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Development of a technical scale PERMCAT reactor for processing of highly tritiated water

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ABSTRACT

In view of future fusion rectors fueled by deuterium–tritium mixtures, highly tritiated water (HTW) of up to $5.2{\cdot}10^{16}$ Bq kg^{-1} will be produced, during routine operation and scenarios as an accidental release of tritium into a glove box. Also in the solid breeder blanket concept, a non-negligible fraction of the tritium produced will be in the tritiated water fraction. To decontaminate HTW the PERMCAT using isotope swamping in a Pd/Ag membrane reactor has been identified as a robust and reliable solution. In order to investigate the decontamination of HTW at flow rates relevant for future fusion power plants, a technical scale, fully tritium compatible PERMCAT consisting of a bundle of finger-type membranes inserted in a single catalyst bed was developed. Nevertheless, it represents only one part of a PERMCAT cascade necessary to achieve the required performance to process HTW on technical scale. By improving the existing PERMCAT geometry using experimental data obtained from isotopic exchange between D_2O and H_2, the performance of the existing PERMCAT reactor was optimised. Based on the optimised geometry a new fully tritium compatible technical scale PERMCAT cascade comprising of two PERMCAT reactors in series was designed, manufactured and commissioned as presented in this paper.

1. Introduction

As described in the previous paper [1] large quantities of tritiated water of low ($0.4{\cdot}10^{12}$ Bq kg^{-1}) to medium ($11.1{\cdot}10^{12}$ Bq kg^{-1}) activity concentration is inevitably produced in a future fusion plant using tritium in its fuel cycle. Options currently under investigation such as the Combined Electrolysis Catalytic Exchange (CECE) process chosen for ITER are able to process tritiated water of a mean specific activity of $0.4{\cdot}10^{12}$ Bq kg^{-1} [2] with a decontamination factor (DF) of $2.5{\cdot}10^{5}$ [2,3]. The additional amount of very highly tritiated water (HTW) of up to $5.2{\cdot}10^{16}$ Bq kg^{-1} (expected from torus cleaning and accidental tritium release into a glove box [4]) are to be processed in a short period of time down to an activity concentration compatible with the CECE process. The high radiotoxicity [5], the characteristic radiochemical decomposition (self radiolysis) [6–8] and the corrosive nature of HTW [9] inevitably require a fast and safe decontamination with a robust and reliable process [1]. Therefore, the TLK has proposed the PERMCAT process to decontaminate HTW [10]. HTW will also be present in the purge gas of the solid breeder blanket. According to the latest design review [11,12] tritiated water shall be removed from the helium purge gas using an adsorption column, i.e. a molecular sieve bed (MSB) containing zeolite material and operated at room temperature. To recover the tritium from this water, the MSB shall be regenerated and this stream further processed, either using a reducing bed or alternatively with a PERMCAT reactor [13].

The PERMCAT process is operated at 673 K and involves a catalytic membrane reactor used in counter current mode for tritium removal via isotopic swamping while maintaining a very low tritium activity at the outlet of the component. H_2 used as swamping gas is fed into the PERMCAT reactors swamping gas side while impurities are fed to the impurity (catalyst) side of the PERMCAT in counter current. Both sides of the PERMCAT are separated by a semipermeable Pd/Ag-membrane selective only to hydrogen isotopes. The swamping gas is enriched with tritium and taken out of the PERMCAT through capillaries while the impurities are decontaminated. This process is abundantly described in several previous publications [10,14,15]. Fig. 1 is showing a schematic view of all the essential PERMCAT components together with a sectional view showing several finger type PERMCAT modules in a catalyst bed.

This paper presents the geometrical optimisation of the existing technical scale PERMCAT reactor [1] to increase its performance. Furthermore, the design, manufacture and functional testing of a new two stage technical scale PERMCAT based on this improved

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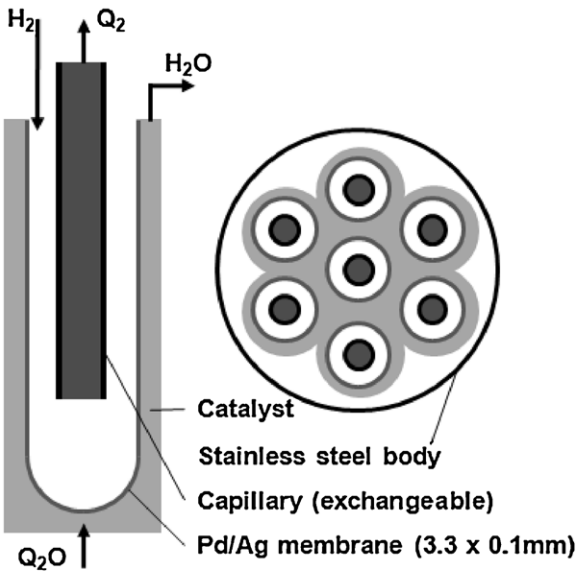

Fig. 1. Schematic view of a finger type PERMCAT reactor to process tritiated water (Q represents either H, D or T) showing the essential components and the counter current processing. Several finger type PERMCAT modules in a catalyst bed are used in the PERMCAT examined in this paper.

geometry is presented. The increase of performance of this new two stage PERMCAT is compared to the original single stage PERMCAT.

