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Parameter study and proposal for tritium control strategy

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Summary

The aim of this task was to perform a parameter study indicating which combination of the different tritium control options are capable of meeting possible future safety requirements, complemented by suggestions for a viable tritium control strategy. We have done this on the basis of the Chinese HTR-PM reactor. For this purpose, a conceptual steam generator lay-out based on the Fort Saint Vrain design (helical coil tube bundle type with Incoloy 800 as the tube material) was assumed, both for the rather conservative HTR-PM temperatures and for higher temperatures. Using the obtained steam generator characteristics in combination with a tritium balance sheet and reasonable assumptions on operating conditions and safety requirements, we described the individual and combined effect of several operating parameters on tritium control. Our results indicate that compliance with plausible tritium control requirements can indeed be achieved with reasonable effort both for electricity generation using a closed steam cycle and for process steam generation with an open steam cycle. However, for new-build HTR, definite country-specific licensing requirements (e.g. chronic and accidental tritium release) are yet to be determined and will shape the required tritium control strategy

Approval

Date	By
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Tritium Transport Modeling for High Temperature Reactors

FP7 ARCHER project
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Deliverable D23·71

Elio D'Agata, Michael A. Fütterer

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Tritium Transport Modeling for High Temperature Reactors

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Elio D'Agata, Michael A. Fütterer

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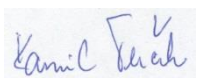
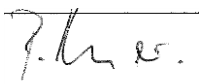
Signature				

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Summary

In a High Temperature Reactor, tritium is produced by a number of mechanisms such as ternary fission and neutron capture on boron (in control rods), lithium (in graphite) and ^{3}He (in the He coolant). Due to its high mobility, some of this tritium ends up in the primary helium cooling circuit from where it can be extracted by the coolant purification system to keep the partial pressure of tritiated compounds low. Nevertheless, the partial pressure of tritium in the coolant is the driving force for permeation across the heat exchanger from the primary cooling system into the secondary cooling system. From there the contamination may further propagate and ultimately end up in the environment.

The aim of this task was to perform a parameter study indicating which combination of the different tritium control options are capable of meeting possible future safety requirements, complemented by suggestions for a viable tritium control strategy. We have done this on the basis of the Chinese HTR-PM reactor.

For this purpose, a conceptual steam generator lay-out based on the Fort Saint Vrain design (helical coil tube bundle type with Incoloy 800 as the tube material) was assumed, both for the rather conservative HTR-PM temperatures and for higher temperatures. Using the obtained steam generator characteristics in combination with a tritium balance sheet and reasonable assumptions on operating conditions and safety requirements, we described the individual and combined effect of several operating parameters on tritium control. Our results indicate that compliance with plausible tritium control requirements can indeed be achieved with reasonable effort both for electricity generation using a closed steam cycle and for process steam generation with an open steam cycle. However, for new-build HTR, definite country-specific licensing requirements (e.g. chronic and accidental tritium release) are yet to be determined and will shape the required tritium control strategy.

Approval

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1 Introduction

In a High Temperature Reactor, several parasitic sources of tritium (e.g., in growing order of importance, ternary fission, boron in control rods, lithium impurities in graphite, ^{3}He traces in the coolant) can contaminate the primary helium coolant. Despite helium purification, the high mobility of tritium may lead to unwanted permeation into a secondary cooling circuit. Assuming for instance an indirect Rankine cycle for power conversion, tritium from permeation across the steam generator (SG) may be found in the secondary steam. The control of this tritium escape is a potential safety and licensing issue and is evaluated here for the European ARCHER project on HTR technology.

Considerations on tritium permeation from a primary helium cooling circuit to a secondary steam circuit and suitable tritium control measures have been analyzed extensively (a good review can be found in [1]) and were published in the past on existing reactors (e.g. Fort Saint Vrain [2], AVR [3]), for planned reactors (e.g. HTR-Modul [3], NGNP [4], [5]) and for fusion [7], [8].

For the purpose of this study we have used the operation characteristics of a power plant similar to the Chinese HTR-PM design [12] and determined the allowable tritium permeation rate through a SG for the cases of a) a closed steam cycle for electricity generation and b) an open steam cycle for process steam. In the former case, the allowable permeation can be quantified from considerations on worker exposure, in the latter from end-user exposure by a contaminated process product.

The design based on the Fort Saint Vrain SG (helical coil tube bundle type) was assumed. This type of SG is considered as a sort of reference design for HTR. It was built by Swiss SULZER AG and was the basis for a numerical design tool named He2SG for "Helium-Heated Steam Generator" written by CEA, France [8] and used courtesy of CEA for the ARCHER project. He2SG was also used for the analysis published in [6].

He2SG essentially computes the temperature profile along a heat exchange surface area between primary He and secondary steam with a tube thickness chosen to accommodate the system pressure. He2SG requires the following information as input: thermal power rating, primary or secondary top cycle temperature, primary bottom cycle temperature, desired He and steam pressure. Other input parameters such as tube diameter, number of tube banks, pitch and helix angle were selected to meet heat transfer and pressure drop requirements on the helium and water side of the SG.

Using the obtained steam generator temperature profile and surfaces, tube thicknesses and using material data for tritium permeability in conjunction with available experience from Fort Saint Vrain and from fusion studies, a 1-d FEM tritium permeation analysis was performed. Based on the acceptable tritium permeation rate, a parametric study allowed us to assess the acceptable tritium partial pressure in the primary circuit. From this we deduced requirements for gas chemistry and helium clean-up system (e.g. required slip-stream fraction, hydrogen swamping, oxidation potential) and performance of possibly required tritium permeation barriers on the steam generator tubing.

Complementary to the study in HTR-PM operating conditions we have performed a similar analysis for an HTR system with a significantly higher reactor outlet temperature of helium and have quantified the required effort on tritium control.

The result of this work is a parameter study, indicating which combinations of the different tritium control options are capable of meeting the safety requirements (worker/end-user exposure). It is complemented by suggestions for a viable tritium control strategy and related R&D needs.

This report first explains the adopted methodology which required the determination of the SG design characteristics. Those were obtained using the computational tool He2SG and it is described how this was done. We also recall tritium transport mechanisms and explain how we quantified tritium permeation across a steam generator. Our methodology requires input related to licensing limits and operational parameters. Those are discussed, and by choosing a number of plausible values we demonstrate the various degrees of freedom that exist to derive a consistent tritium control strategy for the example of the HTR-PM reactor which is dedicated to electricity generation. The case of process steam generation (open steam cycle) is equally addressed.

2 Model and Methodology

For the purpose of this study we consider a simplified model of the primary and secondary cooling circuit of an HTR with a SG as the interface. An overview of tritium transport mechanisms is given in section 8. We assume already here that tritium permeation is a diffusion-controlled process (as opposed to a surface reaction based process) which is justified due to the usually simultaneous presence of relatively large amounts of hydrogen in the helium (cf section 8).

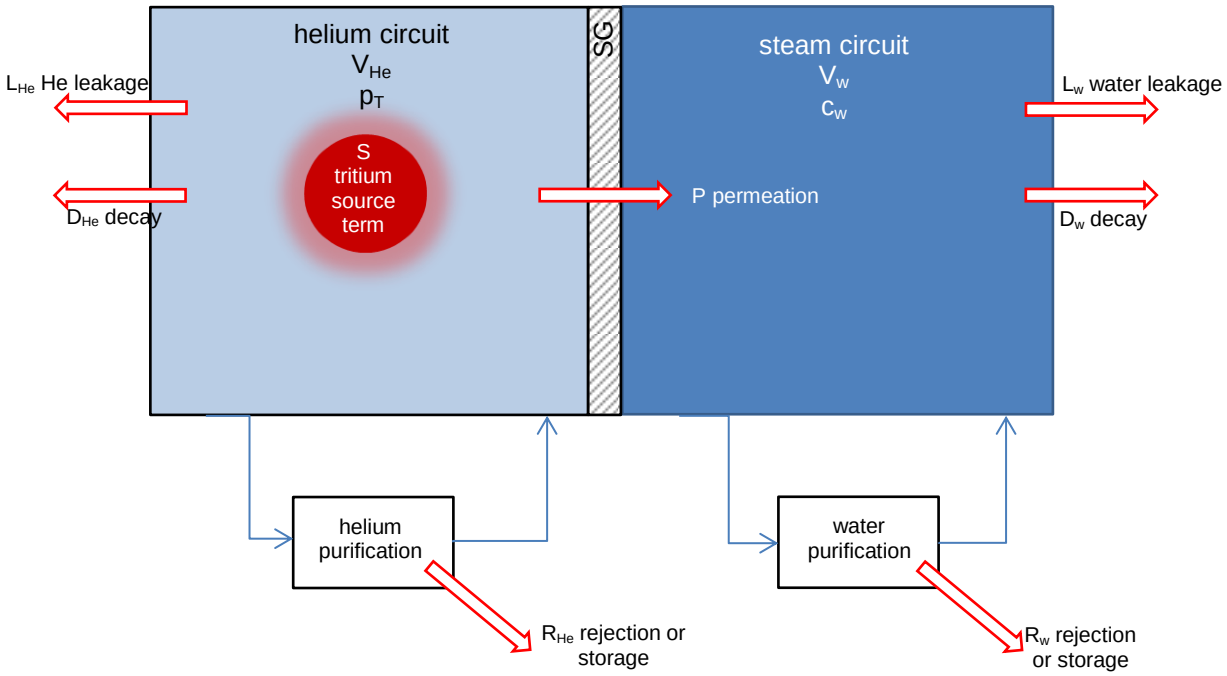

Figure 1: Simplified model of tritium fluxes in an HTR system coupled to a SG

For steady-state conditions, we can draw the following tritium balances:

- for the overall system:

$$S = L_{He}\left(\sqrt{\boldsymbol{p_T}}\right) + D_{He}\left(\sqrt{\boldsymbol{p_T}}\right) + R_{He}\left(\sqrt{\boldsymbol{p_T}}\right) + L_w(\boldsymbol{c_w}) + D_w(\boldsymbol{c_w}) + R_w(\boldsymbol{c_w})$$

- for the water circuit:

$$\boldsymbol{P} = L_w(\boldsymbol{c_w}) + D_w(\boldsymbol{c_w}) + R_w(\boldsymbol{c_w})$$

- for the SG:

$$\boldsymbol{P} = \boldsymbol{C_{SG}}\frac{1}{PRF}\left(\sqrt{\boldsymbol{p_T}}\right)$$

$$\boldsymbol{P} = \frac{2\pi l}{\ln {}^{b}/_{a}}\ \frac{\Phi(T)}{PRF}\left(\sqrt{\boldsymbol{p_T}}\right)$$

with

- $\Phi(T)\ \left[\frac{at}{m\ s\ \sqrt{Pa}}\right]$: tritium permeability of the used SG tube material (from literature)
- PRF [-]: the permeation reduction factor, a measure to quantify the required effort for tritium permeation reduction by permeation barriers and/or gas chemistry
- l [m]: the SG tube length for which the permeation is calculated
- b [m]: the outer SG tube diameter
- a [m]: the inner SG tube diameter

This is a simple set of 4 equations for 4 unknowns (in bold) which are:

- **p_T**: the "equivalent tritium partial pressure" in the helium (we argue in terms of partial pressure because of the gaseous character of the primary fluid); this pressure refers to pure T_2 gas, which in practice does not exist due to the simultaneous presence of H_2 in the helium circuit, thus forming preferentially HT molecules;
- **c_w**: the tritium concentration in the steam circuit (we argue in terms of concentration, because the vast majority of tritium will exist in the form of HTO water molecules in the secondary circuit due to fast isotopic exchange reactions of T with H_2O on the SG surface)
- **P**: the tritium permeation rate through the steam generator
- **C_{SG}**: a characteristic SG constant which is design specific and combines geometry, temperature and permeability characteristics of a SG: $\boldsymbol{C_{SG}} = \frac{2\pi l}{\ln {}^{b}/_{a}} \quad \Phi(T)$

In the following we specify one by one the different terms before solving the mass balances.

