# References

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# Purpose

This document summarizes recommended permeation properties for tritium in materials used for the ITER Internal Components (Blanket and Divertor). The data are either extrapolated from experimental studies or extracted from publications.

In order to model the behavior of particle penetration in materials, this document provides an evaluation of the following variables:

* + - Diffusivity
    - Solubility
    - Recombination coefficient
    - Trap Parameters

The first three parameters will be defined as Arrhenius functions of the temperature, and therefore include two unknowns:

Where T is the temperature and k is Boltzmann’s constant (~ eV/K). Ec (in eV) and C0 are the unknowns. Ec represents the activation energy for the considered event while the pre-exponential factor C0 defines the frequency of the event.

Regarding the trap parameters, both the trap concentration and trapping energy need to be determined.

# Equations used

The diffusion equations hereafter were extracted from the TMAP7 documentation [1]. These equations allow the calculations of mobile and trapped concentrations. The following equation calculates the movement of mobile atoms through structures:

D is the diffusivity (at.m²/s). It follows an Arrhenius dependence on the temperature T. is the mobile concentration of atomic species “s” (at/m³), meaning the concentration of particles that can currently move through the structure. On the opposite, represents the trapped concentration (at/m³), which means the concentration of particles that are currently trapped at a defect site (vacancy, impurities…) and cannot move until they are released.

The traps have an important effect on the permeation of particles in a material. However, it is important to consider that trapping can be highly impacted by the exposition to radiological environment. For example, exposition to neutron irradiation can lead to multiplication of trapping site, as explained in [2]. Ion implantation can also lead to the creation of strong trapping sites in the vicinity of the impacted surface, as shown in [3]. The effects of irradiation are dependent on the environment. In [9], an evaluation of the effect of dpa on deuterium trapping was provided for Eurofer.

The permeation parameters that we are about to present do not take into account any effect of irradiation, or implantation. It is recommended to modify the trap parameters to take these into account, when relevant. An example of this process can be found in [2], where the dpa rate was used to extrapolate the creation of trapping sites in 316L steel.

The following equation calculates the equilibrium trapped/mobile proportion for each trap:

The first part of the formula represents the trapping of particles, while the second part represents their release. is the total trap concentration in the material (traps/m³), which is one of the trapping parameters that we define here. represents the trapping energy (eV), which corresponds to the energy barrier for release from a trap, the other trapping parameter. N, λ and ϑ0 are material properties. N is the atom density (at/m³), λ the lattice parameter (m) of the material and ϑ0 the Debye frequency (s-1). The material properties that we considered in our simulations were:

Table : material properties used for our simulation

|  |  |  |  |
| --- | --- | --- | --- |
| Parameters | Debye frequency (s-1) | Atom density (at/m³) | Lattice parameter (m) |
| Stainless steel 316L |  |  |  |
| Beryllium |  |  |  |
| Copper (and copper alloys) |  |  |  |
| Tungsten |  |  |  |

To describe the boundary conditions, the following two conditions are used:

Sievert’s equation:

In this formula, is the surface concentration of atomic species “s” (at/m²). S is the solubility parameter (at/m².Pa½), defined as an Arrhenius equation. represents the partial pressure of molecules “m” in the gas surrounding the sample (Pa). The square root indicates that the gas species considered here is “s2”. This equation models the absorption of atoms s in the sample from a gaseous atmosphere.

Recombination equation:

is the flux of atomic species “s” out of the sample from the surface (at/.s). is the recombination coefficient (m4/s), written as an Arrhenius equation. This equation can be used to calculate desorption during a TDS. It models the combination of two “s” atoms in an “s2” molecule.

# Isotopic effect

The parameters that we are presenting on this study are aimed toward modelling the tritium behaviour through different materials. If your goal is to model another hydrogen isotope’s behaviour, then the parameters are biased. It is generally admitted that in order to switch from one isotope to the other, you can keep the same permeation parameters, except for diffusivity. For diffusivity, you should then follow the following law [8]:

The mass of the isotope is supposed to hinder the diffusion process. On the other hand, the solubility and recombination coefficients are considered identical same between two isotopes, as their chemical properties are identical.

In all the graph presenting extrapolations from publications in this document, we noted whether the data was originally for tritium, or another isotope. D->T means the studied isotope was originally deuterium and H->T mean it originally was hydrogen. In both cases, we applied the and factors to extrapolate into tritium extrapolations.

# Stainless Steel

We led two extrapolations of permeation parameters for Stainless steel ([2], [4]). Both extrapolations were for 316L (N) Stainless steel. We reckon that the permeation behaviours of tritium through similar stainless steel alloys (such as 304L) should be quite identical.

In both case, we used a calculation code to try and reproduce the experimental results obtained in JET and KAERI respectively. The JET experiments were modelled using TMAP, a 1D calculation code, and the KAERI’s experiments were modelled using Matlab and the pde toolbox, with the same fundamental equations as TMAP. The simulation of these experiments yielded different extrapolations.

Using those two extrapolations to create a confidence interval that could be used to evaluate the variation that may occur when considering different extrapolations, we defined a “mean” set of parameters. These mean extrapolations correspond to the mean value of the JET and KAERI extrapolation.