2. Improving the geometry of the existing PERMCAT

2.1. Goals and requirements

As recently reported, a new technical scale, multi tube, single bed PERMCAT was designed, manufactured and commissioned at TLK [14]. The key process parameters (swamping gas ratio, total water vapor flow rate, water vapor pressure) for operating a PERMCAT were already thoroughly investigated in the last paper presented [1], resulting in a proposal of a three stage PERMCAT cascade to decontaminate HTW. Therefore, to further optimise the PERMCAT performance before starting the design of a three stage cascade it was necessary to improve the geometry of the technical scale PERMCAT reactor.

2.2. Setup and procedures

The experimental setup used to perform isotope exchange experiments between D_2O vapor and H_2 is identical as reported in the previous paper [1]. D_2O vapor produced by an evaporator and He carrier gas from a gas bottle is fed at a 1:1 ratio into the PERMCAT while being processed counter current to H_2 at a fixed swamping ratio of 1:4. The vapor is therefore depleted of D_2 while the swamping gas is enriched with D_2.

After each experiment the DF (quotient of deuterium concentration in the feed water and the water at the impurity outlet) is determined by measurement of the collected water with infrared spectroscopy with an overall uncertainty of 12%. The transient time to reach the equilibrium during the experiment is monitored using a leak detector mass spectrometer. The exact procedure is described in the previous paper [1].

In order to investigate the impact of different geometries on the PERMCAT performance the capillary bundle inserted into the 3.3 mm×0.1 mm diameter Pd/Ag membrane tubes used to extract swamping gas out of the Pd/Ag membranes were manufactured to be easily exchangeable. The principal arrangement of such a component is shown in Fig. 1. By inserting capillaries of different diameters it is possible to change gas velocity and therefore residence time of the swamping gas in the PERMCAT reactor. Capillary bundles of 0.8, 1.4, 2.0, 2.3 and 2.5 mm outer diameter each 325 mm long and consisting of 13 capillaries exactly matching the pattern of the Pd/Ag-membranes were manufactured. In order to eliminate the influence of hydrogen permeation through the capillaries an identical wall thickness of 0.2 mm was chosen. Furthermore the same grade of stainless steel (1.4571, equivalent to SS316TI) was used for all capillaries. Fig. 2 shows the swamping gas side of the reactor and the insertion of a capillary bundle. Experiments were performed at 50, 100, 150, 200 ml min^{-1} (stp) D_2O feed flow rate. Pressure in swamping and catalyst side was kept constant at 75 hPa and 950 hPa respectively.

2.3. Experimental results

Fig. 3 shows the DF as a function of the total D_2O flow rate at a swamping gas ratio of 1:4. The Impact of the capillary diameter is easily recognizable. The highest DF obtained during all experiments was DF = 88.9 ± 10.6 while using the capillary bundle consisting of 1.4 mm×0.2 mm diameter capillaries.

The PERMCAT was fed with 50 ml min^{-1} (stp) D_2O and 200 ml min^{-1} (stp) H_2 as counter current swamping gas (swamping ratio of 1:4). For higher D_2O flowrates, while keeping the swamping ratio at 1:4 the DF is decreasing. This behavior is reproducible for all other capillary bundles, but the DFs reached using 1.4 mm×0.2 mm

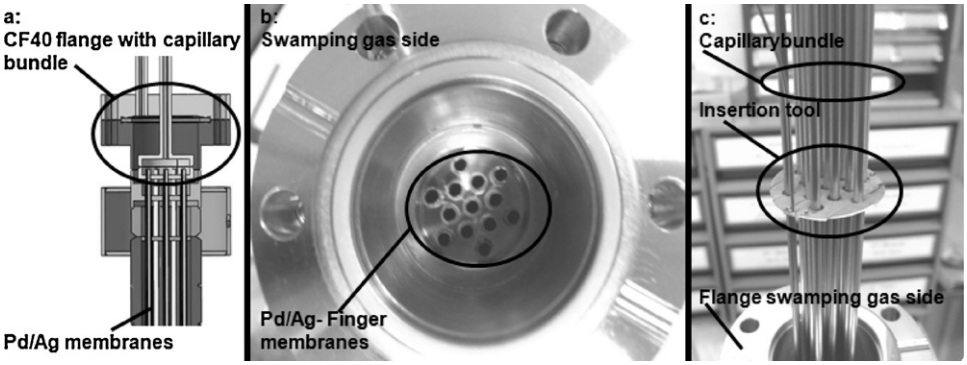

Fig. 2. (a) Upper part of the PERMCAT reactor showing the capillary bundle attached to a CF40 flange inserted into the Pd/Ag finger membranes. (b) Swamping gas side of the PERMCAT reactor during capillary change, the Pd/Ag membrane tubes without capillary inserted are visible. (c) Capillary bundle is inserted using a purpose built insertion tool, the swamping gas side is closed using a CF40 flange after inserting the capillary bundle.