	Step	Method	Variables
1.	S: Tritium source term released to helium	requires a neutronic core design with activation calculation or an experience-based assumption	release fraction from coated particles impurities in graphite control rod design
2.	c_W: Tritium concentration in secondary water/steam circuit	determine acceptable tritium concentration in secondary circuit from radiological or licensing considerations; different for power conversion or process steam generation	type and length of worker exposure, water leak rate, turbine hall ventilation, slip stream fraction for water purification, efficiency of water purification
3.	L_w: Tritium escape from water leakage	use best estimate	water leak rate
4.	D_w: Tritium decay in water circuit		water inventory
5.	R_w: Tritium removed by water purging	use best estimate	water purge slip stream fraction; efficiency of detritiation
6.	C_{SG}: characteristic SG constant $\boldsymbol{C_{SG}} = \frac{2\pi l}{\ln {}^{b}/_{a}} \quad \Phi(T)$	design a SG to obtain surface and temperature profiles and derive a function $P = C_{SG} \frac{1}{PRF} \sqrt{p_T}$	H_2/HT ratio in helium and permeation barriers resulting in a permeation reduction factor PRF
7.	L_{He}: Tritium escape from helium leakage	use best estimate	helium leak rate, helium inventory
8.	D_{He}: Tritium decay in helium circuit		helium inventory
9.	R_{He}: Tritium removed by helium purification	from tritium balance	helium slip stream fraction to purification; efficiency of detritiation
10.	sum up tritium fluxes leaving the system		
11.	adjust input variables (e.g. PRF) such that source term equals sum of tritium fluxes leaving the system		

Table 1: Process steps to determine tritium flux balance

The following subsections refer to these steps in more detail

2.1 S: Tritium source term

The determination of the tritium production by neutronics calculations would go beyond the objectives and possibilities of this study. Therefore, we rather use experience-based assumptions.

Tritium is produced in the core of nuclear reactors by ternary fission of uranium (^{233}U, ^{235}U) and plutonium (^{239}Pu, ^{241}Pu) [28] [29], and by the interaction of neutrons with lithium, boron, and helium impurities in the surrounding core graphite and helium cooling fluid [30]. Table 2 (data from [30]) shows tritium production for Peach Bottom and Fort St. Vrain obtained from data collected during reactor operation. The tritium production for the three other reactors was extrapolated from there. It is not quite clear why this extrapolation has led to a lower tritium production term from ^{3}He in a 3000 MWth HTGR compared to a 1500 MWth HTGR.

	Peach Bottom (66.5 MWth) (measured)	Fort St. Vrain (842 MWth) 6th year (measured)	1500-MWth HTGR (extrapolated)	3000-MWth HTGR (extrapolated)	PNP-500 MWth (extrapolated)	250 MWth HTGR assumptions for this study
Ternary fission [Ci/yr]	283	2730	9950	11000	3190	1500
Power-specific production by ternary fission [Ci/yr-MWth]	4.26	3.24	6.6	3.6	6.38	6
^{6}Li [Ci/yr]	15.1	481	1190	2810	150	1000
^{3}He [Ci/yr]	14.7	370	4540	3680	2000	
^{10}B [Ci/yr]	206	330	Unavailable	919	260	
Total [Ci/yr]	518.8	3911	15680	18409	5600	2500
Power-specific total tritium production [Ci/yr-MWth]	7.8	4.64	10.45	6.14	11.2	10

Table 2: Measured tritium source terms for Peach Bottom and Fort St. Vrain extrapolated to other HTGRs [30] and assumptions used for this study

Although it is unclear how exactly the extrapolated values were obtained, Table 2 shows that the biggest contributor to tritium production is ternary fission which is why the reactor power plays an important role in the tritium production with a typical value of 10 Ci/yr-MWth. Extrapolating from there to a single 250 MWth module (similar to HTR-PM) we expect an annual production of the order of 2500 Ci/yr (i.e. 1.649E15 at/s). Ternary fission contributes to this with values between 3.24 and 6.6 Ci/yr-MWth. We assume here a value of 6 Ci/yr-MWth.

For the purpose of this study we understand as the tritium source term S the quantity of produced tritium released into the helium coolant. Specifically for tritium produced in the fuel particles, there are different approaches to determine how much is actually released into the helium coolant:

- Steinwartz [3] p. 3-15 indicates that SiC is leak tight for tritium below 1000°C and that only defect particles would release tritium, leading to a tritium release fraction of 4E-5.
- Steinwartz also indicates that for the licensing of the THTR a very conservative release of 10% was assumed. This was probably applicable due to the use of BISO fuel which proved to be less performing than modern TRISO fuel. For the licensing of the HTR Modul, also a release fraction of 10% was assumed.
- Oh [5] has assumed a 100% tritium release from ternary fission for a VHTR study, probably because for a VHTR the fuel temperatures were assumed to be significantly above 1000°C.

For consistency with the HTR Modul licensing approach we will use in the following a 10% release fraction for tritium from ternary fission inside coated particles as the reference. This leads to a source term of tritium released into the primary helium coolant of 1150 Ci/yr for our 250 MWth model reactor. The effects of higher and lower release fractions will also be discussed.

^{6}Li impurities in graphite can be further reduced, however at the expense of importing other impurities (in particular Cl) which may actually lead to more hazardous activation products than tritium. Tritium production on ^{10}B in control rods can to some extent be reduced by design while tritium production from ^{3}He impurities in the coolant (today extracted from natural gas) can only be reduced by unrealistically expensive isotopic tailoring.

2.2 c_W: Tritium concentration in steam circuit

To tentatively determine a limit for the tritium concentration c_W in the water circuit we have considered the following approaches:

1. **Based on the maximum admissible dose on a person who may ingest water from the secondary circuit:**
   The admissible dose rate for a radiological worker is 20 mSv/yr [27] and the effective dose factor for ingestion of tritiated water is $e_{ing}(50)$ = 1.81E-11 [Sv Bq^{-1}] [26], [27]. Consequently, the maximum activity that a radiological worker is allowed to ingest in one year is 20E-3/1.81E-11 = 1.11E9 [Bq] (or 3.108E-6 g/yr or 6.23672E17 at/yr or 1.9776E10 at/s). Assuming that it takes 100 radiological workers to ingest 100 m^3 of cooling water inventory (a likely value for HTR-PM), we calculate a maximum tritium concentration in the cooling water c_w = 6.2366E17 at/m^3.

2. **Based on the inhaled dose by workers in the turbogenerator building:**
   Steinwartz [3] discussed different assumptions on release limits and their consequences on the maximum allowable tritium concentration in the secondary cooling circuit. By assuming a 500 mrem/yr (= 5 mSv/yr) limit for non-professionally exposed workers from inhalation of humidity caused exclusively by water/steam leakage into the turbogenerator building, he proposed an upper tritium concentration limit in the secondary cooling system of 9.5E-3 Ci/t, noting that for the German THTR reactor, the allowable value was by a factor 3.8 lower.
   Steinwartz' assumptions were the following:
   - working time: 40 h/week, 50 weeks/yr
   - exposure limit: 500 mrem/yr (= 5 mSv/yr) for non-professionally exposed workers according to German radioprotection legislation valid in 1987; today, this value would be 1 mSv/yr [27];
   - inhaled air volume 2500 m^3/yr
     ➔ allowable tritium concentration in air 4.9E-7 Ci/m^3
   - temperature in turbine hall 40°C
   - humidity in turbine hall 100%
     ➔ water concentration in air of turbine hall 5.1E-5 t_{H2O}/m^3_{air}
     ➔ allowable tritium concentration in secondary cooling circuit: 9.5E-3 Ci/t

   Once the allowable tritium concentration in the cooling water is determined, the allowable tritium permeation from the SG can be deduced by a mass balance.

In the following, we have used Steinwartz' approach for the determination of the allowable tritium concentration in the cooling water, with the exception of the exposure limit.

- **Working time:** 40 h/week, 50 weeks/yr. We kept these extremely conservative assumption although in reality already for noise exposure reasons a worker will never spend that much time in the turbine hall;
- **Exposure limit:** contrary to Steinwartz we assume here that only radiological workers have access to the turbine hall; for those, an exposure limit of 100 mSv in a 5 year period with a maximum annual exposure of 50 mSv/yr is applicable [27], i.e. on average 20 mSv/yr, a value which is 4 times higher than what Steinwartz has assumed;
- **Turbine hall ventilation:** like Steinwartz we conservatively assume absence of turbine hall ventilation which, in practice, however is an effective way to reduce exposure;

- **Water/steam leakage:** the Steinwartz approach does not need to make assumptions on leakage, he simply assumed the same tritium concentration in water as in humidity. But in practice, together with turbine hall ventilation, this leakage will determine the exposure;

Approaches 1 and 2 are deliberately far-fetched and unrealistic so as to underline their conservatism. They produce similar limits for the acceptable tritium concentration in secondary cooling water with a closed steam cycle. Based on approach 2, we assume c_W = 7.904E17 at/m^3 = 3.8E-2 Ci/t = 1.406E6 Bq/l.

In the US, tritium can be rejected at 37 kBq/l [4] which means that for the conditions we used here, a 1:38 dilution with clean water would be required before releasing any secondary cooling water into the environment.

Note that for process steam generation (open cycle) the authors of [5] assumed a similar secondary coolant concentration limit of 120 μCi/m^3 (4.4 kBq/kg) as the HTR Modul limit (5 kBq/kg = 2.81E15 at/m^3).

2.3 L_w: Tritium escape from water/steam leakage

The amount of tritium escaping from the cooling water by leakage becomes:

$$L_w = -c_w V_w l_w$$

We assumed a water volume V_w = 100 m^3 and, initially, a leak rate of l_w = 1E-9 1/s, corresponding to 8.64 l/d. As we will see later, this leak rate may be technologically unrealistic (Steinwartz [3] reports for the THTR a value as high as 0.5% of the SG throughput thus leading to l_w = 4.725E-6 1/s), and overly conservative because it would impose a higher PRF effort. If compatible with licensing requirements, it is thus "beneficial" for the required tritium control effort to let some of the tritium escape by water/steam leakage.

2.4 D_w: Tritium decay in water circuit

The amount of tritium decaying in the water circuit per unit time depends on the total water volume and on the tritium concentration in this volume. The decay constant for tritium is λ = 1.7797E-9 1/s.

$$D_w = -\lambda V_w c_w$$

2.5 R_w: Tritium removed by water purging

The amount of tritium removed from the cooling water by purging (and to be treated, possibly detritiated or diluted before rejection) becomes:

$$R_w = -c_w \eta_w V_w p_w$$

A typical slip stream fraction of the water throughput is 1E-3 which, at a water flowrate in the SG of 94.5 kg/s (from results shown in Table 4 and Table 6) leads to p_w = 9.45E-7 1/s. Steinwartz reports that this value could be by a factor 10 higher, in particular during the start-up phase of the steam circuit. The efficiency of the water purification system can be assumed as 95% (η_w = 0.95).

2.6 C_{SG}: Characteristic SG constant

Having quantified the allowable tritium permeation through the SG in the previous step, we can now determine the tritium concentration (or more precisely the equivalent tritium partial pressure) in the primary He coolant. (The term "equivalent tritium partial pressure" is used here to describe the theoretical T_2 partial pressure resulting from a given number of tritium atoms in a gas. In practice most of the tritium is bound to hydrogen atoms in the form of HT molecules. In this case the equivalent tritium partial pressure would be half of the HT partial pressure.) This is necessary to complete the tritium balance in the helium circuit. It requires the design of a suitable SG (detailed in section 3) coupled to a tritium permeation calculation (described in section 4). More general details on tritium transport are given in section 8 (Annex 1).

