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# Tritium Decay Helium-3 Effects in Tungsten

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ABSTRACT:

Tritium (T) implanted by plasmas diffuses into bulk material, especially rapidly at elevated temperatures, and becomes trapped in neutron induced material defects in the tungsten (W) that act as trapping sites for the tritium. The trapped tritium atoms will decay to produce helium-3 ($^3$He) atoms at a half-life of 12.3 years. $^3$He has a large cross section for absorbing thermal neutrons, which after absorbing a neutron produces hydrogen (H) and tritium ions with a combined kinetic energy of 0.76 MeV through the $^3$He(n,H)T nuclear reaction. The purposed of this paper is to quantify the $^3$He produced in tungsten by tritium decay compared to the neutron-induced helium-4 ($^4$He) produced in tungsten. This is important given the fact that helium in materials not only creates microstructural damage in the bulk of the material but alters surface morphology of the material effecting plasma-surface interaction process (e.g. material evolution, erosion and tritium behavior) of plasma-facing component materials. Effects of tritium decay $^3$He in tungsten are investigated here with a simple model that predicts quantity of $^3$He produced in a fusion DEMO FW based on a neutron energy spectrum found in literature. This study reveals that: (1) Helium-3 concentration was equilibrated to ~ 6 % of initial/trapped tritium concentration, (2) Tritium concentration remained approximately constant (94 % of initial tritium concentration), and (3) Displacement damage from $^3$He(n,H)T nuclear reaction became > 1 dpa/year in DEMO FW.



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## 1. Introduction

A critical challenge for long-term operation of ITER and beyond to a Demonstration reactor (DEMO), or future fusion power reactor, requires the development of plasma-facing components (PFCs) that demonstrate erosion resistance to steady-state/transient heat fluxes and intense neutron/neutral/ion particle fluxes under the extreme fusion nuclear environment, while at the same time minimizing in-vessel tritium inventories and permeation fluxes into the PFC's coolant. Tritium (T) is known to diffuse rapidly into bulk material at elevated temperatures and become trapped in neutron radiation-induced defects that act as tritium trapping sites, with a density of up to 1 at. % T/W [1-3] in tungsten (W). Tritium decays to produce helium-3 ($^3$He) that can play a significant role in microstructural evolution (e.g. helium embrittlement) when compare to the role neutron-induced helium-4 ($^4$He) produces (e.g. $^4$He/dpa ratio of 0.4-0.7 appm [4]) in tungsten. The microstructural evolution from tritium and irradiation effects (e.g. displacement damage, transmutation and gamma radiation) alters the plasma material interaction (PMI) processes of near surface material evolution, erosion, dust production and tritium behavior in the PFCs. As a consequence, advances in tritium and nuclear sciences are crucial for developing PFCs that can withstand the environment produced by long pulse burning plasma operation.

$^4$He induced nanoscopic surface morphology (e.g. bubble and tungsten-fuzz) is known to reduce T diffusion and trapping significantly [5,6], but its effects in neutron-damaged tungsten are still not well known. Radiation damages from 14 MeV D-T neutrons create trapping sites for T of up to 1 at.% T/W (10,000 appm) in tungsten for ITER D-T and DEMO. Concern arises for T decaying into $^3$He when the T accumulates to reach high atom densities in a material through radiation-induced trapping sites, interlayer between two different materials or as a result of a high material solubility for hydrogen. $^3$He has a large cross section for absorbing thermal neutrons, which after absorbing a neutron produces hydrogen (H) and T ions with a combined kinetic energy of 0.76 MeV through the $^3$He(n,H)T nuclear reaction [7]. Accumulations of H, T and $^3$He can lead to hydrogen and helium

embrittlement in tungsten that should be accounted for in addition to that produced by neutron-induced H and $^{4}$He in tungsten [4]. In addition, displacement damage created by the energy imparted to the $^{3}$He(n,H)T reaction products will also create additional trapping sites for hydrogen isotopes. It is important to note that the $^{4}$He induced nanoscopic surface morphology (e.g. bubble and tungsten-fuzz) is directly caused by the D-T reaction produced $^{4}$He, not by the transmutation produced $^{4}$He and T decay produced $^{3}$He. In the meantime, the transmutation produced $^{4}$He and T decay produced $^{3}$He are known to degrade thermal conductivity of materials, and are expected to have indirect effects on nanoscopic surface morphology.

