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On the consequences of neutron induced damage for volumetric fuel retention in plasma facing materials

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ABSTRACT

In burning deuterium–tritium (D–T) fusion experiments, such as ITER, plasma-facing components (PFCs) will for the first time be subject to intense 14 MeV neutron bombardment, which cause displacement damage uniformly distributed throughout PFCs. A literature review indicates these displacements typically lead to hydrogenic trap sites ~1% solid concentration in refractory metals tungsten (W) and molybdenum, a level reached within ~1000 ITER shots. Simple analytic and numerical models indicate this is a concern for reaching the T fuel retention limit in ITER of 350 g, mostly due to the efficient permeation of D/T into the W allowing access to the volumetric trap sites. The sensitivity of the retention results to the incident plasma parameters, PFC temperature, surface flux balance model and plasma duty-cycle is explored. Within the range of experimental and model uncertainties a limit for an all-W ITER divertor is found ~100–1000s of shots. It is shown that ambient temperatures in excess of ~1000 K could control T inventory in a W-clad reactor despite the presence of large trap concentrations, an option which is not possible for ITER with water-cooled walls.

1. Introduction and motivation

The control of bred tritium (T) fuel will be critical for the operation of large-scale burning plasma experiments (e.g. ITER) and future magnetic fusion reactors. A particular concern is in-vessel retention of T, arising both from safety limits of "unaccounted" T, and in reactors, the requirement to recover unburned tritium from the vessel so as to not impact the global fuel cycle. These limits are both important; the former to the public perception of fusion as a safe energy source, and the latter to the scientific viability of operating a steady-state fusion device. ITER, as an example D–T burning plasma, has a regulatory in-vessel tritium inventory limit ~0.3 kg [1]. For an eventual reactor the cycling inventory of T will be ~10s kg, and given that blanket T breeding ratios (TBR) are barely above unity (typical TBR ~ 1.05), an in-vessel accumulation ~10 kg/(TBR-1) ~ 0.5 kg will deleteriously impact the fuel cycle. Therefore a conservative approach is to aim to <0.3 kg of T retention to not impact ITER or reactor operation.

The success of a burning plasma is defined by its ability to generate 14 MeV neutrons from thermonuclear D–T reactions in the plasma core. The plasma-facing components (PFCs) necessarily reside in this neutron (n) flux. The neutrons have long collision distances (typically >0.1 m) in solid materials. Therefore n-induced atomic displacements in the PFC (and other effects such as helium production) will be produced volumetrically, i.e. evenly distributed through the PFC thickness ~1–2 cm. It seems likely that these displacements could produce volumetric hydrogenic (H) traps, i.e. potential wells in the material in which H can reside out of solution.

Simultaneous with n bombardment, PFC materials are under intense H ion and atom bombardment on their plasma-facing sides. If a PFC material allows permeation of the H from the surface through its volume, this evidently raises concerns that the permeating H fuel will "find" the volumetric traps and increase fuel retention (see [2] for a review of this topic). The volume of PFCs present an enormous possible H sink; for example 1% volumetric fraction of D/T in 1 cm thick, 1000 m^2 PFC would represent ~10 kg of tritium retention. The purpose of this article is to explore the issues arising from this concern. We use the refractory metal tungsten (W) as an example PFC of concern due to its high permeation of H, and its projected use in the ITER experiment [1] and future fusion devices (e.g. ARIES-AT [3]). ITER is used as a prototypical large-scale burning plasma experiment, although the exploration is meant to be general to the use of H-permeable PFCs in a neutron environment.

2. Basic estimates

2.1. Displacement-induced volumetric traps

We wish to relate neutron-induced displacements to trap density, n_{tr} (m^{-3}). Unfortunately, no large-scale 14 MeV neutron source is available, so one must rely on experiments that bombard refractory metals, W and molybdenum (Mo) with high-energy (>MeV) ions. Such experiments "simulate" volumetric n-induced

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displacements since their range (>micron) is much larger than plasma ion implantation depths (~nm).

