Reactor modeling and transient behavior of a power-to-gas system based on high temperature electrolysis and CO_2 methanation

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Highlights

- 1D fixed bed reactor model for CO_2 methanation.
- Thermal integration between SOEC and methanation leading to high efficiency (≈85%)
- CO_2 should partly bypass the first reactor to avoid hot spot

1. Introduction

In order to balance the intermittent electricity production from renewables as wind and photovoltaic, carbon dioxide methanation for synthetic (or substitute) natural gas (SNG) production through the "power-to-gas" pathway (P2G) recently gained huge interest. The CO_2 hydrogenation reaction involves hydrogen produced via electrolysis (exploiting the electricity surplus) and carbon dioxide coming from industry, air capture or biogas upgrading processes [1]. The integration between high temperature steam electrolysis through solid oxide cells (SOEC) and methanation reaction seems to be very promising due to the strong thermal integration between the exothermal methanation reaction (Δh=-165 kJ/mol) and steam generation (required before SOEC-based electrolysis).

In this work the process modeling of SOEC-based power-to-gas system and its intermittent functioning are presented. The design of a fixed bed ([2]) methanation reactor (shell and tube configuration) is discussed. Intermittent electric input implies modular functioning: the transient behavior of the two sections is useful to evaluate the outlet SNG composition time profile.

2. Methods

The chosen methanation concept consists in two cooled reactors with an intermediate condensation stage for the produced water removal. A one-dimensional heterogeneous reactor model has been considered. The axial coordinate has been discretized: the overall control volume is thus divided into finite cylindrical volumes. Transport phenomena between catalyst pellet and gas have been considered. Internal mass transfer has been taken into account through the effectiveness factor [3]. Thermal integration between electrolysis and methanation section has been carried out in order to increase the overall electricity-to-SNG efficiency.

Transient behavior of both electrolysis and methanation section has been considered in order to evaluate the time required by the system to ensure the proper SNG outlet composition (fulfilling the pipeline prescriptions). Moreover, also the catalyst deactivation over time should be taken into account in order to evaluate the final methane concentration within the synthetic gas.

3. Results and discussion

One of the most critical aspects is the temperature reached by the catalyst within the first reactor. The methanation reaction is strongly exothermal: even though the reactor is cooled, an initial temperature increase will take place in the first part of the fixed bed, when the reactants concentration is higher. To control the maximum temperature, the ratio between the tube and the catalyst particle can be reduced down to 10, i.e. the lowest suggested value in order to avoid channeling phenomena within the catalytic bed.

A possible option to moderate the pellet temperature increase is a split of the inlet carbon dioxide flow: only a fraction (named χ) of the total CO_2 enters the first reactor. χ has been set equal to 0.7.

Bulk gas temperature reaches its maximum when the heat removed by the cooling fluid is equal to the heat generated through the exothermal reaction, according to the energy balance. As expected, pellet temperature is greater than bulk gas value. In addition to the steam generation via the reactor cooling, a further thermal integration between hot and cold streams enhance the achievable efficiency up to 86% (HHV-based).

The as designed system has been integrated with a renewable fluctuating production scenario (Germany, 2050). The intermittent and partial-load functioning of both electrolysis and methanation section suggest the presence of a H_2 buffer for the temporary storage and to balance the mismatch between instantaneous production and demand.

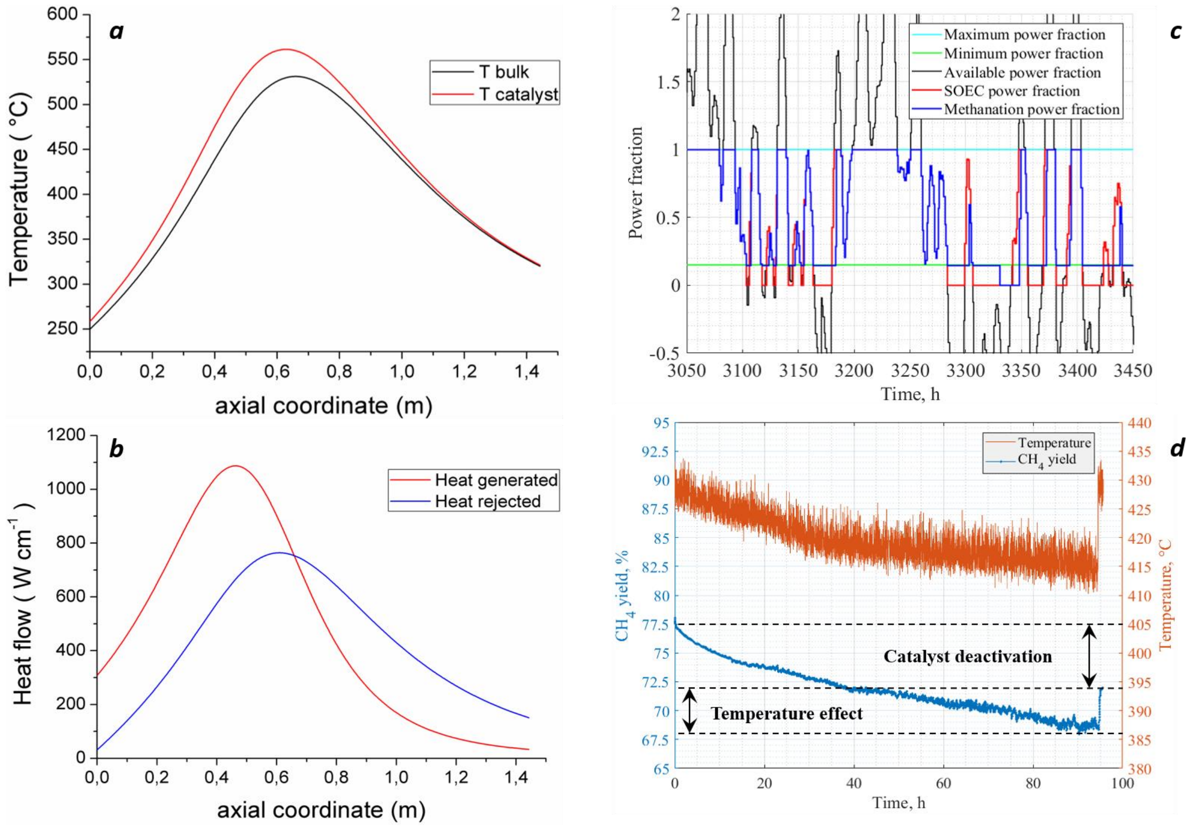

Figure 1. a) temperature profile along the first reactor, assuming a CO_2 bypass factor equal to 30%; b) generated and rejected heat over the first reactor; c) intermittent functioning of the system integrated with a forecast for renewable production in Germany in 2050; d) CH4 yield drop over 100 hours due to the combined effect of decreasing temperature and proper catalyst deactivation.

4. Conclusions

A model for the design of a multi-tubular cooled reactor has been developed. Results showed that the main concern is related to the thermal management of the reactor the temperature increase due to the exothermal reaction is not easily controllable, even with a cooling system with high heat transfer coefficient. The integrated process is characterized by an external energy input of ≈ 2.7% of the electrolysis power, such a strong thermal match can lead to high electricity-to-SNG conversion efficiency.

References

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Keywords
CO_2 methanation, solid oxide electrolysis cell, reactor modeling, system integration.