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Temperature and dose dependences of radiation damage in modified stainless steel

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Abstract

Radiation damage in a modified 316L stainless steel has been investigated as functions of the irradiation temperature from room temperature to 802 °C at 21 dpa and 33 dpa and the irradiation dose up to 100 dpa at room temperature by the heavy ion irradiation simulation and positron annihilation lifetime techniques. The maximum life time was observed at 580 °C for 21 dpa and 33 dpa irradiations, where the vacancy clusters contain 14 and 19 vacancies and have average diameters of 0.68 nm and 0.82 nm, respectively. For irradiations at room temperature up to 100 dpa the size of the vacancy clusters increases with increasing irradiation dose, and the vacancy clusters contain eight vacancies and reach 0.55 nm in diameter at 100 dpa. The experimental results reveal that the radiation damage in this modified 316L stainless steel is more sensitive to the irradiation temperature than the irradiation dose.

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1. Introduction

Stainless steels are often used as target structural materials for spallation neutron sources. The spallation neutron source system is one of the key parts of the accelerator driven system (ADS) [1–3], which provides source neutrons for driving a sub-critical assembly. Stainless steels (SS) are used for the beam window and structural materials of the ADS spallation neutron source system. They are irradiated by high-energy protons and spallation neutrons during operation. The accumulated displacement damage dose could be up to hundred dpa per year. The degradation of mechanical properties of SS induced by radiation damage depends on the irradiation temperature and dose, which could lead to ADS breakdown or even accidents. Therefore, the radiation damage study of SS is of great importance for estimating the lifetime of components and the safe operation of the ADS system. Also, it is desirable to develop a new type of SS that has a good radiation-resistant property.

The present work was motivated to investigate the radiation damage and the radiation-resistant property of a modified 316L stainless steel. Since no neutron and proton sources can be available in a laboratory to directly study radiation damage at such high doses encountered in the ADS, the heavy ion irradiation

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simulation technique was utilized [4]. The radiation damage rate of heavy ions is much higher than that of neutrons and protons. It just takes minutes or hours to reach tens of dpa by energetic heavy ion irradiation rather then several hundred days or years by direct proton or neutron irradiation. Therefore, in the present study the heavy ion irradiation was adopted to simulate the high dose proton and neutron irradiation. The positron annihilation lifetime technique was employed to investigate radiation induced damage microscopically.

2. Experimental

The material used in the experiment was the modified 316 austenitic SS (MSS). The size of samples was ϕ 23 mm × 0.5 mm. The sample thickness was chosen according to the requirement of positron annihilation measurement in which all positrons must annihilate in the sample in question, so the sample thickness is at least five times greater than the positron range. MSS is composed of Cr-15.05%, Ni-14.76%, Ti-0.32%, P-0.007%, S-0.007%, Mn-1.78%, Si-0.52%, C-0.048% and Fe balanced to 100% (all in weight percent) and cold-worked by 20%. The cold-working and minor Ti element addition aimed at improving radiation-resistant property, especially, reducing the radiation swelling. The samples were mechanically polished to a mirror-like surface.

The high-energy heavy ion irradiation was performed to simulate high dose neutron and/or proton irradiation. The samples for the temperature dependence study were irradiated by the 70 MeV ^{12}C ions of ~ 1μA beam current from the HI-13 tandem accelerator at China Institute of Atomic Energy. The irradiation was conducted in a variable-temperature and multi-sample irradiation chamber. The irradiation temperature ranged from room temperature to 802 °C with an accuracy of ±5 °C. The sample was heated by a heater during irradiation. The heating was fully controlled by a temperature controller. Also the irradiating beam deposited a certain amount of heat in the sample, and thus the beam current was kept stable around ~1 μA for an easy control of the temperature. The temperature dependence was performed at two doses of 21 dpa and 33 dpa with a damage rate of 2.1 dpa h^{-1}. In the dose dependence measurements 80 MeV ^{19}F ions were used to irradiate the samples. The irradiation was carried out at room temperature in which the sample was cooled by flowing water. The irradiation dose ranged from 0 dpa to 100 dpa with a damage rate of 3.9 dpa h^{-1}. The displacements per atom generated by the heavy ion irradiation in the samples were calculated using the TRIM program [5,6]. This program was also employed to calculate the irradiation depth, and the radiation damage was mainly located in a ~40 μm and ~20 μm layer under the surface for 70 MeV ^{12}C ion and 80 MeV ^{19}F irradiations, respectively.

