

Deactivation of a Ni/Al₂O₃ catalyst during CO₂ methanation studied in a concentration-controlled recycle reactor

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Introduction: Catalyst deactivation negatively affects the economic efficiency of a chemical plant due to increased energy and reactant consumption during the production process and downtime costs during catalyst exchange [1]. Simulation and optimization studies can assist in developing strategies to reduce these costs. For this, however, knowledge of the deactivation kinetics is required. In the case of CO₂ methanation, accurate descriptions of the reaction kinetics are available [2], yet literature lacks in detailed descriptions of deactivation kinetics. Therefore, this work investigates the deactivation of a Ni/Al₂O₃ catalyst during CO₂ methanation, examining how the temperature and partial pressures influence the deactivation rate, identifying the dominant deactivation mechanism and developing a deactivation model.

Methods: In order to develop appropriate deactivation kinetics based on experimental data, it is necessary to decouple the catalyst activity from the concentration effects on the reaction rates. With constant inlet flow rates to the recycle reactor, partial pressures of species, and thus reaction rates, change due to deactivation, requiring exact knowledge of reaction kinetics to determine catalyst activity. To avoid additional uncertainties from kinetic models, the decoupling of catalyst activity and reaction rates is achieved by measures in the experimental design. Keeping the concentrations constant by adjusting the inlet flow rates allows for observing the catalyst activity [3].

Results and discussion: Utilizing this concentration-controlled recycle reactor (CCRR), the catalyst activity is observed over time. As shown in Figure 1a, the temperature significantly influences the deactivation rate and the deactivation is caused by sintering of the nickel particles. Furthermore, the deactivation is influenced by the partial pressures in the reactor. Figure 1b shows that an increased partial pressure of water increases the deactivation rate under otherwise constant conditions. Based on these investigations, a first deactivation model was developed. Systematic experimental studies and extension of the deactivation model will enable optimization studies on reactor design and operating strategies.

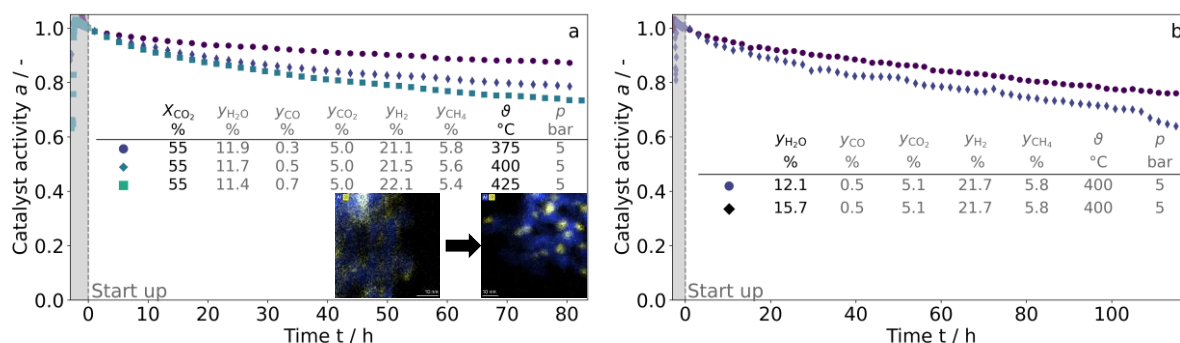


Figure 1: Effect of temperature (a) and partial pressure of water (b) on catalyst activity profiles in a CCRR. Post-mortem observation using EDS (a) reveals sintering of Ni particles.

References

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