HEAVY ION IRRADIATION SIMULATION OF HIGH DOSE IRRADIATION INDUCED RADIATION EFFECTS IN MATERIALS

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Materials used for ADS, ITER, fast reactor, etc suffer very high dose irradiations of protons and/or neutrons. The yearly accumulated irradiation doses could reach a couple of hundred dpa in ADS, ~40 dpa in fast reactors and ~30 dpa in ITER's DEMO, producing severe radiation damage in materials and leading to a breakdown or accident of these installations. Investigation of such high dose irradiation induced radiation damage is a currently interesting topic with great importance. It is deeply hampered for lack of high dose neutron and proton sources. The heavy ion irradiation simulation technique has been developed at HI-13 tandem accelerator to investigate radiation damage encountered in the above mentioned installations. An experiment was carried out to verify the reliability and validity of heavy ion irradiation simulation. A series of experiments were performed by heavy ion irradiation simulation in combination with positron annihilation lifetime spectroscopy to investigate the temperature and dose dependence of radiation damage in stainless steels, tungsten, tantalum, etc. Some experimental results will be presented and discussed.

1. INTRODUCTION

The ADS and ITER are a novel innovative idea for the sustainable development of nuclear power system. However, radiation damage (RD) induced by high dose irradiations is a serious problem. Very high dose protons and/or neutrons irradiate the beam window, target and structural materials in the ADS and the structural, plasma facing and blanket materials in ITER. The yearly accumulated irradiation doses can reach a couple of hundred dpa in ADS, ~30 dpa in ITER's DEMO. In addition, for fast, fission and fusion reactors, the yearly accumulated neutron irradiation dose can arrives at 30-40 dpa in structural materials. The consequence of RD is radiation-induced micro-structural change that in turn causes macroscopic property changes of materials. RD greatly threatens the safe operation of nuclear installations and must be considered in their design and long-term operation.

The consequence of RD is radiation-induced microstructural change that in turn causes macroscopic property changes of materials. RD greatly threatens the safe operation of nuclear installations and must be considered in their design and long-term operation. The investigation of RD induced by high dose neutrons and/or protons is a currently interesting topic with great importance. The lack of high dose neutron and proton sources deeply hampers this investigation in laboratory. The advent of heavy ion accelerators provides a way for the laboratory study of RD produced by high dose neutron and/or proton irradiation. RD is caused mainly by atomic displacement in materials. The displacement rate of heavy ions is ~10^5-10^7 orders higher than that of neutrons and protons. High displacement rate of heavy ions significantly reduces the irradiation time. For examples, it takes 10 years for fission neutrons of 10^{14} cm^{-2}·s^{-1} to conduct an irradiation of ~30 dpa in fission reactors, and 1.5 years for 14 MeV fusion neutrons of 2 MW·m^{-2} in fusion reactors, while only a few hours or days by energetic heavy ion irradiation. Hence, the heavy ion irradiation simulation (HIIS) has been adopted to investigate the high dose neutron and proton induced RD. However, so far, no experiment has been performed to verify its reliability and validity. HIIS is often questioned, especially, for simulation of neutrons.

The HIIS technique has been developed at

China Institute of Atomic Energy (CIAE) to investigate RD encountered in the above mentioned installations in laboratory. A verification experiment for the reliability and validity of HIIS has been carried out for the first time by investigating RD and its thermal annealing behavior in α-Al_2O_3 irradiated at equivalent dose by 85 MeV ^{19}F ions and by $E_n \geq 1$ MeV neutrons, respectively. A series of experiments have been performed by HIIS to investigate RD in stainless steels, tungsten, tantalum, etc at irradiation temperatures ranging from room temperature to 800 ^oC and in the irradiation dose region up to 100 dpa.

2. EXPERIMENTAL VERIFICATION OF HIIS [1]

2.1. Experiment

The α-Al_2O_3 single crystal samples of 11x11x1 mm were used in the experiment. The En≥1 MeV neutron irradiation to a fluence of $3 \cdot 10^{20}$ cm^{-2} was performed at ~75 °C in the JJR-2 reactor at Japan Atomic Energy Research Institute. The heavy ion irradiation was carried out at China Institute of Atomic Energy. The samples were irradiated at room temperature by the 85 MeV ^{19}F ions from the HI-13 tandem accelerator to a fluence of $5.28 \cdot 10^{16}$ cm^{-2}, which is equivalent to the En≥1 MeV neutron fluence of $3 \cdot 10^{20}$ cm^{-2}. The post-irradiation annealing was conducted under nitrogen atmosphere for 40 minutes from 100 to 1050 °C in steps of 50 °C.

