ANALYSIS OF TRITIUM PERMEATION IN A RADIOACTIVE GAS TREATMENT SYSTEM

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INTRODUCTION

Tritium in Nuclear Fusion Systems

Nuclear fusion is the process by which two light nuclei, such as hydrogen and its isotopes, fuse to form a heavier nucleus, releasing large amounts of energy. Several combinations of hydrogen and helium isotopes can be used in fusion reactions, but those involving deuterium and tritium are currently the most extensively studied and preferred. The deuterium-tritium reaction is particularly attractive because it requires less energy to initiate than other isotope combinations that produce the same energy output, resulting in a net energy gain [1].

Since tritium is radioactive, its use is regulated by the radiation protection legislation, which establishes strict limits on the release of radioactive substances into the environment. Consequently, any facility handling tritium must implement and operate an effective radioactive waste treatment system, including dedicated tritium retention and management systems.

The presence of tritium in these systems poses significant challenges for radiological safety and environmental protection. Gaseous protium, deuterium and tritium are diatomic molecules that dissociate, particularly on metallic surfaces, and dissolve into the metal lattice in atomic form. These atoms readily recombine at free surfaces, leading to the permeation of gaseous hydrogen isotopes through metals subjected to a hydrogen concentration gradient [2].

Tritium permeation

Tritium exhibits a high permeation rate through metallic materials, allowing part of the tritium transported through pipes and process equipment to escape into the environment. This phenomenon represents a major concern for radiological safety and emission control in facilities handling tritium.

Permeation occurs through a sequence of processes involving; the adsorption and dissociation of tritium molecules at the material surface, the diffusion of atomic tritium through the crystal lattice, and its subsequent recombination at the opposite surface.

The magnitude of this phenomenon depends on both the material properties and the operating conditions. Factors such as chemical composition, microstructure and the presence of defects may enhance or hinder hydrogen isotope diffusion [2]. Likewise, increasing temperature promotes atomic mobility and consequently increases permeation [2], while pressure gradients drive tritium transport through the material [3]. Surface conditions, chemical interactions and neutron irradiation effects may also significantly modify permeation behaviour [4] [5].

To reduce tritium losses, several low-permeability materials, including oxides, carbides and nitrides, have been proposed as permeation barriers. These coatings limit the transport of hydrogen isotopes through metallic surfaces, thereby improving the safety and performance of radioactive gas treatment systems.

Study objectives

The aim of this work is to analyse tritium permeation in a radioactive gas treatment system using models developed in EcosimPro. To this end, a reference configuration without coating and a modified configuration incorporating an alumina (Al_2O_3) coating are investigated. The results are compared using the Permeation Reduction Factor (PRF) in order to evaluate the effectiveness of the coating as a tritium permeation mitigation strategy.

MATERIALS AND METHODOLOGY

Description of the Detritiation System

The radioactive gas treatment system is a key component responsible for ensuring environmental and worker safety in facilities handling tritium. This section describes in detail the operation of the system, from the inlet of contaminated gas to the release of detritiated gas.

The tritiated gas enters the system and is heated before passing through the catalytic reactor, where gaseous tritium is oxidized into tritiated water vapour (HTO). This process is carried out using a catalyst composed of platinum and palladium supported on an inert alumina substrate, which provides an oxidation efficiency of 99.98% at the operating temperature of 200ºC. The gas stream is then cooled and directed to an adsorption column equipped with a 4A zeolite molecular sieve, capable of retaining HTO with efficiencies above 99% for an adsorption period of approximately 131 days [6]. Finally, if the treated gas meets the discharge limit, it is released to the atmosphere; otherwise, it is recirculated through the system for a new detritiation cycle. This process is controlled by a valve *(Valve_C)*, which directs the flow according to the concentration of the inlet stream. At the system inlet, a second control valve *(Valve_BT)* determines the source of the incoming flow based on the recirculation flow provided by *Valve_C*. When the gas is recirculated, *Valve_BT* selects the recirculation stream as the system inlet. Otherwise, it directs the air collected from the different plant rooms to the system. The complete circuit is illustrated in **Figure 3**.

Once the molecular sieve becomes saturated, the regeneration stage begins. Heated nitrogen is circulated through the adsorption column, promoting the desorption of the tritiated water vapour retained by the adsorbent.

The resulting stream then passes through a condenser, where HTO is separated from the nitrogen by condensation. The recovered tritiated water is stored in a tank for subsequent treatment in the radioactive liquid waste treatment system. Converting gaseous tritium into HTO facilitates its handling and storage while reducing the risks associated with pressure build-up and potential leaks of gaseous tritium.

For modelling purposes, the simulation is divided into three main circuits; the radioactive circuit, the regeneration circuit and the HTO circuit. These three subsystems are integrated into a single overall model referred to as the Detritiation System (DS), as shown in **Figure 1**.

