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Desorption of deuterium from beryllium codeposits using flash heating

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ABSTRACT

As a result of safety concerns, limits will be placed on the allowable tritium inventory retained inside the ITER vacuum vessel. The primary motivation for the present work is to test the proposed method of removing tritium from main chamber codeposits using radiative heat flashing from controlled ITER plasma shutdowns. Detritiation of Be codeposits is studied in the PISCES-B facility using flash-heating by a 10 ms laser with up to 2 MJ/m^2 of absorbed energy density. Three types of codeposits are flash-heated with layer thickness ranging from 0.1 to 1.2 μm. Less than 25% of the D in the Be layer escapes at ITER-relevant flash energy densities and with peak surface temperature up to ~900 °C. Repetitive flashing with peak surface temperature of 400–500 °C results in an increased population of higher energy trap sites, implying that transient heating which causes appropriate surface temperature excursion redistributes D among codeposit trap sites.

1. Introduction

Tritium recovery from plasma-facing components in ITER will be achieved by standard bakeout assuming that most of the in-vessel inventory will accumulate by codeposition with beryllium (Be) on divertor and first wall surfaces [1]. However, the maximum bakeout temperature achievable on the first wall surfaces will only be 240 °C (compared with 350 °C for the divertor) and it is known from laboratory experiments that depending mainly on the surface temperature at which codeposition occurs, the fuel desorption capability may be limited [2,3]. It has been speculated, however, that a degree of codeposit detritiation might be possible as a result of flash heating when impurity radiation is used to dissipate the plasma stored energy by massive gas injection during an induced plasma shutdown [4]. This paper reports on laboratory investigations of the potential of this transient heating for tritium recovery from Be codeposited layers, demonstrating that the process will not yield significant fuel outgassing at the heat flux densities expected in ITER as a result of disruption mitigation.

Previous work on laser flashing of bulk Be exposed to 1.5 keV hydrogen ion implantation showed that hydrogen desorption began to occur for surface temperatures above ~820 °C [5], and it was found that a significant fraction of D could only be released when the surface temperature reached or exceeded the melting temperature of Be [6]. Flash removal of hydrogenic species from carbon codeposits has also been investigated and it was found that desorption occurred for sufficiently high surface temperatures above 1230 °C [7].

2. Experimental setup

An Nd:YAG laser (wavelength of 1064 nm, maximum pulse width of 10 ms, maximum output energy of 50 J per pulse) is used to irradiate the surface of Be codeposits and provide a controlled temperature rise. The codeposits are created on W substrates in the PISCES-B facility [8]. A reference codeposit, which is not subsequently irradiated by the laser, is simultaneously created next to the flash-heated codeposit. The amount of retained deuterium is measured by applying thermal desorption spectroscopy (TDS) to each sample separately. By comparing the retention in the flash-heated sample with the reference sample, the amount of deuterium released due to the laser flash is determined.

The laser is guided by a set of turning mirrors over a distance of ~20 m and a lens is used to obtain the desired spot size at the target. Laser beamline losses are approximately 20%, and the reflectivity of bare W is measured (~40%) in order to give a rough estimate of the absorbed laser power (however, the surface morphology of deposited Be is expected to be different than that of bare W). Codeposits are created using three methods, with more than an order of magnitude variation in layer thickness. Schematics of the experimental setups for the three types of codeposits are shown in Fig. 1:

(a) A W witness plate located outside a D plasma (T_e = 4–15 eV, $n_e = 0.5-3 \times 10^{18}$ m^{-3}) collects sputtered Be and reflected D from a biased Be target bombarded by 55–80 eV plasma ions in PISCES-B. The side-by-side witness plates are located

