

On the modelling of tritium transport phenomena at fluid-structure interfaces

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Abstract. One of the main functions of the DEMO Breeding Blanket (BB) system is to ensure the tritium breeding inside the reactor. Tritium is a beta emitter radioactive isotope, subjected to several processes that determine its permeation across materials and its leakage towards the environment, posing potential safety issues in terms of radiological hazard. Thus, the evaluation of tritium inventories inside components and tritium losses towards the environment plays a key role in the fulfilment of the pertinent BB safety requirements. In this regard, a research activity has been carried out, in close cooperation between the University of Palermo and the Karlsruhe Institute of Technology, focussing on the development of a multiphysic model that might realistically simulate 3D tritium transport phenomena across complex fluid-structure interfaces. Models, source terms and boundary conditions assumed for the analyses are herewith reported and critically discussed, together with the main results obtained.

1. Introduction

The European Roadmap, describing the priorities of fusion energy research development towards electricity production, outlines tritium self-sufficiency and management in fusion power plants among the most critical missions [1]. In DEMO, tritium will be produced inside the Breeding Blanket (BB) system and it is subjected to several processes that determine its permeation across materials. Therefore, the prediction of tritium inventories inside the DEMO BB components and tritium losses towards the environment plays a fundamental role to fulfil the relevant safety requirements.

Tritium transport modelling through the BB system is complex and involves a large number of parameters, most of which have not been determined with an adequate level of accuracy yet. Moreover, the applicability of tritium transport models to the DEMO BB requires the physics to be sufficiently comprehensive to take into account all phenomena that can take place under a wide range of operative conditions [2]. Indeed, within the BB, the T generated into the breeder material (e.g., Li-orthosilicate/metatitanate pebble beds or eutectic alloy of PbLi enriched with ⁶Li) is transported by a T-carrier (He purge gas or the same PbLi) outside the blanket to be extracted. However, the T-carrier is in

contact with tens of thousands of square meters of structural material at high temperatures (e.g. 300-500 °C) and with thin thickness. Therefore, the BB conditions are particularly favourable for the T permeation that can pose a safety concern in terms of contamination of the coolant system and releases into the Tokamak building as well as in the environment. Of course, several mitigation measures are studied to reduce the T permeation (e.g. anti-permeation coatings in alumina) and to control the T inventories. However, the preferable approaches, to investigate complex phenomenon like the T transport, remain the computational ones (although they require strong verification and validation).

All the currently available tritium transport models (e.g. [3], [4]) refer in most of the cases to system-level or microscopic level, being the reason why a component level model (namely a model referring to an intermediate level) is necessary to improve the accuracy of the predictions in terms of tritium inventories, losses and contaminations of coolant flows [5].

Within this framework, a research campaign has been carried out at the University of Palermo, in close cooperation with the Karlsruhe Institute of Technology, in order to develop a 3D model that takes into account transport phenomena involving tritium transport across fluid-structure interfaces.

The study has been carried out following a theoretical-computational approach based on the Finite Volume Method (FVM) and adopting the commercial Computational Fluid-Dynamic (CFD) code ANSYS CFX 2019 R1 [6].

Tritium dissociative and recombinative phenomena occurring at fluid-solid interfaces are governed by a non-linear InterFace Condition (IFC) and the net tritium flux is a function of the local concentration of both diatomic and monoatomic species, which are the variables of the problem. The implementation of non-linear IFCs is not straightforward in the code and, therefore, two novel approaches have been introduced in this work (more details are reported in section 4). As a first step, 1D diffusion problems have been considered and the results of their relevant numerical simulations have been compared against the pertaining analytical solutions (section 3) to assess the predictive potential of the methodologies adopted and validate the modelling approaches (section 5).

Moreover, parametric analyses have been performed and different cases have been studied by varying the model free parameters, to investigate their impact on permeation rates and concentration profiles.

