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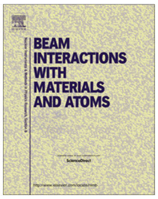

Influence of He ions irradiation on the deuterium permeation and retention behavior in the CLF-1 steel

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ABSTRACT

To evaluate the influence of He ions irradiation on the deuterium permeation and retention behavior in RAFM steels, samples made of the CLF-1 steel was irradiated with 3.5 MeV He ions. Gas driven permeation experiments were performed, and the permeability of virgin sample and pre-irradiated sample were obtained and compared. In order to characterize the effect of He ions irradiation on the deuterium retention behavior, deuterium gas exposure was carried out at 623 K, followed by thermal desorption spectra experiments. The total deuterium retention of the CLF-1 steel increased owing to He ions implantation, which could be attributed to the increase in trapping site for deuterium by the He pre-irradiation.

1. Introduction

Owing to preferable thermophysical properties and irradiation resistance, reduced activation ferritic/martensitic (RAFM) steels have been considered as the leading candidate blanket structural materials for the International Thermonuclear Experimental Reactor (ITER) Test Blanket Module (TBM) and Demonstration Fusion Plant (DEMO) [1]. Permeation and retention behavior of hydrogen isotopes, deuterium (D) and tritium (T) in RAFM steels is vital for the economy and safety of fusion reactors [2]. The high fluence 14 MeV fusion neutrons can produce various kinds of defects as well as gaseous impurity helium (He) in materials, which will influence the behavior of D/T permeation and retention in the materials [3,4]. Due to both extreme lack of 14 MeV neutron sources and troublesome activation of the exposed samples, high-energy ions have been used to simulate the irradiation effects of neutron [5,6]. Jung et al. have done comprehensive work on the effect of irradiation on permeation and diffusion behavior of RAFM steels using high-energy ions [3,7,8]. They found that D permeability and diffusion coefficient of MANET 2 and EUROFER 97 steel were influenced by irradiation.

In this work, high-energy He ions were used to produce cascade damage and to inject high concentration of He gas atoms into the steel specimens. Gas-driven permeation (GDP) and thermal desorption spectra (TDS) experiments were carried out to investigate the effects of irradiation damage on D permeation and retention behavior of a Chinese RAFM steel.

2. Experimental

2.1. Material preparation

The material chosen in the experiments is the CLF-1 steel, developed by Southwestern Institute of Physics (SWIP). Disks with dimensions of Φ 20 mm × 1 mm for the permeation experiments together with disks with dimensions of 10 × 10 × 1 mm for D_2 gas exposure and TDS experiments were cut and first mechanically polished to a mirror finish, and then electro-polished in 10 wt% $HClO_4$ alcoholic solution at ~253 K, finally, the disks were ultrasonically cleaned in acetone and alcohol. The detailed chemical

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Table 1
Detailed chemical composition of the CLF-1 steel samples [9,10].

Element	Cr	Al	W	Ta	V	Y	Zr	C	Mn	Si	Fe
CLF-1 wt%	8.5	<0.03	1.5	0.1	0.3	–	–	0.11	0.5	<0.05	Balance

composition of the CLF-1 steel is shown in Table 1. More detailed information of the CLF-1 steel can be found in Refs. [9,10].

2.2. He ions implantation experiments

He ions implantation experiments were done in the State Key Laboratory of Nuclear Physics and Technology, Peking University using an electrostatic accelerator. The He^+ ions energy is 3.5 MeV, and samples with three irradiation fluences, 6×10^{17}, 6×10^{18}, $3 \times 10^{19}\ m^{-2}$ were obtained. The temperature during ion implantation was nearly R.T. The He atom distribution and DPA distribution was obtained using SRIM 2008 full-cascade simulation code [11] in the “quick Kinchin–Pease calculation” mode with a threshold displacement energy of 40 eV [12]. The implanted He atoms and damage distribution of the $3 \times 10^{19}\ m^{-2}$ sample is shown in Fig. 1. The peak dpa is at a depth of 65,000 A for all three samples. The irradiation information of samples in the experiments is shown in Table. 2.

2.3. Deuterium GDP experiments

GDP experiments were done for both irradiated and virgin samples in the device constructed in Institute of Plasma Physics, Chinese Academy of Sciences (ASIPP). Detailed description of the GDP device and the experiment procedure could be found in our previous papers [13,14].