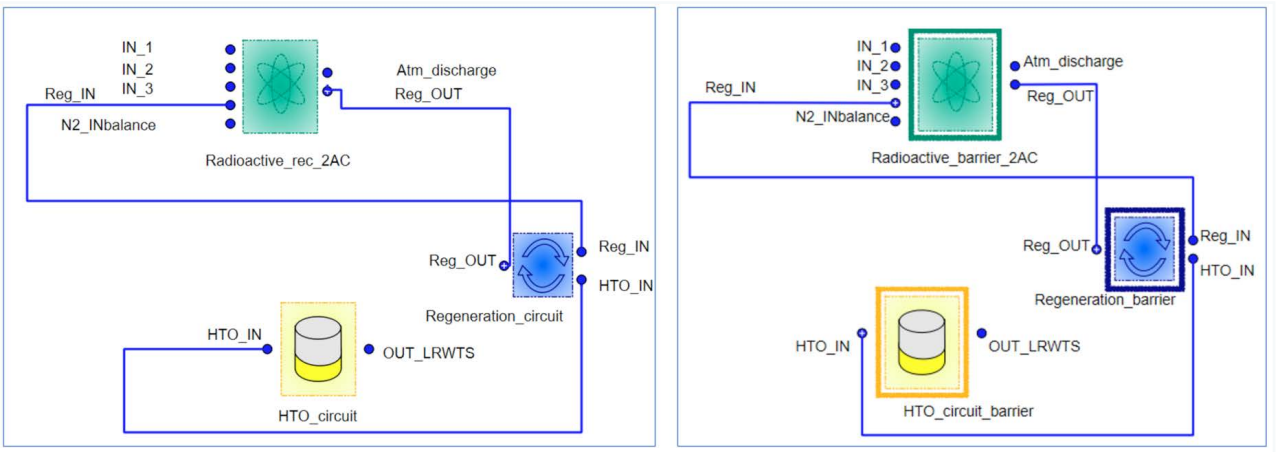

Figure 1. Reference model (left) and coated model (right) in EcosimPro.

Coating selection

Permeation barriers reduce hydrogen isotope transport by limiting the contact area between the gas and the underlying metallic surface, thereby decreasing the overall permeation rate. Current research on tritium permeation barriers mainly focuses on oxides [2], nitrides [7] and metal carbides [8]. However, ceramic materials generally exhibit superior barrier properties. In most cases, the low permeability of ceramics is attributed to the extremely low solubility of hydrogen isotopes in these materials.

Figure 2 compares the behaviour of different ceramic materials under varying temperature conditions, highlighting the differences in their permeation characteristics. It also illustrates the strong influence of temperature on permeability, which is a key factor in evaluating the performance of these materials as permeation barriers.

Among the ceramic materials investigated for use as permeation barriers, two stand out because of their effectiveness; silicon carbide (SiC) and aluminum oxide (Al_2O_3), commonly known as alumina.

Alumina exhibits lower permeability than SiC. However, its behaviour under irradiation must also be considered,

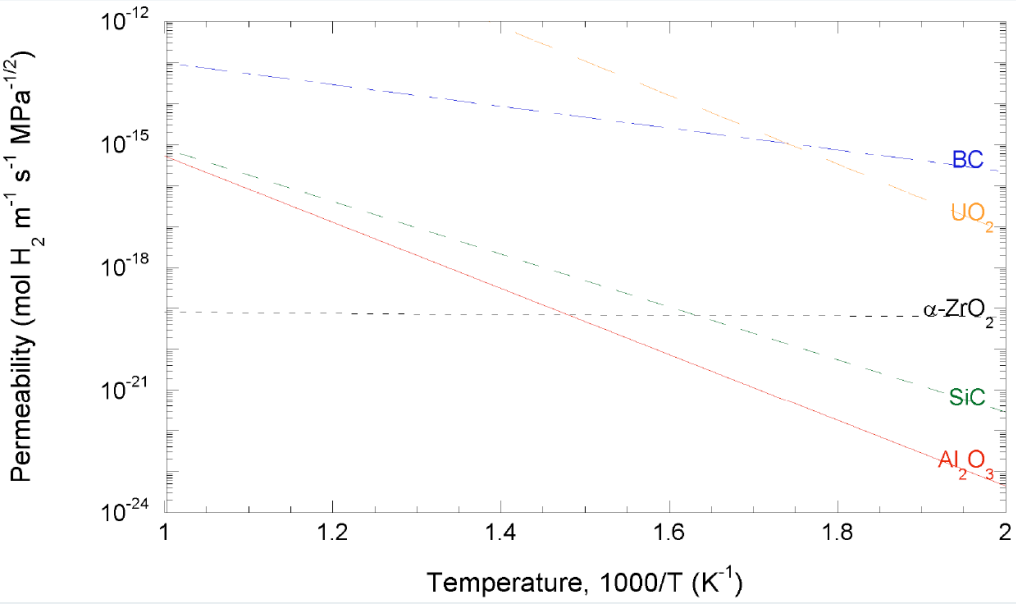

Figure 2. Ceramics permeability [2].

as previous studies have shown that SiC performs better under radiation exposure [2]. Irradiation promotes cracking of the coating, increasing the effective contact area between the gas and the underlying metal, which in turn enhances tritium permeation. In the present study, low radiation levels are expected in the area where the detritiation system is located; therefore, alumina was selected as the coating material. The coefficients required for modelling this material are listed in **Table 1**.