Measured H trap density induced by high-energy ion bombardment of refractory metals is summarized from literature sources in Fig. 1, organized by displacements per atom (dpa). Oliver et al. [4] used 800 MeV protons (p) on W to produce 0.3 and 7 dpa. The high energy of the protons leads to volumetric damage (p ion range in W ~ 0.2 m), of which ~60% is caused by spallation neutrons. Thermal desorption spectroscopy (TDS) and modeling inferred a trap/solid fraction $f_{tr} \sim 7\%$ for both dpa levels ($f_{tr} = n_{tr}/n_{solid}$, $n_{solid} = n_{W/Mo} \sim 6.5 \times 10^{28}\ m^{-3}$ is used throughout). Takagi et al. [5] and Wright et al. [6,7] exposed Mo to ^{3}He ion beams of 0.8 and 3.5 MeV energies, respectively, plus D plasma exposure, and found $f_{tr} \sim 1\%$ as measured by Nuclear Reaction Analysis (NRA). Note f_{tr} is averaged D/Mo from NRA through the ^{3}He implantation range (~1–5 μm), and therefore *assumes* 1 D/H per trap, a convention we adopt throughout for simplicity. Plotted dpa are calculated vacancies per ion (SRIM [8]) as averaged through the ion range, which seems justified given that traps were not particularly located at the end-of-range.

Overall, the trap concentration tends to plateau at $f_{tr} \sim 1\%$ and not increase strongly for >0.3–1 dpa. The reason for this may be related to an equilibrium between displacements both creating and healing traps. The "plateau" $f_{tr} \sim 1\%$ greatly exceeds typical "intrinsic" $f_{tr} \sim 10^{-5}$ for annealed refractory metals [9]. While 800 MeV p-induced spallation neutrons provide the best simulation of truly volumetric damage, TDS is difficult to interpret since the trapped H density, along with other important parameters/assumptions such as trap activation energy, must be obtained from a fit to the thermal desorption spectrum versus temperature [4]. While NRA has the advantage of directly measuring n_T, it can only be obtained up to the depth of the ion range (<10 μm); TDS diagnoses trap density throughout the bulk of the material, which is of the most interest to our study. Therefore we adopt the policy of relying on NRA for the best absolute measure of n_T, but rely on TDS for the trends.

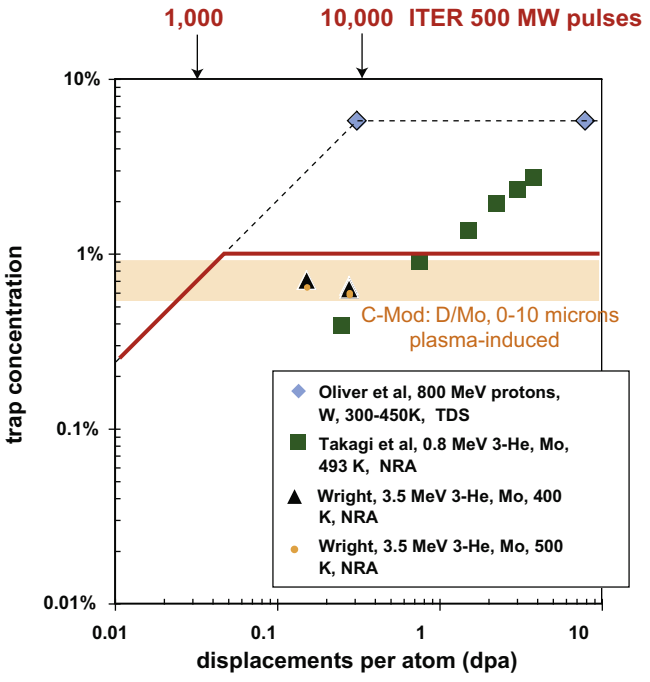

Fig. 1. Database of hydrogenic trap evolution versus displacements per atom. Literature source, irradiation conditions (beam species, energy), exposure temperature and trap diagnosis method are listed in the legend. Equivalent 500 MW ITER discharges are also shown.

For this study we adopt the solid line in Fig. 1 as the trap production rate vs. dpa, which uses Oliver et al. data to interpolate a linear increase of traps/dpa ~ 0.2 starting with un-irradiated samples ($f_{tr} = 0$), arriving at a constant $f_{tr} = 1\%$ for >0.05 dpa as measured by NRA (this sets $f_{tr} \sim 0.7\%$ at 1000 ITER shots). Obviously several other interpretations of Fig. 1 are possible and more data would be desirable, but this seems a reasonable starting point for the study. Note that at the low exposure temperatures in Fig. 1 the traps will not thermally anneal.

