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Estimation of the tritium retention in ITER tungsten divertor target using macroscopic rate equations simulations

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Abstract

Based on macroscopic rate equation simulations of tritium migration on an actively cooled tungsten (W) plasma facing component (PFC) using the code MHIMS (migration of hydrogen in metals), an estimation has been made on the tritium retention in ITER W divertor target during a non-uniform exponential distribution of particle fluxes. Two grades of materials are considered to be exposed to tritium ions: an undamaged W and a damaged W exposed to fast fusion neutrons. Thanks to the simulation, the evolutions of the tritium retention and the tritium migration depth are obtained as a function of the implanted flux and the number of cycles. From these evolutions, extrapolation laws are built to estimate the number of cycles needed for tritium to permeate from the implantation zone to the cooled surface and to quantify the corresponding retention of tritium throughout the W PFC.

I. Introduction

Tritium retention in plasma facing components (PFC) is an important issue for fusion devices such as ITER and DEMO. For instance, the total tritium inventory in the vacuum vessel is limited to 700 g in ITER. In ITER, the main vector of retention is expected to be the retention in Be materials and Be codeposited layers as shown from JET experiments [1, 2] and WALLDYN simulations [3]. On the other hand, the tritium retention in tungsten (W) is also as important concern since the ITER divertor is made of W and in a full W DEMO design, it will be the major vector of tritium retention.

The present study intends to estimate what could be the contribution of tritium retention in W divertor targets to the total tritium inventory in ITER and DEMO. To tackle this issue, one dimensional (1D) macroscopic rate equations (MRE) models are an efficient way to investigate the behavior of tritium in tungsten at the scale of an ITER actively cooled PFC. Such models take into account diffusion and trapping of hydrogen isotopes. They are extensively used to

simulate the migration and the trapping of deuterium in tungsten in well-controlled experiments [4, 5, 6] or tokamak relevant environment [7, 8, 9]. During the deuterium/tritium phase in ITER, fast neutrons (14.1 MeV) will be created, damaging the W PFCs lattice which can change their retention properties. It is thus important to distinguish, in the MRE models, two different types of materials: the materials only exposed to the tritium and the materials which are also damaged by neutrons.

In this study, the code MHIMS (Migration of Hydrogen Isotopes in Metals) [4, 5] is used to simulate the tritium retention in PFCs with these two material properties under ITER relevant conditions of thermal and particle fluxes during steady-state operation for several plasma cycles. Up to 40 cycles have been simulated and from the simulation results, simple extrapolation laws have been built to estimate the tritium retention for larger number of cycles and more precisely, up to the number of cycles needed for the tritium to reach the cooling system.

II. Model parametrization and irradiation conditions

The code MHIMS is based on a standard 1D MRE models that considers two types of particles, the trapped particles and the mobile particles. The mobile particles can diffuse and fall into trapping sites (that represent defects such as vacancies, dislocations, impurities …) feeding the trapped population of particles. The equations have been extensively described in previous paper [4, 5]. The key material parameters of the models are the diffusion coefficient of the mobile particles D(T), the concentration of traps n_i and the detrapping energy $E_{t,i}$ for the traps of type i. The temperature T is of course an important parameter since all the diffusion and trapping processes are thermally activated. In the equations, the implantation of ion in the material is treated as a volume source of mobile particle in the approximation of low trap concentrations. This source depends on the incident flux and the ion energy of incident ions.

