Modeling and Simulation Study of a Fixed-Bed Catalytic Reactor for the Hydrogenation of CO_2 to CH_4

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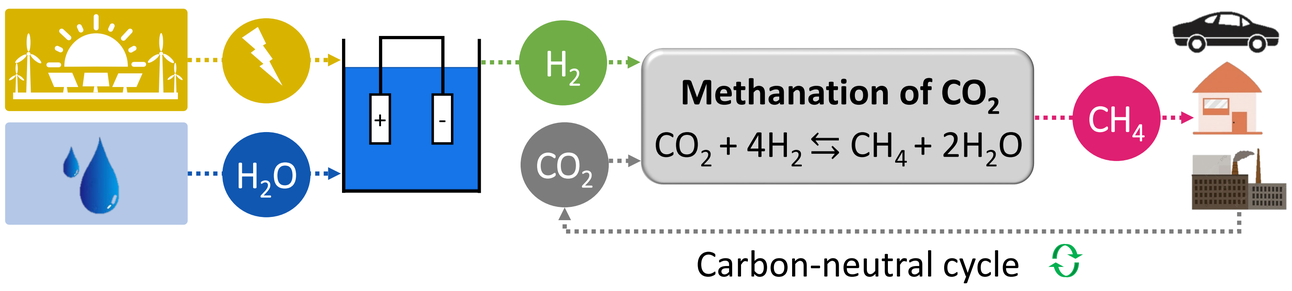
Introduction



The methanation is a highly exothermic and reversible reaction. Thus, efficient heat management of the reactor is necessary to avoid hot spots and maximize CH_4 production at the reactor outlet [1]. The use of simulation software can be a very useful tool to assist the design of efficient reactors [2].

The objective of this work is to develop multiple simulation models for the CO_2 hydrogenation reaction using COMSOL Multiphysics® software. These models utilize the Chemical Reaction Engineering Module and its associated interfaces to simulate the CO2 hydrogenation process within a fixed-bed reactor employing a 10% Ni/alumina catalyst.

Methodology

One-dimensional model

Transport of Concentrated Species

$$\nabla\cdot j_i + \rho(u\cdot\nabla)w_i = R_i \qquad j_i = -\left(\rho D_i^m \nabla w_i + \rho w_i D_i^m \frac{\nabla M_n}{M_n} - j_{c,i}\right)$$

Heat Transfer in Fluids

$$A_c\rho C_p u\cdot\nabla T + \nabla\cdot q = A_c Q + q_0 \quad q = -A_c k_f \nabla T \quad q_0 = A_i\cdot\left[Q - U\cdot(T - T_{ext})\right]$$

IMPORTANT: Inlet boundary conditions

$$w_{bnd,i} : -\int_{\partial\Omega} n\cdot(j_i + \rho u w_{bnd,i})\,dS = J_{in,i} \qquad -n\cdot q = \rho\,\Delta Hu\cdot n$$

Two-dimensional model

Transport of Concentrated Species in Porous Media

$$\nabla\cdot j_i + \rho(u\cdot\nabla)w_i = R_i \qquad D_{e,ik} = \frac{\varepsilon_p}{\tau_f} D_{ik} \qquad \tau_f = \varepsilon_p^{-1/3}$$

Heat Transfer in Porous Media

$$A_c\rho C_p u\cdot\nabla T + \nabla\cdot q = A_c Q + q_0 \qquad k_{eff} = \varepsilon_p k_f + \vartheta_s k_s$$

Brinkman Equations

$$0 = -\nabla p + \nabla\cdot\left[\frac{1}{\varepsilon_p}\left\{\mu\left(\nabla u + (\nabla u)^T\right) - \frac{2}{3}\mu(\nabla\cdot u)I\right\}\right] - \left(\mu\kappa^{-1} + \frac{Q_m}{\varepsilon_p^2}\right)u + F \qquad \nabla\cdot(\rho u) = Q_m$$

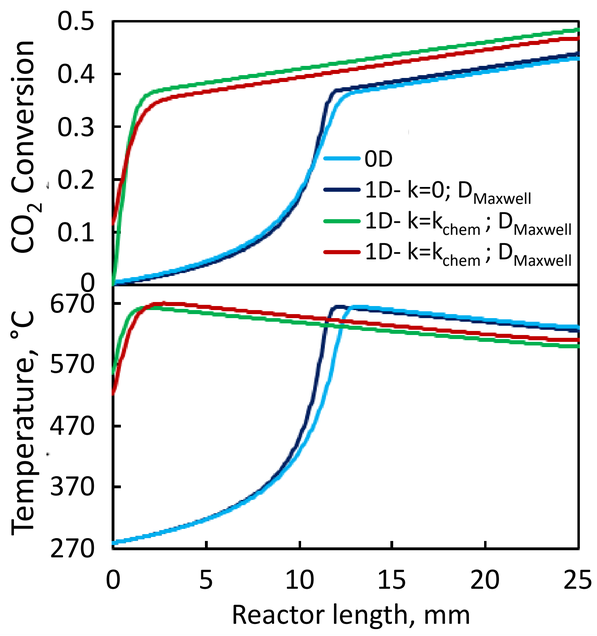
Results and Discussion

One-dimensional model

It is analyzed the impact of mass and heat transport in the longitudinal axis of a reactor, comparing it to ideal zero-dimensional models.

