An Integrated Approach to CO_2 Methanation: catalyst synthesis, advanced reactor design and process simulation.

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The transition from fossil fuels to sustainable energy systems requires innovative solutions to reduce CO_2 emissions while meeting global energy demands. Power-to-Gas (PtG) technologies, particularly CO_2 hydrogenation into methane, offer a promising method for renewable energy storage and integration into existing infrastructure. This study seeks to optimize the methanation process through a multi-phase approach involving thermodynamic analysis, experimental validation, catalyst development and advanced reactor design. A preliminary thermodynamic assessment was performed using Gibbs free energy minimization in Aspen Plus® to identify optimal reaction conditions, exploring variables such as temperature (200–500°C), pressure (1–30 atm) and H_2/CO_2 molar ratios (2:1–6:1). The impact of selective water removal, simulating sorption-enhanced and membrane reactors, was also examined. Based on these results, a literature review was conducted to screen commercial and novel catalysts for CO_2 methanation. Although ruthenium (Ru) exhibited superior activity and CH_4 selectivity, nickel (Ni) was preferred due to its lower cost and comparable performance. To improve Ni-based systems, bimetallic formulations with Fe, Pt, Pd, Rh, Co and Ru were studied, focusing on the Ni-to-metal ratio (Ni-X), as it critically affects the catalytic behavior. Over 350 catalysts were compared and categorized by CO_2 conversion and CH_4 selectivity. This analysis revealed key thermodynamic constraints and opportunities, establishing a theoretical framework for catalyst selection. These insights guided the development of a laboratory-scale fixed-bed reactor for the experimental campaign. A commercial 0.5 wt.% Ru/Al_2O_3 catalyst, widely studied in the literature, was selected for the study. After a thorough literature review, an appropriate kinetic model was identified and implemented in Aspen Plus® for process simulation. The simulation was validated experimentally by varying key parameters such as temperature, gas hourly space velocity (GHSV) and H_2/CO_2 ratio, showing excellent agreement with measured data (error <10%) and confirming the reliability of the kinetic model and simulation framework. The final goal is to develop a high-performance system with CH_4 selectivity near 99% and CO_2 conversion above 90%. This integrated approach highlights how experimental catalysis, reactor design and process simulation can advance the development of efficient scalable solutions for carbon-neutral energy systems.

Keywords: CO_2 methanation, Fixed-bed reactor, Aspen Plus® modeling, Heat management, Bimetallic catalysts synthesis.