Concerning the materials parameters, Density functional theory (DFT) calculations by Fernandez *et al.* [10] show that hydrogen in W diffuses between tetrahedral interstitial sites with an energy barrier of 0.2 eV and $D_H(T) = 1.9 \times 10^{-7} \cdot e^{-\frac{0.2\ (eV)}{k_B \cdot T}}$ $m^2 s^{-1}$. To obtain the diffusion coefficient for tritium (the hydrogen isotope investigated in this study), the pre-exponential factor for H is divided by $\sqrt{3}$, the square root of the mass ratio between tritium and hydrogen. The trapping parameters (concentration of traps and detrapping energies) have been parametrized in previous study [4, 5]. We will distinguish between two kinds of materials: the material only exposed to tritium ions referred to as “undamaged W” and the material damaged by neutrons called “neutron-damaged W”. The detrapping energies and concentrations of traps for the former has been parametrized on annealed W [6] and for the latter, the parameters have been determined on self-damaged W (W damaged by 20 MeV W ions) that is supposed to mimic the

damaging by neutrons [11, 12]. The parametrization results are presented in [4] for the undamaged W and in [5] for the neutron-damaged W and summarized in table 1. Up to 6 different trapping sites are used: 2 intrinsic traps, an ion-induced trap and 3 neutron-induced traps. The ion-induced trap represents defects that are created by the presence of a large concentration of hydrogen isotopes in the lattice during the implantation. Thus, the concentration of this trap increase over time with the incident fluence. The equation giving this evolution was originally proposed by Ogorodnikova *et al.* [6] and implemented in MHIMS [4]. Undamaged W contains only the intrinsic traps and the ion-induced trap, whereas there are all trap types in the neutron-damaged W. It is supposed that the concentrations of the neutron-induced traps are constant over all the depth of the simulated materials which seems to be a not-so-crude approximation according to Gilbert et al. [13]. In addition, it is supposed that the concentrations of neutron-induced traps saturate with the damage dose as in the case of the self-irradiation-induced traps as shown by 't Hoen *et al.* [14] and the concentrations of neutron-induced traps ar considered to be all saturated.

The simulations intend to present the migration of tritium in a 1-cm-thick actively-cooled W PFC in the entire thickness of the PFC. Thus, the actively cooled feature has to be taken into account to calculate the temperature through the PFC material, i.e. from the plasma facing surface to the cooled surface. This is done with a simple 1D thermal diffusion model added to MHIMS that is similar to the one presented by Sang *et al.* [15] from which we took the numerical values of the thermal properties of W.

At the cooled surface, the temperature is controlled by a boundary condition described by Denis *et al.* [16] where the temperature of materials is allowed to rise above 343K, the temperature of the cooling system. At the plasma facing surface, the heat flux Γ_{th} that increases the temperature in all the material is calculated from the incident flux of ions ϕ_{inc} and their energy $\mathrm{E_{ion}}$ via the following formula: $\Gamma_{\mathrm{th}} = \phi_{\mathrm{inc}} \cdot \mathrm{e} \cdot (\mathrm{E_{inc}} + 13.6\ \mathrm{eV})$ with $\mathrm{e} = 1.6 \times 10^{-19}$ C and 13.6 eV the energy of the electron-hydrogen recombination. Thus, the temperature in all the material is determined by the incident flux of particle, their energy and the temperature of the cooling system (343 K).

Several plasma cycles are simulated. A single plasma cycle is divided into 4 phases: a 20 s plasma ramp up during which ϕ_{inc} increases from 0 to its nominal value, a 380 s plasma burning phase with a constant incident flux, a 40 s plasma ramp down during which ϕ_{inc} decreases back to 0 and a final 960 s recovery period during which the fluxes are 0. As the code cannot handle different hydrogen isotopes, the simulated flux is composed of 100 % tritium. In tokamaks such as ITER or DEMO, the flux would be made of deuterium and tritium. A mixed deuterium/tritium flux

would have few effects on the migrations since the diffusion coefficient is just change by a factor $\sqrt{3/2}$ but concerning the retention, it would divided the results obtained by the simulation by 2.

