DESIGN OF AN ACTIVELY-COOLED SABATIER REACTOR FOR THERMOCATALYTIC HYDROGENATION OF CO_2: MODEL-BASED FEASIBILITY ANALYSIS AND EXPERIMENTAL PROOF-OF-CONCEPT

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Abstract

Converting CO_2 into renewable natural gas (RNG) via thermocatalytic hydrogenation is an attractive pathway to reduce our dependence on fossil fuels. The required H_2 can be generated via water electrolysis using renewable electricity (hydro, wind, and solar). However, the process is highly exothermic and thermal management is a major drawback as efficient heat removal is required to drive CH_4 formation and suppress catalyst deactivation by coking. In this study the heat-exchanger type actively-cooled Sabatier reactor was investigated by numerical simulations to assess the techno-economic feasibility. Experimental proof-of-concept of a lab-scale Sabatier reactor cooled by compressed air was successfully conducted. CO_2 conversion above 90% was achieved with 100% selectivity to CH_4 production.

Keywords

CO_2 hydrogenation, Sabatier reactor, thermal management, catalyst deactivation.

Introduction

Converting CO_2 into synthetic fuels is an attractive pathway to decrease CO_2 emissions and to reduce our dependence on fossil fuels. Thermocatalytic hydrogenation provides advantages of fast reaction rates and high conversion efficiencies, thus allowing for compact, high-throughput operation [1]. The required H_2 can be generated via water electrolysis using renewable electricity (hydro, wind, and solar).

Thermal management is a major problem, as the overall process is highly exothermic, Eqs (1-3):

$$CO_2 + 4H_2 \rightleftarrows CH_4 + 2H_2O \quad \Delta H^{\circ}_{298K} = -165 \text{ kJ/mol} \quad (1)$$

$$CO_2 + H_2 \rightleftarrows CO + H_2O \quad \Delta H^{\circ}_{298K} = +41 \text{ kJ/mol} \quad (2)$$

$$CO + 3H_2 \rightleftarrows CH_4 + H_2O \quad \Delta H^{\circ}_{298K} = -206 \text{ kJ/mol} \quad (3)$$

To address this issue, the Sabatier reactor should be designed with a highly-efficient heat removal [2, 3]. In this study, the actively cooled Sabatier reactor was designed and simulated by numerical simulations first, to assess its feasibility. Based on the simulations results, a lab-scale prototype was assembled and tested successfully.

Model Formulation

The reactor configuration is shown in Fig. 1. The dynamic, pseudo-heterogeneous, non-isothermal model was solved using the MATLAB PDE solver:

$$\varepsilon \frac{\partial C_i}{\partial t} = D_{ae} \frac{\partial^2 C_i}{\partial z^2} - \varepsilon \frac{\partial (v_g C_i)}{\partial z} + a(1-\varepsilon)\rho_s \sum_j \eta_j R_{ij} \quad (4)$$

$$(\rho C_p)_{eff}\frac{\partial T}{\partial t} = k_{ae}\frac{\partial^2 T}{\partial z^2} - \varepsilon\rho_g C_{pg} v_g \frac{\partial T}{\partial z} + \\ + a(1-\varepsilon)\rho_s \sum_j (-\Delta H_j)\eta_j R_j - \\ - U_{w,HE} a_{r,HE}(T - T_c) - U_{w,HL} a_{r,HL}(T - T_e) \quad (5)$$

$$\rho_c C_{pc}\frac{\partial T_c}{\partial t} = \lambda_c \frac{\partial^2 T_c}{\partial z^2} - \rho_c C_{pc} v_c \frac{\partial T_c}{\partial z} - \\ - U_{w,HE} a_{c,HE}(T_c - T) \quad (6)$$

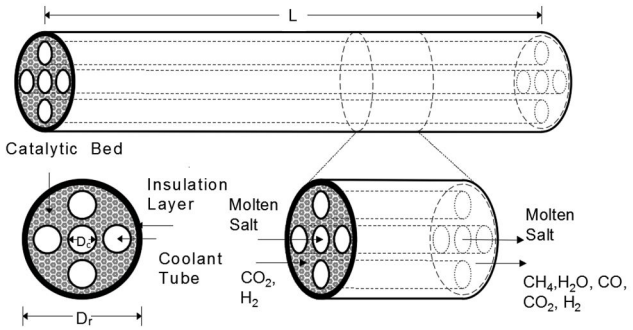

Figure 1. Reactor configuration

Result and Discussion

A typical spatio-temporal temperature profile is shown in Fig. 2.

