Fusion Engineering and Design 88 (2013) 2918–2921

Contents lists available at ScienceDirect

Fusion Engineering and Design

journal homepage: www.elsevier.com/locate/fusengdes

Tritium permeation in EUROFER in EXOTIC and LIBRETTO irradiation experiments

A.V. Fedorov*, S. van Til, A.J. Magielsen, M.P. Stijkel

Nuclear Research and Consultancy Group, Westerduinweg 3, Postbus 25, 1755 ZG Petten, The Netherlands

HIGHLIGHTS

- Tritium permeation in EUROFER97 is studied in EXOTIC-9/1 and LIBRETTO irradiation experiments.
- The irradiation temperature is varied between 300 and 580 °C.
- Method to analyze the tritium and hydrogen co-permeation at temperature transients is presented.
- The obtained effective diffusion coefficients of tritium in EUROFER97 appear to be factor ten lower than the literature data.

ARTICLE INFO

Article history:
Received 10 March 2013
Received in revised form 11 June 2013
Accepted 12 June 2013
Available online 18 July 2013

Keywords:
Tritium
Permeation
Diffusion
EUROFER
Neutron irradiation

ABSTRACT

The reduced activation martensitic steel (RAFM) EUROFER is foreseen as a structural material in test breeder module (TBM) in ITER and breeder blanket in DEMO design. In a number of irradiation experiments conducted in high flux reactor (HFR) in Petten EUROFER was used as a containment wall of the breeder material, through which tritium permeation was monitored on line. Thus in EXOTIC-9/1 (EXtraction Of Tritium In Ceramics) experiment where Li_2TiO_3 pebbles were the breeder material, EUROFER was irradiated up to 1.3 dpa at 340–580 °C. In LIBRETTO experiments (LIBRETTO-4/1, -4/2 and -5) the breeder material was lead lithium eutectic which was in direct contact with the EUROFER containment wall. The neutron damage in steel achieved in the LIBRETTO experiments varied from 2 to 3.5 dpa. The irradiation temperature was 350 °C (LIBRETTO-4/1), 550 °C (LIBRETTO-4/2), and 300–500 °C (LIBRETTO-5).

Tritium permeability was studied by varying the irradiation temperature and hydrogen concentration in the purge gas. From the analysis of the temperature transients performed in all four experiments yielded the tritium diffusion coefficients were derived, which appear to be factor ten lower than the literature data obtained in the gas driven permeation experiments.

1. Introduction

The reduced activation martensitic steel (RAFM) EUROFER is foreseen as a structural material in test breeder module (TBM) in ITER and breeder blanket in DEMO design [1]. Tritium transport in EUROFER is a critical parameter required for the blanket design and safety assessment, which was a subject of multiple studies reported in the literature [2–5]. In a number of irradiation experiments conducted in high flux reactor (HFR) EUROFER was used as a containment wall of the breeder material and tritium permeation was monitored in on line. While the tritium permeability is measured under steady state irradiation conditions, the tritium diffusion coefficient is directly measured in temperature transition experiments [6] and [7]. In order to extract the diffusion coefficient from such experiment one needs to develop a tritium transport model utilizing the cylindrical symmetry of the experiments. Moreover, in the considered experiments the tritium partial pressure at the "high pressure" side is not a step function, as in a typical gas-driven permeation experiment, but rather presents a continuously varying signal.

This work presents a method developed to analyze the tritium transport during temperature transients performed in the EXOTIC-9/1 and LIBRETTO irradiation experiments. The results on the tritium diffusivity measured in these experiments are presented and discussed.

2. Design of EXOTIC-9/1 and LIBRETTO experiments

The aim of the EXOTIC-9/1 experiment was to investigate the in-pile tritium release characteristics of lithium containing ceramic breeder and to measure tritium permeation through the EUROFER97 wall of the pebble bed containment. The pebble

* Corresponding author. Tel.: +31 224568034.
E-mail addresses: fedorov@nrg.eu, goedemorgen90@hotmail.com (A.V. Fedorov).

0920-3796/$ – see front matter
http://dx.doi.org/10.1016/j.fusengdes.2013.06.003

Table 1
Irradiation parameters of EXOTIC-9/1 and LIBRETTO experiments.

	EXOTIC-9/1	LIBRETTO-4/1	LIBRETTO-4/2	LIBRETTO-5
FPD	301	732	732	517
T_{irr}, °C	340–580	550	350	300–500
dpa	1.3	3.4	2.0	3.0
breeder	Li_2TiO_3	Pb-17Li	Pb-17Li	Pb-17Li
⌀ in, mm	12.2	14.1	14.1	14.1
⌀ out, mm	16.6	16.7	16.7	16.7

bed assembly containing lithium titanate (Li_2TiO_3) was enclosed between two sealed concentric EUROFER97 tubes forming the 1st containment. Tritium produced in the pebble bed was extracted by the purge flow with a reference composition (He + 0.1 vol.% H_2). The 2nd containment was formed by a stainless steel tube, which also serves as a guiding tube for the additional heater. The heater is used to perform fast temperature transients. A small gas gap between the 1st and 2nd containment is continuously purged during the irradiation with the reference gas. The purge gas lines are connected to the out-of-core tritium measurement station (TMS), where the tritium concentration is measured in both lines independently using two ionization chambers (IC). While the IC1 signal measured in the 1st containment presents the tritium released from the pebble bed, the IC2 signal measured in the 2nd containment presents the tritium permeated through the EUROFER97. The distance between the reactor core and TMS is about 20 m, the purge gas flow is 100 ml/min and pressure is 2.8 bar.

