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Tritium decontamination of TFTR carbon tiles employing ultra violet light

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Abstract

Tritium decontamination on the surface of Tokamak Fusion Test Reactor (TFTR) bumper limiter tiles used during the Deuterium–Deuterium (D–D) phase of TFTR operations was investigated employing an ultra violet light source with a mean wavelength of 172 nm and a maximum radiant intensity of 50 mW/cm^2. The partial pressures of H_2, HD, C and CO_2 during the UV exposure were enhanced more than twice, compared to the partial pressures before UV exposure. In comparison, the amount of O_2 decreased during the UV exposure and the production of a small amount of O_3 was observed when the UV light was turned on. Unlike the decontamination method of baking in air or oxygen, the UV exposure removed hydrogen isotopes from the tile to vacuum predominantly in forms of gases of hydrogen isotopes. The tritium surface contamination on the tile in the area exposed to the UV light was reduced after the UV exposure. The results show that the UV light with a wavelength of 172 nm can remove hydrogen isotopes from carbon-based tiles at the very surface. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Tritium; Carbon; Waste

1. Introduction

The in situ tritium decontamination is important in controlling resident in-vessel tritium inventory in future deuterium–tritium fusion reactors where carbon tiles are used as plasma facing materials. Tritium used as fusion reactor fuel will accumulate by co-deposition within the vacuum vessel in regions that are not undergoing active erosion by the interaction with plasma [1–4]. Some techniques have been proposed as possible in situ methods to remove tritium from co-deposited layers. These include: baking in vacuum or air/oxygen, abrasive methods such as CO_2 blasting and liquid nitrogen jets, ion cyclotron resonance-discharge cleaning (ICR-DC) and electron cyclotron resonance-discharge cleaning (ECR-DC), helium glow discharge cleaning (He-GDC), isotope exchange with deuterium plasma, deuterium soaking with heated walls, reaction with gaseous radicals (ozone), surface heating to more than 2200 K with continuous wave CO_2 laser, etc. [1]. However, there exist needs to develop more effective and more convenient techniques. Recently, various ultra violet (UV) sources (lamp and laser) have been developed and used to clean up material surfaces in some semiconductor industries and metal surfaces contaminated by tritium in a few tritium facilities [5–7].

Using an ultra violet light source, tritium decontamination of TFTR D–D tiles was investigated at PPPL under the collaborative program between JAERI and US-DOE. In this study, the D–D tiles were exposed to the UV light in a vacuum chamber and the released gases were analyzed using a quadrupole mass spectrometer (QMS) configured to the chamber. The tritium concentrations on D–D tiles before and after the UV exposure were measured with an open wall ion chamber. The effect of the combination of heating the tiles and UV exposure upon tritium decontamination was also investigated.

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2. Experimental

As depicted in Fig. 1, the experimental apparatus consists of a vacuum chamber, a UV light, a QMS, a turbo molecular pump (TMP) and a roughing pump, which was connected to the tritiated vent system of TFTR. The chamber has a sapphire window that minimizes UV light absorption thus allowing greater than 70% of UV light to enter the chamber and interact with the surface of the tile. The ultra violet light source is a xenon excimer lamp, which produces UV lights with a mean wavelength of 172 nm (corresponding to a photon energy of about 7.1 eV) and a maximum radiant intensity of 50 mW/cm^2. The photon energy of the UV light is high enough to cut the chemical bonds between carbon and hydrogen isotopes in the tiles. For instance, the bonding energy between CH_3 and H is 4.5 eV [8].

