Physical Testing and Surface Morphology Studies of Stainless Steel Irradiated with Xe Ions

Chaoliang Xu1, Xiangbing Liu1, *, Yuanfei Li1, Wenqing Jia1, Wangjie Qian1, Qiwei Quan1, Jian Yin1

1Suzhou Nuclear Power Research Institute, Jiangsu province, China

*Corresponding author: liuxbing@cgnpc.com.cn

Abstract. 6 MeV Xe ions were used to irradiate austenitic stainless steel at room temperature. Three displacement damage levels of 2, 7 and 15 dpa were selected. Microstructure and surface morphology were characterized by transmission electron microscopy (TEM), positron annihilation lifetime spectroscopy (PLS) and atomic force microscope (AFM). PLS indicated that vacancy defects were introduced by ions irradiation. Vacancy clusters containing Xe will reduce the positron annihilation lifetime. High density of dislocation loops were observed by TEM. The dislocation loops size and density saturates after 7 dpa and the nature of dislocation loops can be deduced by its distribution. A surface step was detected by AFM measurements between irradiated region (uncovered) and unirradiated region (covered with nickel mesh). This indicate that the irradiation swelling phenomenon occur and swelling is closely related to irradiation damage. According to the step height, the volume swelling is about 1.7% and 4.2% after irradiated to 7 and 15 dpa.

Keywords: austenitic stainless steel; irradiation; microstructure; surface morphology

1. Introduction

Fuel assemblies and control rods are held by internal structures in pressurized water reactor (PWR). This internal structures are often produced by solution annealed 304 austenitic stainless steel (ASS) and cold worked 316 ASS and often exposed to a large neutron irradiation due to its position being closed to the reactor core. Under this condition, the corrosion resistance property and mechanical property will be degenerated. The property degeneration of ASS internals, such as irradiation-assisted stress corrosion cracking (IASCC), irradiation hardening and embrittlement, is detected in many nuclear power plant (NPP) and has been attracted extensive international attention.

Irradiation damage is the key reason of property degeneration according to previous studies. For example, the irradiation hardening and embrittlement is mainly due to the formation of all kinds of defects, especially frank loops. These frank loops will prevent the migration of of dislocations and cause the irradiation hardening phenomenon. In the processing of irradiation, the vacancy and interstitial migration is accompanied by alloying and solute elements movement with a preferential interaction to point defect sinks and cause the IASCC phenomenon. So understanding irradiation hardening and
IASCC should know the vacancy defects, dislocation loops formation and evolution. On the other hand, during irradiation, if there is no complete annihilation between vacancies and interstitials, cavities and microvoids will be formed by additional vacancies and cause the irradiation swelling phenomenon. So defect evolution analysis can provide more information on swelling.

In this studies, Xe ions were used to emulate the irradiation damage in ASS. Then transmission electron microscopy (TEM) under bright field and weak beam dark field condition is conducted to analyze the microstructural changes. Moreover, positron lifetime spectroscopy (PLS) was used to examine the vacancy type defects evolution. At last, the surface morphology was measured by atomic force microscope (AFM).

2. Experiments
The ASS used in this study is provided by Bao Steel. It is solution treated at 1060 °C for 90 min before irradiation. The chemical composition shows in Table 1. The square specimens with a dimension of 15×15×1mm is machined by wire cutting. The silicon carbide paper up to grades 2000 and 0.5 μm diamond spray is used to polish the specimen surface. Then the mirror-like surface is cleaning with deionized water.

