Resistance to He²⁺ irradiation damage in metallic glass Ta₃₈Ni₆₂

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Abstract

Metallic glass Ta₃₈Ni₆₂ strips, metallic W, and V_{87.5}Cr_{4.17}Ti_{4.17}Nb_{4.17} alloy were irradiated using a 500 keV He²⁺ ion beam at different fluence to compare the metallic glass resistance to irradiation. Metallic glass Ta₃₈Ni₆₂ remained amorphous at different He²⁺ irradiation fluence. Transmission electron microscopy analysis revealed the presence of helium bubbles at the end of the range of helium ions in the metallic glass. No significant damage resulted in the metallic glass surface, and the root mean square roughness increased nonlinearly with the increase in fluence. At 1×10¹⁸ ions/cm², metallic W appeared in larger sunken areas on the surface and V_{87.5}Cr_{4.17}Ti_{4.17}Nb_{4.17} alloy experienced multi-layer flaking. The metallic glass Ta₃₈Ni₆₂ resistance to He²⁺ ion beam irradiation was better than that of metallic W, and that of the V_{87.5}Cr_{4.17}Ti_{4.17}Nb_{4.17} alloy was the poorest.

Keyword: He²⁺ irradiation; metallic glass Ta₃₈Ni₆₂; helium bubbles

1. INTRODUCTION

Metallic glass is a non-equilibrium amorphous phase generated by rapid alloy solidification,¹ which is characterized by its arrangement of atoms in the short-range ordered and long-range disordered amorphous structure.² Metallic glass has been applied widely as a new type of structural and functional material with high strength, low magnetic loss, and good corrosion resistance.^{3,4,5} Its extent of application is expanding into the electronics, industrial, machinery, and other fields, and it has attracted much attention and research interest.^{6,7,8} Because metallic glass has material characteristics without lattice defects,⁹ the study of its resistance to irradiation damage is of great interest.¹⁰⁻¹³ Metallic glass has a good irradiation resistance below its crystallization temperature.¹⁴ Ni-based metallic glass has a higher crystallization

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transition temperature, excellent adsorption, and enhanced temperature stability with elemental Ta addition, thus it may have good irradiation resistance.^{15, 16}

Ion beam irradiation is an effective experimental method to study material irradiation resistance.¹⁷ Helium ion beam irradiation exists in the universe and in other irradiation environments.¹⁸ Helium concentrates in irradiated crystalline materials, forms helium bubbles near the peak helium concentration area, and causes the appearance of material swelling, blistering, peeling, embrittling, and cracking from irradiation damage, which results in material aging and mechanical performance degradation.^{19, 20, 21} A study of the irradiation resistance of Ni-based metallic glass with helium ion beam irradiation would therefore be useful. In this paper, a 500-keV He^{2+} ion beam was used to irradiate metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ and study its resistance to He^{2+} ion beam-induced irradiation damage. Polycrystalline W, currently the most promising plasma-facing material for fusion reactors,²² and $\text{V}_{87.5}\text{Cr}_{4.17}\text{Ti}_{4.17}\text{Nb}_{4.17}$ alloy, an important candidate structural material for fusion reactors,²³ were studied for comparison.

2. METHODS

The thickness and width of metallic glass strip $\text{Ta}_{38}\text{Ni}_{62}$ were 30 μm and 2 mm, respectively. The sample dimensions of polycrystalline W were 5 mm $\times$ 10 mm $\times$ 2 mm. The sample dimensions of $\text{V}_{87.5}\text{Cr}_{4.17}\text{Ti}_{4.17}\text{Nb}_{4.17}$ alloy were 2 mm $\times$ 6 mm $\times$ 2 mm. After pre-grinding and polishing the metallic W and $\text{V}_{87.5}\text{Cr}_{4.17}\text{Ti}_{4.17}\text{Nb}_{4.17}$ alloy, metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ strips, metallic W, and $\text{V}_{87.5}\text{Cr}_{4.17}\text{Ti}_{4.17}\text{Nb}_{4.17}$ alloy were cleaned ultrasonically with acetone and alcohol. The $\text{Ta}_{38}\text{Ni}_{62}$, W, and $\text{V}_{87.5}\text{Cr}_{4.17}\text{Ti}_{4.17}\text{Nb}_{4.17}$ were then subjected to irradiation experiments.