With the information available for HTR-PM (thermal power, helium and water/steam temperatures and pressures) we have designed a SG and obtained the heat exchange surface and its temperature profile. We have done the same for higher helium temperatures. With this information we can calculate the tritium permeation as a function of equivalent tritium partial pressure on the helium side. Because geometry and thermal conditions are fixed, we can simplify then the permeation calculation in a way which directly relates the tritium flux to the partial pressure. As the allowable tritium permeation flux through the SG was already determined in the previous step, the corresponding equivalent tritium partial pressure can be calculated.

$$P = C_{SG} \frac{1}{PRF} \sqrt{p_T}$$

The characteristic SG constant C_{SG} combines all geometrical, material and temperature characteristics in a single factor. The PRF is a permeation reduction factor which summarizes the inhibiting effects of He gas chemistry (in particular the ratio H_2/HT) and formation of oxide permeation barriers on the SG tubes.

In later sections, details of the SG design are given and the tritium permeation is calculated. From this we can come up with a C_{SG}

- for HTR-PM conditions:

$$C_{SG}(HTR-PM) = 1.90388 \times 10^{18} \left[\frac{at}{s\sqrt{Pa}}\right]$$

- and for the higher temperature design HTR-PM+ conditions:

$$C_{SG}(HTR-PM+) = 1.96100 \times 10^{18} \left[\frac{at}{s\sqrt{Pa}}\right]$$

Then, the admissible equivalent tritium partial pressure in the helium becomes:

$$p_{Tmax} = \left(\frac{P_{max} \times PRF}{C_{SG}}\right)^2$$

2.7 L_{He}: Tritium escape from helium leakage

Tritium losses due to helium leakage can be calculated as:

$$L_{He} = -c_{He} V_{He} l_{He}$$

The tritium concentration in helium can be calculated from the maximum allowable equivalent partial pressure determined earlier from the allowable permeation, with the primary helium coolant pressure pr_{He} = 7 MPa:

$$c_{He} = \frac{p_T \times 6.022E23 \times 2}{pr_{He} \times 22.4E-3} \left[\frac{at\ T}{m^3 He}\right]$$

Concerning the helium inventory, a typical value is 1 t He/100 MWth. For HTR-PM one can find values in the literature between 2.44 t [37] (13664 m^3 (STP)) and 3 t [36] (16800 m^3 (STP)) the latter value being taken here as the reference value.

Helium leakage is dominated by leakage from the fuel handling system. In the literature one can find widely scattered values between one third of the inventory per year [37] (l_{He} = 1.057E-8 1/s) and 1% of the inventory per day (l_{He} = 1.1574E-7 1/s), the latter being used here as the reference only because it is the current design value for HTR-PM.

2.8 D_{He}: Tritium decay in helium circuit

The amount of tritium decaying in the helium circuit per unit time depends on the total helium volume and on the tritium concentration in this volume. The decay constant for tritium is λ = 1.7797E-9 1/s.

$$D_{He} = -\lambda V_{He} c_{He}$$

2.9 R_{He}: Tritium removed by helium purification

The amount of tritium that has to be extracted is calculated as:

$$R_{He} = -c_{He}\,\eta_{He} V_{He} p_{He}$$

The efficiency of a helium purification system is typically $\eta_{He} > 0.95$.

The helium purification rate p_{He} was determined from [36] where it reads that in HTR-PM 5%/h of the He inventory (150 kg/h) will be diverged to purification: p_{He} = 1.3889E-5 1/s.

Compared to the main helium flow of 346.9 t/h this corresponds to a slip stream fraction of 4E-4. In the AVR this purification rate was 8.3E-6 1/s, in the THTR 1.67E-5 1/s and in DRAGON 1.39E-4 1/s [3], the same as foreseen for HTR-PM. For fusion blankets, a typical value would be 1E-3 1/s due to the deliberately higher ratio tritium production/thermal power.

3 Steam Generator Design

The parameter study on tritium permeation is based on the design principle of the helical coil tube bundle SG of the Fort Saint Vrain HTR (FSV) in Colorado, USA. FSV was a 842 MWth/330 MWe prismatic block type HTR operated between 1974 and 1989. In 1997, it was the first reactor decommissioned in the USA. The SG design is described in detail in [9]. The FSV SGs were placed below the reactor core. Both in the reactor core and in the SGs the He flow direction was downward, so that a counter-current upward water flow in the steam generators could be achieved.

While FSV suffered from a number of teething problems, one of these problems (He circulator and seals leaking bearing water into the primary circuit) actually led to the discovery that controlled oxidation of the steam generator surfaces by humidity exerts an inhibitive effect on tritium permeation.

On the primary side, He flows inside the SG from top to bottom outside the tube banks. On its way the He passes the helical tubes of reheater (RH), super-heater (SH), evaporator (EVAP) and economiser (ECO). The cold helium exits at the bottom of the SG and flows into the suction chambers of the He circulators.

On the secondary side, feedwater is pumped from the bottom to the top through the inside of a fixed number of helical coil tube banks in each exchanger unit. The water flows through the ECO (preheating of water to the boiling point), EVAP (gradual evaporation to dry steam), SH (superheating of dry steam) before leaving the steam generator on the top. In these sections, different heat transfer correlations apply. The small bundle on the top of the SG, the RH reheat section, is not considered here, because it is probably absent in HTR-PM.

FSV used two SG loops with 6 SG modules per loop thus in total 12 SG. The tube bundles were made of concentrically wound multi-start helical coils. The design was such that individual tube helix lengths were very similar thus achieving uniform power uptake, circuit performance and compact design.

By selecting helix angle and longitudinal pitch spacing, it was possible to vary the total number of tube starts and thus the pressure drop (in total 3.47 psi (0.239 bar) on the He side, 590 psi (40.7 bar) on the steam side [10]). No external tube roughing or fins were necessary because the heat fluxes in all parts of the SG could be maintained between 50 and 100 kBtu/hr-ft^2 (158 – 315 kW/m^2). Allowances for fabrication tolerances, uncertainties (e.g. in the used heat transfer correlations) and performance degradation (e.g. by fouling or tube plugging for leak repair) were factored into the design. The material selected for the tubing was Incoloy 800, a well-known Ni-base alloy. A typical outer tube diameter for this type of SG is 25-27 mm as can be derived from [11].

The design principles for the FSV SG were applied by CEA in the 1990s to develop the computer tool He2SG (for Helium Heated Steam Generator) [8]. He2SG had been used for similar tritium permeation studies releated to thermonuclear fusion blankets [6] and was employed here courtesy of CEA to obtain approximate values for heat exchange surface area and for its temperature profile which are decisive parameters for tritium permeation. Note that a SG designer would increase the SG surface typically by 15-20% to account for design uncertainties and to cover operational effects such as fouling and plugging of tubes in case of water leakage. This design margin is not accounted for in this study.

Unfortunately, we were unable to find good illustrations of the FSV SG. But the design principle can very well be seen on the THTR-300 SG (Figure 2). Here, 80 tubes with an outer diameter of 25 mm are divided over 15 tube banks of different diameters leading to a structure of 15 multistart coils inserted into each other. This SG transferred 127 MWth through a heat exchange surface of 923.5 m^2 but, compared to HTR-PM, with somewhat different conditions (helium inlet/outlet temperature 667/293°C, helium pressure 3.81 MPa, water/steam inlet/outlet temperature 180/554°C, steam outlet pressure 19.6 MPa, pinch point temperature difference approx. 90 K) [41].

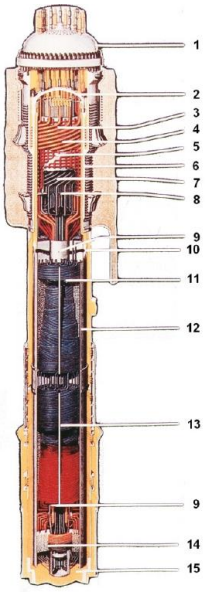

Fig. 2 Steam Generator Cutaway View

1: Outer Closure, 2: Inner Closure, 3: RH Steam Outlet, 4: Expansion compensation zone, 5: HP Steam Outlet, 6: PCRV Penetration, 7: RH Steam Inlet, 8: Feed-Water Inlet, 9: Central Tube, 10: Helium Outlet, 11: HP-I-Bundle, 12: Outer Shroud, 13: HP-II-Bundle, 14: RH Bundle, 15: Helium Inlet

Figure 2: THTR SG layout, pictures from [41]

HTR-PM has to extract about twice the power of a THTR-300 SG in a single SG module. With the FSV design this would have led to a large number of concentric coils, a construction which is difficult to manufacture. HTR-PM has opted for an array of 19 identical smaller multistart tube bundles as shown in Figure 3.

Despite these design differences, we believe that the He2SG tool produces a rather reliable estimate of heat exchange area and temperature profile required for this study.

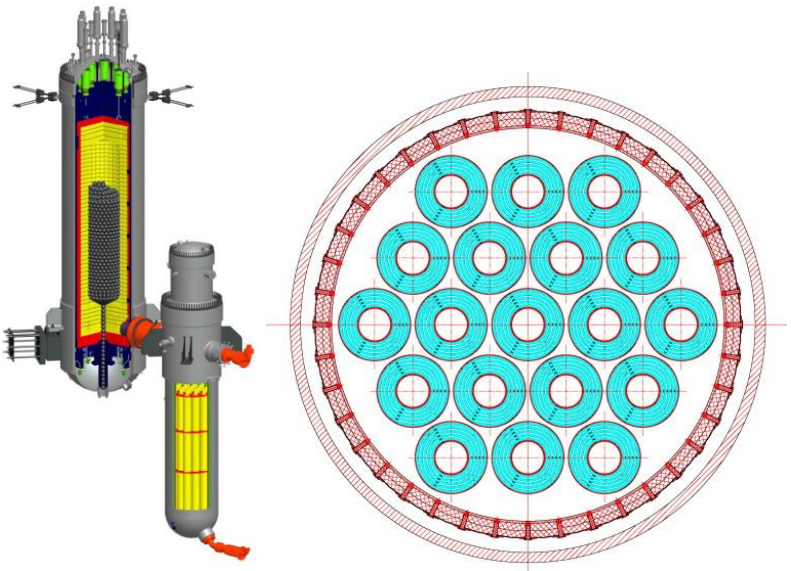

Figure 3: HTR-PM SG layout as currently envisaged by INET [12], [40]

3.1 Steam Generator characteristics for HTR-PM conditions

The present study is based on the operating conditions of HTR-PM [12] where two individual reactor modules (250 MWth each) are equipped with one individual SG each. The two SG deliver live steam to a common turbine generator set.

Parameter	Value
Thermal Power	250 MWth
Electric power	105.5 MWe
Inlet/outlet He temperature	750/250°C
He pressure	7 MPa
Inlet/outlet water/steam temperature	205/567°C
Live steam pressure	13.25 MPa
Steam flow-rate per SG	336.5 t/h

Table 3: Thermal-hydraulic conditions for a single module of HTR-PM [12]

As details on the HTR-PM SG design are not publicly available, we have generated a tentative SG design of the FSV type using He2SG for the HTR-PM operating conditions. The number of tubes was adjusted such that the pressure drops on the primary and secondary side remained within the limits of FSV (see above). The number of iterations was increased until a ± 5% error criterion was fulfilled for the heat exchange surface, which was the case for 90 axial tube sections.

The following results were obtained:

Parameter	HTR-PM Values	He2SG Input	He2SG Results
Thermal power	250 MWth	250 MWth	
Inlet/outlet He temperature	750/250°C	750/250°C	751*/250°C
He pressure	7 MPa	7 MPa	
He flow rate			346.9 t/h
He side pressure drop			1.19%
Inlet/outlet water/steam temperature	205/566°C	205/566°C	
Live steam pressure	13.25 MPa		13.23 MPa*
Water side pressure drop			3.32%
Feedwater inlet pressure			13.69 MPa
Water/steam flow-rate	336.5 t/h		340.1 t/h
Outer tube diameter		18 mm	
Tube thickness			2.10 mm
Number of tubes			444
Longitudinal pitch (longitudinal distance between tube centers divided by outer tube diameter)		1.16	
Radial pitch (radial distance between tube centers divided by outer tube diameter)		1.45	
Helix angle		5°	
Pinch point temperature difference			43.5 K
Total heat exchange surface			1618.1 m^2

Table 4: Tentative steam generator layout for HTR-PM operating conditions
(*: He2SG recalculates these values)

In Figure 4 the resulting temperature profile is plotted for He, wall and water along the heat exchange surface with the pinch point temperature difference at the beginning of the evaporator stage.