Irradiation was performed using 6 MeV Xe ions to fluences of 6.6×10^{14}, 2.3×10^{15} and 5×10^{15} Xe/cm². According to Monte Carlo simulation program SRIM 2012, these fluence correspond to 2, 7 and 15 dpa respectively at peak damage region. In the calculation process, the threshold displacement energy (E_d) of 40 eV is selected. Fig. 1 is the damage profiles obtained from SRIM calculation. According to fig. 1, the peak damage is located at 750 nm. During irradiation, a sheet of nickel mesh with 20 μm hexagon holes is used to cover parts of irradiation area.

| Table 1 Chemical composition of the stainless steel |
|-----------------------------------------------|
| Element | Cr | Ni | Mo | Mn | Cu |
| Wt.%    | 17.28% | 11.65% | 2.49% | 1.24% | 0.46% |
| Element | Si | C | Co | P | S |
| Wt.%    | 0.340% | 0.038% | 0.010% | 0.008% | 0.003% |

*Balance of composition is Fe.*

![Fig. 1. Displacement damage and Xe distribution versus depth after Xe irradiation](image-url)
The PLS were measured with a $^{22}$Na positron source. The BaF$_2$ lifetime spectrometer was used to collect annihilation counts. In each spectrum, more than $10^6$ counts were collected. Several thin TEM specimens were prepared by FEI Quanta 2003D focus ion beam. Then a FEI Tecnai G$^2$ F20 analytical TEM was used to analyze the microstructural characterization under bright field and weak beam dark field condition. Veeco Dimension 3100 AFM is used to give information about the surface morphology of irradiated specimens. The step between irradiated and unirradiated area covered by nickel mesh can give the quantitative information of irradiation swelling.

3. Results and analysis

3.1. PLS analysis

Large amount of vacancy-type defects will be produced by ions irradiation. Vacancy-type defects will capture positrons. The variation of size and concentration of vacancy-type defects will effect the positron lifetime and relative intensity. So we can analyze the size and concentration of vacancy-type defects by means of corresponding annihilation lifetime and intensity. Two lifetime components, namely short lifetime parameter $\tau_1$ and long lifetime parameter $\tau_2$, were used to fit the measured positron lifetime data. The positron mean lifetime $\tau_{av}$ can be expressed as $\tau_{av}=\tau_1I_1+\tau_2I_2$, where $I_1$ and $I_2$ are the intensity of $\tau_1$ and $\tau_2$ and can provide total number of residual defects.

Annihilation lifetime $\tau$ and intensity I variation against irradiation damage are shown in fig. 2$^{[1]}$. Compared to unirradiated specimen, $\tau_1$ show a significant increase after irradiated to 7 dpa. This indicated that Xe irradiation produce large amount of simple open-volume defects such as mono-vacancies and different size vacancy clusters. While after irradiated to 15 dpa, $\tau_1$ shows a significant decrease. Generally, $\tau_1$ will not decrease at present condition because $\tau_1$ response the average size reduction of open-volume defects. So there may be other factors. According to previous studies$^{[2]}$, this factor may be the gas atoms in vacancy clusters, which will reduce the positron annihilation lifetime. So the implanted Xe atoms combining with vacancy clusters can explain the decrease of lifetime $\tau_1$ after irradiated to 15 dpa. On the other hand, when the clusters contain gaseous atoms, its pressure and size would grow larger. In order to release pressure, the lattice matrix atoms will be collided due to the mutual push to decrease the pressure, causing the size of bubble increase. So visible bubbles can be formed. However, because Xe atom concentration (100 appm/dpa) in ASS is very low and irradiation is conducted at room temperature, the size of Xe bubble will not grow very big. This deduction is in consistent with our previous results, who use TEM with underfocus and overfoucs mode and observe several bubbles with a size about 1 nm$^{[3]}$.

![Fig 2. The variations of positron annihilation lifetime parameters and intensity parameters](image-url)
3.2. TEM analysis

