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Deuterium trapping properties of plasma sprayed tungsten layers irradiated by low energy and high flux plasma

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Deuterium retention and trapping in tungsten plasma sprayed layers after low energy plasma irradiation was investigated by means of thermal desorption technique. Plasma irradiation at low temperatures (<650 K) showed the retention and trapping similar to polycrystalline tungsten material. On the other hand, irradiation at high temperatures (>650 K) showed significant increase of the retention in the plasma sprayed layers contrary to the polycrystalline material. Thermal desorption spectra showed extremely high temperatures of the peak positions which are non-typical for the tungsten materials. The possible reasons for such high deuterium retention in the plasma sprayed layers after irradiation at high temperatures and the possible trapping mechanisms are discussed.

Key words: *plasma sprayed tungsten, thermal desorption, retention, plasma irradiation*

1. Introduction

Tungsten material is one of candidates for the divertor plates in the future thermonuclear experimental reactor ITER. Because of difficult machining and welding of bulk tungsten, plasma sprayed (PS) tungsten (W) layer, which has been used in ASDEX and other tokamaks, is also considered as plasma facing material. PS-W has been used in the divertor plates and inner guard limiters of the QUEST spherical tokamak in Kyushu University[1]. The main advantages of PS-W layer are low cost and possibility of coating of large areas even of non-planar shapes[2]. Although a lot of studies on hydrogen isotope retention have been done with polycrystalline tungsten, there are few studies for the PS-W layers. Literature data show large scattering of data for plasma exposed PS-W, retention mechanisms have not been investigated in details. Because divertor materials will be exposed to high flux plasma and heated to high temperatures, investigations under these conditions are of great interest.

It is necessary to study the retention characteristics of the PS-W layer in the laboratory conditions in order to predict its behavior in the plasma confinement machines. In this study, the deuterium (D) retention in PS-W layers exposed to low energy D plasma was investigated at different irradiation temperatures (T_{irr}) and fluences by means of thermal desorption spectroscopy (TDS).

2. Experimental details

VPS (vacuum PS) and APS (atmospheric PS) W samples were used in this study. The thickness of W layer on 1 mm SS substrate was 150 μm. The samples were produced by TOCALO corp. Experiments have been conducted with as received samples as well as with polished ones. The latter were polished mechanically and electro-chemically and then outgassed by linear heating from 300 K up to 1473 K with the rate of 1 K/sec. Polycrystalline W (PC-W) sample with 99.99% purity and Φ10×1 mm dimensions, produced by A.L.M.T.

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corp. was used in the experiments for comparison. It had been polished mechanically to a mirror-like surface and annealed at 1173 K for 1 hour in vacuum prior to the first irradiation.

The samples were exposed to low energy D plasma in the Advanced PWI Simulation Experimental Device and Analysis System (APSEDAS). The scheme of the installation is shown in Fig. 1. The plasma was produced by 13.56 MHz RF field in a steady state magnetic field. The samples were mounted on a copper water-cooled stage and exposed to the various fluences up to 10^{26} D/m² under the constant flux of about 3×10^{21} D/m²sec. The stage was electrically grounded. The energy of ions coming from the plasma to the surface of the sample was estimated to be about 30 eV by the Langmuir probe measurement. During the irradiation the samples were heated by intense plasma particle fluxes and the surface temperature was measured by an infrared thermometer through a pyrex window. Additional substrates were inserted between the sample and the water cooled stage in order to fix the sample surface temperature at a desired level.
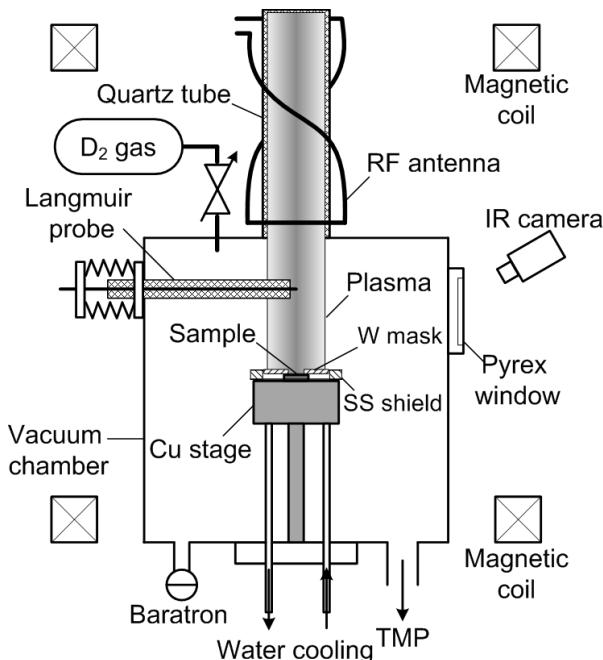