To the best of our knowledge, an assessment of H, T and $^{3}$He accumulations and $^{3}$He(n,H)T in induced displacement damage in fusion material is not available in literature. In this paper, a simple model was developed that treats $^{3}$He loss from the $^{3}$He(n,H)T nuclear reaction like a decay process by deriving an effective "decay" constant through integrating the neutron absorption rate over a fixed neutron energy spectrum, and then applying this "decay" constant in sets of equations that are analytically solve to determine the T and $^{3}$He concentrations in tungsten. It is important to note that the purpose of this study is to perform a scoping study of effects of T decay $^{3}$He (the decay product of T) in tungsten in a simple conservative fashion, that will hopefully motivate neutronics' experts to perform a more self-consistent calculation to accurately estimate T and $^{3}$He effects in tungsten. The main questions to be answered in this study are: 1) Will the concentration of $^{3}$He, H, and T become high enough to cause H/He embrittlement in tungsten for DEMO FW condition, and (2) Will the displacement damage created by $^{3}$He(n,H)T reaction products have a profound effect in tungsten for DEMO FW conditions? The term "T decay $^{3}$He" is used to describe the $^{3}$He created as the decay product of T in this manuscript.

## 2. Theory / Model

### 2.1 Reaction equations for T decay $^3$He and $^3$He(n,H)T nuclear reaction

T decays to a $^3$He atom, an electron ($e^-$) and electron anti-neutrorino ($\overline{\nu_e}$) at a half-life of 12.3 years, as shown in the following equation:

$$ {}_1^3T \rightarrow {}_2^3He + \ e^- + \overline{\nu_e} + Q_1 \qquad \text{(Eq.1)} $$

The decay constant and Q-value for this process are $\lambda_1 = 1.79 \times 10^{-9}$ [s$^{-1}$] and $Q_1 = 18.589$, keV respectively [8].

Under a neutron-irradiation environment, such as those found in a fission or fusion reactor, $^3$He will undergo a neutron ($n$) capture reaction, $^3$He(n,H)T, which produces H and T as shown in Equation 2. The Q-value for the $^3$He(n,H)T reaction is $Q_2 = 0.764$ MeV [7].

$$ n + {}_2^3He \rightarrow {}_1^1H + {}_1^3T + Q_2 \qquad \text{(Eq.2)} $$

Product energies ($E_H$ and $E_T$) of this reaction (H and T ions) were easily calculated by energy and momentum conservation equations Equations 3 and 4.

$$ E_H = \frac{m_H}{m_H + m_T} Q_2 \qquad \text{(Eq. 3)} $$

$$ E_T = \frac{m_T}{m_H + m_T} Q_2 \qquad \text{(Eq. 4)} $$

, where $m_H$ and $m_T$ are the masses of H and T, respectively. The calculated atom energies from these equations are $E_H = 0.573$ MeVfor H and $E_T = 0.191$ MeV for T. These high-energy ions collide with lattice tungsten atoms, coming to rest within a few micrometers, and create lattice damage of tungsten in doing so.

### 2.2 Neutron energy spectrum in fusion reactor and cross section for $^3$He(n,H)T nuclear reaction

To perform a simple estimate of $^3$He(n,H)T reaction, a model , which treats $^3$He(n,H)T like a decay under a fixed neutron energy spectrum, was developed to investigate the effects of T decay $^3$He and the $^3$He(n,H)T nuclear reaction in tungsten. $^3$He has a large cross section for $^3$He(n,H)T nuclear reaction, and its reaction produces H, T and 0.764 MeV energy. The neutron capture cross section is extremely large (> 1000 barns) at thermal neutron energy and exponentially decreases as neutron energy increases. It is important to note that this neutron capture cross section of $^3$He is about an order of magnitude larger than that of $^6$Li, and $^3$He is widely used as a carrier gas for neutron detectors. High concentration of $^3$He in PFCs may also influence neutron energy spectrum and tritium breeding ratio in fusion blanket system. Neutron energy spectra of *Gilbert and Sublet* [9,10], and a cross section of *Knoll* [7] were used in this study. Figure 1 shows the neutron spectra at first wall (FW) for ITER DT, DEMO and pressurized water fission reactor (PWR) along with the cross section for $^3$He(n,H)T nuclear reaction used in this study [7,9,10]. A PWR neutron energy spectrum was included in this study to evaluate whether or not fission reactors (e.g. High Flux Isotope Reactor at Oak Ridge National Laboratory and Advanced Test Reactor at Idaho National Laboratory) can be used to study the effects of T decay $^3$He and $^3$He(n,H)T nuclear reaction in tungsten without the added expensive of thermal neutron shielding.

