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In-pile performance of a double-walled tube and a tritium permeation barrier

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Abstract

In two recent irradiation experiments in the HFR Petten, tritium permeation rates through representative materials to be used as cooling tubes of the water-cooled lithium-lead blanket have been measured in-pile. These latest experiments in the EXOTIC 8 series (E 8.9 and E 8.10) are made of a double wall tube (DWT) and a T91 tube with an Fe–Al/Al_2O_3 layer acting as tritium permeation barrier (TPB). These tubes contain annular pebble beds of ceramic breeder materials for the helium-cooled pebble bed concept blanket as tritium breeding material. Both experiments are built up of two concentric and independently purged containments allowing on-line tritium release rate and permeation rate measurements. In-pile operation has ended in March 2001 after 450 full power days and resulted in an irradiation damage of approximately 2.6 and 3.2 dpa, respectively in T91 steel. This paper reports on the experimental results obtained for in-pile tritium permeation and discusses the influence of purge gas compositions, temperature and irradiation on tritium permeation through the DWT and TPB.

1. Introduction

Within the European Blanket Project, a water-cooled lithium-lead (WCLL) concept and a helium-cooled pebble bed concept (HCPB) are being developed in parallel [1]. Tubes with tritium permeation barriers (TPBs) and a double wall tube (DWT) are being developed as structural materials for the cooling tubes for the WCLL concept [1]. One objective for the development of these tubes is to reduce tritium permeation into the water through the wall of the cooling tube, placed in a tritium generating eutectic mixture of lithium-lead. Because of the stage of development of other components of the WCLL, testing of these tubes was delayed until 2002 [2]. It was therefore decided to use the DWT and the tube with TPB in two new EXOTIC in-pile ceramic breeder tests for the HCPB [3], this allowed simultaneous testing of components of HCPB and WCLL in one experiment. Besides the in-pile performance of the tubes for tritium permeation, the influence of temperature and hydrogen content in the purge gases on the tritium permeation was also studied, which gave information about the tritium transport in the irradiation experiments. The end of these irradiation tests was scheduled for end of July 2000, but because of scientific reasons and promising results, it was decided to prolong these tests until March 2001. This led to a total irradiation time of 450 full power days (FPD) and a total irradiation damage of 2.6 and 3.2 dpa, respectively in T91 steel. The in-pile results of the first 100 FPD of these experiments were already published [4]. To account for trends in tritium permeation and to be conclusive on the in-pile part of these experiments, this paper presents all in-pile results and the results are compared with out-of-pile literature data.

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2. Experiment design and analysis

2.1. Experimental

The E 8.9 capsule consists of a DWT, inner diameter 11.1 mm, outer diameter 16.8 mm, tilled with Li_2TiO_3 pebbles, supplied by ENEA. The DWT functions as a primary containment for the ceramic breeder bed and is made at CEA Grenoble by hot isostatic pressure diffusion welding of two T91 steel tubes, inner tube thickness 1.15 mm, outer tube thickness 1.55 mm, with a copper interlayer, thickness 0.3 mm [5]. The horizontal cross-section of the DWT is shown in Fig. 1. Because of the lower permeability of hydrogen in copper this will act as a permeation barrier. Results on hydrogen/deuterium permeation in a DWT made of 1 mm F82H/0.1 mm Cu/ 1 mm F82H steel showed a reduced permeation of a factor 3 at a temperature of 723 K, compared to bare F82H with a thickness of 2.1 mm, as shown in Fig. 2. The permeability of F82H and T91 are the same because of the similar composition of the materials. The EXOTIC 8.10 capsule is designed as EXOTIC 8.9 except for the wall of the first containment; a T91 steel tube, inner diameter 14.8 mm, outer diameter 16.8 mm, with an external $FeAl/Al_2O_3$ coating of thickness 7 μm – the TPB – was used. This coating was made by the pack-cementation CVD process at CEA. Permeation measurements of a bare T91 steel and a CVD coated T91 steel sample resulted in a reduced permeation with a factor 5000 [6]. These results are also shown in Fig. 2. EXOTIC 8.10 contains Li_4SiO_4 pebbles produced by FZK as a breeder material. In the centre of the annular pebble beds in both experiments, stainless steel guide tubes are placed which contain the gas purge lines for the first and second containments. Temperature transients were performed by means of an electrical heater. By changing the ratio of helium and neon in the third containment, an extra temperature control is obtained for both experiments. The first and second containments are purged with helium gas doped with varying hydrogen concentrations to study the influence of hydrogen on the tritium permeation.

