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Tritium Removal by CO_2 Laser Heating*.

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Abstract — Efficient techniques for rapid tritium removal will be necessary for ITER to meet its physics and engineering goals. One potential technique is transient surface heating by a scanning CO_2 or Nd:Yag laser that would release tritium without the severe engineering difficulties of bulk heating of the vessel. We have modeled the heat propagation into a surface layer and find that a multi–kW/cm^2 flux with a exposure time of order 10 ms is suitable to heat a 50 micron co-deposited layer to 1,000-2,000 degrees. Improved wall conditioning may be a significant side benefit. We identify remaining issues that need to be addressed experimentally.

I. INTRODUCTION

Tritium retention has long been an important issue in DT burning fusion devices, particularly those with carbon first walls. The magnitude of the problem for ITER was brought home by recent calculations of co-deposition rates that predicted the 1 kg in-vessel tritium inventory limit could be reached in as few as 50 pulses[1]. Surface tritium represents a safety hazard as it can be easily mobilized in the event of an accident. In addition, a simple comparison of the tritium fuel required (0.2-0.3 kg per 1,000 s pulse) and the available supply (2-3 kg/year) graphically illustrates the critical importance of efficient removal of tritium retained in the ITER vessel. The development of efficient methods to remove tritium in co-deposited layers was recognized as an outstanding issue in the recent FESAC Report[2] and underlined in a recent review of the tritium inventory in ITER plasma facing components[3]. Ten potential removal methods were listed in [3], however all of these need R&D to resolve various issues before it can be established that they are suitable for ITER. It is important to note that tritium removal is a critical issue for *any* fusion reactor concept based on burning DT.

Tritium is primarily accumulated by co-deposition with carbon on the surface of plasma facing components during the discharge[4]. The amount of tritium retained is a significant fraction of the tritium supplied as fuel[5,6]. In ITER carbon/tritium layers will accumulate due to sputter erosion of the divertor target plate and the subsequent deposition of sputtered material on the divertor plate and/or plenum. Peak growth rates are calculated to be several μm of carbon per 1,000 s pulse[3]. For an operational schedule with ≈10 pulses/day we would anticipate co-deposited layers of thickness of 50 - 100 μm thick after a few day's operations. In vacuum, temperatures in excess of 1,000 K are required to produce rapid release of tritium from the crystallites of graphite[7]. Tritium is released on exposure to air at lower temperatures (≈570 K) [8,9] but baking in air is considered unacceptable due to severe deleterious effects on metal surfaces in the vessel. Unfortunately bulk heating of vacuum vessel components to sufficiently high temperatures for tritium release is not practical for ITER due to excessive water pressure in the cooling system. A dedicated high temperature gas or steam baking method would be large, complex and expensive. However, such bulk heating may be avoided by transiently heating the co-deposited layers on the surface by a scanning CO_2 or Nd:Yag laser. This would enable an elevated surface temperature to be attained without the complications of bulk heating.

Multi-kW, robotically controlled lasers have long been used in industrial applications and are an attractive candidate for the present application. The laser would be external to the ITER vacuum vessel and the beam rastered across the plasma facing components by mirrors on a remotely controlled arm. The laser intensity and exposure duration are key design parameters and these, in turn, are governed by the depth of the co-deposited layer and the desired temperature excursion.

Early work has shown substantial release of hydrogen by heating with electron beams and lasers. A 0.8 s, 2 kW/cm^2 pulse of 8 keV electrons completely purged implanted H from thin TiB_2 coatings[10]. Deuterium implanted in papyex graphite was released by several 50 ns, 2.8 J/cm^2 electron beam pulses[11]. Hydrogen implanted in TiB_2 was removed by 20 ns, 1.5 J/cm^2 pulses from a Nd:glass laser[10].

