Change in the Positron Annihilation Lifetime of Vacancies Containing Hydrogen Atoms in Electron-irradiated Tungsten

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The change in the positron annihilation lifetime (PAL) of vacancies before and after hydrogen charging (hydrogen pressure of 1 MPa) was determined using PAL measurements in electron-irradiated tungsten. The hydrogen atoms were captured by the vacancies that were formed due to electron irradiation during hydrogen charging. Consequently, the PAL of the vacancies decreased from 177 ps to approximately 154 ps. The simulations showed that one/each vacancy captured one or two hydrogen atoms (on average 1.8 atoms). The PAL of the vacancies increased after isothermal annealing at 573 K because the hydrogen atoms were dissociated from the vacancies. We expected one/each vacancy to capture zero or one hydrogen atom (on average 0.8 atoms) during hydrogen charging at a pressure of 1 MPa. Additional experiments under different pressures need to be conducted to further investigate this contradictory result/behavior.

KEYWORDS: Hydrogen, Vacancy, Positron annihilation, Tungsten

1. Introduction

Tungsten is a commonly used material for the plasma-facing components (for e.g. a diverter) of fusion reactors. It is used as the target material for the production of neutrons in spallation neutron sources. Tungsten is irradiated with high-energy protons or neutrons in both systems. The hydrogen and helium atoms from nuclear transmutation interact with the defects generated from irradiation. As a consequence, the gas atoms that remain in the materials [1−3] cause the mechanical properties to deteriorate, leading to swelling or embrittlement [4]. The retention of hydrogen atoms in the diverter materials is of particular significance. Thus, it is important to understand the interaction between the irradiation-induced defects and constituent gas atoms of the...
materials. In this study, we focus on detecting the interaction between the hydrogen atoms and defects using positron annihilation spectroscopy.

Positron annihilation spectroscopy is a very powerful tool that is used for the detection of vacancy-type defects. It has been reported by Troev et al. that the positron annihilation lifetime (PAL) of vacancies or vacancy clusters containing hydrogen or helium atoms decreased as compared to those not containing any gas atoms [5,6]. The change in the PAL of vacancies containing hydrogen atoms in electron-irradiated tungsten has been experimentally determined by Sato et al [7]. The PAL of single vacancies with an initial value of ~175 ps decreased to ~155 ps following hydrogen charging under a high pressure of 5.8 MPa. The PAL of single vacancies containing hydrogen atoms was also calculated using the electron density from the first principle calculation [7]. A comparison between the results from the experiments and simulations showed that one/each vacancy captured one or two hydrogen atoms (on average 1.6 atoms). In this study, we investigated the number of hydrogen atoms that were captured by the vacancies under a pressure of 1 MPa.

2. Experimental Procedure

In this study, high purity tungsten (99.95%, A.L.M.T. Corp. [8]) was used. A wire discharge machine was used to cut samples with 5 mm diameters from 0.2 mm thick sheets, which were then annealed at 1773 K for 1 h in a vacuum (< 10^{-4} Pa) for recrystallization. Vacancies were induced/created using an 8 MeV electron irradiation beam from the electron linear accelerator located at the Institute for Integrated Radiation and Nuclear Science at Kyoto University. Irradiation doses of 1.4 × 10^{22}/m^2 and 3.0 × 10^{22}/m^2 (or 1.4 × 10^{-4} dpa and 2.9 × 10^{-4} dpa, respectively) were used. The irradiation doses were calculated based on [9] with an atomic displacement cross-section of 70.4 barns and displacement threshold energy of 84 eV. Water cooling was used to maintain the irradiation temperature at 363 ± 10 K. All samples were electropolished after electron irradiation to remove the oxidation layers. High-pressure hydrogen charging was performed under a pressure of 1 MPa and temperature of 573 K for a duration of 240 h. The electron-irradiated samples were annealed at 573 K for 240 h in a vacuum prior to hydrogen charging to avoid the formation or growth of vacancy clusters during hydrogen charging. Hydrogen atoms diffused to sufficiently deep regions (>100 μm) during hydrogen charging and subsequent changes in the PAL were measured [7].