Fig.1 APSEDAS PWI simulator scheme

Measurements of thermal desorption from samples were performed in a separate stand. A sample was put on a tantalum bed. A thermocouple was attached on the backside of the bed. Then the bed was evacuated inside a quartz tube and heated by an infrared oven from 300 K up to 1473 K with the rate of 1 K/sec. The desorption rates of various species were measured by a quadrupole mass spectrometer. The calibration of the sensitivity of the spectrometer was performed in each experiment using standard He leak taking into account the ratio of ionization cross-sections $\sigma_{He+}/\sigma_{D2+}$, which was measured experimentally. The amount of desorbed deuterium was calculated as an integral of D_2 and HD desorption signals over the time. HD calibration factor was assumed to be the same as that of D_2.

There might be a certain concern about the temperature distribution in the PS-W layer during the plasma irradiation and also during the TDS heating, since the thermal conductivity of the PS-W layer seems to be low. The temperature of a deeper position in the PS-W layer might differ from that on the surface. From a simple calculation, the maximum temperature difference in the PS-W layer is considered to be only 2–3 degree even for the thermal conductivity of 100 times smaller than that of polycrystalline W. Therefore, errors in the temperature measurement during the irradiation and during the TDS heating seem to be small.

3. Results and discussion

The thermal desorption spectra of deuterium for PCW, VPS-W and APS-W after irradiation at different T_{irr} up to 800 K at the fluence of 2×10^{25} D/m² are shown in Fig. 2.

At low T_{irr} (<600 K), TDS spectra for APS-W and VPS-W are similar to that for PCW.

The contribution of the low temperature part (below 900 K) in the TDS spectra decreases for all materials with the increase of T_{irr}. On the other hand, the contribution of high temperature part (above 900 K) increases with T_{irr} for PS W and is not changed for PCW.

The structure of TDS spectra from APS-W and VPS-W in the high temperature

desorption region is complicated. It consists of many desorption peaks with different amplitudes. The peaks are overlapped and form wide spectrum. Though both materials revealed similar desorption peak positions in this region, the structure of the spectra is different. In other words, for a given T_{irr} the distribution of the peak amplitudes in the spectra are different for APS-W and VPS-W.

Figure 3 shows TDS spectra of deuterium from APS-W irradiated at various fluences up to 4.6×10^{25} D/m^2 at T_{irr} = 760 K. It is seen that all peak amplitudes grow with the fluence. The relative amplitudes of the spectra are not changed with the fluence much. The expression "relative amplitudes" can be applied to a set of spectra and means following. If we normalize a set of TDS spectra and all amplitudes of peaks located at the same temperatures become the same then we say that the relative peak amplitudes in the set of the spectra are the same. The normalization should be performed so that the amplitudes of one chosen peak in every spectrum from the set became the same.

Figure 4 shows the dependence of the deuterium retention on T_{irr} in PC-W, APS-W and VPS-W, where the fluence was 2×10^{25} D/m^2. In the low T_{irr} region (near 500 K) the retention in PS-W is 2-3 times larger, than that in PCW. In the high T_{irr} (above 650 K) the retention for PS W layers is much higher compared to PCW.

The retention in PCW quickly decreases with increase in T_{irr}, while that in PS-W increases up to two orders larger than that of PC-W at T_{irr} = 750 K. There seems to be no sign of saturation of the retention in APS W up to T_{irr} = 800 K.

Figure 5 shows fluence dependence of the retention in APS-W calculated from TDS spectra in Fig. 3. The retention increases as the square root of the fluence up to 5×10^{25} D/m^2 and then it saturates. Irradiation at the fluence of 9.8×10^{25} D/m^2 showed even smaller retention compared to that at the fluence of 4.6×10^{25} D/m^2. Note that all points in this dependence had been measured consequently with the same sample. Moreover, six monthes after a set of experiments with this sample at

