Understanding the Impact of Reaction Conditions on Methanation Catalyst Structure and Performance using in-situ Total Scattering

Benjamin M. D. Fahl, F. Manzoni, Mirijam Zobel
Institute of Crystallography, RWTH Aachen

Objectives

- Investigate disorder and nanostructure in Ni@γ-Al_2O_3 catalyst for CO_2 methanation under dynamic conditions using total X-ray scattering and PDF (Pair Distribution Function) analysis.[1]
- Understanding how structural changes impact catalytic performance.

The Pair Distribution Function

- Allows visual data interpretation: inter atomic distances as a histogram.[2]
- Reveals interfacial metal support interaction and restructuring.
- Insights into thermal motion and static disorder.
- Nanoparticle growth and sintering as well as coordination numbers.
- Complements XRD and XAS analysis by bridging short-range order (Å) to long-range crystallinity. Around a few dozen nano-meter.

Experimental

- Gained from X-ray scattering data, can be measured on laboratory setup or synchrotron.[3]
- Emulate real-world catalytic processes using dedicated flow cell design.
- CO_2 methanation experiment with alternating catalysis and drop-out cycles, simulating rewenable H_2 shortages.[4, 5]
- Using beamline ID15A at ESRF with 65 KeV.

Results

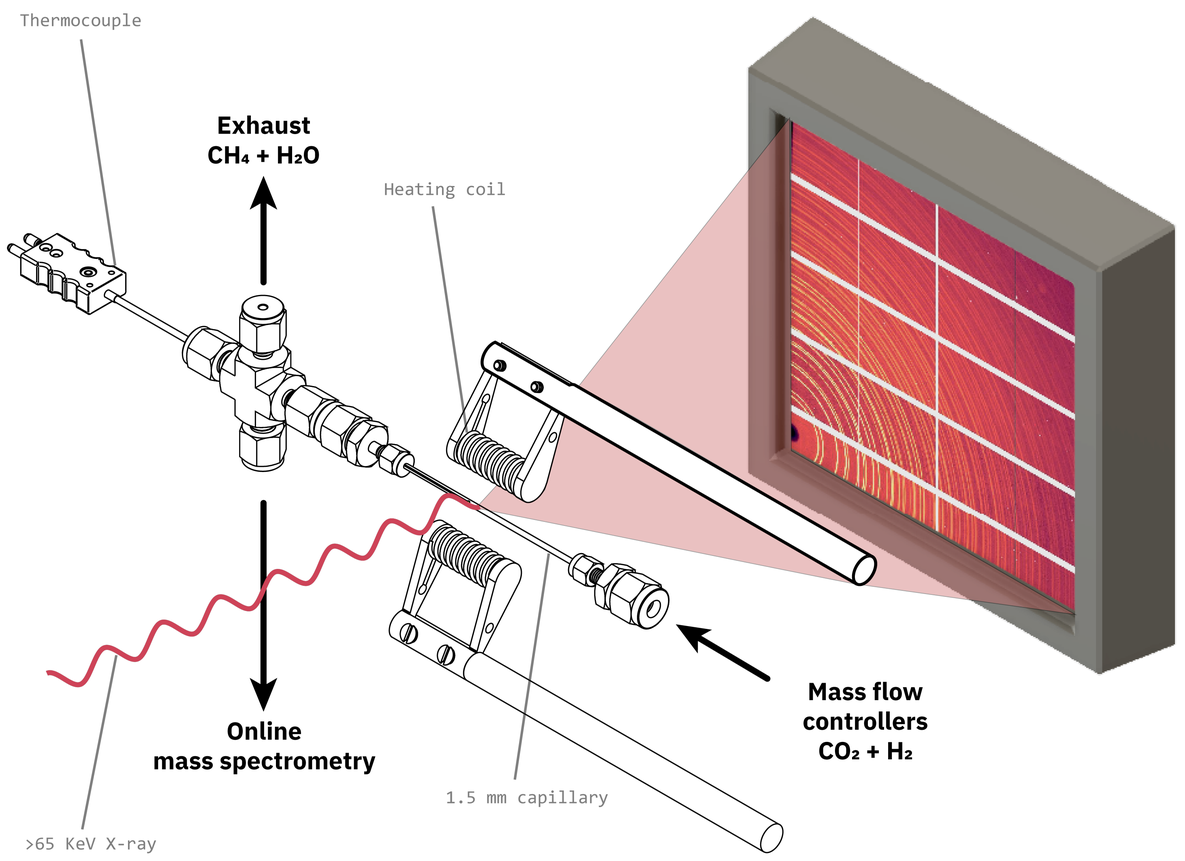
- Capturing **100 ms time resolution**, during dynamic processes, such as lattice expansion during drop-outs, and shrinkage during methanation cycles.
- Discern intricate structural details such as **reversible formation of 2 nm oxide layers** during drop-out conditions.
- Real time **Ni growth observation from 3 to 5 nm**, increased by methanation.
- Jumps in lattice constant only revealed by in-situ experiments. How can this behavior be explained?