2.2. Tritium measurement

The gas lines are connected to the tritium measurement station, where identical ionisation chambers (IC) measure the tritium carried with the purge gas from the containments. In this way the percentage permeated tritium (PPT) of tritium produced in the first containment can be determined directly by:

$$\mathrm{PPT}(\%) = \frac{\mathrm{IC}_{2,\mathrm{signal}}}{\mathrm{IC}_{1,\mathrm{signal}} + \mathrm{IC}_{2,\mathrm{signal}}} \times 100\%, \tag{1}$$

where both IC signals are corrected for the influence of different hydrogen concentrations on the response of the IC (Jesse effect, [7]) and deviations in flow rate and

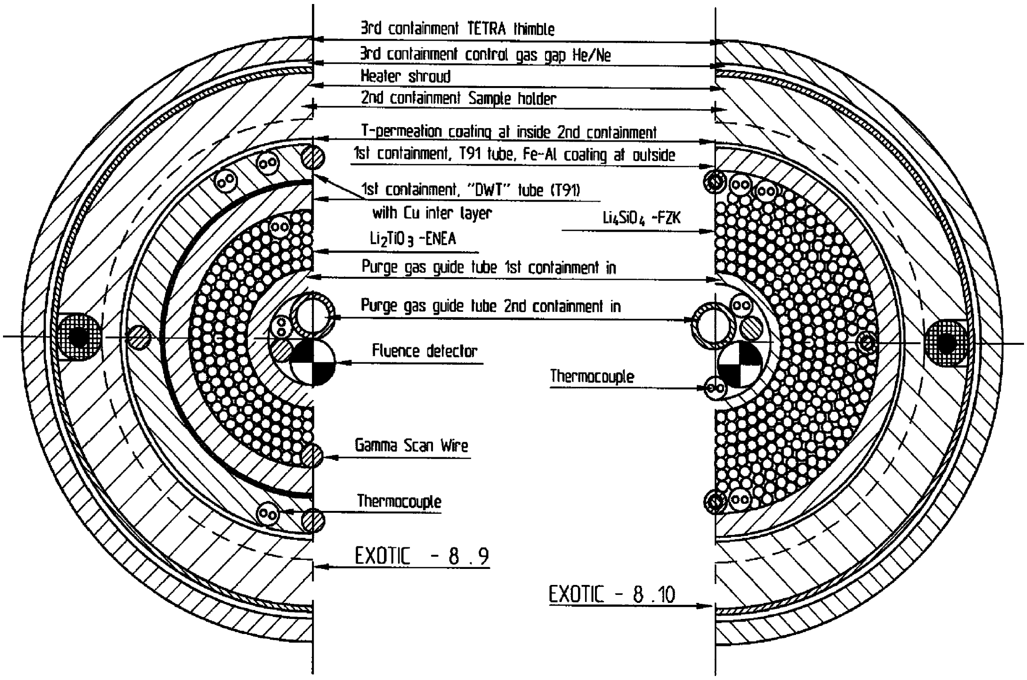

Fig. 1. Cross-section of EXOTIC 8/9 with DWT (left) and EXOTIC 8/10 with Fe–Al/Al_2O_3 TPB.

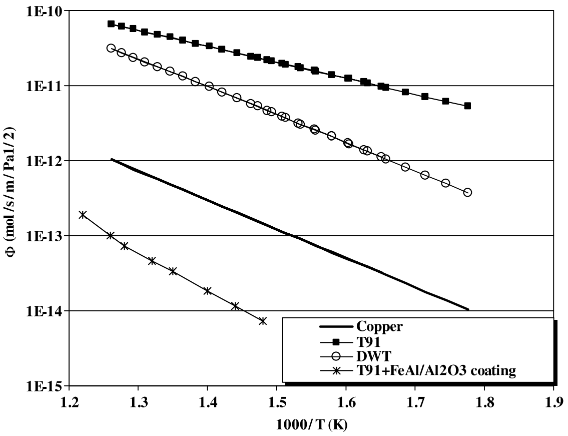

Fig. 2. Out of pile Arhenius plot of tritium permeability as function of temperature [6,7].

pressure of the purge gas. The measured tritium flow in mCi/min in the second containment can be converted in a tritium permeation rate (mol/s). At steady state this calculated rate is equal to the total amount of tritium permeated through the wall of the first containment.

