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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 CO₂ 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 CO₂ to e-fuels have been studied, ranging from traditional heterogeneous catalysis to electrocatalysis and photocatalysis²⁻⁴. While the conversion of CO to jet fuel is one of the largest catalytic processes (Fischer-Tropsch Synthesis, CO-FT), the direct conversion of CO₂ to jet fuel (CO₂-FT) suffers from poor selectivity and low carbon efficiencies (i.e., carbon from CO₂ ending up in the jet fuel). The selective hydrogenation of CO₂ 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 CO₂ activation with C-C coupling reactions. Fe-based catalysts are successful in CO-FT and in this project their activity for CO₂-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.

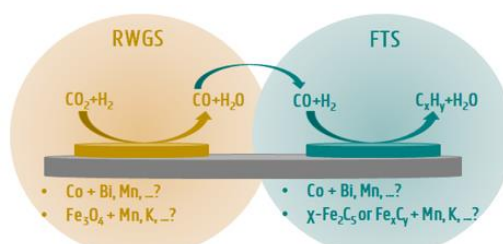


Figure 1. Tandem CO₂-FT mechanism on Fe and Co, coupling RWGS for CO₂ activation with C-C coupling via FTS to form long-chain hydrocarbons

Program

1. Literature review: (1) role of promoters for CO₂-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

1. European Commission, Communication from the Commission to the European Parliament, the European Council, the Council, the European Economic and Social Committee and the Committee of the Regions, The European Green Deal, COM(2019) 640 final., 2019
2. S. Nitopi, et al., *Chem. Rev.*, 2019, **119**, 7610-7672.
3. A. A. Tountas, et al., *Adv. Sci.*, 2019, **6**, 1801903.
4. B. Yao, et al., *Nat. Commun.*, 2020, **11**, 6395.