The Permeation Reduction Factor (PRF) is a key parameter used to evaluate the effectiveness of a permeation barrier. It is defined as the ratio between the permeability of the uncoated substrate and that of the coated substrate. The higher the PRF value, the more effective the coating is in reducing permeation [9].

Equation 1.

$$PRF = \frac{P_{uncoated}}{P_{coated}}$$

Model development

The simulation model is divided into three circuits, each performing a specific function:

Radioactive circuit: the detritiation process converts gaseous tritiated species (HT) into tritiated water.

Regeneration circuit: once the molecular sieve in the adsorption column becomes saturated, the regeneration process begins. During this stage, the HTO adsorbed by the molecular sieve is desorbed and recovered.

HTO circuit: the condensed tritiated water is collected in a storage tank and transferred to the radioactive liquid waste treatment system.

For each component and process stream in the system, it is necessary to define both the atomic species diffusing through the material and the molecular species present in the corresponding stream. These species are listed in **Table 2**.

	Diffusion coefficient		Solubility coefficient		Dissociation coefficient		Ref.
	$D = \alpha * \exp[(-b/RT)]$ m^2/s		$Ks = \alpha * \exp[(-b/RT)]$ $mol/(m^3 Pa^{0,5})$		$Ks = \alpha * \exp[(-b/RT)]$ $mol/(m^2 sPa)$		
	a	b	a	b	a	b	
T	6.36E-09	132000	5.50E-03	22500	-	-	(Rion A. Causey)
H	1.10E-08	132000	5.50E-03	22500	-	-	(Rion A. Causey)
HT	-	-	-	-	1.57E-08	49268	-
HTO	-	-	-	-	0	0	-
T2	-	-	-	-	1.28E-08	40239	-
H2	-	-	-	-	2.22E-08	69611	-

Table 1. Alumina material coefficients.

	Atomic species	Molecular species
	Radioactive gas circuit	
Beginning - End	Atomic_TH	Coolant_gas
	Regeneration circuit	
Beginning - End	Atomic_TH	Coolant_gas
	HTO circuit	
Beginning - End	Atomic_TH	HTO_only

Atomic_TII: T y H atomics.
Coolant_gas: T2, H2, HT, T2O, H2O y HTO.
HTO_only: HTO.

Table 2. Atomic and molecular species in the model.

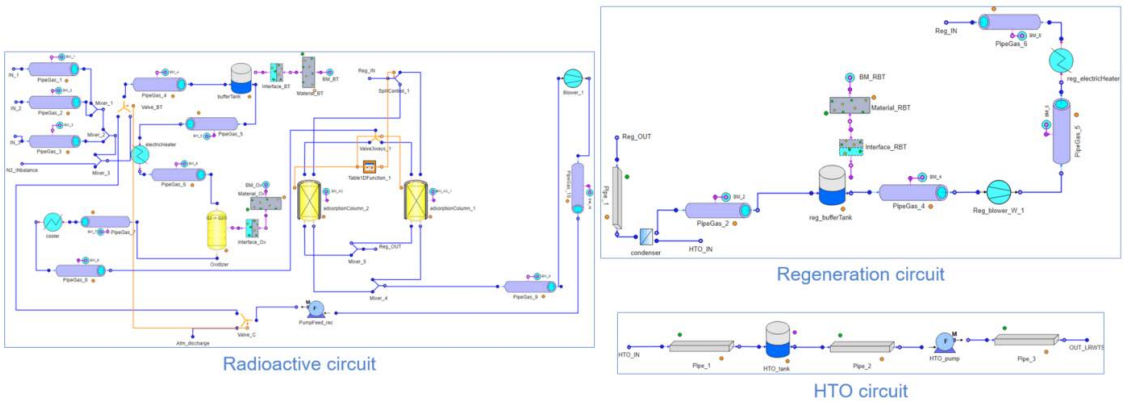

Figure 3. Reference models in EcosimPro. Radioactive circuit, regeneration circuit and HTO circuit.

