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Considerations on techniques for improving tritium confinement in helium-cooled ceramic breeder blankets

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Tritium control issues such as the development of permeation barriers and the choice of the coolant and purge-gas chemistry are of crucial importance for solid breeder blankets. In order to quantify these problems for the helium-cooled ceramic BIT blanket concept, the tritium leakage into the coolant was evaluated and the consequent tritium losses into the steam circuit were determined. Our results indicate that under certain specified conditions the total tritium release from the coolant can be limited to approximately 10 Ci/d, but only on the assumption that experimental data for tritium permeation barriers can be attained under realistic operating conditions. An experimental study on the impact of the gas chemistry on tritium losses is proposed.

1. INTRODUCTION

Within the framework of the European Test Blanket Program CEA and ENEA jointly undertake the development of a helium-cooled ceramic breeder blanket for DEMO. As in most realistic blanket concepts tritium losses from the breeder material into the coolant and eventually into the steam circuit are of major concern. In order to quantify the extent of the problem, the potential sources of tritium contamination of the coolant in normal and faulted operating conditions were evaluated together with an assessment of the tritium permeation through the steam generator. Our studies show that tritium confinement remains a problem even when state-of-the-art techniques such as permeation barriers are applied. This encouraged us to look for other solutions and to outline the related R&D.

2. BLANKET DESIGN

Even though tritium confinement problems are common to *all helium-cooled ceramic breeder* blankets, it is useful to recall some basic design features of the Breeder-Inside-Tube (BIT) concept [1]. Poloidal breeder modules (Fig. 1) are assembled in several rows and contained by a segment box. These modules comprise of bundles of breeder rods surrounded by beryllium blocks and contained in a pressure tube. The breeder rods are filled with a stack of annular pellets made of lithiated ceramics ($LiAlO_2$ or Li_2ZrO_3). All structures consist of martensitic steel.

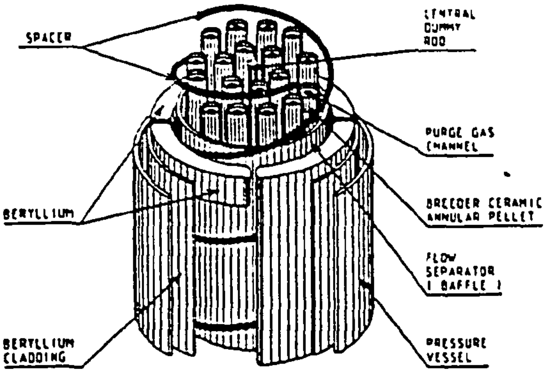

Figure 1: Poloidal breeder module for BIT concept

The annulus in the pellets defines a channel into which helium purge-gas is injected for tritium extraction with a pressure of 5.7 MPa. To improve tritium release from the ceramics, the purge-gas contains either 0.1% hydrogen or 10 ppm H_2O, while the total pressure of tritiated species at the blanket outlet is a few Pascals.

The predicted tritium production for this concept is of the order of 400 g/d.

The helium coolant flows through the first wall and then through the breeder modules at a pressure of approximately 6 MPa with a temperature of 300°C at the blanket inlet and 530°C at the outlet. The temperatures of the breeder rods range between 430°C and 550°C.

3. EVALUATION OF TRITIUM LOSSES

3.1 Tritium permeation into the coolant

Tritium contamination of the helium coolant will occur under normal operating conditions and was previously evaluated using specific computer tools [2]. The three main sources for the contamination were identified to be the first wall, the breeder rods and the beryllium cladding.

Depending on the surface state, steady-state permeation from the plasma through the first wall can range between 1 and as much as 100 g/d. Permeation of the generated tritium from the breeder rods can contribute with a minimum of 25 g/d depending on the efficiency of tritium recovery by hydrogen swamping and the tritium percolation through the pores of the breeder material. Finally, assuming that the tritium generated in the beryllium permeates directly into the coolant, we have to count on an additional 4 g/d. Conservatively, a total tritium contamination of almost 130 g/d must be taken into account, which underlines the significance of the problem.