The detection of vacancies and voids or RD induced by irradiation was accomplished by a positron lifetime technique. The positron lifetime spectra were measured at room temperature after irradiation and annealing at different temperatures, using a conventional BaF_2 fast-fast coincidence positron lifetime spectrometer with a time resolution of 210 ps. Two identical samples were arranged as a sandwich with a 0.8 MBq positron source in the center. The lifetime spectra each contained $1.5 \cdot 10^6$ counts were analyzed with an LT program. Besides the source components, the measured lifetime spectra were fitted with two or three lifetime components. The fitting variance was all less than 1.3.

2.2. Results and discussion

The lifetime spectra were well fitted with two lifetime components τ_1 and τ_2 below ~450 °C, and the third long-lifetime component τ_3 needed to be added above ~450 °C, otherwise, the fitting variance was unacceptable. Fig. 1 shows as a function of post annealing temperature the obtained lifetimes τ_1, τ_2 and τ_3 and their intensities I_2 and I_3 ($I_1+I_2+I_3=1$) in the α-Al_2O_3 irradiated by neutrons and ^{19}F ions of equivalent dose, respectively. It can be seen that all the positron annihilation parameters of lifetime and intensity in the heavy ion irradiated α-Al_2O_3 are in good agreement with the ones in α-Al_2O_3 irradiated by neutrons of equivalent dose. This clearly shows that the RD induced by neutrons and heavy ions of equivalent dose and, even, its detailed thermal annealing behavior are exactly the same. Therefore, a conclusion can be made that the heavy ion irradiation can well simulate the neutron and/or proton irradiation and opens a way to reliably investigate the radiation effects generated by high dose neutron and/or proton irradiation that could hardly be performed within a reasonable time period in laboratory. Though the current experiment was carried out for the insulator α-Al_2O_3 irradiated by heavy ions and neutrons, one can imagine that if the heavy ion irradiation can reliably simulate the neutron irradiation, it can certainly simulate the proton irradiation; also if the heavy ion simulation can be used for insulators, it is no doubt for metals. The physics which can be extracted from the obtained positron lifetimes and intensities in the present experiment will be discussed elsewhere.