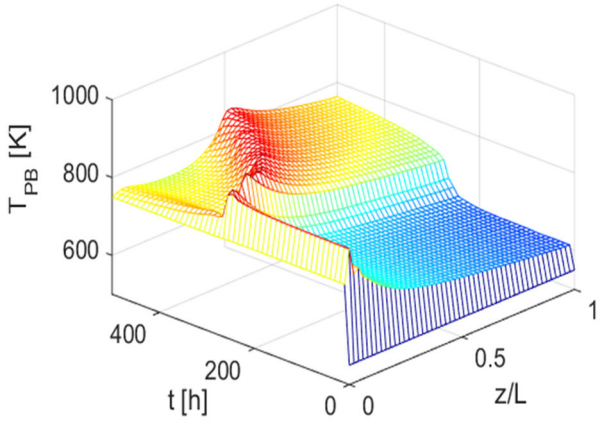

Figure 2. Spatio-temporal temperature profile

Initially, the hot spot is formed at the reactor entrance. As a result of the elevated temperature at this location, the catalyst starts to deactivate due to coke formation, leading in turn to lower reaction rates and, therefore, lower heat release. Under certain condition this phenomenon can results in the hot spot propagation downstream the reactor, which can eventually lead to reactor extinction.

The proof-of-concept unit is shown in Fig. 3 (10 cm active length, 7 g of catalyst), with the corresponding performance presented in Fig. 4. After preheating and initial ignition, the reactor was operated as a standalone unit without any external heating, with heat being removed by compressed air fed through the inner tube. 100% selectivity and 90% CO_2 conversion was achieved.

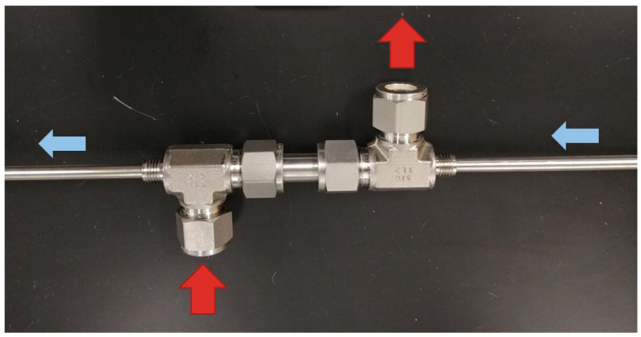

Figure 3. Proof-of-concept unit

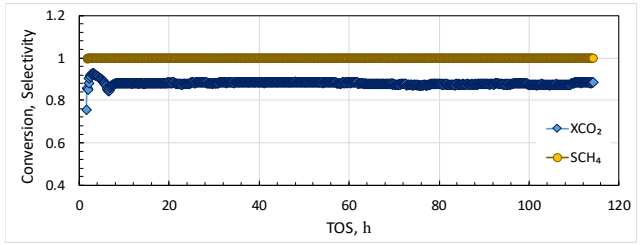

Figure 4. Lab-scale reactor performance

Conclusions

The Sabatier reactor was first investigated using the comprehensive mathematical model and numerical simulations. The lab-scale proof-of-concept unit was successfully tested. Further investigation is underway.

Acknowledgments

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References

Simakov, D. S. A. (2017). Renewable Synthetic Fuels and Chemicals from Carbon Dioxide. *Springer*.

Sun, D., Simakov, D. S. A. (2017). Thermal management of a Sabatier reactor for CO_2 conversion into CH4: Simulation-based analysis. *J. CO_2. Util.*, *21*, 368.

Sun, D., Khan, F. M., Simakov, D. S. A. (2017). Heat removal and catalyst deactivation in a Sabatier reactor for chemical fixation of CO_2: Simulation-based analysis. *Chem. Eng. J.*, *329*, 165.