Tritium retention behavior in tungsten exposed to off-normal plasma condition

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Abstract- A first series of experiment concerning off-normal plasma event were performed under US-Japan collaboration program. The off-normal plasma event was simulated by introducing nitrogen gas into DT plasma. Tritium retention amount and thermal desorption behavior of tritium from tungsten exposed pure DT plasma and nitrogen introduced DT plasma was investigated as a function of nitrogen flow rate. As a result, it was found that tritium retention increased and tritium release temperature rose with increasing nitrogen flow rate. Surface analysis by XPS was also carried out for tungsten specimens after the thermal desorption. Effective chemical change on the surface chemical composition or chemical state was not observed between DT and nitrogen introduced DT plasma.

INTRODUCTION

For fusion reactor development, knowledge of tritium behavior in plasma facing materials (PFM) is a key issue. Tritium inventory and its existing state in the PFM exposed to a deuterium (D) + tritium (T) plasma with normal conditions has been investigated and the construction of a database has progressed [1,2]. However, there are still uncertainties on the behavior of tritium in the PFM in the case of exposure to off-normal plasma conditions. Such conditions include air leakage and ingress of coolant into the vacuum vessel. To date, there are few data on these situations. Therefore, study of tritium behavior in PFM at off-normal plasma conditions is underway in the US - Japan collaboration program.

The first series of experiments concerning the effects of air leakage were performed using the Tritium Plasma Experiment apparatus (TPE) at the Tritium Systems Test Assembly of Los Alamos National Laboratory.

EXPERIMENTAL

Off-normal condition on DT plasma was simulated by injection of nitrogen gas into DT plasma generated in the TPE. Tungsten, which is a candidate for divertor in ITER, was selected for specimen in this experiment. The specimens were the disks, which have 50.0 mm in diameter and 1.0 mm thickness, manufactured by Nilaco Co, Ltd.

The experiment consists of following operations.

1) Plasma exposure simulated off-normal condition using TPE,
2) Tritium measurement by thermal desorption (TDS) methods,
3) Surface analysis of specimen after TDS by XPS.

1. PLASMA EXPOSURE USING TPE

Plasma exposure was carried out using TPE. The TPE is only one apparatus, which can produce high flux tritium plasma by arc reflex ion source. Detail specification of the TPE is described in previous paper [3]. Plasma exposure for off-normal simulation was carried out as follows.
At first, the tungsten specimen was heated up to designated temperature (373 K in this experiment) by D_2 plasma. After the specimen temperature was stabilized, pure DT plasma exposure was initiated and continued for 30 minutes. Nitrogen was then introduced into the plasma for an additional 30 minutes. During the period of pure DT plasma exposure, the D-T ion flux was about 3.2×10^{21} ions/m^2/s at an energy of 100 eV. The total flow rate of DT gas (D_2:T_2=19:1) was kept at 20 sccm and the flow rate of nitrogen gas, for the simulation of leakage, was taken at 0, 1, 3 and 5 sccm in the individual experiment.

2. TRITIUM MEASUREMENT BY TDS METHOD

After the plasma exposure, the tungsten specimen was taken out from TPE and moved to a furnace of the outgassing system. Figure 1 shows a flow sheet of the outgassing system. This system is consists of a furnace, bubblers and ionization chamber. The tungsten specimen was heated in the furnace in Ar/H_2(1%) mixture gas flow to 1473 K at a

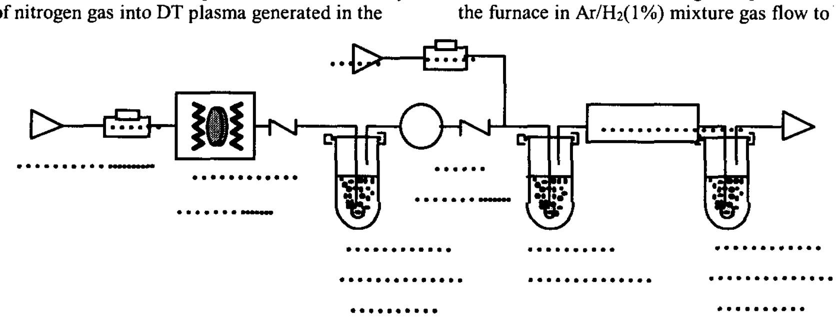

Figure 1 A flow sheet of outgassing system

constant ramp rate of 20K/min. Tritium released from tungsten specimen was measured by an ionization chamber or collected by bubblers. The tritium released in water-soluble form such as vapor was collected by 1st bubbler, and measured tritium amount by a liquid scintillation counter after the TDS operation. The tritium released in hydrogen molecule form was measured by the ion chamber continuously.

3. SURFACE ANALYSIS BY XPS AFTER TDS

Surface analysis by XPS was carried out for the tungsten specimen after TDS operation. The XPS analysis was performed using a PERKIN ELMER model 1600 with a monochromatic X-ray source (Al - Kα). It was impossible to analyze the specimen before TDS operation due to difficulty of contaminated specimen treatment. Atomic elements and chemical states of the elements existing on the tungsten surface were measured by the XPS analysis.

