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DESIGN OF MULTICOMPONENT CATALYSTS BASED ON REDUCIBLE OXIDES FOR GREEN METHANOL

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ETH Hönggerberg, HCI J374 and on Zoom
(https://ethz.zoom.us/j/67997107613)

Project Summary: Methanol synthesis via thermocatalytic hydrogenation of carbon dioxide is a strategic route to enable the sustainable production of this vital commodity and energy carrier. However, the industrial implementation of this approach is limited by a lack of fundamental understanding of the catalytic materials used. To address this critical aspect, this project integrates precision catalyst synthesis, accurate catalytic evaluation, in-depth characterization, theoretical modelling, and kinetic studies to establish synthesis-structure-performance relationships for promising methanol synthesis catalysts based on reducible oxides, such as indium oxide (In2O3) and mixed zinc-zirconium oxides (ZnZrOx). Through this holistic approach, the thesis reveals how CO, a common byproduct in CO2-to-methanol, impacts the performance of In2O3- and ZnZrOx-based catalysts at practically-relevant reaction conditions, identifying distinct promotion and deactivation phenomena that are governed by an interplay of sintering, surface reduction, and CO inhibition. Additionally, the thesis establishes an atomic-level rationalization of In2O3 and ZnZrOx promotion by different metal promoters, offering key guidelines for the design of optimal systems. It was possible to overcome synthetic constraints by engineering multicomponent catalysts using flame spray pyrolysis, creating tailored catalyst architectures with superior performance. This approach led to the development of ternary Pd-In2O3-ZrO2 systems with enhanced oxygen vacancy generation that display unparalleled methanol productivity and stability, as well as ZnZrOx systems with maximized surface area and density of catalytic ensembles that outperform benchmark catalysts attained by coprecipitation. Finally, a systematic investigation using ex situ and in situ electron paramagnetic resonance spectroscopy provides a means to characterize oxygen vacancies on In2O3- and ZnZrOx-based catalysts, advancing understanding of this critical performance descriptor. The findings presented offer design criteria for the development of innovative and efficient catalytic technologies that can propel green methanol production at industrial scale.

CV: T. Pinheiro Araújo received his BSc in Chemistry at the Federal University of Maranhão in 2016. In 2017, he moved to the University of São Paulo and obtained his MSc in Chemistry in 2019. In the same year, he started his PhD studies in the advanced Catalysis engineering group led by Prof. J. Pérez-Ramírez.

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