Inclusion of heat transfer caused significant changes in CO_2 conversion and temperature profiles, shifting the hot spot towards the reactor entrance and raising temperatures to 522 °C at the entrance.

Mass transport, to a minor extent, also influenced these profiles, making them smoother due to back mixing effects.



Presence of porous medium

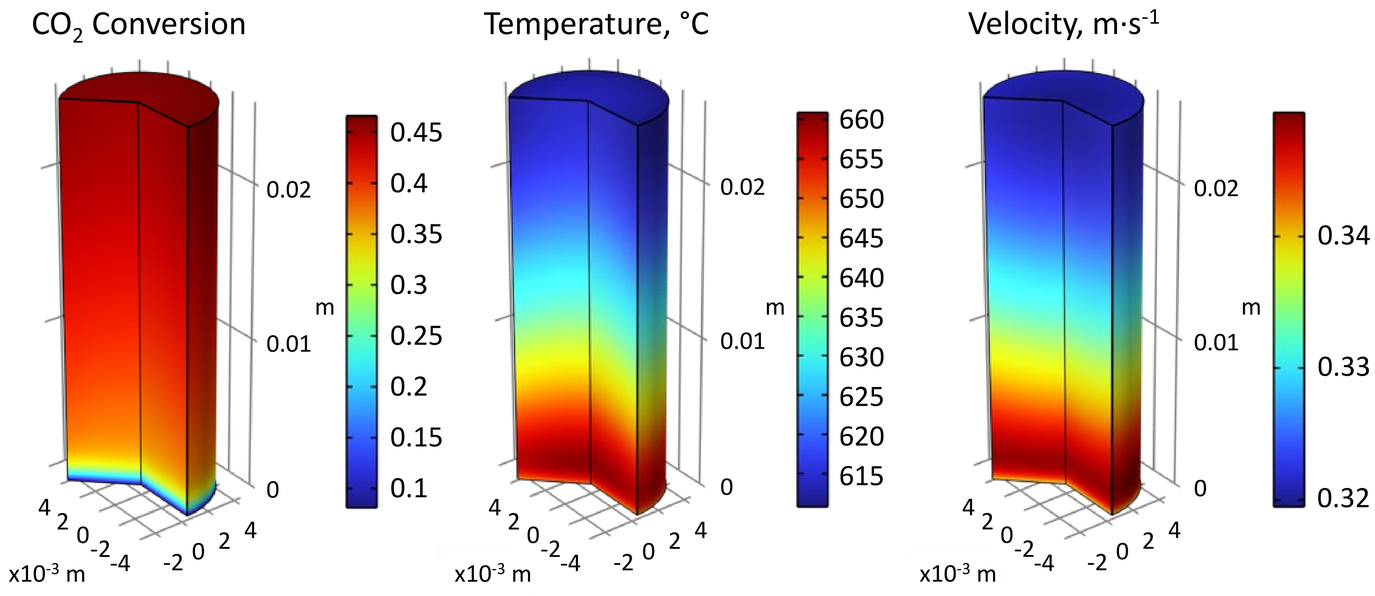
Mass and heat transport in the radial direction

Brinkman equations for velocity profile

Axisymmetric Two-dimensional model

In the radial direction, velocity gradients are absent, while CO_2 conversion and temperature gradients are observable. The reactor walls act as a refrigeration source, causing the gas mixture to be cooler near the reactor wall compared to the axial coordinate. This temperature distribution affects thermodynamics, promoting higher CO_2 conversion near the reactor wall and decreasing conversion towards the axial coordinate of the reactor.

Incorporating a porous medium within the reactor significantly enhances heat transport, primarily due to the higher conduction coefficient of the solid. This alteration results in a further shift of the hot spot towards the reactor entrance, with temperatures reaching 649 °C at the entrance. Also, the hot spot is reduced to 660 °C.



Conclusions

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COMSOL® has demonstrated its effectiveness as a simulation tool. Future work will involve implementing reactor models that consider catalysts, enabling the analysis of mass and heat transfer resistances between fluid and solid phases. Furthermore, temporal studies will be conducted to understand how the reactor responds to variations in hydrogen flow rates, which are contingent on the availability of renewable energy surplus.

References

1. S. Rönsch *et al*, "Review on methanation – From fundamentals to current projects," *Fuel (Guildford),* vol. 166, pp. 276-296, 2016.
2. A. Fache; F. Marias, "Dynamic operation of fixed-bed methanation reactors: Yield control by catalyst dilution profile and magnetic induction." *Renewable energy, vol. 151, pp.* 865-886, 2020.

Acknowledgements

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