2.2. Simplistic permeation of H

Given that neutrons produce volumetric traps, permeation of the H to these empty traps, driven by large H flux densities at the PFC surface, should control retention rates. The standard diffusivity of H in W is taken from [10] as $D(m^2 s^{-1}) = 4.1 \times 10^{-7} \exp(-E_D/kT_{PFC})$ where $E_D = 0.39$ eV, k is the Boltzmann constant and T_{PFC} (K) is the material temperature. A simplistic analytic model [11] of H permeation in W at constant temperature can estimate if the retention rate is of concern. The H permeation "front" will be at a depth $s = (2Dt)^{1/2}$ at time t (s) of the exposure. The effective flux density of traps uncovered by the moving front will be Γ_T (H-m^2 s^{-1}) $= n_{tr}\ v_{front} = n_{tr}\ ds/dt = n_{tr}\ (D/2t)^{1/2}$, which act as a sink to the permeating H. The "source" of H can be estimated by assuming a linear H solute density n_H from the front surface where $n_H \equiv n_{H,0}$ = constant, to $n_H \sim 0$ at s. Therefore the source Γ_H(H-m^2 s^{-1}) $= D\ \nabla n_H = D\ n_{H,0}/s = n_{H,0}(D/2t)^{1/2}$. Examination of Γ_T and Γ_H informs us that if $n_{H,0} \sim n_{tr}$ then the rate of retention is *not* limited by the solute H source, and also indicates that $n_{H,0}$ is a critical boundary condition.

In Fig. 2 we show the permeation front, s, and the cumulative retention of T(g-T) $= (m_T/2)\, A_{PFC} \int \Gamma_T\ dt$, for the case of the ITER divertor (W only PFC, area $A_{PFC} \sim 210\ m^2$, triton mass $m_T \sim 5 \times 10^{-24}$ g, PFC thickness = 2 cm) assuming $n_{H,0} = n_{tr}$ and a 1:1 D:T mix. The trap density is time-dependent as described in Section 2.1, and n_T is considered permanently trapped. Fig. 2 shows that the H is highly permeable in W, reaching 1–10 mm on the timescale equivalent of 100s ITER shots (400 s/shot). As a result the T limit ~350 g can be reached in ~200–600 shots, indicating this issue could be a concern if $n_{H,0} \sim n_{tr} \sim 10^{-3}$–$10^{-2}\ n_W$. Note that here the retention rate is *faster* at higher temperature, solely due to the

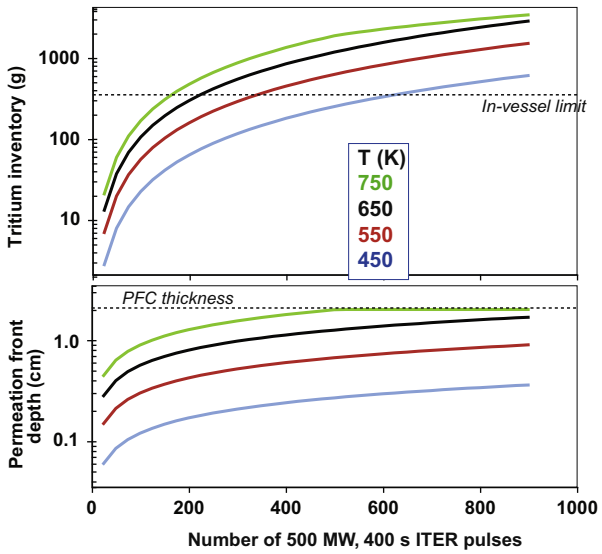

Fig. 2. ITER divertor analytic model with $n_{H,0} = n_{tr}$. Trap/dpa = 0.2. (Lower panel) Permeation distance into W. (Upper panel) Cumulative total T retention with no detrapping.

higher H diffusivity. This simply points out that the benefits of high T_{PFC} (examined below) in reducing retention must be balanced against the enhanced permeation of H to the traps.

