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Modelling and experimental studies of a water–gas shift catalytic membrane reactor

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HIGHLIGHTS

- A 2D-axisymmetric CMR model was developed using *FLUENT*.
- There was close agreement between the model and the prototype.
- Loading extra catalyst would lead to temperatures beyond recommended ranges.
- Reducing permeation side pressure is a key factor to maximise the CO conversion levels.
- The permeation pressure of the CMR defines the maximum limit of H_2 yield.

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ABSTRACT

A 2-dimensional, axis-symmetric CFD model of a tubular CMR has been developed using a commercial software package *FLUENT* for the purposes of guiding the design and operation of a HTWGS-CMR for the processing of coal-derived syngas. Development of the model has been approached in a stepwise manner through the successive incorporation of sub-models for the CMR processes. For each step, performance of model was checked and validated against measurements using a prototype CMR with same set-up applied in simulation.

The optimum catalyst loading, which yield the maximum CO conversion within targeted operating reactor temperatures (350–450 °C), was found to be 11.6 kg/(CO_{mol}/s) for the inlet syngas temperature of 350 °C with a reactor having a 1″ shell diameter. The CMR model was validated experimentally with a simulated coal-derived syngas (64.5% of CO, 33.0% of H_2 and 2.5% of CO_2 with a 3:1 steam to carbon (S:C) ratio) at a total dry gas flow of 4 L_N/min and a feed pressure of 15 bar_g. These tests were performed using a prototype reactor which incorporated with a tubular (0.1 mm thick, 150 cm^2, 3/8″ OD) Pd/Ag23 wt% membrane. The CMR model was simulated using a wider range of operating parameters (namely permeation rate, inlet temperature, catalyst loading, pressure at permeate side and S:C ratios) to examine its sensitivity to these variables. Outcomes of these parametric analyses have enhanced our understanding of CMR operation in order to optimise its performance.

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1. Introduction

Integrated Gasification Combined Cycle (IGCC) with pre-combustion CO_2 capture offers a means of reducing the CO_2 emissions associated with coal-based power systems. The commercialisation barrier for IGCC, however, is its high capital cost. One means of reducing the capital cost is by process intensification, whereby the water-gas shift (WGS) process and CO_2 separation process are combined. In a conventional system, CO from the syngas is converted into CO_2 via two separate WGS reactors: most of the CO in the syngas is shifted within a high temperature WGS reactor and the small amount of CO left is shifted in a low temperature WGS reactor where operating conditions favour higher equilibrium CO conversion. Both WGS reaction processes and the CO_2/H_2 separation process can be combined in a single catalytic membrane reactor (CMR) using a high-temperature WGS catalyst to achieve CO conversion levels higher than that of the two-step WGS reactor configuration. A previous study demonstrated that WGS conversions of >99% can be achieved at 400 °C compared to the equilibrium value of ~94% for coal-derived syngas with a 3:1 steam to carbon (S:C) ratio [1]. This is explained by the continuous removal of one of the reaction products through the selective membrane which drives the equilibrium of the WGS reaction (Eq. (1)) to the right

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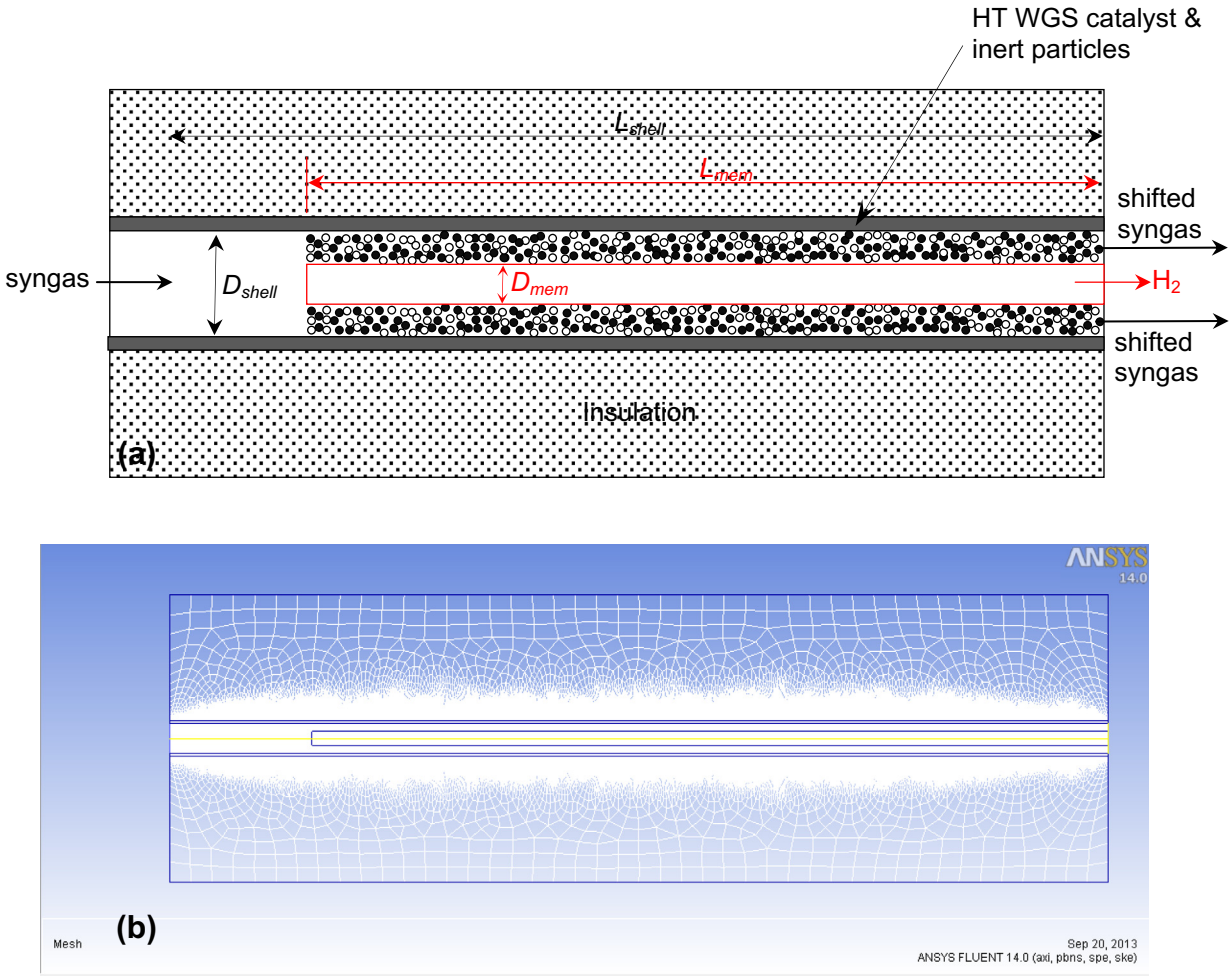

