

## Hydrogen Concentration of Co-deposited Carbon Films Produced in the Vicinity of Local Island Divertor in Large Helical Device

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**Abstract.** It is quite important to evaluate hydrogen concentration of co-deposited carbon film/dust to estimate in-vessel tritium inventory in ITER. The co-deposited carbon films were prepared at the wall of pumping duct in Local Island Divertor experiments of LHD. The hydrogen concentration of the co-deposited carbon film at the wall not facing to the plasma with a low temperature was extremely high, 1.3 in the atomic ratio of H/C. This value is triple times higher than the previous value obtained by hydrogen ion irradiation to graphite. The crystal structure of the co-deposited carbon film observed by Raman spectroscopy showed very unique structure (polymeric a-C:H), which is well consistent with the high hydrogen concentration. The accumulation of in-vessel tritium inventory is also discussed.

### 1. Introduction

In ITER, one of the major concerns is tritium retention in carbon film/flake from the point of view of safety. The tritium inventory in the vacuum chamber, i.e. in-vessel tritium inventory is limited below several hundred grams. Both the carbon produced by erosion of graphite (carbon fiber composite, CFC) divertor target and fuel hydrogen deposit on the wall, and then the co-deposited carbon film is produced. It is believed that the co-deposited carbon film has a high tritium concentration. Hence, it is quite important to evaluate the tritium concentration of co-deposited carbon film.

The tritium concentration can be predicted by the data on hydrogen or deuterium concentration since the chemical binding state of these hydrogen isotopes is very similar. The deuterium concentration of carbon film/flake produced in JET [1] showed a relatively high deuterium concentration, with the atomic ratio of D/C = 0.4. The deuterium concentration of graphite after a deuterium ion irradiation at room temperature (RT) is also D/C = 0.4 [2, 3]. The co-deposited carbon films produced by deuterium arc discharge [4] showed a deuterium concentration, D/C = 0.2 for the ITER

wall condition. It is pointed out that the fuel hydrogen retention in co-deposited carbon film produced at shadow regions (walls not facing plasma) in fusion devices may become higher than the existing value,  $D/C = 0.4$ . However, such the data has not been sufficiently accumulated yet. In the present study, the co-deposited carbon film is produced at the shadow wall with a relatively low temperature in Local Island Divertor (LID) experiments of LHD. The hydrogen concentration of the co-deposited carbon film is investigated using thermal desorption spectroscopy. The surface properties and crystal structures are also investigated and the relation of the hydrogen concentration with the crystal structure is discussed.

## 2. Experiments

In LHD, the LID experiments have been conducted since 2003, in addition to an intrinsic helical divertor experiment [2]. *FIG. 1* shows the LID configuration. A divertor head that is covered by the neutralizer plates made of CFC is inserted into the  $m/n=1/1$  magnetic island in a horizontally elongated cross-section where the width of the island is maximum (about 20cm). The pumping duct surrounds the divertor head, and it works as a baffle. Outer separatrix of the island connects the divertor head as the divertor legs, and the last closed flux surface is determined by the inner separatrix of the island. Plasma-surface interaction occurs only at the divertor head ideally, and thus the wet area ( $< 0.1\text{m}^2$ ) is less than one tenth of that of helical divertor. The divertor head receives relatively high heat and particle fluxes compared with the helical divertor, and the eroded carbon co-deposits with hydrogen on the plasma facing components. Four sets of material probes made of Si were installed at the wall of pumping duct in the vicinity of the LID head from the experimental campaigns in 2005. The number of the hydrogen discharges with the LID configuration in the campaign was 300-700. The locations of the material probes are also shown in *FIG. 1*. One set of probes was placed inside the pumping duct (1U, 1L), and faced to the divertor head. The other set of probes has shallow line of sight to the divertor head (2U, 2L). These material probes were installed in the 9<sup>th</sup> and 10<sup>th</sup> experimental campaigns. After each the experimental campaign, the probes were extracted and their surface morphology, depth profiles of atomic composition, crystal structure and hydrogen retention were investigated using scanning electron microscope (SEM), Auger electron spectroscopy (AES), X-ray diffraction (XRD) and Raman spectroscopy (RS), and thermal desorption spectroscopy (TDS), respectively. The bulk density of carbon film was also estimated.