Fig. 3 is the TEM images of ASS irradiated to 7 and 15dpa under bright field and weak beam dark field condition. It is shown two adjoining contrast lobes, namely one black and one white. There are also some more complex contrast shapes consisting of several lobes. The dislocation loops show a uniform distribution with a size 3-13nm. The number density according to statistics is about $10^{22}$ m$^{-3}$. The size and density of dislocation loops do not show a significant variation after irradiated to 7 dpa. Actually, Etienne et al. [4] focused on the density of dislocation loops and compared the data from literature and experimental results and gave a conclusion that the number density increase up to 1 dpa and saturate for higher damage level. Moreover, he pointed out that the loop size increases with the dpa and saturate between 5 dpa and 10 dpa with the size of 6–12 nm. In fact, because the characteristic of dislocation loops is greatly affected by irradiation parameters, it is possible to obtain different dislocation loops data. For example, the irradiation temperature has a significant influence on dislocation loop size and density once the irradiation temperature is larger than 300 °C. When the irradiation temperature is lower than 300 °C, no significant variation can be observed. The dislocation loop size and density is also affected by ion species because heavy ions will introduce more dislocation loops efficiently. Therefore, considering these factors, the dislocation loops obtained in fig. 3 is similar to previous studies.

![Fig 3. The TEM results of the stainless steel irradiated to displacement damage of 7 (left) and 15 dpa (right) (up: bright field; down:weak beam dark field)](image)

The nature of dislocation loops has not obtained a consistent conclusion. Previous Edwards et al.[5] considered that dislocation loops may be interstitial type or vacancy type. Boulanger et al.[6] believe that dislocation loops are only interstitial. In fact, according to fig. 3, we can observe some stacking fault. The fact that dislocation loops are distributed around the stacking fault can give us more information on the nature of dislocation loops. Comparing to interstitials in steels, vacancies have lower migration rate. The migration energy at room temperature is more than 0.5 eV for vacancies and less than 0.25 eV for interstitials [7]. Interstitials with high migration rate will preferentially move to imperfect area and annihilate with vacancies near stacking fault. Moreover, because interstitial loop nucleation is much less sensitive to vacancy involvement than vacancy loop nucleation to interstitial involvement, interstitial
loop nucleation is easier than vacancy loop nucleation. So from the discussion above, the dislocation loops observed in fig. 3 are supported to be interstitial.

3.3. AFM analysis
Fig. 4 is the variations of surface morphology of unirradiated ASS and ASS irradiated to 7 and 15 dpa. It is indicated that a significant surface modifications were observed and the significance is increase with irradiation damage level. The hillocks with nano-scale formed after irradiated to 7dpa, which is different from the unirradiated specimens. with the irradiation damage increase, the size of hillocks increase and density decrease, which suggest a formation of voids.

![AFM images of unirradiated ASS and ASS irradiated to 7, 15 and 25dpa.](image)

Fig 4. AFM images of unirradiated ASS and ASS irradiated to 7, 15 and 25dpa.

Fig. 5 is a typical cross sectional height obtained from the AFM image. The step with the height of about 10 nm after irradiated to 7dpa. A insignificant ridge can be observed. After irradiated to 15 dpa, the step height increased to 25 nm and a ridge formed with a height 10 nm and width of 0.5 μm. The continuous increase of step height reflect the volume swelling. If assuming the irradiated damage distribution range is about 800 nm according to fig. 1, it can be deduced the volume swelling is about 1.3% and 3.2% after irradiated to 7 and 15 dpa.
Fig 5. the AFM image of step height of ASS irradiated to 7 and 15 dpa

4. Conclusion

6 MeV Xe ions were used to irradiate ASS at room temperature. Three displacement damage levels of 2, 7 and 15 dpa were selected. Microstructure and surface morphology were characterized by TEM, PLS and AFM. PLS indicated that vacancy defects were introduced by ions irradiation. Vacancy clusters containing Xe will reduce the positron annihilation lifetime. High density of dislocation loops can be observed by TEM. The dislocation loops size and density saturates after 7 dpa and the nature of dislocation loops can be deduced by its distribution. A surface step was detected by AFM measurements between irradiated region (uncovered) and unirradiated region (covered with nickel mesh). This indicate that the irradiation swelling phenomenon occur and swelling is closely related to irradiation damage. According to the step height, the volume swelling is about 1.7% and 4.2% after irradiated to 7 and 15 dpa.

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