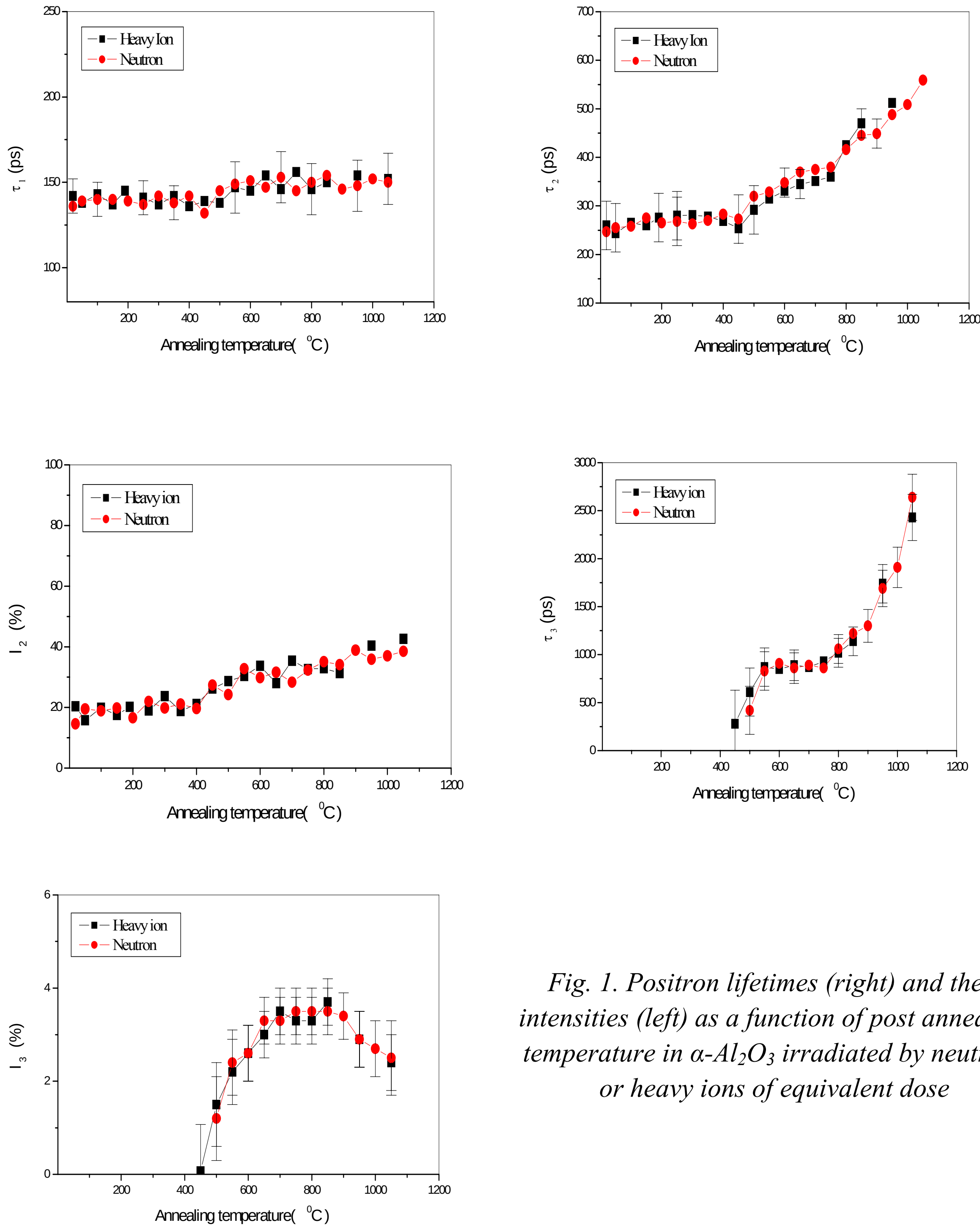

Fig. 1. Positron lifetimes (right) and their intensities (left) as a function of post annealing temperature in α-Al_2O_3 irradiated by neutrons or heavy ions of equivalent dose

3. EXAMPLES OF HIIS INVESTIGATION OF HIGH DOSE IRRADIATION INDUCED RD IN MATERIALS

3.1. Temperature and dose dependence of RD in modified stainless steel [2]

3.1.1. Experiment

The modified 316 austenitic SS (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% (wt.%) and cold-worked by 20%. The cold-working and minor element addition aimed at improving radiation resistant property, especially, reducing radiation swelling. The size of MSS samples was ϕ 23xx0.5 mm. The sample thickness was five times greater than the positron range. The samples were mechanically polished to a mirror-like surface.

In the temperature dependence study the samples were irradiated in a variable-temperature and multi-sample irradiation chamber by 70 MeV ^{12}C ions from the HI-13 tandem accelerator. The irradiation temperature ranged from room temperature to 802 °C with an accuracy of ±5 °C. The temperature dependence was performed at a damage rate of 2.1 dpa·h^{-1} for two doses of 21 and 33 dpa. In the dose dependence study 80 MeV ^{19}F ions were used to irradiate the samples at room temperature. The irradiation dose ranged from 0 to 100 dpa with a damage rate of 3.9 dpa·h^{-1}.

The positron lifetime measurements were performed at room temperature for the un-irradiated samples and the samples irradiated at different temperatures and doses.

3.1.2. Results and discussion

The annihilation lifetime τ_f of free positrons is 110 ps in stainless steel (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. 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 (these definitions of τ_1 and τ_2 are 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. 2 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, τ_1 =128.2 ps and τ_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. 2 that the mono- and di-vacancies, dislocations and different-size vacancy clusters (or voids) were produced at different irradiation temperatures in MSS. 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 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.