2. Tritium permeation phenomena

Tritium permeation at fluid-solid interfaces is mainly governed by different surface and bulk processes as schematized in **Figure 1**. In the following, the most significant phenomena will be described

Surface processes are referred to all the processes occurring on and through the surface of a solid. This category includes adsorption, dissociation, recombination and desorption summarized by the following chemical equilibrium expression:



In general, surface processes occur in a thick layer, just below the gas-solid interface, which is referred to as a "surface layer". This layer exists due to several causes, such as natural oxide formation, deposition of a coating layer or modification by implantation. It is typically some hundreds Å thick and is not uniform, since the oxygen content of the oxide layer and the density of implanted products changes with depth [7].

Bulk processes comprehend atomic dissolution and interstitial diffusion caused by concentration gradients. It is found experimentally that, from a macroscopic point of view, the flux J [$\text{mol m}^{-2} \text{s}^{-1}$] of dissolved tritium atoms inside a solid follows Fick's Law [7]. Two other bulk processes are thermomigration and trapping, the latter occurring when tritium bonds to microstructural features within metals. Both are thermally-driven processes that in this work have been neglected since isothermal conditions have been considered.

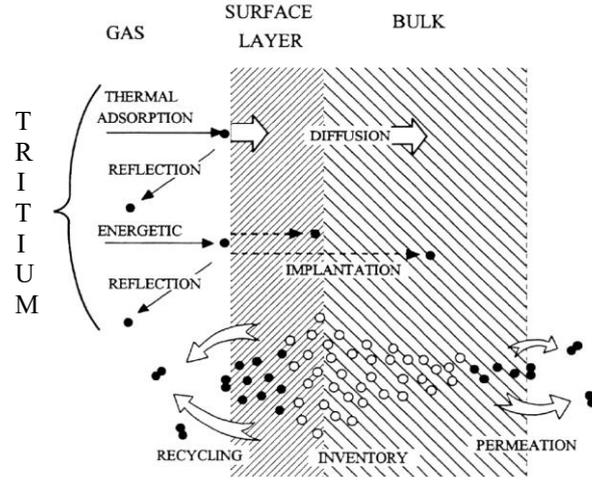


Figure 1. Tritium transport processes across solid boundaries [7].

Permeation can be either limited by surface or diffusion phenomena. The occurring regime is determined by a permeation number W that takes into account tritium partial pressure, surface properties, bulk thickness and permeabilities [8]. Diffusion-limited regime occurs when high partial pressure differences are established across the solid region and, in particular, tritium migration through the metal membrane is limited mainly by diffusion in the metal lattice while the surface processes (such as adsorption, desorption) are significantly faster. On the other hand, when low tritium partial pressure differences occur, a surface-limited regime is established, and the diffusion through the membrane occurs fast enough so that any concentration gradient is cancelled by diffusion [9]. In this case, permeation is limited by the rate at which atoms can recombine back into molecules. Except for extremely high temperatures, this recombination is necessary for tritium to be released from a material.

Tritium dynamics in solid and fluid media is governed by diffusion and/or convective transport phenomena. The set of equations to be solved is strongly simplified by the fact that tritium can be considered a passive scalar since it has no significant influence on the velocity, density and temperature fields. The general transport equation in a generic medium can be derived from the mass continuity equation for the species i in a medium j and it is given by (2):

$$\frac{\partial C_i(\mathbf{x}, t)}{\partial t} + \nabla \cdot \mathbf{u}(\mathbf{x}, t) C_i(\mathbf{x}, t) - \nabla \cdot (D_{ji} \nabla C_i(\mathbf{x}, t)) = S_i(\mathbf{x}, t) \quad (2)$$

where \mathbf{x} and t denote the space and time coordinates, C_i [mol m^{-3}] is the i -th species concentration field, \mathbf{u} is the vector velocity field, D_{ji} is the diffusivity 2nd order tensor of the i -th species in the j -th medium and S_i is the i -th species source term. Obviously, for the case of a solid medium, the governing equation is further simplified for the absence of the convective term.