Under a fixed (non-time-dependent) neutron energy spectrum, $\Phi(E)$, and a fixed cross section, $\sigma(E)$, the integral of product of neutron flux and cross section over neutron energy spectrum gives a reaction coefficient that has identical units [$s^{-1}$] as a decay constant. Treating this neutron capture reaction as a decay process can be approximated by the decay constant determined as shown in Equation 5.

$$\lambda_2 = \int \Phi(E)\sigma(E)dE = \sum_i \Phi_i(E)\sigma_i(E) \qquad \text{(Eq. 5)}$$

Table 1 gives neutron capture reaction coefficients calculated with fixed neutron energy spectra for three different reactors. The calculated neutron capture reaction coefficients ($\lambda_2[s^{-1}]$) are $1.60x10^{-8}$, $2.63x10^{-8}$ , and $7.70x10^{-8}$ for ITER-DT, DEMO FW, and PWR neutron energy spectra, respectively. Despite remarkable difference in neutron energy spectra between fusion (DEMO FW) and fission (PWR) reactors, the difference in neutron capture reaction coefficients was only a factor of 3. The results indicated that fission reactors are suitable to investigate effects of T decay $^3$He and $^3$He(n,H)T in tungsten, and it is important to note the experimental result from fission (PWR) reactors can overestimate effects of T decay $^3$He and $^3$He(n,H)T for fusion (DEMO FW) environment by a factor of 3.

### 2.3 Decay equations for T decay $^3$He and $^3$He(n,H)T nuclear reaction

Equations 6-8 are the zero dimensional (0-D) conservation equations of H, T and $^3$He, the used in the simple model of this study.

$$\frac{dN_T}{dt} = -\lambda_1 N_T + \lambda_2 N_{3He} \quad \text{(Eq.6)}$$

$$\frac{dN_{3He}}{dt} = \lambda_1 N_T - \lambda_2 N_{3He} \quad \text{(Eq.7)}$$

$$\frac{dN_H}{dt} = \lambda_2 N_{3He} \quad \text{(Eq.8)}$$

, where $N_H$, $N_T$ and $N_{3He}$ are atomic concentrations [appm] of H, T and $^3$He in tungsten, respectively.

The above equations can be analytically solved in time ($t$) from $t$=0 to $t$=60 yrs (typical fission reactor lifetime) as shown in Equations 9-11.

$$N_T = \left(\frac{\lambda_2}{\lambda_1+\lambda_2}\right) N_{T,0} + \left(\frac{\lambda_1}{\lambda_1+\lambda_2}\right) N_{T,0} \exp[-(\lambda_1 + \lambda_2)t] \quad \text{(Eq.9)}$$

$$N_{3He} = \left(\frac{\lambda_1}{\lambda_1+\lambda_2}\right) N_{T,0}(1 - \exp[-(\lambda_1 + \lambda_2)t]) \quad \text{(Eq.10)}$$

$$N_H = \left(\frac{\lambda_1\lambda_2}{\lambda_1+\lambda_2}\right) N_{T,0}\left[t + \frac{1}{\lambda_1+\lambda_2}(\exp[-(\lambda_1+\lambda_2)t] - 1)\right] \quad \text{(Eq.11)}$$

### 2.4 Initial concentrations of H, T and $^3$He

In order to avoid complex plasma-material interactions and hydrogen isotope transport (e.g. implantation, trapping, detrapping, diffusion, recombination), an initial concentration ($N_T(t=0)$) of T was introduced at the beginning (*t=0*). T can be trapped in neutron radiation-induced defects with a density of up to 1 at. % T/W (10000 appm) [1-3]. The following simple, and possibly worst case, scenarios were assumed in this study: 1) Trap sites were already saturated with T at three different concentrations ($N_T(t=0) = 100, 1000, 10000\ appm$), 2) No T removal estimates were performed, 3) No H and T diffusion were considered, and 4) all the energy released by $^3$He(n,H)T nuclear reaction was deposited in lattice atoms of tungsten to create displacement damage with $E_{dis} = 90$ eV.