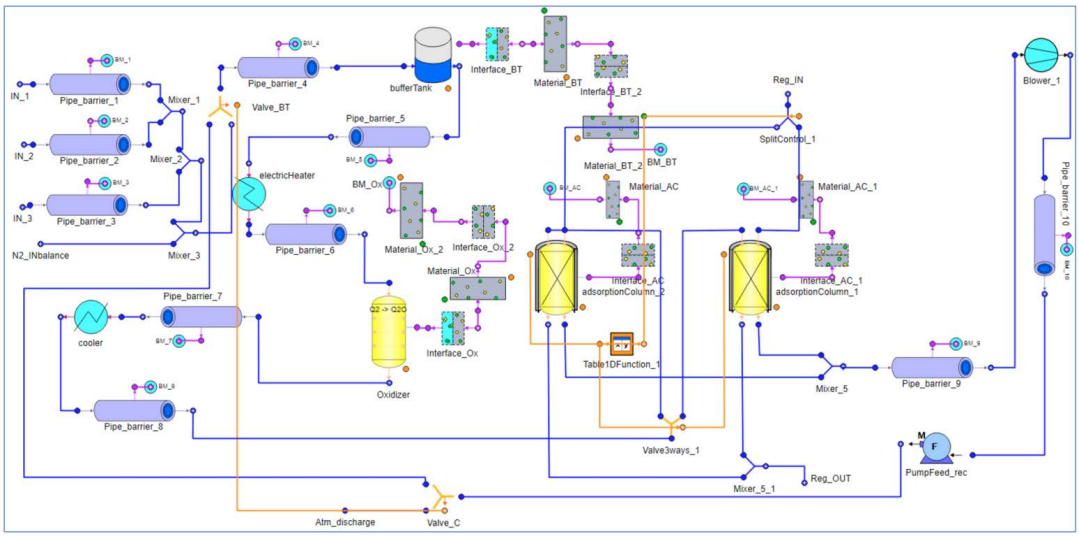

Figure 4. Coated model in EcosimPro. Radioactive circuit.

The system piping, as well as the process equipment, is made of TP304L austenitic stainless steel. In addition, a 99.999% pure Al_2O_3 coating is applied to reduce tritium permeation.

Al_2O_3 coated model

The coated model reproduces the equipment used in the reference model while maintaining the same operating conditions, material properties and boundary conditions. Modifications are introduced only in the radioactive circuit, as shown in **Figure 4**, since the other two circuits do not contain atomic species capable of permeating through the system materials and therefore do not require a protective coating.

A 1 µm thick alumina coating is applied to all components where its implementation is feasible. This thickness was

selected because it corresponds to the value adopted in the study *"Assessment Report on Tritium Term Sources and Different Types of Tritium Permeation Barriers Relevant for Fusion and Fission Reactors"*, carried out within the TRANSAT (TRANSversal Actions for Tritium) project) [10].

RESULTS

Reference model

To characterize tritium permeation in the radioactive waste treatment system, the evolution of the HT and HTO concentrations at the system inlet, together with the corresponding flow rates, was analysed.

The results presented in **Figure 5** and **Figure 6**, show that at the beginning of operation, the inlet stream consists mainly of HT originating from the plant rooms. As the gas circulates through the system, it is oxidized and converted into HTO. Since the discharge criteria is not initially met, the gas stream is recirculated, resulting in a gradual decrease in the HTO concentration until the required discharge limit is achieved. Once these conditions are satisfied, the treated gas is released and the system receives a new HT-rich stream from the plant rooms, repeating the operating cycle.

The behaviour shown in **Figure 7** indicates that the catalytic reactor exhibits the highest permeation rate, which can be attributed to its high operating temperature (200ºC), one of the parameters with the greatest influence on this phenomenon. A significant increase in permeation is also observed in the adsorption column during the regeneration stage, owing to the temperature rise to 350ºC, which promotes the release of the hydrogen and tritium previously retained within the material. Regarding the piping, the highest contribution to permeation corresponds to the section carrying the stream with the highest tritium concentration, highlighting the importance of this parameter in the permeation process. After one year of operation, the total system permeation reaches $3.495^{x}10^{-17}$ g/s, although steady-state conditions have not yet been achieved.

Coated model

As in the reference model, the HT and HTO concentrations at the system inlet, together with the corresponding flow

