Carbon and Hydrogen Accumulation on Exhaust Duct in LHD*⁾

Kazunari KATAYAMA, Naoko ASHIKAWA[1], Keiichiro UEHARA and Satoshi FUKADA

Interdisciplinary Graduate School of Engineering Science, Kyushu University,

6-1 Kasugakoen, Kasuga-shi, Fukuoka 816-8580, Japan

[1]*National Institute for Fusion Science, 322-6 Oroshi-cho, Toki 509-5292, Japan*

To consider carbon balance and hydrogen isotope balance in the fuel cycle system and tritium safety management of a fusion reactor, the evaluation of carbon and hydrogen isotope accumulation not only in the vacuum vessel but also in the exhaust system is necessary. In the present work, type 316 stainless steel substrates were installed at 4 locations in the exhaust duct of the Large Helical Device (LHD) during the 13th experimental campaign. By using the combustion method, the amount of carbon slightly adhering to the substrates, which cannot be measured by electric microbalances, was successfully quantified to be $2\,\mu g/cm^2$. The hydrogen release behavior from the substrate was consistent with that from carbon deposition layer formed by hydrogen plasma sputtering. H/C ratio on the substrate was estimated to be about 1-1.5. Hydrogen incorporated into the metal deposit formed from type 316 stainless steel in the sputtering-deposition device in the laboratory can remain in the deposit even under high vacuum condition in the exhaust duct for a long period.

1. Introduction

In a fusion reactor on which graphite materials are used as plasma facing material, carbon deposition layer including hydrogen isotopes is formed on the plasma facing wall. Sticking coefficients for several kinds of hydrocarbon radical, which are generated by chemical erosion of graphite, have been evaluated by laboratory experiments [1,2]. The hydrocarbon radicals having a large sticking coefficient would be deposited widely in the plasma confinement vessel, but ones having a small sticking coefficient may be transferred to the exhaust system. In JET, carbon flakes including tritium of about 1 TBq/g accumulated at the louvres, which are installed to protect cryopumps, in a region shadowed from plasma [3]. This suggests that a certain amount of hydrocarbon and hydrocarbon radical with tritium were transported to vacuum pumps via the exhaust duct. Investigations of carbon deposition and hydrogen accumulation in the plasma confinement vessel of fusion experimental devises such as LHD [4–7] and JT60 [8, 9] has been performed. However, the understanding of carbon and hydrogen transport to the exhaust system is insufficient.

In a fusion reactor, gaseous hydrocarbon can be processed in fuel purification system but carbon with tritium adhering on the metal wall of the vacuum component and duct will be continuously accumulated in the exhaust system during the operation. Although the amount of accumulated carbon on the metal walls in the exhaust system is speculated to be significantly smaller than that in the plasma confinement vessel, the estimation of the amount of carbon and hydrogen adhering there is important for the evaluation of carbon balance and hydrogen isotope balance in the fusion reactor system and tritium safety management in a maintenance of the exhaust system.

In the present work, in order to understand carbon and hydrogen accumulation behavior in the exhaust duct of the fusion experimental devices, stainless steel (SS316) substrates were mounted in the exhaust duct of LHD, in which graphite material is used in divertor region and SS316 is used in first wall, during the 13th experimental campaign. It has been reported by the present authors that metal deposit formed from stainless steel by hydrogen plasma sputtering can retained a large amount of hydrogen during deposition process [10]. In order to confirm whether hydrogen isotopes co-deposited with metals remain in the deposit for a long time under high vacuum condition of LHD exhaust duct, metal deposit were preliminarily formed on some of SS316 substrates by hydrogen plasma sputtering of a stainless steel plate. This information would be useful for analyzing hydrogen isotope retention in the metallic dust transported to the exhaust duct during the experiment in LHD. After the campaign, the substrates were dismounted from LHD and hydrogen release from the substrate was observed by thermal desorption spectroscopy (TDS) method. Additionally, the amount of carbon adhering on the substrate was roughly estimated by combustion method.

2. Experimental

2.1 Sample location

SS316 substrates, 10 mm × 10 mm in size, 1 mm in thickness, with and without metal deposit formed from SS316 were respectively mounted on the sample holder fixed on the duct wall. The location of the exhaust duct is shown in Fig. 1. To understand the approximate tendency of carbon adhesion distribution, the sample holders were fixed on 4 locations that are different distance or direction from the vacuum chamber as shown in Fig. 2. Sample holder P1 and P2 were fixed at port A, P3 was fixed at port B and P4 was fixed at port C. Two bare SS316 substrates and two metal deposited substrates were set in each sample holder. 4 substrates can be mounted in one holder for the present study. A picture of the sample holder P3 fixed on the duct wall is shown in Fig. 3.

