Isotopic exchange testing of a reactive molecular sieve bed mock-up for tritium recovery applications

Authors: Ana, G, Niculescu, Dr. Alina Elena, Bulubasa, Gheorghe, Craciun, Mrs. Maria Ecaterina, Bucur, Mr. Ciprian, Niculescu, Dr. Alina Elena

Date: 2026-02-03T17:19:40+00:00

Abstract

Efficient recovery of hydrogen isotopes is a critical requirement for purge gas processing systems in fusion breeder blanket applications. This study presents experimental results from isotopic exchange tests performed on a 1:10 scale Reactive Molecular Sieve Bed (RMSB) mock-up, aimed at optimizing deuterium extraction from deuterated water adsorbed on molecular sieves. Experiments were conducted using platinized zeolite with platinum loadings of 0.3, 0.5, and 0.7 wt%. After adsorption of approximately 600 g of deuterated water, isotopic exchange was initiated by flushing the heated bed with hydrogen at temperatures ranging from 25 °C to 120 °C. Deuterium concentration in the outlet stream was continuously monitored via quadrupole mass spectrometry, and extraction efficiencies were evaluated over a reference exchange period of 10 hours. Results demonstrate that both increasing temperature and higher platinum content significantly enhance isotopic exchange performance. Above 80 °C, quasi-steady exchange behaviour was observed, indicating reduced kinetic and mass-transfer limitations. From a combined efficiency-cost perspective, a platinum loading of ~0.5 wt% appears to provide an optimal compromise for practical deployment.

Full Text

Preamble

Isotopic exchange testing of a reactive molecular sieve bed mock-up for tritium recovery applications George Ana, Alina Niculescu*, George Bulubasa, Maria Craciun, Ciprian Bucur alina.niculescu@icsi.ro National Research and Development Institute for Cryogenic and Isotopic Technologies

Abstract

Efficient recovery of hydrogen isotopes is a critical requirement for purge gas processing systems in fusion breeder blanket applications. This study presents experimental results from isotopic exchange tests performed on a 1:10 scale Reactive Molecular Sieve Bed (RMSB) mock-up, aimed at optimizing deuterium extraction from deuterated water adsorbed on molecular sieves. Experiments were conducted using platinized zeolite with platinum loadings of 0.3, 0.5, and 0.7 wt%. After adsorption of approximately 600 g of deuterated water, isotopic exchange was initiated by flushing the heated bed with hydrogen at temperatures ranging from 25 °C to 120 °C. Deuterium concentration in the outlet stream was continuously monitored via quadrupole mass spectrometry, and extraction efficiencies were evaluated over a reference exchange period of 10 hours. Results demonstrate that both increasing temperature and higher platinum content significantly enhance isotopic exchange performance. Above 80 °C, quasi-steady exchange behaviour was observed, indicating reduced kinetic and mass-transfer limitations. From a combined efficiency-cost perspective, a platinum loading of ~0.5 wt% appears to provide an optimal compromise for practical deployment.

1. Introduction

Future fusion power plants such as DEMO will require robust tritium and deuterium management systems to ensure fuel cycle closure and safe operation. In a breeder blanket like Helium Cooled Pebble Bed (HCPB) purge gas systems, water vapor containing hydrogen isotopes (e.g., HDO, HTO) must be efficiently trapped and subsequently regenerated with high isotope recovery performance. Among the candidate technologies, Pt-catalysed zeolites - also referred to as reactive molecular sieves - have been identified as a promising solution for the safe handling and processing of highly tritiated water, as demonstrated in ITER-oriented developments [1].

In the reactive molecular sieve bed (RMSB) process, isotopic water is first adsorbed onto the zeolite, and isotope recovery is subsequently achieved in situ through Pt-catalysed isotopic exchange with a hydrogen purge gas. This exchange reaction can be represented as:

HQO(v)+H2(g) H2O(v) +HQ(g), where Q denotes hydrogen isotopes (D or T) In this study, a RMSB mock-up was experimentally tested using platinized zeolite Y type with a 5.1 SiO_2/Al_2O_3 mole ratio, with different platinum loadings (0.3, 0.5, and 0.7 wt%). The objective was to optimize both catalyst composition and operating conditions in order to enhance isotope recovery efficiency, supporting the application in tritium recovery processes for HCPB or WLCB breeding blanket systems. The present work specifically reports the deuterium recovery efficiency achieved under the investigated conditions using the RMSB mock-up.