2.3. Permeation theory and model

The mass flow of tritium through a cylindrical wall in steady state is described by [8]:

$$J = \frac{2\pi l}{\ln(b/a)} \Phi_{\text{eff}}(T)(p_{\text{in}}^n - p_{\text{out}}^n) \tag{2}$$

with J the net tritium rate from medium 1 to medium 2 (mol/s), b the outer diameter of cylinder (m), a the inner diameter of cylinder, l the length of cylinder, p_{in} the partial pressure of tritium at the inside (Pa), p_{out} the partial pressure of tritium at the outside (Pa), Φ_{eff} the effective permeability of the DWT or TBP (mol m^{-1} s^{-1} Pa^n).

The value of power n lies between 0.5 (diffusion is the rate limiting step in tritium transport) and 1.0 (tritium transport limited by surface effects) [9]. During steady state, the permeation through the DWT and the TPB is assumed to be diffusion limited ($n = 0.5$). The permeability (Φ) of hydrogen in structural materials is dependent on temperature described by the Arrhenius relation as plotted in Fig. 2. Eq. (2) can be used to compute the permeability of the TPB and DWT. However, the inhomogeneous temperature and tritium distribution complicates the analyses. In order to account for this a finite element model (FEM) has been developed which takes these phenomena into account. In this model the analogy between heat and mass transport was used, so the permeability could be regarded to be analogue to heat conduction and the square root of the tritium partial pressure analogue to temperature. This model was used to calculate the temperature distribution of the DWT and further developed to calculate the tritium permeation directly from the literature data. The same approach was used to model the tritium permeation in EXOTIC 8.10. However, the temperature distribution in EXOTIC 8.10 was difficult to interpret because of a dislocation of the thermocouples inside the ceramic breeder bed. Data on tritium permeation for this model was taken from Chabrol et al. [6] who measured the deuterium permeation rate out of pile through a T91 disc with the same coating as on the outside of the first containment of EXOTIC 8.10. These data can be converted into tritium permeation rates by multiplication with the ratio of the square root of the weight of deuterium and tritium ($\sqrt{2/\sqrt{3}}$) [9]. In both FEM the total tritium permeation from the first to the second containment was calculated for the same range of temperatures as measured in-pile and the assumption is made that the permeation process is diffusion limited, because the tritium permeation during steady state is calculated. The model is based on tritium permeation only, so effects of hydrogen and helium on permeation are not taken into account.

2.4. Influence of hydrogen on tritium permeation

The presence of hydrogen on either the inside or the outside of the tube can influence tritium permeation in three different ways [9]: First, because tritium diffusion through a metal is an atomic process, the tritium atom can recombine at the surface by isotopic exchange in the presence of hydrogen. When the hydrogen content in the purge gas in the second containments of both experiments is varied, this results in a change in the permeated percentage tritium. Second, surface modification is the process of altering the surface in a reducing atmosphere. Oxides present on the surface act as a permeation barrier and when reduced, tritium permeation is enhanced. So, as compared to the begin of the experiments, tritium permeation is increased by reduction of oxide compounds at the surface of the DWT of EXOTIC 8.9 and the reduction of the Al_2O_3 coating of the TBP in EXOTIC 8.10. Third, when the hydrogen gas in the first and second containment is varied, the tritium is 'diluted' by hydrogen, therefore the tritium diffusion – and permeation – are reduced. In both experiments the hydrogen concentration in the first containment is varied and the results are analysed.