2.2 Preparation of metal deposit

Metal deposits were formed on the SS316 substrates by hydrogen plasma sputtering by using the capacitively coupled radio frequency (RF) sputtering device which has been shown in the previous paper [11]. A type 316 stainless steel plate, 50 mm × 50 mm in size, 1 mm in thickness and 99.5% in purity, (Nilaco Co.), was mounted on the RF electrode as a target. Several SS316 substrate were mounted on the ground electrode. After mounting SS316 substrates, the plasma chamber was evacuated by a vacuum pump and pure hydrogen gas introduced via a mass flow controller. Hydrogen gas pressure was set to be 10 Pa. RF power was controlled in the range from 150 W to 200 W with 13.56 MHz. The sputtering-deposition process was continued for 107 h. Substrate temperature was 110°C on average.

For instance, SS316 substrates with and without metal deposit mounted on the holder P1 were named as P1dep1, P1dep2, P1ss1 and P1ss2, respectively.

2.3 Analysis of SS substrate installed in LHD

Surface observation by a scanning electron microscope (SEM) and elemental analysis by an energy dispersive X-ray spectrometry (EDX) were carried out. Raman spectroscopy was performed for sample P2ss1 to investigate chemical-bonding state of carbon adhering to the substrate. However, a significant signal was not obtained because of a small amount of carbon.

TDS measurements were carried out for P3ss1, P3dep1, P4ss1 and P4dep1 with a ramping rate of 1°C/s. In order to discuss hydrogen release behavior from the SS316 substrate dismounted from LHD, deuterium release behaviors from metal deposit and carbon deposit were investigated by TDS method. The metal deposit (sample MD) was formed on a tungsten foil substrate from type 316 stainless steel by deuterium plasma sputtering in the same sputtering device used for LHD sample. Deuterium gas
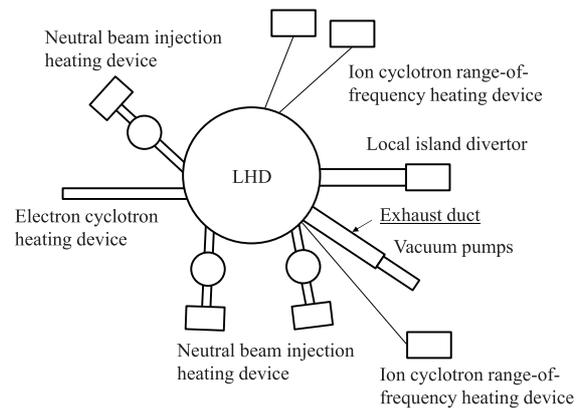

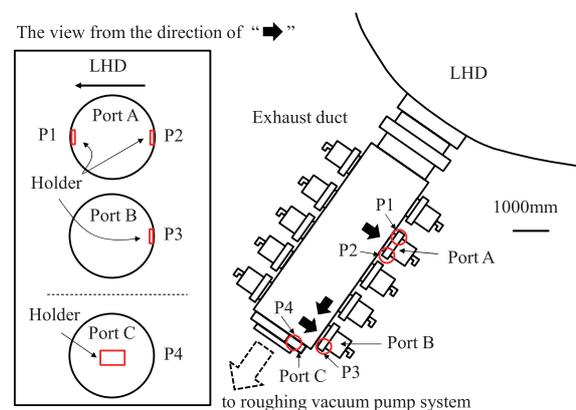Fig. 1 The location of the exhaust duct.

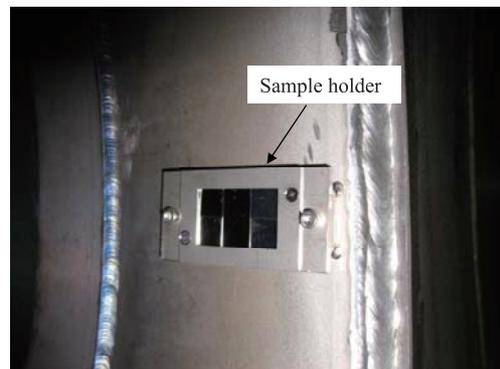Fig. 2 4 locations of the sample holders in exhaust duct.

