blister/bubble-induced crack. The results document that grain boundaries play decisive roles in the irradiation resistance of nanostructured materials, and provide a new perspective to the design of plasma facing materials with excellent irradiation resistance. It is thus suggested that excellent irradiation resistance can be achieved by a meticulous design of grain boundaries based on "interface engineering".

1. Introduction

New generations of fission and fusion reactors require materials with excellent radiation resistance. The evolution of radiation induced degradation in materials is generally understood in terms of three basic regimes. The first regime is characterized by the production of atomic displacement defects (i.e. vacancy and interstitial). The displacement defects are directly produced by energetic displacement cascades when the incident particle energy (E_I) is higher than the displacement energy (E_D) of the host atom (e.g. 0.5 keV for tungsten) [1] or by "trap mutation" [2–4] when $E_I < E_D$, where several implanted atoms agglomerate and force a host atom off its site. In the second regime, when the defects are produced, defect diffusion, accumulation and recombination set in. In this regime, the created defects which survive recombination and annihilation will cluster with the implanted atoms, giving rise to the production of clusters, bubbles, voids, *etc.* With further radiation, the third regime, *i.e.*, macroscopically observable damages, such as cracking, surface blistering, formation of nanostructures

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(e.g. "fuzz"), *etc*. take place, leading to the irreversible degradation in materials.

Over the past 50 years, different strategies for alleviating radiation damage have been developed to achieve better radiation tolerance [5]. The approaches to enhancing radiation tolerance in materials are closely related to the above-mentioned three regimes. For the first regime, the production of point defects in materials under irradiation is intrinsic, and therefore, researchers are mainly focusing on exploring whether certain atomic mass and lattice structure might exhibit inherently superior resistance to the irradiation-induced defect production. For example, body centered-cubic (BCC) metals generally exhibit better radiation resistance than face centered-cubic (FCC) metals. Therefore, tungsten (W), a BCC refractory metal, has been regarded as the most promising candidate for plasma facing materials (PFMs) in future fusion reactors due to its outstanding physical and chemical performances [6,7]. Additionally, high-entropy alloys [8,9] composed of four or more metallic elements mixed in an equimolar or near-equimolar ratio and bulk metallic glasses [10–12] which lack crystalline structure have recently drawn great interest for high temperature fission and fusion applications.

For the second regime, numerous experimental and theoretical attempts have been made to enhance the radiation resistance. Defect migration and annihilation affect the evolution of the irradiation-induced damage in this regime. Therefore, those attempts could be classified into two groups: (i) introduction of defect diffusion barriers to decrease defect accumulation and to increase defect recombination sites; (ii) introduction of defect sinks to promote defect recombination and annihilation. In the first case, if vacancies or interstitials are immobile, not only the diffusion-dominated defect accumulation process that results in damage cascade will be alleviated, but also the number of defect recombination centers will be increased. Another popular approach is via alloying. For example, first-principles calculations show that for the case of W-based alloys, the transition metal solutes with larger electronegativity will be more favorably bonded to vacancies and smaller solutes prefer to be bonded to self-interstitials in W [13]. As such it will affect the defect diffusion behavior. In the second case, the introduction of a large number of surfaces [14], interfaces [15–18] and grain boundaries (GBs) [19–21] that serve as effective sinks for radiation-induced defects and implanted ions (e.g. helium and hydrogen isotopes) has been proven effective in improving radiation resistance. For instance, experiments and molecular dynamics (MD) simulations on nanoscale gold foams show the existence of a window in the parameter space where foams are remarkably radiation-tolerant due to the high density of free surfaces [14]. Oxide-dispersion strengthened steels [22,23] have proven to have excellent radiation resistance due to the high density of interfaces between the matrix and nanoscale oxide precipitates. Recently, two alternative approaches: magnetron sputtering [16] and accumulative roll-bonding (ARB) [24] have also been used to prepare multilayer materials in order to introduce large amounts of interfaces. Results show that the layer interfaces are efficient sinks for absorbing irradiation-induced defects, giving rise to excellent radiation resistance.

