ELSEVIER

In situ diagnostic for monitoring of deuterium and tritium in re-deposited carbon layers by laser induced desorption

B. Schweer *, F. Irrek, G. Sergienko, V. Philipps, U. Samm

Institut für Plasmaphysik, Forschungszentrum Jülich, Association EURATOM–FZJ, Trilateral Euregio Cluster, 52425 Jülich, Germany

Abstract

Tritium retention in the plasma facing components and co-deposition in re-deposited layers is a key issue for ITER, since the operation of the machine will be restricted to a maximum inventory of tritium. Development of methods to detect in real time the amount of retained fuel in the in vessel components is urgently required. A promising method for this is the pulsed laser induced desorption, by which the imbedded gas in such layers is released into the plasma where the injected atoms and molecules are excited and ionised and the characteristic line emission is detected spectroscopically. In this paper the release process is investigated in detail for amorphous hydrocarbon layers in a laboratory experiment, as a prerequisite for the later application and determination of conversion factors between line radiation and desorbed flux in TEXTOR plasmas. A Nd:YAG laser has been used with nearly constant output power leading to a smooth controllable increase of layer temperature, resulting in a complete desorption of D_2 and CD_4 as well as preventing carbon sublimation.
© 2007 Elsevier B.V. All rights reserved.

1. Introduction

The deposition of eroded material in a tokamak is associated with co-deposition of hydrogen. Retention of tritium is limited to 350 g in total in order to satisfy safety regulations [1]. Net deposition areas in tokamaks are observed on plasma facing components (PFC) at the main chamber, the baffles and the divertor as well as on remote areas in shadowed positions, e.g. in pumping port ducts [2]. A quantitative in situ analysis of tritium distribution is of great importance. In particular during the pre-tritium phase in ITER, monitoring of the hydrogen retention at selected areas during and over several discharges is necessary. This will give also the information needed to apply the most efficient removal techniques to continue ITER operation if the tritium contamination limit is approached.

The measurement of the hydrogen content in deposited thin layers by laser induced heating is already described in literature [3,4]. There are mainly two methods considered.

Laser induced ablation with very high power density (several GW/cm^2) destroys a thin layer of deposited material forming a plasma. Characteristic

---

* Corresponding author. Fax: +49 2461 612660.
  E-mail address: B.Schweer@fz-juelich.de (B. Schweer).

for this process is that the released atoms and molecules including the embedded hydrogen isotopes form a jet beam with very high particle energies of a few eV [5]. However, quantitative measurements suffer from uncertainties of the energy absorption at the surface during the short laser pulse of about 10 ns. The formed plasma also might yield to a release of bulk material and in some cases during the pulse itself absorbs the laser radiation.

Laser induced desorption at moderate power densities (up to 100 kW/cm^2) leads to a thermal heating of a thin surface layer and at pulse duration of a few ms surface temperatures up to 1700 °C can be reached on exposed areas, which are not significantly extended by heat diffusion during this time. In this temperature range only the hydrogen isotopes and gaseous constituents are released whereas the bulk material and the deposited layer are not affected. For technical reasons, e.g. guiding the light to locations of difficult access, a laser wavelength should be selected that can be coupled into and transmitted through a fibre without significant power losses.

First attempts for laser induced desorption were made with long pulse (350 μs) ruby lasers [6]. Unfortunately their intensities are focussed in a burst of short spikes with less than about 1 μs duration each producing power densities of more than 1 MW/cm^2 that results in the ablation of layer material. The temporal development of the spikes was arbitrary and did not allow a reliable determination of the hydrogen content in prepared thin layers.

The method proposed here allows a reliable quantitative determination of hydrogen isotope (H, D, T) content in a layer or in the surface of PFC's during plasma operation. The constant radiation of a long pulse Nd:YAG laser is coupled into a fibre, conducted to the tokamak and focussed onto PFC surfaces, resulting in desorption of hydrogen isotopes that are measured in situ spectroscopically. The release of particles into the tokamak plasma is essential for this diagnostic and therefore it is not applicable at remote areas.

