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Journal of Nuclear Materials 290–293 (2001) 469–472

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

A study of tritium decontamination of deposits by UV irradiation

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Abstract

To develop a new method for decontamination of tritium retained in the carbon fiber composites (CFC) tiles and other components of D–T fusion reactors, a preliminary experiment using ultra violet (UV) light source was performed. Samples simulating a co-deposited layer were prepared by glow discharge using C_2H_2 and a tungsten or CFC substrate. The UV light from a xenon excimer lump (172 nm) was irradiated to the samples in 423 K. Small amounts of species were released only by the heating procedure to 423 K. By UV irradiation, large quantities of hydrogen, carbon and hydrocarbons were released from the samples. Fourier Transform Infrared (FT-IR) analysis showed hydrocarbons were formed on the sample by acetylene glow discharge and C–H bonds were decomposed by irradiation with UV light. It is concluded that the combination of heating and UV irradiation causes release of tritium from the surface of the materials.

Keywords: Carbon; Tritium; Waste

1. Introduction

In D–T nuclear fusion reactors using carbon materials as plasma-facing components, the hydrocarbons produced by chemical or physical sputtering of the surface of the carbon materials are accumulated and co-deposited layers are formed. Formation of the co-deposited layers leads to high tritium inventories in the vacuum chamber. Though baking at the low pressure of oxygen [1,2] was proposed as a tritium recovery method from co-deposited layers, this is not the best decontamination method because of the production of moisture with high radioactivity and organic materials by beta ray-induced reactions [3,4]. Since it is desirable not to produce tritiated water, an ideal decontamination method is to recover the tritium in gas form. Ultra violet (UV) irradiation [5–8] whose energy is high enough to dissociate the C–H bond [9], is considered as one of the best ways for tritium decontamination. In this study, a preliminary decontamination experiment was performed.

2. Experimental

2.1. Preparation of substrates

Tungsten substrates as base materials for the preparation of carbon films simulating co-deposited layer were purchased at Nilaco. The size of the substrate was 10.0 mm in diameter and 1.0 mm in thickness. The co-deposit preparation procedures were as follows: The tungsten plate was polished with a SiC polishing paper and washed with acetone in an ultrasonic bath. As another substrate material, the carbon fiber composite: CFC (CX-2002U supplied by Toyotanso) was used. The size of the CFC plate was 25 mm in diameter and the thickness was 1.0 mm in thickness.

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PII: S0022-3115(00)00440-2

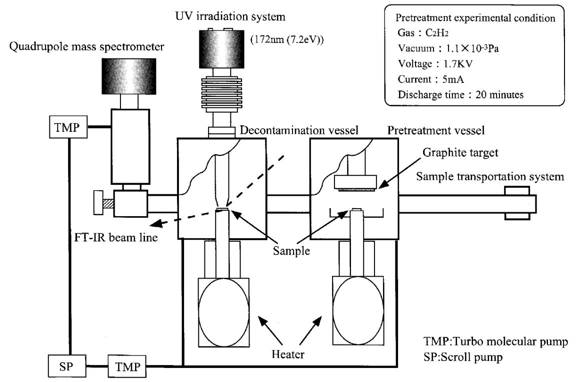

Fig. 1. Schematic view of the experimental system.

A schematic view of the experimental apparatus is illustrated in Fig. 1. Two vacuum chambers, one for the pretreatment and the other for decontamination study were used. For the pretreatment, the sample was introduced and placed in the center of the pretreatment chamber, which was evacuated to less than 10^{-3} Pa. The glow discharge of C_2H_2 was carried out to simulate co-deposition of hydrogen and carbon on the tungsten and CFC. The glow discharge voltage was 1.7 kV and the current was 5 mA. The pressure was kept at about 10 Pa during glow discharge by controlling the C_2H_2 gas flow. The glow discharge was performed for 20 min.