Fig. 3 A picture of the sample holder P3 fixed on the duct wall.

pressure was set to be 10 Pa. RF power was controlled to be 100 W with 13.56 MHz. The sputtering-deposition process was continued for 130 h. Substrate temperature was about 100°C. The carbon deposit was formed on quartz substrates from isotropic graphite IG430U mainly by chemical sputtering with deuterium plasma in the other sputtering device. The sputtered carbon is basically deposited as hydrocarbon radicals. Deuterium gas pressure was set to be 20 Pa. RF power was controlled to be 200 W with 13.56 MHz. One quartz substrate (sample CD1) was

mounted on the center of the ground electrode, where deuterium ion flux is relatively high and the temperature was about 100°C. Another quartz substrate (sample CD2) was mounted on the vicinity of the chamber wall, where deuterium ion flux is quite low and the temperature was a few 10°C.

2.4 Combustion experiment

Weight of each SS316 substrates before and after installation in LHD was measured by an electronic balance with a sensitively of 10 μg. However, weight change was not observed. Therefore combustion method was tried to quantify the amount of carbon adhering to the SS316 substrate.

The schematic diagram of experimental apparatus for combustion experiment is shown in Fig. 4. In order to exclude impurity water vapor in the gas cylinder, an absorbent (MS-5A) bed was equipped in each gas lines. The sample was put into a quartz reaction tube and argon gas including oxygen was introduced. The reaction tube was closed and then heated by an electric furnace. After one hour heating, the reaction tube was opened and pure argon gas was introduced to transport gaseous components to a gas chromatograph (GC:GC-8A, SHIMAZU Co.) with a thermal conductivity detector (TCD).

As preliminary test, carbon deposit, which was prepared by hydrogen sputtering method, was heated in a closed quartz reaction tube with $1 \% O_2/Ar$ gas at 300, 400, 500 and 600°C. It was found that CO_2 was generated above 500°C. P1ss2 and P3ss2 were first heated in $0.1\% O_2/Ar$ gas at 500°C for 3 hours and then heated in $0.1 \% O_2/Ar$ gas at 500°C for 1 hour. P4ss2 was first heated in $1 \% O_2/Ar$ gas at 500°C for 1 hours and then heated in $1 \% O_2/Ar$ gas at 600°C for 1 hour. Two fresh SS316 substrates were heated in each combustion condition in order to obtain the amount of CO_2 generated by the reaction of SS316 substrate with oxygen.

3. Results and Discussion

SEM images for 4 samples without metal deposit are shown in Fig. 5. The clear boundary between the bare region and the edge region which is masked by the holder was observed on each substrate. These indicate the trace of deposition. The atomic ratio on the surface of the bare region for each sample was compared in Fig. 6. On the whole, it was found that approximately 40% of the elements near the surface are carbon. This indicates that carbon is being approximately uniformly adhering to the metal wall of the duct as gaseous components.

Hydrogen release behaviors from P3ss1, P3dep1, P4ss1 and P4dep1 are shown in Fig. 7 (a). For comparison, H_2, HD and D_2 release behaviors from MD, D_2 release behavior from CD1 and CD2 are shown in Fig. 7 (b) and Fig. 7 (c), respectively. The hydrogen release curves from P3ss1 and P4ss1, P3dep1 and P4dep1 showed relatively same shape. This indicates that hydrogen was being retained in P3ss1 and P4ss1, P3dep1 and P4dep1 under the same situation. In Fig. 7 (b), HD and D_2 release rates were magnified 5 times and 20 times respectively because these were much smaller than H_2 release rate. Deuterium release amount from MD was considerably smaller than hy-
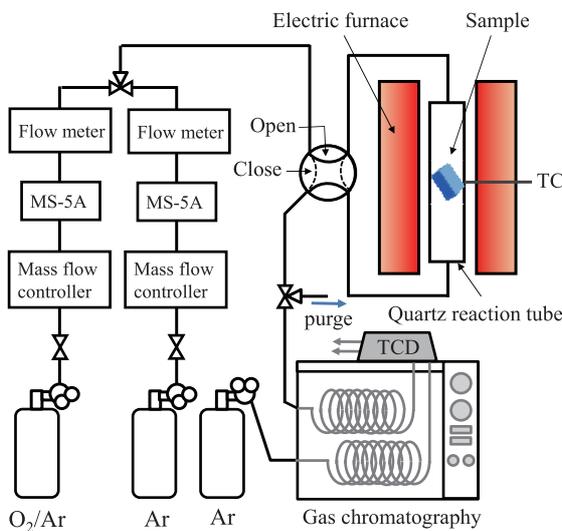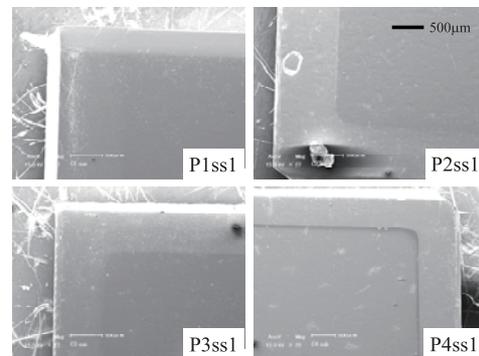
Fig. 5 SEM images of the substrate surface.
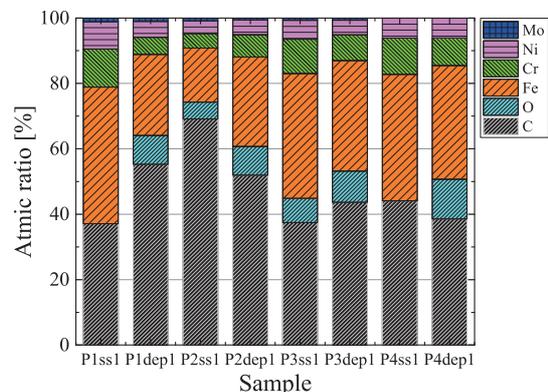
Fig. 6 Atomic ratios on the bare surface of the substrates.