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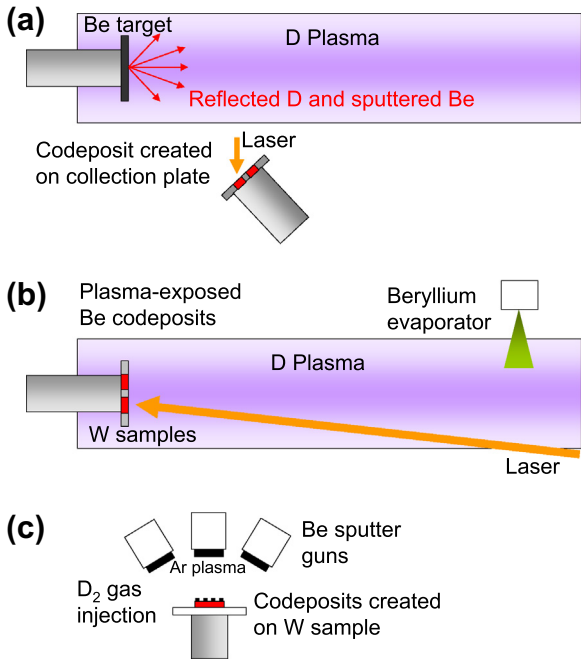

Fig. 1. Schematics of three experimental setups for creating Be/D layers. (a) Codeposit created on W collection plate located outside the plasma using sputtered Be and reflected D from a Be target. (b) Codeposit created inside the plasma with Be seeding. (c) Codeposit created in a magnetron sputtering device.

13 cm away from the target and 7.5 cm from the center of the plasma, with codeposit thickness of approximately 100 nm. A thin Mo mask is used on each W substrate to match the codeposit collection area to the laser wetted area (0.1 cm^2). The collection plate is oriented with its surface normal at an angle of 65° with respect to the laser path, resulting in an oval shaped irradiation pattern as seen in Fig. 2a. The laser irradiates the sample after the plasma is shut off. The W substrate temperature during deposition reached temperatures up to ~200–300 °C. This configuration simulates the build-up of codeposits in a tokamak on surfaces that are not exposed directly to plasma.

(b) Codeposits are also created on 6 mm diameter W samples exposed to a PISCES-B deuterium plasma and Be influx from a Be effusion cell [8]. The W samples are at floating potential, resulting in net Be/D deposition. The effusion cell injects neutral Be which becomes ionized in the plasma and flows toward the W sample, as shown in Fig. 1b. The typical D flux is $0.5-1.0 \times 10^{22}$ m^{-2} s^{-1} with plasma exposure duration ranging from 300 to 1400 s, layer thickness ranging from 100 to 650 nm, and sample temperature during plasma exposure of 40–60 °C (water-cooled samples). The ratio of Be ions to D ions in the plasma is estimated from spectroscopic data and ranges from 0.1% to 1%. The laser path makes an angle of 15° with respect to the codeposit normal. This configuration simulates areas in a tokamak such as the first wall that undergo simultaneous erosion and deposition, but which are in a net deposition regime.

(c) In order to study flashing on thicker codeposits, W samples are coated in a Be magnetron sputtering device [9]. Ar ions sputter Be targets with the W sample biased at −80 V. Deuterium gas is injected in order to trap D in the Be layer, and the flow rate of D_2 is set to ¼ the flow rate of Ar with 6 mTorr of gas pressure. Three Be targets have applied power of 100 W each with the sputtering run for a duration that creates a layer thickness of ~1.2 μm, and the samples reached an estimated temperature of 200 °C during deposition. The sample is moved in air to PISCES-B for laser flashing.

In ITER the expected ion temperature at the first wall is less than 70 eV and the expected energy of charge-exchange neutrals hitting the wall range from tens of eV near the divertor to ~200 eV at the outer midplane [10]. In PISCES-B the approximate energies of D particles arriving at the codeposited sample surface are a few tens of eV for the case of the collection plate outside the plasma [11], ~20 eV for the plasma-exposed case with targets at floating potential, and a few tens of eV for the magnetron sputtering case. Thus, the energies of D are a reasonable simulation of the expected energies of particles forming codeposits in ITER.