Three D–D tiles, R-B-12, G-D-13 and S-A-12, (which are numbered from positions where the tiles had been located in the TFTR vacuum vessel) were used for the UV exposure. About 35% of the tritium produced during the TFTR D–D operation phase remained in the internal vacuum vessel components [9]. Prior to placing the tile into the vacuum chamber for UV light exposure, the surface of the tile was analyzed with an open wall tritium monitor at a detection limit of 4×10^4 Bq/m^2. The pressure of residual gases is kept at about 2×10^{-3} Pa prior to and after the UV exposure. During the exposure to UV light the released gases were analyzed using the QMS attached to the chamber. After the UV exposure the tritium concentration in the same positions on the surface of the D–D tiles was measured again with the open wall ion chamber. A heater was attached to the bottom of the tile, and the effects of the combination of heating the tiles up to 400 K and UV exposure upon tritium decontamination were investigated.

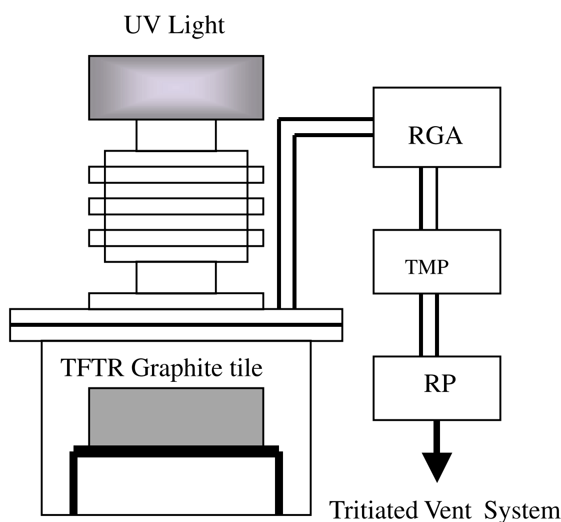

Fig. 1. Schematic representation of UV decontamination apparatus.

3. Results and discussion

3.1. Released gases during UV exposure

The released gases from the tile S-A-12 during the UV exposure were compared with the residual gases in the vacuum chamber prior to exposure. As shown in Fig. 2, the partial pressures of H_2 (mass number: 2), HD (mass number: 3), C (mass number: 12) and CO_2 (mass number: 44) were enhanced more than twice during a 10 min exposure. At that time, the partial pressure of CO (mass number: 28) increased by a small amount. In comparison, the partial pressure of O_2 decreased during the UV exposure. The production of a small amount of O_3 (mass number: 48) was also observed when the UV light was turned on. The similar changes of mass spectra were observed also in the UV exposure of the tile G-D-13.

The decontamination mechanism seems to be two photosensitized processes, namely desorption process and decomposition process. In the desorption process, adsorbed contaminants (CO, CO_2 and H_2O are excited and/or dissociated by the absorption of the UV light with a short wavelength. Simultaneously, in the decomposition process some hydrocarbons are decomposed into hydrogen and free carbon by cutting the

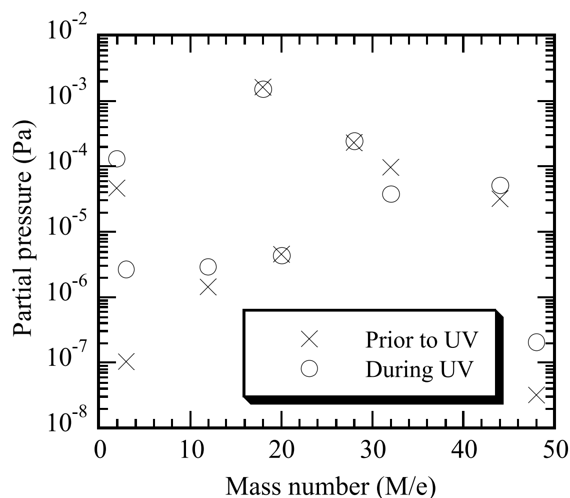

Fig. 2. Comparison between the quadrupole mass spectrum of gases released from the tile S-A-12 during the UV exposure and that of the residual gases in the vacuum chamber prior to exposure ($M/e = 2, H_2$; $M/e = 3$, HD; $M/e = 12$, C; $M/e = 18$, H_2O; $M/e = 20$, D_2O; $M/e = 28$, CO; $M/e = 32$, O_2; $M/e = 44$, CO_2; $M/e = 48$, O_3).

