

H₂/He and H₂O/He Separation Experiments with Zeolite Membranes for Nuclear Fusion Applications

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Abstract : In future nuclear fusion reactors, tritium self-sufficiency will be ensured by tritium (3H) production via reactions between the fusion neutrons and lithium. To favor tritium breeding, a neutron multiplier must also be used. Both tritium breeder and neutron multiplier will be placed in the so-called Breeding Blanket (BB). For the European Helium-Cooled Pebble Bed (HCPB) BB concept, the tritium production and neutron multiplication will be ensured by neutron bombardment of Li₄SiO₄ and Be pebbles, respectively. The produced tritium is extracted from the pebbles by purging them with large flows of He (~ 104 Nm³h⁻¹), doped with small amounts of H₂ (~ 0.1 vol%) to promote tritium extraction via isotopic exchange (producing HT). Due to the presence of oxygen in the pebbles, production of tritiated water is unavoidable. Therefore, the purging gas downstream of the BB will be composed by Q₂/Q₂O/He (Q = 1H, 2H, 3H), with Q₂/Q₂O down to ppm levels, which must be further processed for tritium recovery. A two-stage continuous approach, where zeolite membranes (ZMs) are followed by a catalytic membrane reactor (CMR), has been recently proposed to fulfil this task. The tritium recovery from Q₂/Q₂O/He is ensured by the CMR, that requires a reduction of the gas flow coming from the BB and a pre-concentration of Q₂ and Q₂O to be efficient. For this reason, and to keep this stage with reasonable dimensions, ZMs are required upfront to reduce as much as possible the He flows and concentrate the Q₂/Q₂O species. Therefore, experimental activities have been carried out at the Tritium Laboratory Karlsruhe (TLK) to test the separation performances of different zeolite membranes for H₂/H₂O/He. First experiments have been performed with binary mixtures of H₂/He and H₂O/He with commercial MFI-ZSM5 and NaA zeolite-type membranes. Only the MFI-ZSM5 demonstrated selectivity towards H₂, with a separation factor around 1.5, and H₂ permeances around 0.72 μmolm⁻²s⁻¹Pa⁻¹, rather independent for feed concentrations in the range 0.1 vol%-10 vol% H₂/He. The experiments with H₂O/He have demonstrated that the separation factor towards H₂O is highly dependent on the feed concentration and temperature. For instance, at 0.2 vol% H₂O/He the separation factor with NaA is below 2 and around 1000 at 5 vol% H₂O/He, at 30°C. Overall, both membranes demonstrated complementary results at equivalent temperatures. In fact, at low feed concentrations (≤ 1 vol% H₂O/He) MFI-ZSM5 separates better than NaA, whereas the latter has higher separation factors for higher inlet water content (≥ 5 vol% H₂O/He). In this contribution, the results obtained with both MFI-ZSM5 and NaA membranes for H₂/He and H₂O/H₂ mixtures at different concentrations and temperatures are compared and discussed.

Keywords : nuclear fusion, gas separation, tritium processes, zeolite membranes

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