Imaging is used to verify the alignment of the laser pulse and targeted codeposit. In some cases a fast camera framing up to 10,000 frames/s is used to record the laser flash. Fig. 2a–c shows the samples and laser spot in each of the three codeposit cases described above. For case (c), multiple masked 2.5 mm diameter codeposits (as seen as the dark circles) are created on a sample which, after laser flashing, is separated into individual codeposits for TDS of each codeposit. The outermost codeposits are reference samples.

The temperature of the sample during laser flashing is measured with a high speed pyrometer consisting of beamsplitters, laser-blocking filters, four detectors, and four interference filters each with bandpass ranging from 30 to 100 nm and central wavelengths at 1300, 1550, 2725 and 3468 nm. The pyrometer collection optics are aligned by back-illuminating the pyrometer fiber and overlapping the resulting spot onto the target. Thermal emission during the laser flash is collected with a CaF_2 lens and is transmitted to the pyrometer using 10.5 m of visible/NIR metal fluoride glass optical fiber. The two shorter wavelength detectors are Hamamatsu photo-multiplier tubes with adjustable gain; the two longer wavelength detectors are liquid nitrogen cooled InSb photovoltaic devices from Teledyne Judson. The signal from the InSb detectors is typically too small to be processed due to limited amplification and possibly due to coatings on the vacuum window from plasma–material interaction experiments. The temperature of the sample is thus determined using a ratio technique for the two near infrared signals using the assumption of gray body thermal emission over the detected wavelength range from 1300 to 1550 nm, with a minimum detectable temperature of ~400 °C. The time response of the detectors and pre-amps is approximately 100 microseconds.

3. Results

The TDS curves for the three types of codeposits are shown in Fig. 3. The blue data points show the D flux from D_2 thermal desorption for the reference samples with no laser flash, and the red lines show the TDS curves for samples flashed with a 10 ms laser pulse. One to four laser pulses are typically fired for all flashed codeposits; however, the Be-seeded plasma exposed sample shown here in Fig. 3b received 50 laser flashes (each 10 ms in duration) to study the effect of multiple pulses. The linear temperature ramp during TDS is also shown in the figures.

The laser pulse results in some desorption of D from the low temperature release peaks, but a significant amount of D remains in the codeposit after flashing. The codeposits created on the collection plate and in the Be-seeded plasma have a prominent release peak at ~500 °C, while the magnetron codeposit has a large release peak at 560 °C. The difference in release temperatures is attributed to the −80 V bias applied to the sample during deposition and the presence of Ar ions in the magnetron coater. Likewise,