### 2.5 Displacement damage from $^3$He(n,H)T nuclear reaction

The $^3$He(n,H)T nuclear reaction produces H and T, and the Q-value for the $^3$He(n,H)T reaction is $Q_2 = 0.764$MeVwith $E_H = 0.573$MeVand $E_T = 0.191$MeV. As a conservative assessment, all the energy released was deposited in tungsten as shown in Equation 12.

$$dpa = \frac{Q_2}{E_{dis}}\frac{N_H}{N_W} \quad \text{(Eq. 12)}$$

where dpa, $N_W$, and $E_{dis}$ are the displacement per atom [dpa], atomic density [appm] of tungsten, and displacement energy on tungsten. The displacement energy of $E_{dis} = 90$ eV was used in this study. It is important to note that this assumption of 100 percent of the energy deposition in tungsten was used as a simple and conservative estimate.

## 3. Results and discussion

First, only the T decay $^3$He was considered in the simple model, and the decay equations (Equations 9-11) were analytically solved in time ($t$) from $t$=0 to $t$=60 yrs with $\lambda_2 = 0$ [s$^{-1}$]. Figure 2 showed (a) T, $^3$He and $^4$He concentrations, (b) H concentration and (c) Displacement damage without $^3$He(n,H)T effect ($\lambda_2 = 0$ [s$^{-1}$]). Figure 2(a) showed that concentrations of T and $^3$He became identical at $t$=12.3 yrs, and majority of T were replaced by $^3$He after $t$=20 yrs. It is interesting to note that T decay produced more than 500 appm of $^3$He in a year ($t$=1 yr) for $N_T(t = 0) = 10000$ appm case compared with only 1 appm production of $^4$He by (n,α) reactions in a year. Since the $^3$He(n,H)T nuclear reaction was not considered, Figure 2 (b) and 2(c) showed no H concentration and no additional displacement damage in tungsten.

Second, both the T decay $^3$He and $^3$He(n,H)T nuclear reaction were considered in the simple model, and the decay equations (Equations 9-11) were analytically solved in time ($t$) from $t$=0 to $t$=60 yrs with $\lambda_2 = 2.8 \times 10^{-8}$ [$s^{-1}$] for DEMO FW neutron energy spectrum. Figure 3 showed (a) T, $^3$He and $^4$He concentrations, (b) H concentration and (c) Displacement damage with $^3$He(n,H)T effect ($\lambda_2 = 2.8 \times 10^{-8}$[s$^{-1}$]. Figure 3(a) showed that concentrations of T and $^3$He became equilibrium after approximately $t$=3 yrs. $^3$He concentration was equilibrated to ~ 6 % of initial/trapped T concentration, and T concentration stayed approximately constant (94 % of initial T concentration) in DEMO FW. It is important to note that the $^3$He(n,H)T nuclear reaction reduced the $^3$He concentration to approximately 600 appm after approximately $t$=3 yrs for $N_T(t = 0) = 10000$ appm case. Figure 3(b) showed that H concentration reached approximately 1000 appm at $t$=3 yrs in tungsten and kept increasing in time. Figure 3(b) showed that additional displacement damage by the $^3$He(n,H)T nuclear reaction became comparable to that by neutron in tungsten after approximately $t$=5 yrs. Displacement damage from the $^3$He(n,H)T reaction introduces additional concern (> 1 dpa/year) in DEMO FW.