The permeation regime mainly depends on the tritium partial pressures in the fluid mean which is strongly related to the examined problem. To avoid any a-priori choice of the permeation regime a more general model derived from classical kinetic theory has been adopted here to calculate the net flux J_i of the atomic species i through a metal surface [10].

For the aim of this work, only the molecular species T_2 has been considered, neglecting for the sake of simplicity the presence of H_2 and HT . Tritium molecules are adsorbed at the interface and dissociated into two T atoms. Then, atoms may desorb from the surface and recombine back into gas as T_2 molecules. The dissociation and recombination fluxes at interfaces (from the fluid side) can be expressed as:

$$J_{T_2,d} = K_d \cdot p_{T_2} \quad (3)$$

$$J_{T_2,r} = K_r \cdot C_T^2 \quad (4)$$

where K_r and K_d are phenomenological constants called recombination and dissociation constant respectively, p_{T_2} is the T_2 partial pressure and C_T is the concentration of the T species.

If the system is not in an equilibrium state, dissociation and recombination fluxes at fluid-structure interfaces are not counterbalanced, and net transport of tritium occurs from one side to the other. The net flux is thus:

$$J_{T_2} = K_d \cdot p_{T_2} - K_r \cdot c_T^2 \quad (5)$$

Considering a gas with a temperature T_g , Dalton's law (6) expresses the relationship between the concentration of T_2 (C_{T_2}) and its partial pressure:

$$C_{T_2} = \frac{1}{RT_g} p_{T_2} = K_g p_{T_2} \quad (6)$$

This relation allows fluxes to be expressed as a direct function of the concentrations as follows:

$$J_{T_2} = K \cdot C_{T_2} - K_r \cdot C_T^2 \quad (7)$$

where K is the ratio of K_d to K_g .

Since the concentration in solid and gas is referred to different species, the continuity of the fluxes at the interface cannot be applied, but looking at the reaction (1), the stoichiometry of the dissociation reaction defines a relationship between the species fluxes, that can be expressed as:

$$J_T = 2 \cdot J_{T_2} \quad (8)$$

Summarizing the conditions at the interface, under the hypothesis of homogeneous and isotropic media, the Cauchy IFCs between gas and solid domains are given below.

$$\begin{cases} -D_{s,T} \nabla C_T \cdot \hat{n}|_{g \rightarrow s} = 2 \cdot (K \cdot C_{T_2} - K_r \cdot C_T^2) \\ (-D_{g,T_2} \nabla C_{T_2} + \vec{u}_g \cdot C_{T_2}) \cdot \hat{n}|_{s \rightarrow g} = K \cdot C_{T_2} - K_r \cdot C_T^2 \end{cases} \quad (9)$$

From the IFCs it is possible to notice how there is neither continuity in the concentration nor in the flux, and furthermore that the fluxes depend non-linearly on the concentrations of the two species. Therefore, these equations significantly differ from those adopted in standard IFCs and their implementation into commercial computational fluid-dynamic codes results quite complex. To this purpose, two novel approaches have been considered and validated against analytical solutions to open the way to the assessment of tritium permeation properly considering 3D transport phenomena across fluid-solid interfaces.

3. Analytical model definition

In mathematical form, the transient mass conservation law (2), resolving the space-time distribution concentration C_i of the i -th species, can be easily solved in one-dimensional geometries, with a reference frame system defined by the space coordinate x (the scalar distribution is supposed to depend only on the x coordinate for the plane geometry and on the radial coordinate r for the cylindrical and the spherical geometries). Note that in the following dimensionless equations the generalized coordinate \tilde{X} has been normalized to the total length of the domain L and, analogously, the problem variables, i.e. concentrations c_i , have been normalized to a reference concentration C_0 .