Thermal desorption induced by a 30 ns laser pulse has been developed as a tool to analyze hydrogen located at the surface of solids[12]. This data are relevant to the present work and will be further discussed below. The technique was applied to in-situ analysis of TdeV[13] and the TFTR limiter tiles[14]. Pulse lengths longer than nanoseconds will be needed to heat a thick co-deposited layer. The goal of the present work is to estimate the heat flux and duration needed to transiently heat the co-deposited region to a temperature sufficient to release tritium and to explore the associated engineering issues.

II. SURFACE HEATING

The temperature excursion required to release co-deposited tritium depends on the transport of tritium at elevated temperatures, a process complicated by the anisotropic structure of the codeposited layer and trapping. Tritium release in transient temperature excursions is sensitive to the slowest kinetics such as multistep low activation energy diffusion and it is not always clear how to extrapolate low temperature isochronal thermal desorption data to transport on

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fast time scales at very high temperatures[15]. Fortunately data is available from pulsed laser desorption experiments on deuterium implanted graphite. A 30 ns pulsed ruby laser was incident on graphite samples containing deuterium implanted in the top 100 nm of the surface[15]. The laser fluence was 0.1 mJ/cm^2 to 3 J/cm^2 and the heating and cooling cycle took less than 300 ns. The surface temperature, as calculated by a finite difference code, ranged up to the melting temperature of 4,300 K. The laser energy density required to desorb the deuterium decreased with increasing deuterium density in the graphite, indicating that high order detrapping processes are involved in the re-emission process[15]. A surface temperature of 2,000 K released 90% of the implanted deuterium in vitreous carbon and pyrolytic carbon *'a'* and 30% in isotropic and pyrolytic *'c'*. For the present study we take 2,000 K as a target surface temperature. The results can be easily extrapolated to other temperatures since the temperature is linearly dependent on the laser power.

Continuous wave lasers offer two significant advantages over pulsed lasers. One is that pulsed lasers at high fluence ($\approx$ 1 J/cm^2) release particulate matter from the impact of the laser pulse. The accumulation of particulates in the ITER vessel is a safety concern due to their reactivity. The second advantage is that high average powers and laser efficiencies are needed to cover the ITER divertor in a reasonable time (hours) and these are available in CO_2 and Nd:Yag lasers.

The average power required to heat the surface is estimated as follows. In ITER the lower half of the divertor target and line-of-sight plenum region opposite the target has an area $\approx 5\times10^5$ cm^2 and the overnight temperature is expected to be 400 K. Using the (temperature dependent) specific heat from the database in the HEAT1DS code[16] we estimate 3×10^7 J is required to heat a 100 µm surface layer from 400 K to 2000 K. This energy corresponds, for example, to the output of a 1 kW laser in an overnight tritium clean up lasting 8 hours. The actual power required will be somewhat higher depending on laser beam transmission losses, heat lost by radiation and by conduction to deeper layers.

The thermal conductivity of co-deposited graphite is a key parameter in the present study. Graphite is manufactured in a variety of forms and its thermal conductivity varies over a wide range depending on the micro structure and crystallite orientation. Table 1 shows thermal coefficients of three kinds of manufactured graphite at 400, 1,200 and 2,000 K[16]. The micro structure of co-deposited graphite is unlike manufactured graphite. Sections of co-deposited layers on TFTR tiles show a wave-like layered structure with a surface texture that varies with poloidal location[17]. Unfortunately there do not appear to be any thermal coefficients for co-deposited graphite in the literature. However, the conductivity is expected to be low and in the range between 0.015 and 0.1 W/cm-K[18]. In the present study we use conductivities from these materials with the expectation that pyrolytic carbon perp. is likely to be closest to that of co-deposited graphite. 'perp.' refers to heat flow in a direction perpendicular to the layer planes. Experimental measurements on co-deposited graphite are needed to resolve this issue.