3. Experimental results

3.1. Irradiation and comparison with FEM

Most data for the study of the temperature influence on the tritium permeation through a TPB and a DWT were obtained with helium purge gas doped with 1000 ppm hydrogen in both containments. This hydrogen concentration in helium is called reference gas. In order to evaluate the DWT and TPB performance during irradiation, the experimentally obtained PPT values are plotted in Figs. 3 and 4 as a function of the average wall temperature. In the temperature range 350–500 °C and for reference gas, the T91 tube with the Fe–Al/Al_2O_3 coating performs a factor 70 better in preventing tritium permeation than the DWT. Tritium permeation in the DWT is strongly influenced by temperature. The influence of temperature on the PPT value for the TPB is not seen clearly, partly due to a low permeability of the coating, which results in a low signal-to-noise ratio in the tritium measurement for the second containment of EXOTIC 8.10. The FEM calculated tritium percentages are plotted together with the experimental results as a solid line. For the DWT the experimental data are in reasonable agreement with the calculated permeation data. The in-pile and the FEM results of EXOTIC 8.10 are in the same order of magnitude, but the in-pile results they show somewhat higher tritium permeability. This is probably due to the estimation made in the temperature distribution, and therefore the calculated tritium permeation should be regarded as an estimation. The in-pile tritium permeation results were obtained in 0.1 vol.% hydrogen in helium environment. It can be concluded that when the reference gas is purged at a flow rate of 100 ml/min through both containments, tritium permeation behaves as if only tritium is present at low pressure and no additional purge gas flow is present. Trends in the PPT values due to irradiation time are not observed when all data are compared to the results of the first 100 days of irradiation [4] which indicates that the coating and the DWT did not deteriorate during irradiation.

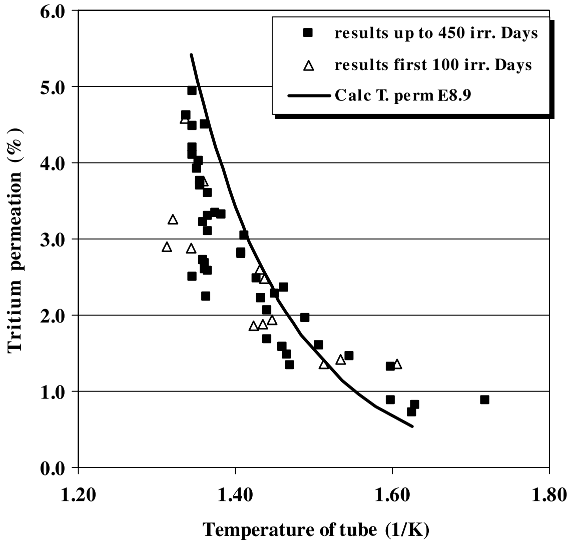

Fig. 3. Tritium permeation through DWT in EXOTIC 8/9 in-pile and FEM results.

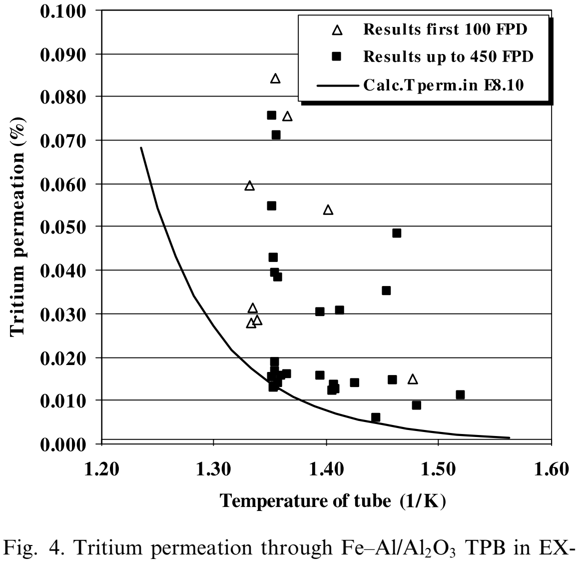

Fig. 4. Tritium permeation through Fe–Al/Al_2O_3 TPB in EXOTIC 8/10, in-pile and FEM results.

