HYDROGEN AND DEUTERIUM STORAGE IN TUNGSTEN WHEN IRRADIATION WITH PLASMA BEAM

Bauyrzhan RAKHADILOV [1], Mazhyn SKAKOV [2], Arman MINIYZOV [2], Aidar KENESBEKOV [1]

[1]*Sarsen Amanzholov East Kazakhstan state university, Ust-Kamenogorsk, Republic of Kazakhstan, rakhadilovb@mail.ru*

[2]*Institute of Atomic Energy of NNC RK, Kurchatov, Republic of Kazakhstan, skakovmk@mail.ru*

Abstract

The paper devotes to study of hydrogen and deuterium storage and research of capturing hydrogen and deuterium in tungsten samples irradiated with plasma beam. The paper shows that some changes occur in the surface like relief propagation caused by heterogeneous surface etching after irradiation with hydrogen plasma. Ratio of change in relief and structure of irradiated samples' surface layer herewith depends on the irradiation temperature. Hydrogen and deuterium storage in tungsten have been studied while their irradiation with hydrogen and deuterium plasma. Thermo-desorption analysis of tungsten samples irradiated with hydrogen and deuterium plasma at 1000°C degrees showed that tungsten surface became saturated with deuterium and did not become saturated with hydrogen. The data obtained by emission spectrometry and thermo-desorption spectrometry verified, that the basic share of captured deuterium is accumulated at the depth of up to 7 μm.

Keywords: Tungsten, accumulation, hydrogen, deuterium, plasma, irradiation

1. INTRODUCTION

Tungsten is thought of as a priority material to use it in diverter region of ITER reactor owing its high melt temperature and high threshold energy of physical sputtering. In addition, application of tungsten in fusion neutron source and DEMO reactor has alsobeen considered [1-4]. Today, one of the main requirements imposed for the materials contacting with fusion plasma, is reducing of tritium concentration in those materials [5,6]. The problem of storage and retention of hydrogen isotopes in metals and alloys is a very complicated and multi-side degradation process takes place in nuclear reactor materials that needs to be systematically studied with view of today's achievements in reactor material science. Therefore, study of H isotopes' storage in tungsten materials affected by hydrogen and deuterium plasma is of big interest to measure radioactive tritium concentration in reactor elements when operation of fusion facility. In view of the above, the given paper is focused on study of deuterium storage in tungsten materials after their irradiation with hydrogen and deuterium plasma.

2. MATERIAL AND RESEARCH METHOD

According to the problem formulated, WP tungsten (straight pure tungsten) was chosen as test object. Cylindrical samples of 5 mm high with dia of 10 mm were cut out by EDM machine. Then they were ground and polished. Tungsten samples were irradiated by H plasma and D plasma with ion energy of 2 keV for 180 minutes. Target temperature was varied herewith. In the course of irradiation the in-chamber pressure was $2 \cdot 10^{-3}$ tor.

The samples were irradiated with H plasma in beam-plasma discharge machine which simulates plasma-surface interactions of KTM Tokamak. The beam-plasma discharge machine (BPDM) was designed to support

creation and operation of Kazakhstan's Material Testing Tokamak (KTM) to test small-scale samples of KTM materials and equipment [7,8].

OLIMPUS BX41M Optical microscope and JSM-6390 Scanning-electron microscope were used to examine W samples microstructure before and after irradiation. To identify W microstructure, there has been applied chemical etching in solution containing 50 per cent of fluohydric acid and 50 per cent of nitric acid.

Element analysis of irradiated tungsten surface layer was conducted by opto-emission spectrometry using Profiler HR glow-discharge spectrometer.GD-Profiler emission glow-discharge spectrometer is designed for determination of elements' mass fraction in solid samples with pre-polished surface.

