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Deuterium thermal desorption from vacancy clusters in tungsten

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ABSTRACT

Deuterium interaction with vacancy clusters in tungsten was studied by means of thermal desorption spectroscopy (TDS). A recrystallized W foil was used as a sample, and the vacancy clusters were formed in the bulk by irradiation with 10 keV/D ions to the fluence of 3×10^{19} D/m^2 and subsequent annealing at the temperature of 800 K. Then the sample was loaded with deuterium (0.67 keV/D ions with a fluence of 1×10^{19} D/m^2), and TDS measurements with varying heating rates β in the range of 0.25–4 K/s were performed. The high temperature peak with the maximum at around 700 K was attributed to deuterium desorption from vacancy clusters and the detrapping energy for this type of defects was determined from the slope of the Arrhenius-like plot $\ln(\beta/T_m^2)$ versus $1/T_m$, where T_m is the peak position. The detrapping energy calculated this way is 2.10 ± 0.02 eV.

1. Introduction

The investigation of hydrogen (H) isotopes behavior in materials is of great interest for development of fusion reactors. Hydrogen isotopes retention in plasma-facing components (PFC) and permeation into the coolant system must be controlled from the safety point of view. Dynamics of hydrogen desorption from PFC can also seriously influence plasma operation. Tungsten (W) will be used as a plasma-facing material for the divertor region of ITER, and its use in future fusion devices is also very likely.

Tungsten has a very low solubility for hydrogen [1], so the retention of H isotopes in W is mainly determined by defect structure of the material [2,3]. Some intrinsic defects, such as dislocations, grain boundaries and impurities, are always present in the material. The H binding energy with these kinds of defects is relatively low [4], while much higher energy values correspond to vacancies, vacancy clusters and inner voids [5–7]. These defects can be effectively formed in plasma-facing materials (PFM) of fusion devices due to irradiation by ions or neutrons. Moreover, in the case of neutrons, radiation defects are formed in the whole bulk of PFM and can significantly increase the T accumulation.

Although H isotope retention in various W grades under different conditions have been studied quite extensively, most of quantitative characteristics of H–W interaction are not well known yet. For example, the value of H detrapping energy from a single vacancy in W varies among different researchers in the wide range of 1.3–1.6 eV [5,6,8], and from vacancy cluster – in the range of 1.7–2.2 eV [5–7,9–11].

The method of thermal desorption spectroscopy (TDS) is often used for investigation of hydrogen-defect interaction. The most common technique of determination of H detrapping energy from defects is based on fitting simulated thermal desorption spectra to experimental ones using numerical diffusion-trapping codes [5–6,12–14]. A large uncertainty in determination of characteristics of defects in such approach is given by dependence of TDS spectrum simulation on many input parameters, such as characteristic frequencies for trapping or detrapping, depth distribution of traps and trapped H. The necessity to consider all of these parameters in models leads to the result that one experimental spectrum can be fitted by using several combinations of different parameters.

However, it is possible to determine the detrapping energy from the slope of the Arrhenius-like plot $\ln(\beta/T_m^2)$ versus $1/T_m$, where β is the heating rate, T_m is the temperature corresponding to the TDS peak of interest, in a series of experiments with different heating rates β without knowing other parameters (the so-called Kissinger method [15]). This technique is applicable in the case of a high recombination rate at the surface, i.e. when the recombination at the surface does not influence the hydrogen release from the material [16].

In our recent work [17], the method described above was successfully applied for determination of the deuterium (D) detrapping energy from the single vacancies in W. In that work, the radiation damage was produced by 10 keV D^+ ions with a

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low fluence, which produced mainly point defects, i.e. Frenkel pairs – single vacancies and interstitial atoms, within the sample [18]. At elevated temperatures vacancies become mobile and start to agglomerate in vacancy clusters. According to results of positron annihilation measurements by Eleveld et al. [19], this process takes place at around 600 K. It was also confirmed by our recent experiments with different annealing temperatures [20]. The transformation of the TDS spectra and appearance of the peaks corresponding to the D release from vacancy clusters was observed at annealing temperatures higher than 550 K. Therefore, ion irradiation with subsequent heating can be used to produce vacancy clusters within the sample.

This work is devoted to the determination of the D detrapping energy from vacancy clusters in W from the shift of the desorption maximum in a series of TDS measurements of identical samples performed with different heating rates. This method doesn't include any fitting procedure, and, thus, it is independent of free parameters.

2. Experimental

The experiments were carried out in MEDION ion-beam facility (NRNU MEPhI, Moscow). A detailed description of the experimental setup is given in [21]. The duoplasmatron-type ion source is used for generation of ions with energies in the range of 0.5–10 keV/D. After extraction from the source, the ion beam goes through a lens system, a separating magnet, and then is directed onto the sample through a 3 mm diaphragm in order to reduce the spatial beam flux variations.