Table 1
Examples of thermal coefficients for graphite

Material	Density g/cm^3	Temperature K	Heat capacity J/g-K	Thermal conductivity W/cm-K
pyrolytic perp.	2.2	400	1.77	0.082
"		1200	1.88	0.018
"		2000	1.99	0.010
2D C/C comp perp.	1.7	400	1.77	0.369
"		1200	1.88	0.19
"		2000	1.99	0.10
3D C/C isotropic	1.96	400	1.77	1.14
"		1200	1.88	0.65
"		2000	1.99	0.41

III. HEAT PROPAGATION

We use both analytical and numerical methods to find the dependence of the heat penetration depth on exposure time. This will determine the laser spot size and scanning speed. An initial estimate of the scaling of the heat propagation depth with exposure time may be obtained using Schmidt's method for unsteady heat flow[19]. A prescribed temperature distribution on an equally spaced grid will relax on time scale:

$$\Delta t = c\rho a^2 \big/ 2K \tag{1}$$

where c is the specific heat, ρ the density, a the grid spacing and K the thermal conductivity. Using thermal coefficients for pyrolytic perp. in Table 1 the temperature of a 100 µm layer is found to relax on a time scale of 5 ms.

Analytic solutions exist for some well defined heat transfer problems. They avoid issues with fine spatial grids, numerical stability and the parametric dependence of the variables is transparent. Equation (2) describes the temperature, θ, vs. depth, x, of a semi-infinite solid at zero initial temperature under a constant heat flux at $x = 0$ beginning at $t = 0$ [19]:

$$\theta = \frac{2F}{K}\left\{\left(\frac{\kappa t}{\pi}\right)^{1/2} \exp\left(\frac{-x^2}{4\kappa t}\right) - \frac{x}{2}\operatorname{erfc}\frac{x}{2\sqrt{\kappa t}}\right\} \tag{2}$$

Here 'erfc' is the complimentary error function, K is the thermal conductivity, $\kappa = K/\rho c$ the thermal diffusivity.

The surface temperature is:

$$\theta_{x=0} = \frac{2F}{K}\left(\frac{\kappa t}{\pi}\right)^{1/2} \tag{3}$$

We rewrite this as:

$$F = \theta K\left(\frac{\pi}{4\kappa t}\right)^{1/2} \tag{4}$$

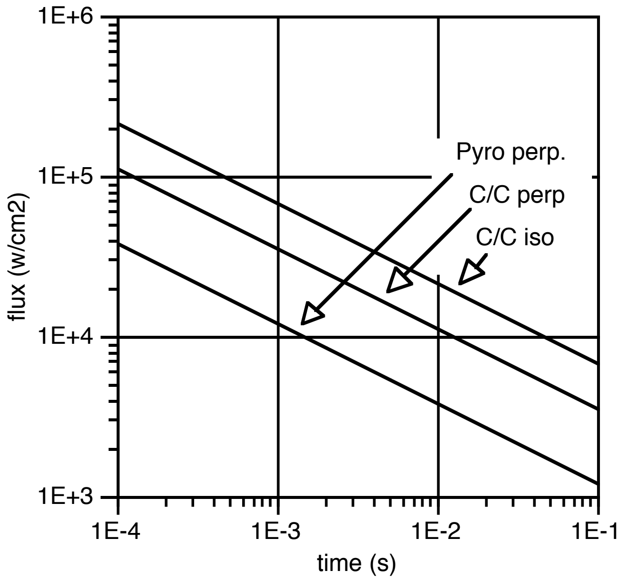

Fig. 1. Flux necessary to attain a 400 K - 2,000 K surface temperature rise for different graphites (4).

where F is the flux necessary to raise the surface temperature by θ degrees in a time t. (see Fig. 1) This approach neglects the temperature dependence of the heat capacity and radiation cooling of the surface but does give useful physical insight into the problem. Black body radiation at 2,000 K is 90 W/cm^2 and is small compared to the incident flux. A 1D expression is appropriate since the laser spot width is likely to be larger than the depth of the heated layer.