According to scaling laws proposed by Eich *et al.* [17], the thickness of the scrap-off layer at the mid plane is expected to be 1 mm. Taking an expansion factor of 10, the thickness of the flux deposition can then be expected to be 1 cm in ITER. Thus, we decided to simulate an exponential distribution of flux with the coordinate s along the target in a poloidal section with a decay length of $\lambda_\phi = 1$ cm: $\phi_{\mathrm{inc}}(s) = \phi_{\mathrm{inc}}^{\mathrm{MAX}} \cdot \exp(-\frac{s}{\lambda_\phi})$ (black diamond in figure 1). The maximal incident flux we use is 10^{24} $m^{-2}s^{-1}$ and hit the target at what is called strike point in the next (s=0). Here, the energy of the impinging tritium ions is 25 eV/ion which is the order of magnitude of incident ion energy in tokamaks [8, 18]. Thus, the maximal heat flux is 6.2 MW/m^{-2} and the maximal temperature, called T_H, is 970 K. Due to the non-uniform ion flux distribution, the temperature is also non-uniform along the target (inset in figure 1). The reflection coefficient r for 25 eV ions implanted on a W surface being $r \approx 0.7$ [19], the maximum implanted flux $\phi_{\mathrm{imp}} = (1 - r) \cdot \phi_{\mathrm{inc}}$ is equal to 3×10^{23} $m^{-2}s^{-1}$. It has to be pointed out that the Eich scaling law are focused on attached divertor conditions. In semi-detached plasma operations foreseen in ITER, a further broadening and a lower maximum particle flux is expected. Thus, the simulation results presented here can be considered as an upper limit for the estimation of the tritium retention.

III. Simulation results

The MHIMS simulations give the evolution of the tritium inventory, $\mathrm{Inv}_{\mathrm{trit}}$ in tritium/m^2 over time/cycles. Figure 2 shows the distribution of $\mathrm{Inv}_{\mathrm{trit}}$ given by the MHIMS simulations for different ϕ_{imp} in undamaged W (a) and neutron-damaged W (b). The coordinate s is also display as well as the temperature T_H on the top x axis.

In the undamaged W, the tritium retention exhibits a maximum for $\phi_{\mathrm{imp}} = 6 \times 10^{22}$ $m^{-2}s^{-1}$ ($T_H = 449$ K). Intuitively, a monotonous increase of tritium retention with the flux is expected since it means an increase of the fluence which implies an increase of the retention [6]. However, the behavior we observe in our simulations is due to the non-uniform temperature distribution on the target. Indeed, in undamaged W, tritium can only be trapped in the low detrapping energy traps (intrinsic) and the ion-induced traps (see table 1). With such detrapping energies and for a constant fluence, the retention generally decreases with temperature increase and drops by several order of magnitude above 600 K [4]. Thus, in the simulation, we observe a balance between the effect of an increasing fluence (that makes the retention grow) and an increasing temperature (that makes the retention drop).

In the neutron-damaged W the tritium retention increase with ϕ_{imp} as intuitively expected. In fact, there is also a maximum of the retention which is not observed precisely at the position of the maximum implanted flux, but very close to it ($\phi_{imp} = 2.7 \times 10^{23}$ $m^{-2}s^{-1}$ with $T_H = 894$ K). The difference as compared to undamaged W is due to the presence of high detrapping energy neutron-induced traps. They can easily retain tritium up to 900 K [5] and the balance between the increasing fluence and the increasing temperature occurs at a much higher temperature than in undamaged W.

The ITER major radius is $R = 6$ m and there will be two strike points (inner and outer divertor) that we suppose identical. Assuming a toroidal symmetry of the flux deposition, and by integrating the curves $Inv_{trit}(s)$ along the coordinate s for undamaged W (figure 2 (a)) and for neutron-damaged W (figure 2 (b)) at 40 cycles, the tritium inventory in the divertor of the tokamak INV_{div} (gram of tritium) can be obtained as: $INV_{div} = 2 \cdot \left(2\pi \cdot R \cdot \int Inv_{trit}(s) \cdot ds\right)/(2 \times 10^{23}\ \text{tritium}.\,g^{-1})$. 2×10^{23} (tritium·g^{-1}) is the number of tritium atoms in 1 gram of tritium. After 40 cycles, the INV_{div} is 0.21 g (or 0.105 g for a mixed deuterium/tritium flux) in undamaged W and 0.49 g (or 0.245 for a mixed deuterium/tritium flux) in neutron-damaged W which is only 2.3 times higher. However, at the strike point, the ratio between the neutron damaged W and undamaged W is much higher (15.7) underlining the impact of the non-uniform distribution of implanted flux and temperature.