The domain has been split into two parts, a fluid domain from $\tilde{X}=0$ to $\tilde{X}=\Xi$ and a solid domain, from $\tilde{X}=\Xi$ to $\tilde{X}=1$, connected by their common interface.

To simplify the resolution, the steady-state condition has been considered. Under the hypothesis of homogeneous and isotropic media, incompressible fluid at rest and a uniform volumetric source term inside the fluid domain Σ , equation (2) is particularized into two ordinary differential equations (ODEs), i.e. the diffusion equation for fluid domain and the diffusion equation for the solid domain.

The set of BCs assigned to analytically solve the Cauchy's problem consists of a Neumann BC, imposed at the fluid domain (at $\tilde{X} = 0$) due to symmetry constraint, a Dirichlet BC in the solid domain (at $\tilde{X} = 1$), and of the two non-linear IFCs at Ξ . The resulting set of ODEs together with their BCs is reported in equations (10).

$$\text{a) } \left\{ \begin{array}{l} \frac{1}{\tilde{X}^m} \frac{d}{d\tilde{X}} \left(\tilde{X}^m \frac{dc_{T_2}}{d\tilde{X}} \right) + \Sigma = 0 \\ \left. \frac{dc_{T_2}}{d\tilde{X}} \right|_{\tilde{X}=0} = 0 \\ \left. \frac{dc_{T_2}}{d\tilde{X}} \right|_{\tilde{X}=\Xi} = -(\Lambda c_{T_2} - \Gamma c_T^2) \end{array} \right. \quad \text{b) } \left\{ \begin{array}{l} \frac{1}{\tilde{X}^m} \frac{d}{d\tilde{X}} \left(\tilde{X}^m \frac{dc_T}{d\tilde{X}} \right) = 0 \\ c_T(\tilde{X}=1) = \frac{C_X}{C_0} \\ \left. \frac{dc_T}{d\tilde{X}} \right|_{\tilde{X}=\Xi} = -2\delta \cdot (\Lambda c_{T_2} - \Gamma c_T^2) \end{array} \right. \quad (10)$$

where $m = 0, 1, 2$ for Cartesian, cylindrical and spherical coordinate systems, respectively.

As regards the sensitivity analyses, an additional set of BCs has been considered for the Cartesian geometry (see equations (13)), maintaining the fluid left boundary at a fixed non-zero concentration (at $\tilde{X} = 0$), without any source term inside the fluid domain. The set of BCs adopted in equations (12) entails that the tritium flux across the interface is uniquely determined by the imposed volumetric source term. Enforcing Dirichlet-Dirichlet BCs at the ends of the domain, on the contrary, the tritium flux becomes a result of the problem and can be used to assess the quality of the different modelling strategies.

$$\text{a) } \left\{ \begin{array}{l} \frac{d^2 c_{T_2}}{d\tilde{x}^2} = 0 \\ c_{T_2}(\tilde{x}=0) = 1 \\ \left. \frac{dc_{T_2}}{d\tilde{x}} \right|_{\tilde{x}=\xi} = -(\Lambda c_{T_2} - \Gamma c_T^2) \end{array} \right. \quad \text{b) } \left\{ \begin{array}{l} \frac{d^2 c_T}{d\tilde{x}^2} = 0 \\ c_T(\tilde{x}=1) = 0 \\ \left. \frac{dc_T}{d\tilde{x}} \right|_{\tilde{x}=\xi} = -2\delta \cdot (\Lambda c_{T_2} - \Gamma c_T^2) \end{array} \right. \quad (11)$$

Dimensionless coefficients that figure in the two sets of equations are given in **Table 1**.

Table 1. Coefficients of the dimensionless form equation.