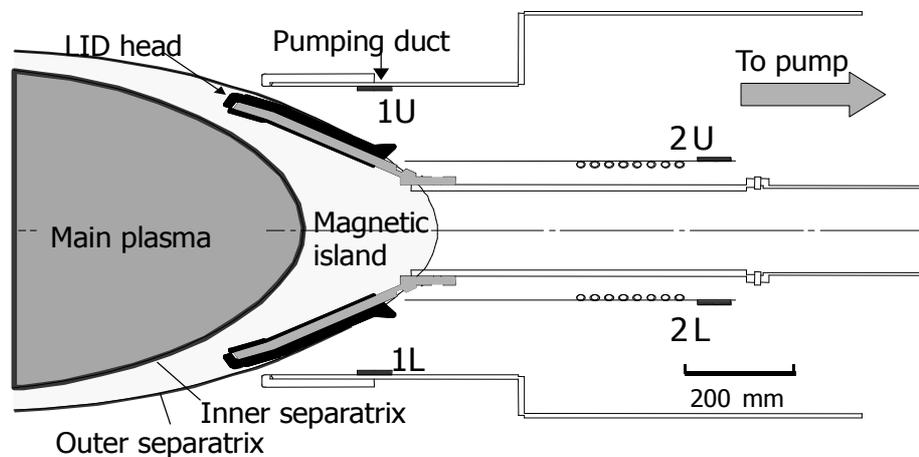


FIG.1 A schematic view of Local Island Divertor.

### 3. Results

The SEM photographs showed that the surface of 2U was very smooth, but sub-micron size protuberant structure was observed on the surface of 1U (FIG. 2(a)). This difference in the surface morphology might have been caused by the deposition angle of carbon and hydrocarbon to the probe surface. If the incident angle of carbon to the probe surface is shallow, the carbon may deposit selectively on the protuberant parts. The smooth surfaces of 2U and 2L might have been caused by the deposition of hydrocarbons, and/or low energy carbon atoms and deposition of carbon reflected by the wall. FIG. 3 shows the depth profile of atomic composition obtained by Auger electron spectroscopy. The carbon concentration was 99 at% and the other content such as oxygen was below 1 at%. Very uniform carbon composition was observed in the every co-deposited carbon film.

(a) 1U

(b) 2U

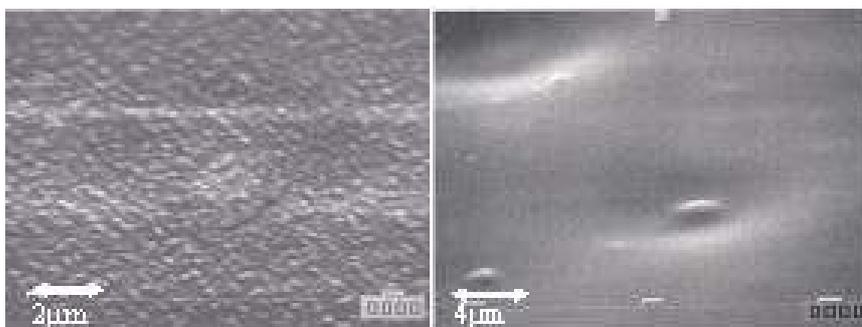


FIG. 2 SEM images of the surface morphologies of the probes, (a) 1U and (b) 2U.

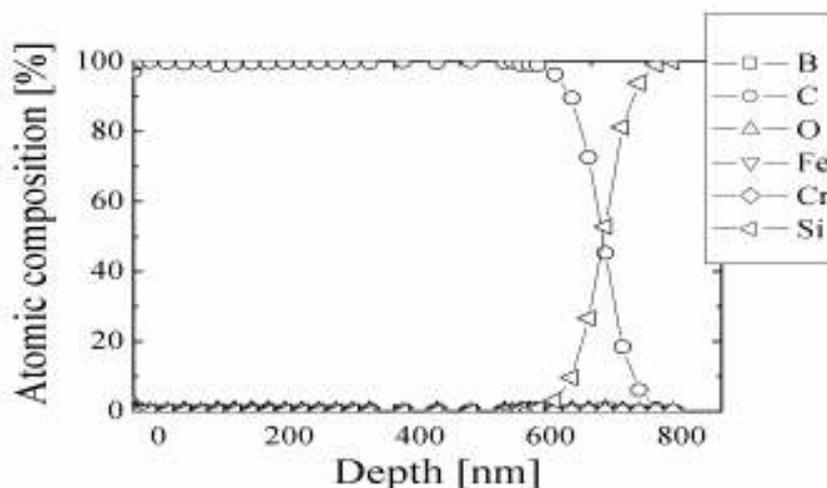


FIG. 3 The depth profile of atomic composition for carbon film produced at sample location, 1U.

