Effect of Helium Irradiation Fluence on He\textsubscript{n}V\textsubscript{m} Cluster Evolution in Nickel Studied by Positron Annihilation Spectroscopy

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Polycrystalline nickel was irradiated using 50 keV He ions at room temperature. The irradiated fluences were $5 \times 10^{13}$ He$^+$ cm$^{-2}$, $5 \times 10^{14}$ He$^+$ cm$^{-2}$, and $5 \times 10^{15}$ He$^+$ cm$^{-2}$, respectively. Positron annihilation Doppler broadening spectroscopy (DBS) was used to characterize the irradiation-induced defect evolution. The DBS results show that a large amount of vacancy defects were introduced in the specimens after helium irradiation. In addition, the DBS data could be also interpreted as the formation of helium-vacancy (He\textsubscript{n}V\textsubscript{m}) clusters due to combination between vacancies and helium atoms.

1. Introduction

Nickel based alloys are potential structural materials in thorium molten salt reactors (TMSR) and supercritical water reactors (SCWR), due to their high molten salt corrosion resistance, creep resistance and high temperature strength [1–4]. The working circumstance of such structural materials is extreme, including high energy and flux neutron irradiation at high pressure and temperature. It is well known that interstitials and vacancies are produced by neutron displacement cascade [5]. Meanwhile, helium atoms are easily generated by $(n, \alpha)$ nuclear reactions [6]. The combination of vacancies and helium atoms forms He\textsubscript{n}V\textsubscript{m} clusters which are the primal nucleation sites of helium bubbles in structural materials. The existence of helium bubbles can enhance void swelling, surface blistering and degradation of mechanical properties [7–9]. Therefore, it is very important to investigate He\textsubscript{n}V\textsubscript{m} cluster evolution in nickel based alloys. Nickel based alloys are complicated and it is difficult to investigate the effect of helium irradiation fluence in these structural materials. Therefore nickel, as a typical face centered cubic material, was used in order to avoid interference from other components in the alloys. In the present work, positron annihilation Doppler broadening spectroscopy (DBS) was used to investigate the evolution of irradiation-induced defects since positrons are easily trapped by open space type defects [10].

2. Experimental detail

Polycrystalline nickel (purity: 99.999 \%) was used and the specimens were cut to $10 \times 10 \times 0.3$ mm$^3$ in size. The surface of each specimen was polished to a mirror-like finish through a combination of mechanical polishing and electrochemical polishing. All specimens were then annealed at 1223 K for 2 h at high vacuum ($\sim 10^{-5}$ Pa). In the present work, helium ion irradiation was performed at the HI-13 tandem accelerator facility at the China Institute of Atomic Energy (CIAE). Irradiation was carried out using 50 keV helium ions at room temperature, and irradiation fluences were $5 \times 10^{13}$ He$^+$ cm$^{-2}$.
The stopping and range of ions in matter (SRIM) software code was used to calculate the helium atom and vacancy distribution in nickel, and the results are shown in Fig. 1. The displacement energy of nickel was set to 40 eV in the SRIM simulation [11].

Mono-energetic positrons were implanted into the specimens with the implantation energy varied from 0.1 keV to 20 keV. Positrons are generated by a 0.713 GBq $^{22}$Na radiation source and moderated by a solid Ne moderator. A high-purity germanium detector (HPGe) was used to detect one of the annihilation $\gamma$-rays from the positron annihilation. The positron momentum is around 0.025 eV after thermalization so that the energy of the $\gamma$-ray photon mostly conveys information on the electron momentum [12]. The shape of the 511 keV $\gamma$-ray peak can then be parameterized with parameters which characterize the momentum distribution of annihilated electron. The $S$ parameter is defined as the ratio of counts in the low-momentum region (zero to $3 \times 10^{-3} m_0c$, where $m_0$ is the electron rest mass and $c$ is the speed of light) and the $W$ parameter as the ratio of counts in high-momentum region ($10.0 \times 10^{-3} m_0c$–$23.0 \times 10^{-3} m_0c$) to the total counts in the annihilation peak (zero to $\pm38.4 \times 10^{-3} m_0c$). For each spectra, a total of $2.0 \times 10^6$ counts were accumulated in order to reduce the statistical error.

3. Results and discussion

The SRIM result shows that the maximum irradiation depths of helium implantation and vacancy production are about 310 nm and 300 nm, and the peaks of helium atoms and vacancies are located at about 200 nm and 150 nm, respectively. The displacement damage (vacancies produced/atom = displacement per atom or dpa) and the vacancy distribution have the following relationship [13]:

$$V \times \left( \frac{10^7 \text{[nm cm}^{-1}\text{]} \times \text{Fluence [ions cm}^{-2}\text{]}}{D_{\text{atoms}} \text{[atoms cm}^{-3}\text{]}} \right) = \text{dpa},$$

where $V$ is the value of the vacancy distribution (0.5488 vacancies (ions nm)$^{-1}$), $D_{\text{atoms}}$ is the atomic density of the material (the value of $D_{\text{atoms}}$ for nickel is $9.127 \times 10^{22}$ atoms cm$^{-3}$). The helium irrad-
Fig. 2  $S$ parameter as a function of incident positron energy (mean implantation depth) for nickel. The data points of $S$ parameter are fitted by VEPFIT program.