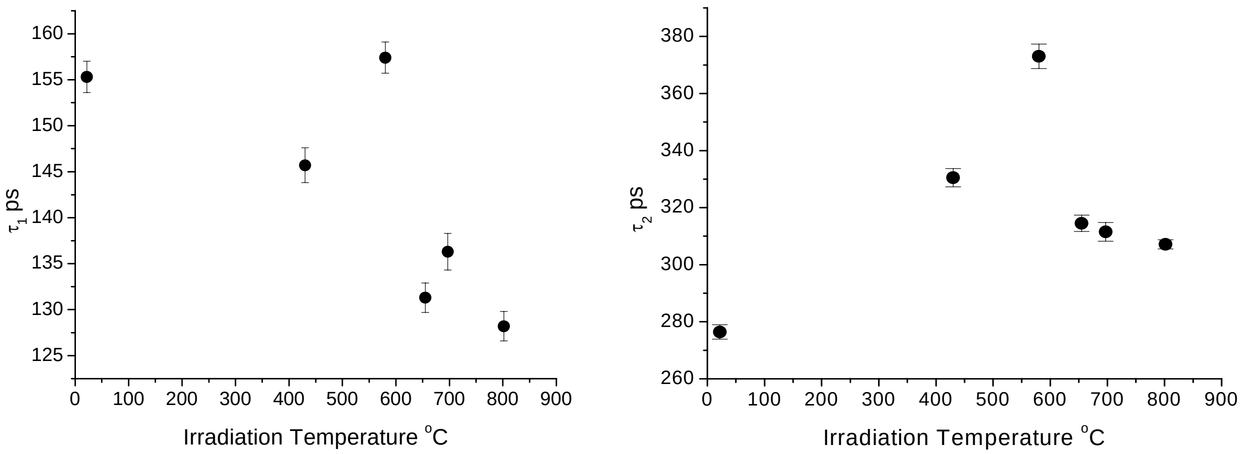

Fig. 2. Comparison of positron lifetime τ_2 in irradiated and unirradiated MSS

Fig. 3 shows the irradiation temperature dependence of positron lifetime τ_2 in MSS irradiated to 21 and 33 dpa. The temperature dependence obtained at 33 dpa also exhibits a peak at 580 ^{o}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 the increase of lifetime τ_2 with irradiation dose. This also indicates that the RD depends more sensitively on irradiation temperature than on irradiation dose.

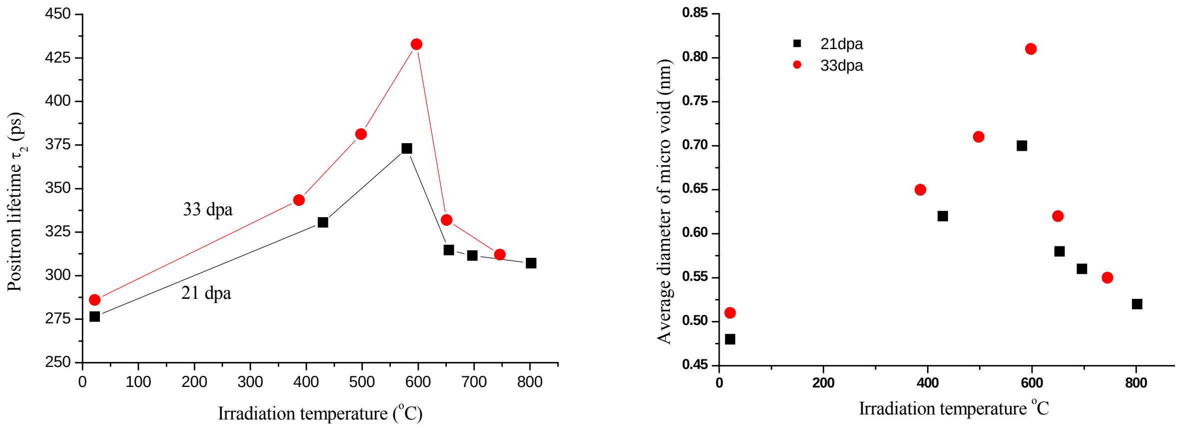

Fig. 3. Temperature dependence of positron lifetime τ_2 and average void diameter in MSS irradiated to 21 and 33 dpa

The radius of voids can be estimated by $R_v=(NZ)^{1/3}$ r_s or $R_v=(N)^{1/3}R_{ws}$, where R_{ws} is the Wigner-Seits radius, N is the number of vacancies contained in a void that can be deduced from τ_2, 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} = $2.67a_0$. Fig. 3 also shows the calculated average diameters of the observed voids or clusters at different irradiation temperatures up to 802 ^{o}C at the 21 and 33 dpa. 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 of SS occurs usually in 450-650 ^{o}C, depending on the SS-type. 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 ^{o}C and the corresponding voids contain 14 and 19 vacancies and have average diameters of 0.68 and 0.82 nm for 21 and 33 dpa irradiations, respectively. As mentioned above, the cold-working treatment and adding of minor stabilizing elements can greatly suppress radiation swelling in SS. Though a radiation-swelling peak was detected at 580 ^{o}C, and the average diameter of voids is just 0.68 or 0.82 nm at 21 dpa or 33 dpa. This swelling is much smaller than that in common stainless steels, in which the void with an average diameter of 25.8 nm was found for the irradiation at 560 ^{o}C to a total dose of $3.2\cdot10^{22}$ $n\cdot cm^{-2}$, which is equivalent to the present dose of heavy ion irradiation.