bonds between H (or D) and C. Almost all atoms of the hydrogen isotopes liberated by the UV light react with each other to form molecules of hydrogen isotopes but there are still a few atoms of hydrogen isotopes that react with free carbon or atomic oxygen (atomic oxygen is generated when molecular oxygen is dissociated by the UV light) to form hydrocarbon or water. As the partial pressure of O_2 decreased during the UV exposure, molecular hydrogen is considered not to be produced from the photolysis of water. Hydrogen isotopes absorbed on top of the surface and bound to carbon atoms in one or several monolayers below the surface were released by the UV exposure.

Chiu and Haasz [4] analyzed the reaction products formed by exposing an a-C:D film to $^{18}O_2$ at 470 K, and concluded that essentially all of the deuterium is removed via D_2O formation (no D_2 was observed) and C is removed with the formation of $C^{18}O$ and $C^{18}O_2$ by the method of baking in oxygen. Sowell et al. [5] found that exposure of a metal or ceramic surface to UV radiation for a prolonged time in air or oxygen removes adsorbed hydrocarbon contaminants. Vig [6] argued that the combination of short wavelength UV light plus ozone produces a clean surface substantially faster than either short wavelength UV light without ozone or ozone without UV light. Krasznai and Mowat [7] determined the relative efficiency of UV and ozone in removing tritiated species from the surface of stainless steel by substituting nitrogen gas for oxygen. Their results showed that the tritium decontamination effectiveness was reduced from 93–94% to 31–46% when oxygen was replaced with nitrogen.

Unlike the method of baking in air or oxygen and the technique of UV exposure in air or oxygen mentioned above, the UV exposure in vacuum of this work removes hydrogen isotopes from the tile predominantly in the form of gases of hydrogen isotopes. This feature of the UV exposure technique is very attractive from the point of view of both radiation safety and tritium processing, because the radiation hazard of tritiated water is four orders greater than that of tritium gas and tritium recovery process from hydrogen isotope gases is much simpler than that from water containing tritium.

3.2. Surface tritium concentration and heating effects

The tritium concentrations in seven positions of the surface of the tile G-D-13 were measured in the atmosphere with an open wall ion chamber at the detection limit of 4×10^4 Bq/m^2 prior to placing the tile into the vacuum chamber for UV exposure. After the UV exposure for 50 min, the tile was removed from the chamber and analyzed again in atmosphere with the open wall ion chamber. As shown in Fig. 3, the tritium concentration in position 7 which experienced the intense UV exposure (50 mW/cm^2) showed the largest decrease due to the UV exposure. The concentration decrease after the UV exposure was observed also for tiles R-B-12 and S-A-12.

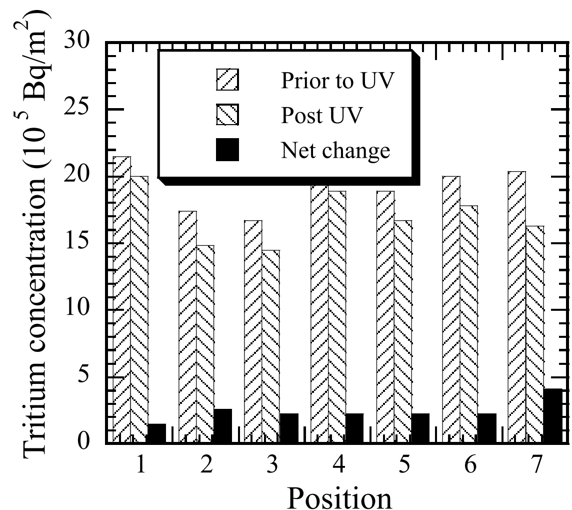

Fig. 3. The tritium concentrations (prior to UV exposure, after UV exposure and net change) in seven positions on the surface of the tile G-D-13.