Experimentally, the prevalent method for increasing GB density is via grain size refinement. For instance, nanocrystalline W (NC-W, grain size $d < 100$ nm) [25–27] and ultrafine grained W (UFG-W, $100\text{ nm} < d < 1000$ nm) [27,28] have been achieved by severe plastic deformation (SPD) methods including high pressure torsion (HPT) [25], equal-channel angular pressing (ECAP) [29] and ARB [30]. The versatility and practicability of those methods open innovative pathways to tailoring material performances, and are applicable for investigating the underlying mechanisms during plastic deformation. Results have shown that both mechanical properties (e.g. high strength, high ductility) and improved radiation resistance of the aforementioned materials can be achieved because of their high density of GBs, which are sinks for the irradiation-induced defects. Those defect sinks also facilitate recombination of the implanted ions and irradiation-induced defects [20,26,31]. Additionally, the efficiency of GBs as sinks depends on the GB character, including both misorientation and GB plane orientation [32]. Theoretically it has been shown that the efficient annealing of radiation damage near GBs is via defects-GB interaction [33,34]. The self-healing capability of the nanostructured material is closely related to the coupling of the individual segregation and annihilation processes of vacancies and interstitials near the GBs. Upon irradiation, interstitials are loaded into the GBs which then act as a source, emitting interstitials to annihilate vacancies in the bulk.

The irradiation performances of various materials with lager numbers of defect sinks have been widely studied as summarized in the above discussion. However, previous studies are mainly focused on the first two regimes of the radiation-induced degradation. The degradation mechanism in the third regime of nanostructured materials and how it differs from that of the coarse-grained counterparts are still poorly understood. It is particularly true that a direct comparative study is still lacking. In other words, introducing high density of defect sinks could delay the onset of macroscopically observable damages by providing numerous sites for defect annihilation. But how does the subsequent microstructural evolution proceed as the defects approach a saturation concentration under severe ion irradiation? For instance, how does the large number of GBs affect the blistering behavior on the surface of nanostructured materials under severe ion irradiation? Additionally, towards the application, the macroscopically observable damage of materials during the third stage is directly related to their performance and service life. For instance, in the operation of tokamaks, blister bursting leads to dust production, resulting in various hazards such as (i) plasma contamination; (ii) explosion in the case of air ingress (accident by loss of vacuum); (iii) dusts containing tritium or activated materials may be highly radioactive [35]. Thus, a fundamental question arises: does it work for decreasing blistering and dust generation via GB design based on "interface engineering"?

Herein, considering the importance of tungsten in the future nuclear reactors, it is selected as the model system in the present work. To the best of the knowledge of the authors, this is the first comparative study on the *in situ* helium irradiation behavior of W: CG-W versus HPT-W in a helium ion microscope (HIM). Taking advantage of the accurate and controllable micro-area He implantation technique in a helium ion microscope, different helium doses can be implanted into the location of interest [36,37]. This study reveals that the degradation mechanisms of HPT-W and CG-W exposed to helium irradiation are very different. Blistering has been observed on the surface of CG-W as the irradiation dose is increased to $5.0 \times 10^{21}\ \text{m}^{-2}$, while crack formation along GBs is the main degradation mechanism of HPT-W even when the irradiation dose is increased up to $1.0 \times 10^{23}\ \text{m}^{-2}$. In this context, the effect of helium and helium clusters on the crack propagation in W bicrystals with different GB characters has also been investigated by molecular dynamics (MD). The MD results show that the crack propagation behavior and the resulting crack morphology are, to a large extent, controlled by the presence of high-angle GBs and helium clusters. Finally, by comparing the experimental and computational results, it is concluded that a meticulous design of GBs can offer a great potential in improving the performance of materials under harsh ion irradiation.