Dimensionless coefficients				
Γ	Λ	δ	Ξ	Σ
$\frac{K_r L}{D_{g,T_2}} C_0$	$\frac{KL}{D_{g,T_2}}$	$\frac{D_{g,T_2}}{D_{s,T}}$	$\frac{X_1}{L}$	$\frac{S_{T_2} L^2}{D_{g,T_2} C_0}$

Directly integrating the ODEs with constant coefficients - and homogeneous for the solid domain- and applying the related BCs, the concentration distribution profiles c_{T_2} and c_T have been obtained.

4. Numerical model definition

The aforementioned model has been implemented using the commercial code ANSYS CFX which allows solving numerically the tritium transport equation using the FVM approach.

Equation ma

In ANSYS CFX, two adjacent domains can be connected only if the flux continuity condition is imposed. If this condition is not met (8), the two domains are treated as independent, and field variables belonging to one domain cannot be retrieved from the other. Consequently, the imposition of an IC defined as a function of local concentrations on both sides of the interface is not foreseen by the code.

To overcome this issue, two alternative approaches have been conceived and they have been reported in the following paragraph. The first approach investigated consists of the definition of a thin layer - which, hereafter, is referred to as "dummy layer"- derived from the solid volume, between the fluid and

the solid domain. Atomic concentration C_T and molecular concentration C_{T_2} have been defined separately in solid and gas domains as specific passive scalars while both concentrations have been defined inside the dummy layer.

The dummy layer thickness should be of the same order of magnitude as the surface layer (discussed in §2) but, for numerical reasons, the domain cannot be simulated with such a small layer. Consequently, it has been chosen significantly thinner than the characteristic size of the domain. The effect of layer thickness assumption has been investigated separately by running a set of dedicated parametric analyses.

Inside this layer, the surface phenomena occurring at fluid-structure interfaces are simulated as bulk phenomena by introducing properly defined effective source terms in the transport equations of the two scalars C_T and C_{T_2} , reported as follows:

$$S_T = \frac{2 \cdot (K \cdot C_{T_2} - K_r \cdot C_T^2)}{\delta} \cdot \rho_s \quad (12)$$

$$S_{T_2} = -\frac{(K \cdot C_{T_2} - K_r \cdot C_T^2)}{\delta} \cdot \rho_g \quad (13)$$

where δ is the dummy layer thickness, ρ_s and ρ_g are the solid and the gas density, respectively.

This approach holds because, integrating the above-defined source terms over the dummy layer thickness, the results are equal to the fluxes exchanged at the interface, under the assumption that the concentration of the two species is uniform along the dummy layer width. Consequently, the dummy layer has to be small enough to avoid the development of profile concentrations along its thickness.

Furthermore, due to the lack of the physical properties that characterize the surface layer (**Figure 1**), different sensitivity analyses have been carried out to verify how the several free parameters of the dummy layer approach may influence the results.

The other approach investigated is based on the implementation of a *Fortran User Routine* in the code ANSYS CFX. The subroutine makes the concentration of the i -th species, initially defined in a domain, available on the interface side belonging to the adjacent domain. Thus, it becomes possible to enforce a flux condition at the interface boundaries as a function of concentrations on both sides (9).

The two novel numerical methods have been tested considering simple model problems, consisting of a fluid and a solid domain, in which tritium diffuses, connected by means of an interface. Steady-state isothermal analyses have been performed considering three elementary geometries - Cartesian, cylindrical and spherical - to validate the numerical results against the relevant analytical models.

Concerning the geometrical features, for each one of the three cases, the solid domain length has been chosen as half of the fluid domain width. In particular, fluid domain width in plane geometry is 1 [m], while the radius of the fluid domain in cylindrical and spherical geometries has been chosen equal to 0.5 [m]. The analyzed scenario consists of a fluid at rest with a constant and uniform tritium source (fixed at 1 [mol·m⁻³·s⁻¹]) and a solid external surface maintained at a constant concentration equal to zero. It has to be highlighted that the geometrical features, the Boundary Conditions (BCs) and volume sources are not physically representative of the major engineering applications related to this methodology and, consequently, these model problems have to be intended just for validation purposes.