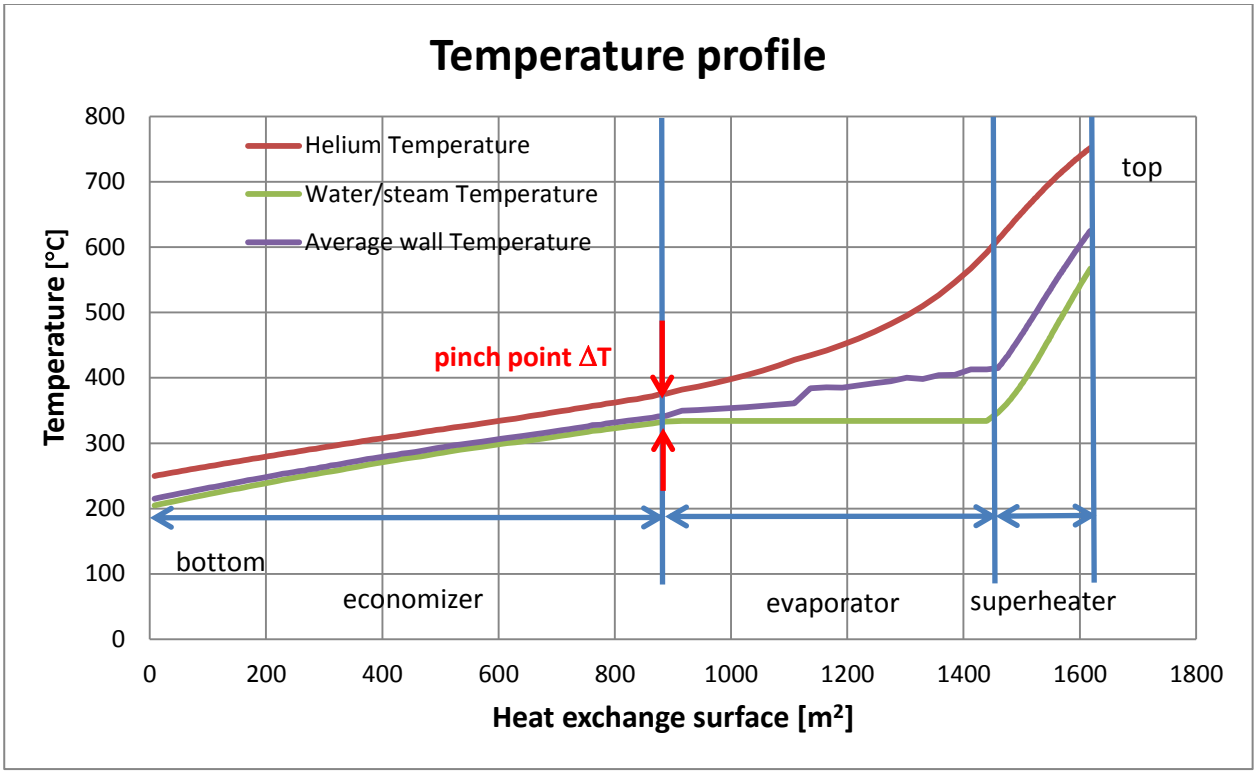

Figure 4: Temperature profile of He, wall and water along heat exchange surface

Note that He2SG uses the local heat transfer coefficients to calculate for each of the 90 tube sections the temperatures of He, wall and water which were assumed constant. This T vs. A plot together with the tube thickness is now used to perform the tritium permeation calculation described in section 4.

Comments on HTR-PM operating conditions:

From our calculations it seems that the HTR-PM Steam Generator and operating conditions should be defined in more detail because with our calculations we obtain a very low pinch point temperature difference thus leading to unnecessarily big and expensive components. The pinch point ΔT is the smallest temperature difference between primary and secondary coolant. In a SG, this occurs at the end of the economizer section.

From a pure SG optimization point of view we would also recommend to somewhat reduce the water pressure to shorten the economizer section, although we acknowledge that in this case a standard turbogenerator set as for coal-fired power plants can no longer be used and would have to be modified. Another possibility would be to somewhat increase the helium outlet temperature from the SG (currently 250°C), but this would require a proportionally higher helium throughput.

3.2 Steam Generator characteristics for higher temperatures

To better appreciate the effect of temperature on tritium permeation, we have raised the He inlet temperature from 750 to 950°C with all other parameters constant and obtained a different SG design with different heat exchange surfaces and temperature profiles. We call this case HTR-PM+.

Parameter	Value
Thermal Power	250 MWth
Electric power	105.5 MWe
Inlet/outlet He temperature	950/250°C
He pressure	7 MPa
Inlet/outlet water/steam temperature	205/567°C
Live steam pressure	13.25 MPa
Steam flow-rate per SG	336.5 t/h

Table 5: Thermal-hydraulic conditions for HTR-PM+ steam generator

Due to the higher helium temperature we expect a smaller heat exchange surface but at the expense of higher wall temperature and thus higher tritium permeation. This was to be expected because the heat exchange surface scales in first instance linearly with temperature but permeation scales exponentially.

The following results were obtained:

Parameter	HTR-PM Values	He2SG Input	He2SG Results
Thermal power	250 MWth	250 MWth	
Inlet/outlet He temperature	950/250°C	950/250°C	962*/250°C
He pressure	7 MPa	7 MPa	
He flow rate			247.8 t/h
He side pressure drop			0.47%
Inlet/outlet water/steam temperature	205/567°C	205/567°C	
Live steam pressure	13.24 MPa		13.24 MPa*
Water side pressure drop			2.33%
Feedwater inlet pressure			13.37 MPa
Water/Steam flow-rate	336.5 t/h		340.1 t/h
Outer tube diameter		18 mm	
Tube thickness			2.10 mm
Number of tubes			444
Longitudinal pitch (longitudinal distance between tube centers divided by outer tube diameter)			1.16
Radial pitch (radial distance between tube centers divided by outer tube diameter)			1.45
Helix angle			5°
Pinch point temperature difference			93.1 K
Total heat exchange surface			1148.8

Table 6: Tentative steam generator layout for HTR-PM+ operating conditions
(*: He2SG recalculates these values)

In Figure 5 the resulting temperature profile is plotted for He, wall and water along the heat exchange surface. Here, the pinch point temperature difference of 93 K appears acceptable.

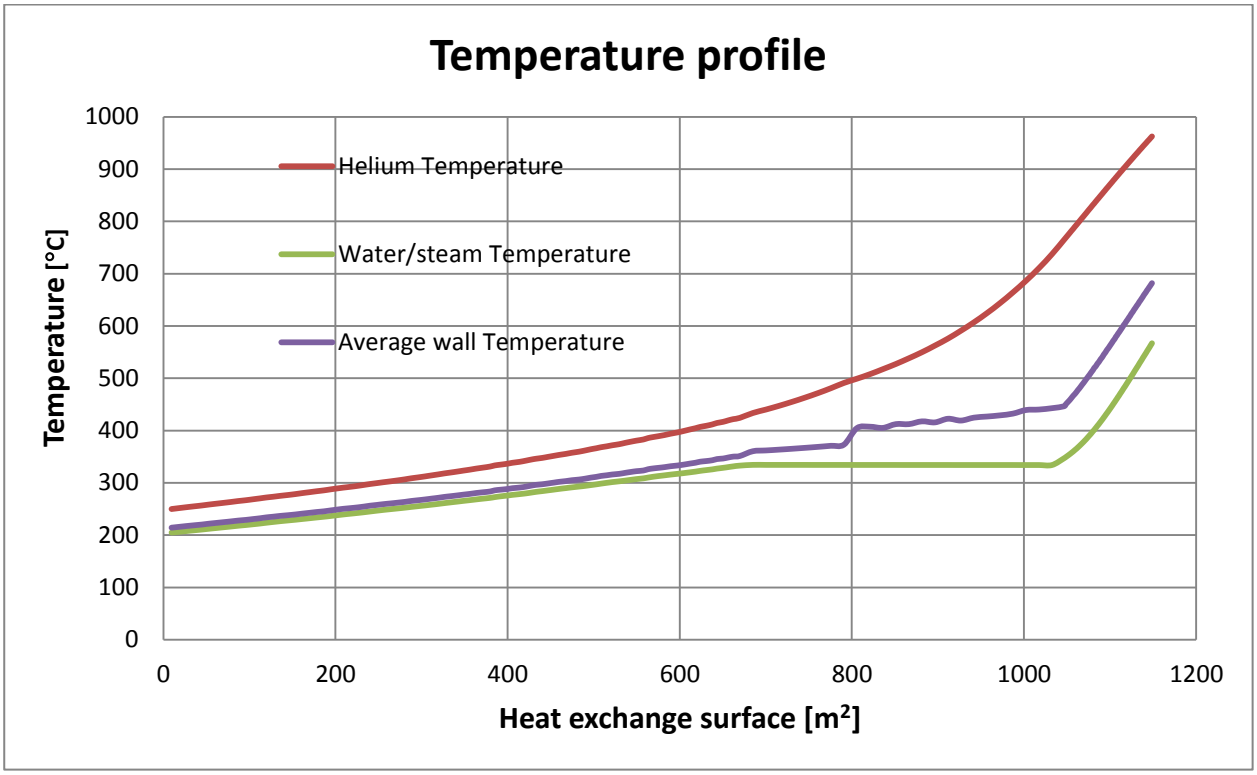

Figure 5: Temperature profile of He, wall and water along heat exchange surface for HTR-PM+ operating conditions

4 Tritium Permeation Calculation through Steam Generator

The temperature profile along the heat exchange surface in the SG obtained in Section 3 enables the calculation of tritium permeation for a tubular geometry:

$$P = \frac{2\pi l}{\ln(b/a)} \Phi(T)\left(p_T^n - p_w^n\right)$$

where:

P is the tritium permeation rate [at s^{-1}];
l is the tube length [m];
b is the outer tube diameter [m];
a is the inner tube diameter [m];
$\Phi(T)$ is the permeability [at m^{-1} s^{-1} Pa^{-n}]; it is usually expressed in the form of an Arrhenius function

$$\Phi = C \exp\left(-\frac{E_a}{RT}\right)$$

With:

C a characteristic constant for each material [at m^{-1} s^{-1} Pa^{-n}];
E_a the activation energy for the process [J/mol];
R the gas constant [J/(mol K)];
T the temperature in [K];

p_T is the equivalent tritium partial pressure in He [Pa];
p_w is the equivalent tritium partial pressure in water (≈ 0) [Pa];
n is the power of pressure dependence ($0.5 \leq n \leq 1.0$).

The power *n* of the pressure dependence must be determined experimentally. It ranges between 0.5 when the rate-determining process is pure bulk diffusion of atomic tritium, and 1.0 when only surface processes are rate-determining. The equivalent tritium partial pressure in He, p_T, depends on the contamination of the coolant, tritium extraction from the coolant and on the coolant chemistry.

The calculation is conservatively based on the assumption of constant equivalent tritium partial pressure on the He side (high He flow velocity), whereas the downstream tritium partial pressure in the water/steam was assumed to be nil. It was further assumed that the wall temperature was constant in each mesh element and that the radial temperature gradient in the wall had no slowing effect on tritium permeation due to thermal diffusion effects.

