Available online at www.sciencedirect.com

SCIENCE DIRECT®

ELSEVIER

Journal of Nuclear Materials 343 (2005) 330–332

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Positron annihilation lifetime spectroscopy on heavy ion irradiated stainless steels and tungsten

Zhu Shengyun [a,*], Xu Yongjun [a], Wang Zhiqiang [a], Zheng Yongnan [a], Zhou Dongmei [a], Du Enpeng [a], Yuan Daqing [a], M. Fukuda [b], M. Mihara [b], K. Matsuta [b], T. Minamisono [b]

[a] *China Institute of Atomic Energy, P.O. Box 275-50, Beijing 102413, China*

[b] *Department of Physics, Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan*

Abstract

Radiation effects have been studied in the modified 316L stainless steel and the commercially available stainless steel and tungsten by the heavy ion irradiation simulation and positron annihilation lifetime techniques. The measured positron lifetime data show that the radiation resistant property of stainless steels is much better than that of tungsten, and the modified 316L stainless steel possesses the best radiation resistant property among them. The present experimental results demonstrate that the commercially available stainless steel is better than the commercially available tungsten for a beam window material of an ADS spallation neutron source system, and the modified 316L stainless steel is the best among the three.

1. Introduction

The accelerator driven nuclear power system (ADS) is a novel innovative idea for the sustainable development of nuclear power system [1,2]. The spallation neutron source system is one of the key parts of an ADS [1,3], which provides intense neutrons to drive a sub-critical reactor. Stainless steel and tungsten are important candidate materials for the beam window and the spallation neutron source target. High-energy and high flux protons and/or neutrons irradiate them during operation. The accumulated dose could reach a couple of hundred displacement per atom (dpa) per year. A radiation damage study of stainless steel and tungsten for use in the ADS spallation neutron source system is of great importance for their usable lifetime evaluation and the ADS safe operation.

The present work was stimulated to investigate radiation damage in the modified 316L stainless steel and the commercially available stainless steel and tungsten at irradiation doses up to 20–30 dpa and to compare their radiation resistant properties. It would take hundreds of days to reach such a high dose by proton and neutron irradiations with the use of currently available proton and neutron sources. The displacement rate of heavy ions is much higher than those of neutrons and protons [4], which makes it possible to study radiation damage at high doses in the laboratory. Therefore, the heavy ion irradiation was adopted to simulate the proton and/or neutron irradiations in the present experiment. The radiation damage produced was examined by a positron

---

[*] Corresponding author. Tel.: +86 10 69358003; fax: +86 10 69357787.
E-mail address: zhusy@iris.ciae.ac.cn (S. Zhu).

0022-3115/$ - see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2004.11.024

annihilation lifetime spectroscopy that is a very sensitive tool for investigating radiation damage on an atomic scale.

2. Experiment

The samples used in the experiment were modified 316 austenitic stainless steel (MSS) and commercially available stainless steel (SS) and tungsten (W). The size of all the samples was ∅ 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. SS was the18Cr–9Ni–Ti stainless steel. The purity of W was 99.9%. MSS was made 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%, and treated by 20% cold-working. The cold-working and Ti element-addition can greatly improve the resistance to swelling of stainless steels. All the samples used in the experiments were mechanically polished to a mirror-like surface. The samples were irradiated at room temperature by 80 MeV ^{12}C or 85 MeV ^{19}F ions from the HI-13 tandem accelerator at China Institute of Atomic Energy. In order to ensure that the irradiation was carried out at room temperature, the irradiating beam current was limited to ~0.3 μA and the samples were firmly contacted with the irradiation chamber wall of metal that was cooled by an electric fan. 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. The displacement per atom (dpa) created by the heavy ion irradiation in the sample was calculated using a TRIM program [5,6]. This program was also employed to calculate the irradiation depth. In the present experiment the radiation damage was mainly located in a ~23 μm and ~18 μm layer under the surface for stainless steel and tungsten, respectively. The radiation depth was within the positron range. In general the irradiation depth and dose increase with the increasing of ion energy. Practically, one has to make multiple considerations of the ion energy that accelerator can provide, the ion beam current, the irradiation depth and positron range and the irradiation dose etc.

