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Irradiation Behavior in High Entropy Alloys

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Abstract: As an increasing demand of advanced nuclear fission reactors and fusion facilities, the key requirements for the materials used in advanced nuclear systems should encompass superior high temperature property, good behavior in corrosive environment, and high irradiation resistance, etc. Recently, it was found that some selected high entropy alloys (HEAs) possess excellent mechanical properties at high temperature, high corrosion resistance, and no grain coarsening and self-healing ability under irradiation, especially, the exceptional structural stability and lower irradiation-induced volume swelling, compared with other conventional materials. Thus, HEAs have been considered as the potential nuclear materials used for future fission or fusion reactors, which are designed to operate at higher temperatures and higher radiation doses up to several hundreds of displacement per atom (dpa). An insight into the irradiation behavior of HEAs was given, including fundamental researches to investigate the irradiation-induced phase crystal structure change and volume swelling in HEAs. In summary, a brief overview of the irradiation behavior in HEAs was made and the irradiation-induced structural change in HEAs may be relatively insensitive because of their special structures.

Key words: high entropy alloy; irradiation behavior; self-healing; structure change; volume swelling

The rapidly growing energy demand and more considerations in environmental issues impel nuclear energy to play an important role among other energies. Currently, about 430 commercial nuclear power reactors in the world provide about 11% of the world's supply of electricity. A vast majority of structural materials are playing important roles in fission reactors. To improve the safety and efficiency of nuclear reactors, the development of novel and advanced nuclear structural materials with high resistance to irradiation damage is necessary. In many cases, a key strategy for designing high resistance to irradiation damage materials is based on high temperature phase stability, high temperature strength, and dimensional stability under irradiation conditions, because irradiation can result in the increase of defect densities, which impedes dislocation motion and increases flow stress. During plastic loading, a vast majority of dislocation glide can cause the absorption of defects, leading to strain softening and yield drop. In addition, the interaction between solute atoms and point defects can cause coupled transport of solute atoms by point defect fluxes[1], giving rise to solute segregation phenomena and leading to void swelling and structural changes. The structural materials for advanced nuclear systems need to endure much higher neutron doses, higher temperatures and extremely corrosive environment, which are beyond the performances of materials used in current nuclear systems. In general, conventional materials for nuclear reactors include various ferritic/martensitic steels[2], austenitic stainless steels[3], zirconium alloys[4-6], ceramics, and composites, etc. They might have a limit to endure the high irradiation dose and the severe environment in future nuclear system.

In order to endure the irradiation threats to the operation of structural materials used in nuclear systems, novel structures, which possess high irradiation resistance, should be developed. Recently, high entropy alloys (HEAs)[7-12], also termed multi-base alloys (MBAs), are being developed. As equi-atomic or approximately atomic, multi-element metallic systems, the MBAs generally have at least 3 major metallic elements with high concentrations (5 at.% to 35 at.%), and the multiple elements with different crystal structures can crystallize as a relatively simple phase, because the configurational

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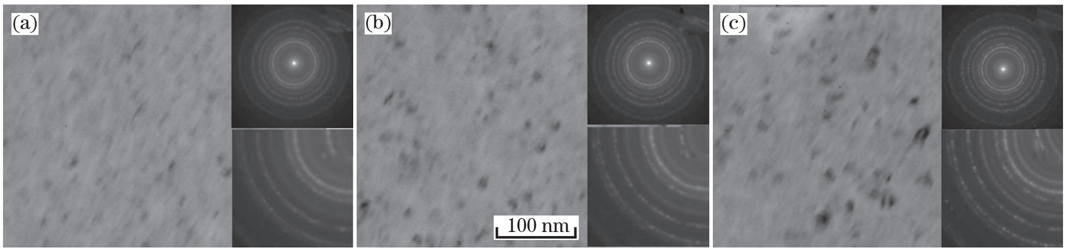
entropy has the contribution to the total free energy in alloys. High entropy may stabilize the disordered solid-solution state rather than multi-phase microstructures, which distinguishes HEAs from conventional alloys, and the mixing of various elements leads to the possibility of high irradiation resistances via the unique "self-healing" mechanisms[13]. In contrast to conventional alloy systems, HEAs tend to form a simple solid solution structure, such as face-centered cubic (FCC), body-centered cubic (BCC), or a mixture of both, which is due to the high entropy of mixing of the solution phases[14]. Recent results in HEAs are presented to demonstrate how atomic-scale characteristics can provide a high irradiation resistance. In this article, the irradiation behavior of HEAs with high phase stability under fast electron or accelerated MeV heavy ion irradiations was reviewed. Different structures of HEAs were described in more detail with an emphasis on the irradiation behavior of recent correlative researches. Besides, the "self-healing" process in HEAs was proposed.