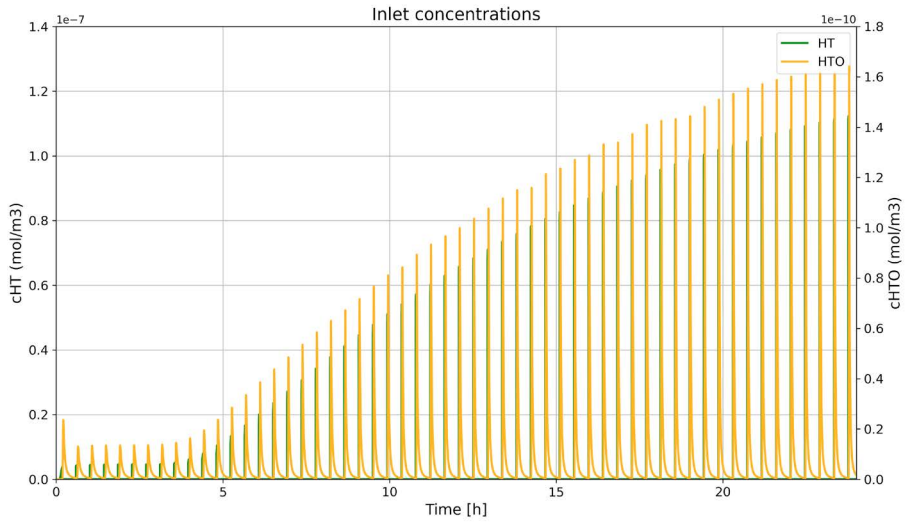

Figure 5. Radioactive circuit's inlet's concentrations. Reference model.

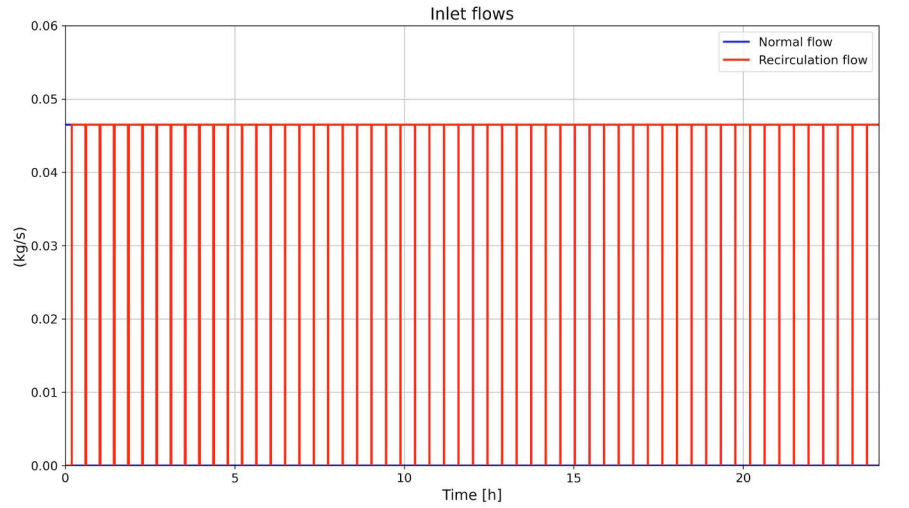

Figure 6. Radioactive circuit's inlet's flow rate *(Valve_BT)*. Reference model.

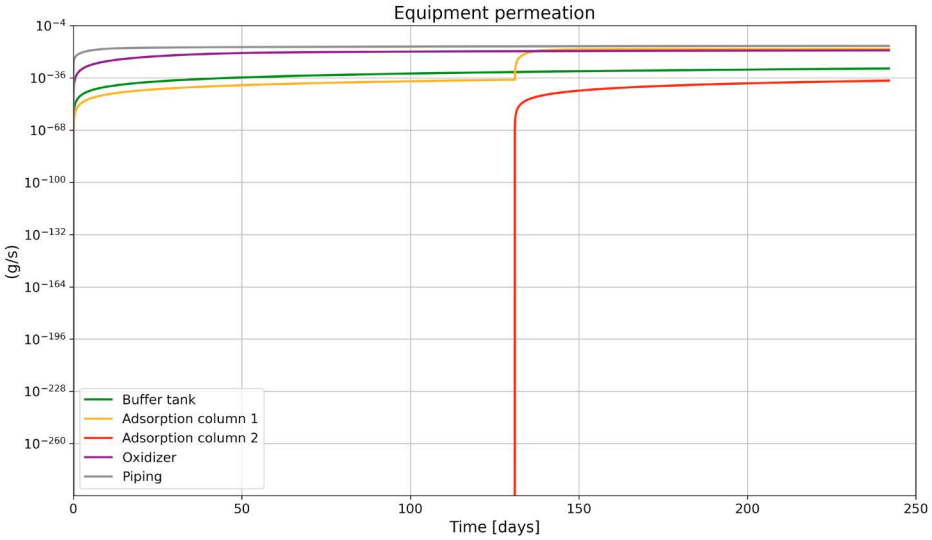

Figure 7. Radioactive circuit's permeations. Reference model.

rates, are first presented in order to analyse their evolution under the established operating conditions.

Figure 8 and **Figure 9** show that the HT concentration is lower in the coated configuration. This behaviour is attributed to the permeation barrier, which reduces tritium losses to the surroundings, thereby increasing the amount of tritium remaining within the process stream and extending the recirculation period. As a result, a larger fraction of the tritium is converted into HTO before the gas can be discharged, reducing the frequency at which new HT-rich streams from the plant rooms enter the system and leading to lower HT concentrations over time.

