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Experimental study of binary mixture permeation of hydrogen and helium in nanocomposite MFI-alumina membrane for tritium processes

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Introduction

The nuclear reaction between hydrogen's isotopes deuterium and tritium is the most effective reaction for future fusion power plants. Tritium being very rare, it will be produced via ^{6}Li (n, α) within the breeder blanket to achieve self-sufficiency. In the solid blanket concept, the bred tritium is extracted using a helium stream of about 10^4 m^3/h doped with 0.1% H_2. Most of the tritium recovery processes developed up to now are based on conventional technologies using condensation and adsorption/desorption techniques [1]. Alternatively, the Tritium Laboratory Karlsruhe (TLK) proposes a combination of catalytic membrane reactor PERMCAT with a pre-concentration stage using zeolite membranes. This would ensure a simple and fully continuous operation and smallest achievable tritium inventory [2].

Single gas tests with helium and hydrogen (replacing radioactive tritium) through MFI-alumina hollow fibre have been performed [3]. Since the breeder blanket stream contents only traces of tritium, the effect of feed composition on the separation is a key issue. This paper presents the first results of permeation experiments obtained using mixtures of H_2 and He.

Experimental

The nanocomposite MFI-alumina hollow fibre is prepared by IRCELYON via the pore-plugging synthesis of a porous asymmetric support (15 cm-long, o.d. 1.65 mm, i.d. 1.44 mm). The surface area of the membrane is 4.97 cm^2. The equivalent thickness of zeolite layer is less than 1 μm [4].

At first the hollow fibre is conditioned by heating at 400°C under vacuum during 6 h to remove any species adsorbed in the pores. During the experiment, He and H_2 are mixed dynamically via separated mass flow controllers and fed inside the hollow fibre. The permeate flow is measured by digital mass flow meter. The pressures at both sides can be adjusted using needle valves. The gas composition at the permeate side is analyzed using a quadrupole mass spectrometer. The pressure on the permeate side is kept constant at 500 mbar using vacuum pump. The membrane module is thermostated in a cylindrical oven from room temperature up to 400°C. The permeance Π^i [$mol/m^2 \cdot s \cdot Pa$] of component the component "i" is calculated according to following equation

$$\Pi^i = \frac{P \cdot y_p^i}{A \cdot (p_r \cdot y_r^i - p_p \cdot y_p^i)}$$

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where y_p and y_r are the molar fractions [no dim] in the permeate and retentate, respectively; A [m^2] is membrane area; p_p and p_r [Pa] are the total pressures in the permeate and retentate, respectively; P [mol/s] is the total permeate flow.

Results and discussion

The observed permeances of hydrogen and helium as a function of temperature are presented in Fig. 1 for single gases and for mixtures with 20 and 50 %vol of H_2. The permeance of hydrogen increases significantly at decreasing of hydrogen concentration in feed from 50 to 20 %vol. in the mixture. This makes the membrane attractive for breeder blanket applications. As expected from results reported in previous study for single gases [3], the permeance of hydrogen decreases with a temperature for both mixtures. At the same time the permeance of helium is practically independent from feed mixture composition and temperature and remains nearly constant.

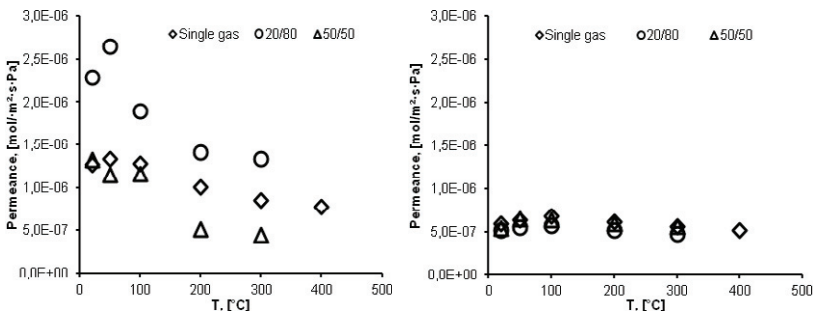

Fig. 1. Permeance of hydrogen (left) and helium (right) for single gases and mixtures as a function of temperature.

Conclusions and perspectives

For the first time the permeance of H_2 - He mixtures through a MFI-alumina hollow fibre membrane has been measured for different compositions and temperatures. Such highly permeable membrane, although it shows a limited selectivity, appears attractive for tritium recovery in the blanket. This will imply its operation in a permeation cascade, for which simulation work is ongoing. Next experimental campaigns will explore lower hydrogen concentrations and study the influence of water vapour in the feed stream.

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