3. Scoping T retention in ITER with a numerical model

Beyond the simple analytic model above, one must use a numerical model to scope the sensitivity of the expected T retention to $n_{H,0}$, $T_{PFC,}$ exposure history, etc. The purpose of the model is to have a simplified material model (compared to say TMAP [12]) but with highly flexible time and spatial dependencies and rapid execution. The 1-D slab model solves the coupled continuity equations for solute H, n_H, and trapped H, n_T, namely:

$$\frac{\partial n_H}{\partial t} + D\nabla_s^2 n_H = -R_{trap} n_H + R_{de\text{-}trap} n_T \quad (1)$$

$$\frac{\partial n_T}{\partial t} = +R_{trap} n_H - R_{de\text{-}trap} n_T \quad (2)$$

Traps, and therefore trapped H, are taken to be immobile (uniform in s). The form of trapping and de-trapping rate are taken from [12] as

$$R_{trap} = \frac{D n_{tr}}{\lambda^2 n_W} \qquad R_{de\text{-}trap} = \nu \exp\left(\frac{-E_{tr}}{kT_{PFC}}\right) \quad (3)$$

where λ is the distance between *empty* traps, $\nu \sim 10^{13}\ s^{-1}$ is the bounce frequency of H in traps and $E_{tr} \sim 1.5$ eV is the trap activation energy, typical of E_{tr} found in the studies of Section 2.1. The W PFC thickness is 2 cm. A zero-gradient n_H is imposed on the back of the deepest slab, which is equivalent to placing a permeation barrier between the W PFC and heat sink.

The numerical model is used to study the effect of changing the ratio $n_{H,0}/n_{tr}$ in Fig. 3 for a simple case of constant f_{tr} and no de-trapping ($R_{de\text{-}trap} = 0$). The analytic model of Section 2.2 (dashed line in Fig. 3) is close to the $n_{H,0}/n_{tr} = 1$ case, as expected. If $n_{H,0}/n_{tr} > 1$ then the exponential-like leading trail ahead of s_{front} (from actually solving the diffusion equation) allows the deep traps to fill even faster, with all the traps being filled eventually. Conversely if $n_{H,0}/n_{tr} < 1$ the retention continues to increase with time (again due to the finite H source caused by the leading trail) but at a decreased rate.

We use two models to establish a reasonable bound on $n_{H,0}/n_{tr}$. In general we expect $n_{H,0}$ to be set by the dominant rate-limiting process for release of H from the surface, which is roughly equal to the incoming H flux of ions/atoms, $\Gamma_{H,in}$ from plasma contact (small retention rates normalized to incident flux density are assumed here). The "implantation" model assumes that diffusion of the implanted H back to the surface through the distance $s_{implant}$ is the rate-limiting process, so that in equilibrium $n_{H,0} = \Gamma_{H,in} s_{implant}/D$, assuming $n_H = 0$ at $s = 0$. This model tends to produce the minimum possible $n_{H,0;}$ D_H in W is large and shallow implantation depths (~1–10 nm) are expected in the divertor and surface recombination is not required. The "recombination" model assumes the rate limiting process is recombination of H into volatile H_2 at the surface, so that in equilibrium $n_{H,0} = (2\Gamma_{H,in}/R)^{1/2}$. The recombination rate coefficient $R\ (m^4\ s^{-1}) = 5 \times 10^{-30} \exp(-0.19\{eV\}/kT)$ is taken from DIONISOS for 30 eV D^+ exposure [6]. It is important to note that this R was *inferred* based on direct DIONISOS measurements of $n_{H,0}$ during plasma exposure, which found $n_{H,0}/n_{Mo} \sim 0.1$–1% for $\Gamma_{in} \sim 2 \times 10^{21}\ m^{-2}s^{-1}$. While arising from a direct measurement of $n_{H,0}$, the DIONISOS R trends towards lower value of R from other literature sources [9], thus we consider this an *upper* bound on $n_{H,0}$. Note the two models have different power dependencies on Γ_{in} and different T_{PFC} trends due to different activation energies. For ITER simulations we use the values for Γ_{in} and surface T_{surf} as listed in Table 1, which distribute the total divertor flux $\sim 10^{25}\ s^{-1}$ [13] and divertor power (~50 MW) into three discrete zones. This is meant to be indicative of expected exposure conditions in a large-scale burning plasma, rather than strictly predictive of ITER.

