LABORATORY FOR CHEMICAL TECHNOLOGY
Technologiepark 125, 9052 Gent, Belgium

Coaches	**Supervisor(s)**	**Funding**
Dr Dipika R. Kanchan	Prof. Mark Saeys	

Modeling the Impact of Catalyst Structure and Surface Coverages on CO_2 Hydrogenation to Methanol over $ZnZrO_2$ catalysts

Aim

Analysing effect of the catalyst structure and surface coverages on the CO_2 hydrogenation to methanol over inverse oxide catalysts using:

1. Density functional theory (DFT) calculations
2. Machine Learning
3. Microkinetic modeling

Justification

Electrification is a key strategy in the EU Green Deal. Direct electrification is however not always feasible, and converting H_2 and CO_2 into methanol offers a cost-effective and scalable alternative to store renewable energy and convert CO_2 into chemical building blocks. Methanol is compatible with existing infrastructure for transport, storage, and industrial use.

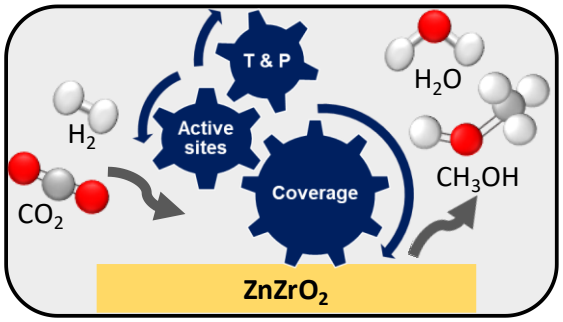

Figure 1. Illustration of the CO_2 to methanol reaction over $ZnZrO_2$ catalyst.

Oxide-based catalysts such as $ZnZrO_2$ catalyst show promising selectivity to methanol over CO compared to commercially used $Cu/ZnO/Al_2O_3$ (CZA) catalysts.[1-3] Surface coverages, catalyst structure, and reaction conditions significantly influence catalyst performance. Researchers claim that the superior activity is a result of unique synergy between ZnO and ZrO_2. However, a detailed atomistic understanding of the catalytic surface and its evolution under industrially relevant reaction conditions remains uncovered.[4] The availability of various crystal structures, surface sites and surface species make it challenging to identify the key character of surface which govern catalytic activity. Understanding how surface properties influence reactivity requires integrating molecular modeling with microkinetic simulations to bridge theoretical insights with experimental observations.

In this proposal, we aim to bridge the gap between computational models and realistic catalyst behavior by leveraging advanced computational modeling, machine learning (ML), and compare with experimental data obtained at the LCT.[3] We will systematically explore how various surface coverages, catalyst reconstructions, and active sites influence CO_2 hydrogenation to methanol.

Program

1. Literature review: Experimental and theoretical studies on CO_2 hydrogenation to methanol over oxide-based catalysts, with a focus on selectivity and active sites.
2. Computational surface science studies to investigate the synergy between ZnO and ZrO_2 and understanding roles of various surface sites and species.
3. Developing machine learning potentials to expedite understanding of surface evolution and activity.
4. Microkinetic modeling for the data developed in above steps to estimate surface-activity.

References

1. T. Pinheiro Araújo et al., *Nat. Commun.*, 2024, **15**, 3101.
2. T. Pinheiro Araújo et al., *Adv. Energy Mater.*, 2023, **13**, 2204122.
3. Z. Zanganeh et al., *Catal. Sci. Technol.*, 2025, **15**, 563.
4. H. Zada et al., *ChemSusChem*, 2024, **18(4)**, e202401846.