The tritium permeability Φ of the tubes depends on the choice and the surface state of the SG material, as well as on the temperature. Data for a variety of classical SG materials are available (a selection is given in [13] - [24]). A discussion of the results can be found in [6] and deals with materials, suitable oxidation conditions, power dependence of permeability and self-healing of oxide coatings formed in-situ. Here, only an element of particular importance in the current context is repeated:

Between 1976 and 1980 Bell and coworkers published a number of articles ([16] - [20]) which focused on the development of suitable oxidation conditions for Incoloy 800. In order to determine the activation energy for the permeability, Bell varied the test temperatures only by approximately ± 50 K in order to preserve the chemical composition of the surface oxide. He found identical activation energies for clean and oxidized Incoloy 800 of the order of 67.5 kJ mol^{-1}. This indicates that the oxide barrier created was partly defective and works by blocking the adsorption sites on the surface rather than by the oxide's own low permeability. This could also explain why the pressure dependence for Incoloy 800 oxides remained 0.5-power dependent even at very low tritium partial pressures (<< 1 Pa) whereas other authors found 1.0-power dependence for ideal oxides with no defects. The tritium permeabilities were measured in $He/H_2/T_2/HT$ mixtures and related to the square root of the residual molecular T_2 pressure which was calculated from

thermodynamic equilibrium data. From [17] we can determine the tritium permeability for clean Incoloy 800 as

$$\Phi_{clean} = 4.3924 \times 10^{17} \exp\left(-\frac{67491}{RT}\right)\left[\frac{at}{m s\sqrt{Pa}}\right]$$

and for pre-oxidized Incoloy 800 with defects (barrier factor 400) as $\Phi_{ox}=\Phi_{clean}/400$.

The 0.5-power dependence of the permeability leads to conservative results in tritium permeation calculations, but corresponds more realistically to SG operating conditions (defects in oxide scales) and is therefore applied in this work.

We conclude that Incoloy 800 is a well-established SG material with the potential to form efficient self-healing tritium permeation barriers by pre-oxidation. Therefore, we adopted Incoloy 800 as a reference SG material for this study reflecting the choice of several SG manufacturers for HTR in the past and for the Chinese HTR-PM reactor.

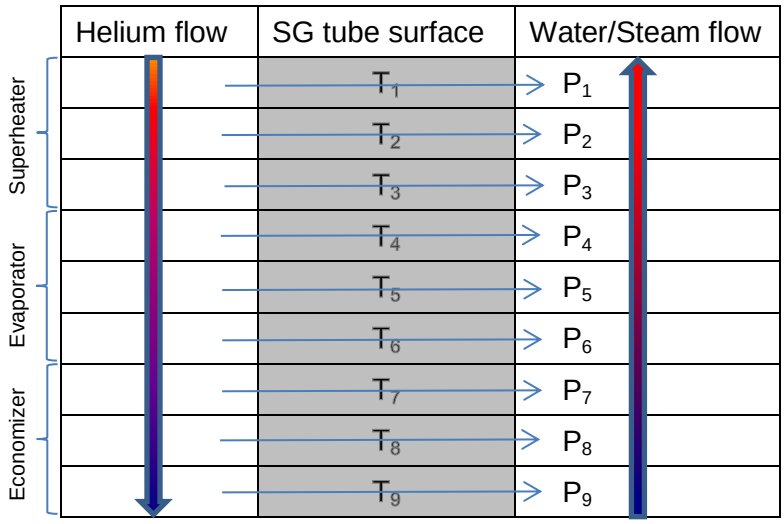

Figure 6: Schematic of 1-d FEM calculation of tritium permeation, for simplicity's sake only 9 mesh elements are shown here.
The sum of the individual mesh permeations yields the total permeation: $P = \sum_{i=1}^{n} P_i$

4.1 C_{SG}: Determination of characteristic SG constants

Based on the SG designs for HTR-PM conditions and for the higher temperature version HTR-PM+ we have calculated the tritium permeation for clean (PRF=1) and oxidized SG surfaces (PRF=400).

By collapsing the geometrical, thermal and resulting permeability characteristics of this SG we can calculate a characteristic SG constant C_{SG} according to:

$$P = \frac{2\pi l}{\ln(b/a)}\Phi(T)\left(p_T^n - p_w^n\right) = C_{SG}\sqrt{p_T}$$

For HTR-PM conditions, we obtained:

$$C_{SG}(HTR-PM) = 1.90388 \times 10^{18}\left[\frac{at}{s\sqrt{Pa}}\right]$$

and for the higher temperature design HTR-PM+ conditions:

$$C_{SG}(HTR-PM+) = 1.96100 \times 10^{18}\left[\frac{at}{s\sqrt{Pa}}\right]$$

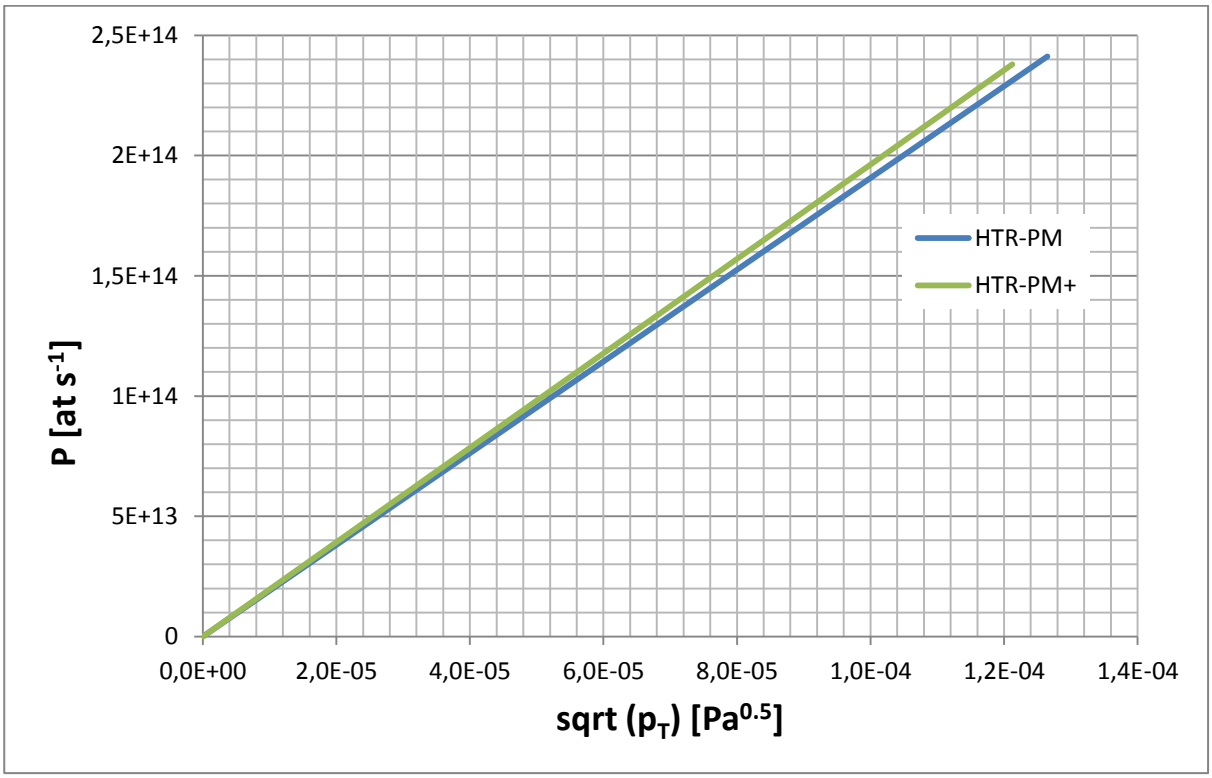

Figure 7: Tritium permeation vs. the square root of equivalent tritium partial pressure in helium for HTR-PM and HTR-PM+ conditions with clean surface (PRF=1)

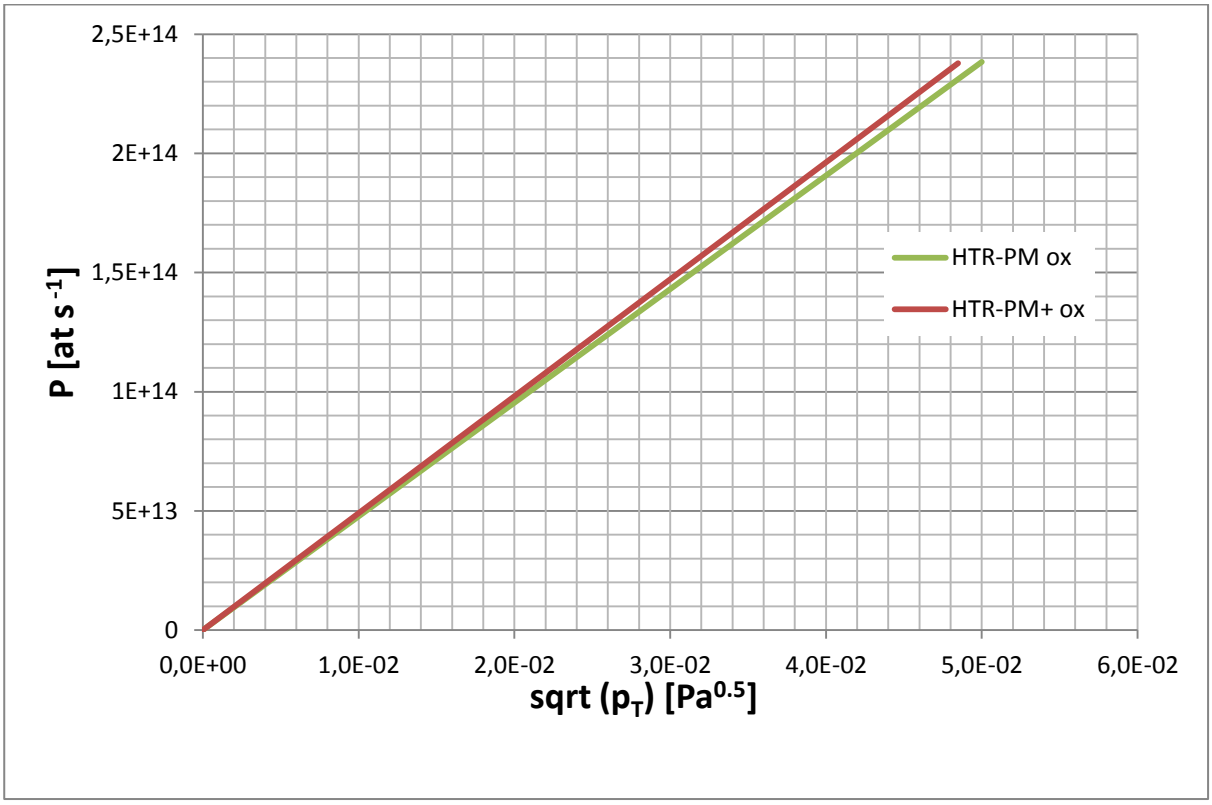

Figure 8: Tritium permeation vs. the square root of equivalent tritium partial pressure in helium for HTR-PM and HTR-PM+ conditions with oxidized surface (PRF=400)

5 Results and Discussion

The results presented here apply mainly to an HTR with a closed steam cycle for electricity generation. Considerations on an open steam cycle are given in section 0. Using the mass balances and "nominal" input data, the applicability of which we have discussed in the earlier sections, as an initial reference we have set up a simple spread sheet and compared the tritium source term to the tritium fluxes leaving the system. Then we have modified various parameters such that the tritium balance is matched. In the following plots, the nominal conditions from the tables below are marked with a red box.