We have scoped the effect of changing exposure parameters, such as imposing a realistic T gradient across the PFC from the front surface to the heat sink (T = 428 K in all cases) and the effect of evolving versus constant f_{tr}. Due to limited space, we highlight an interesting sensitivity of the retention to plasma duty cycle in Fig. 4. For simplicity, retention calculations often assume the plasma exposure and T_{PFC} are constant, however in reality ITER will be a pulsed device with a nominal duty cycle of 20%. One may hope that the periodic removal of Γ_{in}, which drives the permeation through $n_{H,0}$, would impede the access of H to the traps. However, Fig. 4 indicates that the retention (per shot) can be *increased* in many cases. This is due to two effects. First the removal of the plasma flux also removes the power flux and the PFC returns to low T = 428 K, which slows diffusion and precludes significant de-trapping. This tends to "freeze" the H profile. Secondly, since D_H is finite between shots, the n_H gradient tends to relax both towards the surface *and* deeper into the bulk. However the "emptied" n_H profile near the surface is nearly instantaneously refilled when the plasma flux returns, continuing to drive permeation. The result is a ratcheting effect for solute and trapped H deeper in the surface. This result highlights the importance of *ambient* (i.e. heat sink) PFC temperature, although the relative sensitivity to cycling changes from one exposure condition to the next.

Sample results for a W divertor in ITER are shown in Fig. 5. This highlights the general result of the scoping study that the retention is most sensitive to the surface model. Shown is the "realistic" exposure model: linear temperature gradients through the PFC, 20% duty cycle and evolving trap density. This can be compared in Fig. 5 to the limit reached using the usual "simple" exposure: constant $T(s) = T_{surf}$, 100% duty cycle and fixed trap fraction of 1%.

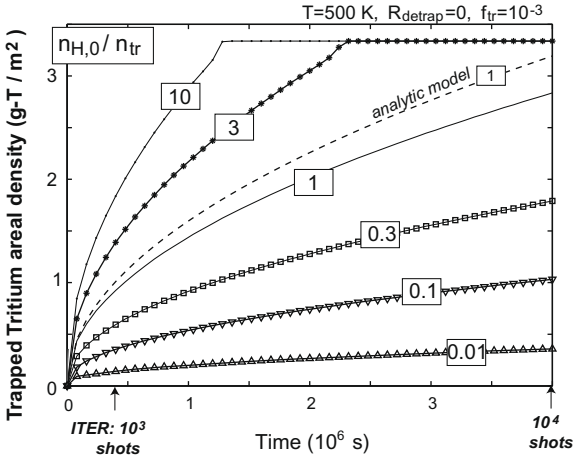

Fig. 3. Effect of changing ratio $n_{H,0}/n_{tr}$ in cumulative tritium retention from numerical model. Case shown: T = 500 K, constant $f_{trap} = 10^{-3}$, $R_{detrap} = 0$.

Table 1
Description of plasma flux, incident particle energy and surface temperature for three ITER W-divertor "zones" used in numerical simulations. Total particle flux to divertor (area = 210 m²) is 10^{25} ion/s (strikepoint + baffle) and 10^{25} atoms/s (baffle) and total power to divertor delivered by particles is 54 MW. Two centimeter thick W PFC, thermal conductivity ~180 W/m/K is used.

Region	Area (m^2)	Γ_i ($m^{-2}s^{-1}$)	$E_{incident}$ (eV)	q (MW m^{-2})	T_{surf} (K)
Strikepoint	8	6×10^{23}	30	2.9	~760
Baffle	112	4.8×10^{22}	30	0.23	~460
Dome	90	5.6×10^{22}	5	0.04	~440