This paper presents detailed investigations of the process of desorption from amorphous hydrocarbon layers deposited on graphite irradiated by a long pulse Nd:YAG laser. In the first part the experimental set-up of a laboratory experiment, the preparation of the sample targets and the properties of the laser radiation are described. The second chapter describes the dependence of desorption of different species on laser power and energy depending on

the layer thickness. In the last chapter the consequences for application of the method in TEXTOR are discussed.

2. Laboratory set-up

The laboratory experiment for the laser induced desorption is shown schematically in Fig. 1. The light of an Nd:YAG laser at a wavelength of 1064 nm is coupled into a 35 m long fibre with 400 μm core diameter. The maximum power density and pulse duration of this laser that is commonly used for welding applications are 20 kW and 20 ms, respectively but the energy is limited to 60 J per pulse and an average power of 200 W. Typically about 85% of the input power is decoupled at the other end of the fibre and imaged by a lens with 30 cm focal length onto the target in a vacuum chamber. The optical elements are coated to minimize transmission losses. A target holder with four samples of 22×22 mm^2 can be rotated to vary the angle of incidence of the laser beam at the target surface. Additionally, the target can be moved in vertical and horizontal direction to give access of the whole surface to the laser radiation. The chamber with 85 l volume is evacuated to a base pressure of 10^{-7} mbar by a turbo pump system. During measurements a gate valve can be closed to determine in a simple manner the increase of partial pressure of deuterium and CD$_4$ induced by the laser desorption. The pressure increase is measured 15 cm from the target surface by a quadrupole mass spectrometer (QMS) that is mounted at equatorial plane on a port tilted 45° from the laser beam. At the opposite side a fast linear CCD array camera with 128 elements and 100 kHz maximum line repetition rate
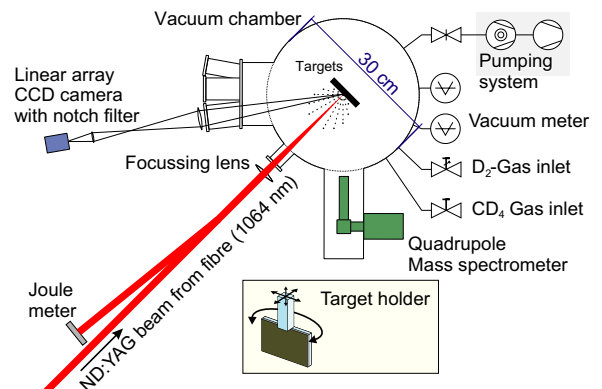

Fig. 1. Experimental set-up in laboratory.

is imaged vertically onto the laser exposed spot measuring the spatial distribution of the temperature with 10 μm spatial resolution. Reflected laser stray light from the target is suppressed by a notch filter. In the experiment temperatures up to 2700 °C were achieved measured with the shortest integration time of 10 μs. At long integration times of 100 μs (10 kHz) the lowest detectable temperature is about 700 °C. The camera was calibrated by black body radiation from an oven.

Amorphous hydrocarbon layers with thicknesses from 50 nm to 250 nm were deposited on carbon targets in a dc assisted glow discharge. Reflectrometry was used to determine the layer thickness. Its deuterium content was measured by absolutely calibrated thermal desorption [7]. For the layers investigated a value of 1.2×10^{15} D-atoms/cm² nm was deduced that was used for the absolute calibration of the QMS signal in this experiment. Inside the vacuum chamber a spot on the target was exposed by laser pulses until the QMS deuterium partial pressure signal was negligible. With the sum of all signals before, the known spot size and layer thickness the calibration factor for the QMS signal was determined. The relative QMS sensitivity for the different gases was cross calibrated by filling the chamber with D_2 or CD_4 to a partial of pressure of 10^{-5} mbar.

3. Experimental results

3.1. Laser properties

The laser properties have been deduced from signals obtained from a linear array camera exposed by the beam after attenuation with neutral density filters. The temporal evolution of the laser power for 1.2 ms pulse duration after passing the fibre is shown in Fig. 2(a). The Nd:YAG laser power fluctuates only within ±10% around the average power. The temperature deviations caused by these fluctuations are negligible and do not disturb the determination of the hydrogen content.

