High-dose neutron induced radiation swelling simulated by heavy ion irradiation and its microscopic study with positron annihilation technique

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Abstract 70 MeV-carbon-ion irradiation is used to simulate the radiation swelling induced by neutron irradiation of $3.2\times10^{22}\,n\cdot cm^{-2}$ in domestically-made 316 austenitic stainless steels modified by a 20%-cold-working and Ti-adding from room temperature to 802°C. The created swelling is microscopically examined by the positron annihilation lifetime technique. A radiation swelling peak is observed at 580°C and the corresponding void has an average diameter of 0.7 nm which is hardly probed by macroscopic methods.

Keywords Stainless steel, Radiation swelling, Heavy ion irradiation simulation, Positron annihilation lifetime technique

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1 Introduction

Stainless steel (SS) is an important reactor structural material. SS used in reactors experiences neutron irradiation and the accumulated dose may reach $10^{22\sim23}\,n\cdot cm^{-2}$ after years of reactor operation. Consequently, severe radiation effects occur in SS and would lead to a reactor breakdown or even accident. Radiation swelling is a major radiation effect in SS which should be taken into account in reactor design and operation.

Severe swelling generally occurs at neutron doses of $\geq 10^{20} n\cdot cm^{-2}$. The lack of neutron producing sources of such high doses hampers its investigation. The advent of heavy ion accelerators opens up a way to study the radiation swelling at such high doses[1]. The production mechanism of radiation damage is almost the same for neutrons and heavy ions, but the radiation damage rate of heavy ions is much higher. An irradiation dose of ~20 dpa (equivalent to $10^{22}\,n\cdot cm^{-2}$)takes a year or more in a reactor, whereas only a few minutes or hours by heavy ion irradiation. Therefore, use can be made of heavy ions to simulate severe radiation swelling.

The domestically made 316 austenitic SS modified by the 20% cold-working and Ti-addition is expected to have a good property of radiation swelling resistance, which needs to be tested experimentally. Thus, the present work was motivated to test the high-dose-neutron induced radiation swelling in it by a heavy ion irradiation simulation and a positron annihilation lifetime technique which is a very sensitive tool of investigating defects on an atomic scale.

2 Experiments

The samples used in the experiment were the home-made 316 austenitic SS, the

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size of which was 15 mm×15 mm×0.4 mm. They were made from 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% and treated by a 20% cold-working. The cold-working and Ti- addition aimed at reducing radiation swelling. The samples were mechanically polished to a mirror-like surface.

The samples were irradiated by 70 MeV carbon ions from the HI-13 tandem accelerator at China Institute of Atomic Energy. The sample irradiation was performed in a variable-temperature and multi-sample irradiation chamber from room temperature to 802°C with an accuracy of ±5°C. At each temperature the irradiation time was ~10 h at a beam current of 1μA and total irradiation dose was ~1.61×10^{17} C·cm^{-2} which generated 21dpa in SS and corresponded to 3.2×10^{22}n·cm^{-2} neutron irradiation.

The induced radiation swelling in SS was examined by a positron annihilation lifetime technique. The positron lifetime measurements were performed for both irradiated and unirradiated but annealed samples. The annealing of the unirradiated samples was done for~1 h at the same temperatures as the irradiation temperatures.

The positron annihilation lifetime measurements were carried out at room temperature, using a fast-fast coincidence positron lifetime spectrometer consisting of a pair of BaF_2 scintillation detectors. Its time resolution is 210 ps to ^{60}Co γ rays. Two identical samples were arranged as a sandwich with a 1.48 MBq ^{22}Na positron source in the center. Besides the source components, all measured lifetime spectra were well fitted by two lifetime components with a fitting variance of less than 1.3.

3 Results and discussion

The free annihilation lifetime, τ_f, of positrons is 110 ps and the annihilation lifetimes of positrons trapped at the monovacancy and divacancy are $\tau_{1v}=1.3\tau_f$ and $\tau_{2v}=1.5\tau_f$, respectively[2]. τ_1=147 ps and τ_2=271 ps were obtained for the unirradiated SS samples without annealing. τ_1 is assumed to be a weighted average of annihilation lifetimes of free positrons and positrons trapped at the mono- and di-vacancies and τ_2 to the small vacancy clusters. τ_1 decreases drastically with annealing temperature above 400°C and approaches to 110 ps at 800°C and τ_2 slowly decreases and reaches 255 ps at 800°C.

