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Homogeneous Catalysis is Up for the Challenge

Lutz Ackermann*^[a] and Jean-Baptiste Sortais*^[b, c]

The articles in this Special Issue illustrate how homogeneous catalysis can meet and overcome the current challenges in the field, ranging from the valorization of biomass and the forma-

tion of advanced materials to the synthesis of sustainable fuels, and the step-economical preparation of novel value-added chemicals.

In recent years we have witnessed the emergence of exciting new directions in homogeneous catalysis research. Innovative concepts have set the stage for major advances in supramolecular catalysis and the development of Earth-abundant 3d metal and main group element catalysts. Likewise, momentum was recently gained by establishing novel strategies for photo-redox- and electro-catalysis, thereby enabling the sustainable use of renewable energies. The articles that comprise this Special Issue illustrate how homogeneous catalysis can meet and overcome the current challenges in the field, which include the valorization of biomass and the formation of advanced materials to the synthesis of sustainable fuels, as well as the step-economical preparation of novel value-added chemicals. This Special Issue comprises 31 contributions from world-renowned research groups, including one Review, three Minireviews, one Concept article, fourteen Communications and twelve Full Papers.

The transformative nature of supramolecular catalysis is illustrated by the rational redesign of ligands to enable the supramolecular substrate preorganization for regioselective hydroformylations, homochiral molecular helices for asymmetric amplification, and host-guest complexation with sulfonatocalixarenes. In contrast, the challenges and opportunities of metal-free approaches are summarized for halogen and chalcogen-bond donor organocatalysis, tetrazine-catalyzed pericyclic reactions, phenol phenothiazine coupling, and NHC-catalyzed ester syntheses with abundant CO₂. The valorization of abundant

feedstock materials is efficiently accomplished by transition metal catalysts, including innovative tantalum complexes for the hydroaminoalkylation of easily-accessible alkenes. Ruthenium complexes catalyze the base-free reduction of CO₂, the dehydrogenative coupling of alcohols and amines, and they enable the use of methanol as C1 source for the β -methylation of alcohols. Furthermore, the mechanochemical synthesis of a series of Ru olefin metathesis catalysts was reported. Formates were identified as viable CO surrogates, and could be employed in reductive N-alkylations. The CO₂ reduction is likewise accomplished by novel main-group element hydrides. Abun-

Lutz Ackermann studied chemistry at the Christian-Albrechts-University Kiel, and obtained his PhD in 2001 with Alois Fürstner at the MPI für Kohlenforschung in Mülheim/Ruhr. He was a postdoctoral fellow with Robert G. Bergman (University of California at Berkeley) before initiating his independent research program in 2003 at the Ludwig-Maximilians-University München. In 2007, he became Full Professor at the Georg-August-University Göttingen, where he served as the Dean of Research and Dean of the Faculty of Chemistry. The development of novel concepts for homogeneous catalysis and their applications to sustainable organic synthesis as well as molecular labeling are among his main current research interests.



Jean-Baptiste Sortais was educated at the École Normale Supérieure de Lyon. He received his MSc in organic chemistry from the Université Louis Pasteur, Strasbourg, where he completed his PhD in 2007 under the supervision of Dr. M. Pfeffer. After a first post-doctoral stay with Prof. J. E. Bäckvall at Stockholm University, he then joined the group of Prof. G. Erker at the University of Münster. In 2009, he was appointed as an Associate Professor at the Université de Rennes 1. In 2017, he was promoted to Full Professor at the Université Paul Sabatier, Toulouse 3, performing his research at the Laboratoire de Chimie de Coordination (LCC-CNRS). His research mainly focuses on the design and development of new complexes based on Earth-abundant transition metals for catalysis.



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dant H₂O is efficiently oxidized in terms of oxygen evolution by well-designed NHC-iridium catalysts, and the formation of iridium nanoparticles during water oxidation was conveniently monitored by real-time small angle X-ray scattering for a number of molecular iridium catalysts. Less expensive copper catalysts can likewise be employed for H₂O oxidation enabled by electrocatalysis. The enormous potential of and topical interest in Earth-abundant 3d metal catalysts is reflected by various contributions, highlighting the major advances in copper, nickel, cobalt, manganese and iron homogeneous catalysis. Hence, copper catalysts have enabled efficient radical cross-couplings of oxime esters and sulfonates as well as transformative Suzuki-Miyaura reactions of fluorinated aryl boronate esters. While nickel catalysts were designed for ethylene (co)-polymerizations, homogeneous iron catalysis set the stage for styrene aziridinations, alkyne aminocarbonylations, alkene epoxidations, and Het-H transformations, with the latter featuring detailed mechanistic insights of the carbene transfer reaction. In contrast, notable advances have been witnessed in manganese catalysis by the elegant design of effective hydrogenation

and C–H activation catalysts. The increasingly potent nature of homogeneous cobalt catalysis is mirrored by effective aminations with organozinc pivalates and its recent merger with photoredox- and electro-catalysis towards step-economical C–H activations, among others. Given the transformative power of homogeneous catalysis summarized herein, we feel that exciting future developments are expected in this rapidly evolving research area.

Finally, we would like to take this opportunity to sincerely thank all the authors and reviewers for their great contributions to this Special Issue dedicated to the new directions in homogeneous catalysis. We hope you will enjoy reading this issue as much as we did.

Keywords: Asymmetric catalysis • Electrocatalysis • Homogeneous catalysis • Organocatalysis • Photocatalysis • Supramolecular catalysis