Nominal input data for HTR-PM conditions:

T source term released to He	S	[at/s]	7.5850E+14	1150 Ci/yr
T concentration in water circuit	c_w	[at/m^3]	7.9040E+17	
characteristic SG constant	C_{SG}	[at/(s Pa$^{1/2}$)]	1.9039E+18	
tritium decay constant	l	[1/s]	1.7797E-09	
water volume	V_w	[m^3]	100	
water leak rate	l_w	[1/s]	1E-09	8.64 l/d
water purge rate	p_w	[1/s]	9.45E-07	slip stream fraction 1E-3
efficiency of water purification	h_w	[-]	0.95	
He volume (STP)	V_{He}	[m^3]	1.68E+04	3.0 t
He leak rate	l_{He}	[1/s]	1.1574E-07	1%/day
He purge rate	p_{He}	[1/s]	1.3889E-05	
efficiency of He purification	h_{He}	[-]	0.95	
He pressure	pr_{He}	[Pa]	7E+06	

Nominal output data:

T escape from water leakage	L_w	[at/s]	7.9040E+10	
T decay in water circuit	D_w	[at/s]	1.4067E+11	
T removed through water purging	R_w	[at/s]	7.0958E+13	
T permeation through SG	P	[at/s]	7.1178E+13	
T partial pressure in He	p_T	[Pa]	4.0000E-04	
T concentration in He	c_{He}	[at/m^3]	3.0725E+15	
T decay in He circuit	D_{HE}	[at/s]	9.1864E+10	
T escape from He leakage	L_{He}	[at/s]	5.9742E+12	
T removed through He purification	R_{He}	[at/s]	6.8107E+14	
sum of T fluxes leaving the system		[at/s]	7.5832E+14	
mass balance check (target is 1)		[-]	1.0002E+00	use goal seek to vary any parameter until this value is 1
required PRF		**[-]**	**535**	
T inventory in He	I_{He}	[at]	5.1618E+19	
T inventory in He	I_{He}	[Ci]	2.48	
T inventory in water	I_w	[at]	7.9040E+19	
T inventory in water	I_w	[Ci]	3.80	

Using the nominal conditions mentioned here, a PRF = 535 would be required. The equivalent tritium partial pressure in helium would become of the order of 4E-4 Pa. With typical hydrogen swamping (cf. explanation in section 8.3.4), the overall pressure of hydrogen species in helium would be of the order of 100 Pa thus fully justifying a diffusion-based tritium permeation approach as used here.

In these conditions, 9.4% of the tritium source term would permeate into the secondary cooling circuit. Because the leakage and decay terms are relatively small, the remaining approx. 90% must be captured by the helium purification system.

The tritium inventory in helium would be < 2.5 Ci and the inventory in the cooling water < 4 Ci.

5.1 Permeation Reduction Factor PRF vs. helium purge rate

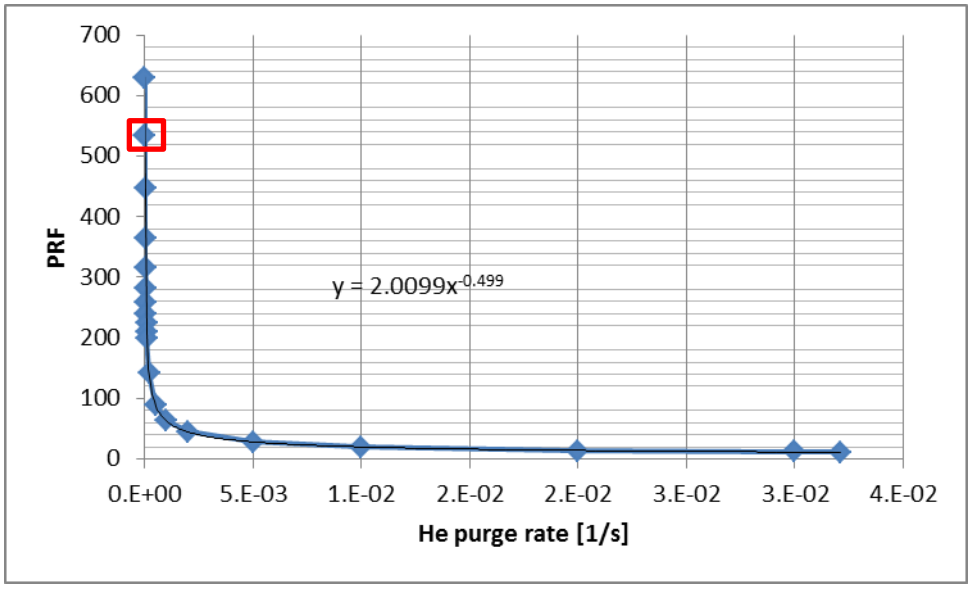

Figure 9: Required Permeation Reduction Factor PRF vs. helium purge rate

Figure 9 shows the relationship between required PRF and the He purge rate.

We can make 4 observations:

- with the nominal purge rate, the required PRF is as high as 535
- even if the entire He flow through the SG was purged (which is completely unrealistic) the required PRF would still be 11
- a realistic value for PRF=400 can be achieved with a He purge rate of 2.5E-5 [1/s] which is about twice as high as the reference value for HTR-PM (1.3889E-05 1/s).
- the proportionality is of the type $PRF \approx \frac{1}{\sqrt{p_{He}}}$

5.2 Permeation Reduction Factor PRF vs. water purge rate

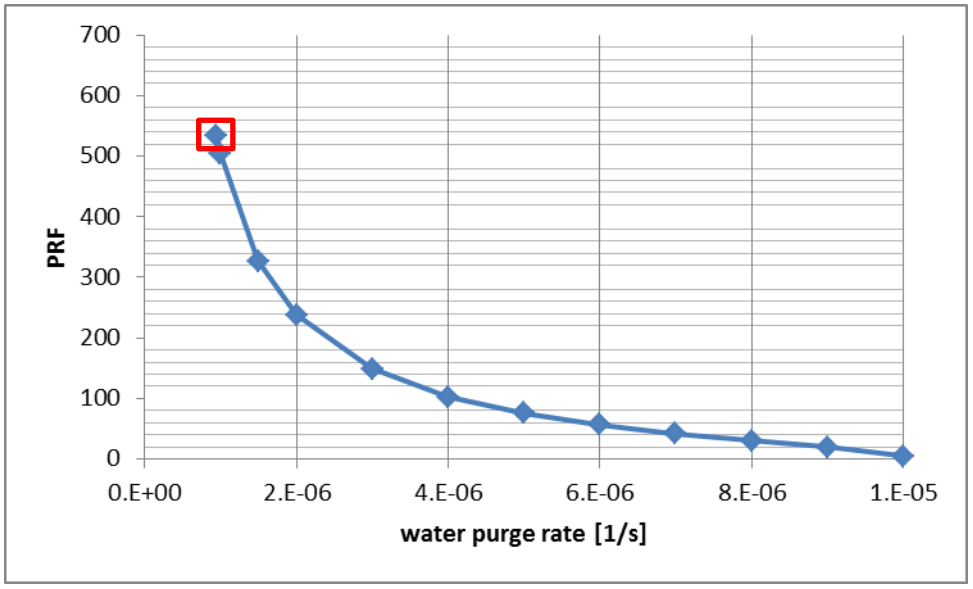

Figure 10: Required Permeation Reduction Factor PRF vs. water purge rate

5.3 Permeation Reduction Factor PRF vs. water leak rate

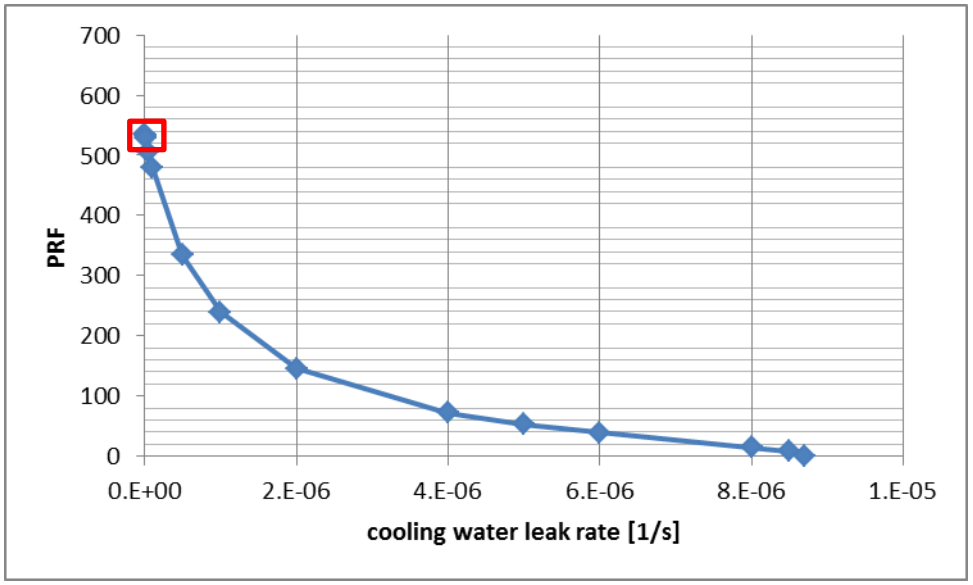

Figure 11: Required Permeation Reduction Factor PRF vs. water leak rate

5.4 Permeation Reduction Factor PRF vs. allowable tritium concentration in secondary water circuit

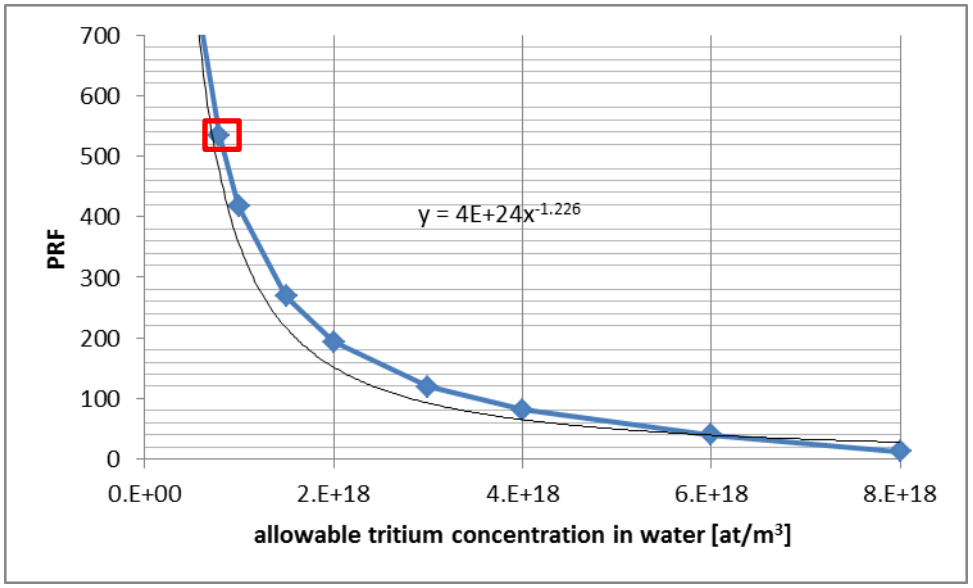

Figure 12: Required Permeation Reduction Factor PRF vs. allowable tritium concentration in secondary water circuit

As expected, the effort on tritium management expressed in PRF is strongly depending on the allowable tritium concentration in the secondary water. For the development of a solid tritium control strategy, it is thus necessary to clarify this question with the concerned licensing authority.

5.5 Permeation Reduction Factor PRF vs. tritium source term

Steinwartz [3] stated that a significant amount of the tritium generated in an HTR is actually not released into the helium stream due to retention in the coated fuel particles (our reference value is 10% release of the tritium generated in the fuel) and due to recoil and sorption on graphite with chemical fixation on the graphite. This could reduce the apparent source term by a factor 5-10 and thus the requirements for tritium control measures as shown in Figure 13.