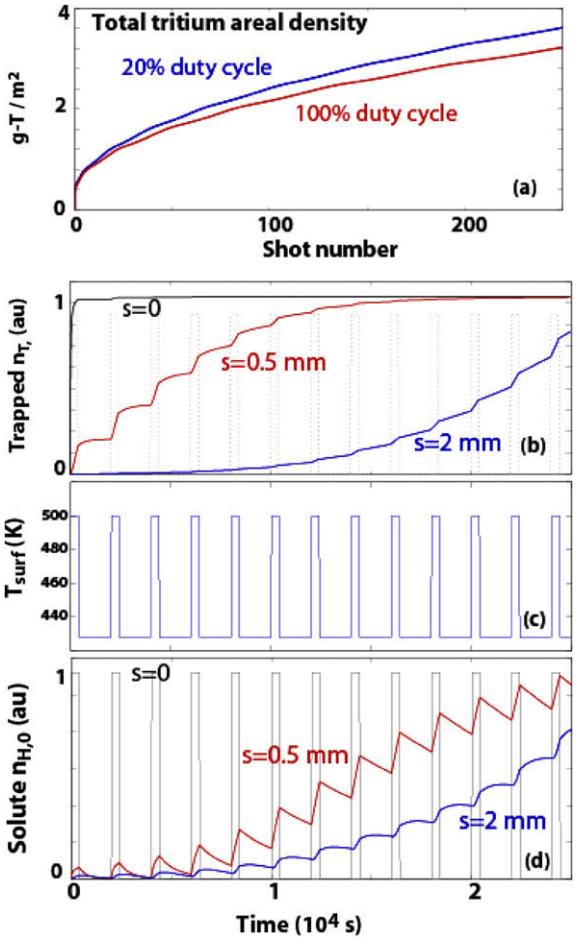

Fig. 4. Effect of 20% duty cycle plasma exposure. Case shown: $f_{trap} = 7 \times 10^{-3}$, "baffle" region of Table 1. (a) Cumulative retained T areal density between 100% and 20% duty cycle versus plasma shot number (b)–(d) 20% duty cycle. (b) Trapped H density, n_T, at different depths s, (c) surface temperature showing thermal pulsing, (d) solute H density, n_H at different depths. For different s values the amplitudes of n_T and n_H have been arbitrarily scaled for viewing clarity.

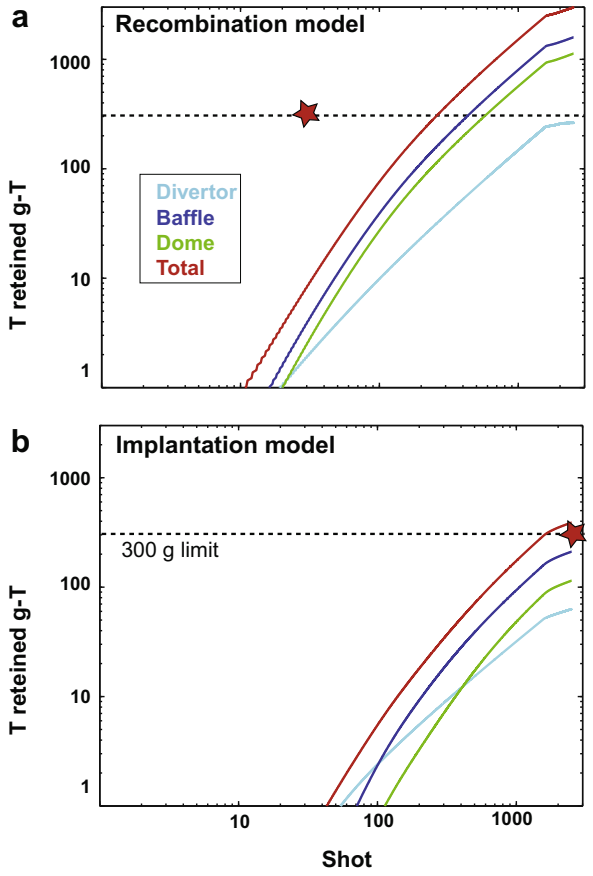

Fig. 5. Summary of T retention results for ITER. Lines show "Full" exposure model: T gradient through PFC, 20% duty cycle, evolving f_{tr} as in Fig. 1. (a) Recombination surface model (b) Implantation surface model. Stars indicate shot where limit T = 350 g is reached with "simple" exposure model described in text, but the same surface model.

In fact the exposure model has opposite effects with the two surface models: increasing the limit to ~200 shots from ~20 shots for the recombination model and decreasing the limit to ~1000 shots from ~2500 shots for the implantation model. We note this last result is in good agreement with DIFFUSE calculations [14] which indicate a limit ~2500 shots using a similar surface and exposure model (benchmark cases for specific exposure conditions of our model are also within < factor of two of DIFFUSE results). The model is therefore deemed numerically accurate, however this exercise indicates that the expected results for retention shot limits in ITER are sensitive to assumptions in the modeling.

4. Discussion and conclusions

The results of Fig. 5 indicate ITER scenarios where the T limit may be met in 100s–1000s of shots due to T retention in a W divertor. It is important not to view these as "predictions", since no present model (including our simple one) can capture the true complexity of the evolutions of PFC surface and materials in ITER (for example it will not be true that all plasmas will be the same Table 1, low-Z impurities will likely be present in the divertor, disruptions are ignored, etc.). *Rather this serves as a gross indication that volumetric trap production is a concern with refractory metal PFCs due to a simple picture of permeation to volumetric traps.* For ITER this serves to indicate that one may likely require recovery of tritium from the bulk of W PFCs during its operational lifetime, which is not addressed in the present operational scenario (for example maximum divertor bake T ~ 240 C [1] is inadequate for T desorption from the bulk). Looking past ITER to a steady-state device the optimistic limit estimates presented here of ~2500 shots are the equivalent of only ~10 days exposure, obviously a concern if operation for ~1 year are expected.