The radiation damage produced in the samples was examined by a positron annihilation lifetime technique. The positron lifetime measurements were performed at room temperature for the un-irradiated samples and the samples irradiated at different irradiation temperatures and doses, using a fast–fast coincidence positron lifetime spectrometer consisting of a pair of BaF_2 scintillation detectors. The time resolution of the spectrometer is 210 ps for ^{60}Co γ rays. Two identical samples irradiated at the same condition were arranged like a sandwich with a ^{22}Na positron source of 1.1 MBq in the center. Besides the source components, all measured positron lifetime spectra were fitted by the PATFIT [7] or LT [8] program with two lifetime components, and the fitting variance was less than 1.3.

3. Results and discussion

The annihilation lifetime τ_f of free positrons is 110 ps in SS and the annihilation lifetimes of positrons trapped at the mono-vacancy, di-vacancy and dislocation are $\tau_{1v} = 1.3\tau_f$, $\tau_{2v} = 1.5\tau_f$, and $\tau_{dis} = 169$ ps, respectively [9].

For the un-irradiated MSS samples $\tau_1 = 147$ ps and $\tau_2 = 271$ ps were obtained. Here the obtained τ_1 is assumed to be a weighted average of annihilation lifetimes of the free positrons and the positrons trapped at the mono- and di-vacancies and dislocations, and τ_2 is attributed to the vacancy clusters or voids (this definition of τ_1 and τ_2 is used throughout this paper). The annealing temperature dependence of τ_1 and τ_2 was first measured for the un-irradiated MSS samples. τ_1 decreases with increasing the annealing temperature and reaches 110 ps at 800 °C, and τ_2 approaches to 255 ps at 800 °C.

For the irradiated MSS samples, the dependence of τ_1 and τ_2 on irradiation temperature is shown in Fig. 1 for 21 dpa irradiation. At room temperature τ_2 is almost the same as τ_2 in the un-irradiated samples, while τ_1 is 155.3 ps. Both τ_1 and τ_2 reach their peak values of 157.4 ps and 373.0 ps at 580 °C. At 802 °C, $\tau_1 = 128.2$ ps and $\tau_2 = 307.1$ ps, and both of them are larger than the values of τ_1 and τ_2 in the un-irradiated sample annealed at 800 °C. It can be seen from Fig. 1 that the mono- and di-vacancies, dislocations and different size vacancy clusters (or voids) were produced at different irradiation temperatures in MSS irradiated by 70 MeV carbon ions to a dose of 21 dpa. The fractions of the mono- and di-vacancies and dislocations decrease with increasing the irradiation temperature except at 580 °C, which can be interpreted by the fact that τ_1 is a weighted average of annihilation lifetimes of the free positrons and the positrons trapped at the mono- and di-vacancies and dislocations and the free positron

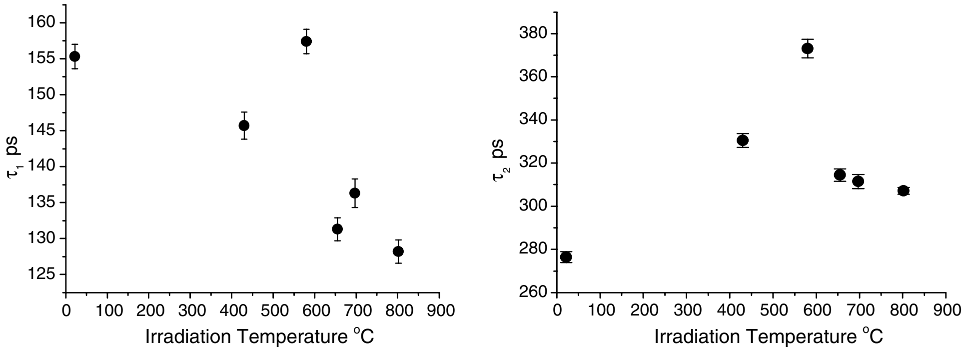

Fig. 1. Positron annihilation lifetimes τ_1 and τ_2 as a function of irradiation temperature in MSS irradiated by 70 MeV carbon ions to 21 dpa.

lifetime is the smallest among them. τ_1 shows a peak at 580 °C. The competition between the defect combination by thermal motion and the defect thermal annealing causes an increase of the di-vacancy fraction, resulting in a peak at this temperature (see below). The biggest voids or clusters characterized by τ_2 were also observed at 580 °C, which will be discussed below.