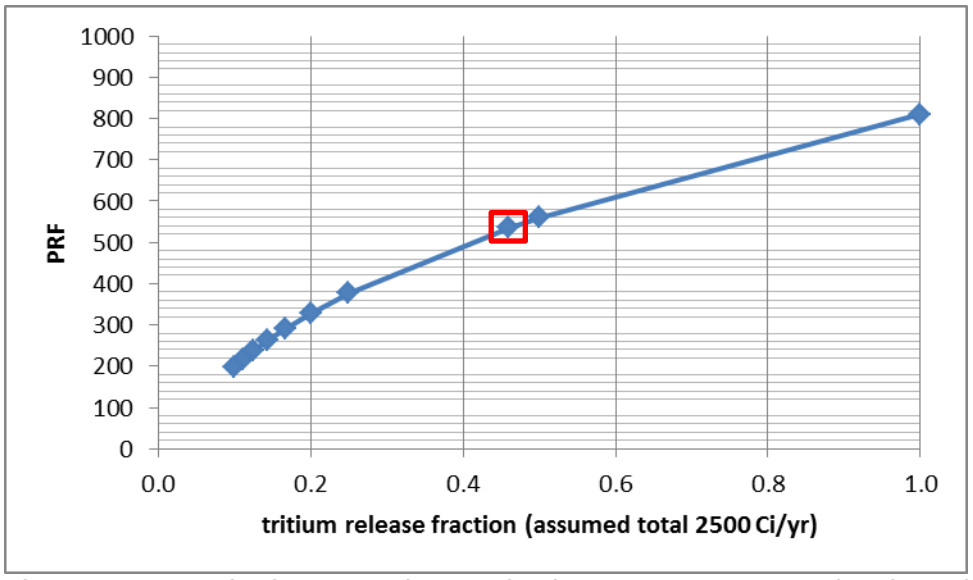

Figure 13: Required Permeation Reduction Factor PRF vs. fraction of total tritium generation (assumed 2500 Ci/yr)

However, the uncertainty is such that sorption and recoil were so far not used in the licensing process but only given as a justification of the conservatism of the assumed tritium release. Indeed, it is very difficult to model this effect reliably because it depends on time-dependent surface conditions of the graphite, temperature profiles, graphite type and other factors. Tritium release from the fuel may also be better than the assumed 10% (cf. section 2.1).

5.6 Combination of parameter changes

When combining reasonable changes to the parameters, one can estimate which additional effort has to be made in other areas. For instance, by doubling the helium purge rate, by doubling the allowable tritium concentration in the cooling water (e.g. by improving turbine hall ventilation) and by doubling the assumed water purification rate), the additionally required PRF drops to 79.

If in addition we assume a technologically more realistic water/steam leak rate of 1E-6 instead of 1E-9, the required PRF drops to a value as low as 41, which can be achieved by moderate hydrogen swamping of the primary helium with a $p_{HT}/p_{H2} = 10^{-3}$ (a typical value used in the AVR is $p_{HT}/p_{H2} = 10^{-4}$) not accounting for any other effects.

5.7 Consequences for tritium release

Our calculations corroborate the practically confirmed experience that the major pathways for tritium release are the helium and water purification systems of an HTR, whereas leakage is relatively insignificant and not easily controlled either. The effluents from the helium and water purification systems are mainly tritiated water (HTO) and hydrocarbons. Depending on the licensing limits of the power plant, these effluents can either be diluted and released into the environment or stored for decay.

In Europe, there are no generic tritium release limits for nuclear power plants. Such limits are power plant specific and encompass tritium in the form of gas or tritiated water, but those depend strongly on national legislation. As an example [25] the French Cattenom power plant (4×1300 MWe ≈ 15758 MWth) may release 10,000 GBq/yr of tritiated gas and 140,000 GBq/yr of tritiated water. Scaling down with power to HTR-PM conditions (which in itself is already a debatable approach because licensing limits may be rather site-specific than power-specific), this would lead to an allowable release of tritiated water of 2221 GBq/yr (= 3.96E13 at/s = 60 Ci/yr).

To illustrate the bandwidth of possible limits and for comparison, the license limit for the German THTR (760 MWth) was 1000 Ci/yr, so that power scaling to HTR-PM conditions would lead to a release limit of 330 Ci/yr. For LWR, Steinwartz [3] also mentions a reasonable experience-based limit of 200 Ci/yr-100 MWe, which, scaled with electric power, would yield a "European limit" for an HTR-PM type reactor of 211 Ci/yr.

All these tritium release limits are significantly lower than the source term assumed here (2500 Ci/yr) which may mean that in certain conditions effluents (e.g. from the helium purification plant) may need to be stored for decay. However, considering that most operated HTR plants used only a fraction of their licensed tritium release limits, we must indeed assume that much of the produced tritium is actually captured inside the reactor, mainly in the graphite. While this is beneficial in terms of chronic tritium release, the accumulation effect has to be accounted for during thermal transients and in the radioactivity source term for severe accident analysis.

5.8 Process steam generation

If using an HTR for process steam generation, the contamination of the process product (either by heat exchange or by participation in the reaction) must be limited. The main objective is, therefore, to keep the tritium contamination of the process steam low by minimizing the tritium permeation through the SG.

For the HTR Modul a tritium contamination limit of process steam of 5 kBq/kg (= 2.81E15 at/m^3) was assumed on the basis of an allowable end-user exposure. Postulating a similar SG rating, design and operating conditions as for a closed cycle (steam flow rate of 94.5 kg/s), the allowable tritium permeation through the SG would become 2.66E14 at/s.

To determine the operational requirements to achieve this target, the mass balance from section 2 which we have used in previous sections can be simplified to the helium system alone:

$$S = P\left(\sqrt{p_T}\right) + L_{He}\left(\sqrt{p_T}\right) + D_{He}\left(\sqrt{p_T}\right) + R_{He}\left(\sqrt{p_T}\right)$$

From the allowable tritium concentration in the process steam one can calculate the allowable equivalent tritium partial pressure in helium (with a PRF as a parameter) and further the leak, decay and purification terms (with the purification rate as a parameter).

Input data for HTR-PM conditions:

T source term	S	[at/s]	7.585E+14	1150 Ci/yr
T concentration in process steam	c_w	[at/m^3]	2.8100E+15	
characteristic SG constant	C_{SG}	[at/(s Pa$^{1/2}$)]	1.9039E+18	
tritium decay constant	l	[1/s]	1.7797E-09	
process steam flow rate	V_{PS}	[kg/s]	9.4500E+01	
He volume (STP)	V_{He}	[m^3]	1.68E+04	3.0 t
He leak rate	l_{He}	[1/s]	1.1574E-07	1%/day of inventory
He purge rate	p_{He}	[1/s]	1.3889E-05	
efficiency of He purification	h_{He}	[-]	9.5000E-01	
He pressure	phelium	[Pa]	7.0000E+06	

Output data:

T permeation through SG	P	[at/s]	2.6555E+14	
T partial pressure in He	p_T	[Pa]	2.8684E-04	
T concentration in He	c_{He}	[at/m^3]	2.2032E+15	
T decay in He circuit	D_{HE}	[at/s]	6.5874E+10	
T escape from He leakage	L_{He}	[at/s]	4.2840E+12	
T removed through He purification	R_{He}	[at/s]	4.8838E+14	
sum of T fluxes leaving the system		[at/s]	7.5828E+14	
mass balance check (target is 1)		[-]	1.0003E+00	use goal seek to vary PRF until this value is 1
required PRF		[-]	121	
T inventory in He	I_{He}	[at]	3.7014E+19	
T inventory in He	I_{He}	[Ci]	1.78	

One immediately realizes that compared to a system with a closed steam cycle for electricity generation, the required tritium control effort, here expressed in PRF, is actually somewhat lower for an open steam cycle for process heat generation.

This PRF can quite easily be achieved by hydrogen swamping on the helium side and by in-situ formation of oxide coatings on the SG tubes. The helium purification system should be designed such that the purge rate is flexible enough to cope with uncertainties and transients.

Again, conservatively, this analysis does not account for the beneficial effects of tritium sorption and recoil effects which we know are significant but difficult to quantify.

6 Conclusions and Recommendations

Based on tritium mass balances, using a steam generator design tool and making justified initial assumptions based on HTR-PM operating conditions as an example, we have developed a method to study the interrelationship between different operating parameters and their effect on tritium control requirements. The tool is flexible and can be adapted to any other operating conditions. We have shown the individual impact of these parameters and given a few examples for what we think could be sensible combinations of them.

For a closed steam cycle (electricity generation) we assumed that for radioprotection reasons the acceptable tritium concentration in the secondary cooling water is the decisive boundary condition. From that we found several combinations of operating parameters which would allow meeting plausible tritium control requirements even without postulating any permeation barrier on the SG tubes, which will rather easily form in situ given the usual impurities in helium.

For an open steam cycle (process steam generation) the contamination of the process steam is the decisive boundary condition, which determines the allowable tritium permeation rate. Although the allowable tritium contamination is very low, this condition is actually even more easily met than for a closed steam cycle.

If in addition we consider the sometimes coarse conservatism of our assumptions (e.g. neglecting tritium sorption/recoil in graphite which are difficult to quantify and were thus never used in the licensing process), then we may conclude that a viable tritium control strategy is actually not problematic to achieve. However, plant designers should build in some flexibility in operating conditions to allow compensating for the various uncertainties.

Obviously, the elaboration of a more refined tritium control strategy for HTR requires the knowledge of the licensing conditions for new-build HTR, in particular the allowed chronic and accidental tritium release. As these limits affect system design, they should be known and approved well before the final design stage of a reactor. Any changes later on may necessitate design modifications with a very detrimental effect on timing and cost, as it was experienced during the construction of the THTR. Once those limits are known, the methodology presented in this report can be used to define the effort to be made by the different systems which influence the effectiveness of tritium control. Those are in particular helium and water purification, allowable water/steam leak rate, helium chemistry and permeation barriers on the SG tube walls.

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Annexes

Annex 1 – Tritium transport through metals and permeation barriers

Annex 2 – Document approval by beneficiaries' internal QA

8 Annex 1: Tritium Transport through Metals and Permeation Barriers

Metals and permeation barriers can interfere with tritium transport in two different ways: In the following subsections, the basic transfer equations for surface- (1) or diffusion-limited (2) permeation are outlined. A mixed regime of both surface and diffusion limited transfer has also been observed which is often connected to the tritium partial pressure in the system.

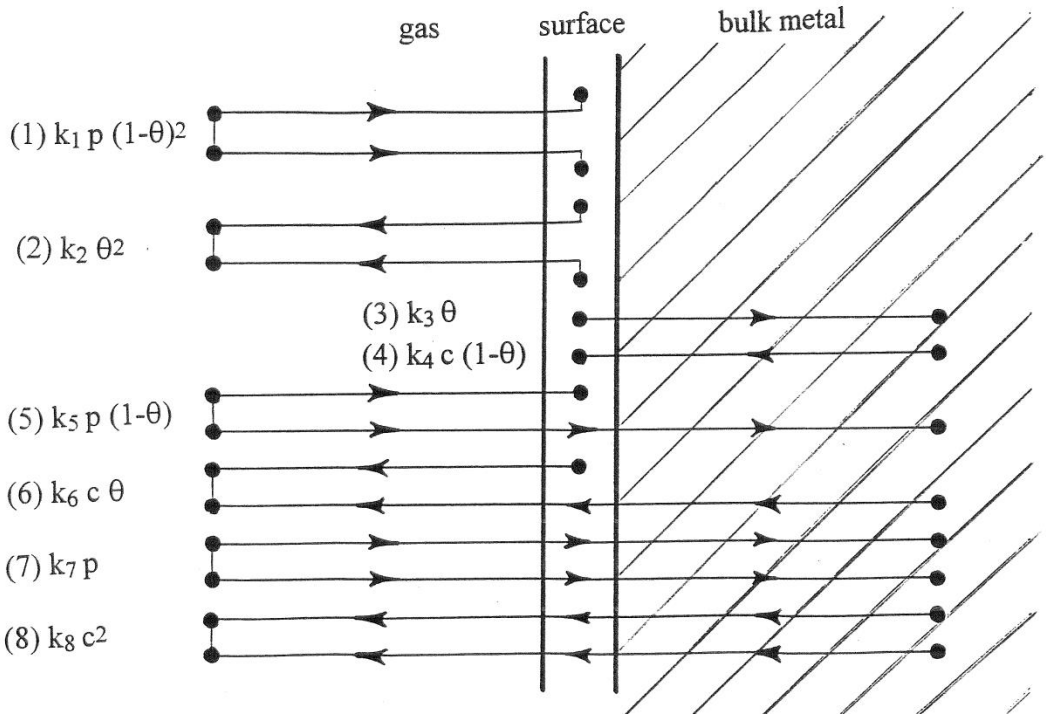

Figure 14: Hydrogen interaction processes with metals according to [31]

8.1 Permeation limited by surface reactions

Richards [31] investigated the basic processes on a metal/gas interface, he discussed the findings of Baskes [32] and Pick [33] and gave expressions for the fluxes which are associated to them. These processes are adsorption, desorption, jumping into solution and back from solution into the surface (cf. Figure 14). The kinetic constants k_i are temperature dependent. Surface processes are slow (i.e. rate determining) at low hydrogen concentrations in the metal and low pressures in the gas phase. For example, for AISI 304 or 316 type stainless steel these processes are important between 300°C and 600°C and at pressures lower than 100 Pa. It is theoretically and experimentally proved that hydrogen enters a metal in atomic form. This means that a dissociation or recombination process with its associated electron transfer is needed every time a hydrogen atom enters or leaves a metal lattice. In Figure 14, c is the subsurface hydrogen concentration, p is the pressure of molecular H_2 gas and θ is the surface coverage which is defined as the ratio of occupied to available adsorption sites. When all adsorption sites are occupied and $\theta=1$, the processes (1)-(4), and (5) become unimportant. Because the remaining processes are all quite fast, the transfer rate will then be limited by diffusion in the bulk material.

