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Production of higher alcohols from CO and H₂

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Combined mechanistic and catalytic reactor studies were applied to gain mechanistic insights into the catalytic cycle of producing C₂⁻oxygenates (C₂O) from CO and H₂ over supported single metallic Rh catalysts.

Background

Supported metallic Rh has been shown to form C₂O with a wide range of selectivities and CO conversion levels [1,2]. The present study seeks to understand the origin of this wide spread and become able to make catalysts with high C₂O selectivity at simultaneous high syngas conversion at an acceptable level of metal loading.

Catalytic performance during the CO−H₂ reaction for 1% Rh dispersed on SiO₂ or ZrO₂ – impact of CO/H₂ pressure

Role of in situ formed MeOH as reactant

Significant levels of ester products were found at high CO/H₂ pressure conditions. It was shown, that surface C₂O precursor adsorbates can react in a H₂/MeOH atmosphere to methylate.

Topological changes to the Rh catalyst

RhCl₃-derived catalysts are reported to be better C₂O producing catalysts. Remaining Cl is assumed to mobilize the Rh through involvement in formation of Rh-carbonyl complexes.

• 50 bar CO−H₂ reaction treatment of 3% Rh/SiO₂ (from RhCl₃) resulted in a complete depletion of the Rh. Most likely due to volatility of formed carbonyls.

Outlook

• In situ EXAFS: Rh/support exposed to different CO pressures.
• Environmental TEM – identifying possible topological changes of the Rh during the exposure to CO atmospheres.

References