2. Experimental setup and methodology

RMSB mock-up description The RMSB mock-up was designed and manufactured at a 1:10 scale with respect to the DEMO reference system, with the sizing primarily driven by the expected water trapping capacity. Since the Q2 O content in the purge gas stream of the DEMO breeding blanket is estimated to be approximately 6.1 kg/day, the mock-up was dimensioned to retain about 0.6 kg/day of isotopic water, ensuring representative loading conditions for scaled experimental investigations.

The mock-up consists of three sequential sections (figure 1 [Figure 1: see original paper]), each containing an independent zeolite bed of approx. 5 kg, with tube-and-shell heat exchangers installed between adjacent sections. This segmented configuration was adopted to reproduce the different operating modes foreseen for a DEMO-scale RMSB, namely Q2O trapping, isotope recovery via catalytic exchange, and complete regeneration of the adsorbent. The functional roles of the three sections are as follows:

First section: primary adsorption zone, where isotopic water is trapped and where partial isotopic exchange is initiated during the recovery phase.

Second section: supplementary exchange zone, intended to further enhance isotope recovery and complete the exchange process.

Third section: safety retention zone, designed to prevent the release of isotopic water vapours from the system during both adsorption and elevated-temperature recovery operation.

The configuration of all bed sections allows heating by hot gas circulation rather than electrical heating, providing improved operational reliability. To ensure similarity with DEMO operating conditions, particularly regarding gas-phase hydrodynamics during the isotopic exchange phase, the cross-sectional area of the molecular sieve bed was also scaled according to the 1:10 ratio. This approach allows the mock-up to reproduce comparable gas velocities and mass-transfer characteristics to those expected in the full-scale RMSB.

Figure 1 RMSB mock-up Testing Procedure The experimental campaign was conducted in order to evaluate the isotopic exchange performance of the Reactive Molecular Sieve Bed (RMSB) mock-up under controlled and reproducible conditions. Each test followed a well-defined sequence consisting of three main operational phases: (i) adsorption of isotopic water, (ii) isotope recovery by catalytic isotopic exchange, and (iii) regeneration of the molecular sieve bed. A reference exchange duration of 10 hours was adopted for all experiments to allow consistent comparison of recovery efficiencies across the investigated operating conditions. In the same time, this duration is considered as maximum having in view the overall duration of 24 hours for the DEMO scaled RMSB to be ready for a new adsorption cycle.

Adsorption of deuterated water vapor (loading phase) The first step of each experiment consisted of trapping a controlled amount of deuterated water on the molecular sieve bed. A stable vapor generation system was used to produce a deuterated water feed rate of approximately 30 g/h, which was mixed with a nitrogen carrier gas stream at a flow rate of 23 Nm^3/h.

The resulting humid nitrogen mixture was circulated through the RMSB mock-up, enabling the adsorption of deuterated water vapor predominantly within the first section of the zeolite bed. The loading phase was continued until approximately 600 g of deuterated water had been introduced into the system, corresponding to the design trapping capacity of the scaled mock-up. During the adsorption phase, the gas stream exiting the first bed section was continuously monitored for humidity. No detectable moisture was observed at the outlet, confirming that the first section was not fully saturated and that water vapours did not break through into the downstream sections during loading.

Isotope recovery by catalysed isotopic exchange (exchange phase) Following the adsorption step, isotope recovery was initiated by heating the top of first bed section using hot gas circulation to the target operating temperature, together with feeding the bed with a hydrogen swamping gas stream at a controlled flow rate of 0.3 Nm^3/h. This resulted in an axial temperature gradient of approximately 22oC along the RMSB first section for all temperatures envisaged for the experiments. Experiments were performed over a temperature range of 25 °C to 120 °C, in view of assessing the impact of temperature onto the deuterium extraction efficiency for each Pt content. The hydrogen purge was maintained for a constant exchange period of 10 hours.

Under these conditions, isotopic exchange took place between the HDO vapours and the hydrogen gas, according to the reaction:

HDO(v)+H2(g) H2O(v) +HD(g) The outlet gas stream from the first section was continuously analysed using a quadrupole mass spectrometer, allowing real-time measurement of the deuterium concentration released as HD. The integrated deuterium inventory recovered in the outlet stream over the 10-hour period was used to quantify the isotope recovery efficiency for each test condition (Figures 2 -5).

