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In-pile tritium-permeation measurements on T91 tubes with double walls or a Fe–Al/Al_2O_3 coating

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Abstract

Two new irradiation projects are being performed at the HFR Petten, named EXOTIC-8.9 and EXOTIC-8.10. Issues such as tritium release from candidate ceramic breeder pebbles for the HCPB blanket and tritium permeation through cooling tubes of the WCLL blanket are investigated simultaneously. In EXOTIC-8.9, the tritium release behaviour of a Li_2TiO_3 pebble bed is measured along with the tritium-permeation rate through a double-wall tube (DWT) of T91 with a Cu interlayer. In EXOTIC-8.10, the tritium release behaviour of a Li_4SiO_4 pebble bed is measured along with the tritium permeation rate through a T91 tube with a Fe–Al/Al_2O_3 coating as tritium permeation barrier (TPB). Tritium permeation phenomena are studied by variations of temperatures and purge gas conditions. This paper reports on the results of the first 100 irradiation days. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Within the European Blanket Project, the European associations jointly develop a water-cooled lithium–lead (WCLL) concept and a helium-cooled pebble-bed (HCPB) concept [1]. For the WCLL concept, components like the double-walled tube (DWT) and the tritium permeation barrier (TPB) are being developed [2]. These components have to be qualified in out-of-pile and in-pile tests. Pre-design studies have shown that dedicated irradiation tests for DWT and TPB qualifications are feasible in the HFR Petten [3] but considering the stage of component development, such tests will not start before 2001. Therefore, an idea has been developed to irradiate presently available DWT and TPB as part of the newly planned EXOTIC-8 in-pile tests of ceramic breeder pebbles for the HCPB programme [4].

The objectives of EXOTIC-8.9 and EXOTIC-8.10 for the HCPB programme are to measure in situ the tritium release characteristics and thermal behaviour of Li_2TiO_3 and Li_4SiO_4 pebble beds. Simultaneously, the tritium permeation rate through an uncoated DWT (EXOTIC-8.9) and through a T91 tube with TPB coating (EXOTIC-8.10) is measured in gas–gas conditions, using lithium-ceramics as the tritium source. The experiments have been designed to cope with temperature requirements for the two blanket concepts. Results for both experiments obtained in the first 100 irradiation days are now reported. Irradiation will be continued up to 300 days and followed by post-irradiation examination.

2. Experiment design and analysis

2.1. Irradiation of a DWT and a TPB

The EXOTIC-8.9 capsule contains Li_2TiO_3 pebbles supplied by the ENEA. The DWT acts as the primary containment at the outer side of the annular pebble bed, see Fig. 1. The DWT is fabricated at the CEA Grenoble using hot isostatic pressure (HIP) diffusion welding of

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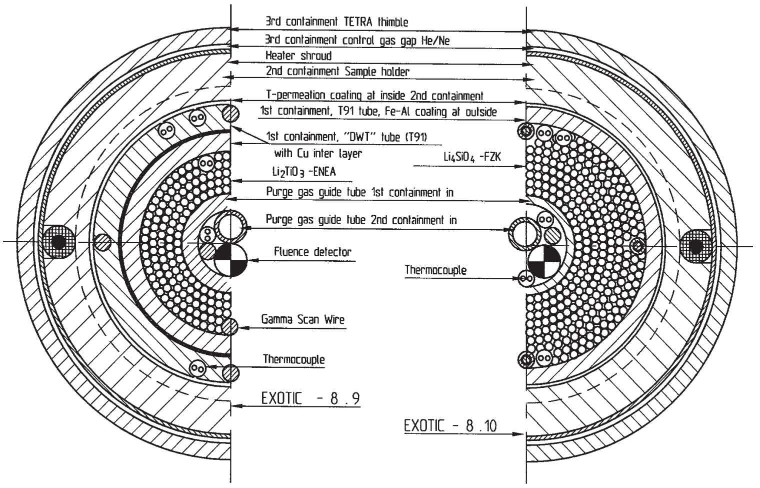

Fig. 1. Horizontal cross-sections of experiments EXOTIC-8.9 and EXOTIC-8.10. The DWT of EXOTIC-8.9 provides the primary containment for a ceramic breeder pebble bed. The TPB of EXOTIC-8.10 is located at the outer side of the thin-walled T91 tube that provides the primary containment for the ceramic breeder bed.

two 9% Cr steel (T91) tubes with an electroplated Cu interlayer of about 0.1-mm thickness [5]. The inner and outer diameters are 11.1 and 16.8 mm, respectively.