2.2. *UV irradiation experiment*

After the glow discharge, the sample was transferred into the chamber for decontamination treatment. The chamber was evacuated to less than 10^{-4} Pa by turbo molecular pump. The Polarization Modulation Reflection Infrared Spectroscopy (PMRIRS) using a Fourier Transform Infrared (FT-IR) spectrometer (Mattson Instruments.: Infinity GOLD) was applied to the analysis of the chemical states of hydrogen and carbons by comparing the spectra before and after UV irradiation. The infrared rays were directed on the sample at an angle of 80° to the surface normal. The typical spot size of the IR ray on the surface of the sample was about 3×10 mm^2 along the axis of the ray.

The specimen was heated from room temperature to 423 K at a ramp rate of 10 K/min. The temperature was kept at 423 K during the UV irradiation experiment. The xenon excimer lamp (Ushio.: UER20H-172 (wavelength: 172 nm)) was used as the UV light source. The UV source, corresponding to 7.2 eV, has enough energy to dissociate C–H bond (about 4.5 eV [9]). UV irradiation was carried out for 15 min. During heating and UV irradiation, the exhausted gases from the chamber were analyzed continuously by a quadrupole mass spectrometer, MICROVISION PLUS (SPECTRA International).

3. Results and discussion

3.1. *Mass analysis of evolved gases from deposit samples*

Fig. 2 shows the mass spectra of the released gases from the film deposited on the tungsten substrate before and during the UV irradiation at 423 K. Some peaks, namely, hydrogen, water, carbon monoxide and carbon dioxide, were observed with intensities much higher than those before UV irradiation. To check the effect of UV irradiation, the differences of mass intensities before and during UV irradiation were obtained by subtraction and are illustrated at the bottom of Fig. 2. Release of gases at *m/e* 2, 18, 28 and 44 were enhanced by the UV irradiation. The mass peak 18 can be assigned to water, according to the cracking pattern of water at *m/e* 16 and 17. The gas species of the mass numbers 28 and 44 can be assigned to carbon monoxide and carbon dioxide. It seems that the adsorbed particles on the surface of the substrate would be desorbed by UV

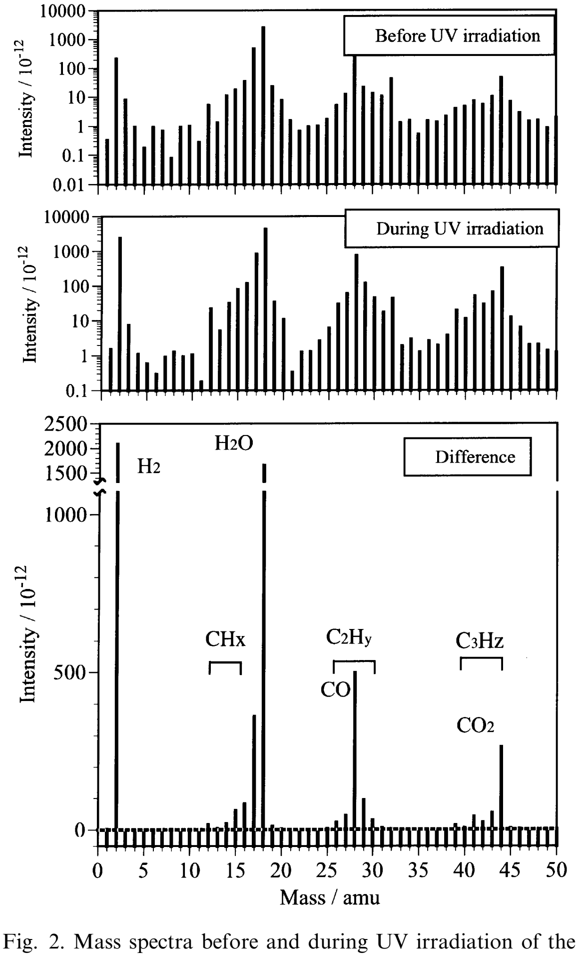

Fig. 2. Mass spectra before and during UV irradiation of the acetylene discharge deposit on tungsten.