Molecular Sieve Bed Regeneration (Desorption Phase) After completion of the isotope exchange step, the molecular sieve bed was regenerated in order to restore its adsorption capacity for subsequent experimental cycles. Regeneration was performed by heating the entire bed to temperatures exceeding 150 °C, while flushing with dry nitrogen.

The nitrogen purge gas was characterized by an extremely low moisture content, corresponding to a dew point below −50 °C, ensuring effective desorption of residual water from the zeolite structure.

Regeneration was considered complete once the dew point of the nitrogen exiting the bed decreased below −30 °C, indicating that the molecular sieve had been sufficiently dried and reactivated for reuse.

Experimental Matrix and Repeatability The above procedure was repeated systematically for platinized zeolite samples with platinum loadings of 0.3 wt%, 0.5 wt%, and 0.7 wt%, at multiple operating temperatures. Selected experiments were repeated in order to confirm measurements and to improve the consistency and reliability of the consolidated dataset.

3. Results

The isotopic exchange experiments performed on the RMSB mock-up demonstrated effective deuterium recovery over the full investigated temperature range (25–120 °C) for all tested platinum loadings. The results show a clear dependence of exchange efficiency on both operating temperature and catalyst composition (Figures 2 –5). An increase in temperature systematically enhanced deuterium extraction, with the most significant improvement observed between 25 °C and 80 °C. At higher temperatures (80–120 °C), the process approached a quasi-steady regime, indicating reduced kinetic and mass-transfer limitations. 0.3% Pt 0.5% Pt 0.7% Pt Time [h] 0.3% Pt 0.5% Pt 0.7% Pt Time [h] Figure 2 [Figure 2: see original paper] Deuterium profile at the outlet of the first RMSB mock-up section (left) and corresponding extracted deuterium fraction (right), measured at 25 °C for different platinum loadings of the platinized zeolite 0.3% Pt 0.5% Pt 0.7% Pt Time [h] 0.3% Pt 0.5% Pt 0.7% Pt Time [h] Figure 3 [Figure 3: see original paper] Deuterium profile at the outlet of the first RMSB mock-up section (left) and corresponding extracted deuterium fraction (right), measured at 65 °C for different platinum loadings of the platinized zeolite 0.3% Pt 0.5% Pt 0.7% Pt Time [h] 0.3% Pt 0.5% Pt 0.7% Pt Time [h] Figure 4 [Figure 4: see original paper] Deuterium profile at the outlet of the first RMSB mock-up section (left) and corresponding extracted deuterium fraction (right), measured at 80 °C for different platinum loadings of the platinized zeolite 0.7% Pt 0.5% Pt 0.3% Pt 1200C Time [h] 1200C 0.3% Pt 0.5% Pt 0.7% Pt Time [h] Figure 5 [Figure 5: see original paper] Deuterium profile at the outlet of the first RMSB mock-up section (left) and corresponding extracted deuterium fraction (right), measured at 120 °C for different platinum loadings of the platinized zeolite Platinum loading also had a strong influence on performance. Higher Pt contents (0.5 and 0.7 wt%) consistently provided improved recovery compared with the reference 0.3 wt% material, confirming the catalytic role of platinum in promoting isotopic exchange. Repeated tests at selected conditions improved data consistency and resolved discrepancies observed in earlier measurements.

Overall, the results indicate that a Pt loading of approximately 0.5 wt% represents a favorable compromise between recovery efficiency and catalyst cost, while higher loadings may be advantageous when reduced operating temperature or enhanced performance is required.

4. Discussion

The experimental results confirm that the Reactive Molecular Sieve Bed (RMSB) concept provides an efficient method for isotope recovery from adsorbed deuterated water through Pt-catalysed isotopic exchange. The observed increase in recovery efficiency with temperature indicates that the process is governed by a combination of surface reaction kinetics and mass-transfer limitations within the porous zeolite structure. In particular, the quasi-steady behaviour reached above approximately 80 °C suggests that elevated temperatures significantly reduce these limiting effects, enabling faster and more complete exchange.

