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Simulation of binary gas separation through multi-tube molecular sieving membranes at high temperatures

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HIGHLIGHTS

- A model was validated for a multi-tube membrane module for binary gas separation.
- H_2 driving force is greatly affected by process conditions and module design.
- The radius of the module influences significantly on H_2 purity and recovery.
- Similar counter and co-current flows due to high gas-through-gas diffusion.

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ABSTRACT

An experimentally validated theoretical model was developed to investigate the influence of operating conditions on the performance of a multi-tube membrane module containing cobalt oxide silica (COxS) membranes with molecular sieving properties. The model investigated the separation process for a binary gas mixture consisting of H_2 and Ar at 400 °C. Engineering parameters such as feed flow rate, feed pressure, module size and flow configuration were systematically varied in order to optimise the separation performance promoting three main goals: H_2 yield, H_2 purity and H_2 recovery. Changing these parameters led to different flows and H_2 fractions in the feed domain, thus altering the driving forces for the preferential permeation of H_2. The simulated results suggest that gas separation was greatly improved by reducing the module radius which meets all of the three aforementioned optimisation criteria. Interestingly, increasing the feed flow rate and feed pressure were found to be beneficial but the former led to lower H_2 recovery whilst the latter did not deliver the same purity when compared to lower feed pressure. In addition, two flow configurations, counter-current and co-current, were compared. It was observed that the results of counter-current were effectively the same as the co-current. This was attributed to the high gas-through-gas diffusion for high-temperature membrane operation. Finally, neglecting diffusion effects, or considering advection only, leads to over prediction of H_2 permeate molar fraction.

1. Introduction

Hydrogen is widely accepted as a clean energy carrier, particularly in fuel cell deployment, whereby reaction with oxygen from air generates water, an environmentally benign emission. Conventional gas separation technologies using organic membranes require low temperature operation due to the poor thermo-stability and anti-oxidation properties of polymers. As H_2 is often generated at high temperatures by natural gas reforming or coal gasification [1], separation of H_2 attracts energy penalties due to the required cool down of high temperature streams to meet the operating requirements of polymeric membranes. Inorganic membranes derived from ceramics, silica or metal alloys [2] and zeolites [3,4] can be employed in high temperature gas separation due to their thermo-stability and good resistance to chemical attack. Molecular sieving membranes for H_2 separation are quite promising due to superior performance at elevated temperatures [5,6], especially those derived from metal oxide silica matrices [7]. Therefore, there has been a major concerted effort of the research community to improve the gas separation performance by primarily focusing on membrane synthesis.

However, membrane performance is intrinsically linked to the operating conditions and design of membrane modules, which play a significant role in gas separation processes. It is well known that higher driving forces are favourable for the efficient operation of membrane systems in most cases. Moreover, operating conditions and the membrane module design (size and flow configuration)

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