T_{irr} = 750 K the repetitive measurements of the retention at the fluences of 2×10^{25} D/m^2 and 4×10^{25} D/m^2 have been conducted. These measurements showed 4 times less values and
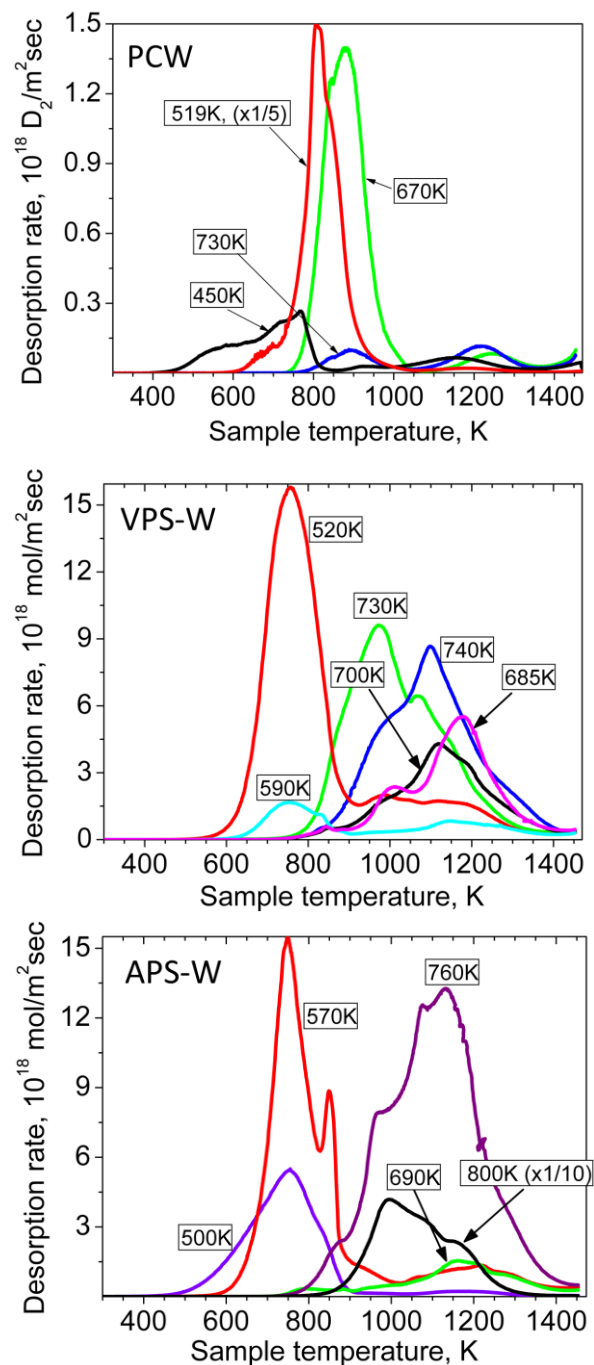

Fig.2 TDS spectra of D$_2$+0.5HD from PCW, VPS W and APS W samples irradiated up to the fluence of 2×10^{25} D/m^2. The irradiation temperatures are shown in the plots.

than that of earlier measurements performed after irradiation at similar fluences (Fig. 5), though the peak positions of the corresponded

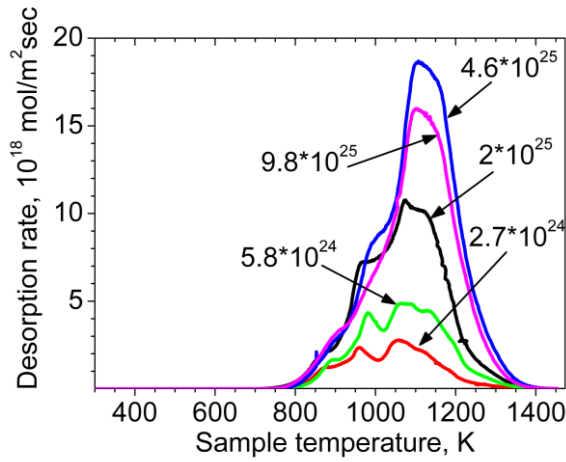TDS spectra as well as their shape remained the same.

Fig.3 TDS spectra of D_2+0.5HD from APS W irradiated by D plasma at the irradiation temperature 760 K. The fluences are shown in the plot in D/m^2.