A systematic improvement in performance was also obtained with increasing platinum loading, highlighting the key catalytic role of Pt in promoting the exchange reaction. While the highest recovery efficiencies were achieved with 0.7 wt% Pt, the results indicate that 0.5 wt% provides a substantial enhancement compared with the reference 0.3 wt% catalyst, representing a favourable compromise between recovery efficiency and catalyst cost.

In the present experimental campaign, isotopic exchange was carried out only over the first bed section, without switching the process flow to the second section. This operational choice was motivated by the different recovery behaviours observed for the investigated catalysts. For the reference 0.3 wt% Pt material, achieving higher recovery efficiencies would require exchange durations longer than the 10- hour reference period. In contrast, for the 0.5 and 0.7 wt% Pt catalysts, recovery efficiencies exceeded 90% within the same exchange time, making the use of additional bed sections unnecessary under the tested conditions.

An important feature of the mock-up configuration and hot-gas heating approach is that it enabled the establishment of a longitudinal (axial) temperature gradient along the first bed section. Bed temperatures were monitored at multiple axial positions, confirming representative thermal profiles during operation. Such longitudinal gradients have also been identified as beneficial in ITER-oriented engineering-scale Pt–zeolite columns, where enhanced isotope exchange factors were achieved through improved water vapour redistribution and catalyst utilization [1–3].

Overall, the present mock-up results provide a solid experimental basis for further scale-up toward DEMO-relevant RMSB operation. Future work should focus on extended cycling behaviour, optimization of thermal management strategies, and integration of the RMSB process within breeder blanket tritium extraction and recovery systems.

5. Conclusions

The experimental campaign performed on the 1:10 scale RMSB mock-up successfully consolidated the isotopic exchange performance of platinized zeolite beds for isotope recovery from adsorbed deuterated water. Tests carried out over a temperature range of 25–120 °C and with platinum loadings of 0.3, 0.5, and 0.7 wt% demonstrated that the isotopic exchange process is effective under all investigated conditions.

Both increasing operating temperature and higher Pt catalyst content led to systematically improved deuterium recovery efficiencies. The strongest enhancement was observed up to approximately 80 °C, above which the process approached a quasi-steady regime, indicating reduced kinetic and mass- transfer limitations. From a practical perspective, a Pt loading of around 0.5 wt% appears to provide a favorable balance between performance gains and catalyst cost. The experiments also demonstrated the feasibility of hot-gas heating, which generated a longitudinal temperature gradient along the bed, representative of thermal management strategies envisaged for larger-scale RMSB systems. These results provide a strong foundation for future scale-up activities toward DEMO-relevant tritium recovery applications, including extended cycling tests, optimized thermal control, and system-level integration within breeder blanket tritium extraction and recovery architectures.

References

[1] W. M. Shu, I. Cristescu, R. Michling, D. Demange, R. S. Willms &M. Glugla - Highly Tritiated Water Processing by Isotopic Exchange, Fusion Science and Technology, Vol. 67, Issue 3, pg. 563-566, 2015, https://doi.org/10.13182/FST14-T80 [2]Robert Michling, Adalbert Braun, Ion Cristescu, Helmut Dittrich, Manfred Glugla, Nando Gramlich, Nancy Lohr, Wataru Shu &Scott Willms - Capture and Isotopic Exchange Method for Water and Hydrogen Isotopes on Zeolite Catalysts up to Technical Scale for Pre-Study of Processing Highly Tritiated Water, 483-486, https://doi.org/10.13182/FST14-T60 [3] Ion Cristescu, Mirela Draghia - Developments on the tritium extraction and recovery system for HCPB, Fusion Engineering and Design, Volume 158, 111558, 2020, https://doi.org/10.1016/j.fusengdes.2020.111558

Acknowledgments

Technology 67, April Science Fusion Pages 2015, This work has been carried out within the framework of the EUROfusion Consortium, funded by the European Union via the Euratom Research and Training Programme (Grant Agreement No 101052200 –EUROfusion). Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Commission. Neither the European Union nor the European Commission can be held responsible for them.

This work was conducted using infrastructure funded by the European Union via POC/448/1/1 and PCID- IF/315/PCIDIF_P1/OP1/RSO1.1/PCIDIF_A11 Programs, Grant No. 127931 & 323091 -Extinderea PESTD pentru dezvoltarea de aplicatii de cercetare-dezvoltare în domeniul tritiului (TRI-Valcea).

Note: Figure translations are in progress. See original paper for figures.