RESULTS

1. RESULT OF PLASMA EXPOSURE

Figure 2 shows the trends of the ion current at the specimen and target temperature during plasma exposure experiment. When nitrogen gas was injected into DT plasma, the ion current was enhanced and gradually decreased after that. The ion current increased again for 3 and 5sccm N_2 injection case. During the plasma exposure, target temperature corresponded to the ion current because the specimen was heated only by plasma. The ion current at just after stopping N_2 introduce was lower than that before N_2 introduce even the plasma parameter was not changed during the plasma exposure. This behavior indicates that the N_2 injection cause change at sample surface such as nitride formation, irradiation defects etc.

2. RESULT OF TRITIUM MEASUREMENT

In the TDS experiment, it was found that almost tritium was released in a form of hydrogen molecule from tungsten by the result of bubbler measurement. In some case, the result showed that the tritium released in the vapor form was less than 1%.

Figure 3 shows the result of ionization chamber measurement of the TDS experiment for individual

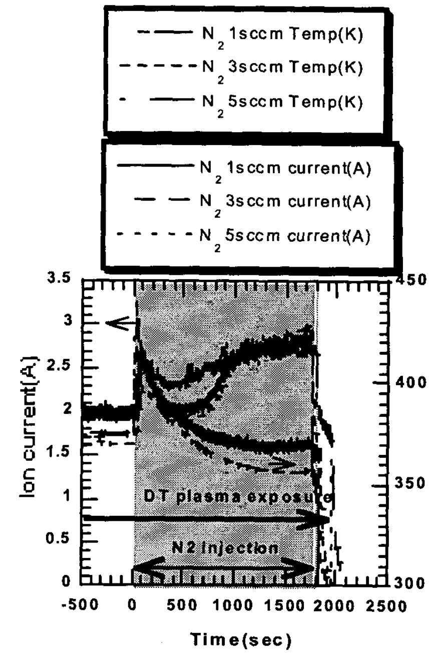

Figure 2. The trends of the ion current and temperature during plasma exposure

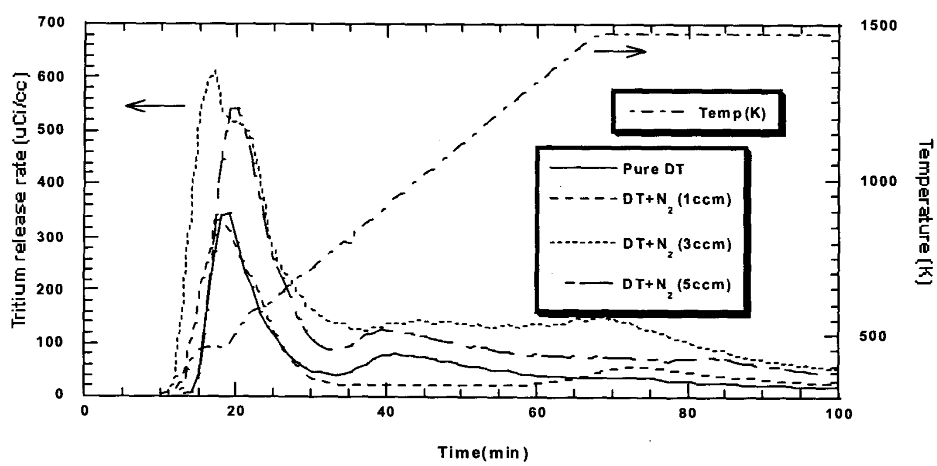

Figure 3. The result of TDS of tritium from tungsten measured by ionization chamber

experiment, where, right axis shows tritium concentration in the carrier gas in a form of hydrogen molecule and left axis shows the specimen temperature.
The TDS result taken with the sample exposed to pure DT plasma showed a major peak around 460 K. On the other hand, it can be seen a shoulder around 560K in a case of the specimen exposed to the DT plasma with 1 sccm N_2 flow rate and minor peak around 600 K in a case of 3sccm N_2 flow into DT plasma.

Table 1 summarized that the results of the TDS experiment. The peak temperatures were derived from the curve fitting using asymmetric gaussian method. This table also lists the surface trap energies on tungsten surface predicted by a TDS mode of TPERM code [4], which was calculated in an assumption that the tritium is trapped only in surface region (5nm) of tungsten for individual case using literature's value of physical properties for tungsten [5]. It can be seen two things in this table. One is that total tritium retention amount has a tendency to increase with N_2 introduce flow rate into DT plasma. And another one is that the peak at higher temperature (peak 2) is increasing with N_2 introduce flow rate. Where, it has a low reliability in the peak temperature for 5 sccm N_2 injection case due to scattered temperature during TDS operation. Obtained trap energy of tungsten exposed pure DT plasma was almost corresponded to one reported by Anderl et al [6]. The obtained surface trap energy was increased with the N_2 injection flow as well as peak temperature.