A simple model indicates that the *key to controlling T retention in W with n-induced traps is, not surprisingly, ambient temperature*. If one considers a steady-state reactor/DEMO we can reasonably expect that (a) the PFC/blanket temperature will be required to be high (>900 K) for thermal–electric conversion efficiency (e.g. ARIES [3]) and (b) therefore the H will be fully permeated through the PFC depth. An unknown is the equilibrium n_{tr} that would be achieved by combining continuous n damage plus annealing from relatively high T_{PFC} (compared to T_{PFC} of data in Fig. 1). Rather we consider the pessimistic assumption that f_{tr} will remain ~1%, but that de-trapping will be sufficiently strong to leave the traps empty. Taking a steady state solution ($\partial/\partial t \to 0$), Eq. (3) can be rewritten to solve for f, the fraction of traps that are filled, as:

$$\frac{f}{(1-f)^{2/3}} = \frac{D\, n_{tr}^{2/3}\, n_{H,0}}{n_W\, v_{bounce}\, \exp(-E_{trap}/kT)} = \frac{n_{tr}^{2/3}\, \Gamma\, s_{implant}}{n_W\, v_{bounce}\, \exp(-E_{trap}/kT)} \quad (4)$$

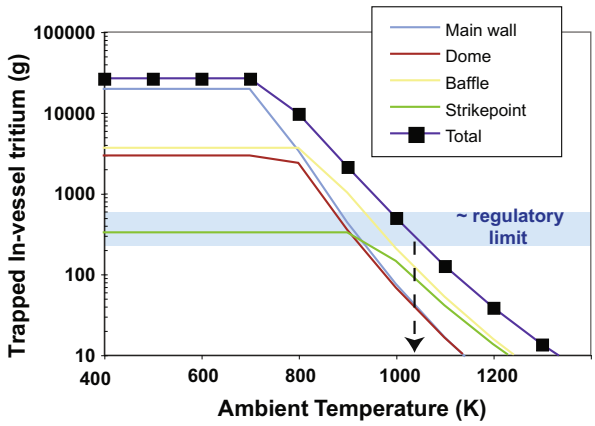

Fig. 6. Total T retention for ITER as if operating steady-state with varying ambient constant PFC temperature. Case: f_{tr} = 1%, implantation model (Eq. (4)).

where the spacing between empty trap sites is estimated as $\lambda \sim (n_{tr}-n_T)^{-1/3} = n_{tr}^{-1/3}(1\text{-}f)^{-1/3}$. In the last term of Eq. (4) we substitute the implantation model for $n_{H,0}$ which removes the dependence on D. Taking $f_{tr} \sim 1\%$ and the ITER exposure/T_{PFC} model (Table 1) plus a low flux-density W main wall (area $\sim$600 m^2, $\Gamma_{in} \sim 10^{21}$ m^{-2}s^{-1}) produces the results shown in Fig. 6. This calculation indicates that ambient $T > 1000$ K are required to reduce the retained T to acceptable levels, but this can be achieved even in the presence of large $f_{tr} \sim 1\%$. Again this should be taken as indicative (rather than predictive) that for a fixed exposure condition there will exist a sharp minimum ambient T_{PFC} for operating the W PFCs in order to *assure* acceptable T retention levels.

The preceding exercise also illustrates the difficulty in vetting PFC T retention in ITER or any pulsed, water-cooled device where maximum ambient T < 500 K. In such devices, the PFC is heated only simultaneously with plasma flux, while intra-shot periods freeze the solute and trapped H deep in the PFC. This is the worst-case scenario for driving permeation in the presence of volumetric traps caused by neutrons in a successful burning plasma experiment. Therefore, one cannot expect the T retention of W in ITER to be indicative of what will occur in a reactor. This observation also affects the rationale for using W in ITER; one should not necessarily expect W to readily "solve" tritium retention, but rather one is motivated to test other aspects of W such as its erosion resistance, the effect of W on the burning plasma performance and neutron-damage resistance.

Beyond the obviously strong influence of temperature, the present study clearly motivates more experiments and modeling towards better understanding trap production/healing mechanisms in different radiating environments, deep permeation of H in W, and the complex coupling of the two.

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