In order to measure the amount of retained hydrogen in the co-deposited carbon film, the sample was placed in the vacuum chamber of thermal desorption spectroscopy device and heated up to 1273 K by infrared furnace. The emitted gas species such as  $H_2$  and  $CH_4$  during the heating was quantitatively measured using quadruple mass spectrometer (QMS). Most of the hydrogen retained in the co-deposited carbon film desorbed in form of  $H_2$ . Approximately 1% of the hydrogen retained in the co-deposited carbon film desorbed in form of  $CH_4$ . FIG. 4 shows the  $H_2$  desorption spectra for four samples. The spectra of samples 1U and 1L placed at the position close to the divertor head have a sharp peak at 1000 - 1100 K. This spectrum pattern is often observed in the case that graphite is irradiated by hydrogen or deuterium ions. However, the spectra of the samples 2L and 2U placed far from the divertor head showed a different spectrum pattern with a broad peak at around 950 K. In the later case, the desorption of hydrocarbon gas,  $CH_4$ , was relatively dominant compared with the cases of samples 1U and 1L. This implies that the deposition of hydrocarbon gas such as  $CH_4$  contributes the formation of the co-deposited carbon film. The thickness of carbon films was measured by using a surface roughness meter, and it was in the range from 200 to 700 nm. In FIG. 5, the amount of retained hydrogen and the film thickness are shown for four samples. In the sample 2L or 2U, the amount of retained hydrogen per unit thickness is larger than that in the sample 1U or 1L, so that the hydrogen concentration of the sample 2U or 2L become higher.

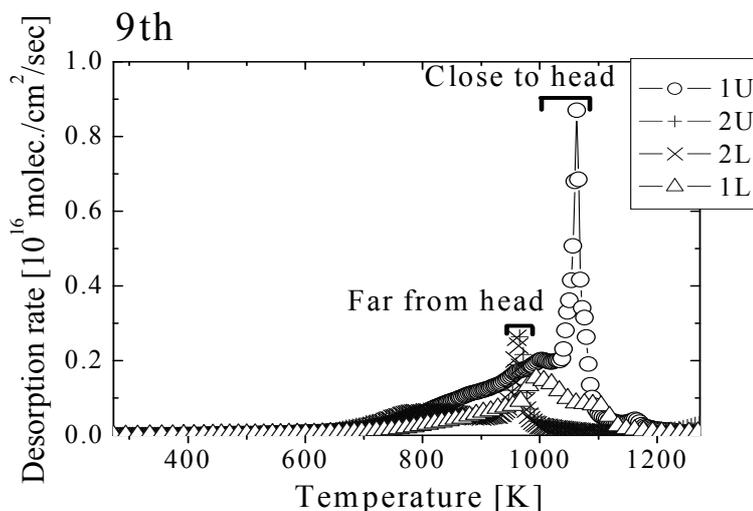


FIG.4 Thermal desorption spectra of hydrogen of the four probes.

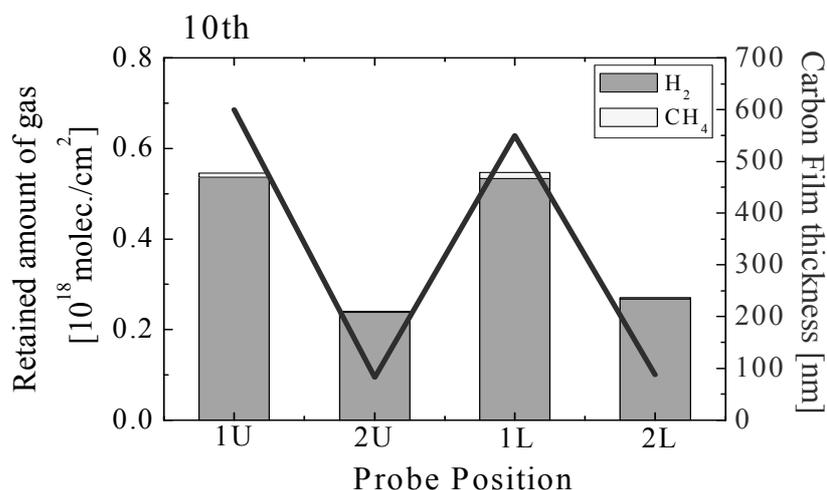
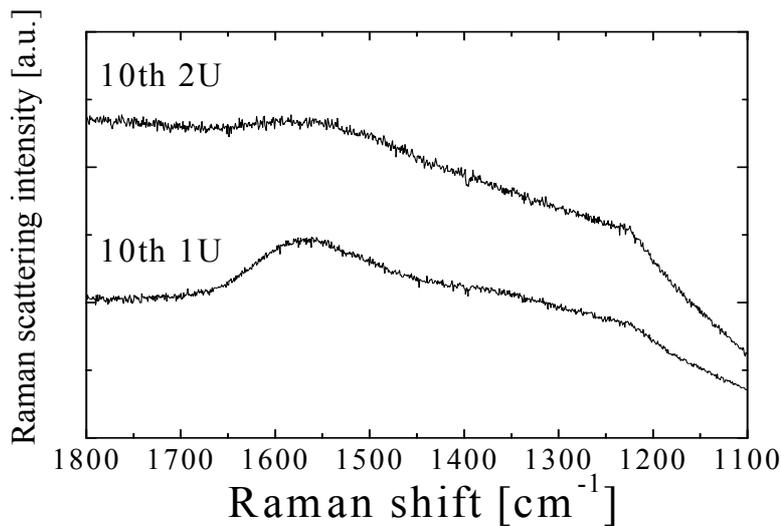


