

Is Tritium an Issue for High Temperature Reactors?

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Abstract – In a High Temperature Reactor, tritium is produced by a number of mechanisms. 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. The remaining 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 escape into the environment.

This paper summarizes a study on the different tritium control options capable of meeting possible future safety requirements. 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.

I. 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, ³He 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 was examined 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 thermonuclear fusion [6], [7].

For the purpose of this study we have used the operation characteristics of a power plant similar to the Chinese HTR-PM design [8] 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 [7] and used courtesy of

CEA for the ARCHER project. He2SG was also used for the analysis published in [9].

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.

This paper 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.

II. 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.

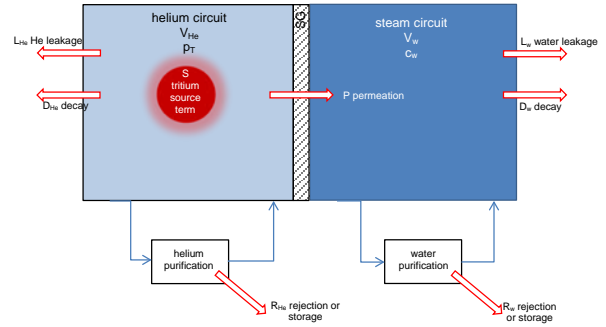


Figure 1: Simplified model of tritium fluxes in an HTR system coupled to a SG

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.

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

- for the overall system:

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

for the water circuit:

$$P = L_w(c_w) + D_w(c_w) + R_w(c_w)$$

- for the SG:

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

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

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; some of the tritium is also bound in CH_3T (tritiated methane) 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:

$$C_{SG} = \frac{2\pi l}{\ln b/a} \Phi(T)$$

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

Table 1: Process steps to determine tritium flux balance

| | 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 $C_{SG} = \frac{2\pi l}{\ln b/a} \Phi(T)$ | 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 | | |

II.A. 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) [10] [11], and by the interaction of neutrons with lithium, boron, and helium impurities in the surrounding core graphite and helium cooling fluid [12]. Table 2 (data from [12]) 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 3He in a 3000 MWth HTGR compared to a 1500 MWth HTGR.

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.

Table 2: Measured tritium source terms for Peach Bottom and Fort St. Vrain extrapolated to other HTGRs [12] and assumptions used for this study

| | Peach Bottom (66.5 MWth) (measured) | Fort St. Vrain (842 MWth) 6 th 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 |

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.

⁶Li 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 ¹⁰B in control rods can to some extent be reduced by design while tritium production from ³He impurities in the coolant (today extracted from natural gas) can only be reduced by unrealistically expensive isotopic tailoring. The

ratio ³He/⁴He from natural gas typically ranges between 0.5 and 5E-6.

II.B. 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 [13] and the effective dose factor for ingestion of tritiated water is $e_{ing}(50) = 1.81E-11$ Sv Bq⁻¹ [13], [14]. 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 m³ 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/m³.

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 [13];
- inhaled air volume 2500 m³/yr
- allowable tritium concentration in air 4.9E-7 Ci/m³
- temperature in turbine hall 40°C
- humidity in turbine hall 100%
- water concentration in air of turbine hall 5.1E-5 t_{H2O}/m³_{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 [13], 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 $c_w = 7.904E17 \text{ at/m}^3 = 3.8E-2 \text{ Ci/t} = 1.406E6 \text{ Bq/l}$.

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 $\mu\text{Ci/m}^3$ (4.4 kBq/kg) as the HTR Modul limit (5 kBq/kg = 2.81E15 at/m³).

II.C. 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 $V_w = 100 \text{ m}^3$ and, initially, a leak rate of $l_w = 1E-9 \text{ 1/s}$, 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 $l_w = 4.725E-6 \text{ 1/s}$), 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.

II.D. 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 $\lambda = 1.7797E-9 \text{ 1/s}$.

$$D_w = -\lambda V_w c_w$$

II.E. 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 leads to $p_w = 9.45E-7 \text{ 1/s}$. 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% ($\eta_w = 0.95$).

II.F. 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 coupled to a tritium permeation calculation which is explained in more detail in [15].

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:

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

II.G. 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 $p_{r_{He}} = 7$ MPa:

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

For the helium inventory, a typical value is 1 t He/100 MWth. For HTR-PM one can find numbers in the literature between 2.44 t [16] (13664 m³ (STP)) and 3 t [17] (16800 m³ (STP)) the latter value being taken here as the reference.

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 [16] ($l_{He} = 1.057E-8$ 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.

II.H. 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 $\lambda = 1.7797E-9$ 1/s.

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

II.I. 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 [17] 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.

III. STEAM GENERATOR DESIGN

Because little information on the HTR-PM SG is available publicly, this study on tritium permeation is based on the design principles of the helical coil tube bundle SG of the Fort Saint Vrain HTR (FSV) in Colorado, USA. This SG design is described in detail in [18] and analysed in [19] and [20].

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) [7]. He2SG had been used for similar tritium permeation studies related to thermonuclear fusion blankets [8] 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 leakage. This margin is not accounted for in this study.

III.A. Steam Generator characteristics for HTR-PM conditions

The present study is based on the operating conditions of HTR-PM [8] 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.

