

<b>Coaches</b> Konstantijn Rommens Dengxin Yan Lukas Hiel	<b>Supervisor(s)</b> Prof. Mark Saeys	<b>Funding</b> CO2MBS
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## Computational investigation of the structure and activity of supported Ni- and Co-based catalysts

### Aim

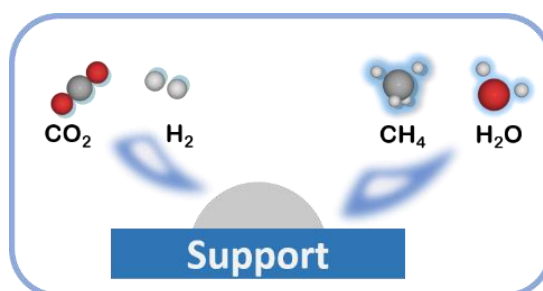
Analysis of the surface structure and activity of Ni- and Co-based catalysts in the CO<sub>2</sub> hydrogenation to methane (and olefins) using:

1. Density functional theory (DFT) calculations
2. Microkinetic modelling

### Justification

Recently, the European Commission presented its Green Deal to become the 'first climate-neutral continent'. Electrification of the economy is a key component of the Green Deal, but direct electrification is not the best option for many sectors. One of the routes explores the use of hydrogen as a possible energy carrier, but high transportation and storage costs are associated with hydrogen. Therefore, routes to convert hydrogen into molecules that are compatible with the current infrastructure such as methane or other valuable chemicals, like olefins, are highly desirable.

Nickel is often used as a catalyst for the methanation of CO<sub>x</sub>, while cobalt also has a strong methanation potential. Both catalysts will be studied with a detailed first-principles microkinetic model, where the influence of temperature, pressure and feed composition will be investigated. This model builds on the extensive microkinetic model for CO hydrogenation developed at the LCT.



**Figure 1.** Illustration of the CO<sub>2</sub> methanation reaction over supported Ni- and Co-based catalysts.

In the next stage, the role of the support and of promoters will be investigated using DFT calculations. Ceria-Zirconia supports have been proven to boost CO<sub>2</sub> methanation for Co-based catalysts<sup>1, 2</sup>, while TiO<sub>x</sub> supports tune the selectivity towards C<sub>2+</sub> hydrocarbons<sup>3, 4</sup>. Modelling of the effects of the support has been limited, leaving a knowledge gap in our understanding of the reaction mechanism. Currently, catalysts are assumed to be metallic under reaction conditions, but the shift to CO<sub>2</sub>-rich feeds can alter the stability of oxides and might impact the stability of active sites. Additionally, the support-metal interface and support itself are promising alternative active sites.

### Program

1. Literature review: role of the support and promoters on CO<sub>2</sub> methanation over Ni and Co
2. Microkinetic modelling of the CO<sub>2</sub> methanation reaction on Ni and Co catalysts
3. Computational surface science studies to investigate the effect of the support and promoters on the active sites of Ni and Co catalysts
4. DFT calculations to link structure and activity

### References

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2. A. Parastaev, et al., *Nat. Catal.*, 2022, **5**, 1051-1060.
3. I. C. t. Have, et al., *Nat. Commun.*, 2022, **13**, 324.
4. M. Xu, et al., *Nat. Commun.*, 2022, **13**, 6720.