An example of scanning electron microscope (SEM) picture of chemically polished surface of APS-W is shown in Fig. 6. It is seen from the figure that the material consists of tungsten beads with the voids between them. Sometimes the beads contain a
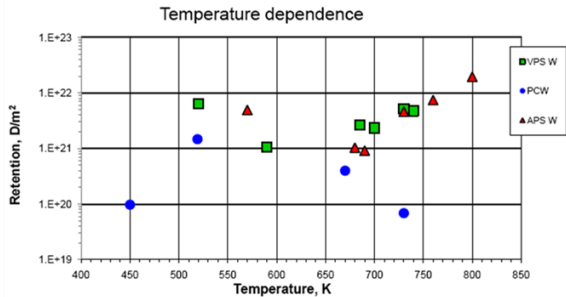

Fig.4 Irradiation temperature dependence of D retention in PCW, APS W and VPS W irradiated by D plasma up to the fluence of $2×10^{25}$ D/m^2

lot of small bubbles and voids. The net of voids forms the pores or paths for the deuterium atoms and molecules from the surface into the bulk. Thus, deuterium can migrate in the form of molecules or atoms along the internal surfaces toward the top surface. Since closed pores and voids exist in the material, deuterium gas penetrated and accumulated inside them needs to overcome the surface barrier to release to the vacuum or to the open pores. Most probably, the internal surfaces of

the pores and voids are enriched by the impurities, which are accumulated on the surface layer of each grain during the deposition process.

Usually, the desorption of hydrogen isotopes implanted by ion beams in PC-W as well as by plasma has been observed within 300 K–1000 K region[3)-6)]. Contrary to the polycrystalline tungsten material, high temperature (>1000 K) desorption from plasma sprayed tungsten layers has been reported in several studies[7)-9)].
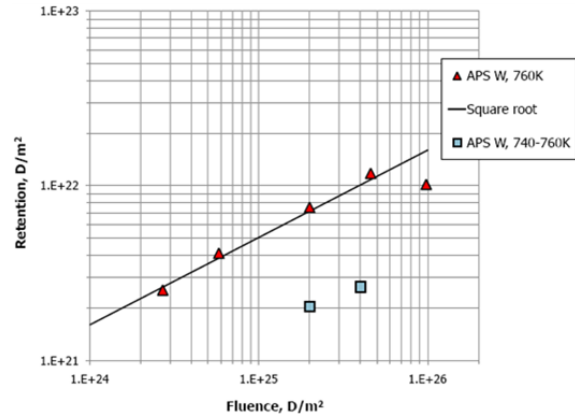

Fig.5 Fluence dependence of D retention in APS W exposed to D plasma at 760 K (triangles). The repetitive points are shown by squares, the square root behavior is shown by the line
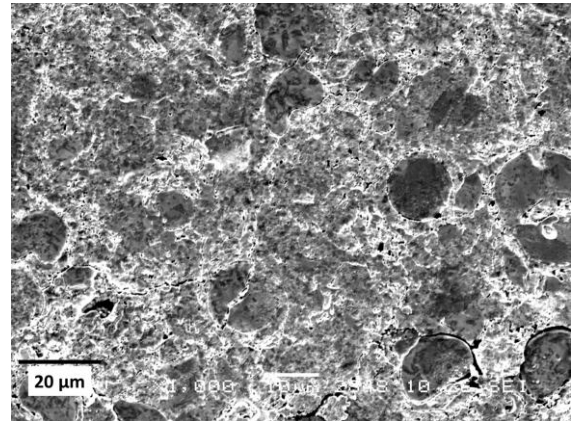

Fig.6 SEM picture of APS W chemically polished surface

In ref. 10, Otsuka et al. have measured tritium depth profiles in plasma exposed APS-W and VPS-W layers deposited on F82H by means of tritium imaging plate technique. It has been shown that the contribution of the F82H substrate in the total deuterium retention is small and decreases with T_{irr}. Even at the T_{irr} = 453 K tritium distributes

throughout whole 1 mm deposited APS and VPS layers. This depth is extremely large compared to deuterium permeation inside polycrystalline W material at these conditions. Deep tritium migration was attributed to tritium diffusion (penetration) through open pores and/or migration along grain boundaries followed by adsorption on grain surfaces facing the pores and dissolution of T in the grains.