The PAL measurements were conducted at room temperature. A fast-fast coincidence system with a time resolution of 190 ps at the full width at half maximum (FWHM) was employed [10]. Na-22 was used as the positron source. A total of ~3 × 10^8 PAL spectra were collected and analyzed using the PALSfit package [11]. The amount of positron annihilation in the matrix can be quantified by the short lifetime component (τ_1). The long lifetime component (τ_2) corresponds to the size of the vacancy-type defects, while the density is given by the relative intensity of the long lifetime component (I_2). In this study, only τ_2 and I_2 are shown/considered to signify the importance of the PAL change in the vacancy-type defects. Isochronal annealing for a period of 1 h was conducted between 473 to 573 K at every step-size of 50 K. Subsequently, isothermal annealing was carried out at 573 K for a total duration of 140 h.
3. Results and Discussion

Table I shows the change in the PAL of tungsten irradiated with electrons at doses of $1.4 \times 10^{-4}$ dpa and $2.9 \times 10^{-4}$ dpa. A $\tau_2$ value of ~177 ps after annealing indicates that irradiation causes the formation of single vacancies and the annealing process at 573 K for 240 h before hydrogen charging does not lead to the formation of vacancy clusters. The $\tau_2$ value decreases after hydrogen charging as the single vacancies have been decorated with hydrogen atoms. The $\tau_2$ and $I_2$ values are almost constant until the isochronal annealing process at 573 K. There is an increase in $\tau_2$ and decrease in $I_2$ after annealing at 573 K for 10 h. This increase in $\tau_2$ is caused by the dissociation of hydrogen atoms from the vacancies. The $\tau_2$ and $I_2$ values of tungsten irradiated with electrons at doses of $1.4 \times 10^{-4}$ dpa and $2.9 \times 10^{-4}$ dpa, respectively.

| Treatment                      | $\tau_2$ (ps) | $I_2$ (%) |
|--------------------------------|---------------|-----------|
| Irradiation dose               | 1.4 $\times 10^{-4}$ dpa | 2.9 $\times 10^{-4}$ dpa |
| Before irradiation             | 110           | 110       |
| Annealing at 573 K for 240 h    | 177 (34)      | 178 (49)  |
| Hydrogen charging              | 156 (33)      | 152 (59)  |
| Isochronal annealing for 1 h    | 473 K         | 148 (44)  |
|                                | 523 K         | 153 (40)  |
|                                | 573 K         | 152 (47)  |
| Isothermal annealing at 573 K   | 10 h          | 159 (38)  |
|                                | 20 h          | 160 (39)  |
|                                | 44 h          | 169 (31)  |
|                                | 80 h          | 175 (25)  |
|                                | 140 h         | 175 (27)  |

The trapping and de-trapping reaction rate of the hydrogen atoms at the vacancies is equal under thermal equilibrium. The trapping rate is defined as the product of the hydrogen concentration, hydrogen jump frequency for trapping, and concentration of the vacant sites around the vacancy available for trapping the hydrogen atom. The de-trapping rate is defined as the product of the concentration of the vacant interstitial hydrogen sites, hydrogen jump frequency for de-trapping, and concentration of hydrogen trapped at the vacancies. Based on [7] and Sieverts’ law, we set the maximum number of hydrogen atoms (Z) that the vacancies can capture at 6, while the binding energy of hydrogen to the vacancies and hydrogen concentration were 1.19 eV and 4.6 $\times 10^{-12}$ (Pressure: 1 MPa, Temperature: 573 K), respectively. Furthermore, the jump frequencies for trapping and de-trapping were assumed to be equal. The number of
hydrogen atoms that were trapped in one vacancy was 0.8. Thus, it was expected that the value of $\tau_2$ after hydrogen charging at 1 MPa would decrease to half as seen in a previous study (5.8 MPa) [7]. However, the experimental results did not generate the expected value. Hydrogen charging at several pressures (for e.g. 0.1 MPa or 100 MPa) will be conducted in future studies to investigate the reason behind the above-mentioned discrepancy.

4. Summary/Conclusion

We determined the change in the PAL of single vacancies using high-pressure hydrogen charging and annealing in electron-irradiated tungsten. We detected the trapping and de-trapping of hydrogen atoms in vacancies using PAL measurements after hydrogen charging and isothermal annealing were performed. The number of hydrogen atoms trapped in the vacancies was estimated assuming a state of thermal equilibrium. However, the proposed estimation did not correspond to the experimental results and would require further experiments to clarify/investigate the discrepancy.

Acknowledgments

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