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Experimental and simulation study on adsorption of hydrogen isotopes on MS5A at 77 K

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Abstract

Extraction of tritium from the sweep gas of the ITER Helium Cooled Pebble Bed (HCPB) Test Blanket Module is proposed to be carried out in a two-step process: trapping of water in a cryogenic cold trap, and adsorption of hydrogen isotopes (H_2, HT, T_2) in a Cryogenic Molecular Sieve Bed (CMSB) at 77 K. A CMSB mock-up in a semi-technical scale (1/6 of the flow rate of the ITER-HCPB) was designed and constructed at the Forschungszentrum Karlsruhe. In this work, the adsorption isotherms of hydrogen and deuterium on the MS5A adsorbent were investigated by the volumetric method. The correlation of the experimental adsorption isotherms of hydrogen and deuterium on the MS5A adsorbent were performed. To validate the cryogenic adsorption process, breakthrough experiments were performed using the CMSB mock-up. The experimental breakthrough curves obtained were analyzed with model mass balance equations, and scale-up numerical simulations were carried out.

Keywords: Tritium; Adsorption; Sweep gas; Liquid nitrogen; Fuel cycle

1. Introduction

To validate design concepts of tritium breeding blankets used for the generation of fuel in fusion machines such as ITER and DEMO, experimental investigation and tests of these concepts are necessary. According to the design concepts, ceramic breeder materials such as lithium orthosilicate generate tritium via nuclear reaction between the neutrons and the lithium atoms. The tritium produced in the breeder material has to be removed by means of a Tritium Extraction System (TES) in which a helium sweep gas containing 0.1% of hydrogen is used to assist the tritium release from the breeder materials via isotope exchange reactions. The TES foreseen for the ITER Helium Cooled Pebble Bed (HCPB) Test Blanket Module (TBM) will

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Nomenclature

a surface area of adsorbent particle per unit volume of packed bed, $a = 3(1-\varepsilon)/R_p$ (m^{-1})
$\bar{a}_i$ partial molar surface area (m^2/mol)
b_i Henry's law constant (mol/g Pa)
c adsorbate concentration in gas phase (mol/m^3)
c^* concentration in gaseous phase equilibrated with adsorbate concentration in solid phase (mol/m^3)
D diffusivity in gas phase (m^2/s)
D_L axial dispersion coefficient (m^2/s)
D_P effective pore diffusivity (m^2/s)
ΔG_i° standard Gibbs energy for adsorption (J/mol)
k_g mass transfer coefficient at fluid film (m/s)
n_i^s number of moles in surface phase (mol/g)
$n_i^{s,\infty}$ maximum number of moles of i in surface phase (mol/g)
n_m^s total number of moles of mixture in surface phase (mol/g)
$n_m^{s,\infty}$ maximum total number of moles of mixture in surface phase (mol/g)
P total pressure (Pa)
p_i partial pressure of i in vapor phase (Pa)
Pe Peclet number ($=2R_p u/D_L$)
q adsorbate concentration in solid phase (mol/g)
$\bar{q}$ adsorbate concentration in solid phase averaged over a particle (mol/g)
r length in radial direction in adsorbent particle (m)
R gas constant (=8.31) (J/(mol K))
R_P radius of adsorbent particle (m)
Re Reynolds number ($=2R_P u\rho/\mu$)
Sc Schmidt number ($=\mu/(\rho D)$)
Sh Sherwood number ($=2R_P k_g/D$)
t time (s)
T temperature (K)
u superficial gas velocity (m/s)
x_i molar fraction in vacancy-free adsorbed phase
y_i molar fraction of vacancy-free vapor phase
z length in axial direction in adsorption column (m)

Greek symbols
α_{ij} NRTL parameter
γ packed bed density (g/m^3)
γ_i^s activity coefficient in adsorbed phase
ε void fraction of packed bed
θ fractional coverage
Λ_{ij} Wilson parameters for interaction between i and j
μ viscosity (g/(m s))
π_i spreading pressure (N/m)
ρ density of gas (g/m)
ρ_s apparent density of adsorbent particle ($\rho_s = \gamma/(1-\varepsilon)$) (g/$m^3$)
τ_{ij} NRTL parameter
ϕ_i fugacity coefficient in gas phase

Subscript
i number of component (1: H_2, 2: D_2 and 3: vacancy in binary adsorption system)

remove all tritiated species present in the He sweep gas in a two-step process such as (1) trapping of water in a cryogenic cold trap and (2) adsorption of all hydrogen isotopes (H_2, HT, T_2) in a Cryogenic Molecular Sieve Bed (CMSB) operating at liquid nitrogen temperature [1,2]. In a first series of experiment, the efficiency of a semi-technical scale cold trap (1/6 of the ITER operating conditions) has been demonstrated, which can remove efficiently water vapor from the He stream [3,4]. Then, the second part of the extraction process for removal of all hydrogen isotopes was validated. A CMSB mock-up in a semi-technical scale (1/6 of the flow rate of the ITER-HCPB) has been designed and manufactured in the Forschungszentrum Karlsruhe and was installed in an experimental rig at the Tritium Laboratory Karlsruhe (TLK). The adsorbent container filled with 20 kg of molecular sieve (MS5A) was arranged inside a liquid nitrogen-cooled vacuum-isolated vessel. During the operation of the bed, the MS5A adsorbent is uniformly cooled down to 77 K. To comply with the ITER requirements and to