5. Results

Results have been reported in terms of concentration distribution profile along the x -axis for the Cartesian geometry and along the r -axis for the cylindrical and spherical geometries. The computational results calculated by means of ANSYS CFX code have been compared to their analytical equivalents described in §4. Nodal concentration values obtained with the two approaches are compared to the analytical results and reported for the most significant case, i.e. the cylindrical geometry, in **Figure 2**.

Both concentration and the r coordinate are normalized to the reference variables defined in §4 and for the three geometries studied with a dummy layer thickness of 2 mm, which corresponds to 0.4% of the solid domain thickness.

In this case, results have been validated against the Dirichlet-Dirichlet analytical model. In particular, the concentrations and the net flux at the interface on both sides have been compared to the analytical values.

Results have been reported in terms of relative errors on concentration at the fluid and solid sides of the interface and error on the net flux of T_2 . They have been summarized in **Table 4** where cases A, B, C and D stand for dummy layer diffusivities of $1E-5$ [m^2/s], $1E-6$ [m^2/s], $1E-7$ [m^2/s] and $1E-8$ [m^2/s] respectively.

Table 4. Errors between numerical and analytical models with $S = 3.33E-4$.

# Cells	$\varepsilon(C_T) [\cdot 10^{-3} \%$]				$\varepsilon(C_T) [\cdot 10^{-3} \%$]				$\varepsilon(\phi_s) [\cdot 10^{-3} \%$]			
	A	B	C	D	A	B	C	D	A	B	C	D
1	0.2	0.2	0.2	0.1	21.8	22.3	23.4	34.5	5.0	4.8	3.7	7.5
2	0.4	0.4	0.4	0.2	12.3	12.3	12.9	19.2	14.8	14.8	14.1	7.5
3	0.4	0.4	0.4	0.3	10.2	10.2	10.8	16.5	16.7	16.5	16.0	10.4
4	0.4	0.4	0.4	0.3	9.7	9.7	10.2	15.5	17.3	17.3	16.7	11.3

Errors have been reported just for the case with a normalized thickness of $3.33E-4$ which represents the best case with the lowest errors. This was expected since, as mentioned before, the dummy layer thickness should be of the same order of magnitude as the surface layer.

Looking at the effect of diffusivity, it can be observed that the best configurations are those with the highest diffusivity. The reason is related to the hypothesis of no gradient in concentration across the dummy layer; indeed, if the diffusivity is higher, the profiles inside the layer are flatter, resulting in a better agreement with the analytical models. On the other hand, a decreased diffusivity would play an important role in the case of 3D solutions, since it could be responsible for an increased tritium diffusion perpendicularly to the interface normal direction.

The last parameter is the number of cells across the layer; as it can be observed, just one cell is the optimal configuration, once again for the hypothesis of no concentration gradients, and here, the positive effect is only related to the numerical approximation of the problem in case just one cell is used to discretize the interface, the gradients are inhibited by the averaging properties of the FVM.

To summarize, the best combination of parameters in terms of permeation flux to be adopted is the one with a thin dummy layer, one cell across its length and higher diffusivity. Under these assumptions, it is possible to obtain a configuration where the maximum error is related to the concentration in solid and it is equal to 0.0218%, while the errors on flux and concentration in the gas are practically equal to zero.

As shown in the previous sensitivity analyses, the layer thickness significantly influences the results and, therefore, this parameter has been studied more in details. In particular, its effect has been evaluated on the cylindrical geometric to see how the curvature of the geometry could affect the results in terms of concentration distribution.

For this reason, six different layer thicknesses, ranging from $8E-3$ to $8E-2$, have been investigated and results in terms of peaks of concentration distributions compared to analytical solutions of equations (10), are reported in **Figure 3**.