References

[1] Christiansen et al, *Nanoscale Adv*, **2020**, 2, 2234-2254
[2] S. Billinge; I. Levin, *Science* **2007**, 316, 5824
[3] Thomä et al, *Rev. Sci. Instrum*, **2019**, 90 (4), 043905
[4] Prinz et al, *New J. Chem*, **2023**, 47, 11623-11635
[5] Prinz et al, *Nanoscale*, **2020**, 12, 15800-15813

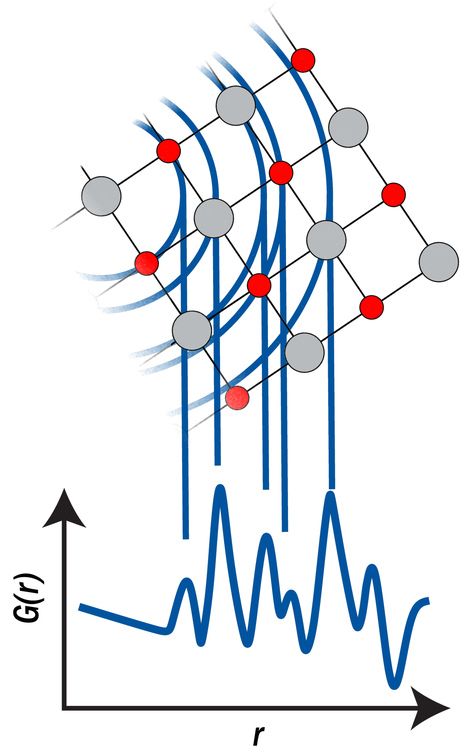
Contact: zobel@ifk.rwth-aachen.de