The simulations also give the evolution of the tritium depth profile for the different implanted fluxes. Here, we are interested in R_m^c which is the depth reached by tritium after a given number of cycles. Figure 3 shows the evolution of R_m^c extracted from the MHIMS simulations as a function of the coordinate s for the undamaged W (a) and the neutron-damaged W (b). As for the tritium inventory, the results for the highest number of cycles are an extrapolation that will be discussed in the next section. In addition to the coordinate s, the temperature is also displayed in the y axis as well as the position of the cooling system.

First, after 40 cycles, the value of R_m^c in undamaged W (8 mm) is much higher than in neutron-damaged W (300 µm). This is due to the presence of high energy traps in neutron-damaged W which slows down the migration of tritium.

In both cases, the value of R_m^c decreases as the coordinate s increases as could be expected: as the temperature decreases, the diffusion of tritium slows down. However, the decrease of R_m^c is smooth in the neutron-damaged W while it is not in the undamaged W. In the latter, a stronger decrease appears between 0.005 m and 0.020 m which correlates with the bump observed in the retention-versus-coordinate plot (figure 2 (a)). In this area, due to the decrease of temperature with the coordinate s, there is a quick transition between a favorable detrapping regime (i.e. a fast

migration regime) and a favorable trapping regime (i.e. a slow migration regime). The position at which there is a maximum retention is in this transition zone and more precisely at its beginning (figure 3 (a)): a favored trapping process is more important for the total retention than the depth up to which tritium is trapped. This transition zone is small on the neutron-damaged W again because of the high detrapping energy traps: the trapping is favored in all the temperature range used in these simulations.

IV. Discussion and conclusions

For a given implanted flux, both the tritium inventory Inv_{trit} and the depth reached by tritium R_m^c grow with the number of cycles N_c. From the simulation results, power exponential extrapolation laws can be built: $Inv_{trit}(N_c, \phi_{imp}) \propto N_c^{\alpha_{Inv}(\phi_{imp})}$ and $R_m^c(N_c, \phi_{imp}) \propto N_c^{\alpha_R(\phi_{imp})}$. The dependence of α_{Inv} and α_R with ϕ_{imp} are extracted from the simulation results and displayed on figure 4 for both materials. For a migration limited regime, these exponents should be around 0.5 as already discussed in many papers [4, 19, 20] which is the case for both materials in the medium flux range. For low implanted flux, the migration depth is in the range of the area where ion-induced traps are created (table 1 and figure 3). Because of that, the migration is slowed down ($\alpha_R < 0.5$) and the increase of the retention is enhanced ($\alpha_{Inv} > 0.5$) compared to a pure migration limited regime [4]. In undamaged W, for high value of ϕ_{imp} we have $\alpha_{Inv} \approx \alpha_R < 0.5$ (less than 0.2 at the strike point). This behavior is due to the temperature gradient in the material and the 10 times faster migration of tritium in undamaged W (figure 3) explaining why it is not observed in neutron-damaged W.

From these extrapolation laws, we estimate the number of cycles needed for tritium to reach the cooling system. For undamaged W, this occurs after 347 cycles. The estimated distribution of the retention along the target is displayed in figure 2 (a) and the estimated R_m^c is shown on figure 3 (a). There is still a maximum of retention for the same flux value and there is still the strong decrease between 0.005 m and 0.020 m. The retention in the divertor of the tokamak is estimated to be 0.55 g (or 0275 g for a mixed deuterium/tritium flux). For the neutron-damaged W, the permeation of tritium occurs after a much higher number of cycles, 52, 519 cycles. The estimated distribution of the retention along the target is displayed in figure 2 (b) and the estimated R_m^c is shown on figure 3 (b). The retention in the divertor of the machine is estimated to be 14.91 g (or 7.455 g for a mixed deuterium/tritium flux). This is also 27 times higher than the retention in undamaged W. If one only takes into account the retention at the strike point, a much higher ratio of 267 would be obtained. This shows once more the importance of taking into account the non-uniform distribution of flux and temperature. These non-uniform distributions tend to increase the retention in undamaged W