FIG. 5 Amount of retained deuterium and thickness of co-deposited carbon films.

The crystal structure was investigated by using both X-ray diffraction and Raman spectroscopy. In the X-ray analysis, there were no dominant peaks in every sample, indicating no regular crystal surfaces which cause the diffraction. However, in the Raman spectroscopy the crystal structures of 1U, 1L, 2U and 2L showed very unique structure, polymeric structure (polymeric a-C:H). FIG. 6 shows the Raman spectra of the samples 1U and 2U. The typical structure of the carbon film produced in the fusion devices so far is an amorphous structure consisting of broad peaks of graphite and defective graphite bands. The similar amorphous structure is also observed in the graphite after irradiation of energetic ions and in the carbon film produced by plasma chemical vapor deposition. The present polymeric structure contains various sizes of poly-acetylene, olefin chain and aromatic clusters, so that the hydrogen concentration becomes high owing to many bonds of C-H. In the polymeric structure of 1U and 1L,

the peak at graphite band is relatively high, which indicates that the sputtered carbon deposited on the film in addition to hydrocarbons such as  $\text{CH}_4$ . In the samples of 2U and 2L, the major deposited species of carbon is regarded to be hydrocarbon. Thus, the hydrogen concentration of 2U and 2L becomes higher than that of 1U and 1L. The hydrogen concentration in the samples produced at the shadow region was high as shown in *FIG. 5*, which is consistent with the crystal structure.



*FIG. 6* Raman spectra of co-deposited carbon films (1U: bottom, 2U: upper).

In order to determine the hydrogen concentration in the atomic ratio of H/C, the mass or bulk density of the co-deposited carbon film has to be known. The weight gain due to the film deposition could not be measured by using a mass balance since the relative weight gain was too small compared with the total sample weight. In order to evaluate the mass density, the co-deposited carbon film was prepared by using magnetron sputtering device with a carbon target and  $\text{Ar}+\text{H}_2$  mixture plasma. Argon sputter-etching rate in unit of  $\text{nm}/\text{min}$  was obtained for the film with the known mass density ( $0.98 \text{ g}/\text{cm}^3$ ). Argon sputter-etching was conducted for the co-deposited films produced in LHD. Since the sputter-etching rate is inversely proportional to the mass density, the mass density of the co-deposited carbon films produced in LHD can be obtained. The mass density of the co-deposited carbon films was in the range from  $0.90$  to  $0.93 \text{ g}/\text{cm}^3$ . The mass density of 2U and 2L was slightly lower than that of 1U and 1L.

Based upon the data of the amount of retained hydrogen, the film thickness and the mass density, the hydrogen concentration in the atomic ratio of H/C was obtained.

The hydrogen concentrations of the co-deposited carbon films at 1U and 1L became  $H/C = 0.55$  and at 2L and 2U  $H/C = 1.25$ . During the LID discharge, the sample temperature was monitored using a thermocouple. The temperature at 2U and 2L was 300 K, and the temperature at 1U and 1L was 600 K. FIG. 7 shows the hydrogen concentration of the co-deposited carbon films against the wall temperature. The hydrogen concentration of the co-deposited carbon film produced at the shadow region with a low temperature, 300 K, had an extremely high. This value is twice of the hydrogen concentration in the co-deposited carbon film produced at the wall with a temperature of 600 K. The hydrogen concentration becomes  $H/C \sim 0$  at 800 K if the data is extrapolated to the higher temperature range.