For this study, the experimental temperature ranged from 623 to 823 K, which was set on account of the service temperature range for RAFM steels as the structural materials in the blanket. To take the thermal induced recovery of irradiation defects into account, the GDP experiments were performed from low temperature to high temperature and then back to low temperature again. The upstream D_2 gas pressures were set to three different values ranging from 10^3 Pa to 10^5 Pa for each temperature.

2.4. Deuterium gas exposure and TDS experiments

To investigate the bulk D retention behavior in the irradiated CLF-1 steel, samples with a He^+ ion implantation fluence of $3 \times 10^{19}\ m^{-2}$ and virgin samples were exposed to D_2 gas with a pressure of 8×10^4 Pa at 623 K for 4 h. After exposure, all samples were kept in D_2 gas and cooled with furnace to R.T. TDS experiments were followed. Samples were heated in an infrared furnace up to 1273 K with a heating rate of 1 K/s. Desorption signals of mass 4 was monitored with a quadrupole mass spectrometer, which was calibrated with a D_2 standard leak.

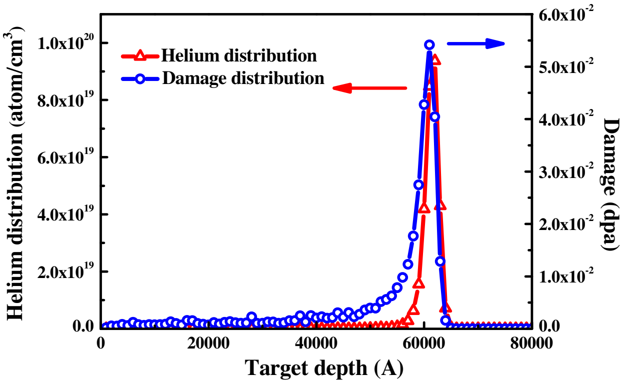

Fig. 1. The implanted He atoms and damage distribution of the $3 \times 10^{19}\ m^{-2}$ sample calculated by SRIM 2008 full-cascade simulation code in the “quick Kinchin–Pease calculation” mode with a threshold displacement energy of 40 eV.

Table 2
The irradiation information of samples in the experiments.

Sample No.	Implanted fluence	Peak dpa value	Peak He content
1	$6 \times 10^{17}\ m^{-2}$	0.001	$1.9 \times 10^{18}\ m^{-3}$
2	$6 \times 10^{18}\ m^{-2}$	0.01	$1.9 \times 10^{19}\ m^{-3}$
3	$3 \times 10^{19}\ m^{-2}$	0.05	$9.5 \times 10^{19}\ m^{-3}$

As all the samples underwent the same preparation, D_2 gas exposure and TDS procedure in one treatment, the difference in TDS measurements should result from different He implantation doses.

3. Results and discussion

3.1. Deuterium gas driven permeation

Shown in Fig. 2 are the GDP permeation fluxes as a function of the square root of the upstream D_2 pressure for the virgin CLF-1 steel sample. A linear relation between the steady GDP flux and the square root of upstream D_2 pressure at all the temperatures examined, which reflects the D GDP process through the CLF-1 steel in the temperature range is controlled by a volume diffusion process [15]. As the steady state permeation flux (J_∞) is related to the square root of driving gas pressure ($p^{1/2}$) through:

$$J_\infty = \frac{P}{d} p^{1/2}, \qquad (1)$$

where P represents the permeability of D in material; d is the thickness of the sample [16,17], the temperature dependence of measured permeability (mol/m/s/Pa$^{1/2}$) of D in CLF-1 steel can be obtained:

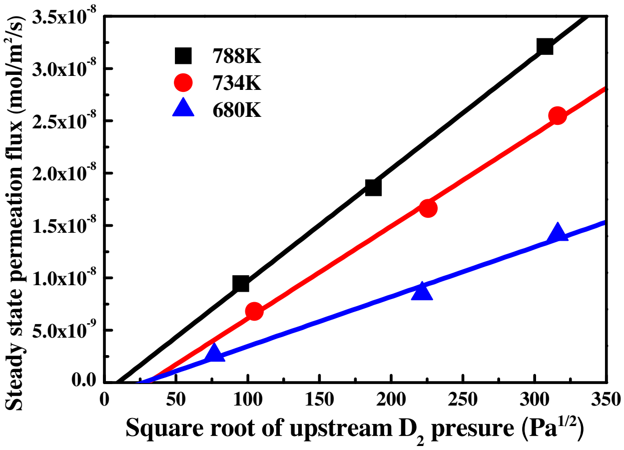

Fig. 2. GDP permeation fluxes as a function of the upstream D_2 pressure for the virgin CLF-1 steel sample.