(due to a visible transition between detrapping favorable and trapping favorable regimes) and it tends to decrease the retention in neutron-damaged W (due to a less visible transition and a decreasing fluence along the coordinate from the strike point). In both cases, extrapolations tend to show that tritium retention in the divertor of ITER alone will not reach the 700 g safety limit before the tritium atoms permeate trough the full W PFC. However, what is presented here is a semi-2D study that does not take into account the gradients of both temperature and particle concentrations transversally to the surface. The former would decrease the maximum temperature leading to a higher retention and the latter would rather flatten the tritium concentration decreasing the behavior observed in figure 3 (a) in undamaged W. In addition, the study does not include the effect of transients such as edge localized mode, the possible change of the divertor during maintenance period or any tritium removal technique (baking, heating or isotope exchange). It also does not take into account tritium retention in the first wall and in deposited layers which, has already said in the introduction, will be the main contribution to the tritium retention in ITER.

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Reference

[1] S. Brezinsek *et al*., *Nuc. Fusion 53,* p. 083023, 2013.

[2] K. Heinola *et al*., *J. Nucl. Mater. 463,* pp. 961-965, 2015.

[3] K. Schmid *et al*., *Nuc. Fusion 55*, p. 053015, 2015.

[4] E. A. Hodille *et al*., *J. Nucl. Mater. 467,* pp. 424-431, 2015.

[5] E. A. Hodille *et al*., *Nucl. Fusion*, p. 056002, 2017.

[6] O. V. Ogorodnikova *et al*., *J. Nucl. Mater. 313-316*, pp. 469-477, 2003.

[7] X. Bonnin *et al*., *J. Nucl. Mater. 463*, pp. 970-973, 2015.

[8] K. Schmid, *Phys. Scr. T167*, p. 014025, 2016.

[9] A. Hu and A. Hassanein, *J. Nucl. Mater 465,* pp. 582-589, 2015.

[10] N. Fernandez *et al*., *Acta Mater. 94,* pp. 307-318, 2015.

[11] A. Zaloznik *et al*., *Phys. Scr. T167,* p. 014031, 2016.

[12] S. Markelj *et al*., *J. Nucl. Mater. 469,* pp. 133-144, 2016.

[13] M. R. Gilbert *et al*., *Nucl. Fusion 52,* p. 083019, 2012.

[14] M. H. J. 't Hoen *et al*., *Nucl. Fusion 52,* p. 023008, 2012.

[15] C. Sang *et al*., *Nucl. Fusion 52,* p. 043003, 2012.

[16] J. Denis *et al*., *Phys. Scr. T167,* p. 014073, 2016.

[17] T. Eich *et al*., *Nucl. Fusion 53,* p. 093031, 2013.

[18] J. Roth *et al*., *J. Nucl. Mater. 390-391,* pp. 1-9, 2009.

[19] O. V. Ogorodnikova *et al*., *J. Appl. Phys. 119,* p. 054901, 2016.

[20] R. Bisson *et al, J. Nucl. Mater. 467,* pp. 432-438, 2015.

Table Caption

Table 1. Trap parameters used in the simulations. For undamaged W, the neutron-induced traps are not used.

Figure Caption

Figure 1. Non-uniform distribution along the W target of the implanted tritium flux ϕ_{imp} (black diamonds) and corresponding temperature T_H (grey circles in the inset).

Figure 2. Evolution of the tritium retention as a function of the implanted flux for different number of cycles in undamaged W (a) and in neutron-damaged W (b). The coordinate s from the strike point and the corresponding temperature are displayed on the top x axis as well as the position of the maximum of retention for the undamaged W.