The radiation damage generated in the samples was examined by a positron annihilation lifetime technique. The positron annihilation lifetime measurements were performed at room temperature by means of a conventional fast–fast coincidence positron lifetime spectrometer consisting of a pair of BaF_2 scintillation detectors, the time resolution of which is 210 ps for ^{60}Co γ rays. Two samples irradiated to the same irradiation dose were arranged as a sandwich with a 1.5 MBq ^{22}Na positron source in the center. Besides the source components, all measured positron lifetime spectra were well fitted by two lifetime components with a fitting variance of less than 1.3.

3. Results and discussion

The extracted lifetimes τ_1 and τ_2 and their relative intensities I_1 and I_2 for MSS, SS and W are shown in Fig. 1 as a function of irradiation dose.

The annihilation lifetime τ_f of free positrons is ~110 ps in stainless steel and ~105 ps in tungsten. In general the annihilation lifetimes of positrons trapped at mono- and di-vacancies are $\tau_{1v} = 1.3\tau_f$ and $\tau_{2v} = 1.5\tau_f$, respectively. The lifetime of positrons trapped at dislocations is a bit larger than τ_{2v}. Vacancy clusters or voids possess much longer annihilation lifetimes. Usually, the larger the vacancy cluster, the longer the annihilation lifetime. In the present work we assume that the short lifetime τ_1 is a weighted average of the annihilation lifetimes of free positrons and positrons trapped at the mono- and di-vacancies and dislocations, and the long lifetime τ_2 is related to the positrons trapped at the vacancy clusters or voids. It can be seen

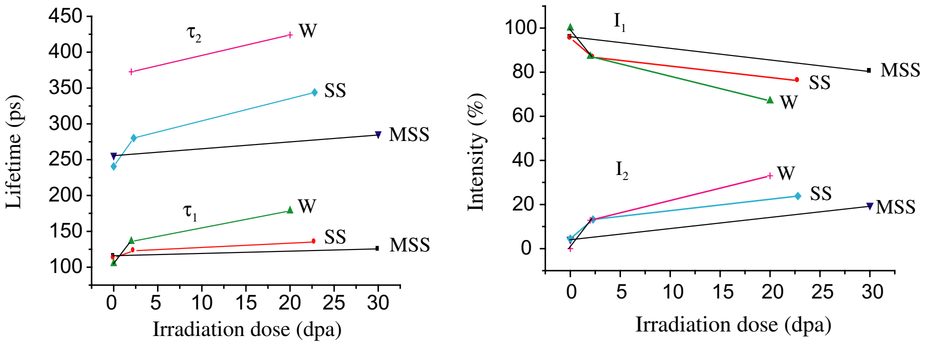

Fig. 1. Dependence of positron lifetimes (left) and their intensities (right) on irradiation dose in MSS, SS and W.

from Fig. 1 that only τ_1 was obtained for the un-irradiated W the value of which is in good accord with the free positron annihilation lifetime, and $\tau_1 = 112.9$ ps and $\tau_2 = 240.0$ ps and $\tau_1 = 115.0$ ps and $\tau_2 = 256$ ps were observed for the un-irradiated SS and MSS, respectively. The τ_1 values of 112.9 and 115.0 ps are a little larger than the free positron annihilation lifetime, indicating there are a very small amount of mono- and di-vacancies and dislocations. τ_2 is related to the small vacancy clusters, and vacancy cluster is a bit larger in MSS than in SS. The above measured data show that before irradiation W is defect-free and SS and MSS contain vacancy-clusters. MSS is treated by the cold-working which produces vacancies, vacancy clusters and dislocation in it [7]. Even after thermal annealing at 800 °C the vacancy clusters still exist [8]. $\tau_1 = 108.8$ ps and $\tau_2 = 255.7$ ps were obtained for the MSS annealed at 800 °C for about 40 min, demonstrating that the mono- and di-vacancies and dislocations were annealed away, and the vacancy clusters are not annealed.