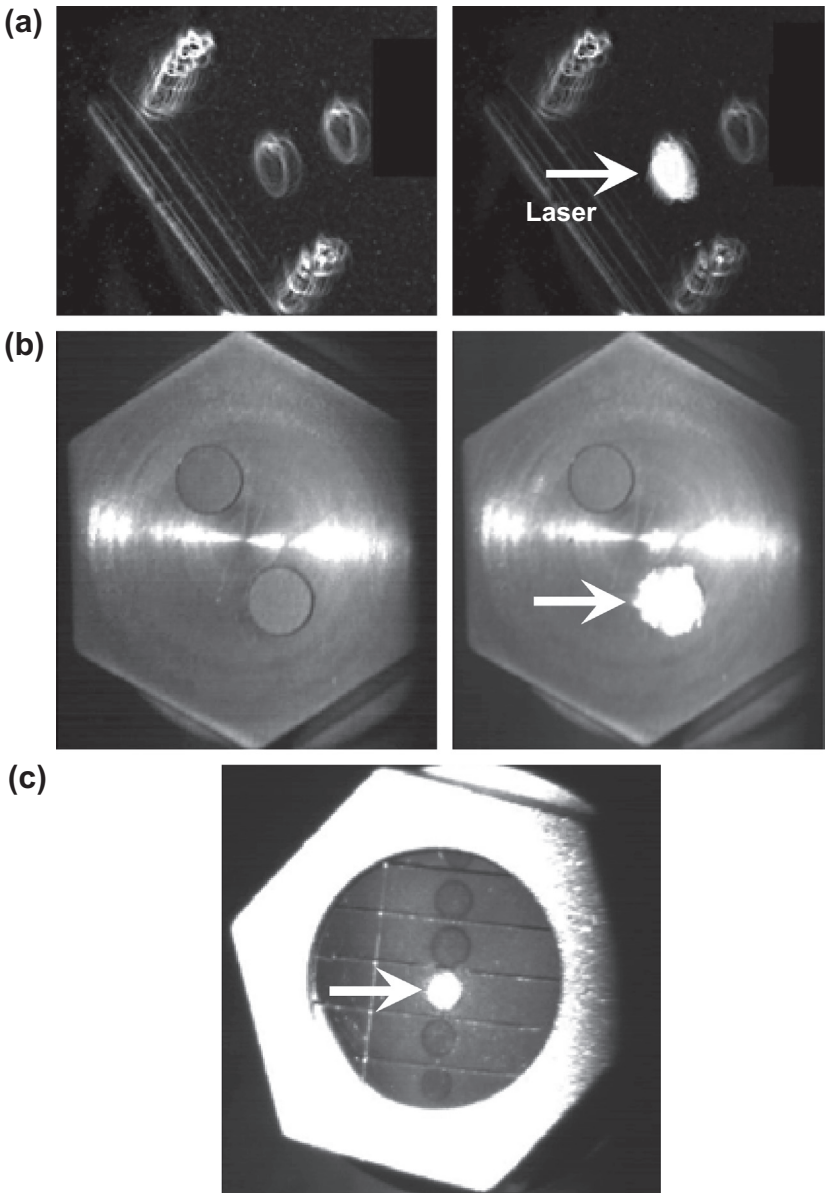

Fig. 2. Side-by-side codeposits showing reference samples and the laser-irradiated samples for the three types of codeposits: (a) collection plate, (b) plasma-exposed, and (c) magnetron sputtering device.

the low temperature release peaks seen in cases (b) and (c) have a similar temperature difference. The low temperature release peak is not present in case a), presumably because the temperature of the sample during the formation of the codeposit (200–300 °C) exceeded the release temperature. The amount released due to the laser flash does not exceed 25% even for the highest energy density case shown in Fig. 3a, which reached a peak surface temperature during the flash of ~900 °C. Cases (b) and (c) reached peak surface temperatures during the flash of ~450 °C and ~700 °C, respectively. The planned first wall thermal bakes in ITER are currently 240 °C, which may remove the low temperature peak in the TDS curves but will not be sufficient to remove the high energy peak.

In the plasma-exposed codeposit shown in Fig. 3b, a third peak is visible at 280 °C which has been identified as the decomposition of beryllium deuteride (BeD_2) by Reneilt et al. [12]. Previous work has shown that in a codeposit a significant amount of BeD_2 can be created [13]. The TDS curves for the flashed and un-flashed cases suggest that BeD_2 is effectively dissociated by the laser heat pulses.

The TDS curves shown in Fig. 3b, in which the sample received multiple laser flashes, reveals interesting behavior. The higher energy traps appear to become increasingly populated after multiple heat pulses, but the total retention remains nearly unchanged after flashing. The heat pulse de-traps D from lower energy traps and we speculate that some of the mobilized D falls into deeper energy traps. D remains in these higher energy traps provided that the heat pulse is insufficient to detrap. In the case shown in Fig. 3b the peak surface temperature during the flash is lower than the release temperature of the higher TDS peak, and thus D trapped in the high energy trap sites is not desorbed by the laser.

For reference, a spatially uniform disruption flash which deposits ~350 MJ of stored thermal energy over 700 m^2 of the first wall in ITER would yield an energy density of ~0.5 MJ/m^2, with a predicted flash duration of ~10 ms [14]. The summary of flash heating data shown in Fig. 4 indicates that for such ITER-relevant energy densities and flash duration, the fraction of desorbed D is ~10%. The fractional release increases to only ~25% at the maximum absorbed laser energy density in PISCES-B.