As in the uncoated configuration, **Figure 10** shows that the catalytic reactor exhibits the highest permeation rate during normal operation due to its high operating temperature. However, during the regeneration stage, the adsorption column becomes the component with the highest permeation as a result of the temperature increase associated with this process.

Regarding the piping, the largest contribution to permeation is observed in pipe 6. Unlike the reference model, the longer recirculation periods reduce the flow of HT-rich streams through the inlet pipes, making temperature the dominant factor and causing pipe 6 to become the most permeable section of the system.

A significant increase in permeation is also observed during the regeneration of the adsorption columns. This effect is more pronounced in the coated configuration because the diffusion coefficient of alumina is more sensitive to temperature, although the material continues to exhibit excellent performance as a tritium permeation barrier.

Overall, both the piping and the process equipment show a substantial reduction in permeation due to the presence

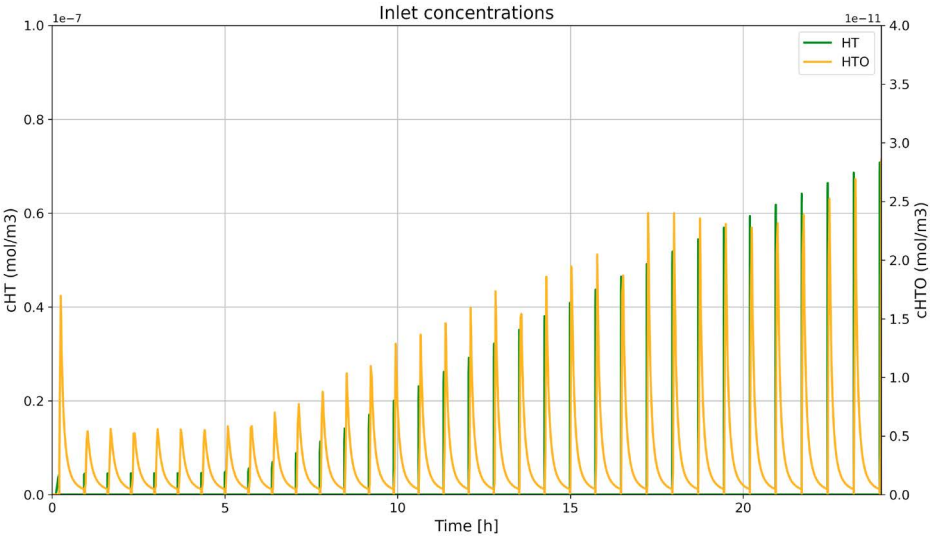

Figure 8. Radioactive circuit's inlet's concentrations. Coated model.

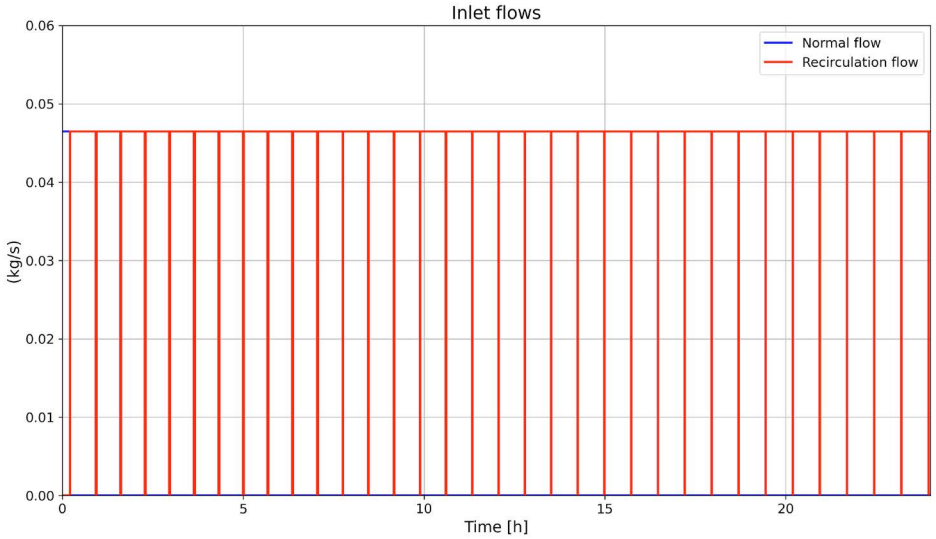

Figure 9. Radioactive circuit's inlet's flow rate *(Valve_BT)*. Coated model.