3.2. Tritium permeation into the steam circuit

In order to define the specifications of reliable measures to improve tritium confinement, the tritium permeation from the helium coolant through the steam generator into the steam circuit was evaluated [3]. This included the definition of the steam generator operating conditions by thermodynamic cycle calculations, and its thermal-hydraulic design. The obtained geometry, surface area, and temperature profiles along the heat exchanger tubes were then used to estimate the daily tritium permeation into the steam circuit. Steam oxidised Incoloy 800 austenitic stainless steel was identified as the best suited existing material. It enabled, within the given operating conditions, to restrict the daily tritium losses to less than 10 Ci/d. The conditions for this are:

- that experimental data for the oxide barrier efficiency (permeation reduction factor 400) of steam preoxidised Incoloy 800 are applicable in the long run and in realistic steam generator operating conditions (thermal cycling, pressure transients, vibrations, adapted gas and water chemistry); a proof for this is not yet available;
- that the coolant chemistry and tritium extraction processes allow to keep the tritium activity in the helium coolant on a level corresponding to a tritium partial pressure of 3×10^{-3} Pa; various possibilities have been identified, but a substantial lack of knowledge persists concerning the reaction kinetics and their impact on tritium losses, inventory and recovery under the particular boundary conditions of a fusion reactor.

4. MEASURES AGAINST TRITIUM LOSSES

4.1. Permeation barriers

Permeation barriers are the result of a surface treatment and work either by reducing the number and/or availability of adsorption sites, by the reduction of the surface recombination constant, or by their own low permeability. Already natural oxide layers (passivation films) possess some barrier qualities, but the best overall results achieved in laboratory experiments stem in general from aluminisations [4].

Permeation barriers are still under development but do not yet meet the performance requirements, in particular under irradiation [5]. Substantial progress is still needed if permeation barriers should remain the only means for tritium control.

4.2. Gas chemistry optimisation

A parallel method for tritium control could consist in the application of a proper gas chemistry in the blanket. When using $LiAlO_2$ or Li_2ZrO_3 in the blanket, the composition of the purge-gas is practically fixed and defined by the requirements for tritium extraction from the lithium ceramics and must carry hydrogen and/or water vapor, yet it must be essentially free from oxygen. The helium coolant will probably also contain some water vapor from small steam generator leaks.

A number of experiments showed qualitatively that the choice of the gas chemistry could reduce tritium losses through at least four mechanisms:

1. "Isotopic swamping", which means the dilution of tritium with hydrogen in the gas phase. In the case of a breeder rod, isotopic swamping can occur on the inside of the rod (hydrogen addition to the purge gas) or on the outside (hydrogen addition to the helium coolant). When tritium and hydrogen are present in the purge gas (approx. 0.1% H_2 are recommended to facilitate tritium extraction from most ceramic breeders) they supposedly compete for the available adsorption sites on the breeder rod. This theoretically reduces the probability for a tritium atom of finding an adsorption site and to diffuse into the helium coolant. The permeation of tritium can be expressed as

$$J = \frac{A}{t} \Phi (p_1 - p_2)^n \tag{1}$$

with

J	tritium permeation rate [at/s]
A	exchange surface area [m^2]
t	thickness of permeated material [m]
Φ	permeability $\left[\frac{at}{m \cdot s \cdot Pa^n} \right]$
p_1	upstream tritium partial pressure [Pa]
p_2	downstream tritium partial pressure [Pa]
n	power of pressure dependence, $0.5 \leq n \leq 1$

By hydrogen addition, the equivalent tritium partial pressure p_1 (or in other words the chemical activity of tritium) is reduced according to

$$[p_1] = \frac{[p_{HT}]^2}{K \cdot [p_{H_2}]} \tag{2}$$

where K is the temperature dependent equilibrium constant for the reaction $H_2 + T_2 \leftrightarrow 2 \cdot HT$ (e.g. K = 3.45 at 600 K [6]). The partial pressure of HT, p_{HT}, reaches its theoretical upper limit when all T_2 molecules have reacted with H_2, i.e. for a given T_2 partial pressure of 1 Pa, the maximum attainable HT partial pressure would be 2 Pa. When at the same time the H_2 partial pressure is kept at 1000 Pa, the residual equivalent tritium partial pressure would be reduced to 1.16×10^{-3} Pa and the resulting tritium permeation would drop to 1/30 of the initial value when diffusion limited transport is assumed (n=0.5).

