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Ablative removal of codeposits on JT-60 carbon tiles by an excimer laser

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Abstract

The codeposits on JT-60 tiles experienced hydrogen plasma burning were irradiated by focused beams of an excimer laser. The removal rate of the JT-60 codeposits was low when the laser energy density was smaller than the ablation threshold (1.0 J/cm^2), but reached to 1.1 μm/pulse at the laser energy density of 7.6 J/cm^2. The effective absorption coefficient k in the JT-60 codeposits at ArF excimer laser wavelength was determined to be 1.9 μm^{-1}, which is almost one order smaller than the optical absorption coefficient at the same wavelength in graphite (16.4 μm^{-1}). In the process of ablative removal of the codeposits, hydrogen was released predominantly in the form of hydrogen molecule and water formation could be ruled out. The temperature rise on the surface was measured on the basis of Planck's law of radiation, and the temperature during the irradiation at the laser energy density of 0.5 J/cm^2 decreased from 3570 K at the beginning of the irradiation to 2550 K at 1000th pulse of the irradiation.

Keywords: Plasma-wall interactions; Laser ablation; Laser application; Carbon; Tritium; Codeposits

1. Introduction

Tritium will be retained inside the ITER vacuum vessel mainly by codeposition with carbon eroded from carbon fiber composite (CFC) tiles [1–6]. CFC is chosen because it sublimes during disruption thermal quenches, thereby avoiding melting and generation of surface irregularities that might later form hot spots in normal steady-heat flux operation. With this material selection, however, the codeposition of tritium may severely limit the operational availability of ITER. Thus, it is a pressing issue to develop efficient techniques for codeposit removal [1]. An irradiation technique has been developed in JAERI for this purpose by applying an ArF excimer laser [7,8]. In this study, the irradiation effect of the excimer laser was investigated using codeposits on JT-60 carbon tiles that had experienced hydrogen plasma burnings. The removal rate of the codeposits was measured as a function of the laser energy density, and the releases gases during the irradiation and the temperature rise on the surface were analyzed.

2. Experimental

Three experiments were performed using an excimer laser, which operates on ArF to produce laser beam with a wavelength of 193 nm (corresponding to a photon energy of about 6.3 eV), a repetition rate of 5 or 20 Hz, a pulse duration of 25 ns and a beam size of 23 mm by 7 mm. The samples used were codeposits on carbon tiles that had experienced hydrogen plasma burnings in JT-60.

In the first experiment, the relationship between the removal rate of the codeposits and the laser energy

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density was investigated. The laser beam was focused with a lens and the focused beam was irradiated to different locations of codeposits on a JT-60 graphite tile. The laser energy density at each position was changed by adjusting the distance of the surface from the focus of the lens. At the laser energy density of 2.0–7.6 J/cm^2 the irradiation lasted for 50 pulses, while 1000 pulses were irradiated at the laser energy density of 0.1–1.0 J/cm^2. The eroded depth was measured after the laser irradiation with a CCD laser displacement meter which sampled a distance of 10 mm with a resolution of 0.1 μm.

In the second experiment, the released gases during the laser irradiation were measured with a quadrupole mass spectrometer (QMS). A cube cut from a JT-60 tile was placed into a vacuum chamber, in which a QMS and a quartz window were installed. The background pressure of the QMS was kept at about 6×10^{-7} Pa by a small orifice and the molecular turbo pump. The plasma facing surface of the cube was irradiated at the laser energy density of 2.0 J/cm^2 by a focused laser beam going through the quartz window.

Additionally, in the third experiment, the temperature rise on the surface during the laser irradiation was measured on the basis of Planck's law from the infrared (IR) radiation signals. The laser beam was focused with a lens and then irradiated onto the JT-60 tile at an angle of 45° to the surface normal. The laser energy density on the surface of the JT-60 tile was changed by adjusting the distance of the surface from the focus of the lens. The IR radiation was collected with a lens at an angle of 90° to the laser beam and the IR signals going through a filter was detected by an InGaAs photo sensor with a response time of 3 ns and a fast sampling oscilloscope (5 GS/s).

3. Results and discussion

3.1. Removal rate of the codeposits

When the laser energy density is larger than the ablation threshold, the ablative removal rate (μm/pulse) R can be expressed, using Beer's law, by

$$R = d \ln(E/E_{th}), \quad (1)$$

where d is the laser penetration depth in μm (inverse of the laser absorption coefficient k, i.e., $d = k^{-1}$), and E and E_{th} are laser energy density and the ablation threshold, respectively.