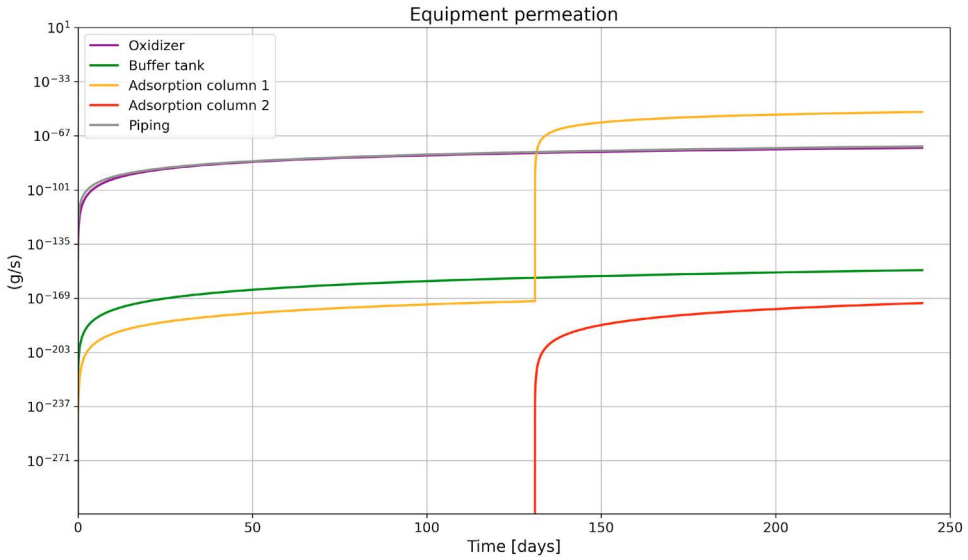

Figure 10. Radioactive circuit's permeations. Coated model.

of the alumina coating, demonstrating its effectiveness in limiting the transport of permeating species.

Permeation Reduction Factor (PRF)

The determination of the Permeation Reduction Factor (PRF) requires permeation to reach steady-state conditions. For this purpose, an additional simulation was performed simulating only pipe 6, which operates at the highest temperature and exhibits the highest permeation rate in the coated model.

Steady-state is reached when the material, or materials in the coated configuration, have absorbed the maximum amount of hydrogen and tritium they can retain, after which the permeation rate remains essentially constant.

As shown in **Figure 11** and **Figure 12**, the metallic substrate in the coated configuration reaches steady-state conditions in a shorter time because the ceramic barrier limits permeation to the surroundings, promoting the accumulation of hydrogen isotopes within the metal and accelerating its saturation. In contrast, the ceramic coating requires a longer time to become saturated, since it acts as a barrier to hydrogen isotope transport and delays diffusion.

By comparing both configurations under steady-state conditions, as shown in **Figure 13**, the PRF is calculated as the ratio between the permeation of the uncoated system and that of the coated system. This parameter provides a direct quantitative measure of the reduction in tritium permeation achieved by the alumina coating.

Equation 2: PRF Equation

$$PRF = \frac{P_{uncoated}}{P_{coated}} = \frac{1.2032e^{-16}}{1.2712e^{-24}} = 9.46e^{7}$$

A PRF of this magnitude demonstrates that the applied alumina coating is highly effective in reducing tritium permeation. Specifically, the tritium permeability of the coated material is $9{,}46 \times 10^{7}$ times lower than that of the uncoated material. This result indicates that the coating dramatically decreases the rate at which

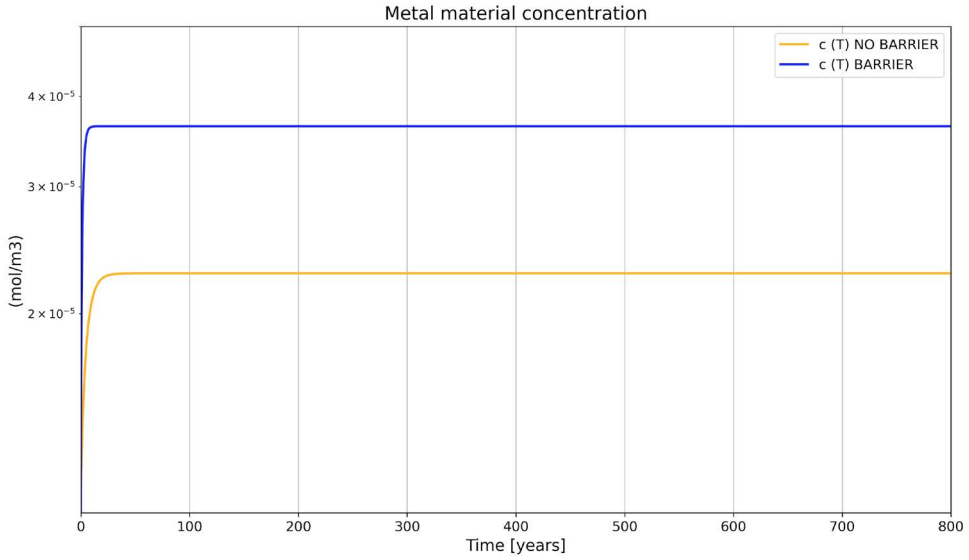

Figure 11. Tritium concentrations in the metal material of the test pipe.

