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PERMCAT experiments with tritium at high helium flow rates relevant for the tritium extraction systems using the CAPER facility at TLK

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HIGHLIGHTS

- We examined PERMCAT reactor efficiency processing tritiated water at high Helium carrier flow rates.
- We have found that – as expected from previous studies – that the swamping ratio (ratio between the impurity and purge side flow rates) has a key effect on the decontamination factors.
- On the other hand, some rather unexpected effects tend to show that the limiting phenomena of such specific operation of PERMCAT reactors (at high impurity flow rates, thus short residence time) lies on the kinetics of the isotope exchange reactions.

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ABSTRACT

Experiments are still necessary to consolidate the processes retained for the Tritium Extraction Systems of the European ITER Test Blanket Modules (TBM). A PERMCAT reactor combines a catalyst promoting isotope exchange reactions and a Pd/Ag membrane allowing tritium recovery from complex gaseous mixtures containing tritium in different chemical forms. Originally developed for the Tokamak Exhaust Processing, the PERMCAT process is also candidate to detritiate the water arising from an adsorption column installed in the TBM ancillary systems. We discuss the results of an extensive experimental campaign using a PERMCAT reactor to process Q_2O containing impurity gas mixtures at high flow rates. Two different experimental configurations were studied, namely PERMCAT stand-alone, and PERMCAT in combination with a zeolite molecular sieve bed (MSB, previously loaded with Q_2O) under regeneration. On the one hand, many expected behaviors were observed, such as the key influence of the swamping ratio (ratio between the impurity and purge side flow rates) on the decontamination factors. On the other hand, some rather unexpected effects tend to show that the limiting phenomena of such specific operation of PERMCAT reactors (at high flow rates, thus short residence time) lies on the kinetics of the isotope exchange reactions.

1. Introduction

A PERMCAT reactor is a catalytic membrane reactor combining a Pd/Ag membrane exclusively selective for molecular hydrogen isotopes and a catalyst bed promoting isotopic exchanges between tritium in molecules and gaseous hydrogens. The working principle is based on counter-current isotope swamping. The gas to be detritiated is fed into the "impurity side" of the reactor that contains the catalyst bed, while fresh (tritium-free) hydrogen is supplied in "purge side" in a counter current mode. A more detailed description of the PERMCAT process can be found in [1].

Thanks to the counter current isotope swamping mode, a PERMCAT reactor can be operated such as a very low tritium activity can be maintained at the impurity outlet, thus enabling very high decontamination/detritiation factors (DF) to be reached. The DF is very sensitive to many factors that make this process very flexible but also a bit difficult to design and master. In general, the following parameters play a crucial role:

- The length of the reactor: at given operating conditions, the DF increases exponentially with the length of the reactor (DF to the square is expected by doubling the length; thus, the length of the reactor is crucial for detritiation performances),

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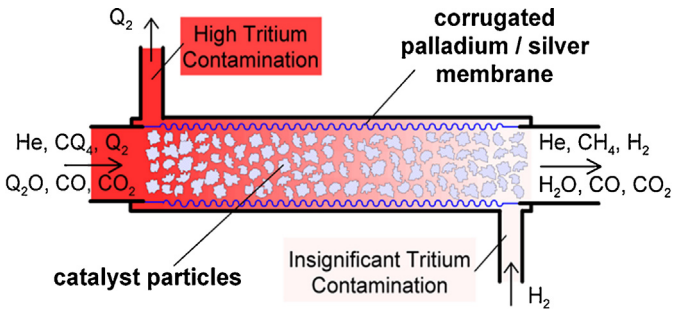

Fig. 1. PERMCAT reactor (single-tube unit comprising a corrugated membrane).

- Ratio between impurity and purge sides flow rates (called swamping ratio): increasing H_2 purge flow rate increases DF, but it can be detrimental if too much H_2 is used, optimization of this ratio is key for any PERMCAT operation.

In this work, we discuss the results of processing Q_2O in Helium carrier gas up to 800 mL/min flow rates.

2. Experimental setup

The PERMCAT reactor under operation in CAPER has been installed in 2007 and underwent several experimental campaigns [2]. The PERMCAT reactor is of special and advanced design comprising "corrugated" Pd–Ag membranes [3], see Fig. 1. The initial tubular membrane (100 μm thickness, 280 mm length, 6.3 mm outer diameter) provides a permeation area of 55.0 cm^2; the actual membrane length is a bit shorter (about 225 mm) after the convolution process. The nominal PERMCAT catalyst is used (Nikki 111) and 9 g is hosted in the inner part of the membrane (inside the membrane). Two identical units are mounted in line on a CF flange offering a sufficient length and the possibility for placing a monitoring system (ionization chamber, IC) at the mid-pathway of the process.