Quantity of deuterium and hydrogen stayed in tungsten after irradiation was measured by thermal desorption spectrometry (TDS) in experimental facility VIKA.This facility enables conducting experiments on studying gas release from different materials by temperature-programmed desorption within the temperature range from 390 Kto 1750 Kwith mass-spectrometry recording of gases released. Research technique was as follows: cut-out sample of irradiated tungsten was de-oiled and loaded into the crucible of VIKA working chamber [9]. Afterwards the walls of working chamber were degasified at 473 K-500 K for 2.5 hours while continuous pumping-out with vacuum Penning-type pump NORD-250 and turbomolecular pump EXT 75DX. Then, chamber walls were cooled down up to room temperature followed by the experiment on linear heating of tested sample up to the temperature of 1750 K under heating velocity of 15 K/min while continuous pumping-out of gases released from the working chamber and registration of partial gas in chamber. Release ofH_2andD_2molecules was measured with QMSRGA-100. In the experiments, time dependences of changes in partial gas pressure were obtained inside of vacuum chamber as a result of gas release from tungsten samples while linear heating of tested samples at the temperature from 390 Kto 1500 K. Before conducting TDS-experiments with tested tungsten samples, one experiment on gas release from empty crucible was performed with heating of the crucible up to 1750 Kwith heating velocity of 15 K/min and continuous pumping-out of VIKA vacuum chamber and mass-spectrometry registration of partial pressure changes inside of the working chamber. **Figure 1**shows diagram of all-metal high-vacuum water-cooled working chamber designed for TDS-experiments with small samples. The chamber is equipped with a heater as tantalum plate with fixed tantalum crucible loaded with tested sample. Sample temperature is recorded by two thermocouples entered the chamber through the pressure-sealed connector and fixed on the crucible.

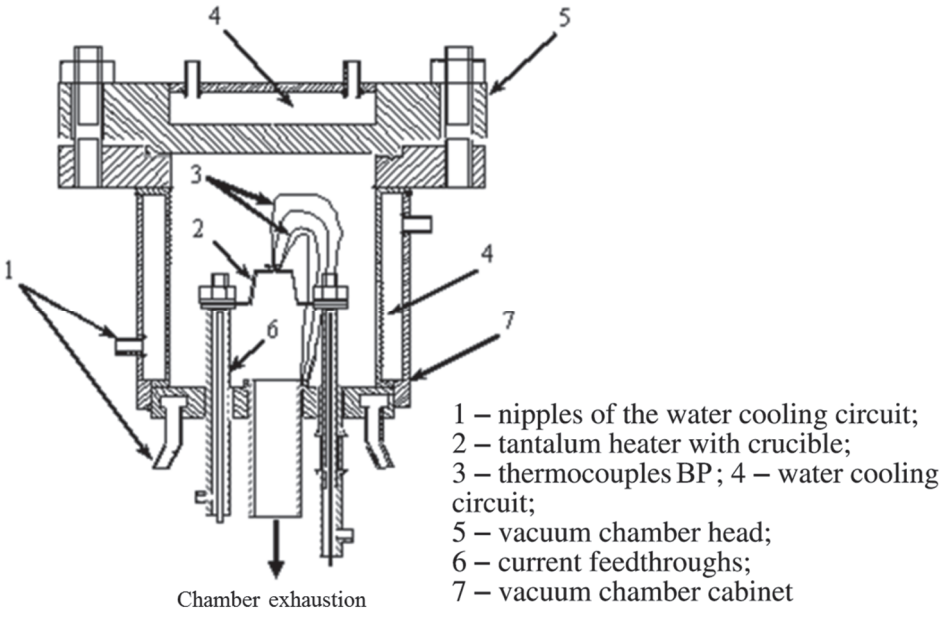

Figure 1 Vacuum working chamber of VIKA facility

3. RESULTS AND DISCUSSIONS

To obtain such a defect structure, tungsten samples were irradiated with hydrogen plasma at 700 °C, 1000 °C and 1500 °C degrees. **Figure 2** shows microstructure of tungsten surface irradiated with hydrogen plasma at 700 °C, 1000 °C and 1500 °C degrees. As shown from the **Figure2**, when irradiation at T = 700 °C degrees there are some changes in the surface like relief propagationcaused by heterogeneous surface etching. This relief consists of chaotically located claws and caves. Formation of relief propagation on the tungsten surface due to different erosions of near zones while plasma irradiation is connected with the fact, that diversely orientable surface crystals are characterized with different sputtering yields. Tungsten samples irradiated at T = 1000 °C and 1500 °C have lower degree of relief propagation in comparison with that which was irradiated at T = 700 °C. However, some small cracks are shown in these samples and the cracks become bigger and more when temperature rising. It was found that the strongest failure occurs after irradiation of samples with hydrogen and deuterium plasma at T = 1000 °C and T = 1500 °C degrees.