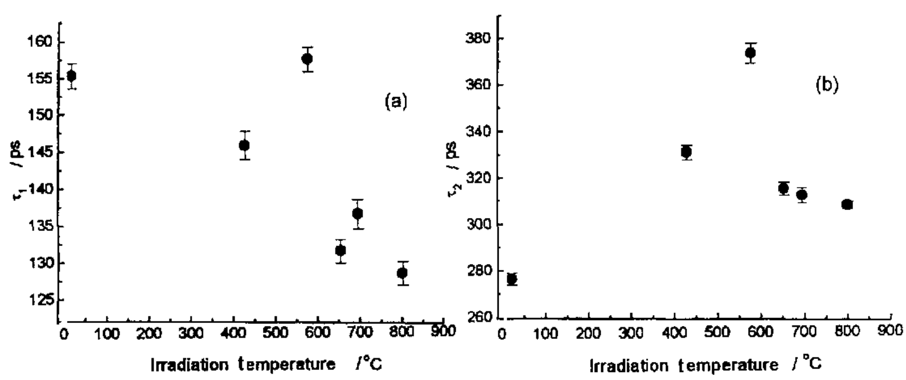

Fig.1 Lifetimes τ_1 and τ_2 as a function of irradiation temperature in the modified SS irradiated by 70 MeV carbon ions of 1.61×10^{17}C·cm^{-2}

The dependence of τ_1 and τ_2 on irradiation temperature is shown in Fig.1 for the irradiated samples. At room temperature τ_1 is 155.3 ps and τ_2 is 276.4 ps. For the 580°C irradiation, both τ_1 and τ_2 reach their peak values of 157.4 ps and 373.0 ps, respectively. At 802°C τ_1 takes a value of 128.2 ps and τ_2 a value of 307.1 ps, and both of them are larger than the values of τ_1 and τ_2 in the unirradiated sample but annealed at 800°C. It can be seen from Fig.1 that di-vacancies and different size vacancy clusters (voids) were created in these modified stainless steels irradiated by 70 MeV carbon ions of 1.61×10^{17} C·cm^{-2} from room temperature to 802°C. The biggest void was observed at 580°C.

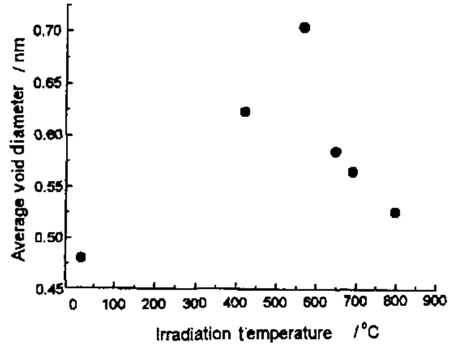

Fig.2 Average void diameter as a function of irradiation temperature in the modified SS irradiated by 70 MeV carbon ions of 1.61×10^{17} C·cm^{-2}

The radius of voids can be estimated by $R_v = (NZ)^{1/3}\ r_s$ or $R_v=(N)^{1/3}R_{ws}$[3,4], where R_{ws} is the Wigner-Seits radius, N is the number of vacancies contained in a void, Z is the valence, $r_s=(0.75\pi n)^{1/3}$ is the density parameter in unit of Bohr radius a_0 and n is the number density of conduction electrons. For iron $r_s=2.12a_0$, Z=2 and $R_{ws}=2.67a_0$[5,6]. Fig.2 shows the calculated results of the average diameter of the observed clusters at different irradiation temperatures. The void contains 14 vacancies and has an average diameter of 0.7 nm at 580°C, which is hardly inspected by the macroscopic ways. The radiation swelling is dependent on temperature and a swelling peak in SS is usually located in a temperature region of 450~600°C. In the present case the radiation swelling peak was detected at 580°C by considering the void size and its relative intensity obtained in the data fitting. This amount of swelling can be practically neglected. It indicates that the cold-working treatment and adding of minor stabilizing impurity such as Ti and Nb can greatly suppress radiation swelling in SS.

In summary, the heavy ion irradiation can be used to simulate high-dose-neutron induced radiation swelling and the positron annihilation technique to examine microscopically the produced swelling. The experimental results also clearly show that the proper cold-working and minor-impurity-addition will effectively improve the SS property against radiation swelling.

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