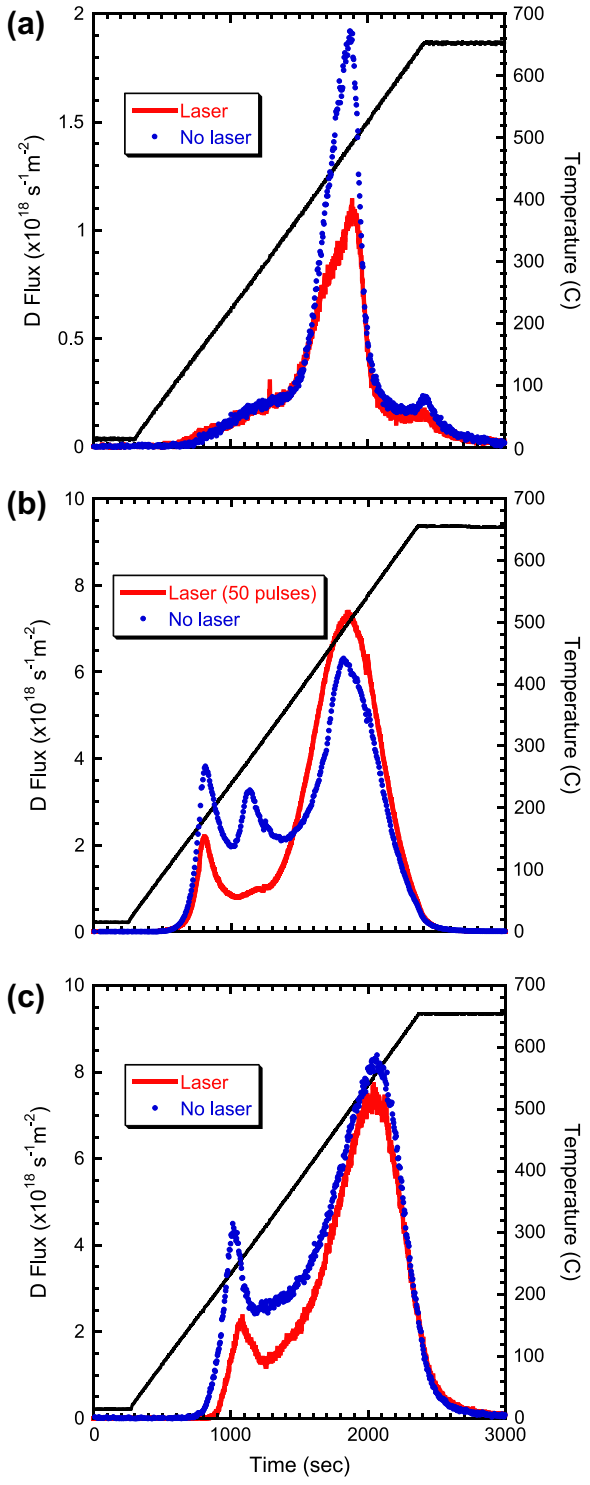

Fig. 3. TDS curves for the three types of codeposits: (a) collection plate, (b) plasma-exposed codeposit irradiated by 50 laser flashes, and (c) magnetron sputtering device.

A recent simulation of a controlled plasma shutdown in ITER using Ne gas injection predicts toroidal and poloidal peaking factors of ~3 and peak wall temperatures of 800–900 °C [14]. The flash heating data shown in Fig. 5, now compiled in terms of peak surface temperature rise on the different samples, indicates that only ~25% of the D would be desorbed from local hot spots which reach these temperatures, and the average fraction of desorbed D over the entire first wall would be even smaller due to peaking factors larger than unity. Fig. 5 has fewer data points than Fig. 4 because the pyrometer was not installed for all experiments. In both Figs. 4 and 5, the fraction of desorbed D is calculated as $1 - \phi_{flash}/\phi_{reference}$, with retention calculated from D flux (Γ_D) measured from TDS: $\phi = \int \Gamma_D dt$. Negative values for the fraction of desorbed D are due to the lack of exact reproducibility in the deposition for the flashed and reference samples.

