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Kinetic modelling of the Sabatier reaction to produce synthetic natural gas

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This project addresses the efficient conversion of renewable hydrogen into storable and dispatchable energy carriers through CO_2 methanation (Sabatier reaction) integrated within a power-to-gas (P2G) framework. A central component of the work is a comprehensive kinetic study of CO_2 hydrogenation over a Ni/CeO_2–ZrO_2 catalyst, aimed at developing reliable models for reactor design and system integration.
Experiments are designed using a Central Composite Design to systematically explore the effects of temperature, pressure, gas hourly space velocity (GHSV), and H_2/CO_2 ratio. The study spans a wide operating window representative of industrial conditions, with repeated central points ensuring statistical robustness and reproducibility. A total of approximately 40 experimental conditions are investigated, with CO_2 conversion ranging from 10% to 90%, thus enabling the identification of both kinetic and equilibrium-limited regimes.
In addition to the main Sabatier reaction ($CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$), the system accounts for the reverse water gas shift (RWGS) reaction and CO methanation, allowing a complete description of the reaction network. The inclusion of these parallel and consecutive reactions is essential to capture selectivity trends and intermediate species formation under varying conditions.
Kinetic modelling is carried out using Python-based tools and implemented in Aspen Plus, combining parameter estimation, regression, and model validation. Two modelling approaches are explored and compared: (i) power-law rate expressions, providing a simplified and computationally efficient description, and (ii) Langmuir–Hinshelwood–Hougen–Watson (LHHW) models, incorporating adsorption phenomena and competitive surface reactions. The latter approach enables a more mechanistic interpretation, particularly relevant for understanding hydrogen and CO_2 adsorption, surface coverage effects, and the role of CO as an intermediate.
Model discrimination is performed based on statistical criteria (e.g., residual analysis, confidence intervals) and predictive capability across the full experimental domain. The resulting kinetic models are validated against the complete dataset and used to identify optimal operating conditions and rate-limiting steps.
The final outcome is a robust kinetic framework capable of describing CO_2 methanation over Ni/CeO_2–ZrO_2 catalysts under realistic conditions, suitable for integration into reactor and process simulations.