The EXOTIC-9/1 was irradiated for 301 full power days (FPD) with the temperature varied between 340 and 580 °C, reaching a lithium burn up of 3.5% and 1.3 dpa of damage in steel.

In three LIBRETTO experiments, LIBRETTO-4/1, -4/2 and -5, the breeder material was lead lithium eutectic which was in direct contact with the inner surface of the EUROFER97 containment wall. The LIBRETTO-4/1 and LIBRETTO-4/2 are identical experiments operated under 550 °C and 350 °C, respectively. In LIBRETTO-5 the irradiation temperature varied between 300 and 500 °C. Similar to EXOTIC-9/1, in LIBRETTO experiments the lead lithium eutectic was enclosed between two sealed concentric tubes. The outside tube manufactured from EUROFER97 presented the wall of the 1st containment. The tritium was removed from inside and outside the containment with two independent purge gas lines with He + 0.1 vol. % H_2 connected to the TMS. The irradiation conditions of all four experiments are summarized in Table 1.

3. Results and discussion

3.1. Temperature transients

In EXOTIC and LIBRETTO experiments the temperature transients were produced by changing the temperature of the breeder in-pile by using the heaters located in the shroud of the irradiation assembly. An example of a temperature transient is demonstrated in Fig. 1. A step-wise increase of the breeder temperature results in a spike of the tritium partial pressure (HT-molecular form) measured with the ionization chamber IC1. After a delay depending on the tritium diffusivity the spike is measured in the 2nd purge gas line by the ionization chamber IC2. From the measured time delay the diffusion coefficient is determined.

3.2. Analysis of transients

The steady state diffusion equation in cylindrical geometry has the following solution [9]:

$$C_{ss} = C_1 - \frac{C_1 - C_2}{\log(r_2/r_1)} \times \log(r/r_1) \quad (1)$$

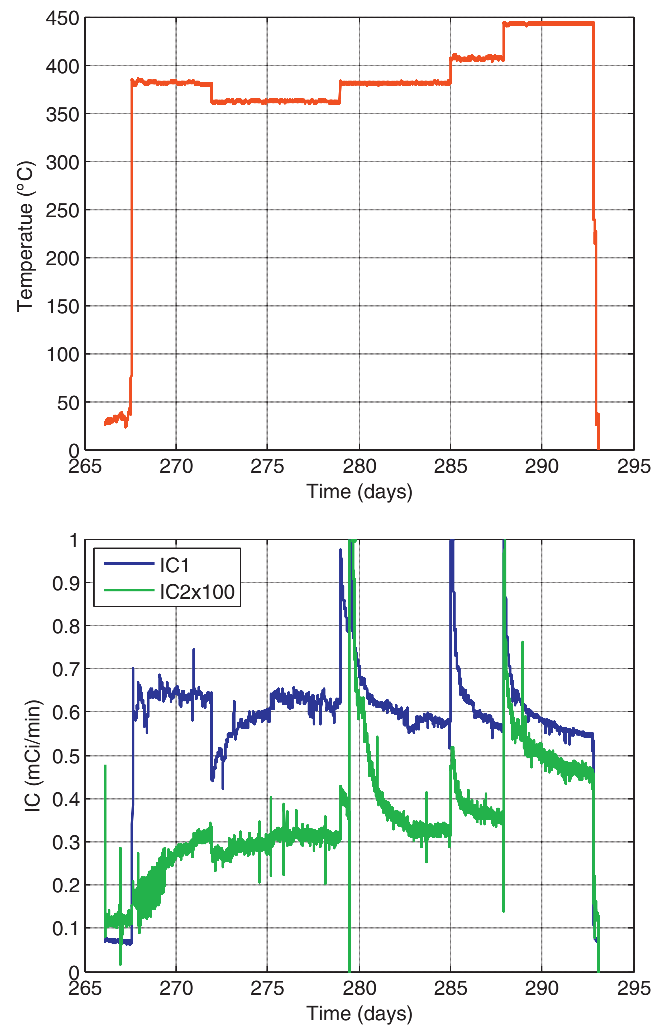

Fig. 1. Top: variation of the EUROFER97 wall temperature. Bottom: tritium concentrations measured in the purge lines, IC1-tritium release from the breeder; and IC2-tritium permeation through the EUROFER97 wall.

where C_1 and C_2 are the tritium concentrations at the inner, "high pressure", boundary, r_1, and at the outer boundary, r_2, respectively. In diffusion limited regime the surface concentration is related to the partial pressure of tritium in molecular form HT via the following equation [8] and [11]:

$$C_{1(2)} = 0.5 \times S \times \frac{P_{HT,1(2)}}{\sqrt{P_{H_2,1(2)}}} \quad (2)$$

where S is the tritium solubility in EUROFER97, P_{HT} and P_{H_2} are the partial pressures of HT and H_2 in the purge gas. As in the previous equation, index 1 refers to the inner boundary and index 2 to the outer boundary. In further discussion the partial pressure of HT in the 2nd purge line, $P_{HT,2}$ is ignored because of a very low value. The partial pressure $P_{HT,1}$ is derived from the activity measured by the ionization chamber (IC1) as follows:

$$P_{HT} = \frac{A}{\lambda} \times \frac{RT}{N_{av}} \times \frac{1}{F} \quad (3)$$

where A is the tritium activity in the purge gas flow in a unit of time (Bq/s), $\lambda = 1.79 \times 10^{-9}\ s^{-1}$ is the decay constant for tritium, R = 8.31 J/mol/K is the universal gas constant, T is the absolute temperature in the pebble bed in K, $N_{av} = 6.022 \times 10^{23}\ mol^{-1}$ is the Avogadro number and F is the gas flow in m^3/s.