Fusion Engineering and Design 87 (2012) 547–550

Contents lists available at SciVerse ScienceDirect

Fusion Engineering and Design

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

Inactive commissioning of a micro channel catalytic reactor for highly tritiated water production in the CAPER facility of TLK

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ARTICLE INFO

Article history:
Received 15 September 2011
Accepted 19 January 2012
Available online 3 March 2012

Keywords:
Fuel cycle
Highly trititated water
Micro-reactor

ABSTRACT

In future DT fusion machines, several events will generate highly tritiated water (HTW). Among potential techniques for HTW processing, isotopic swamping in a catalytic membrane reactor (PERMCAT) appears promising. The experimental demonstration of PERMCAT for HTW processing has started in the CAPER facility at the Tritium Laboratory of Karlsruhe (TLK). Without any HTW source, such water has to be produced on purpose.

Catalytic HT oxidation would ensure clean operation but could be critical for operation due to possible occurrence of explosive mixture. A tritium compatible micro-channel catalytic reactor (μCCR) has been designed and manufactured to produce up to 10 mL min^{-1} of HTW with very high specific tritium activity (stoichiometric DTO: 5.2×10^{16} Bq kg^{-1}). Prior to its integration in CAPER for tritium operation, this reactor has been commissioned at different feed flow rates, gas composition (air or Helium), and temperature. The results demonstrate the good performances of the μCCR in producing water.

The combination of the μCCR with the O_2 sensor represents a reliable system able to produce HTW in a safe way and without radioactive waste. Accordingly, the CAPER facility can be upgrade in order to continue the R&D activity on HTW processing with PERMCAT.

1. Introduction

Highly tritiated water (HTW) will be inevitably produced in a D-T fusion machine, like ITER, due to the reaction between tritium and oxygen. This reaction can occur during tritium recovery from the first wall or in case of accidental release of tritium in a glove box or in a room. Although the amount of such HTW will be small, a suitable detritiation process has to be identified both for safety and economic reasons [1]. From a safety point of view, HTO is very dangerous not only for its high radiotoxicity but also because its self radiolysis leads to the formation of explosive mixture of HT and O_2 [2]. Moreover, as reported by Bellanger [3], tritiated water causes corrosion of stainless steel by pitting and crevice attack. According to these considerations, the storage of HTW should be avoided.

A possible way to recover tritium from water is the isotopic exchange, which can be achieved by using a PERMCAT reactor: a catalytic membrane reactor that combines a Pd/Ag membrane exclusively permeable to hydrogen isotopes (Q_2) and a catalyst bed promoting the isotopic exchange to liberate tritium from molecules (Q_2O). By operating in counter-current isotope swamping mode, the PERMCAT reactor allows the tritium removal from the inlet gas stream and ensures a low tritium activity at the outlet [4,5].

To evaluate the feasibility of the PERMCAT in processing HTW, an experimental demonstration has started in the CAPER facility of the Tritium Laboratory Karlsruhe (TLK) [6]. In a first approach, the HTW was obtained by using a metal oxide reactor (MOR) but, due to the problem related with the final disposal of the MOR, the tritium content of the HTO produced was limited to 1%. However, the results obtained during these experiments provided promising information about the decontamination factor (DF) and thereby proof of principle. Alternatively to the use of MOR, the HTW necessary to feed the PERMCAT (tritiated water having a tritium content up to 50%) can be produced by catalytic HT oxidation. In this view, a specific micro-channel catalytic reactor (μCCR) has been designed by the Institute for Micro Process Engineering of KIT. This reactor is coated with a Pt catalyst that promotes the reaction between O_2 and Q_2. The velocity of the reaction together with the small dimension of the channels avoids any risk of explosion-detonation inside the device. This paper describes the inactive commissioning of the μCCR required before the integration into the CAPER facility.

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doi:10.1016/j.fusengdes.2012.01.021

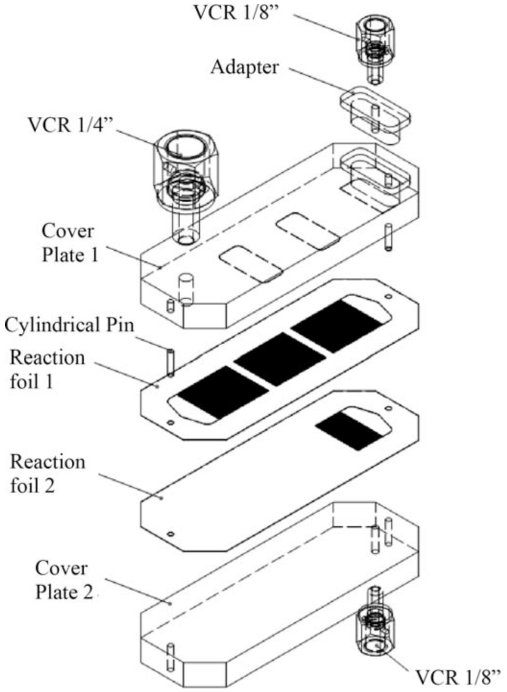

Fig. 1. Technical drawing of the microstructured catalytic reactor (μCCR).