Ion beam irradiation experiments were conducted on a 320-kV electron cyclotron resonance experimental platform at the Institute of Modern Physics, Chinese Academy of Sciences. During irradiation, samples were fixed to a rotating target platform in a vacuum target chamber at room temperature. The vacuum degree was maintained at 5×10^{-6} Pa. The beam intensity was 12 μA . The He^{2+} ion energy was 500 keV. The irradiation fluences were 1×10^{17} , 2×10^{17} , 5×10^{17} , and 1×10^{18} ions/ cm^2 .

A stopping and range of ions in matter (SRIM)²⁴ program was used to simulate the irradiation of incident helium ions in $\text{Ta}_{38}\text{Ni}_{62}$. After irradiation, grazing incidence

X-ray diffraction (XRD) was used to measure the phase structures of metallic glass Ta₃₈Ni₆₂ before and after He²⁺ ion irradiation. Metallic glass Ta₃₈Ni₆₂ cross-sectional transmission electron microscopy (TEM) by focused ion beam was used to observe microstructural changes and helium bubble distribution. Scanning electron microscopy (SEM) and atomic force microscopy (AFM) were used to observe the surface morphology and roughness.

3. RESULTS AND DISCUSSION

3.1 SRIM-calculated simulation results

The simulation of ion beam irradiation of incident helium ions in metallic glass Ta₃₈Ni₆₂ at 500 keV was calculated by Monte Carlo-based SRIM, which achieved a range of 1.01 μm and a sputtering yield of 0.002234 atoms/ion.²⁵ From the SRIM-simulated interaction of incident ions and materials, the incident depth-dependent distribution curves of material interior vacancies, replacement collisions, and helium atoms were calculated. Metallic glass has no vacancies unlike crystal, so the vacancies here refer to vacancy-type defects after atomic displacement.²⁶ According to the distribution of vacancy-type defects and replacement collisions, the displacement per atom (DPA) parameter was calculated to characterize the irradiation damage.²⁷ The distribution of helium concentration was calculated from the helium distribution and metallic glass Ta₃₈Ni₆₂ density (6.7715×10^{22} atom/cm³). Figure 1 shows the incident depth-dependent distribution curve for the DPA and the helium concentration caused by He ion beam irradiation of Ta₃₈Ni₆₂ at 500 keV and 1×10^{18} ions/cm². The incident depths that correspond to the peak DPA and peak helium concentration of Ta₃₈Ni₆₂ were in the vicinity of the range. The area where the helium concentration was greater than 10% is located 0.84–1.24 μm from the surface, with a thickness of 400 nm.

3.2 Evolution of microstructure with irradiation

Figure 2 shows the XRD pattern of metallic glass Ta₃₈Ni₆₂ before and after different He²⁺ irradiation fluences. The XRD spectrum shows a diffuse amorphous hillock, and metallic glass Ta₃₈Ni₆₂ maintained an amorphous state at different irradiation fluences. Metallic glass Ta₃₈Ni₆₂ had a higher glass transition temperature

T_g (993 K) and initial crystallization temperature T_x (1056 K),²⁸ but the temperature increase during He^{2+} irradiation was not obvious, and the target surface temperature was much lower than the glass transition temperature of $\text{Ta}_{38}\text{Ni}_{62}$. The He^{2+} beam irradiation resistance of metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ below its glass transition temperature was good and $\text{Ta}_{38}\text{Ni}_{62}$ maintained an amorphous state even at the highest He^{2+} irradiation fluence of 1×10^{18} ions/cm².