Fig. 2 shows the irradiation temperature dependence of positron lifetime τ_2 in MSS irradiated to 21 dpa and 33 dpa. The temperature dependence obtained at 33 dpa also exhibits a peak at 580 °C. It can be seen that below the peak temperature the variation of positron lifetime τ_2 with irradiation dose increases with irradiation temperature. The higher the irradiation temperature, the larger increase of lifetime τ_2 with irradiation dose. This also indicates that the radiation damage depends more sensitively on irradiation temperature than on irradiation dose.

The radius of voids can be estimated by $R_v = (NZ)^{1/3} r_s$ or $R_v = (N)^{1/3} R_{ws}$ [10,11], where R_{ws} is the Wigner–Seits radius, N is the number of vacancies contained in a void that was given by Puska et al. [12], Z is the valence, $r_s = (0.75\pi n)^{1/3}$ is the density parameter in the unit of Bohr radius a_0 and n is the number density of conduction electrons. For iron we have $r_s = 2.12a_0$, $Z = 2$ and $R_{ws} = 2.67a_0$ [12,13]. Fig. 3 shows the calculated results of average diameters of the observed voids or clusters at different irradiation temperatures up to 802 °C for the 21 dpa and 33 dpa irradiations. The temperature affects significantly the void diameter and thus radiation swelling. There is a swelling peak of SS in a temperature region of fast reactor operation. The swelling peak occurs usually in 450–650 °C, depending on the SS type. The swelling peak can be explained as follows: at lower irradiation temperatures the defects are less mobile and the probability to form larger clusters is small, at higher irradiation temperatures the vacancy annealing takes place, and the swelling occurs only in a certain temperature. In the present case the radiation swelling peak was observed at 580 °C and the corresponding voids contain 14 and 19 vacancies and have average diameters of 0.68 nm and 0.82 nm for 21 dpa and 33 dpa irradiations, respectively. As mentioned above, the cold-working treatment and adding of minor stabilizing elements such

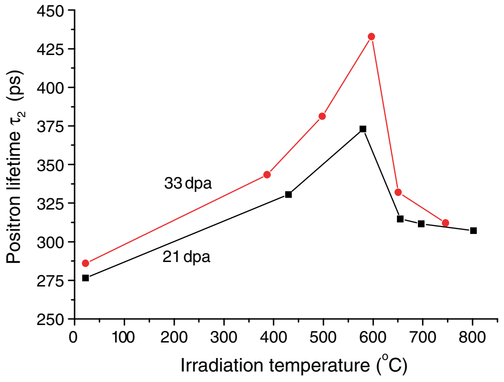

Fig. 2. Temperature dependence of positron lifetime τ_2 in MSS irradiated to 21 dpa and 33 dpa.

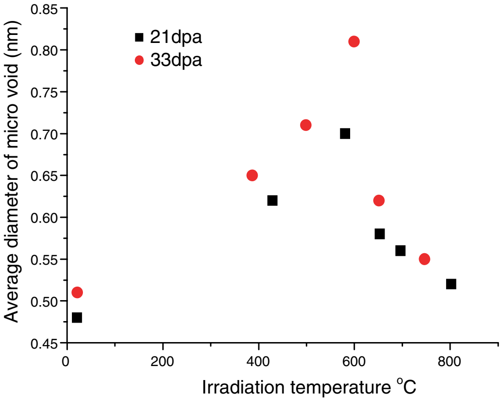

Fig. 3. Dependence of average void diameter on irradiation temperature in MSS irradiated to 21 dpa and 33 dpa.

as Ti etc can greatly suppress radiation swelling in SS. Though a radiation-swelling peak was detected at 580 °C, and the average diameter of voids is 0.68 or 0.82 nm at 21 dpa or 33 dpa. This swelling is much smaller than that in a common stainless steels, in which the void with an average diameter of 25.8 nm was found for the irradiation at 560 °C to a total dose of $3.2 \times 10^{22} n\ \mathrm{cm}^{-2}$ [14], which is equivalent to the present dose of heavy ion irradiation. This comparison was made based on the experiment, which demonstrates that the equivalent dose irradiations of neutrons and heavy ions produce the same radiation damage in the irradiated samples [15].