2. Experimental

2.1. Description of the μCCR

The microstructured reactor consists of two stainless steel foils with microchannels (Fig. 1). The foils have been structured by wet chemical etching combined with laser drilling. Hydrogen/tritium and oxygen-rich gas are mixed via small holes in the bottom of the channels of the top foil (reaction foil 1) inside the microreactor. The top foil is the reaction zone and is thus to be coated with catalyst. The bottom foil (reaction foil 2) is distributing the oxygen-rich gas to the mixing holes in the top foil. Two gas windows are milled inside the top cover plate, to allow a cross mixing of the individual gas streams, i.e., the reaction mixture, between the channels sections. This design was regarded to avoid different conversion in the individual channels and to remove the influence of the manufacturing tolerance of channel geometry, hole size and catalyst coating thickness. A bottom plate, the top cover plate (each 5 mm thick), and the foil stack are joined together by diffusion bonding. VCR connections are connected by electron beam welding. The geometrical dimensions of the microreactor are summarized in Table 1. Subsequently to diffusion bonding, the reaction air and hydrogen passages have been impregnated with a $Pt(NO_3)$ solution. The catalyst is dried in air at 100 °C for 3 h and reduced in an Ar flow containing 5 vol.% of H_2 at 200 °C for 1 h.

Table 1
Geometrical data of the μCCR.

Channel width	300 μm
Channel height	100 μm
Wall width	150 μm
Channel per foil	45
Channel length	12 mm
Number of foils	2
Hole diameter	50 μm

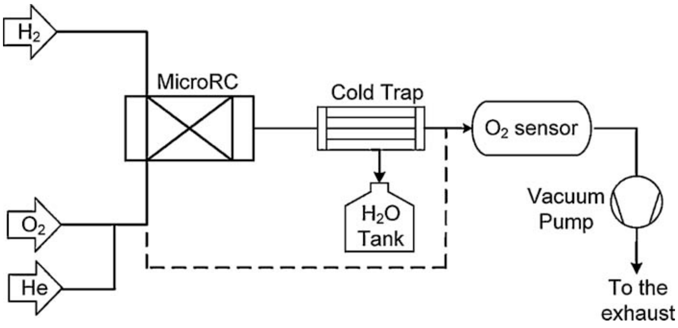

Fig. 2. Schematic view of the experimental setup (the dash line is the bypass used for calibrate the oxygen sensor). The μCCR converts the H_2 and O_2 into water which is condensed and collected trough the cold trap, while the oxygen sensor measures the un-reacted O_2. To reduce risk of explosion in case of a malfunction of the μCCR, O_2 is mixed with He.

2.2. Experimental setup

To perform the inactive commissioning, the micro-channel catalytic reactor has been integrated in the experimental setup reported in Fig. 2. It is equipped with instruments for measuring and controlling flow and pressure of the gases, the temperature of the reactor and the un-reacted oxygen. Particularly, two gas streams, composed of hydrogen and a mixture of oxygen and helium, entered the μCCR, which is operated at up to 120 °C. Inside the reactor, oxygen and hydrogen are instantaneously converted into water, according to Eq. (1).

$$H_2(g) + 1/2O_2(g) \rightarrow H_2O(v) \qquad (1)$$

The water produced is condensed and collected in a cold trap, while the other gases (i.e., oxygen and hydrogen non reacted and helium) are measured by paramagnetic based oxygen sensor (M&C TechGroup Version PMA 45 K). The whole system is connected to a vacuum pump able to evacuate the entire system down to $<10^{-3}$ hPa.

2.3. Experimental procedure

Before each experiment, the apparatus is evacuated and flushed with He; this procedure is repeated until the O_2 sensor measures no oxygen. Once this condition is reached, a mixture of helium and oxygen is sent into the reactor until the oxygen sensor reaches a constant value. Afterwards hydrogen is fed into the reactor at constant flow. In this way is possible to verify the response of the sensor at the different oxygen concentrations. In fact, as soon as the reaction occurs, water is formed and the oxygen is consumed so that its concentration downstream of the system decreases.

By using this experimental setup, the performances of the μCCR have been evaluated at different operative conditions. Particularly, the reaction conversion has been evaluated at various O_2/H_2 feed flow ratios and several reactor temperatures.

3. Results

3.1. First functional tests at IMVT with air

To investigate the activity of the catalyst inside the microreactor, tests were performed with hydrogen and air at a temperature of 120 °C. This temperature was selected to keep permeation through the outer reactor wall in the tritium application at a tolerable value. The air flow was 25 ml min^{-1} Standard Temperature and Pressure (STP). Due to the fact that tritium and oxygen excess should be avoided for the tritium application, the dependence of the conversion around an air ratio of 1 was investigated. The H_2 feed was thus