The deuterium concentration in graphite after deuterium ion irradiation has been well investigated so far by numerous authors. In this case, the deuterium concentrations in graphite at 300 K and 600 K are  $D/C = 0.4$  and 0.2, respectively [2, 3]. FIG. 7 also shows the deuterium concentration in graphite after deuterium ion irradiation. The hydrogen concentrations of the present co-deposited carbon films are approximately triple of the graphite case. Thus, it is clearly shown that the hydrogen concentration of the co-deposited carbon film with polymeric structure produced at the wall not facing to plasma is extremely higher than that after the deuterium or hydrogen ion irradiation for graphite.

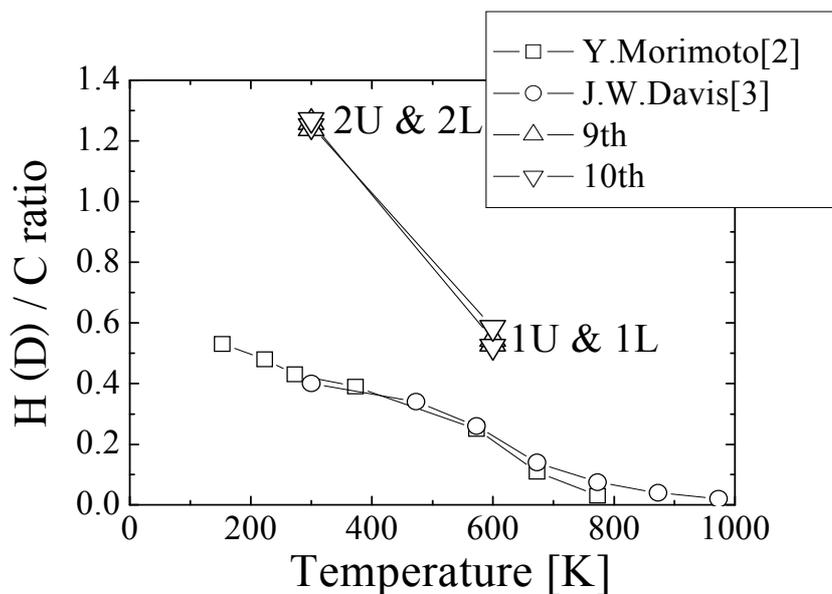


FIG. 7 Hydrogen concentration of the present co-deposited carbon films and deuterium concentration in graphite after deuterium ion irradiation [2, 3] vs wall or substrate temperature.

#### 4. Conclusion

The co-deposited carbon films were produced at the wall of pumping duct in the LID configuration of LHD. Two sets of the samples (1U and 1L) were placed close to the divertor head, and the other two sets of the samples (2U and 2L) were placed far from the head. The wall temperatures during the LID discharges were 300 K for 2U and 2L and 600 K for 1U and 1L. The hydrogen concentrations in the atomic ratio of H/C were 0.55 for 1U and 1L, and 1.25 for 2U and 2L. Such the high hydrogen concentration has not been observed for the carbon film/dust produced in the fusion devices. In addition, the wall temperature dependence of hydrogen concentration in the co-deposited carbon film was newly obtained. The crystal structures observed by Raman spectroscopy showed a quite new type structure, polymeric structure, for 1U, 1L, 2U and 2L. Many C-H bonds are included in this structure, so that the hydrogen concentration becomes extremely high. In fact, the hydrogen concentration of the present film was triple of that in graphite after hydrogen or deuterium ion irradiation. In this study, it was shown that the carbon film/dust with polymeric structure could have a quite high hydrogen concentration.

Based upon the present data and the previous our data [2], we now estimate the accumulation of in-vessel tritium inventory. The present data show that the tritium concentration of co-deposited carbon film/dust produced at the wall not facing to plasma becomes  $T/C = 0.28$  for the wall temperatures of 600 K in the DT discharge. In our previous study [2], the tritium concentration of the co-deposited carbon film facing to plasma was  $T/C = 0.1$  when the wall temperature was 600 K. We here assume that the wall temperature is 600 K, and the amount of eroded carbon is 50 g/shot, 45 g/shot of the film/dust facing to plasma and 5 g/shot of that not facing to plasma. The shot number becomes 350 shots for the tritium amount to reach the limited amount, 500 g. This is the estimation for the wall temperature of 600 K. In ITER, the wall temperature is significantly higher than 800 K. In this case,  $T/C$  becomes  $\sim 0$  as seen in *FIG. 7*. Then, the shot number for the tritium inventory to reach 500 g becomes at least a few times larger than 350 shots. Thus, the periodic cleaning for the tritium can be conducted when the divertor cassettes are replaced.

#### References

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