Isotopic swamping could also be applied on the coolant side to make hydrogen and tritium compete for adsorption sites on the steam generator.

It can, however, be argued that hydrogen addition will not exert any effect on tritium permeation because one has to consider the permeation of hydrogen and tritium separately. The authors are currently not aware of any experiment that could support either of these theories but the potential of this measure would certainly deserve an experimental verification. An excellent summary on tritium transport theory and some related experiments can be found in [7].

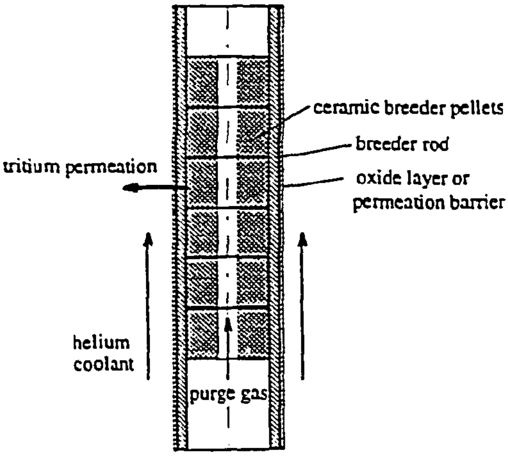

Figure 2: Sketch of a breeder rod

2. Reactions of certain purge-gas additives with the surface of the structural material could form a possibly self-healing permeation barrier. The addition of water vapor is a typical example and has proven beneficial against tritium permeation at least for certain steam generator materials [8]. The resulting passivation film is not only acting as a permeation barrier but also impedes the catalytic reduction of HTO to permeable HT. It is not clear so far if the same water vapor will also form an efficient permeation barrier on the structural material of the breeder rods (martensitic steel). Other substances but water should be tested as well. One should, however, keep in mind the possible consequences of the gas chemistry on the whole system, e.g. with respect to corrosion or tritium recovery.

3. Blockage of adsorption sites on the surface by contaminants can also reduce permeation. This effect has been observed e.g. with CO on Pd-Ag permeators as they are used in tritium processing units. It is not clear if the same effect can be reproduced on passivated stainless steel as it will be encountered on the breeder rods and in the steam generator.

4. The conversion of mobile gaseous tritium (HT or T_2) into non-permeable species like tritiated water in the presence of water vapor could be yet another way for permeation reduction. Water vapor will be present in the helium coolant in any case (small steam generator leaks) or has to be injected so as to maintain an efficient oxide barrier on the steam generator tubes. At the same time the permeated tritium can partly be desorbed as unpermeable tritiated water by isotopic exchange reactions, at least on low temperature surfaces where water can be chemisorbed. This would reduce the amount of permeable tritium in the coolant while it would increase the overall water vapor concentration and thus facilitate the extraction of HTO (by molecular sieves or cold traps). Two problems which are difficult to quantify must be mentioned in this context:

- The strong dilution of the HTO with H_2O might cause a waste water problem because the tritium concentration in the water traps of the coolant purification units will be too low to justify an economical tritium recovery.
- Even when isotopic exchange reactions are known to be rather fast, it seems questionable if they are (without catalysis) fast enough to convert most of the tritium to HTO in the short time available (approx. 1 second) before it reaches the steam generator. The oxidation of HT to HTO, e.g. by a copper oxide bed, is in principle feasible but, in return, would cause an unacceptable coolant pressure drop. Oxygen addition to the helium coolant for HTO formation is deemed little attractive because of the slow reaction kinetics while a catalyst would induce too high pressure drops.

5. CONCLUSION

Tritium permeation losses from ceramic breeder blankets to the environment still present a serious issue when available techniques such as permeation barriers are used alone. Supplemental measures might consist in the optimisation of the chemical composition of the gas phases, in particular of the helium coolant, which can have a strong impact onto permeation by various physical-chemical mechanisms. In principle all of them go in the right direction, i.e. they can qualitatively reduce tritium permeation into the coolant and could thus lower the requirements for permeation barriers. However, a quantitave evaluation of the impact of the mechanisms is missing so far and should be the objective of a more detailed experimental program.

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