The dose dependence of the positron annihilation lifetimes τ_1 and τ_2 and the variation of produced void size are shown in Fig. 4 for MSS irradiated at room temperature by 80 MeV ^{19}F ions with a damage rate of 3.9 $dpa\cdot h^{-1}$. It can be seen that τ_1 and τ_2 all increase with the increasing of irradiation dose, indicating that irradiation generates the mono- and di-vacancies, dislocations and vacancy clusters or voids. 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 obta lifetime τ_2 we arrive at that the vacancy clusters contain 8 vacancies and reach an average diameter of 0.55 nm 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, illustrating good radiation resistant property. Fig. 4 also shows a tendency that the lifetime τ_2 or vacancy cluster size approaches its saturated value at 75 dpa.

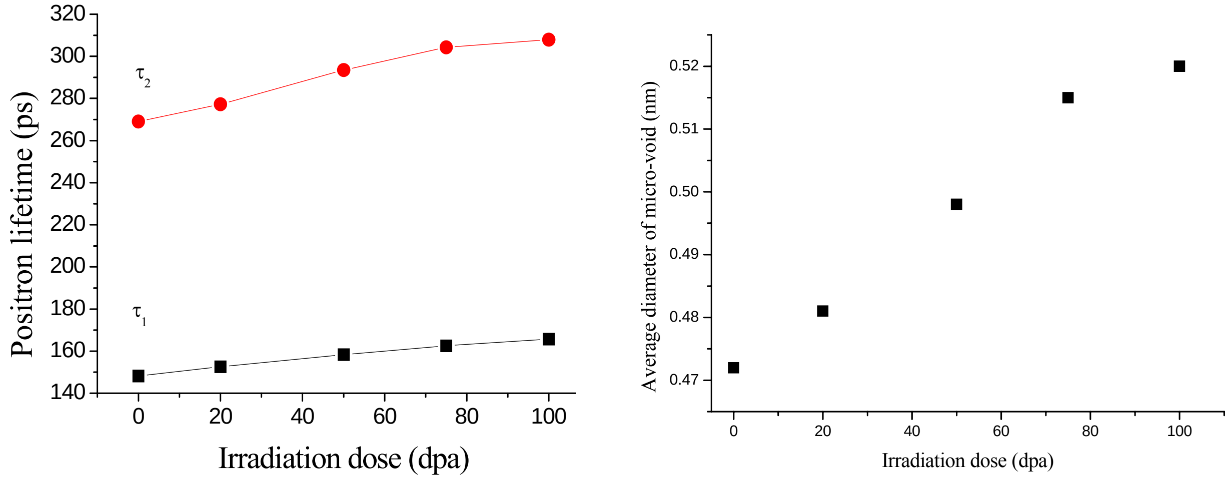

Fig.4. Dose dependence of positron lifetime τ_2 and the produced void size in MSS irradiated by 80 MeV ^{19}F ions at room temperature

3.2. Comparison of Radiation Effects in Stainless Steels and Tungsten [3]

3.2.1. Experiment

The samples used in the experiment were modified 316 austenitic stainless steel (MSS) and commercially available stainless steel (SS) and tungsten (W). SS was the18Cr-9Ni-Ti stainless steel. The purity of W was 99.9%. The samples were irradiated at room temperature by 80 MeV ^{12}C or 85 MeV ^{19}F ions from the HI-13 tandem accelerator. The irradiation doses were 2.0 and 20.0 dpa for W, 2.28 and 22.8 dpa for SS, 30.0 dpa for MSS.

3.2.2. Results and discussion

The positron annihilation lifetimes τ_1 and τ_2 and their relative intensities I_1 and I_2 were obtained for MSS, SS and W. The long lifetime τ_2 is attributed to the positrons trapped at vacancy clusters or voids. The longer the lifetime τ_2, the larger the vacancy cluster or void.