The dependence of the positron annihilation parameters τ_1 and τ_2 and I_1 and I_2 on irradiation dose is shown in Fig. 4 for MSS irradiated at room temperature by 80 MeV ^{19}F ions with a damage rate of 3.9 dpa h^{-1}. Here τ_1 and τ_2 are the weighted average positron lifetimes mentioned above, and I_1 and I_2 are the relative intensities of τ_1 and τ_2, respectively. I_1 and I_2 always satisfy the relation of $I_1 + I_2 = 1$. For the un-irradiated samples, the obtained τ_1 and τ_2 and I_2 are the same as those in the un-irradiated samples discussed before. It can be seen from Fig. 4 that τ_1 and τ_2 and I_2 all increase with increasing irradiation dose. This tells that irradiation generates the mono- and di-vacancies, dislocations and vacancy clusters or voids. Though the relative intensity I_1 decreases with increasing dose, the increase of τ_1 means the increase of the fractions of the produced mono- and di-vacancies and dislocations with increasing the irradiation dose. The lifetime τ_2 is closely connected to the size of vacancy clusters or voids, and longer lifetime τ_2 corresponds to a larger size of vacancy clusters. Therefore, the increase of τ_2 indicates the formation of larger size vacancy clusters. From the obtained lifetime τ_2 we arrive at that the vacancy clusters contain eight vacancies and reach 0.55 nm in diameter at 100 dpa. Compared to the vacancy cluster size in the un-irradiated MSS, the size increase at 100 dpa is less than 0.1 nm. Fig. 4 also shows a tendency that the lifetime τ_2 or vacancy cluster size approaches its saturated value at 75 dpa.

4. Summary

In summary, the dependence of radiation damage in the modified 316L stainless steel on irradiation temperature from room temperature to 802 °C and on irradiation dose up to 100 dpa has been investigated by heavy ion irradiation simulation and positron annihilation lifetime techniques. Under the heavy ion irradiation conditions, the variation of the produced vacancy clusters with irradiation temperature shows a peak corresponding to the largest size of vacancy clusters at 580 °C. The clusters contain 14 and 19 vacancies and have average diameters of 0.68 nm and 0.82 nm for the 21 dpa and 33 dpa irradiations, respectively. For the irradiation performed at room temperature the size of the vacancy clusters increases with increasing irradiation dose, and the vacancy clusters contain eight vacancies and the cluster size reaches 0.55 nm in diameter at 100 dpa. There is a tendency that the saturation of the vacancy cluster size starts at 75 dpa. It seems that the variation of radiation effects with irradiation dose increases with irradiation temperature and, the radiation damage in MSS is more sensitive to irradiation temperature than irradiation dose. The experimental results demonstrate to some extent that this modified 316L stainless steel (MSS) has a good radiation-resistant property, and the positron annihilation lifetime technique is a powerful tool to investigate the radiation damage on an atomic scale.

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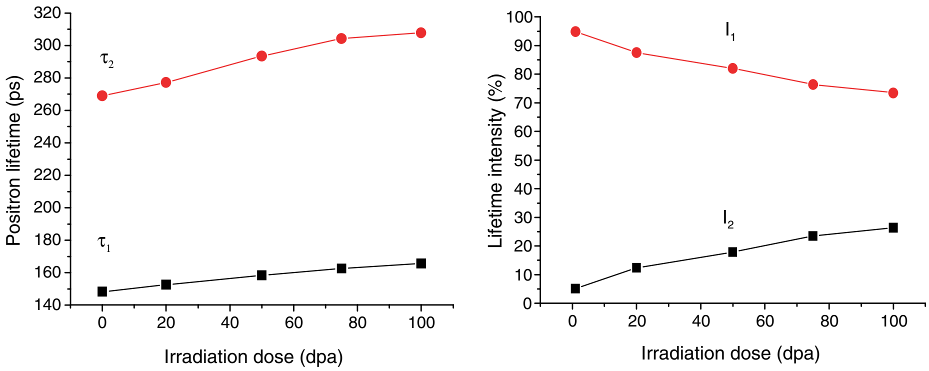

Fig. 4. Dose dependence of positron lifetimes and relative intensities in MSS irradiated by 80 MeV ^{19}F ions at room temperature.

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