2.1. PERMCAT standalone configuration

For PERMCAT standalone experiments, a metal oxide reactor (MOR) containing CuO and operated at 400 °C is used to produce the tritiated water vapor carried with He; this configuration mirrors the first experiments on highly tritiated water performed in CAPER in 2010 [3] and uses the process configuration as shown in Fig. 2. The Helium/Q_2O mixture is fed into the PERMCAT reactor for detritiation. The moisture content of the decontaminated feed stream after the PERMCAT reactor is trapped in a small MSB, and finally handled in the TLK infrastructure. The H_2 purge stream (after removing most of the tritium content from the feed stream) is stored for later handling in the Q_2 recovery vessel.

In total, eight experimental runs were performed in this configuration to study the effects of different parameters on the DFs.

2.2. PERMCAT with zeolite MSB under regeneration

For this experimental campaign the setup was modified by replacing the MOR module with a zeolite MSB for temporary Q_2O storage (see Fig. 2). The role of Q_2O production was taken over by a μ-channel catalytic reactor (CCR, placed upstream of the zeolite MSB storage) which oxidized the prepared Q_2 mixture using a separate O_2 flow. These experiments required two different steps. First, Q_2O production and storage in the zeolite MSB (upstream of PERMCAT) were done. As second step, Q_2O was released by heating the storage MSB (up to 400 °C) and He carrier gas transported it to the PERMCAT for detritiation. Downstream of the PERMCAT unit, at the impurity side, another MSB was placed as a water trap to capture the detritiated Q_2O. Seven experimental runs were done in this configuration.

2.3. Sensors and measurement methods

The required Q_2 gas mixtures were prepared and their specific activities were determined using p-V-T and gas chromatograph (GC) measurements. Along the experiments, different ionization chamber sensors online monitored the feed activity up- and downstream to the PERMCAT (IC1&IC4, IC3), and the feed stream between the two internal PERMCAT units (IC2). A humidity sensor (HM) was placed just between the PERMCAT unit and the small MSB trap. The indicated dew point (Q_2O partial pressure) along with the measured flow rate determined the amount of tritiated water in the gas stream.

Offline gas samples from the feed stream (either via the small MSB trap or bypassing it) were taken, using a detachable gas vessel ("sample tube" in Fig. 2). The specific activities of these offline samples were measured using an oxidation method: transforming the tritium in the sample into water via oxidization and then measuring it with liquid scintillation counting (OX measurements).

Two MSB traps (located downstream of PERMCAT unit) were used along the seven experimental runs releasing stored Q_2O from the zeolite MSB located upstream of the PERMCAT unit. One trap for six experimental runs and another exclusively for the last run

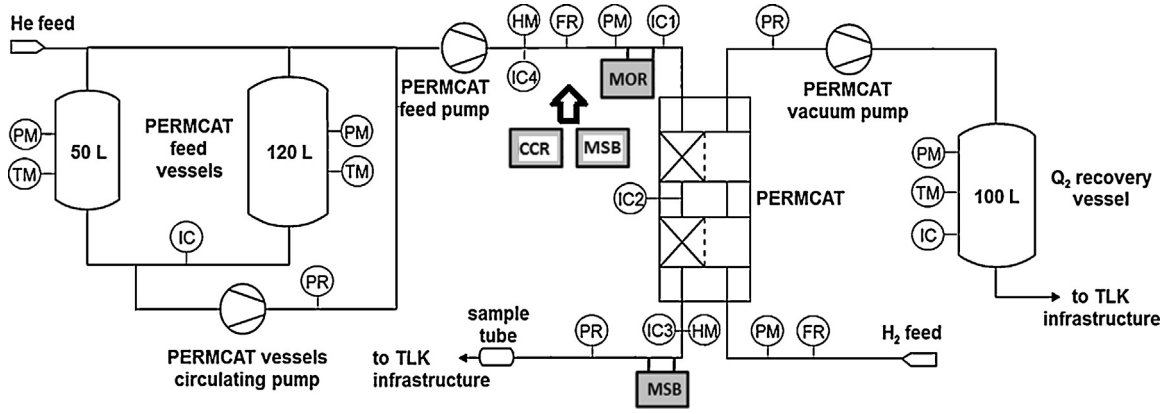

Fig. 2. Simplified schematic view of the PERMCAT process in the CAPER facility using a metal oxide reactor (MOR) and a small molecular sieve bed (MSB, downstream of PERMCAT unit) used as a Q_2O trap. The micro channel catalytic reactor (CCR) and the zeolite MSB indicate the replacement of the MOR for the second type of configuration (PM: pressure measurement; TM: temperature measurement; PR: pressure regulation; FR: flow regulation; IC: ionization chamber, HM: humidity sensor).

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