Table : permeation parameters for stainless steel 316L

|  |  |  |  |
| --- | --- | --- | --- |
| Parameters | KAERI experiment | Mean parameters | JET experiment |
| Diffusivity (m²/s) |  |  |  |
| Solubility (at/m³.Pa½) |  |  |  |
| Recombination (m4/s) |  |  |  |
| Trap concentration  (atom fraction) | 0.08 | 0.08 | 0.08 |
| Trap energy (eV) | 0.7 | 0.7 | 0.7 |

The trapping energy and concentration should represent a vacancy-type trap. Vacancies are expected to be created while the stainless steel is irradiated.

One may notice that these extrapolations are quite different. In order to compare them, we drew them hereafter as function of 1/T (K-1). We also drew other extrapolations we found in various publications.

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| --- |
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|  |
| Figure : comparison of the permeation coefficients for our study VS other publications for Stainless steel |

You can notice that our extrapolations are similar to the results obtained in publications, which is reassuring. In the same way, we are reassured to notice that our extrapolations are well within the range observed in publications.

In order to ascertain that this mean extrapolation could represent the behaviour of tritium in stainless steel, we modelled the KAERI and JET experiments using the mean parameters, and generally observed a difference of ~10% between the results obtained with the mean and respectively extrapolated parameters. This kind of difference is common with TDS experiment. We considered from these considerations that the mean parameter model is indeed able to simulate tritium migration in Stainless steel correctly.

# Pure copper

Regarding pure copper, we decided to use an extrapolation from a publication. The following graphs present the extrapolations we were able to gather for diffusivity, solubility and recombination:

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|  |
| Figure : comparison of the permeation coefficients from various publications for pure copper |

As one might notice, the publications are quite in agreement about the solubility and diffusivity values, but the recombination coefficient is subject to variation. It is better to consider a set of three parameters from the same publication if possible, as they should work well together. We choose to consider the Reiter et al 1993 extrapolation [10], which provided an estimation of all three parameters and does not present an important deviation with the other publication. Reiter et al extrapolations are coloured in bright red in the previous graphs.

The permeation parameters for Reiter et al correspond to those Arrhenius equations:

Table : permeation parameters for pure copper

|  |  |
| --- | --- |
| Parameters | Equations |
| Diffusivity (m²/s) |  |
| Solubility (at/m³.Pa½) |  |
| Recombination (m4/s) |  |

Trapping in pure copper is considered to be very energetically low. The intrinsic traps in copper are generally considered to have energies under 0.5eV. In [5], mono-vacancy or small vacancies cluster trapping was evaluated around 0.42eV.

We consider using the data from Wilson 1987, presented in [5]: “DIFFUSE calculations that considered a 0.5-eV trap with a concentration of 0.5 appm and a 0.93-eV trap with a concentration of 25 at parts /1E9 yielded an approximate fit to the TDS data.” in order to choose the trap concentration. Wilson 1987 evaluated hydrogen isotope retention and release from copper under ion bombardment. Deuterium was implanted at ambient temperature in copper. We consider that the traps that were modelled in order the simulate their results originate from two sources: The 0.5eV trap is representative of the vacancy cluster that were intrinsically present in copper, while the 0.9eV traps originates from implantation and bubble generation in the metal.

From this assumption, we consider that we can model copper with the parameters from Table 3, and 0.5appm of traps at 0.42eV.

# CuCrZr

For the CuCrZr alloy used in ITER, here are the parameters that we were able to extract from publications:

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|  |
| Figure :comparison of the permeation coefficients from various publications for CuCrZr |

The extrapolation numbered far less than for the other materials. Copper is not very documented, and even more so for this specific alloy. Moreover, regarding the particular alloy used in ITER, only the ELBRODUR G and ELBRODUR HF alloy meet the requirements from [6] (see 10.1). This means that we only have two reliable extrapolations for the diffusion and solubility parameters. Moreover, only one recombination extrapolation was found for the CuCrZr alloy (and not with IT grade alloy). If we compare this recombination coefficient with the recombination for pure copper, we observe:

|  |
| --- |
| Figure : comparison of the permeation coefficients for pure copper VS CuCrZr |

We can observe that the recombination coefficient is in the same order of magnitude than the extrapolations for copper in the extrapolation’s range of temperature. The slope of the curve is steeper, which could be explained with the presence of copper. In a study presented in [5], Cr in copper matrix was found to affect the surface-oxide effect. Of course, it is difficult to create a definitive opinion with only one extrapolation.

The equations corresponding to the ELBRODUR G and ELBRODUR HF are the following:

Table : permeation parameters for the ELBRODUR G and ELBRODUR HF alloys

|  |  |  |
| --- | --- | --- |
| Parameters | ELBRODUR G | ELBRODUR HF |
| Diffusivity (m²/s) |  |  |
| Solubility (at/m³.Pa½) |  |  |

In order to evaluate a confidence interval for further calculation, we can complete these data with Anderl et al’s recombination coefficient as follow:

Table : permeation parameters for CuCrZr

|  |  |  |
| --- | --- | --- |
| Parameters | ELBRODUR G | ELBRODUR HF |
| Diffusivity (m²/s) |  |  |
| Solubility (at/m³.Pa½) |  |  |
| Recombination (m4/s) |  |  |

Although this is the case, Serra et al’s ELBRODUR HF values can be considered more trusted, as several publications referred to their work.

