COMPARISON BETWEEN FLUIDIZED BED AND PACKED BED MEMBRANE REACTORS FOR H2 PRODUCTION – EFFECT OF MASS TRANSFER LIMITATIONS

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Summary

In this paper hydrogen production via methane steam reforming carried out in membrane reactors has been studied. A direct comparison between the performances of packed bed and fluidized bed membrane reactors is proposed. In particular, the effects of mass transfer limitations on the membrane area required in the different membrane reactors have been evaluated with detailed models by using different correlations. It has been found that the extent of bubble-to-emulsion phase mass transfer limitations increases by increasing the reactor length. On the other hand, bed-to-wall mass transfer limitations are the major mass transfer resistances in packed bed membrane reactors and they increase by increasing the membrane permeation flux. The comparison shows that the fluidized bed membrane reactor requires lower membrane area than the packed bed membrane reactor for a given conversion and hydrogen recovery.

Keywords

Membrane reactors, Hydrogen production.

Introduction

The recent advances in Polymer Electrolyte Membrane Fuel Cells (PEMFC) for small or medium scale applications make the production of ultra-pure hydrogen a challenging topic in energy conversion. On an industrial scale, most of the hydrogen is currently produced via steam reforming of methane (SRM).

A high degree of process integration and process intensification can be accomplished by integrating hydrogen perm-selective membranes in the steam reformer (1,2). Via the integration of hydrogen perm-selective membranes, the number of process units can be strongly decreased and the total required reactor volume can be largely reduced, while higher methane conversions and hydrogen yields beyond thermodynamic equilibrium limitations can be achieved, at lower temperatures and with higher overall energy efficiencies (3-6).

The use of both packed bed membrane reactors (7) and fluidized bed membrane reactors (8,9) has already been presented in literature for the reforming of methane and pros and cons of both concepts have already been discussed. In this paper a direct comparison between the two concepts has been performed for ultra-pure hydrogen production via methane reforming using detailed theoretical models. The extent of mass transfer limitations in the different reactors have been evaluated, and strategies to decrease (or avoid) these limitations have been proposed. A comparison with an ideal isothermal plug flow membrane reactor has been carried out for both reactor configurations.

Reactor configuration

Fluidized bed membrane reactor concept

A schematic representation of the fluidized bed membrane reactor configuration considered is reported in Fig. 1. Pure hydrogen is recovered via Pd-based membranes inserted into the fluidized bed. With the fluidized bed membrane reactor a virtually isothermal condition can be achieved and bed-to-membrane mass transfer limitations are largely avoided. On the other hand, bubble-to-emulsion phase mass transfer limitations and the extent of gas back-mixing could deteriorate its performance. In particular, the use of membranes inside the reactor could decrease the extent of back-mixing and can also help decreasing the bubble diameter, enhancing the bubble-to-emulsion phase mass transfer. With the help of a two-phase phenomenological reactor model, the effect of bubble-to-emulsion phase mass transfer limitations and gas back-mixing have been quantified. Different correlations have been tested and the effects of bubble-to-emulsion phase mass transfer limitations on reactor performances have been studied.

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Packed bed membrane reactor concept

The typical tube-in-tube packed bed membrane reactor configuration was considered (see Fig. 2).

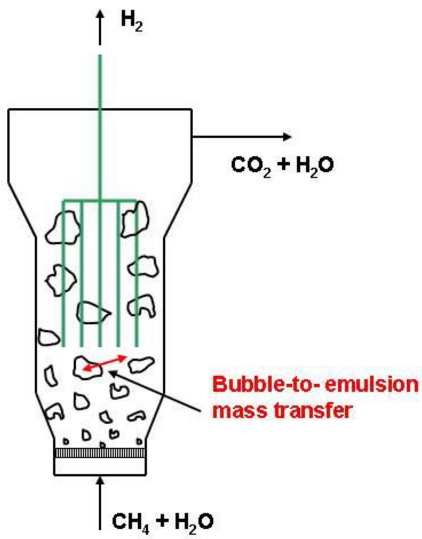

Fig. 1 Typical fluidized bed membrane reactor scheme

The reactor has been studied with a detailed 2D model in order to identify the extent of bed-to-membrane mass transfer limitations (concentration polarization) and their effect on reactor performance. The influence of the reactor and particle dimensions has been investigated.

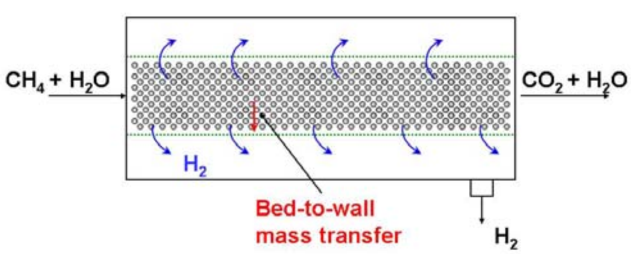

Fig. 2 Typical packed bed membrane reactor scheme

Results and discussion

Fluidized bed membrane reactor concept

Due to the low pressure drop in fluidized bed reactors, small catalyst particles can be used and intra-particle mass transfer limitations are circumvented. On the other hand the good mixing in the emulsion phase make the catalyst to membrane mass transfer limitations unimportant in fluidized bed membrane reactors. The important mass transfer limitations occurring in a fluidized bed membrane reactor are the thus the only bubble-to-emulsion phase mass transfer limitations.

As a matter of fact, the bubble-to-emulsion phase mass transfer limitation increases with increasing bubble diameter, which itself increases by increasing the reactor length.

The longer is the reactor, the bigger are the bubbles and the greatest are the bubble-to-emulsion mass transfer limitations.

As an example, a direct comparison between a 1.5 m long ideal isothermal packed bed reactor and a similar long fluidized bed show that, in case of bubble-to-emulsion phase mass transfer limitations calculated as fluidized bed without internals, the fluidized bed membrane reactor requires up to 2.5 times the membrane area required for a given conversion of 97%.

Different correlations for bubble rise velocity and bubble diameter have been tested in the fluidized bed model and the results will be discussed. The effect of H2 extraction on the bubble diameter has also been considered. A simple strategy for decreasing bubble-to-emulsion phase mass transfer limitation in the fluidized bed will be also discussed and validated through modelling.

Packed bed membrane reactor concept

A big difference between a packed bed and a fluidized bed is the mass transfer limitations between the catalytic bed and the membrane wall which was of no importance for fluidized bed but which plays a key role in the packed bed membrane reactor.

For packed bed membrane reactors the extent of bed-to-wall mass transfer limitations (also called concentration polarisation) has been evaluated with the 2D model considering different dispersion correlations and porosity models. The effects of operating conditions on the extent of concentration polarisation have been evaluated and the results in terms of membrane area required for a given conversion have been reported.

It has been demonstrated that, depending on the operating conditions, a packed bed membrane reactor requires more that 2 times the ideal membrane area as a consequence of mass transfer limitations.

It is not surprising that the effects of bed-to-wall mass transfer limitations increase by increasing the hydrogen membrane flux. The study suggests that better membranes in terms of higher permeation fluxes require an appropriate reactor concept to avoid the detrimental effects of mass transfer limitations.

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