Figure 3. Evolution of the migration depth of tritium atoms R_m^c for different coordinate s from the strike point (for different implanted fluxes) and for different number of cycles in undamaged W (a) and in neutron-damaged W (b). The temperatures are displayed on the undamaged W case (they are the same for the neutron-damaged) as well as the position of the maximum of retention for the undamaged W only. For each cycle, the results of the simulations (symbols) defined a line: on the left side of this line, tritium is present and there is none on the right side of this line.

Figure 4. Dependence of power laws exponents for the tritium inventory α_{Inv} (circles) and for the depth rached by tritium α_R (square) with the implanted flux ϕ_{imp} for undamaged W (grey) and neutron-damaged W (black) extracted from the MHIMS simulations.

Table 1

Intrinsic 1	$E_{t,1} = 0.85$ eV $n_1 = 0.13$ at. %
Intrinsic 2	$E_{t,2} = 1.00$ eV $n_2 = 0.035$ at. %
Ion-induced	$E_{t,3} = 1.5$ eV $n_3^a = 8$ at. % up to 10 nm $n_3^b = 1$ at. % up to 10 µm
Neutron-induced 1	$E_{t,4} = 1.65$ eV $n_4 = 0.11$ at. %
Neutron-induced 2	$E_{t,5} = 1.85$ eV $n_5 = 0.2$ at. %
Neutron-induced 3	$E_{t,6} = 2.06$ eV $n_6 = 0.05$ at. %

Figure 1

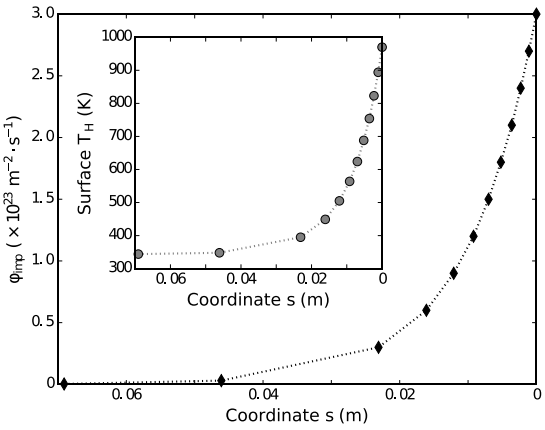

3.0
2.5
2.0
1.5
1.0
0.5
0
Φ_{imp} ($\times 10^{23}$ $m^{-2} \cdot s^{-1}$)
0.06
0.04
0.02
0
Coordinate s (m)
1000
900
800
700
600
500
400
300
Surface T_H (K)
0.06
0.04
0.02
0
Coordinate s (m)

