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Application of machine learning in the design of Co-based CO2 to jetfuel catalysts

Aim

Analyse the effect of promotors on cobalt-based catalysts to facilitate CO_2 hydrogenation to jetfuel by using:

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

Justification

Recently, the European commission presented its Green Deal to become the 'first climate-neutral continent'[2]. 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[1, 3-4]. While the conversion of CO to jetfuel is one of the largest catalytic processes (Fischer-Tropsch Synthesis, CO-FT), the direct conversion of CO_2 to jetfuel (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.**)[1]. An ideal catalyst for this will need to balance CO_2 activation with C-C coupling reactions. Cobalt-based catalysts are successful in CO-FT and in this project their activity for CO2-FT will be investigated. However, to promote fast CO_2 activation, an additional type of active sites will need to be introduced to the Co system. This will be investigated by performing selected DFT calculations to uncover the catalytic site structure and reactivity. And the latter can be used to predict the activity. Moreover, this project will also explore the use of machine learning potential (MLPs) in predicting the morphology Co nanoparticles under reaction conditions and by extension the nature of available catalytically active sites. Lastly, the MLPs predicted catalyst structure will be compared with earlier findings based on thermodynamic analyses[5-7] as well as used to generate a more realistic view of CO_2.

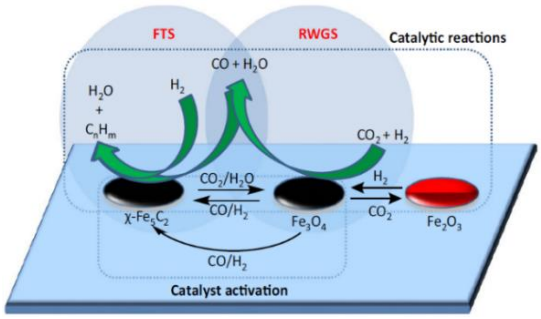

Figure 1. Tandem CO_2-FT mechanism on Fe, coupling RWGS for CO_2 activation with C-C coupling via FTS to form long-chain hydrocarbons[1]

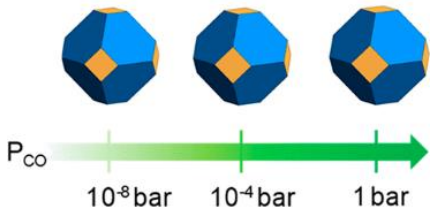

Figure 2. Wulff equilibrium shapes for cobalt nanoparticles for a CO pressure of 10^{-8}, 10^{-4} and 1 bar at 500K[6]. Co(111) facet (blue) and Co(100) is (yellow)[6]

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 particle structures and surface restructuring of (un)promoted Co-based catalysts under reaction conditions
3. DFT Calculations on uncovered active sites to link structure and activity
4. Implementation of the promotor kinetics in the dual-site microkinetic model developed at LCT

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