Homogeneous Catalysis for Sustainable Chemistry

Martin Nielsen

DTU Chemistry,
Technical University of Denmark, Kemitorvet 207, 2800 Kgs
Lyngby, Denmark email: marnie@kemi.dtu.dk

To reach an environmentally sustainable society, fundamental new developments must be made within the vast field of chemistry. For example, CO_2 utilization, e.g. for use as building block in the chemical industry rather than simple CO_2 storage, can significantly contribute to CO_2 abatement, biomass upgrade to commodity chemicals can lead to a carbon negative footprint, and liquid chemical H_2 storage can revolutionize the energy- and fuel- grids. However, despite these colossal potential benefits, the developments of all three sustainable transformations are in their infancy and are yet to become viable industrial processes.

The use of homogeneous catalysis has long been recognized as the superior choice with respect to reaction selectivity and efficacy. Nevertheless, it remains to be seen that this type of catalysis can mediate any of the above-mentioned transformations under sustainable conditions while maintaining a high efficacy and long catalytic-system longevity.

The Nielsen lab has repeatedly shown how biogenic compounds such as ethanol, levulinic acids, furanics, mono- and polysaccharides, and even raw biomass all are converted to industrially value-added compounds such as primary- and secondary alcohols, hydrocarbons, furfuryl alcohols, and γ-valerolactone (GVL) using organometallic catalysis under mild condition and with best-in-class catalyst performances.

In my talk, I will show how several of these reports not only represent state-of-the-art reactivity profiles, but also disclose novel catalytic behaviour. For example, we demonstrate the first example of ethanol upgrading to secondary alcohols [1], which will open for the usage of bioethanol as platform compound to a much larger library of target molecules than we see today.

We furthermore show that Noyori-type pincer catalysts can be rendered catalytically active for hydrogenation in the presence of an acid, even though this catalyst type has long been recognized to require either neutral or basic conditions to stay functional. I will demonstrate its high usefulness by showing that this unique combination of acid and a hydrogenation catalyst allows for unprecedented valorisation of numerous biomass waste sources to GVL [2].

I will also show how a novel combination of an organometallic catalyst and ionic liquid provides energy-efficient reversible CO_2 hydrogenation to formic acid [3]. Such a system is a potential powerful tool for H_2 storage, e.g. in H_2 batteries, and in hydrogen-based transportation.

Finally, I will share recent findings on how a novel in-house developed organometallic complex salt shows different chemoselectivities for catalytic transfer hydrogenation of aldehydes, ketones, alkenes, and alkynes depending on the choice of the seemingly innocent counter anion to the complex cation [4].

References

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[3] L. Piccirilli, B. Rabell, R. Padilla, A. Riisager, S. Das and M. Nielsen, *J. Am. Chem. Soc.*, **2023**, *145*, 5655.
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