For the same shot in Fig. 2(b) the radial distribution of the power density of a laser spot at 1 ms is presented. The power density is constant within ±10% over the spot diameter of 1.2 mm. The influence of the 'wings' can be reduced by a careful alignment. Nevertheless the pulse duration should not exceed 4 ms to minimize the influence of lateral (horizontal) heat transfer that is important for the calculation of the deuterium content. The absolute surface temper-
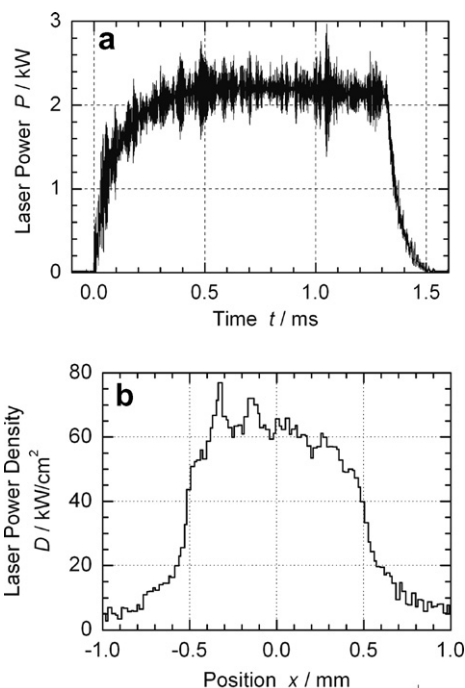

Fig. 2. Performance of a laser pulse. (a) Temporal evolution of total power, (b) radial distribution of power density for the same shot at 1 ms.

ature is deduced from a one-dimensional heat conduction model with material constants for graphite and proportional to the absorbed power and the square root of the exposure time.

The experiments were performed with laser power densities in the range from 40 to 160 kW/cm² at the target surface. An example for a rapid heating is shown in Fig. 3 where the absorbed power density is about 130 kW/cm², which is sufficient to reach surface temperatures on carbon above 2800 °C in 1.7 ms. However, the measured tempera-
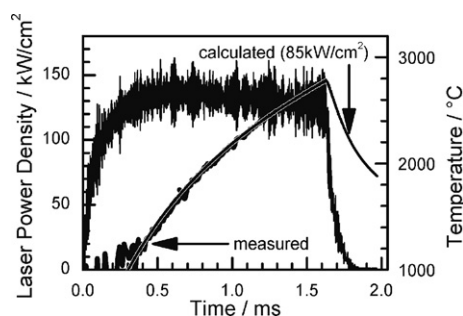

Fig. 3. Comparison of measured and calculated surface temperature on carbon.

ture time evolution could be reproduced only assuming 50% laser power absorption. For the investigated hydrocarbon layers ($\leqslant 250$ nm) the laser power is mainly absorbed in a bulk graphite layer (about 100 μm for a 4 ms pulse) and changes in reflection coefficients of hydrocarbon layers during exposure might not be considered in the first order.

3.2. Deuterium and methane release

The laser induced desorption can lead to the evaporation of atoms and molecules of all hydrogen isotopes. Therefore deuterated layers were used to clearly separate mass 4 in the QMS. The measurement of D atoms can still be disturbed by H_2 released from the carbon bulk material. Atomic hydrogen species are released mainly at temperatures above 900 °C [8]. The laser power density and the corresponding temperature increase at the surface of an a-C:D layer on graphite are shown in Fig. 4(a) and (b), respectively. In Fig. 4(c) the partial pressure, given in total number of released D_2 molecules from a 250 nm thick hydrocarbon layer for a laser power density of 42 kW/cm^2 is shown. The maximum value where the time derivative (flux) approaches nearly zero is achieved at 2.6 ms where the temperature is about 1100 °C. This again fits with theoretical values if only 50% of the laser power is absorbed. Although the temperature still rises the signal is nearly constant indicating that all retained D_2 molecules are evaporated. This is also verified by a second laser shot on the same spot where less than 20% D_2 of the first shot is released.