We consider a temperature rise from 400 K to 2,000 K (i.e., from $\theta = 0$ to $\theta = 1600$ K) for the three materials listed in Table 1 and plot equation (4) using the thermal conductivities at 1,200 K. The penetration of the heat pulse into the material will change with the exposure time. Fig. 2 shows the temperature profile for two materials after a heat flux specified to raise the surface temperature from 400 K to 2,000 K. To raise the top 50 µm of pyrolytic perp to a temperature above 1,000 K, a flux of 3.8 kW/cm^2 and a duration of 10 ms is appropriate. For the more conductive 2D C/C perp., a higher heat flux for a shorter duration (35 kW/cm^2 for 1 ms) is necessary to concentrate the heating in the top 50 µm.

Numerical solutions were also obtained using the HEAT1DS code[16]. This is a 1D finite difference code which uses temperature dependent thermal conductivities. The spatial grid was set at 10 µm and time step at 10 µs. The material depth was 1 mm and initial temperature 400 K. The front and back surfaces were cooled by radiation with the emissivity set to 0.98. The solution in the region of interest did not change if the time step size or material depth was decreased by a factor of 2. The calculated surface temperatures were within 30% of the analytic solution and the differences consistent with the temperature variation of the thermal conductivity.

Figure 3 shows the temperature profiles under a heat flux aimed at heating the top 100 microns to a temperature greater than 1,000 K. A calculation for 2D C/C perp., but with 10x higher flux for 1/10 of the duration, yielded a similar heat deposition profile. In ITER the co-deposited material will lay on top of a higher conductivity substrate of carbon, beryllium or tungsten and this is likely to depress the temperatures near the interface.

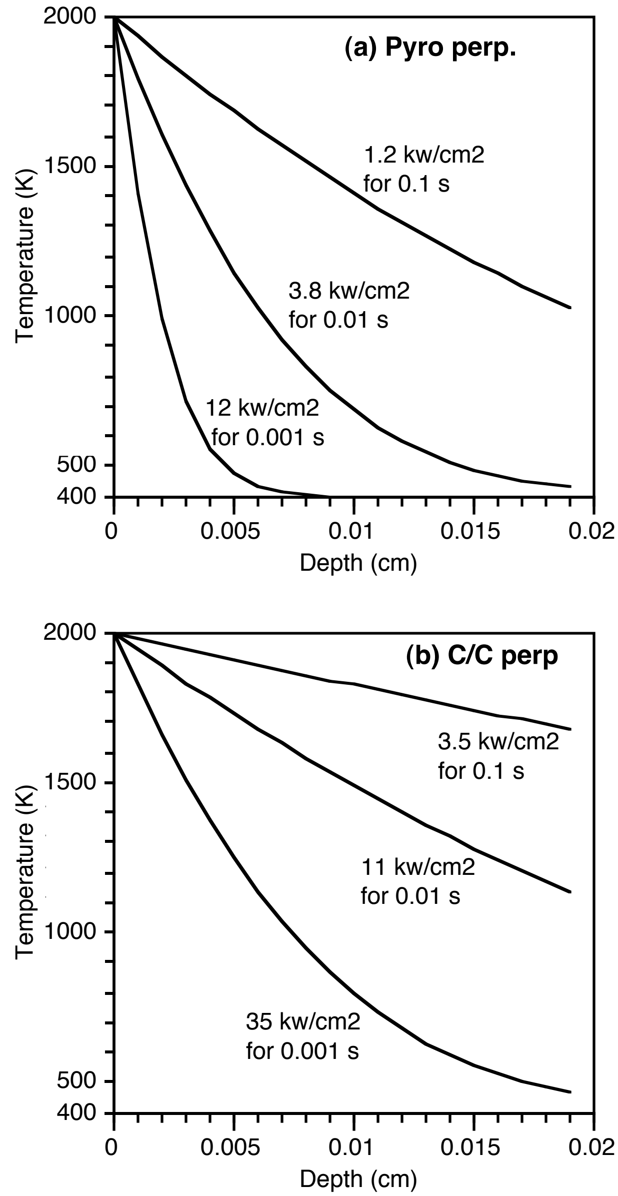

Fig. 2. Temperature vs. depth for two materials immediately after exposure to a constant heat flux of duration sufficient to raise the surface temperature from 400 to 2,000 K (from (2)).