At the highest fluence (1×10^{18} ions/cm²), irradiated metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ was studied to determine the microstructure change and helium bubble distribution by TEM. Figure 3 shows the TEM cross-sectional image, electron diffraction image, and energy-dispersive X-ray spectroscopy (EDS) line scanning analysis image of metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ at 1×10^{18} ions/cm². The electron diffraction spectrums for an area near the surface (I), an area in the central ion trajectory (II), an area at the end of the range (III), and an area out of range (IV) of metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ are the same that there is only a amorphous diffraction ring as Fig.3b shows. Different depths of $\text{Ta}_{38}\text{Ni}_{62}$ after He^{2+} ion beam irradiation maintain a good amorphous state, and no crystals were generated.²⁹ It can be seen from the TEM cross-sectional image in Fig. 3a that a whitish belt layer was formed approximately 1 μm from the surface of the metallic glass $\text{Ta}_{38}\text{Ni}_{62}$. Its depth inside the $\text{Ta}_{38}\text{Ni}_{62}$ was approximately equal to the SRIM-calculated depth of peak helium concentration. Figure 3c shows the depth-dependent EDS analysis for metallic glass $\text{Ta}_{38}\text{Ni}_{62}$. The elemental Ta and Ni contents at the end of the range were slightly less than those in the central ion trajectory, which may be because of the retention of helium ions at the end of the range.

Figure 4 shows the through-focal TEM images for the whitish belt layer of He^{2+} -implanted metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ at 1×10^{18} ions/cm². In the under-focused condition (Fig. 4a), the bubbles exhibited bright Fresnel fringes, whereas in the over-focused condition (Fig. 4b), they exhibited dark Fresnel fringes. This phase contrast is associated with a change in electronic potential at the round defects. Such a change in electronic potential usually manifests as bubbles or voids.³⁰ The helium concentration for implantation under the experimental conditions was above the

threshold fluence for bubble formation, so the round defects are therefore He bubbles. As can be seen from the images, helium bubbles gathered at the end of the ion range. The helium bubble layer width was approximately 400 nm, which is similar to the thickness of the He concentration area, which is greater than 10% by SRIM calculation. Most helium bubbles were round, whereas some had small bars or irregular shapes after connection and coalescence in the growth process. The helium bubble size was largest in the central depth of the helium bubble layer and decreased gradually to the top and bottom sides.

Figure 5 shows high-resolution TEM images of He^{2+} -irradiated metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ at different depths at 1×10^{18} ions/cm². Figure 5a shows the high-resolution TEM image of $\text{Ta}_{38}\text{Ni}_{62}$ near the surface area (Fig. 3I). Tiny helium bubbles are visible and the target atoms are disordered, which confirms that $\text{Ta}_{38}\text{Ni}_{62}$ is amorphous even under irradiation at maximum fluence.³¹ Figure 5b shows a high-resolution TEM image of $\text{Ta}_{38}\text{Ni}_{62}$ in the central ion trajectory area, approximately 0.4 μm away from the surface (Fig. 3II). In this area, only a small amount of tiny helium bubbles existed, the atomic arrangement was disorderly, and the amorphous state was maintained. Figure 5c shows a high-resolution TEM image of the helium bubble layer at the end of the He ion range (Fig. 3III). The helium bubbles were distributed uniformly in this area, and their diameter was approximately 5 nm. Figure 5d shows a high-resolution TEM image of the area outside the He ion range (Fig. 3IV). Typically, the arrangement of atoms was amorphous, so the state before irradiation was maintained.

3.3 Evolution of surface morphology with He^{2+} irradiation

Figure 6 shows the SEM surface morphology images of metallic glass $\text{Ta}_{38}\text{Ni}_{62}$, metallic W, and $\text{V}_{87.5}\text{Cr}_{4.17}\text{Ti}_{4.17}\text{Nb}_{4.17}$ alloy before and after He^{2+} irradiation. The surface of metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ before and after irradiation was smooth, without apparent irradiation damage even for maximum fluence. The polishing-induced sunken areas on the metallic W surface became larger at 1×10^{18} ions/cm². The $\text{V}_{87.5}\text{Cr}_{4.17}\text{Ti}_{4.17}\text{Nb}_{4.17}$ alloy was scratched slightly by polishing before irradiation, blistered, and peeled seriously after irradiation, and multi-layer peeling even occurred in the peeled area at 1×10^{18} ions/cm².