irradiation. However, large amounts of hydrogen, mass number 2, were released from the specimen. This fact cannot be explained only by the desorption of hydrogen adsorbed on the surface. In addition, some observable differences at the peaks, such as 26, 27, 29, 41, 42 and 43 were also observed. They indicate that some minor contributions to the peak intensities around mass numbers, 28 and 44 can be attributed to the C_2H_x ($x = 2$–4), C_3H_y ($y = 4$–8) hydrocarbons evolved. The time evolution of mass numbers 2 and 12 are displayed in Fig. 3. The bottom graph shows the temperature change during the experiments. The UV light was turned on at 23 min after the experiment was started. There was no significant release of m/e 2 and 12 observed during only heating up to 423 K. After reaching up to 423 K, all the mass intensities went back to the initial intensities. When the UV light irradiation to the substrate started, the mass intensities were increased suddenly. This result suggests the UV light decomposes the accumulated hydrocarbons on the substrate. By shutting off the UV light, the mass intensities dropped to the initial level.

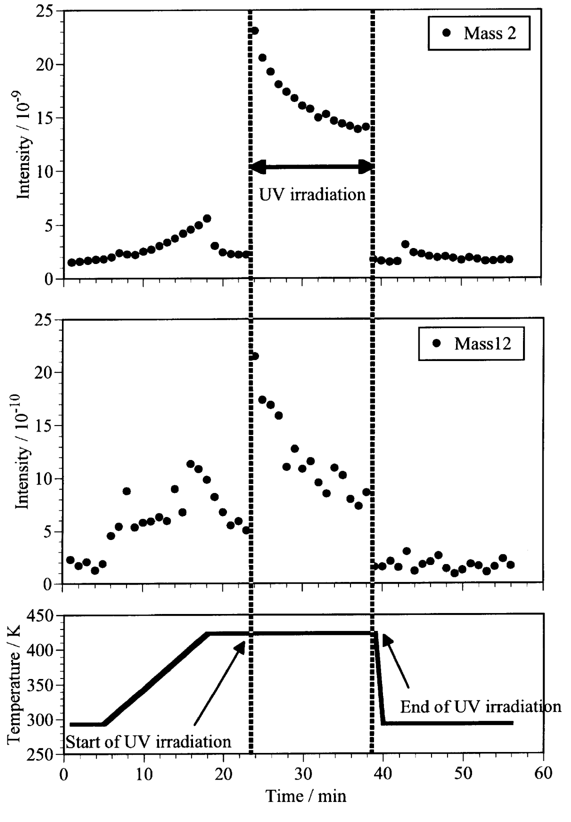

Fig. 3. Time evolution of m/e 2 and 12 during heat treatment and exposure to UV light for the acetylene discharge deposit on tungsten.

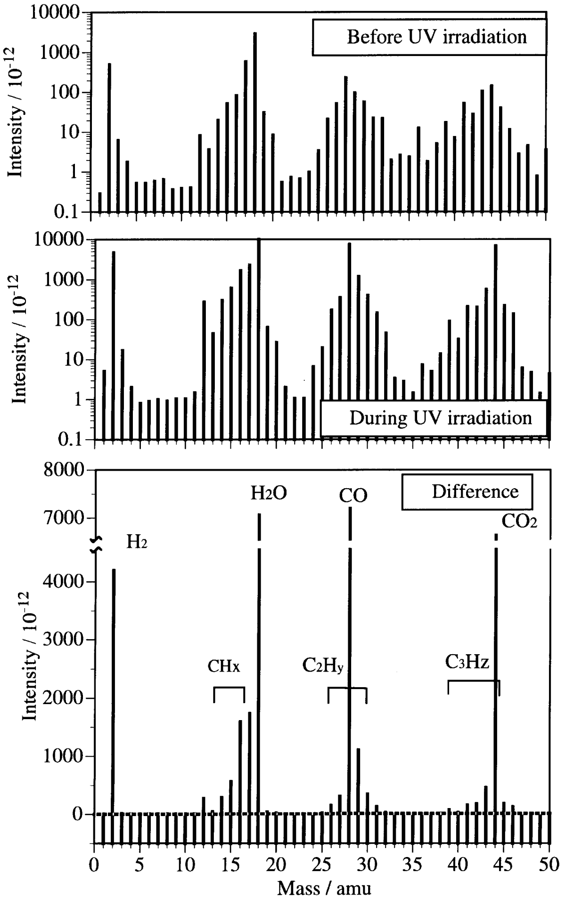

Fig. 4. Mass spectra before and during UV irradiation of the acetylene discharge deposit on CFC.