Fig. 1. (a) Schematic diagram of catalytic membrane reactor used in this study. (b) Geometry and mesh of CMR used for 2D-axisymmetric model.

$$CO\ (g) + H_2O\ (g) \leftrightarrow CO_2\ (g) + H_2\ (g)\ \Delta H_{298K} = -41.1\ \text{kJ mol}^{-1} \quad (1)$$

Successful integration of the WGS reaction and the gas separation process into a single unit requires an understanding of heat and mass transfer processes, catalytic WGS reaction rates and H_2 permeation across the membrane under different operating conditions to maximise the advantages of the CMR concept. Mathematical modelling is an effective tool for the design, evaluation and optimisation of any reactors and reaction processes. In the past two decades, a significant number of numerical models have been used to investigate the performance of CMRs [2–29]. Among them, most studies focused on the catalytic water-gas shift reaction inside the CMRs. Some studies modelled for the partial oxidation and catalytic steam-reforming of methane [20–26]. Some developed their models to study the performance of the catalytic membrane reactor for the dehydrogenation of ethylbenzene to styrene [27–29]. Previous models included those with very simplified assumptions that use one dimensional, isothermal and isobaric conditions at steady state excluding the kinetics of the WGS reaction rate [2], an extension to include the pressure drop along the reactor [3,4] and kinetics of the WGS reaction [5]. Other CMR models were relatively more complex taking into account the effects of heat and mass transfer, heat loss, and WGS kinetics but still in one-dimensional in space without considering the radial gradients [6–9]. One-dimensional models can provide fast simulation and quick results with reasonable accuracy compared to two dimensional models [10,11]. Two-dimensional, 2D-axisymmetric or even 3D models, however, are more suitable for H_2 permeation through the membrane in CMRs as this process is truly two-dimensional. Therefore, recent CMR models and membrane module models include the effects of mixing and heat/mass transfer in the radial direction [12–17,30]. Most of the CMR models were developed to study the reactor performance at steady state conditions, whereas only a few of the CMR models were designed to investigate the dynamic behaviour of the CMR [18,19,26].

Over the last decade, CFD modelling with higher space dimensions (i.e., 2D-axisymmetric, 3D) coupled with a detailed understanding of the mechanism and kinetics of the catalytic WGS shift reaction together with mechanisms of H_2 permeation across the membrane has been one of the key tools in understanding of insights into the flow field and temperature profiles within the CMRs that will lead to improving and optimising of its performance. Basile et al. [12] developed a 2D CMR model to investigate the effect of co-current and counter-current flow configurations of CMR. Markatos et al. [13] used *PHOENICS CFD* software to investigate the role of mass and heat dispersion effects on the performance of a large-scale CMR module. Chiappetta et al. [14] investigated the sensitivity of their 2D CMR model in order to define the role of some variables on the performance of a membrane rector for maximising the system efficiency. Chen et al. [15] used a commercial CFD software, *FLUENT*, to develop their WGS reactor model to investigate the performance of both high-temperature and low-temperature WGS reactions. It has to be noted that the H_2 separation process was not included in their model. Byron Smith and Muruganandam [16] also used *FLUENT*

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