Table 3: Thermal-hydraulic conditions for a single module of HTR-PM [8]

| 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 4: Tentative steam generator layout for HTR-PM operating conditions

| 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 |

(*: He2SG recalculates these values)

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 $\pm 5\%$ error criterion was fulfilled for the heat exchange surface, which was the case

for 90 axial tube sections. The results in Table 4 were obtained.

In Figure 2 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.

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 temperature vs. surface plot together with the tube geometry is now used to perform the tritium permeation calculation.

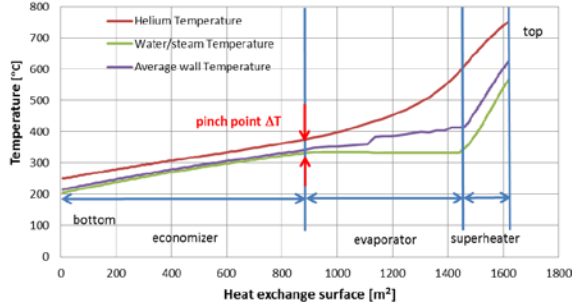


Figure 2: Temperature profile of He, wall and water along heat exchange surface

IV. TRITIUM PERMEATION THROUGH STEAM GENERATOR

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

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

with:

P : tritium permeation rate [at s⁻¹];

l : tube length [m];

b : outer tube diameter [m];

a : inner tube diameter [m];

$\Phi(T)$: 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 : characteristic material constant [at m⁻¹ s⁻¹ Pa⁻ⁿ];

E_a : activation energy for the process [J/mol];

R : gas constant [J/(mol K)];

T : temperature in [K];

p_T : equivalent tritium partial pressure in He [Pa];

p_w : equivalent tritium partial pressure in water (≈ 0) [Pa];

n : 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 [21] - [28]). A discussion of the results can be found in [8] 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 recalled:

Between 1974 and 1980, numerous results were published by several authors (e.g. [22]-[28]) focusing on the development of suitable oxidation conditions for SG materials, especially 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 worked 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 ($\ll 1$ Pa) whereas other authors found 1.0-power dependence for ideal oxides with no defects. The tritium permeabilities were measured in He/H₂/T₂/HT mixtures and related to the square root of the residual molecular T₂ pressure which was calculated from thermodynamic equilibrium data. From [24] 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.

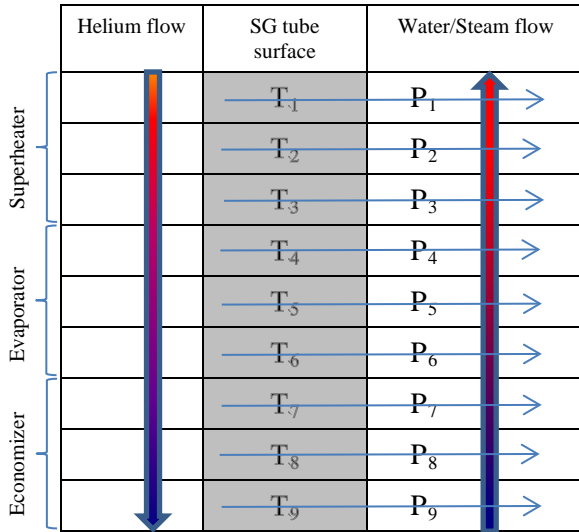


Figure 3: Schematic of 1-d FEM calculation of tritium permeation; for simplicity's sake only 9 mesh elements are shown here.

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 3 shows schematically the 1-d FEM calculation of tritium permeation (for simplicity's sake only 9 mesh elements are shown). The sum of the individual mesh permeations yields the total permeation: $P = \sum_{i=1}^n P_i$.

IV.A. C_{SG} : 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:

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

For HTR-PM conditions, we obtained:

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

V. RESULTS AND DISCUSSION

V.A. Closed steam cycle

The results presented in Table 5 and Table 6 apply to an HTR with a closed steam cycle for electricity generation. Using the mass balances and "nominal" input data, the applicability of which we have discussed above, as an initial reference, we have set up a simple spreadsheet 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.

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, 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.

Table 5: Nominal input data for HTR-PM conditions with closed steam cycle

| | | | | |
|----------------------------------|--------------|-----------------------------|------------|---------------------------|
| 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 | $p_{r_{He}}$ | [Pa] | 7E+06 | |

Table 6: Nominal output data for HTR-PM conditions with closed steam cycle

| | | | | |
|------------------------------------|----------|----------------------|------------|---|
| 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 | |

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.

In [15] the individual and combined effect of several parameters on the PRF is analysed and discussed.

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 $p_{HT}/p_{H_2} = 10-3$ (a typical value used in the AVR is $p_{HT}/p_{H_2} = 10-4$) not accounting for any other effects.

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. 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%.

Our analysis corroborates 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 [29] 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.

V.B. Process steam generation

If using an HTR for process steam generation without condensate return (i.e. when the steam is used as feedstock), 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/m³) 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 which we have used in previous sections for a closed secondary cycle can be simplified to the helium system alone:

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

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). The results are summarized in Table 7 and Table 8.

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.

Table 7: Nominal input data for HTR-PM for open process steam generation

| | | | | |
|----------------------------------|--------------|-----------------------------|------------|---------------------|
| 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 | p_{helium} | [Pa] | 7.0000E+06 | |

Table 8: Nominal output data for HTR-PM for open process steam generation

| | | | | |
|-------------------------------------|----------|----------------------|------------|--|
| 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 | |

VI. CONCLUSIONS

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 without condensate return) 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 study 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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