As shown in **Figure 3**, numerical results fit better to the analytical results when the layer width gets thinner and it is possible to see how, increasing the layer thickness, the concentration peak gets lower and far from the analytical results.

Moreover, it can be observed how the errors remain limited up to a normalized dummy layer thickness of $3.2E-2$. Above this threshold value, a departure from linearity is observed and the error increases significantly, due to the combined effect of curvature and the development of a concentration profile inside the layer. It can be thus concluded that the effects of curvature remain negligible when the dummy layer thickness is smaller than 1% of the local curvature radius.

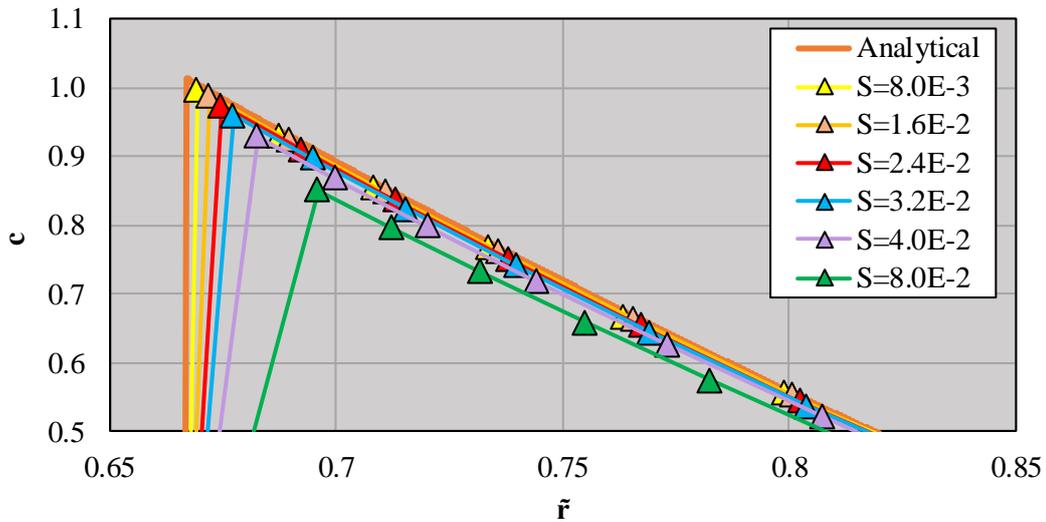


Figure 3. Zoom of concentration peaks near the gas/solid interface for different values of the layer thickness.

6. Conclusions

In the frame of the European DEMO BB studies, a research campaign has been carried out at the University of Palermo in cooperation with the Karlsruhe Institute of Technology, aimed at developing a 3D tritium permeation model that could take into account the processes occurring at the fluid-structure interfaces.

A theoretical-computational approach based on the FVM has been followed and the commercial code ANSYS CFX has been adopted. Due to the code limitations for the implementation of non-linear IFCs, two novel approaches have been developed. The first consists of the implementation of surface phenomena in a thin layer (referred to as “dummy layer”) as bulk phenomena, while the second approach is based on the implementation of a specific *Fortran User Routine* in the code.

Both methodologies have been fully validated against analytical results, with very small errors, in several diffusion problems. Furthermore, sensitivity analyses have been carried out to assess the impact on the results of the free parameters of the dummy layer approach. In particular, the best configuration (i.e. leading to the smallest numerical errors) comprehends a thin dummy layer, with one cell volume discretization and a higher diffusivity. In the case of cylindrical geometries, it has been found that the effects of curvature remain negligible when the dummy layer thickness is smaller than 1% of the local curvature radius.

Outstanding topics for the future are mainly the integration of the thermofluid-dynamics in the model and its extension to more complex geometries. Further developments of this work will aim at the implementation of other phenomena such as trapping, which is significant for fusion-relevant applications. The second step will be the expansion of the model to a multispecies model that includes the hydrogen molecules' transport and their recombination and dissociation at interfaces.

Disclaimer

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