Table shows the comparison of the lifetime τ_2 in W, SS and MSS irradiated to 20, 22.8 and 30 dpa, respectively. Fig. 5 shows the deduced average diameters of the voids produced by the irradiation in MSS. SS and W. It can be seen that RD is very severe in W, sizeable in SS and very small in MSS.

Comparison of the lifetime τ_2 in MSS, SS & W

Sample	MSS	SS	W
Irradiation dose (dpa)	30.0	22.8	20.0
Lifetime τ_2 (ps)	286.0	343.9	424.1

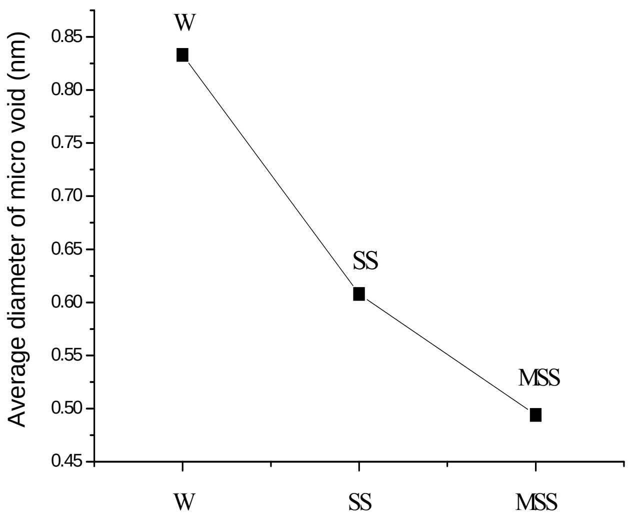

Fig. 5. Average diameter of void or vacancy cluster produced in MSS, SS and W

The irradiation induces the voids of 19-vacancy with an average diameter of 0.79 nm clusters in W, 10-vacancy clusters with a diameter of 0.57 nm in SS and 5-vacancy clusters with a diameter of 0.47 nm in MSS and their relative intensities are in turn 33, 23.8 and 19.6%. One can say that among the three samples investigated in the present experiment the modified 316L stainless steel (MSS) has the best radiation resistant property, and the radiation resistant property of the commercially available tungsten (W) is worst.

3.3. Dose dependence of RD in tungsten & tantalum

3.3.1. Experiment

W and Ta samples used in the experiment were all commercial products with a purity of better than 99.9%. They were irradiated at room temperature by 80 or 90 MeV ^{19}F ions from the HI-13 tandem accelerator in the dose range from 0 to 90 dpa. The RD created in the samples was examined by a positron annihilation lifetime technique.

3.3.2. Results and discussion

It can be seen that from the obtained lifetimes and their relative intensities that the mono- and di-vacancies, dislocation and different-size vacancy clusters (or voids) were produced by irradiation. The concentrations of the mono- and di-vacancies, dislocation and vacancy clusters and the size of the vacncy clusters increase with the increasing of the irradiation dose. Fig. 6 illustrates the size variation of produced voids or vacancy clusters with the irradiation dose for W and Ta. It shows that the void diameter rises rapidly below 20 dpa and then increases slowly with W faster than Ta. This variation with dose is much different from that for the modified 316 austenitic stainless steel (MSS) as shown in Fig.4.

