Advanced Catalyst and Reactor Engineering for Efficient Hydrogen Release from Perhydro-Benzyltoluene

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Abstract

Hydrogen storage via liquid organic hydrogen carriers (LOHCs) such as perhydro-benzyltoluene (H12-BT) is a promising route for safe, reversible, and compact hydrogen storage under ambient conditions.[1] Recent developments in both catalyst design and reactor engineering have significantly enhanced dehydrogenation performance, a critical step for hydrogen release. In this study the performance of bimetallic platinum-rhenium (PtRe) catalysts[2] supported on Al_2O_3 was investigated and optimised for dehydrogenation in a continuous three phase slurry reactor. The slurry reactor represents a promising alternative to conventional fixed-bed reactor designs. Superior heat management and rapid separation of released hydrogen mitigate the dewetting effects commonly encountered in endothermic LOHC dehydrogenation processes.

Modification of the PtRe/Al_2O_3 catalysts focussed on the influence of support morphology, surface chemistry, and synthesis conditions. By tailoring synthesis procedures—including support calcination parameters and the order of metal precursor during sequential impregnation—and analysing the resulting catalysts by ICP-AES, XRD, CO-pulse chemisorption, NH_3-TPD, TEM, and N_2 physisorption, observed effects on catalyst productivity and dehydrogenation activity were characterised. Additional selective sulphur poisoning resulted in improved by-product suppression. The findings provide critical insight into the optimisation of metal-support interactions and preparation methods to enhance hydrogen release kinetics and catalyst lifetime.

The slurry reactor demonstrated stable long-term operation at elevated temperatures (330 °C) and pressures (4.7 barg), with high platinum-based productivity and low by-product formation. Operation parameters, such as stirrer speed, temperature, pressure, and feed rate, were systematically varied to optimise space-time-yield and degree of dehydrogenation. Kinetic analyses based on Arrhenius parameters were conducted to deepen mechanistic understanding. Catalyst and reactor performance were benchmarked using both commercial and custom-designed platinum-based catalysts, including S-poisoned Pt/Al_2O_3 and PtRe/Al_2O_3 formulations. Ultimately, the synergy between advanced Pt-based catalysts and optimised slurry reactor design enabled a significant enhancement in hydrogen release efficiency from H12-BT.

This study highlights a holistic approach to LOHC dehydrogenation by combining rational catalyst design with innovative reactor engineering. The integration of tailored PtRe catalysts and continuous slurry reactor operation establishes a robust framework for improving the viability and scalability of hydrogen storage technologies in real-world energy systems.

1. E. Herzinger, M. Wolf, *Chem. Ing. Tech.* **2024**, *96*, 65.
2. D. Strauch, P. Weiner, B. B. Sarma, A. Körner, E. Herzinger, P. Wolf, A. Zimina, A. Hutzler, D. E. Doronkin, J.-D. Grunwaldt P. Wasserscheid, M. Wolf, *Catal. Sci. Technol.* **2024**, *7*, 1775.