The largest decrease in the ion chamber reading was only 20%. However, this does not mean that the tritium on the surface dropped by 20%. The reason is that UV light can remove tritium on surface (only one or a few monolayers) and that ion chamber counts not only the tritium on the surface but also the β-ray (tritium) in the area whose depth is a few micrometers.

An empirical formula for the maximum range R corresponding to the maximum energy E (0.01 MeV $< E <$ 2.5 MeV) of β-ray was given by [10]

$$R = 0.412\ E^{1.265-0.0954 \ln E}, \quad (1)$$

where R in g/cm^2 and E in MeV.

The maximum range for tritium was calculated to be 5.86 g/m^2 corresponding to the maximum energy of 0.0186 MeV for β-ray of tritium decay. Considering that the density of carbon materials ranges from 1.7 to 2.3 g/cm^3, the maximum depth where β-ray of tritium decay can reach the surface of carbon tiles is 2–3 μm. In addition, only 50% of the β-rays emitted at this maximum depth can reach the surface and enter the ion chamber, the other β-rays move away from the surface.

For the tile G-D-13, the effects of the combination of heating and UV exposure was investigated. The tile was heated up to 400 K in atmosphere with a heater attached to the bottom of the tile. The tile was then cooled rapidly down to room temperature in a few minutes by purging with argon. The surface of the tile was then analyzed for the tritium concentration at seven positions with the ion chamber. Subsequently, the tile was placed into the

Table 1
Ion chamber readings of the tile G-D-13 (10^6 Bq/m^2)

Position #	Before heat	Post heat	Post UV
1	2.18	2.22	2.18
2	1.63	1.74	1.63
3	1.63	1.78	1.63
4	2.04	2.18	2.00
5	1.78	1.89	1.67
6	1.89	1.96	1.81
7	1.78	2.00	1.81

vacuum chamber and exposed to the UV light in vacuum. As shown in Table 1, the tritium concentrations in different positions increased by 4×10^4 to 2.2×10^5 Bq/m^2 after heating the tile to 400 K, while they dropped to almost the same values as those before heating, after the tile was subjected to UV exposure.

The above results imply that a small amount of tritium moved to the surface during the heating and was removed during the UV exposure. However, tritium in the layers a few micrometers deep cannot be removed by the UV light as indicated by the ion chamber measurements. In order to remove the tritium from the layer of a few micrometers, a more powerful UV source is required. A new UV laser (Lambda Physik, COMPex 102) has been installed in JAERI/TPL (Tritium Engineering Laboratory, Japan Atomic Energy Research Institute). Although the wavelength of the UV laser (193 nm) is a little bit longer than the UV light (172 nm), the largest pulse energy is about 250 mJ with a pulse duration of 25 ns. The preliminary experiment indicates that an a-C:D film (a few micrometers thick) formed on stainless steel substrate by C_2D_2 glow discharge was removed by the UV laser within a minute (300 pulses). This UV laser is being shipped to PPPL to continue in the development of the tritium decontamination experiments.

4. Conclusion

Three TFTR D–D tiles were exposed to UV light with a photon energy of about 7.1 eV, which is sufficiently high to cut C–H bonds. The gases released to a vacuum chamber during UV exposure were analyzed with a QMS and the change in tritium concentration on the surface was measured with an open wall ion chamber. During the UV exposure, hydrogen and deuterium were liberated from the tile surface. Unlike the method of baking in air or oxygen, the UV exposure removed hydrogen isotopes from the tile to vacuum predominantly in the form of gases of hydrogen isotopes. The largest decrease in the ion chamber reading was only 20%. However, this does not indicate that the tritium on the surface dropped by 20% considering that the maximum depth where β-rays of tritium decay can reach the surface of carbon tiles (2–3 μm). A small amount of tritium moved to the surface during heating of the tile to 400 K, and was removed during the subsequent UV exposure. A more powerful UV source (laser) is required to remove the tritium from the layer of a few micrometers.

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