The EXOTIC-8.10 contains Li_4SiO_4 pebbles supplied by the FZK. This experiment is designed similar to EXOTIC-8.9; the breeder bed is, however, surrounded by a single-walled T91 tube coated on the outer side of the tube with a TPB. The TPB has been produced by a pack-cementation process (Fe–Al alloy) and CVD (Al_2O_3) at the CEA, Grenoble [6]. The tube has inner and outer diameters of 14.8 and 16.8 mm, respectively, while the coating is about 7-μm thick.

The pebble beds surround a stainless steel guide tube containing the purge gas lines for the primary and secondary containments. Both containments are purged independently. The purge gas can be varied between pure helium and helium with 1% H_2. The continuous monitoring of tritium in ionisation chambers (IC) allows the determination of the tritium permeation flux through the tubes in a temperature range of about 340–500°C. Additional TPBs have been applied to the inside of the stainless steel secondary containments in order to reduce parasitic tritium flow. Furthermore, the tube ends and the feedthrough welds are much colder than the tube near the breeder bed. The capsules are instrumented with thermocouples and neutron detectors to monitor temperatures and to determine the neutron fluence after irradiation.

2.2. Permeation theory

The tritium permeation through a tube can be described – in 'ideal' circumstances, i.e. with homogeneous temperature fields and constant partial pressures of a single hydrogen species on both sides – by [7,8]

$$J = \frac{2\pi l}{\ln(b/a)} \phi(T)(p_{\mathrm{in}}^{n} - p_{\mathrm{out}}^{n}), \tag{1}$$

where J is the tritium permeation (atoms/s), l the tube length (m), b the outer diameter (m), a the inner tube diameter (m), (T) the permeability (atoms m^{-1} s^{-1} Pa^{n}), p_{in} and p_{out} the equivalent tritium partial pressures in He (Pa) inside and outside the tube, respectively, and n is the power of the pressure dependence ($0.5 \leqslant n \leqslant 1.0$). The power n of the pressure 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 [8]. In the presented experiments, two types of purge gas have been used, namely, pure helium and helium containing 0.1% H_2, which is the reference purge gas mentioned in this paper. The aim of the present experiments is to find the fitting parameters of Eq. 1 for various hydrogen contents in the purge gases and to investigate 'competition' effects between the hydrogen and the tritium atoms in the permeation process.

2.3. A coupled temperature and permeation model

The present experiments have been designed to predict operational temperatures and [parasitic] permeation fluxes. The temperature distribution in the experiments has been computed using finite element modelling (FEM) with ANSYS 5.3. The thermal model has been transformed into a FEM tritium permeation model. Each element in the FEM model is given a permeability

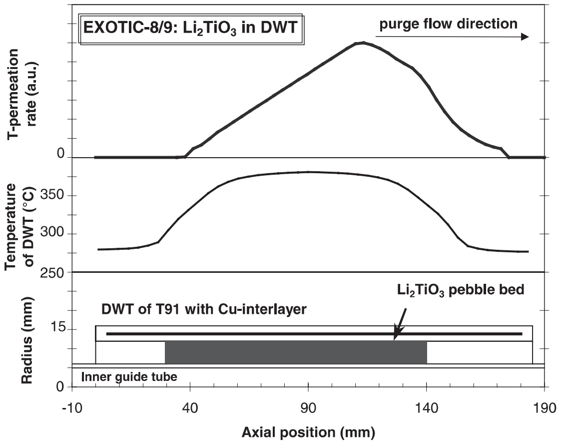

Fig. 2. The calculated geometrical distribution of the tritium permeation through the DWT. The figure shows an axial cross-section of the DWT and at the top and the bottom, the stainless steel parts are included.

value taking into account its temperature and the temperature dependence of the permeability. Fig. 2 shows the calculated distribution of the tritium permeation through the DWT along with the axial temperature profile. The axial asymmetry in the permeation flux is in line with the purge gas flow direction: it is higher at the gas outlet. The present computations show that the parasitic permeation (e.g., through the inner guide tube and the tube ends) does not play a significant role under purge flow conditions.

3. Experimental results

The permeation experiments are performed in two 'modes', being either a continuous purge gas flow or a stagnant gas in the first containment, with a variety of purge gases and/or temperature transients. In all cases, the second containments are continuously purged.