In the case of surface limited transfer, diffusion in the bulk is quick enough to instantaneously cancel any spatial concentration gradient. Therefore, the processes (1, dissociative adsorption) and (2, recombinative adsorption) are the most important and the hydrogen flux can be calculated from a flow balance:

$$J_s = A\left[k_1 p\left(1-\theta\right)^2\right) - k_2\theta^2\right]$$

where:

J_s is the net hydrogen flux from medium 1 to medium 2 [at s^{-1}]

k_1 is the dissociation constant [at m^{-2} $s^{-1}Pa^{-1}$]

k_2 is the recombination constant [at m^{-2} s^{-1}]

θ is the surface coverage $0 < \theta < 1$

p is the driving H_2 partial pressure [Pa]

A is the surface area [m^2]

When the surface coverage is very small at low pressures and concentrations (θ<<1) this equation simplifies to:

$$J_s = A\left[k_1 p - k_2 c^2\right]$$

where:

c is the H subsurface atomic concentration [atfrac]

Note that in equilibrium ($J_s = 0$) Sievert's law holds which relates hydrogen concentration to partial pressure:

$$c_{eq} = K_s\sqrt{p}$$

where:

$K_s = \sqrt{\frac{k_1}{k_2}}\left[\frac{atfrac}{\sqrt{Pa}}\right]$ which is the Sievert's constant

8.2 Permeation limited by diffusion

When the pressure at the interface is sufficiently high so that all surface sites are always occupied, (usually at a total hydrogen pressure, including tritium > 1 Pa, thus covering the conditions of an HTR) bulk processes, which are usually slower than surface reactions, will be the rate determining step. The involved processes are interstitial and grain boundary diffusion due to spatial concentration gradients, thermodiffusion due to temperature gradients, trapping at defects, and chemical trapping by hydride formation. The relative importance of these processes depends on the individual bulk material.

In metals for which trapping or hydride formation is unimportant (e.g. Fe, steels with relatively low thickness), tritium transfer at higher partial pressures is governed by diffusion. In this diffusion limited regime we can measure that the hydrogen permeation through a metal is proportional to the square root of the partial pressure difference between the upstream and the downstream side. The equation below is the solution of Fick's law for a slab and is often referred to as Richardson's equation:

$$J_d = \frac{A}{t} D K_s \left(\sqrt{p_1} - \sqrt{p_2} \right)$$

where:

J_d is the net diffusive flux from medium 1 to medium 2 [at s^{-1}]

A is the exchange surface area between the media [m^2]

t is the thickness of the permeated medium [m]

D is the diffusivity of hydrogen in the permeated material [m^2 s^{-1}]

K_s is the Sievert's constant of hydrogen in the permeated material [at m^{-3} $Pa^{-0.5}$]

$\Phi = D K_s$, often called permeability [at m^{-1} s^{-1} $Pa^{-0.5}$]

p_1 is the upstream hydrogen partial pressure [Pa]

p_2 is the downstream hydrogen partial pressure [Pa]

These equations are used to determine the permeability of materials by imposing a hydrogen pressure on the upstream side of a membrane while measuring the resulting hydrogen flow rate through the membrane or the pressure rise on the downstream side.

8.3 Effect of Hydrogen on Tritium Transfer

A rather complete phenomenological description is given in [39].

8.3.1 Isotopic exchange reactions, increase of surface recombination

When a purge gas sweeps a metallic surface (liquid or solid) and desorbs tritium, the addition of hydrogen to the purge gas can increase the tritium desorption rate. This happens because a tritium atom arriving on a surface adsorption site needs a recombination partner to leave the metal and hop into the gas phase. The probability to find such a partner is higher when molecular hydrogen in the gas phase is present and ready for an isotopic exchange with a tritium atom. Of course, an increase in the desorption rate can only be achieved when the overall tritium transfer through the metal is limited by surface recombination and not by diffusion through the metal or a surface oxide layer.

Fast isotopic exchange can also occur between tritium atoms on an adsoption site and water molecules thus forming an HTO molecule. For the specific case of a SG, surface recombination steps on the water side can quite safely be ruled out as the rate determining step.

8.3.2 Surface modification

The increase of tritium desorption from a surface by hydrogen doping of the purge gas was also observed when tritium transfer was diffusion limited, which happened at high tritium concentrations. This phenomenon must be related to a reaction of the hydrogen with existing surface impurities. Surface oxides, for instance, which usually represent a permeation barrier, may be reduced and thus tritium transfer enhanced. If one is interested in slowing down tritium permeation, then the adjustment not only of a hydrogen concentration must be considered but also the adjustment of an oxidation potential. This can be done by suitable mixtures of H_2 and H_2O. It is also known that gas impurities containing carbon (e.g. CO, CO_2, CH_4) block surface adsorption sites for hydrogen.

8.3.3 Hydrogen counter pressure

It was speculated in the past that another way of reducing tritium transfer could be by imposing a counter-current hydrogen flow. However, as diffusion is a statistical process and as shown by

experimental evidence [34], [35], [38], the simultaneous counter-current diffusion of hydrogen and tritium does not affect tritium transfer, at least at very low pressures. At high hydrogen counter pressures, tritium permeation would become proportional to p_{HT} (not to its square root), i.e. surface effects become again rate-determining, and would decrease proportionally to $\frac{1}{\sqrt[4]{p_{H2}}}$.

However, hydrogen diffusion into the He can lead to hydrogen swamping on the He side (see section 8.3.4).

8.3.4 "Isotopic swamping"

The presence of additional hydrogen on the upstream side can change the statistics of diffusive tritium transfer. When the upstream side (here the primary He side of the SG) is "swamped" with hydrogen, all hydrogen species compete for available adsorption sites on the surface. Thus, the probability for tritium adsorption (with consecutive permeation) is expected to be reduced when hydrogen prevails. This assumption implies that the permeation rates of different hydrogen isotopes cannot be considered independent of each other and simply be added. It is argued that an adsorbing metal surface cannot distinguish different hydrogen isotopes and regards them as chemically identical. Already low hydrogen concentrations in the gas phase (approx. $p_{HT}/p_{H2} = 10^{-5}$ in the AVR [3]) would cause tritium permeation to drop significantly.

To quantify this effect, and as explained in [3] one can write the equilibrium law for hydrogen and tritium as

$$\frac{1}{2}H_2 + \frac{1}{2}T_2 \leftrightarrow HT$$

with the equilibrium constant

$$K_{eq}^2 = \frac{p_{H2} \times p_{T2}}{p_{HT}^2}$$

or

$$p_{T2} = \frac{K_{eq}^2 \times p_{HT}^2}{p_{H2}}$$

That means that in the tritium permeation equation we have to use now the term

$$\frac{K_{eq}^2 \times p_{HT}^2}{p_{H2}}$$

as the driving partial pressure.

If we assume a ratio $p_{HT}/p_{H2} = 10^{-5}$ as in the AVR [3] and knowing that in typical HTR conditions the equilibrium constant is only weakly temperature dependent and equal to 0.5, we can estimate the effect of the hydrogen swamping on tritium permeation assuming the same number of tritium atoms in the gas phase. Note that here we make use again of the "equivalent tritium partial pressure" of pure T_2 molecules (which would be half the pressure of HT molecules) and compare it to a mixture of HT and H_2 molecules:

The ratio between the square roots of the driving partial pressures without and with hydrogen swamping can be expressed as an apparent permeation reduction factor:

$$\frac{\sqrt{p_{T2}}}{\sqrt{\frac{K_{eq}^2 \times p_{HT}^2}{p_{H2}}}} = \frac{\sqrt{p_{T2}}}{K_{eq}\sqrt{p_{HT}\frac{p_{HT}}{p_{H2}}}} = \frac{\sqrt{p_{T2}}}{K_{eq}\sqrt{2p_{T2}\frac{p_{HT}}{p_{H2}}}} = \frac{1}{K_{eq}\sqrt{2\frac{p_{HT}}{p_{H2}}}} = \frac{1}{0.5\sqrt{2\times 10^{-5}}} = 447$$

In other words, the presence of 10E5 times more H_2 than HT would reduce tritium permeation by a factor 447.

While this proportionality was demonstrated in the AVR, the absolute benefit is more difficult to quantify, because there is no AVR data available for comparison without H_2 and with pure T_2.

Other authors [38] on the contrary argue with dissociation constants and find for typical material properties and geometries that tritium permeation through bare metal is only weakly dependent on hydrogen swamping. Assuming a perfect oxide barrier on the upstream side and assuming that the barrier works as a dissociation inhibitor rather than a diffusion inhibitor, the authors found that then the permeation reduction factor is no longer influenced by hydrogen swamping.

Waelbroek [39] states that co-streaming of hydrogen in the same direction as tritium (as it is the case with "isotopic swamping") is more efficient than counter-streaming. The resulting permeation reduction factor would be proportional to $\sim\sqrt{p_{H2}}$ (in accordance with [3]) instead of $\sim\sqrt[4]{p_{H2}}$.

8.3.5 Presence of humidity or hydrocarbons in helium circuit

In normal operation of an HTR, other chemical species are present in the primary helium gas, for instance small quantities of tritiated methane (CH_3T, approx. 1-20% of H_2, decreasing with temperature), tritiated water (HTO, approx. 0.5% of H_2) and small concentrations of CO and CO_2 [3]. These molecules must first be broken up by a chemical reaction with the surface, before the containing hydrogen atoms can permeate into the SG wall, thus slowing down tritium transfer. In particular small amounts of humidity also favour formation of self-healing oxide layers on the SG surface. This effect is also difficult to quantify and depends very much on the operating conditions, but it is clearly contributing to a reduction of tritium permeation.

Unit conversions for tritium:

1 Ci = 2.08×10^{19} atoms = 3.7×10^{10} Bq = 1.036×10^{-4} g

1 g = 9650 Ci

λ: decay constant = $\ln2/T_{1/2}$ (with $T_{1/2}$ = 12.35 yr) = 1.7797×10^{-9} [1/s]

9 Annex 2: Document approval by beneficiaries' internal QA

Fill involved beneficiaries as appropriate (mandatory for Milestones and Deliverables, but optional for other document type)

#	Name of beneficiary	Approved by	Function	Date
1	JRC	Kamil Tuček	Reviewer	
2	JRC	Karl-Fredrik Nilsson	Action Leader	
3	JRC	Peter Hähner	Head of Unit	
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