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and diffusion area) according to the SRIM result. The positron trapping rate $k$ is proportional to the vacancy density $C_v$ [17]:

$$k = \mu C_v,$$

(3)

where $\mu$ is the defect trapping coefficient. There is a positive correlation between the positron trapping rate $k$ and the vacancy density $C_v$. Vacancies are easily aggregated and form vacancy clusters, and the value of the positron trapping rate $k$ has a positive correlation with the size of the vacancy cluster [18, 19]. It is known that vacancies and vacancy clusters are helium atom trapping states, which can form $\text{He}_n\text{V}_m$ clusters. The stability of $\text{He}_n\text{V}_m$ clusters is related to their structure and the vacancy-to-He ratio. The $\text{He}_n\text{V}_m$ clusters always have highly symmetric structures corresponding to their minimum energy configurations of $\text{He}_n\text{V}_m$ clusters. The clusters have lowest energy configuration when the ratio of vacancy-to-He of $\text{He}_n\text{V}_m$ clusters is 1 : 1 [20]. $\text{He}_n\text{V}_m$ clusters can generate vacancy-interstitial pairs when the ratio of vacancy-to-He is less than 1 and can absorb helium atoms when the ratio of vacancy-to-He is more than 1. According to the SRIM result, the mean ratio of vacancy to helium atoms is 131 which is significantly more than 1. Therefore, the $\text{He}_n\text{V}_m$ clusters also belong to open space defects, and the characteristics of positron trapping at these clusters are similar to vacancy clusters. In the conventional positron trapping model, the total number of positrons annihilating in defects ($N_d$) has the following relationship with positron trapping rate $k$ [17]:

$$N_d = \frac{1}{1 + \lambda_f/k} \cdot N_0,$$

(4)

where $N_0$ is the total number of incident positrons and $\lambda_f$ is the positron annihilation rate in the perfect lattice. The $S$ parameter can be divided into two parts, $S_f$ and $S_d$ corresponding to the value of $S$ when all positrons annihilate in the crystal lattice or in defect states, respectively. Therefore, the $S$ parameter can be written as [21]:

$$S = \frac{S_f \cdot (N_0 - N_d) + S_d \cdot N_d}{N_0}$$

(5)

Fig. 3 $S$–$W$ plot for a non-irradiated reference nickel specimen, together with plots for the specimens irradiated with different helium irradiation fluence.
According to Eq. (3), Eq. (4) and Eq. (5), it could illustrate that the $S$ parameter has a positive correlation with positron trapping rate $k$. In the damage area, the density and size of open space defect clusters (He$_n$V$_m$ clusters) increases with increasing helium irradiation fluence [22]. The positron trapping rate $k$ and the number of positrons annihilated around defects increase with increasing defect density and the size of open space defects (He$_n$V$_m$ clusters). Therefore, the $S$ parameter also has a positive correlation with the density and sizes of defects. The migration energy of vacancies and helium atoms in nickel are 1.17 eV and 0.078 eV, respectively. Hence, helium atoms can migrate deeper than vacancies. The helium atoms tend to agglomerate with each other and form helium atom clusters, and such clusters could induce a vacancy and self-interstitial when the number of helium atoms exceeds 5 [20, 23, 24]. In the diffusion area, the density of open space defects was lower than that of the damage area. Therefore, the $S$ parameter keeps decreasing in the diffusion area.

Further information about the positron annihilation mechanism with different types of defects can be obtained by plotting the $S$ and $W$ parameters against each other, the so-called $S$–$W$ plot [25]. $S$–$W$ plots for a non-irradiated reference nickel specimen, together with the 3 specimens irradiated with different helium irradiation fluence are shown in Fig. 3. The $S$–$W$ curve of the reference specimen shows a linear relationship suggesting that the positron annihilation mechanism is similar at each annihilation site. After helium irradiation, the $S$–$W$ plots deviate from the reference plot and show a linear relationship (solid arrow) in the surface region because most positrons are trapped by vacancy type defects and annihilate with electrons around this defect. In the damage area, the linear relationship fades away and the data points tend to aggregate together. The gathering of data points along the direction of the dashed arrow suggests that the positron annihilation mechanism is similar in each of these irradiated specimens. After helium irradiation, many vacancies were introduced in the specimen, and He$_n$V$_m$ clusters were formed due to combination between vacancies and helium atoms. In the helium irradiation process, higher fluence needs a longer irradiation time for the same flux. Therefore, the helium atoms and vacancies have more time to diffuse, nucleate and grow, resulting in larger sized He$_n$V$_m$ clusters [22]. The size of open spaces increases with increasing size of He$_n$V$_m$ clusters when the ratio ($m/n$) is constant. The positron trapping rate and the number of positrons annihilated around He$_n$V$_m$ clusters are increased with increasing He$_n$V$_m$ clusters size. Therefore, the $S$–$W$ plots indicate the relative change of the size of He$_n$V$_m$ clusters, suggesting that their size increased with increasing helium irradiation fluence (dashed arrow).

4. Conclusion

Helium irradiation was carried out using 50 keV He$_{2+}$ ions in nickel at room temperature, and irradiation fluences were $5 \times 10^{13}$ He$^+$ cm$^{-2}$, $5 \times 10^{14}$ He$^+$ cm$^{-2}$, and $5 \times 10^{15}$ He$^+$ cm$^{-2}$. DBS was used to investigate the effect of helium irradiation fluence on He$_n$V$_m$ clusters evolution. After helium irradiation, the existence of open space defects seems to weaken the influence of the surface effect. In the damage area, the helium atoms combined with vacancies to form He$_n$V$_m$ clusters and the size of He$_n$V$_m$ clusters increased with increasing helium irradiation fluence.

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References