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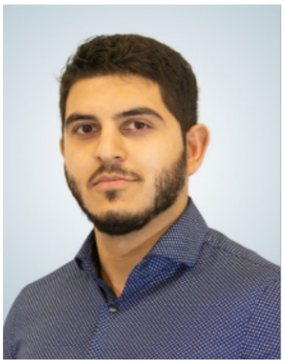
CATALYST DESIGN FOR THE HYDRODEHALOGENATION OF HALOMETHANES IN NATURAL GAS UPGRADING



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Co-examiner: Prof. Dr. Christoph Müller

ETH Hönggerberg, 15/07/2021, 10:00 am

Zoom Meeting ID: 320 031 1319

Project Summary: In the context of natural gas upgrading, halogen-mediated processes have emerged as attractive routes for the on-site valorization of methane into fuels and chemicals. Nonetheless, the formation of dihalomethanes (CH2X2; X = Cl, Br) during methane halogenation is detrimental, contributing to halogen and carbon-losses in the downstream halomethanes (CH3X) upgrading step. The reforming of these byproducts into CH3X via selective catalytic hydrodehalogenation is a realistic approach. However, limited understanding hinders the development of promising catalysts. This project addresses this aspect by a fundamental catalyst-by-design approach, combining synthetic strategies with experimental catalyst evaluation and kinetic studies, in depth characterization, and advanced mechanistic studies complemented by computational modelling and statistical tools in order to derive synthesis-structure-performance relationships. This approach revealed a global performance descriptor, based on the adsorption strength of the halogen/carbon fragment on the active metal phase (Fe, Co, Ni, Cu, Ru, Rh, Ag, Ir, and Pt), allowing the rationalization of reactivity patterns over the benchmark SiO2-supported nanoparticles, which still suffer from limited selectivity and/or stability. Engineering the metal phase at the nanoscale resulted in catalysts with enhanced performance, including highly selective carbon-supported single atoms of Ru, Ir, and Pt that govern the availability of surface H-atoms, epitaxially-directed Ir morphologies on rutile TiO2 for increased hydrodechlorination reactivity and stability against active phase sintering, as well as carbon-supported bimetallic Ir-Ru nanoparticles that display unparalleled selectivity and stability in hydrodebromination. The results provide design criteria for the development of superior hydrodehalogenation catalysts that can bring current halogen-mediated technologies one step closer to industrial realization.

CV: A. Saadun received a double degree LLM in International Business Law at Tilburg University and an MSc in Chemical Engineering at the Eindhoven University of Technology in 2017. In the same year, he joined the group of Prof. J. Pérez-Ramírez to pursue his doctoral studies.

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