Fig. 4 The schematic diagram of experimental apparatus for combustion experiment.
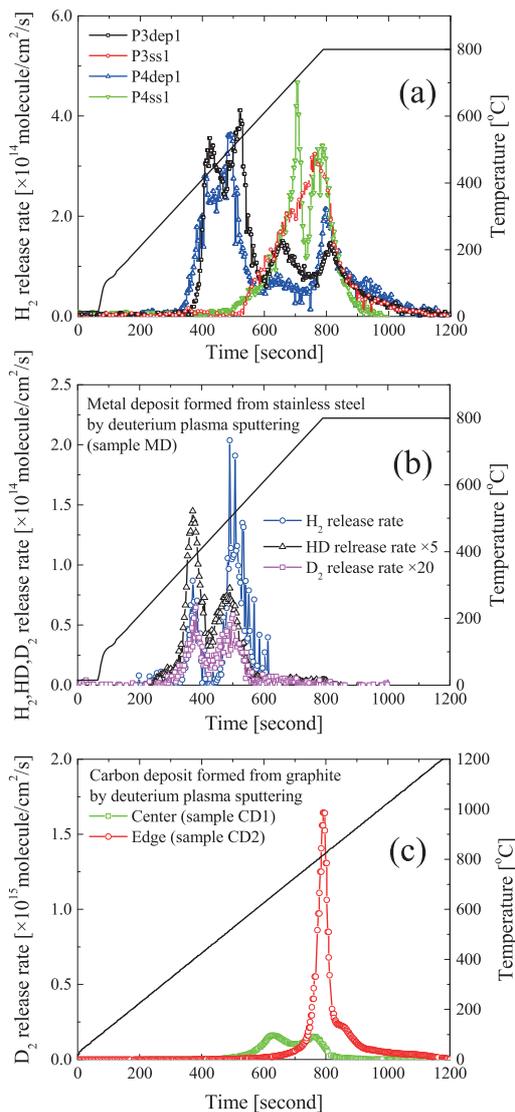
Fig. 7 H$_2$ or D$_2$ release behaviors from the samples installed in LHD (a), metal deposit formed from 316SS (b), and carbon deposits formed from graphite (c).

Fig. 8 The signal intensity from a thermal conductivity detector in the gas chromatograph with the retention time for combustion of sample P4ss2.

drogen release amount although MD was formed by deuterium plasma sputtering. The similar phenomenon for tungsten sputtering-deposition was discussed in the previous paper but the mechanism is not enough clarified yet [12]. It was found that two release peaks of hydrogen isotopes from MD appears at 370°C and 500°C. Deuterium release from CD1 has two broad peaks at 600°C and 800°C. From CD2, one sharp peak appeared at 800°C. This suggests that hydrogen isotope retained in carbon deposition is approximately released in the temperature range between 600°C and 800°C, depending on deposition condition. From comparison of release peaks, hydrogen released from P3ss1 and P4ss1 can be concluded to be retained in the carbon adhering to the substrate. The hydrogen release from P3dep1 and P4dep1 at the temperature range from 400°C to 600°C indicates that hydrogen incorporated into metal deposit during the sputtering-deposition process can remain in the deposit under high vacuum con-
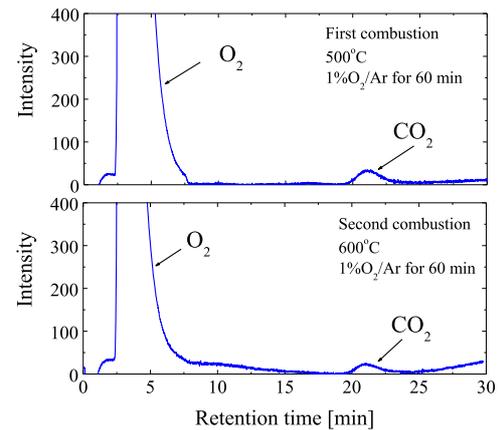
dition for a long period. The amounts of hydrogen released from P3dep1 and P4dep1 above 600°C, which is retained in carbon, were smaller than that from P3ss1 and P4ss1. A part of hydrogen released from metal deposit would react with carbon adhering on the metal deposit and be released as hydrocarbon, although signal of m/e = 16 was under the detection limit of a quadruple mass spectrometer because of low sensitivity.