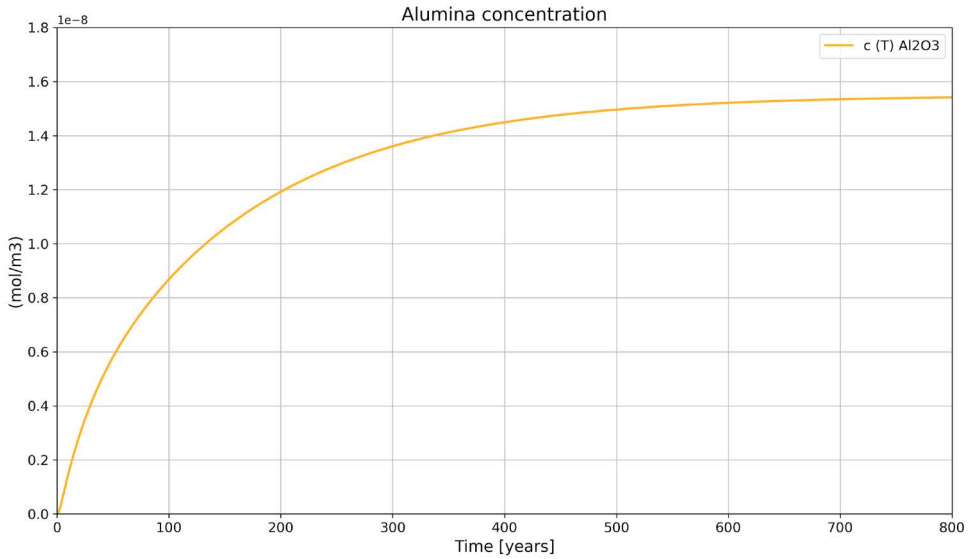

Figure 12. Tritium concentration in the ceramic material of the test pipe.

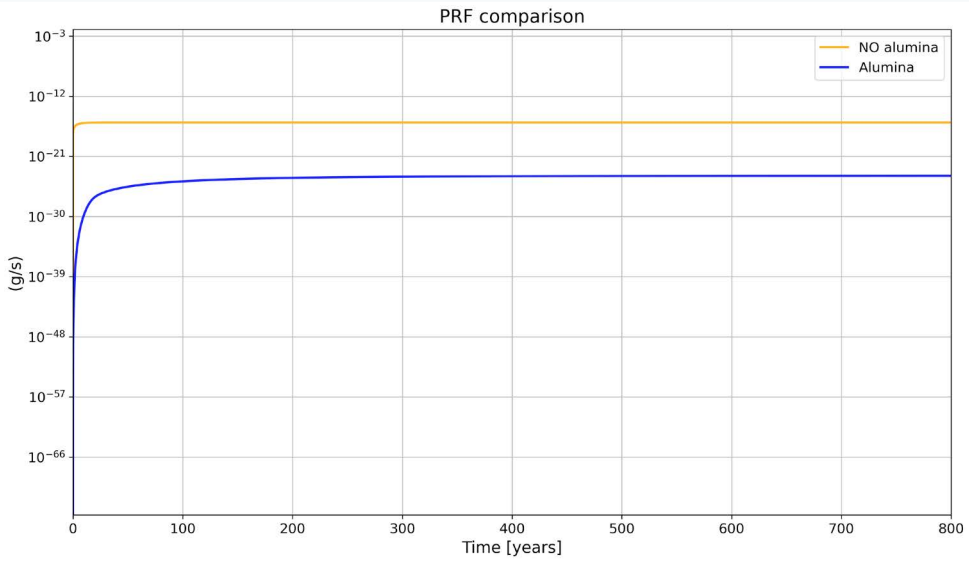

Figure 13. Permeation comparison.

tritium is able to permeate through the material, confirming its suitability as an effective permeation barrier.

CONCLUSIONS

The present work has investigated tritium permeation in a radioactive gas treatment system by means of numerical simulation and evaluated the effectiveness of an alumina (Al_2O_3) coating as a permeation barrier. The results show that temperature is one of the most influential factors governing tritium losses, with the highest permeation rates occurring in components operating at elevated temperatures. The incorporation of the coating was also found to modify the system behaviour by promoting the accumulation of hydrogen and tritium within the material, leading to earlier saturation and consequently longer recirculation periods. Nevertheless, the alumina coating proved to be highly effective, achieving a PRF of $9{,}46 \times 10^7$. These findings demonstrate the potential of ceramic coatings to significantly reduce tritium permeation in radioactive gas treatment systems and provide valuable information for the design and optimization of future fusion nuclear facilities.

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