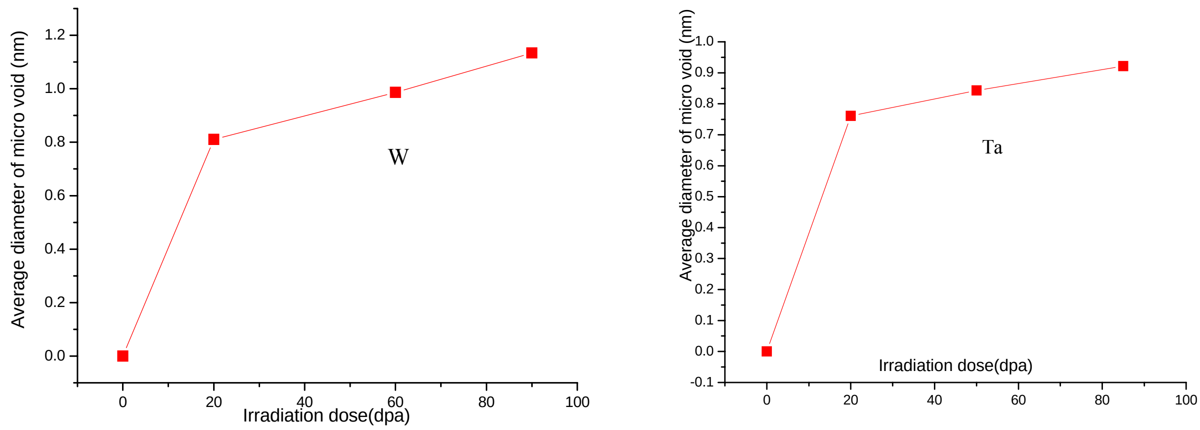

Fig. 6. Variation of average void diameter with irradiation dose in W and Ta

4. SUMMARY

The HIIS makes it possible to investigate RD induced by high dose neutron and/or proton irradiations in lab in a reasonably short time period. A verification experiment has been performed for the first time to demonstrate the reliability and validity of HIIS. A series of HIIS experiments have been performed at China Institute of Atomic Energy, which provide useful RD data of SS, W, Ta, etc. for use in ADS, ITER, etc.

Acknowledgment

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МОДЕЛИРОВАНИЕ РАДИАЦИОННЫХ ЭФФЕКТОВ, ВЫЗВАННЫ

ОБЛУЧЕНИЕМ МАТЕРИАЛОВ БОЛЬШИМИ ДОЗАМИ, С ИСПОЛЬЗОВАНИЕМ ОБЛУЧЕНИЯ ТЯЖЕЛЫМИ ИОНАМИ

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Материалы, используемые в ADS, ITER, быстром реакторе, подвержены очень высоким дозам облучения протонами и/или нейтронами. Это облучение вызывает серьёзное радиационное повреждение материалов, что приводит к разрушению этих установок или их аварии. Исследование радиационных повреждений, вызванных облучением большими дозами, является весьма актуальной и важной задачей. Однако это исследование тормозится из-за отсутствия нейтронных и протонных источников, имеющих высокие плотности потока частиц. Моделирование с использованием облучения тяжелыми ионами предоставляет эффективный путь такого исследования. Метод моделирования с использованием облучения тяжелыми ионами на основе тандемного ускорителя HI-13 применялся в Институте атомной энергии Китая для исследования радиационных повреждений, встречающихся в вышеупомянутых установках. Проверена надежность и достоверность моделирования с помощью облучения тяжелыми ионами; выполнен ряд экспериментов путем моделирования облучения тяжелыми ионами в сочетании со спектроскопией времени жизни позитронов для изучения зависимости радиационных повреждений от температуры и дозы для нержавеющих сталей, вольфрама, тантала и т.д. Представлены и обсуждаются некоторые экспериментальные результаты.

МОДЕЛЮВАННЯ РАДІАЦІЙНИХ ЕФЕКТІВ, ЗУМОВЛЕНИХ ОПРОМІНЕННЯМ МАТЕРІАЛІВ ВЕЛИКИМИ ДОЗАМИ, З ВИКОРИСТАННЯМ ОПРОМІНЕННЯ ВАЖКИМИ ІОНАМИ

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Матеріали, що використовуються в ADS, ITER, швидкому реакторі, піддані високим дозам опромінення протонами та/або нейтронами. Це опромінення викликає серйозне радіаційне пошкодження матеріалів, що призводить до руйнування цих устаткувань або їх аварії. Дослідження радіаційних пошкоджень, зумовлених опроміненням великими дозами, є досить актуальним та важливим завданням. Однак це дослідження гальмується внаслідок відсутності нейтронних та протонних джерел, що мають високі щільності потоків часток. Моделювання з використанням опромінення важкими іонами представляє ефективний шлях такого дослідження. Метод моделювання з використанням опромінення важкими іонами на основі тандемного прискорювача HI-13 застосовувався в Інституті атомної енергії Китаю для дослідження радіаційних пошкоджень, що спостерігаються у вищезазначених устаткуваннях. Перевірена надійність та достовірність моделювання за допомогою опромінення важкими іонами; виконано низку експериментів шляхом моделювання важкими іонами у поєднанні зі спектроскопією часу життя позитронів для вивчення залежності радіаційних пошкоджень від температури і дози для нержавіючих сталей, вольфраму, танталу і т.д. Представлені і обговорюються деякі експериментальні результати.