As an example, the change of the signal intensity from TCD with the retention time are shown in Fig. 8, where P4ss2 was heated. A first large peak is attributed to O$_2$ and a second peak is attributed to CO$_2$. For three samples, P1ss2, P3ss2 and P4ss2, the amount of CO$_2$ generated in the second heating was approximately consistent with the amount of CO$_2$ generated by the reaction of fresh SS316 substrate with oxygen. This indicated that almost all carbon adhering to the sample was converted to CO$_2$ in the first heating. From the quantified amount of CO$_2$, the amount of carbon adhering to the surface of P1ss2, P3ss2 and P4ss2 were estimated to be 1.28, 1.73 and 2.09 μg, respectively. When it is assumed that carbon is adhered on the bare surface area of the substrate, which is approximately 0.90 cm^2, the amounts of carbon per cm^2 are 1.42 μg/cm^2 on P1ss2, 1.92 μg/cm^2 on P3ss2 and 2.32 μg/cm^2 on P4ss2, respectively. It can be said that about 2 μg/cm^2 of carbon adhered on the metal wall of LHD exhaust duct during 13th experimental campaign.

The density of hydrogen rich carbon films formed on the remote area of the inner JET divertor has been reported to be about 1.0 g/cm^3 [13]. When the density of carbon adhering to the substrates is assumed to be 1.0 g/cm^3, the thickness of carbon deposition on the SS316 substrates is estimated to be about 20 nm. This value is significantly smaller than the thickness of co-deposited carbon film (the maximum thickness is about 400 nm) produced in the vicinity of LHD divertor tiles on the material probe during the 13th experimental campaign [7]. Here, the atomic

ratio of hydrogen to the other deposited elements (carbon, boron and iron) were shown to be in the range from 0.1 to 0.6. From the estimated amounts of carbon on P3ss2 and P4ss2, and from the released amount of hydrogen from P3ss1 and P4ss1, H/C ratios in the deposit on the duct were obtained to be 1.52 and 1.04, respectively. These values are larger than the H/C in co-deposited carbon film produced in the vacuum vessel. This result suggests that the H/C in the precursor of the deposit formed in the exhaust duct is higher than that formed in the vacuum vessel. From the rough estimation, the surface area of metal wall in the duct region shown in Fig. 1 was obtained to be approximately $50\,m^2$. Therefore, carbon accumulation in this region is estimated to be about 1 g and hydrogen accumulation is estimated to be 0.043 - 0.63 mol-H_2. When carbon adhesion in the roughing vacuum pump region following the region investigated in this work is taken into consideration, total amounts of carbon and hydrogen accumulation in whole exhaust system would increase more.

When H_2 is replaced to T_2 by using a conversion factor of 2.15×10^{15} Bq/mol-T_2 without considerations of isotope effect and isotope mixing ratio in order to approximately evaluate tritium accumulation on the exhaust duct in a fusion reactor, the amount of tritium in this region is about 100 TBq and 200 MBq/cm^2. This amount is not ignorable from a viewpoint of tritium safety management although its amount would be significantly smaller than that in the vacuum vessel.

4. Conclusions

Carbon is uniformly accumulated with hydrogen on the metal wall of exhaust duct of LHD although its amount is significantly smaller than that in the vacuum vessel. The amount of carbon adhering to the metal wall is about $2\,\mu g/cm^2$ and H/C ratio is 1~1.5. When total surface area of the duct wall in the region investigated is assumed to be $50\,m^2$, carbon accumulation in the 13th campaign is about 1 g and hydrogen accumulation is about 0.043~0.063 mol-H_2.

Hydrogen incorporated into the metal deposit formed from SS316 in the sputtering-deposition device in the laboratory can remain in the deposit under high vacuum condition in the exhaust duct for a long period.

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