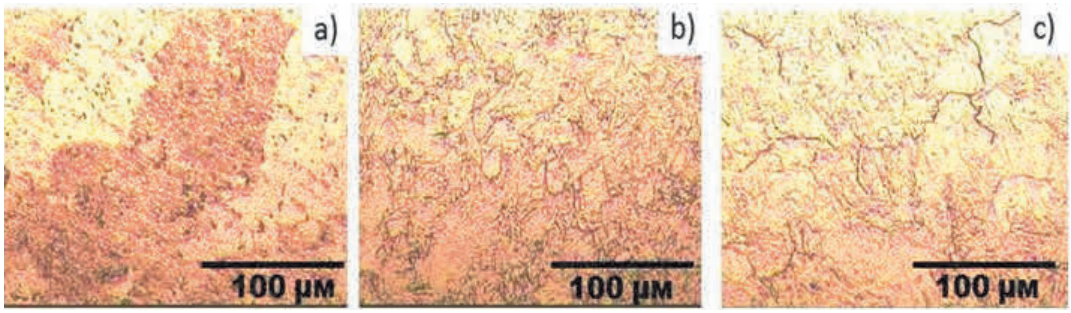

Figure 2 Microstructure of tungsten surface irradiated with hydrogen plasma at T = 700 °C (a), T = 1000 °C (b) and T = 1500 °C (c)

The permeability of tungsten upon contact with gaseous hydrogen has been studied relatively widely. However, the permeability data on irradiation with hydrogen ions are limited by the temperature interval from 610 K to 823 K. The question of the permeability of tungsten when irradiated with ions of hydrogen isotopes at temperatures above 973 K has not been studied. In [10] it is reported that the diffusion coefficient of deuterium in tungsten is much smaller than the values obtained for tungsten saturated with protium upon contact of the surface with gaseous protium. This means that in the temperature range 823 K -1023 K there is trap hydrogen that slows down the transfer process. Apparently, deuterium is captured on vacancy-type defects created in the process of ion irradiation due to the compressive stress fields arising from the supersaturation of the near-surface layer of tungsten by the deuterium atoms being introduced. In this connection there has been studied capturing of hydrogen isotope in these ion-induced and thermal-induced defects of the samples irradiated with hydrogen and deuterium plasma at 1000 °C degrees.

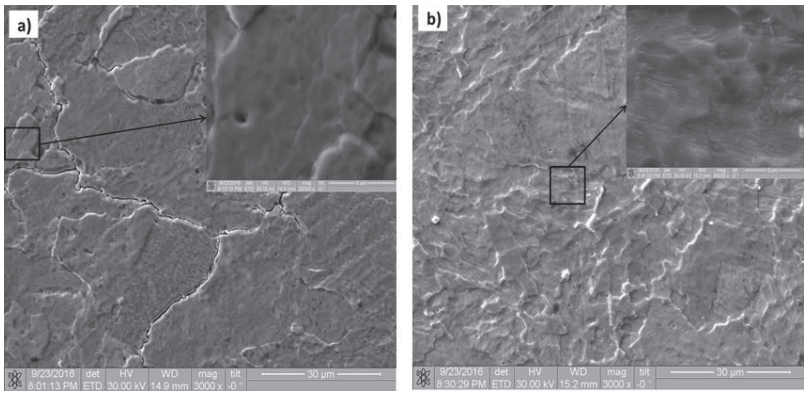

Figure 3 REM-image of tungsten surface irradiated with hydrogen(a) and deuterium (b) plasma at T = 1000 °C

Figure 3 illustrates REM-images of tungsten surface irradiated with hydrogen and deuterium plasma at T = 1000 °C. Topography of irradiated tungsten surface testifies its severe erosion. Tungsten-hydrogen plasma interaction resulted in etching of tungsten surface, development of grain boundaries and their orientation along with formation of fine cracks.

TDS was used to research gas accumulation in tungsten samples while irradiation. The purpose of TDS experiments was to obtain time dependences of changes in partial gas pressures in VIKA vacuum chamber as a result of gas release from tungsten samples while linear heating of tested samples.

Figure 4a shows temperature dependences of H_2 (amu 2) release while heating of empty crucible, sample W-H-1 and sample W-D-1 at the velocity of 15 K/min. making analysis of H_2 gas release diagram shown in **Figure 5a**, it can be noted that there is no visible hydrogen release while heating of tested samples; moreover, integral background release from the crucible exceeds in size with integral value for release of those gases in the experiments with the samples. In this case, recording of H_2, release from samples in TDS experiments was failed. It might be because of high irradiation temperature that exceeds the temperature of the crucible when maximum hydrogen release takes place.