Both τ_1 and τ_2 increase for the SS irradiated to a dose of 2.28 dpa. Their values indicate that the mono-vacancy was mainly produced and that the vacancy cluster becomes larger and its intensity increases by ~9%. In the case of W irradiated to 2.0 dpa $\tau_1 = 136$ ps reveals the production of the mono- and di-vacancies, and the rapid increase of τ_2 to 372.6 ps indicates the formation of rather large vacancy clusters.

In W irradiated to 20.0 dpa τ_1, τ_2 and I_2 arrive at the values of 178.4 ps, 424.1 ps and 33%, respectively. $\tau_1 = 178.4$ ps illustrates the generation of the tri- and quadri-vacancies besides the mono- and di-vacancies, otherwise, one cannot obtain such a large τ_1. Here, τ_1 is the average value of lifetimes of free positrons and positrons trapped at the mono-, di-, tri- and quadri-vacancies. In metals the positron lifetime of ~500 ps means a zero-density limit of electron density. $\tau_2 = 424$ ps clearly demonstrates the formation of large size voids the intensity of which is 33%. In SS irradiated to 22.8 dpa τ_1, τ_2, and I_2 are 135.1 ps, 343.9 ps and 23.8%, respectively. $\tau_1 = 135.1$ ps shows the creation of the mono- and di-vacancies. The lifetime τ_2 of 343.9 ps, the intensity of which reaches 23.8%, can be explained by the generation of the ~10-vacancy cluster [9]. For the MSS irradiated to 30 dpa τ_1 increases to 125 ps, indicating the production of the mono-vacancies, and τ_2 increases to 286 ps with an intensity of 19.6%. This value of τ_2 is corresponding to the lifetime of positrons trapped at the 5-vacancy clusters [9].

Table 1
Comparison of the lifetime τ_2 in MSS, SS and W irradiated to ~20 or 30 dpa

Sample	MSS	SS	W
Irradiation dose (dpa)	30.0	22.8	20.0
Lifetime τ_2 (ps)	286	343.9	424.1

Table 1 shows the comparison of the lifetime τ_2 in W, SS and MSS irradiated respectively to 20, 22.8 and 30 dpa. It can be seen that radiation damage is very severe in W, sizeable in SS and very small in MSS. The irradiation induces large size voids in W, 10-vacancy clusters in SS and 5-vacancy clusters in MSS and their relative intensities are in turn 33%, 23.8% and 19.6%. Therefore, from our positron lifetime data, 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. The present experimental results demonstrate that the commercially available stainless steel is better than the commercially available tungsten for a beam window material of an ADS spallation neutron source system, and the modified 316L stainless steel is the best among the three. More data are needed to further confirm this conclusion and the investigation of radiation effects for the modified 316 austenitic stainless steel and the commercially available stainless steel and tungsten irradiated by spallation neutrons is under way.

Acknowledgement

The present work was supported by the State Major Basic Research Development Program in China under Grant No. G1999022600.

References

[1] C. Rubbia et al., CERN/AT/95-44(ET), 1995.
[2] Ding Dazhao, in: Zhao Zhixiang (Ed.), Selected Works of Concept Research of Accelerator Driven Radioactive Clean Nuclear Power System, Atomic Energy Press, Beijing, 2000, p. 3, in Chinese.
[3] C.D. Bowman et al., Nucl. Instrum. and Meth. A 320 (1992) 336.
[4] K. Exel, W. Humbach, K.H. keister, et al., in: K. Bethge, H. Baumann, et al. (Eds.), Nuclear Physics Methods in Materials Research, 1980, p. 478.
[5] J.P. Biersack, L.G. Haggmark, Nucl. Instrum. and Meth. B 174 (1980) 257.
[6] J.P. Biersack, W. Eckstein, Appl. Phys. A 34 (1984) 73.
[7] Ahmat Polat, Fan Zhiguo, Luo Qi, et al., Nucl. Sci. Tech. 11 (2000) 52.
[8] Ahamat Polat, Fan Zhiguo, Luo Qi, et al., Atomic Energy Sci. Technol. 37 (2003) 18, in Chinese.
[9] M.J. Puska, R.M. Nieminen, J. Phys. F 13 (1983) 333.