Table 1 A summary of TDS experiment

	Tritium retention amount	Peak 1	Peak 2	Surface trap energy calculated by TPERM code	
		Release temperature	Release temperature		
		ratio	tratio	Peak 1	Peak 2
Pure DT	0.5 mCi	463 K	• •	0.96 eV	• •
		1.0	• •		
$DT+N_2$ (1sccm)	0.5 mCi	481 K	561 K	1.00 eV	1.24 eV
		0.867	0.133		
$DT+N_2$ (3sccm)	1.6 mCi	496 K	602 K	1.05 eV	1.36 eV
		0.747	0.253		
$DT+N_2$ (5sccm)	1.4 mCi	510 K	• •	1.10 eV	• •
		1.0	• •		

The experimental results of TDS, which mean increase of tritium retention amount in tungsten, rise of the peak temperature of tritium release from tungsten and increase of tritium release ratio at higher temperature with nitrogen injection flow rate into DT plasma, imply that nitrogen injection induces the stronger binding energy of tritium in tungsten compared to the case of pure DT plasma exposure.

3. RESULT OF SURFACE ANALYSIS

Figure 4 shows the results of the XPS analysis on the tungsten surface exposed pure DT and DT $+N_2$ (3sccm) plasma after the TDS operation. It can be seen major W peaks and O, C & minor N peak on both specimen surfaces in fig. 4 (a). It was not observed clear difference between pure DT and DT + N_2 plasma exposure specimen. On the other hand, although chemical shift of 15% of W peak was observed on both surfaces, observed chemical shift was agreed with ones of tungsten oxide [7] as shown in fig. 4 (b). Therefore, the XPS result would be concluded the difference between pure DT and DT + N_2 plasma exposure specimen was not observed on the specimen after the TDS experiment.

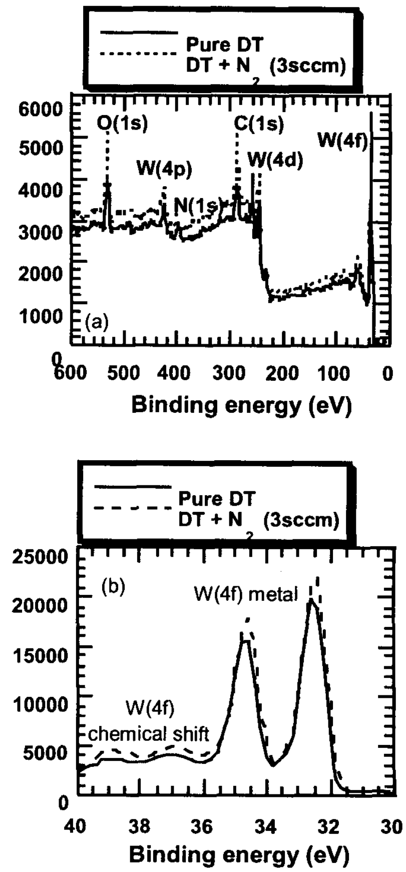

Figure 4. The result of surface analysis by XPS

DISCUSSIONS

Summarizing the results of experiments such as plasma exposure, TDS, and surface analysis, it is obvious that DT + N_2 plasma exposure to tungsten causes changes of chemical or physical state on the tungsten surface or near surface. Where, the change of chemical state of surface means tungsten nitride formation etc., and change of physical state means production of trap site by the damage produced by nitrogen implantation etc.

With regard to the chemical state, although it is known the instability of tungsten nitride production, the successful tungsten nitride formation was reported using plasma reaction method and ion bombardment [8,9]. It would be possible to be created tungsten nitride on the surface by plasma enhanced reaction. However, one paper reported nitrogen is released form tungsten nitride around 900 to 1100K [9]. As mentioned previously, the XPS analysis was carried out after TDS experiment, which heated tungsten up to 1473K. Therefore there is a possibility that tungsten nitride created by the plasma exposure was decomposed during the TDS experiment. It is necessary the knowledge concerned to the chemical state of the surface before TDS operation. Because surface contamination of tungsten specimen after DT experiment is quite high level, the additional experiment with $D_2 + N_2$ plasma is desired to verify the chemical state after plasma exposure.

On the other hand, for the physical state, it is appropriate that the surface trap energy of tungsten exposed to pure DT plasma obtained in this experiment agreed with the trap energy in the bulk tungsten reported Anderl et al. And it is well known that the damages produced ion implantation into the materials sometimes act as a trap site of hydrogen isotopes [10,11]. However, there are some difficulties on understanding above description, which is whether low energy (lower than 100eV in this experimental condition) implantation could cause damages or not.

The surface observation using SEM is progressed for the tungsten specimen exposed to DT and DT $+N_2$ plasma in this position. This result might give useful information for the physical state change.

It can not be determined the mechanism of tritium behavior under the nitrogen injection condition into DT plasma in this position. It will be planned additional experiment, such as plasma exposure at higher temperature, surface analysis by XPS and SEM after plasma exposure, to acquire the knowledge of the mechanism of tritium behavior.

SUMMARY

Tritium retention and TDS behavior from tungsten specimen exposed to DT plasma with nitrogen introduction was carried out as a preliminary experiment of off-normal plasma simulation study under the US - Japan collaboration program.

It was found that the nitrogen introduce into DT plasma induced the enhancement of tritium retention amount, rise of tritium release temperature and increase the ratio of tritium release at higher temperature from tungsten.

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