Figure 4b shows temperature dependences of deuterium gas release. According to the **Figure 4**, D_2 gas release diagram (amu4) has explicit peak corresponding to D_2release from the sample irradiated with deuterium plasma at T_{sample} = 1223 K. Tungsten sample temperature during which deuterium is at most released, is close to the temperature of irradiated sample.

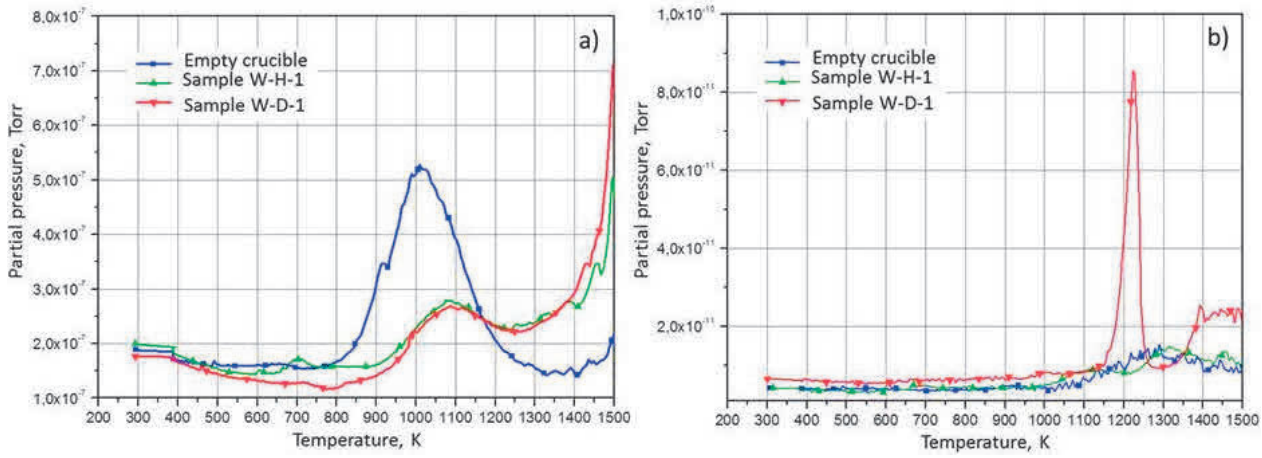

Figure 4 Dependence of change in partial hydrogen pressure (amu 2) (a) and D_2 (amu 4) (b)on the crucible (sample) temperature in VIKA working chamber

Table 1 Number of deuterium released from empty crucible and tested samples

	Empty crucible	Sample W-H-1	Sample W-D-1
Quantity of deuterium released to T = 1500K, mol	$2.84 \cdot 10^{-11}$	$3.28 \cdot 10^{-11}$	$6.93 \cdot 10^{-11}$
Quantity of deuterium released to T = 1500 K minus background resulting from crucible heating, mol	-	$4.42 \cdot 10^{-12}$	$4.09 \cdot 10^{-11}$
Quantity of deuterium released to T = 1500 (referred to sample mass)	-	$9.04 \cdot 10^{-12}$	$8.95 \cdot 10^{-11}$

In this way, for tungsten sample irradiated with hydrogen plasma, recording of visible hydrogen release failed due to high background resulting from hydrogen release from the crucible and structural elements of vacuum chamber while linear heating. Integral release of hydrogen from empty crucible was $8.8016 \cdot 10^{-7}$ mol; integral hydrogen release from tested sample irradiated with hydrogen plasma was $6.3142 \cdot 10^{-7}$ mol. For tungsten sample irradiated with deuterium plasma there is a peak of deuterium release (amu 4) within the range of

tested sample temperatures from 1150 K to 1275 K. Total number of deuterium released of tested sample was equal to $4.09 \cdot 10^{-11}$ mol. Deuterium release peak was like classical diffusion peak which as a whole is typical for deuterium diffusion activation energy in the sample from 130 to 200 kJ/mol. **Table 1** provides Number of released deuterium calculated from obtained TDS dependences

Figure 5 demonstrates results obtained in Optical-emission spectrometer. Hydrogen accumulated on the surface layer up to 7 μm deep is clearly seen. In samples herewith, irradiated with deuterium plasma, accumulation of carbon can be observed. Hydrogen distribution diagram shows that there is a clearly visible peak related to hydrogen near-surface accumulation at the depth of 0.2 - 0.3 μm. Then the intensity is declined. Tungsten sample irradiated with deuterium plasma, intensity peak is observed at the depth of 2 μm. Taking into account TDS analysis it can be concluded, that observed hydrogen peak corresponds to deuterium peak since glow-discharge spectrometer is not capable to determine isotopes.