1 Phase Stability of HEAs against Irradiation

The structural changes in an FCC CoCrCuFeNi high entropy alloy obtained by sputtering (thickness: 25 nm, 100 nm and 1 μm) under MeV electron irradiation were investigated with a high voltage electron microscope (HVEM)[15]. The results confirmed that the FCC solid solution exhibited high phase stability against irradiation, and the FCC phase remained as the main constituent phase over 40 dpa (displacement per atom) of irradiation at 298 K and 773 K, respectively. Moreover, although the irradiation produced in this alloy was similar to the structural changes induced by thermal annealing, the irradiation-induced grain coarsening did not occur at 773 K as well as at 298 K. Fig. 1 shows the in-situ transmission electron microscopy (TEM) images of structural changes in a 100 nm thick specimen of CoCrCuFeNi HEA irradiated by MeV electron at 773 K[15]. The grain coarsening caused by irradiation could not be seen in bright field (BF) images (Figs. 1(a)−1(c)) at 773 K, and the corresponding selected-area diffraction (SAD) patterns indicated the irradiation-induced structural changes. The number and the position of Debye rings in SAD patterns tended to increase with increasing the irradiation dose. To understand the irradiation-induced changes observed in SAD patterns, the electron diffraction intensity profile was analyzed and the results are shown in Fig. 2[15]. Before irradiation, the main constituent phase was identified as the FCC phase accompanied with a minor phase of BCC solid solution; the BCC phase increased with increasing irradiation time, but the FCC phase remained as the main constituent phase. Similarly, another different structure HEA, BCC solid solution Zr-Hf-Nb[16-18], did not exhibit any obvious structural change after about 10 dpa irradiation at 298 K, revealing a high phase stability against irradiation damage.



(a) Before irradiation; (b) After irradiation for 180 s, 6.8 dpa; (c) After irradiation for 1.2×10^3 s, 45.6 dpa.

Fig. 1 In-situ TEM images showing the changes in the structure of a 100 nm thick specimen of CoCrCuFeNi HEAs irradiated by MeV electron at 773 K

The present authors prepared a five-element high entropy alloy, Al_xCoCrFeNi (x = 0.1, 0.75, and 1.5 in molar ratio, for simplicity, they are denoted by Al0.1, Al0.75 and Al1.5, respectively) by vacuum levitation melting (VLM) with varying Al contents, which can form different structures[19]. Then, the $Al_{1.5}$CoCrFeNi HEA with BCC structure was further studied by TEM. The TEM micrographs of the $Al_{1.5}$CoCrFeNi alloy sample before and after Au-ion irradiation were compared and are shown in Fig. 3[20]. The insets in the TEM micrographs, as shown in Figs. 3(a) and 3(b), are the corresponding SAD patterns. It showed that after irradiation, the sample structure did not change. The TEM image also indicated that there was no obvious mixing between the matrix and particle under irradiation. However, numerous black spots could be observed in Fig. 3(b), which may be caused by the irradiation-induced element segregation. Moreover, the authors' research also verified that the value of void swelling in $Al_{1.5}$CoCrFeNi alloy was quite lower than those of other typical nuclear alloys under similar irradiation conditions[20]. In contrast, in some amorphous alloys or bulk metallic glasses (BMGs), such as Zr-based binary BMGs[21], electron irradiation could lead to the formation of nano-crystalline. It indicated that the amorphous phase in Zr-based binary BMGs was not stable under electron irradiation, but the crystalline phase which required to resist electron irradiation might be stable. In addition, in nano-structured Cu-based alloys, such as $Cu_{93.5}W_{6.5}$ and