TDS spectra evolution with T_{irr} (Fig. 2) from APS-W, VPS-W and PCW suggest that PS-W layers have high energy trapping sites additionally to lower energy ones, typical for PC-W. Complicated shape of TDS spectra from PS-W in the high temperature desorption region indicates that there are multiple high energy traps with different detrapping energies. Similar desorption peak positions in the high temperature region of TDS spectra from APS-W and VPS-W layers leads to the conclusion that the both materials have similar high energy trapping sites. Different relative peak amplitudes in the TDS spectra from APS-W and VPS-W can be explained by different concentrations of high energy traps in the materials. At low T_{irr} the trapping mechanisms in PS-W layers are similar to those in the PCW, but still small amout of D atoms is trapped in the high energy traps. The role of the high energy traps increases substantially in PS-W layers with T_{irr} increase. This allows us to suggest that at higher T_{irr} D atoms can reach the regions which they cannot reach at low T_{irr} and can be trapped there.
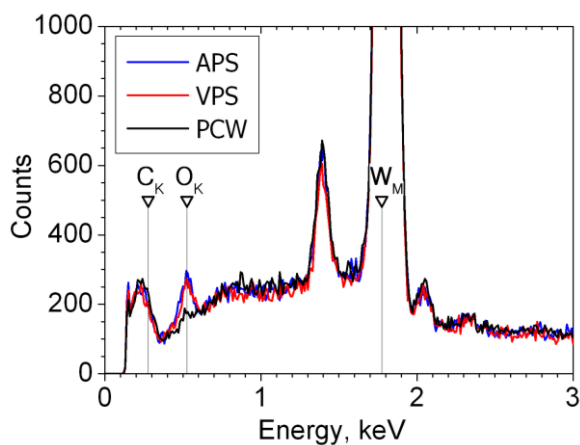

Fig.7 EDS spectra of plasma irradiated surface of non polished APS W, VPS W and PCW. CK, OK, WK – the energy level of X-rays corresponded to the K shells of C, O and W atoms.

Surface analysis of plasma irradiated VPS-W and APS-W layers was performed by Energy Dispersive Spectroscopy (EDS). The both surfaces were contaminated by oxygen. The oxygen concentration was higher than that of PCW as shown in Fig. 7. It seems that the surface of each W drop in PS-W is enriched by impurities, mainly oxygen, bounded to the beads during the deposition process. Other studies have also showed the presence of oxygen on the surface of W beads in PS-W layers, e.g. ref. 9.

At high plasma irradiation temperatures plasma sprayed layers can store huge amount of hydrogen isotopes. In order to remove it from the material heating up to high temperatures is necessary. Probably, the internal structure of the layers plays crucial role in the hydrogen isotopes accumulation. In further investigations the D retention in PS-W layers deposited in different conditions should be investigated. The deposition conditions should be changed to keep lower concentration and size of pores and cracks between W beads and lower contamination level of the W beads surfaces.

4. Conclusion

Deuterium trapping at low energy plasma irradiation in plasma sprayed layers was investigated by means of thermal desorption spectroscopy. It was observed that retention in the plasma sprayed layers strongly increases for the irradiation temperatures higher 650 K, contrary to polycrystalline tungsten. The high temperature part (>1000 K) of TDS gives a large contribution in the total retention after high temperature irradiations. High oxygen concentration on the surfaces of PS-W layers was observed.

High storage ability of hydrogen and high detrapping energies could be negative for using this material as plasma facing in the fusion reactor especially at elevated divertor working temperatures. It is proposed that the contamination level and the porous structure have strong influence on the retention

properties in the PS-W layers. In order to improve their characteristics it is necessary to reduce the number of voids and cracks and contamination level in plasma sprayed layers by appropriate deposition procedure.

References

1) S.K. Sharma, H. Zushi, N. Yoshida et al., Fusion Engineering and Design, 87 (2012) 77–86

2) Jirı Matejıcek, Pavel Chraska, and Jochen Linke, Thermal Spray Coatings for Fusion, Applications – Review, Journal of Thermal Spray Technology, Volume 16(1) March 2007-83

3) M. Poon, A.A. Haasz, J.W. Davis, Journal of Nuclear Materials, 374 (2008) 390-402

4) O.V. Ogorodnikova, J. Roth, M. Mayer, Journal of Nuclear Materials, 313–316 (2003) 469–477

5) A. Rusinov, Yu. Gasparyan, N. Trifonov et al., Journal of Nuclear Materials, 415 (2011) S645–S648

6) J.P. Sharpe, R.D. Kolasinski, M. Shimada et al. Journal of Nuclear Materials, 390–391 (2009) 709–712

7) O V Ogorodnikova, T Schwarz-Selinger, K Sugiyama et al., Physica Scripta, T138 (2009) 014053

8) V.Kh. Alimov, H. Nakamura, B. Tyburska-Püschel et al., Journal of Nuclear Materials, 414 (2011) 479–484

9) Yaran Niu, Sachiko Suzuki, Xuebin Zheng et al., Journal of Nuclear Materials, 417 (2011) 551-554

10) T. Otsuka, T. Tanabe and K. Tokunaga, Physica Scripta, T145 (2011) 014035