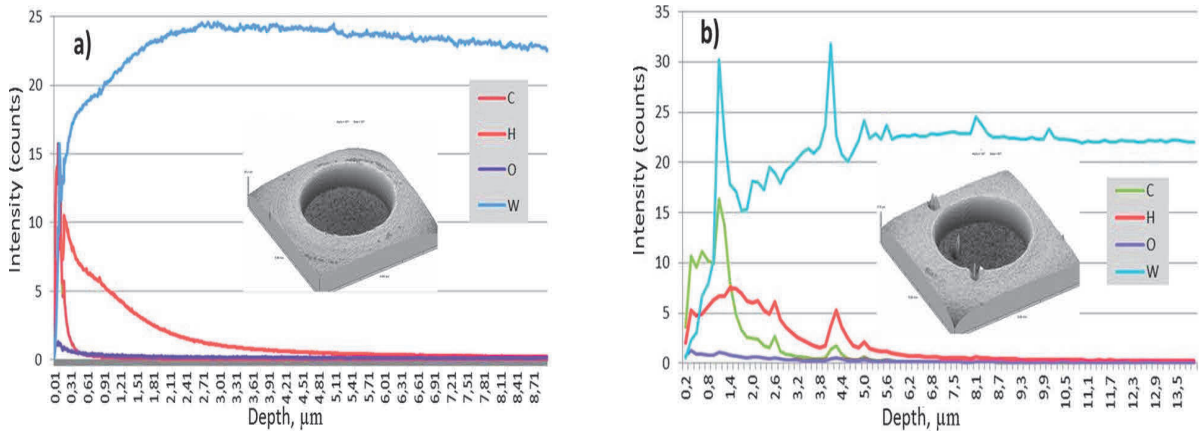

Figure 5 Profiles of intensive distribution of hydrogen yield for tungsten samples irradiated with hydrogen (a) and deuterium plasma (b)

In the course of given research it is established, that in spite of the irradiation temperature is close to the temperature of maximum gas release, deuterium is accumulated on the surface layer. It might be associated to defect structure formed while irradiation under high temperatures T = 1000 °C. So much deuterium is not accumulated on the surface layer of unaffected tungsten at this heating temperature.

4. CONCLUSION

1) It is established, that when irradiation there are some changes in the surface like relief propagation caused by heterogeneous surface etching. Ratio of change in relief and structure of surface layer of irradiated samples depends on the irradiation temperature. Metallographic analysis showed that tungsten samples irradiated at T = 1000 °C and 1500 °C have lower degree of relief propagation in comparison with those which were irradiated at T = 700 °C. However, some small cracks are shown in these samples.
2) In this connection we have studied accumulation of hydrogen and deuterium on the defects as fine cracks formed in tungsten at high irradiation temperatures with hydrogen and deuterium plasma. Thermo-desorption analysis of tungsten samples irradiated with hydrogen and deuterium plasma at T = 10000 °C showed that tungsten surface is saturated with deuterium and isn't saturated with hydrogen.
3) For tungsten sample irradiated with hydrogen plasma, recording of visible hydrogen release failed due to high background resulting from hydrogen release from the crucible and structural elements of vacuum chamber while linear heating. For tungsten sample irradiated with deuterium plasma there is a peak of deuterium release (amu4) within the range of tested sample temperatures from 1150 K to 1275 K. Total number of deuterium released of tested sample was equal to $4.09 \cdot 10^{-11}$mol.

4) Results obtained in optical-emission spectrometer showed hydrogen accumulation on the surface layer of tungsten samples irradiated with hydrogen and deuterium plasma up to 7 μm deep. In samples herewith, irradiated with hydrogen plasma at the depth of 0.2 - 0.3 μm there is a clearly visible peak related to hydrogen near-surface accumulation. Then the intensity is declined. Tungsten sample irradiated with deuterium plasma, intensity peak is observed at the depth of 2 μm; hydrogen distribution depth is smaller than by the samples irradiated with hydrogen plasma.
5) Comparison of the data obtained by thermo-desorption spectrometry and optical-emission spectrometry gives reason to believe that structure imperfection appeared during irradiation leads to deuterium accumulation on the surface tungsten layer in spite of the high irradiation temperature.

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