3.1. Continuous flow in first containment

A typical result of both temperature and purge gas transients in EXOTIC-8.9 is shown in Fig. 3. It can be seen that the amount of tritium coming from the first containment is influenced by the gas and temperature transients, which is due to the properties of the Li_2TiO_3 pebble bed. A temperature decrease or a hydrogen concentration decrease increases the time constant of the tritium release, causing a temporary dip (at $t = 5.5$ and 13.5 days) in the tritium concentration of the gas in the first containment. This effect can be corrected in the permeation computations. Various effects are observed, both for EXOTIC-8.9 and EXOTIC-8.10, from the tritium-release signal of the second containment:

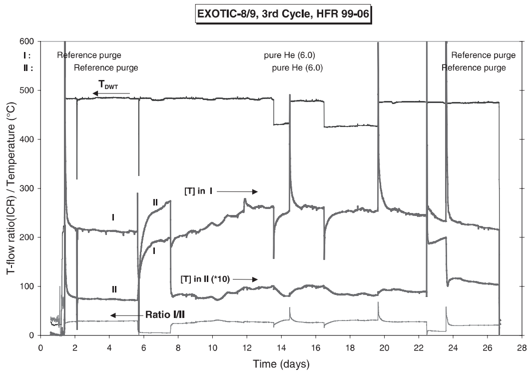

Fig. 3. The IC signal from the first containment, the second containment (multiplied by 20), the ratio of both quantities and the temperature. The purge gas conditions are included in the top of the figure and mark the various periods in time in which these conditions are present.

- Changing the purge gas in the first containment from the reference gas (helium with 0.1% H_2) to pure helium enhances the tritium permeation.
- Decreasing the temperature causes a gradual decrease of the amount of tritium permeating into the second containment, as can be seen after the temperature decrease at $t = 13.5$ days. The inverse effect is seen after the temperature increase at $t = 14.5$ days. Due to the relatively slow permeation processes, a rather rapid change of the tritium concentration in the first containment is seen not to affect the second containment or only to affect it much later.

Fig. 4 shows the ratio of the tritium flows in both containments as a function of the DWT temperature (average at centre) with reference purge gas in both containments. The tritium permeation increases with increasing temperature. A similar behaviour is observed for the TPB in EXOTIC-8.10. The temperature dependence of the permeation rate is similar to the DWT, but the absolute permeation is about 50 times smaller.

In the first 100 days of irradiation, the observed values for fractional permeation through the DWT and TPB do not show evidence of changes with increasing time and irradiation dose. Such analysis will be performed in detail on completion of the irradiation.

3.2. Stopped flow in first containment

Inhibiting the purge gas flow through the breeder bed causes a homogeneous tritium distribution, which increases the tritium production rate (Fig. 5). This increase causes the tritium permeation to increase with time, until it saturates in the case of the DWT, when the tritium permeation through the DWT becomes nearly equal to the tritium production. In saturated conditions, the tritium release from the second containment is about 60% of the initial value from the first containment.

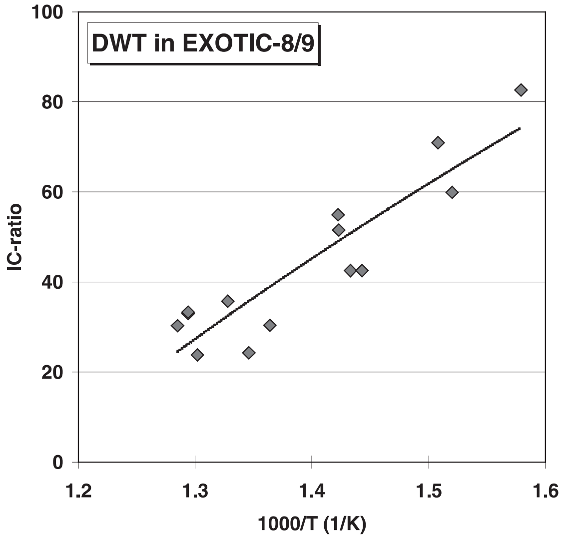

Fig. 4. The ratio of the IC signals of both containments as a function of temperature for EXOTIC-8.9. The values have been derived from the stabilised conditions after a temperature transient, with the reference purge gas at both sides of the tubes. The tritium concentration in the purge gas in the first containment is about 0.3 ppm.

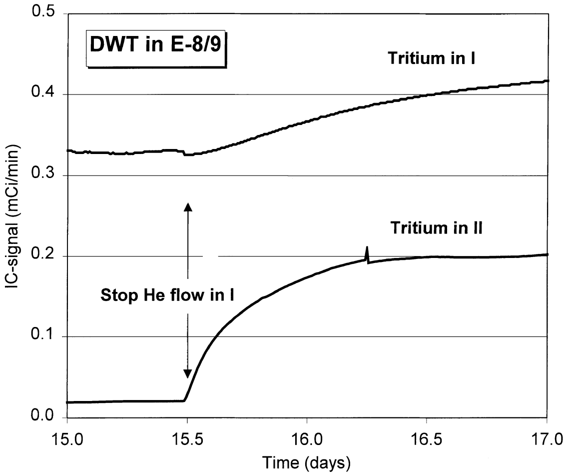

Fig. 5. The tritium flow out of the first and second containment for EXOTIC-8.9. Before the stop of the flow, the purge gas in the first containment was helium. The purge gas of the second containment was the reference gas, which flows continuously.