$$P_{CLF\text{-}1} = 6.36 \times 10^{-8} \exp\left(\frac{-0.43[\mathrm{eV}]}{kT}\right). \tag{2}$$

Diffusion coefficients can be acquired using the time-lag method [17,18], the diffusion coefficient (m^2/s) of D in virgin CLF-1 steel can be expressed by:

$$D_{CLF\text{-}1} = 3.71 \times 10^{-7} \exp\left(\frac{-0.23[\mathrm{eV}]}{kT}\right). \tag{3}$$

The permeability and diffusion coefficient of CLF-1 steel are close to the literature data for Eurofer 97 [19] and F82H steel [20].

Temperature dependence of permeability and diffusion coefficient of D in CLF-1 steel irradiated by 3.5 MeV He^+ ions with fluences 6×10^{17}, 6×10^{18}, 3×10^{19} m^{-2} compared with virgin CLF-1 steel is shown in Figs. 3 and 4, respectively. No significant difference was detected in all samples except the one irradiated with the highest dose, whose permeability together with diffusion coefficient was decreased by irradiation at low temperature. As most of irradiation damages are possible trapping sites for D, D will fill all the trapping sites after steady-state permeation. The D diffusion will be refrained by these trapping sites, leading to the reduction of D permeation [21]. However, after the experiments at high temperature, the permeability and diffusion coefficient went back to normal, which could be explained by the recovery of irradiation-induced defects at high temperature [22,23].

3.2. Thermal desorption spectra experiments

Fig. 5 shows the TDS spectra of mass 4 for samples after D_2 gas exposure at 623 K for 4 h with a pressure of 8×10^4 Pa. The samples include virgin CLF-1 steel and sample irradiated by 3.5 MeV He^+ ions with a fluence of 3×10^{19} m^{-2}. As no mass 4 peak was observed in TDS spectra from room temperature to 1273 K for samples irradiated by He^+ ions with a fluence of 3×10^{19} m^{-2} without D_2 gas exposure, it can be concluded that the mass 4 signal monitored by the quadrupole mass spectrometer is from D_2. For the virgin sample, one main desorption at 484 K was observed. The major D-trapping sites in the virgin 9Cr RAFM steels were reported to be vacancies, dislocations, or certain types of impurities [24]. The main desorption peak of the irradiated sample shifted to higher temperature slightly, which was also seen in previous investigation of SSNS (Swiss Spallation Neutron Source) and He ions irradiation effect on steels [25,26]. A new desorption peak at around 570 K can be found. There are two possible reasons for the formation of new desorption peak, (i) a new trap is being created, (ii) the number density of a particular defect present in the virgin material is being enhanced. In this case, we believe the former reason is more suitable because the implantation of He ions will inevitably induce formation of new traps for hydrogen isotopes [27]. It should be noted that the amounts of D retained in the irradiated sample was higher than that in the virgin one. As mentioned above, the difference in TDS measurements should result from the difference between samples, in this case, He implantation. The higher D inventory in the irradiated sample may result from the increase in trapping site for D by the He pre-irradiation [28].

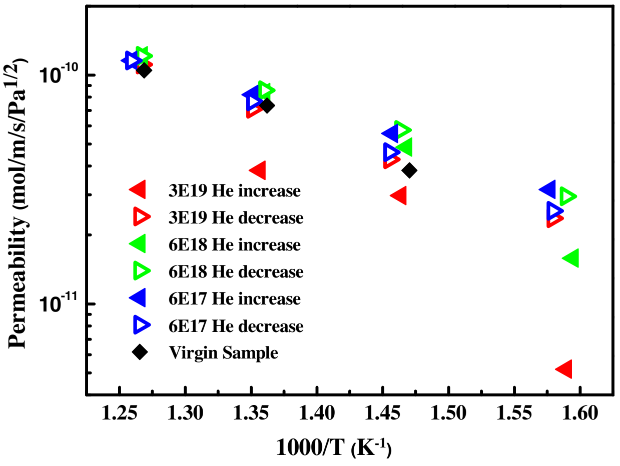

Fig. 3. Temperature dependence of permeability of D in the CLF-1 steel irradiated by 3.5 MeV He^+ ions with fluences 6×10^{17}, 6×10^{18}, 3×10^{19} m^{-2} compared with the virgin CLF-1 steel.