The above results showed unique interplay between T, $^{3}$He and neutron in fusion and fission nuclear environment. In a non-nuclear environment, the $^{3}$He(n,H)T nuclear reaction does not need to be considered. T decay $^{3}$He effects and aging effects in material for a non-nuclear environment have been investigated that show the effects of $^{3}$He embrittlement in material under high T charging conditions [11]. Fortunately for fusion and fission application, the $^{3}$He(n,H)T nuclear reaction reduces $^{3}$He concentration significantly (e.g. to the percent level of initial/trapped T concentration), but it introduces additional H, T and displacement damage. Displacement damage from the $^{3}$He(n,H)T nuclear reaction introduces additional concern in fusion and fission nuclear environments, and this additional displacement damage is associated with the local T concentration. *Causey et.al.* discussed tritium behavior in fusion and fission reactor conditions, and poor tritium permeation barrier performances in fusion and fission reactor conditions [12]. The T decay $^{3}$He and $^{3}$He(n,H)T nuclear reaction can play a crucial role in understanding tritium behavior and tritium permeation barrier performances in fusion and fission reactor conditions since T accumulates in interlayer between two different materials and/or in tritium permeation barrier materials. Radiation damage from $^{3}$He(n,H)T nuclear reaction in the T accumulated interlayer can degrade tritium permeation barrier performance.

## 4. Conclusions

Trapped T atoms decay into $^{3}$He atoms with a half-life of 12.3 years. $^{3}$He has a large cross section for $^{3}$He(n,H)T nuclear reaction, producing H and T ions with 0.764 MeV energy. An assessment of H, T and $^{3}$He accumulations and $^{3}$He(n,H)T induced displacement damage was performed by the simple model, which treats the $^{3}$He(n,H)T nuclear reaction as a decay process for a fixed neutron energy spectrum. Sets of conservation equations were analytically solved to investigate the T decay to $^{3}$He effects in tungsten with DEMO FW neutron energy spectrum. The results showed that (1) Concentrations of $^{3}$H and $^{3}$He became equilibrium after approximately *t=3 yrs*, (2) $^{3}$He concentration was equilibrated to ~ 6 % of initial/trapped T concentration, and T concentration stayed approximately constant (94 % of initial T concentration), (3) H concentration reached approximately 10 % of initial/trapped T concentration at *t*=3 yrs in tungsten and kept increasing in time, and (4) Additional displacement damage by the $^{3}$He(n,H)T nuclear reaction became comparable to that by neutron in tungsten after approximately *t*=5 yrs.

Displacement damage from the $^{3}$He(n,H)T reaction introduces additional concern (> 1 dpa/year) in DEMO FW. This study shows profound effects of T decay into $^{3}$He in tungsten even when the $^{3}$He(n,H)T nuclear reaction is taken into account by a simple model in a conservative manner; but given the simplicity of this model a more comprehensive neutronics calculation is required to confirm and accurately estimate the effects of T decay $^{3}$He and $^{3}$He(n,H)T nuclear reaction in tungsten in fusion nuclear environments.