The averaged removal rate of the JT-60 codeposits was plotted against the laser energy density. As shown in Fig. 1, the removal rate was very small when the laser energy density was smaller than 1.0 J/cm^2, but a rapid increase was observed for the laser energy density larger than 1.0 J/cm^2. The removal rate of the JT-60 codeposit

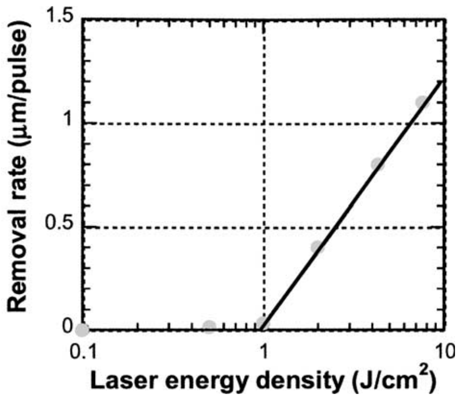

Fig. 1. The relationship between the removal rate of JT-60 codeposits and the laser energy density.

increased from 0.4 to 1.1 μm/pulse as the laser energy density increased from 2.0 to 7.6 J/cm^2. From Eq. (1) and Fig. 1, the effective absorption coefficient k of the JT-60 codeposit for the excimer laser was determined to be 1.9 μm^{-1}, which is almost one order smaller than the absorption coefficient of graphite at 193 nm (16.4 μm^{-1}) [9]. This difference should be caused by the significantly different structure of an amorphous codeposit compared to that of graphite. In contrast, the threshold of laser energy density for ablation of the JT-60 codeposit irradiated by the excimer laser (193 nm) is obtained from Fig. 1 to be 1.0 J/cm^2, which is quite close to that of graphite (1.12 J/cm^2) [9].

3.2. Released gases during the laser irradiation

For the sample of JT-60 tile irradiated in vacuum, the released gases during the laser irradiation at the laser energy density of 7.6 J/cm^2 were analyzed by a QMS. The net increases of partial pressures during the laser irradiation were plotted against the mass number (M/e) in Fig. 2. Hydrogen showed the maximum value of the partial pressure in the released gases, and the partial pressure of H_2 during the laser irradiation (1.33×10^{-6} Pa) was enhanced 66.5 times in comparison with that before irradiation (2.00×10^{-8} Pa). On the contrary, the partial pressure of water (H_2O) during the laser irradiation (5.77×10^{-7} Pa) was only 1.5 times of that before irradiation (3.85×10^{-7} Pa). Water should be absorbed in the codeposits during the storage (long exposure to air). This suggests that the excimer laser irradiation in vacuum removes hydrogen isotopes from the codeposits predominantly in the form of molecules of hydrogen isotopes. This feature of the laser ablative technique is very attractive from the viewpoint 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

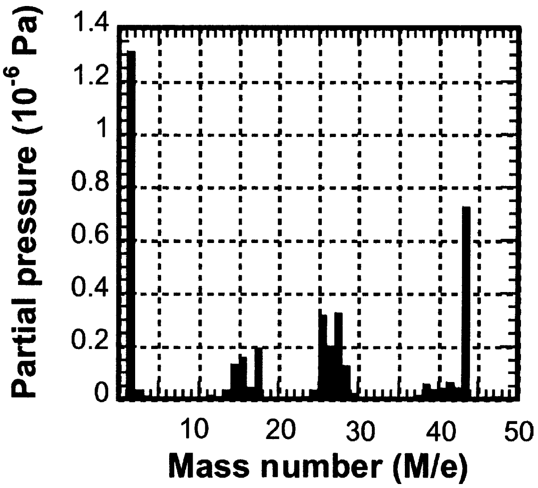

Fig. 2. The partial pressures of gases released during the irradiation.

hydrogen isotope gases is much simpler than that from tritium oxide.

Besides hydrogen, mass number of 44 (CO_2 or C_3H_8) also showed a large increase in the value of the partial pressure (from 1.87×10^{-8} Pa before the irradiation to 7.43×10^{-7} Pa during the irradiation). Since the ratio of partial pressure (39.7) during the irradiation against that before irradiation for mass number 44 was much greater than that (1.87) of mass number 43 (C_3H_7), the main part of the partial pressure for the mass number 44 could be attributed to CO_2. In other words, carbon was predominantly released in the form of CO_2 during the laser ablation. This implies that carbon dissolved from hydrocarbon recombines predominantly with oxygen dissolved from water in the laser ablation. In addition, since the increase in the partial pressure was observed during the irradiation for the mass numbers of 28, 26, 16, ... some carbon was released in the form of hydrocarbons like (C_2H_4, C_2H_2, CH_4, ...). This suggests that a small portion of codeposits removed during the laser ablation would re-deposit on the surfaces of the vacuum chamber. The re-deposition was examined by surrounding the sample with a small aluminum box where a slit was opened for the laser beam, and visible re-deposition was not observed after the laser ablation.

The results of time-of-flight mass spectrometric measurements of the plume dynamics of laser ablation of graphite [10] showed that C^+ was the major ionic species at the laser energy density of 0.9–2.8 J/cm^2 and the wavelength of 266 nm, and that the major neutral species (C and C_3) were ionized by ArF laser. Because the laser energy density used in this study was 2.0 J/cm^2 and the wavelength (193 nm) was shorter than 266 nm, C^+ would be the major species. The released species from the JT-60 codeposits irradiated by both ArF (193 nm) and KrF (248 nm) lasers will be measured in a future experiment.