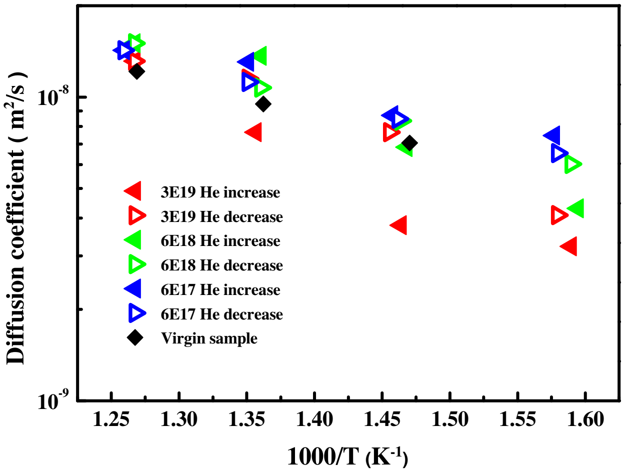

Fig. 4. Temperature dependence of diffusion coefficent of D in the CLF-1 steel irradiated by 3.5 MeV He^+ ions with fluences 6×10^{17}, 6×10^{18}, 3×10^{19} m^{-2} compared with the virgin CLF-1 steel.

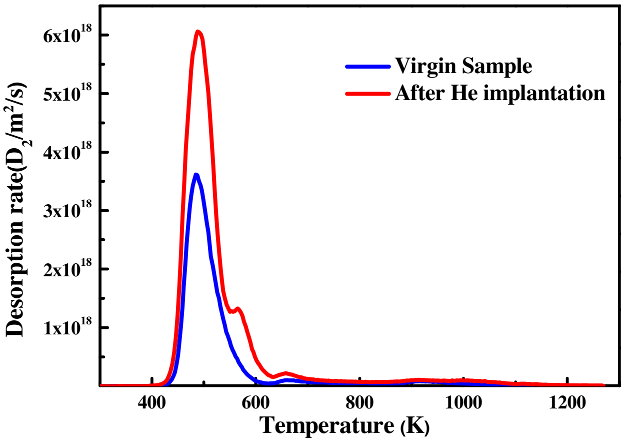

Fig. 5. TDS spectra of D_2 for samples after D_2 gas exposure at 623 K for 4 h with a pressure of 8×10^4 Pa, the samples include virgin CLF-1 steel and sample irradiated by 3.5 MeV He^+ ions with a fluence of 3×10^{19} m^{-2}. The rate of temperature ramp was 1 K/s during TDS experiments.

4. Conclusions

The focus of this work is on understanding the influence of He ions irradiation on the D permeation and retention behavior of CLF-1 steel. He ions implantation experiments were performed using 3.5 MeV He^+ ion beam. GDP experiments of virgin CLF-1 steel sample and pre-irradiated CLF-1 steel samples were performed, the permeability and diffusion coefficient of virgin CLF-1 steel were given and compared with the pre-irradiated samples. The D permeability and diffusion coefficient of the sample with a He implanted fluence of $3 \times 10^{19}\ \mathrm{m}^{-2}$ were slightly decreased at low temperature. After the experiments at high temperature, the permeability and diffusion coefficient went back to normal. To characterize the influence of He pre-irradiation on the D retention behavior of the CLF-1 steel, virgin sample and the pre-irradiated CLF-1 steel sample underwent D_2 gas exposure and TDS procedure in one treatment. The total D inventory of the sample with a He implanted fluence of $3 \times 10^{19}\ \mathrm{m}^{-2}$ was much higher than the virgin one.

The presented results suggest that the He implanted may slightly decrease the D permeability, diffusion coefficient and increase the total D retention of the CLF-1 steel. From the point of view of applying RAFM steels as the structure material in the future devices, it is very important and necessary to further investigate the irradiation effect on the hydrogen isotope transportation behavior. In this article, the investigation of the influence mechanism of irradiation damages on hydrogen isotope transportation is preliminary, more characterization of the irradiation defects will be done in the future. Meantime, it should be noted that the fluence of He implanted in the experiments is too small to give a fair conclusion on the trend of irradiation effect on D permeation behavior, more work will be done in the future.

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