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## Figures

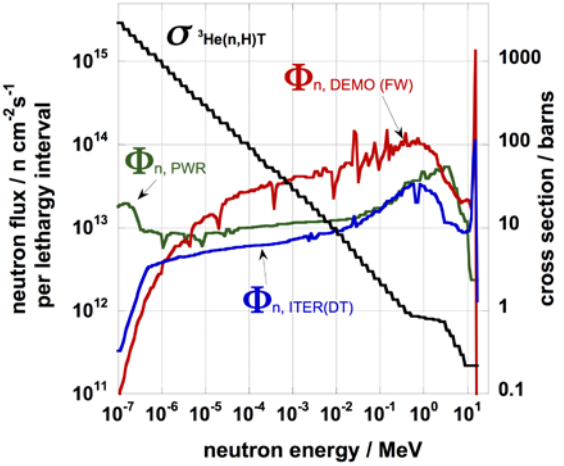


**Figure 1: Neutron spectrum [9,10] and cross section [7] used in this study**

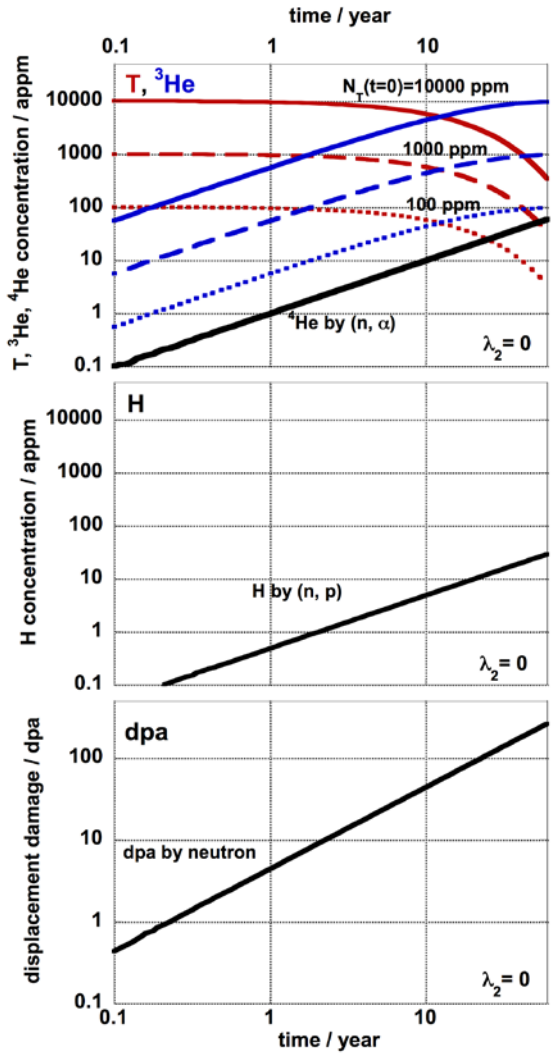


**Figure 2: (a) T, $^3$He and $^4$He concentrations, (b) H concentration and (c) displacement damage in the case of no $^3$He(n,H)T effect ($\lambda_2$= 0 [$s^{-1}$]).**

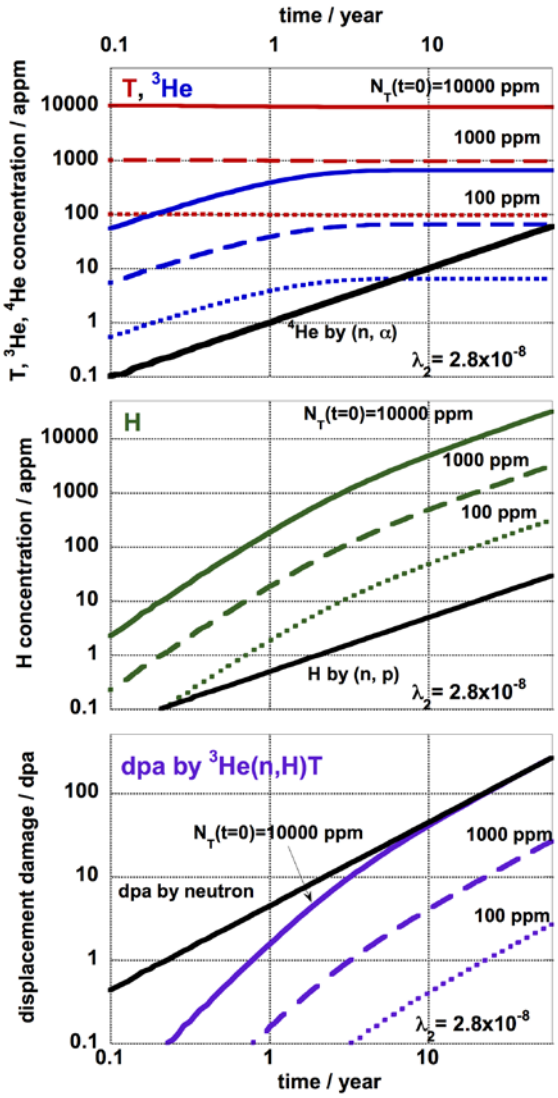


**Figure 3: (a) T, $^3$He and $^4$He concentrations, (b) H concentration and (c) displacement damage in the case of $^3$He(n,H)T effect ($\lambda_2$= 2.63x10$^{-8}$ [s$^{-1}$]) in DEMO FW**

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