3.3. Temperature rise on the surface during the laser irradiation

According to Planck's law of radiation, the radiation energy density, $\rho_\lambda d\lambda$ for the wavelength interval $(\lambda, \lambda + d\lambda)$ can be expressed as:

$$\rho_\lambda d\lambda = 8\pi ch d\lambda / \lambda^5 [\exp(ch/k_B \lambda T) - 1], \quad (2)$$

where c, h and k_B are light velocity in vacuum, Planck constant and Boltzmann constant, respectively.

To know the temperature rise on surface during the laser irradiation is a key for understanding the mechanisms of codeposit removal. The transient surface temperature in nanoseconds was measured from Eq. (2). The thermal emission signal from the irradiated surface was collected into the InGaAs photo detector with a response time of 3 ns through an IR filter of 1202 ± 6 nm. As shown in Fig. 3, at the laser energy density of 0.5 J/cm^2, the IR intensity decreased with increasing number of pulses.

The temperature rise due to pulsed irradiation of a homogeneous material like graphite was calculated based on the resolution of the following one-dimensional heat flow equation where thermal diffusion is dominant [11].

$$\kappa \partial^2 T(x,t)/\partial x^2 + H(t)/(\rho C_p) = \partial T/\partial t, \quad (3)$$

where $T(x,t)$ is temperature at x, the distance from the surface and at t, time, $H(t)$ is time-varying volumetric heat source, and κ, ρ and C_p are thermal conductivity, density and specific heat, respectively.

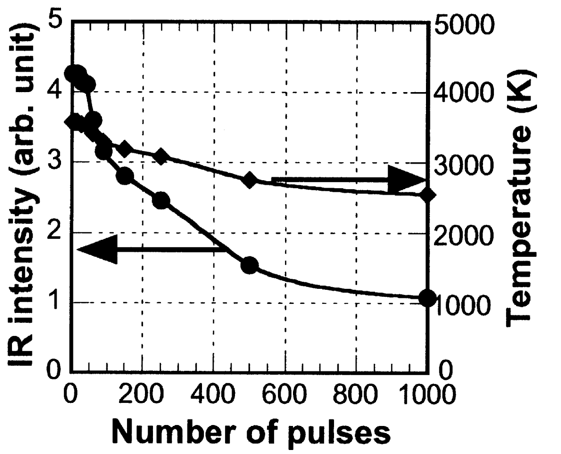

Fig. 3. The IR intensity and temperature as functions of the number of pulses at the laser energy density of 0.5 J/cm^2.

Giving properly selected input data, some physical properties of the material and laser data, the time evolution of the surface temperature of the material can be obtained. At the laser energy density of 0.5 J/cm^2, the maximum temperature for the graphite was then calculated to be about 2550 K from the graphite's properties (the density $\rho = 2$ g/cm^3; the specific heat $C_p = 0.8$ J/g K; the thermal conductivity $\kappa = 1$ W/cm K) [12].

Then the value of temperature rise calculated by the code was used as the temperature at the 1000th pulse irradiation, since the codeposits were confirmed to be completely removed after the irradiation by both the IR intensity data and the depth data. Subsequently, the temperature at other pulses was calculated from the IR intensity and Eq. (2). The temperature at the beginning of the laser irradiation was 3570 K, which is approaching the sublimation point (4000 K) [13] and the melting point of graphite (4450 K) reported by Heremans et al. [14]. The decreasing temperature with increasing number of pulses suggests that the thermal conductivity increases with the depth of the codeposits.

4. Conclusions

Three experiments using JT-60 tiles were performed with an excimer laser with a wavelength of 193 nm, and the removal rate, released gases and temperature rise were discussed. The following conclusions were deduced:

(1) The effective absorption coefficient of the JT-60 codeposits for the excimer laser was determined to be 1.9 μm^{-1}, which is almost one order smaller than the optical absorption coefficient of graphite at 193 nm (16.4 μm^{-1}). At a laser energy density of 7.6 J/cm^2, an erosion rate of 1.1 μm/pulse was reached.
(2) Hydrogen isotopes were released predominantly in the form of hydrogen molecule in the laser ablation process of the JT-60 codeposits, while the formation of water could be ruled out. This feature of the ablation technique using the excimer laser looks very attractive from the viewpoint of effective and safe tritium recovery.
(3) At a laser energy density of 0.5 J/cm^2, the surface temperature decreased from 3570 K at the beginning of the irradiation to 2550 K at the 1000th pulse irradiation, which suggests that the thermal conductivity increased with the depth of the codeposits.

The above results show that excimer laser irradiation can provide a viable technique for the removal of codeposits.

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