

Supervisor(s)	Period	Funding
Prof. Mark Saeys	4 years	FWO

Design of a composite catalyst system for CO₂-assisted light alkane dehydro-aromatization. 2 PhD positions.

Aim

Design of an efficient CO₂-Oxidative Dehydro-Aromatization (ODA) catalytic process which couples the production of BTX from light alkanes with the conversion of CO₂ to CO.

Justification

Benzene and Xylene are key building blocks for robust polymer materials. They are typically obtained as side products of the production of high-octane gasoline and of the naphtha cracking for light olefins. Dehydro-aromatization of light alkanes provides an alternative on-purpose process to produce these aromatic building blocks. Direct alkane dehydro-aromatization (DDA) suffers from severe coke formation and unfavorable equilibrium conversions. The addition of CO₂ during propane dehydrogenation (CO₂-PDH) improves the equilibrium propane conversion by removing hydrogen via the reverse water gas shift reaction (RWGS), while at the same time producing valuable CO and enhancing the stability of the catalyst.

In recent work, we found that the presence of CO₂ reduces coke formation and modifies the state of the GaO_x/HZSM-5-based catalyst to a more active and stable phase. However, the complex product composition of CO₂-PDH complicates product separation and process development. CO₂-ODA potentially combines the advantages of CO₂-PDH with easier separation and produces more valuable aromatics. Aromatization of light alkanes is accomplished by a synergy of Brønsted (B) and Lewis (L) centers. The acid type, strength and distribution greatly influence selectivity and coke formation, providing opportunities for catalyst design. GaO_x/HZSM-5 emerges as a promising catalyst system able to achieve high BTX selectivity at high conversions. To couple alkane dehydro-aromatization with CO₂ conversion, additional active sites need to be introduced since GaO_x/HZSM5 shows limited RWGS activity.

In this project, we develop composite catalyst systems, combining the various catalytic functions in a balanced way to exploit catalytic coupling between the reaction steps. The project brings together advanced synthesis, characterization and catalytic testing using state-of-the-art steady-state and transient (SSITKA) kinetic studies available at the LCT. The combination of standard characterization tools with Density Functional Theory (DFT) calculations provides unprecedented insight in the kinetic role of the different active sites. One PhD will focus on synthesis and testing, the other on characterization and DFT modeling.

Program

- Preparation and testing of composite catalyst systems.
- Investigation of structure-reactivity relationships for the CO₂-ODA reaction.
- Unraveling the kinetic role of the various active sites and their interactions; identification of coke precursors by steady-state and transient kinetic measurements and by DFT calculations.