The reduction of the signal after the laser shot shown in Fig. 4(d) can be explained by temperature relaxation of the 'hot' molecules to room temperature. Measurements on mass 2 (D or H_2) produced similar results indicating that mainly H_2 molecules from the bulk material were released and not atomic deuterium that should appear at higher temperatures.

Methane molecules are suggested to be an important channel of deuterium release too. Under the same condition as before the release of CD_4 molecules (mass 20) was investigated. The temporal development shown in Fig. 5 is nearly identical with that of the D_2 molecules but the flux is lower by a factor of 12. The amount in a second shot is negligible. That means that the major part of deuterium is leaving in two channels. For the layers investigated here the ratio for D_2 to CD_4 was 6–1.
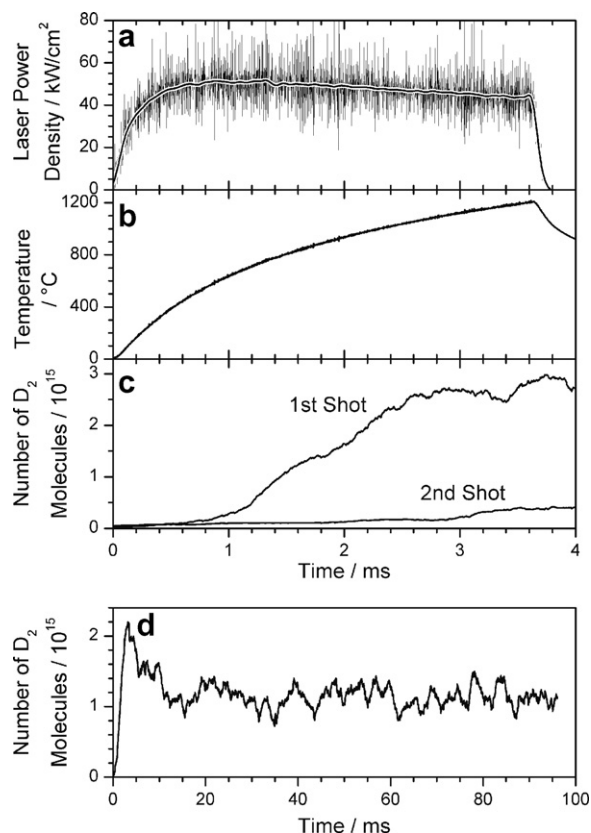

Fig. 4. Exposure of a 1.2 mm^2 spot on a 250 nm thick a-C:D layer on graphite with a 1.9 J laser pulse. Temporal evolution of (a) laser power density, (b) surface temperature, (c) desorbed deuterium molecules during 2 consecutive laser pulses, and (d) desorbed deuterium molecules from same layer after laser exposure (relaxation effect).
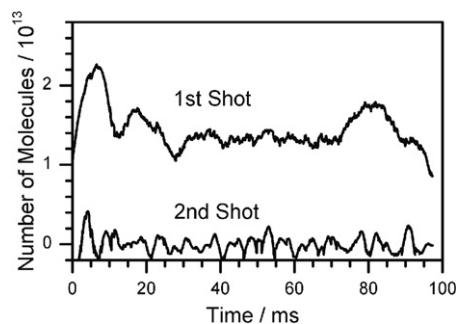

Fig. 5. Temporal evolution of desorbed methane molecules in 2 consecutive laser pulses.