The target chamber is separated from the ion source by a differential pumping stage and has a base pressure below 5×10^{-9} mbar which increases to about 10^{-8} mbar during the ion irradiation. Release of gases from the sample is monitored by a quadrupole mass-spectrometer (QMS), which is installed in the target chamber. This allows performing TDS measurements just after irradiation without air exposure. A QMS signal for mass 4 (D_2) is calibrated using a helium leak with a known leak rate after every TDS measurement. The relative D_2–He sensitivity of QMS was determined previously and was assumed to be stable (details are given in [21]).

The sample is mounted on two water cooled current feedthroughs. It can be resistively heated by DC current up to 1800 K during TDS measurements with the linear heating ramp in the range of 0.25–4 K/s. The heating rate deviation at the temperatures of interest (from 400 to 1000 K) did not exceed 2%. A higher deviation is only at the temperatures below 400 K. Temperature was measured by a W-Re thermocouple spot-welded to the sample close to the beam strike area [21].

A polycrystalline 15×50 mm^2 W foil with thickness of 25 μm and the purity of 99.97 wt.% (Plansee AG, Austria) was used as a sample. It was annealed at 1800 K for 30 min before the start of the experimental series in order to minimize the amount of intrinsic defects. Since one sample was used for all experimental series, it was annealed at the temperature of 1700 K for 30 min before each experiment in order to remove radiation damage caused by previous irradiation between experiments [17,22]. This procedure led to the full annealing of radiation damage in the sample after its multiple use, and all the experiments showed a good reproducibility of the experimental results.

The vacancies were first introduced in the sample by irradiation with 10 keV D^+ ions to the fluence of 3×10^{19} D/m^2. The energy of the ions used was high enough to produce displacement damage in the sample and such low fluence resulted in mainly point defects [17]. The irradiated sample was then annealed at high temperature in the range of 550–800 K for 5 min in order to remove D from the sample. Annealing at temperatures higher than 550 K also allowed agglomerating of vacancies in vacancy clusters [20].

The irradiated sample was then implanted by a 2 keV D_3^+ (0.67 keV/D) ion beam to the fluence of 1×10^{19} D/m^2. This irradiation, as well as high-energy damaging one, was performed at normal ion incidence and at room temperature. The ion flux on the sample was in the range of $3–6 \times 10^{16}$ D/m^2 s. The value of 0.67 keV/D is below the threshold of the formation for the displacement damage in W [2], so it can be expected that no new damage was produced during this irradiation, and only the defects existing in the sample will be decorated with D. As was said before, according to [18], such experimental procedure allows obtaining a TDS spectra with prominent high temperature desorption peaks. Three hours after the implantation, when signals of all monitored masses returned to their initial values before the implantation, the TDS measurements were performed with varying heating rates in the range of 0.25–4 K/s.

All the experiments were performed in UHV conditions without any contact with air, so it can be assumed, that the condition of high H–H recombination rate at the surface was satisfied. This fact ensures the applicability of the method of determination of detrapping energies described in Section 1. All the time intervals between the experimental stages were fixed in all experiments in order to exclude the possible influence of partial deuterium release.

3. Results and discussion

The comparison of thermal desorption spectra obtained in a series of experiments performed with different annealing temperatures in the range of 550–800 K is shown in Fig. 1. In the case of annealing at 550 K and 600 K one can see two well resolved desorption peaks in the TDS spectrum. The first one, at around 400 K, is suggested to be composed of several more narrow peaks [21], and can be attributed to the D release from the defects with low binding energy (grain boundaries, dislocations) [4,22]. Though the experimental procedure (the ions energies, fluencies, the lag time between irradiation and TDS etc.) was strictly controlled in all experiments, the first peak didn't show a good reproducibility. The position, shape and amplitude of the first peak varied from one experiment to another performed in identical conditions. The origin of such variations is not completely clear. Partly, this is

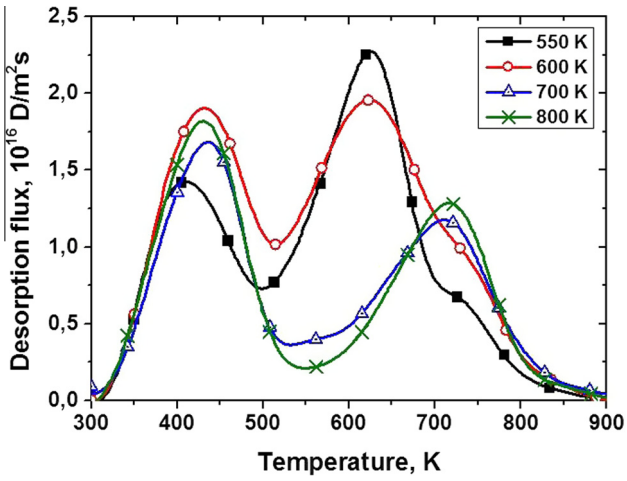

Fig. 1. The comparison of thermal desorption spectra of D_2 molecules from recrystallized W irradiated by 10 keV/D ions to the fluence of 3×10^{19} D/m^2, subsequently annealed at different temperatures of 550 K, 600 K, 700 K and 800 K, and then implanted with 0.67 keV/D ions to the fluence of 1×10^{19} D/m^2. The heating rate was 2 K/s.