Fig. 4 shows the mass spectra of released gases from the deposited film on CFC graphite substrate before and during the UV irradiation. The results of subtraction of intensities between before and during UV irradiation are shown at the bottom of Fig. 4. The major released species are hydrogen, water, carbon monoxide and carbon dioxide. The spectra obtained from the deposited film on CFC substrate were the same as that of the spectra of deposited film on the tungsten substrate. Some weak species around mass numbers, 16, 28 and 44 can be assigned to hydrocarbons as mentioned in the case of tungsten. From these results, the component of the deposited layer on tungsten is almost as same as that of the deposited layer on CFC.

3.2. FT-IR analysis of tungsten substrate

Fig. 5 shows the FT-IR spectra of the film deposited on the tungsten substrate. The thin line and the bold line denote the spectrum before and after the UV irradiation,

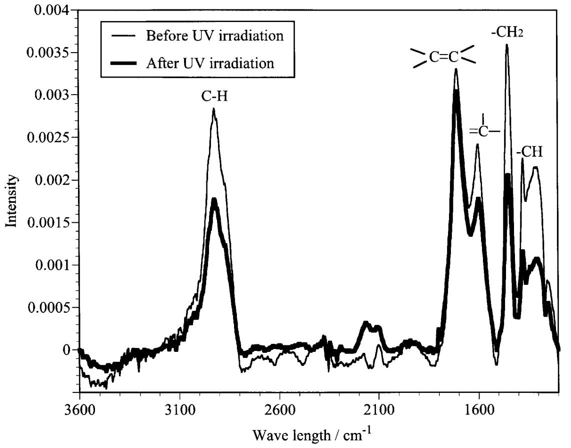

Fig. 5. FT-IR spectra of the acetylene discharge deposit on tungsten before and after UV irradiation.

respectively. The peak at around 2900 cm^{-1} can be attributed to C–H stretching. The peaks below 1800 cm^{-1} were assigned to amorphous carbon structure. The peak at 1450 cm^{-1} can be assigned to degenerated deformation vibration of saturated hydrocarbons and that at 1380 cm^{-1} corresponds to the symmetric deformation vibration of saturated hydrocarbons, respectively. The peak at 1680 cm^{-1} can be attributed C=C stretching. The peak intensities of 1450 and 1380 cm^{-1} were decreased by UV irradiation and it is clear that the C–H bonds (1450 and 1380 cm^{-1}) were broken by UV irradiation. But the C=C bonds (1680 cm^{-1}) of unsaturated hydrocarbons corresponding to 7.24 eV remained unaffected compared with C–H bonds after the UV irradiation in Fig. 5. In our previous study [8], the dissociation efficiency of the C–H bond was similar to that of the C–Cl bond. The chemical bond whose energy is much lower than UV energy, would be broken with the same efficiency by UV irradiation. The energy of C=C bonds is slightly larger than that of an UV photon and the bond cannot be dissociated practically. This result implies that the UV whose energy is 172 nm, may dissociate the hydrogen only and the hydrocarbons would not be produced so much. It is suitable for decontamination of tritium because the UV light only cut the C–H bonds with larger efficiency comparing with the C=C bonds.

4. Conclusion

To develop tritium decontamination technique without the production of tritiated water, some preliminary UV irradiation experiments were performed. The deposited layers on tungsten and CFC were obtained using a glow discharge in gaseous C_2H_2 to study the decontamination property of UV irradiation. UV irradiation experiments were then performed and the released gas analyzed with a mass spectrometer. After some amounts of gas species were released by heating up to 423 K, large amounts of hydrogen, carbon, carbon monoxide and carbon dioxide were released during UV irradiation. By the FT-IR analysis, it was found that the C–H bond was broken and the co-deposited hydrogen bounded to the carbon on the surface was evolved by the UV irradiation. It is concluded that the UV decontamination method can be one of the effective ways to decontaminate tritium from the surface of the materials without heating over 513 K, which temperature corresponds to the current design baking temperature of ITER. The combination of heating and UV irradiation seems to be the best way for tritium decontamination.

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