Layers with different thicknesses have been investigated in the experiment. A linear behaviour between the released D content and thickness up to 250 nm has been observed, as expected.
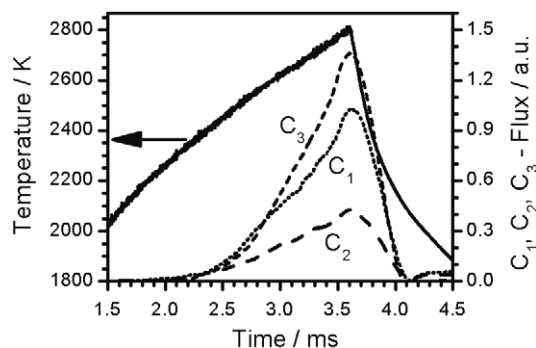
Fig. 6. Temporal evolution of the flux of sublimated carbon atoms C_1 and cluster C_2 and C_3 (no correction of ionisation probabilities, 50% absorption).

3.3. Carbon release

The aim of the diagnostic is the in situ determination of the hydrogen isotopes. Even at low layer thicknesses the bulk material should not removed. To study this, the carbon sublimation as a function of temperature was investigated. The species C (mass 12), C_2 (mass 24) and C_3 (mass 36) were measured at the quadrupole and the results are shown in Fig. 6. Significant carbon flux appears at 2.5 ms where the surface temperature passes 1900 K. At the end of the laser pulse the temperature decreases rapidly and the signals drop immediately since only line of sight carbon particles can enter the QMS.

4. Discussion

Nd:YAG laser light coupled into fibres offers a high flexibility to desorb co-deposited hydrocarbon layers in a laboratory and a tokamak. The experiments have shown that almost all the deuterium can be desorbed in a single shot. Nearly all the embedded deuterium is released in form of D_2 and CD_4 molecules leaving the surface in the temperature range from 700 °C to 1200 °C practically independent on the temperature rise time and the thickness of the layers (up to 250 nm). No significant fraction of atomic deuterium was desorbed. At temperature below 1200 °C the release of carbon atoms and clusters can be neglected. Hydrocarbon

layers with larger thickness (several μm) as one can expect in a tokamak have not been investigated yet. The heat conductance of such layers to the bulk material might be reduced leading under pulsed laser irradiation to a non predictable increase of the temperature with strong evaporation of material.

For application at TEXTOR at the maximum available power, a spot of $0.5\ cm^2$ can be heated up to 1200 °C within 4 ms and about 10^{17} D atoms might penetrate into the edge plasma of TEXTOR sufficient for a reliable spectroscopic detection. In addition the optical imaging allows the production of smaller spot sizes associated with shorter exposure times. This can be an advantage for measurements in tokamak plasmas because of a better definition of the spot size and of a larger pulsed Balmer line intensity above the background radiation in the plasma edge.

Acknowledgement

References

[1] M. Shimada, A.E. Costley, G. Frederici, K. Ioki, A.S. Kukushkin, V. Mukhovatov, A. Polevoi, M. Sugihara, J. Nucl. Mater. 337–339 (2005) 808.

[2] J. Roth, A. Kirschner, W. Bomeyer, S. Brezinsek, et al., J. Nucl. Mater. 337–339 (2005) 970.

[3] C.H. Skinner, N. Bekris, J.P. Coad, C.A. Gentile, M. Glugla, J. Nucl. Mater. 313–316 (2003) 496.

[4] D.D.D. Summers, M.N.A. Beurskens, J.P. Coad, G. Counsell, W. Fundamenski, G.F. Matthews, M.F. Stamp, J. Nucl. Mater. 290&291 (2001) 496.

[5] A. Pospieszczyk, P. Bogen, H. Hartwig, Y.T. Lie, J. Nucl. Mater. 93&94 (1980) 378.

[6] B. Schweer, A. Huber, G. Sergienko, V. Philipps, F. Irrek, H.G. Esser, U. Samm, M. Kempenaars, M. Stamp, C. Gowers, D. Richards, J. Nucl. Mater. 337–339 (2005) 570.

[7] J. Winter, H.G. Esser, P. Wienhold, V. Philipps, E. Vietzke, K.H. Besocke, W. Möller, B. Emmoth, Nucl. Instrum. and Meth. B 23 (1987) 538.

[8] S. Brezinsek, G. Sergienko, A. Pospieszczyk, Ph. Mertens, U. Samm, P.T. Greenland, Plasma Phys. Control. Fusion 47 (2005) 615.