IV. ENGINEERING ISSUES

The practical realization of this scheme needs research and development on three scales. Laboratory measurements, guided by the above calculations, are needed on co-deposited deuterium/carbon layers on tokamak tiles to measure the dependence of gas release on heat flux and duration. The change in deuterium content of the tiles would be measured by nuclear analysis. These experiments would resolve the uncertainty associated with the thermal conductivity of co-deposited graphite. Temperature limits and possible changes in the surface under intense heat would be investigated. The next step would be the implementation on a small tokamak.

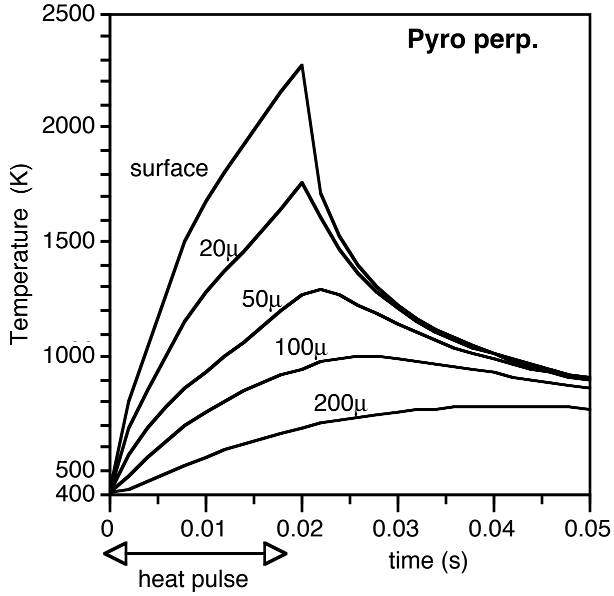

Fig. 3. Numerical calculation of temperature vs. time for pyrolytic perp. under a 3,000 W/cm^2 heat flux for 20 ms. The curves represent the temperature at the depths indicated below the surface.

Here the issues associated with beam delivery and scanning systems would be addressed. The temperature excursion up to 2,000 K would result in outgassing of hydrocarbons and other contaminants trapped in the top 100 µm of the graphite and aid plasma density control. Wall conditioning would be a major potential benefit. Major advances in plasma performance have resulted from past improvements in wall conditioning. The technique may also be useful for co-deposited carbon/tritium layers on other materials such as tungsten[20]. Pulsed laser-induced desorption has also been applied to Si, SiC, and Be[21] (although melting would be an issue with Be). The third step would be an assessment of engineering feasibility for ITER and comparison to other candidate methods for tritium removal.

Both Nd:Yag and CO_2 laser systems are potential candidates for this task. Nd:Yag lasers are available with up to 2 kW average power and the 1 µm laser wavelength has the advantage that the beam can be conveyed via fiber optics to the divertor. For the case of Fig. 3 the beam would be converted to a line focus 0.1 cm wide and 6 cm long that would be swept at 5 cm/s over the surface. Higher fluxes could be achieved with narrower line foci. The fiber optics would be retracted to a shielded container during plasma operations to avoid radiation damage. CO_2 lasers have the advantage of higher efficiencies (wall plug efficiency 10-17% compared to 2% for Nd:Yag) and higher available powers (up to 6 kW). The 10 micron wavelength beam would be directed by reflective optics. A robotic boom or vehicle would be programmed to track the divertor surfaces and a IR camera would monitor the surface temperature. The camera signal could be used to feedback control the scan speed and temperature. To permit cleaning in the presence of the magnetic field, the boom or vehicle would be vacuum compatible and be fabricated from non-magnetic materials. Significant activation and remote handling issues need to be addressed for this method to become a routine and frequent operation.

V. CONCLUSIONS

We believe that transient surface heating by a scanning laser beam merits serious consideration as a candidate for tritium removal in ITER. As with all the present schemes there are some areas that need R&D and we have outlined a graded approach to resolve the various issues.

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