Figure 2

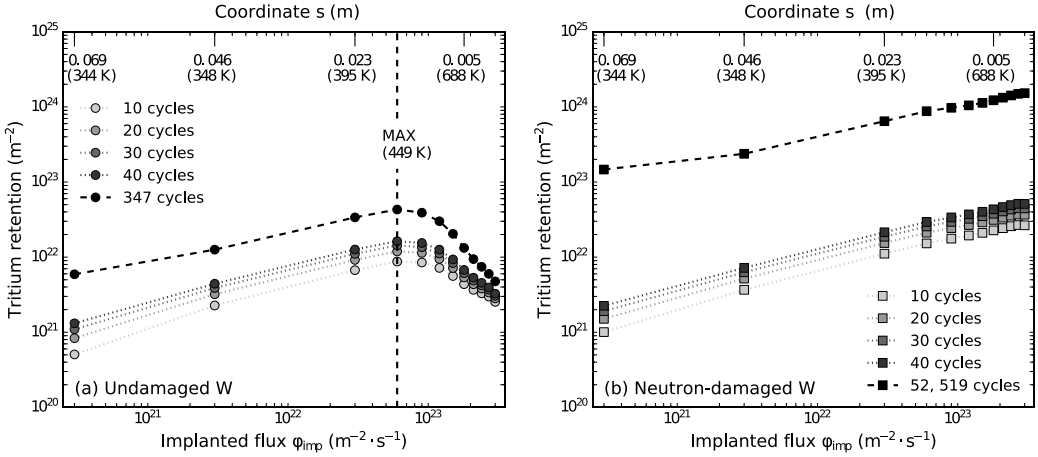

Coordinate s (m)
0.069 (344 K)
0.046 (348 K)
0.023 (395 K)
0.005 (688 K)
MAX (449 K)
10 cycles
20 cycles
30 cycles
40 cycles
347 cycles
(a) Undamaged W
Tritium retention (m^{-2})
Implanted flux φ_{imp} (m$^{-2}\cdot$s^{-1})
Coordinate s (m)
0.069 (344 K)
0.046 (348 K)
0.023 (395 K)
0.005 (688 K)
10 cycles
20 cycles
30 cycles
40 cycles
52, 519 cycles
(b) Neutron-damaged W
Tritium retention (m^{-2})
Implanted flux φ_{imp} (m$^{-2}\cdot$s^{-1})

Figure 3

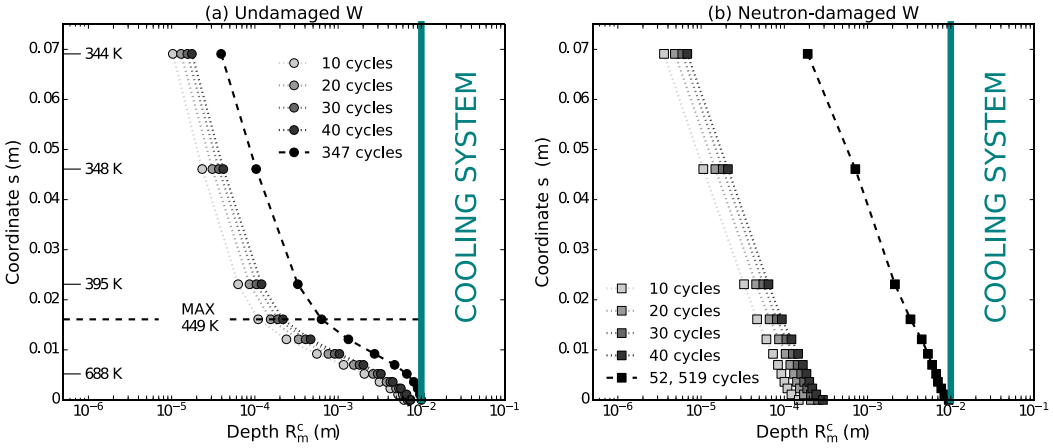
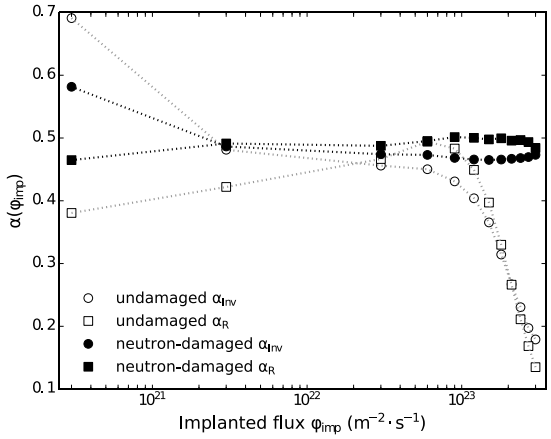

(a) Undamaged W
(b) Neutron-damaged W
Coordinate s (m)
Depth R_m^c (m)
344 K
348 K
395 K
688 K
MAX
449 K
10 cycles
20 cycles
30 cycles
40 cycles
347 cycles
52, 519 cycles
COOLING SYSTEM

Figure 4


0.7
0.6
0.5
0.4
0.3
0.2
0.1
α(φimp)
undamaged αInv
undamaged αR
neutron-damaged αInv
neutron-damaged αR
10^21
10^22
10^23
Implanted flux φimp (m^-2 · s^-1)