Regarding trapping in the copper alloy, the intrinsic traps of copper are still present, but we have to take into account the effect of the zirconium. A study presented in [5] estimated a trap binding energy of 0.83eV for Zr in copper matrix. Cr apparently had no effect on bulk-trapping.

It is difficult to evaluate how the presence of zirconium will translate in terms of trap concentration. If we consider the data from [6], the zirconium concentration in the copper alloy is between 0.07 and 0.15 wt%. This represents, in terms of atom fraction, 0.0019 and 0.004 atom fraction. If we take the mean value, we get to 0.003 atom fraction.

# Beryllium

Regarding the Beryllium on the tokamak First wall, we chose some publicized extrapolation that various publications showed a good agreement on. We also used some data from JET, which experimented deuterium migration in ion-implanted beryllium, to extrapolate the traps parameters that we may use with these parameters. The permeation parameters were:

Table : permeation parameters for Be

|  |  |
| --- | --- |
| Parameters | Equations |
| Diffusivity (m²/s) | D=6.5e-9\*exp (-0.364 /(k\*T)) |
| Solubility (at/m³.Pa½) | S= 2.3e22\*exp (-0.174 / (k\*T)) |
| Recombination (m4/s) | Kr=3.4e-29\*exp (-0.28/(k\*T)) |

These parameters were already used to calculate tritium migration in co-deposited Beryllium.[2] The major difference between co-deposited and solid beryllium lays in the energy and concentration of the traps.

When we compared to extrapolations extracted from publications, the agreement was good. The comparison is presented hereafter, with our extrapolation being bright red:

|  |
| --- |
|  |
|  |
| Figure : comparison of permeation parameters from various publications, for Beryllium |

One can notice that the extrapolation for Beryllium from publications are more scattered than for the other materials.

According to previous analysis led in [8]the traps parameters used to model Beryllium co-deposits were:

Table : trap parameters modelling Be co-deposits

|  |  |  |
| --- | --- | --- |
| Trap n° | Trap parameters | Value |
| 1st trap | **Concentration  (atom fraction)** | 0.028 |
| **Energy (eV)** | 0.778 |
| 2nd trap | **Concentration (atom fraction)** | 0.009 |
| **Energy (eV)** | 0.99 |
| 3rd trap | **Concentration  (atom fraction)** | 0.011 |
| **Energy (eV)** | 1.43 |

Of course, the trapping parameters for solid Beryllium are expected to be different, especially with implantation. In order to establish the trap parameters for solid Beryllium, as well as make an estimation of the impact of implantation on Beryllium, we modelled a TDS experiment on the Beryllium limiters used in the JET reactor. The implantation parameters are different from the one expected for ITER, so our conclusion on the implantation traps are not directly applicable on ITER’s Beryllium, especially the trap concentration. The trapping energy should still be relatively comparable.

On the other hand, this simulation brought us the intrinsic trapping parameters of Beryllium. The intrinsic traps that we extrapolated from the study of JET limiter’s Beryllium had the following parameters:

Table : generic trap parameters for solid Beryllium

|  |  |  |
| --- | --- | --- |
| Trap n° | Trap parameters | Value |
| 1st trap | **Concentration  (atom fraction)** | 0.005 |
| **Energy (eV)** | 0.93 |
| 2nd trap | **Concentration  (atom fraction)** | ~3.75\*10-4 |
| **Energy (eV)** | 1.6 |

Our opinion is that the first trapping site encompasses the effect of grain joints and dislocations, while the second trapping site can be explained through the effect of vacancy in the crystallographic mesh. The vacancies in Beryllium are more energetic than the vacancies in stainless steel, generally considered around 1.4~2eV. Vacancies are expected to be created while the beryllium is irradiated.

# Tungsten

# Conclusion

# Annex

## CuCrZr alloys

Table : specification for Iter-grade alloy

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Alloy Designation | Base Alloying Elements and Impurities (wt.%) | | | |
| Cu | Cr | Zr | Impurities |
| CuCrZr -IG | base | 0.6 – 0.9 | 0.07-0.15 | Total < 0.1 |
| Including Co < 0.05 |
| O – as low as possible |

Table : elements in the alloys presented in CuCrZr chapter

|  |  |  |  |
| --- | --- | --- | --- |
| Alloy designation | Base Alloying Elements (wt.%) | | |
| Cu | Cr | Zr |
| ELBRODUR G | base | 0.65 | 0.1 |
| ELBRODUR N | base | 0.65 | 0.05 |
| ELBRODUR HF | base | 0.65 | 0.08 |
| AMPCOLOY 972 | Base | 1.1 | 0.1 |
| Anderl 1999 | Base | 1.1 | 0.1 |

## Comparison Cu VS CuCrZr