3.2. *Influence of purge gas chemistry*

The influence of different hydrogen concentrations on tritium permeation in the purge gas was also studied. To examine the influence of hydrogen content on one side of the permeation barrier on tritium permeation, the temperature was kept constant and the hydrogen content in the purge gas of the first containment was varied. This ‘sub-experiment’ was performed within 30 days. The tritium permeation at steady state is plotted as function of hydrogen concentration in the first containment in Fig. 5. The tritium permeation in E 8.10 is increased at a hydrogen concentration in the first containment of 100 ppm. Changes in permeation rates at higher hydrogen concentrations are not evident. No effect of hydrogen concentration in first containment is shown in E 8.9. In a next range of in-pile measurements, the hydrogen concentration on the outside of the DWT was varied and the temperature of the wall and purge gas composition in the first containment was kept constant. Increasing the hydrogen content in the second containment enhanced the tritium permeation by 40%. In Fig. 6 these data are shown together with the tritium permeation under reference gas condition.

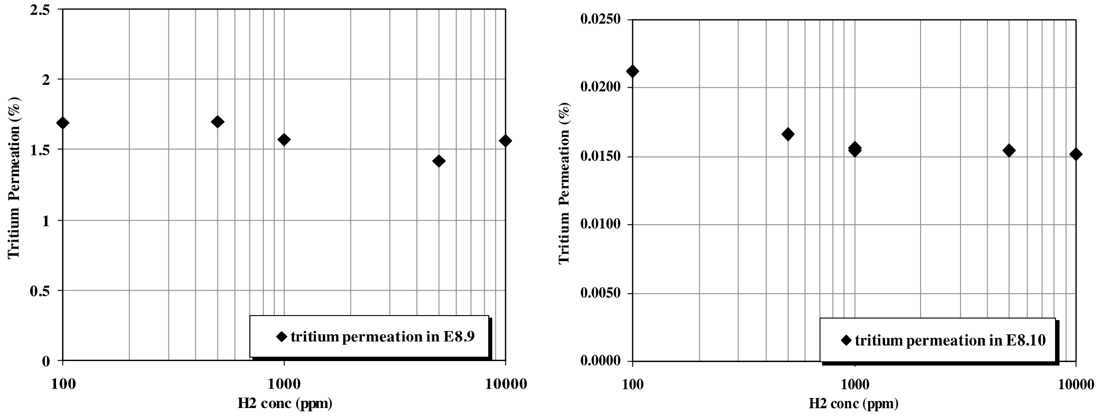

Fig. 5. Influence of hydrogen in first containment on tritium permeation.

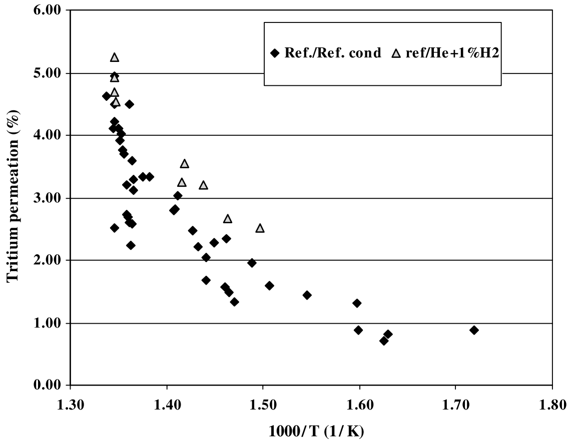

Fig. 6. Influence of hydrogen in second containment in EXOTIC 8/9 on tritium permeation.

4. Conclusions and recommendations

- Two TPBs for the WCLL-blanket concept were tested in-pile as first containments in EXOTIC 8.9 and 8.10, with on-line ability to measure tritium permeation in the second containment, from May 1999 till March 2001.
- Both TPBs showed no increase in tritium permeation trends during irradiation, which implies that the barriers did not deteriorate with respect to tritium permeation.
- The Fe–Al/Al_2O_3 TPB at the outside of a T91 tube performed about a factor 70 better in reducing tritium permeation than the T91 DWT with copper interlayer.
- The tritium permeation through the DWT increased significantly (about 40%) with increased hydrogen concentration in the second containment.
- The change in hydrogen concentration in the first containment had only an effect on tritium permeation at low hydrogen concentrations.
- Post-irradiation examination will be performed in order to obtain valuable information of irradiation impact on the TPB and the DWT.

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