4. Discussion

4.1. Analysis of purge gas effects

Increasing hydrogen concentration at the inside of the tubes affects the tritium behaviour in various ways:

- The tritium residence time in the tritium-producing Li_2TiO_3 or Li_4SiO_4 ceramic decreases which causes a temporary increase in the tritium concentration in the inside of the tubes.
- A modification in the calibration factor of the ICs (Jesse effect), which can easily be taken into account (factor 1–1.5).
- The simultaneous presence of hydrogen and tritium in the inside of the tubes decreases the tritium permeation due to competition between tritium and hydrogen atoms in the adsorption or diffusion effects that determine the total tritium permeation. Fig. 3 (at $t = 6$ days) shows that changing the purge gas of the first containment from 0.1% H_2 to helium causes an increase in the permeation by a factor of about four.

Changing the gas at the outside of the TPB in EXOTIC-8.10 and the DWT in EXOTIC-8.9 from helium to the reference purge gas enhances the tritium permeation rate, as can be seen in Fig. 3 at $t = 22.5$ days. This shows that for very low hydrogen or tritium concentrations, surface effects (e.g., desorption) at the outside of the tubes decrease the permeation rate.

4.2. Comparison with literature data

In the field of deuterium and tritium permeation through fusion materials, various experiments have already been performed. Most of these experiments have been performed out-of-pile using deuterium [9]. Only a few in-pile experiments have been performed involving tritium [10]. These in-pile experiments were performed on other types of coatings under different test conditions and cannot be compared with the present results. Furthermore, some evidence exists of enhanced hydrogen permeation in strong radiation fields [11].

Serra et al. [9] studied the out-of-pile tritium permeation through a material sandwich that has been prepared by HIP and representative for a DWT similar to the presently studied one. They observed that the copper interlayer reduces the deuterium permeation rate by a factor of 5 as compared to a sandwich without copper. This reduced permeation is clearly due to the lower permeability of the copper.

Chabrol et al. [6] measured deuterium permeation for Fe–Al/Al_2O_3 by pack-cementation + CVD coatings deposited on T91 martensitic steel, similar to the TPB now being irradiated in EXOTIC-8.10. Their results show a rather high barrier efficiency, which when expressed in terms of a permeation reduction factor (PRF), was about 5000 at 673 K and 2500 at 773 K (with deuterium pressure of 7.5×10^4 Pa on one side and vacuum on the other). The present results, obtained in rather different geometrical and test conditions, confirm a rather low permeability of such a coating. Further quantitative analyses will be made when more in-pile data become available.

5. Conclusions

- As a contribution to the European Blanket Project, NRG and JRC-IAM conducted in-pile experiments with a unique design that combines key components for the WCLL blanket concept together with ceramic breeder materials for the HCPB blanket. Both a DWT and a TPB with todays technology have been included in the ongoing EXOTIC-8 series.
- In EXOTIC-8.9, a DWT forms the primary containment for a Li_2TiO_3 pebble-bed, and its in-pile tritium permeation efficiency is being determined. Tritium permeation data will be obtained for temperatures in the range of about 600 to 800 K and for different purge gases. In EXOTIC-8.10, a TPB-coated T91 tube forms the primary containment for a Li_4SiO_4 pebble bed. The parasitic tritium permeation is small in both experiments. Although the post-irradiation examinations on the TPB are the primary test objective, valuable data on its in-pile tritium-permeation behaviour have been obtained.
- In He + 0.1% H_2 purge gas conditions, the permeated fraction of tritium through the DWT (EXOTIC-8.9) ranges from about 1.2% at 360°C to about 3% at 500°C. The permeated fraction through the TPB-coated tube ranges from about 0.03% at 350°C to about 0.1% at 480°C. This reduction in the permeated fraction by a TPB coating is consistent with previous findings in out-of-pile experiments with deuterium.
- A model has been constructed that is capable of computing the temperature dependence of the permeability through the DWT and the TPB-coated tube, taking the impact of the exact experimental conditions (temperature distribution, tritium distribution and geometry) into account.
- The planned irradiation time of the experiment is about 300 days, about 1.5–2 dpa for the T91. A more detailed analysis will be made after completion of the experiment. Post-irradiation examination will be performed in order to obtain additional information on the impact of irradiation on the TPB and the DWT.

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