The surface roughness of metallic glass Ta₃₈Ni₆₂ before and after different fluences of He²⁺ ion irradiation was researched further, and the AFM images of surface topography and surface roughness changes are shown in Fig. 7. The surfaces of the metallic glass Ta₃₈Ni₆₂ strip were smooth, and the root mean square (Rms) roughness values were small. The surface morphology of Ta₃₈Ni₆₂ after irradiation was rougher than that before irradiation, and the Rms roughness after irradiation was also greater than before. The greatest increase in Rms occurred from 1×10^{17} to 2×10^{17} ions/cm².

4. DISCUSSION

Ion bombardment could lead to sputtering in the material surface.³² Surface morphology evolution of the ion beam-irradiated solid material was caused by competition between ion bombardment-inducing roughness and internal material transportation-inducing smoothness.³³ The metallic glass surface was smooth because of its disordered structure, without the orientation-dependent sputtering phenomenon in crystal materials.³⁴ Therefore, there was no obvious sign of irradiation damage in the metallic glass Ta₃₈Ni₆₂ surface. The roughening of metallic glass by ion bombardment played a dominant role as the fluence increased, and the Rms roughness increased slightly. The surface of metallic W had small sunken areas, and the crystal material displayed an orientation-dependent sputtering phenomenon, which caused roughness and larger sunken areas on the metallic W surface.

The accumulation of incident helium ions could form bubbles in the material.³⁵ Cascade collision occurred inside the material during ion irradiation, and helium was retained at the end of the ion range. Helium as an inert gas atom has low solubility and diffusivity in metal. When helium ions were implanted into the material, most eventually stopped at the end of the ion range after a series of collisions, and the incident helium was captured mainly in grain boundaries, vacancies, and other defects. As the fluence increased, helium nucleated with high concentration. Once nucleated, the helium easily adsorbed other surrounding helium atoms and vacancies or merged to form helium bubbles, which grew in combination and connection. When the bubble pressure reached a certain critical value, the stress generated by internal pressure

could be released only through surface deformation, and resulted in blistering.³⁴ $V_{87.5}Cr_{4.17}Ti_{4.17}Nb_{4.17}$ alloy had a low helium solubility and diffusivity and therefore experienced multi-layer peeling. While the atoms inside metallic glass $Ta_{38}Ni_{62}$ were arranged disorderly in space without point defects, helium was limited by the free volume after incidence, and as the fluence accumulated, helium was difficult to gather in high amounts to form helium bubbles. Ni-based metallic glass had a higher absorbability and solubility for helium, thus no obvious blistering phenomenon resulted on the metallic glass $Ta_{38}Ni_{62}$ surface.

Under the same ion energy and fluence, metallic glass strip $Fe_{80}Si_{7.43}B_{12.57}$ ³⁷ and bulk metallic glass $Zr_{64}Cu_{17.8}Ni_{10.7}Al_{7.5}$ ³⁸ also maintained an amorphous state. Helium bubble layers of metallic glass $Fe_{80}Si_{7.43}B_{12.57}$ and $Zr_{64}Cu_{17.8}Ni_{10.7}Al_{7.5}$ were located approximately in the depth of the ion range. The largest helium bubble diameter of Fe-based metallic glass under the same fluence was approximately one fifth of that which we have found previously.³⁶ Research by Wang et al. focused on a larger fluence. The helium bubble diameter in Zr-based metallic glass in their study was approximately 10 times larger than in ours, and blistering, peeling, and flaking did not occur.³⁸ As a result of the SRIM calculation, the metallic glass $Fe_{80}Si_{7.43}B_{12.57}$ density was 8.4599×10^{22} atoms/cm³, that of metallic glass $Ta_{38}Ni_{62}$ was 6.7715×10^{22} atoms/cm³, and that of metallic glass $Zr_{64}Cu_{17.8}Ni_{10.7}Al_{7.5}$ was 5.3242×10^{22} atoms/cm³. The different amount of free volume in the metallic glass made their helium solubility different, thus the helium bubble size was different. Under their respective irradiation conditions, all three types of metallic glass conditions resisted He^{2+} ion irradiation better than W. The peak DPA of metallic glass $Ta_{38}Ni_{62}$ under He^{2+} irradiation at 1×10^{18} ions/cm² was 29 dpa, whereas the peak DPA of metallic W was 16 dpa, which was the DPA required for W in the demonstration power plant as the divertor for 5 years.³⁹ Although the peak DPA of metallic glass was greater than that of W, because of the short-range ordered and long-range disordered structure, metallic glass could not generate lattice defects like the crystal, and ion irradiation damage in metallic glass was smaller than that in the crystal. The He^{2+} ion irradiation damage in metallic glass was therefore smaller than metallic W. These results provide reference for the

