LABORATORY FOR CHEMICAL TECHNOLOGY
Technologiepark 125, 9052 Gent, Belgium

Coaches	**Supervisor(s)**	**Funding**
Konstantijn Rommens	Prof. Mark Saeys	FWO

Application of machine learning potentials for the discovery of stable Fe-based CO2 to jet fuel catalysts

Aim

Analyse the surface structure of Fe-based catalysts to model CO_2 hydrogenation to jet fuel by using:

1. Density functional theory (DFT) calculations
2. Machine learning potentials

Justification

Recently, the European commission presented its Green Deal to become the 'first climate-neutral continent'[1]. Electrification of the economy is a key component of the Green Deal, but direct electrification is not the best option for many sectors. Air travel and long-distance transport are amongst the most difficult sectors to electrify. A lot of routes to convert CO_2 to e-fuels have been studied, ranging from traditional heterogeneous catalysis to electrocatalysis and photocatalysis[2-4]. While the conversion of CO to jet fuel is one of the largest catalytic processes (Fischer-Tropsch Synthesis, CO-FT), the direct conversion of CO_2 to jet fuel (CO_2-FT) suffers from poor selectivity and low carbon efficiencies (i.e., carbon from CO_2 ending up in the jet fuel). The selective hydrogenation of CO_2 to long chain hydrocarbons is a challenging catalytic reaction, which requires multi-functional catalysts. Coupling the reverse water gas shift (RWGS) and CO-FT reactions in a single catalytic process can in principle lead to higher carbon utilization efficiencies (**Error! Reference source not found.**)[4]. An ideal catalyst for this will need to balance CO_2 activation with C-C coupling reactions. Fe-based catalysts are successful in CO-FT and in this project their activity for CO_2-FT will be investigated. Iron is already active in the RWGS, but a wide range of phases and surface facets have been proposed as the active phase. By performing selected DFT calculations the catalytic site structure and reactivity can be uncovered. And the latter can be used to predict the activity. Moreover, this project will also explore the use of machine learning potentials (MLPs) in predicting the stability of Fe surface facets under reaction conditions and by extension the nature of available catalytically active sites. Lastly, the MLPs predicted catalyst structure will be compared with the earlier findings based on thermodynamic analyses.

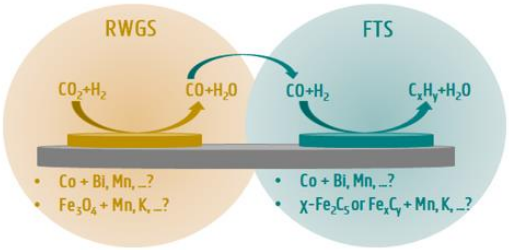

Figure 1. Tandem CO_2-FT mechanism on Fe and Co, coupling RWGS for CO_2 activation with C-C coupling via FTS to form long-chain hydrocarbons

Program

1. Literature review: (1) role of promotors for CO_2-FT and (2) application of MLPs in catalysis.
2. Surface science studies with MLPs to uncover surface stability and possible restructuring of unpromoted Fe- based catalysts under reaction conditions
3. DFT Calculations on uncovered active sites to link structure and activity
4. Development of machine learning potentials for uncovering the stability of FeC_xO_y surfaces

References

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3. A. A. Tountas, et al., *Adv. Sci.*, 2019, **6**, 1801903.
4. B. Yao, et al., *Nat. Commun.*, 2020, **11**, 6395.