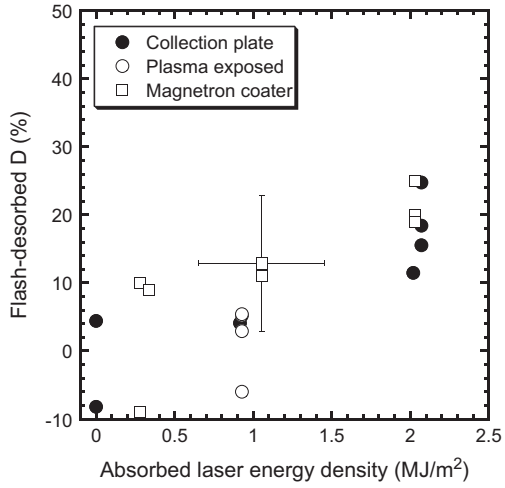

Fig. 4. The fraction of flash-desorbed D as a function of absorbed laser energy density. The error bars apply to all data points.

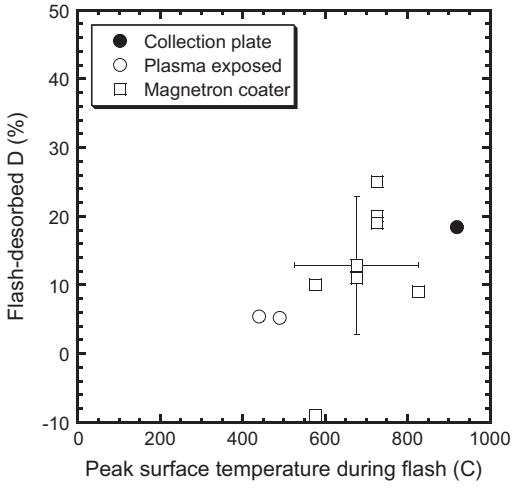

Fig. 5. The fraction of flash-desorbed D as a function of peak surface temperature during the laser flash. The error bars are typical for all data points.

4. Discussion and conclusion

The fraction of deuterium trapped in Be codeposits is known to decrease with increasing steady-state surface temperature [3], and TDS releases all trapped D with slow (0.3 K/s) thermal ramp rates up to 650 °C [15]. The transient laser flash creates a surface temperature that in some cases exceeds that maximum temperature reached during TDS, yet only a small fraction of D is released. The experiments presented here thus demonstrate that D desorption from Be codeposits is much reduced when heating occurs on the timescale of milliseconds. Modeling flash heating and TDS with the Tritium Migration Analysis Program (TMAP) shows that the re-

lease of D due to a short-duration heat pulse is trap-limited rather than diffusion or surface recombination limited [16].

As shown by De Temmerman and Doerner [17], D retention and the slow-thermal-ramp release of D depend on the deposition conditions. This is in fact nicely demonstrated by the variation of the total retention and the detailed shapes of the TDS curves of the three different types of codeposits shown in Fig. 3. However, for short duration thermal transients, the fraction of released D for the three deposition conditions appears similar to within the experimental uncertainty. This implies that during transient flash-heating the rate limiting cycle of trap-depletion and re-trapping does not depend of the details of the codeposition creation process.

Our results and those of [5,6] show that for short timescale heating, a significant amount of D can only be removed by melting or approaching the melting temperature of Be. Since the disruption mitigation system in ITER is being designed specifically to avoid Be melting, our conclusion is that full detritiation during controlled plasma shutdowns will not be possible. We find that radiative flash heating on the timescale of milliseconds removes only $\sim$10% of deuterium from Be codeposits at flash energy densities expected for a spatially-uniform plasma shutdown in ITER. We thus recommend that alternate methods should continue to be pursued to address the challenging issue of detritiation in future tokamaks. Future work includes investigating the effects of longer flash duration on the removal of hydrogenic species from Be codeposits.

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