selection of irradiation-resistant materials.

5. CONCLUSION

After exposure to different He^{2+} irradiation fluences at 500 keV, metallic glass strip $\text{Ta}_{38}\text{Ni}_{62}$ remained in an amorphous state, and irradiated atoms at different depths were characterized by their disordered amorphous structure. At 1×10^{18} ions/cm², no significant damage occurred on the metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ surface, and the Rms roughness value increased nonlinearly with increase in fluence. Helium bubble layers appeared approximately 1 μm away from the surface, with a depth that was approximately equal to the ion range, and many helium bubbles were distributed at the end of the ion range. The helium bubble size was largest in the central depth of the helium bubble layer and decreased gradually to the top and bottom sides. Larger sunken areas appeared in the metallic W surface, and multi-layer peeling occurred in the $\text{V}_{87.5}\text{Cr}_{4.17}\text{Ti}_{4.17}\text{Nb}_{4.17}$ alloy surface at 1×10^{18} ions/cm². The He^{2+} beam irradiation resistance of metallic glass $\text{Ta}_{38}\text{Ni}_{62}$ below its glass transition temperature was better than that of metallic W and $\text{V}_{87.5}\text{Cr}_{4.17}\text{Ti}_{4.17}\text{Nb}_{4.17}$.

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Figure captions

Figure 1. Incident depth-dependent distribution curve for DPA and helium concentration caused by He ion beam irradiation of Ta₃₈Ni₆₂ at 500 keV and 1×10^{18} ions/cm².

Figure 2. XRD patterns of metallic glass Ta₃₈Ni₆₂ before and after application of different He²⁺ irradiation fluences: (a) pristine, (b) 1×10^{17} ions/cm², (c) 2×10^{17} ions/cm², (d) 5×10^{17} ions/cm², and (e) 1×10^{18} ions/cm².

Figure 3. TEM images of metallic glass Ta₃₈Ni₆₂ at 1×10^{18} ions/cm²: (a) TEM cross-sectional image, (b) electron diffraction image, and (c) EDS line-scanning analysis.

Figure 4. Through-focal TEM images for whitish belt layer of He²⁺-implanted metallic glass Ta₃₈Ni₆₂ at 1×10^{18} ions/cm²: (a) under-focused view of He bubble layer in the damage zone and (b) over-focused view of He bubble layer in the damage zone.

Figure 5. High-resolution TEM images of He²⁺-irradiated metallic glass Ta₃₈Ni₆₂ at different depths at 1×10^{18} ions/cm²: (a) area near the surface, (b) area in the central ion trajectory approximately 0.4 μ m away from the surface, (c) area in the helium bubble layer at the end of the He ion range, and (d) area outside the He ion range.

Figure 6. SEM surface morphology images of metallic glass Ta₃₈Ni₆₂, metallic W, and V_{87.5}Cr_{4.17}Ti_{4.17}Nb_{4.17} alloy before and after He²⁺ ion irradiation.

Figure 7. AFM images of surface topography and Rms roughness changes of metallic glass Ta₃₈Ni₆₂ before and after different He²⁺ ion irradiation fluences: (a) pristine, (b) 1×10^{17} ions/cm², (c) 2×10^{17} ions/cm², (d) 5×10^{17} ions/cm², (e) 1×10^{18} ions/cm², and (f) change in Rms roughness with increase in fluence (z-scale 30 nm/division).

Figure 1.

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