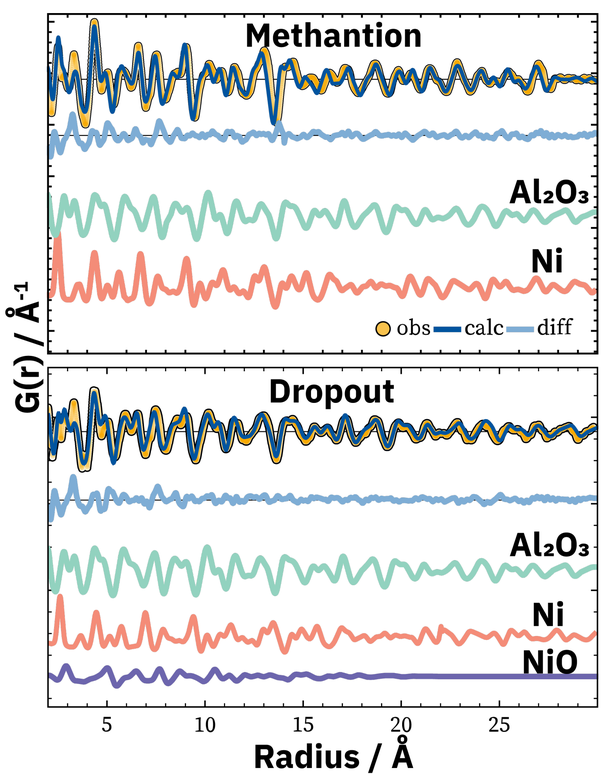
RWTH AACHEN UNIVERSITY

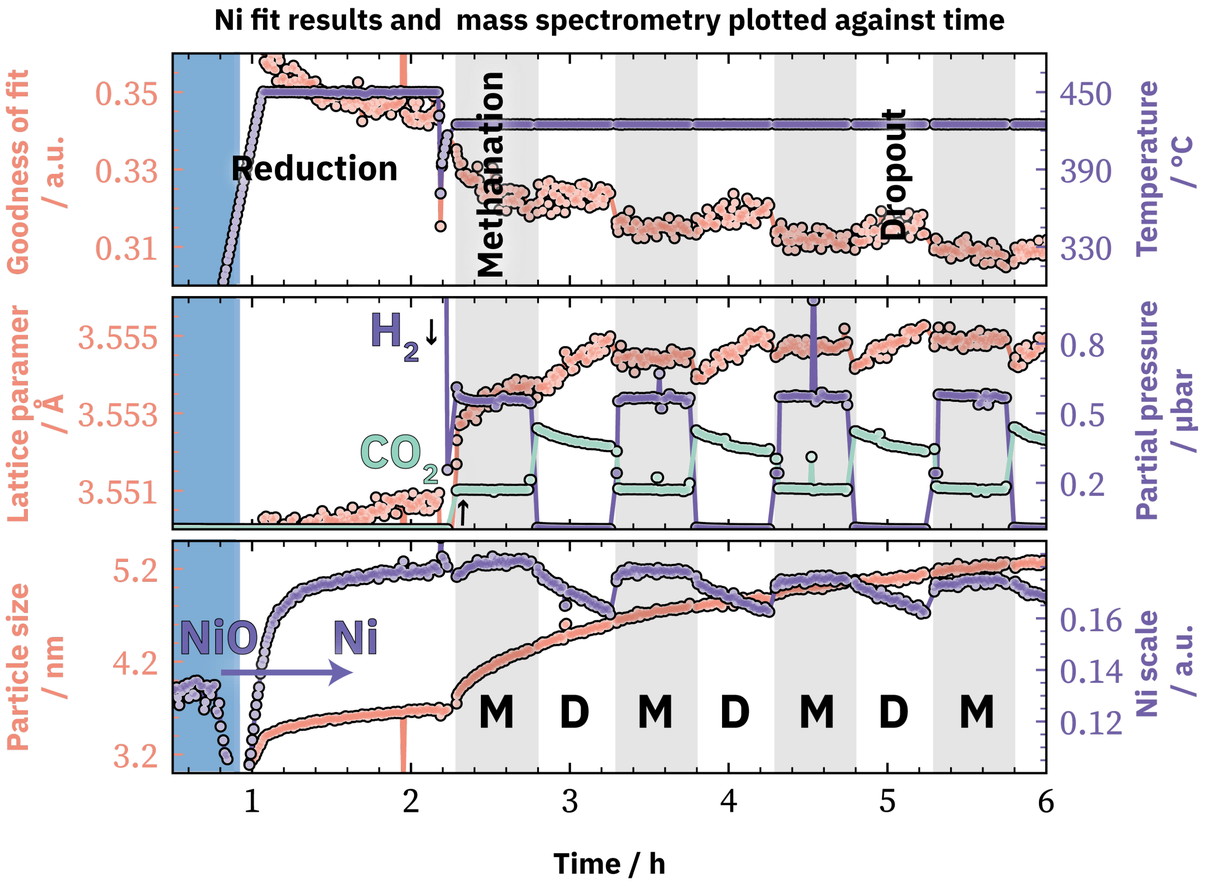


The Pair Distribution Function

$$G(r) \approx \mathbf{FT}[I_{2\theta}] \approx \widehat{f} \cdot \sum_{\mu\nu} \delta(\mathbf{r}_{\mu\nu}) - 4\pi\rho_0 r$$







Institut für Kristallographie

SPP 2080 DynaKat

ESRF The European Synchrotron

FSC The Fuel Science Center

Ben@Fahl.io