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Sustainable catalytic conversions of renewable substrates

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The conversion of biomass and the platform molecules derived from it into more sustainable chemicals and fuels, is a rapidly evolving field of research. Efficient valorization of such renewable feedstocks will require many innovations in the entire process chain ranging from crop optimization by bioengineering, improved pretreatment processes to separate the biomass feed into its components and finally the selective, energy- and resource-efficient conversion of the constituents into value-added fuels and chemicals. While many disciplines of science are involved in this effort, one field is considered to be particularly instrumental to the eventual realization of these biomass-based conversion routes: catalysis. Indeed, catalysis is considered as a key, enabling technology for the development of future, competitive biorefineries. These biorefineries will need to be developed in an analogous manner to the current petrochemical refineries, wherein both chemicals and fuels are produced in a collection of highly integrated and highly optimized processes. Today, virtually all fuels and over 85% of the chemicals that come out of petrochemical refineries have seen at least one catalytic conversion step in their production process. Importantly, such conversions are performed with extremely high carbon efficiencies; that is, most of the carbon atoms in the various fractions of the feed are found in the end products. Biorefineries should therefore also aim to leave no component behind, to produce little to no waste, and to produce both biobased fuels as well as chemicals.

For biofuel production, one should realize that these fuels will constitute only part of the future renewable energy mix, and might even be a transitional or niche solution as longer-term technological developments will almost certainly be based on the direct conversion and storage of energy from sources such as the sun and wind. The important, but partial contribution that biofuels will make to the sustainability challenge that we face should certainly be factored in when biomass availability and sourcing is considered.

For chemical production, biomass has the potential to become a dominant feedstock in the future chemical industry. High value-added products derived from biomass can provide an important driver for the desired and necessary transition to sustainable fuels and commodity chemicals. As such, two conversion paths can be followed: either convert biomass into existing chemicals (*e.g.*, 'drop-in' or chemically identical green replacements), or generate new building blocks/end products for the chemical industry.

Drop-in biobased chemicals offer the advantage of addressing an existing market and using an existing infrastructure, but have to be produced in a cost-competitive manner. From the point of view of economic viability, biobased chemical production will benefit from the recent surge in shale gas production. On the one hand, hydrogen availability will continue to increase, thus decreasing restrictions on reductive processes; and on the other, the product distribution from traditional refeineries will continue to change, thus generating new opportunities for biobased processes. For instance, the replacement of naphtha with lighter feeds from shale gas in crackers for ethylene production has resulted in a drastic change in the chemical composition of the cracker output and, consequently, has affected the supply of key bulk chemicals such as butadiene and BTX. These scenarios clearly open up opportunities for the development of competitive biobased routes for the production of chemicals.

Biobased routes to new products offer the possibility of improved or unprecedented material properties in addition to the inherent sustainability of the product. Many new products have been reported over recent years, but have been often derived from relatively few building blocks (*e.g.*, polylactic acid). Newly developed platform molecules, such as 5-hydroxymethylfurfural or γ-valerolactone, are thought to have the potential to ultimately replace the olefins and aromatics that are the building blocks of the chemical industry of today. However, market penetration for new products is not an easy task, and

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the development of central building blocks from viable biorefinery schemes is critical for establishing the foundation for sound economical and technological assessments.

Within the boundaries, set by these developments in society and industry, many fundamental challenges are faced by those working on the development of new catalysts and new catalytic routes for the conversion of biomass to chemicals and fuels. First, the highly complex, highly oxygenated, highly polar and often polymeric nature of biomass is clearly quite different from the petrochemical apolar hydrocarbons for which most of the catalysts that we use today have been developed. Thus, selective conversion of biomass components often involves controlled defunctionalization and deoxygenation of the feed, which requires selective activation of specific functional groups in the presence of many others without resorting to the extensive protection/deprotection protocols typically used in fine chemicals production. The highly functionalized nature of biomass components also means that catalytic conversions will typically take place in the liquid phase, as the substrates cannot be readily volatilized without decomposition. Such liquid phase reactions, involving highly polar environments, high temperatures, and high pressures, put considerable demands on the stability of the catalyst. The challenges and opportunities for catalyst development are vast. Solutions are being developed from within the various fields of catalysis, including biological, homogeneous and heterogeneous catalysis. In some recent and exciting developments, multidisciplinary routes and approaches that cross the traditional boundaries are also being explored with considerable success.

Research efforts on catalytic biomass valorization are often part of national or international collaborations, be it (virtual) research schools, public-private partnerships, or transnational training programs. This themed issue is the result of one such program, Subicat. Subicat is a Marie Curie Initial Training Program that has started to forge a long-lasting collaboration between the Dutch 'Catalysis for Sustainable Chemicals from Biomass' consortium led by Utrecht University, and the cluster of excellence 'Tailor-Made Fuels from Biomass' (TMFB) from RWTH Aachen, Germany and the 'Critical Mass grouping' of Sustainable Biomass Catalysis at EaStCHEM in the UK and several industrial partners. The multidisciplinary overview of state-of-the-art technologies for the use of renewable resources given by world leaders in the field at the 2013 symposium 'Sustainable Catalytic Conversions of Renewable Substrates' at the University of St. Andrews, formed the basis of this themed issue. The various disciplines relevant to sustainable catalytic conversions of renewable feedstocks, including homogeneous, heterogeneous and biocatalysis, industrial catalysis, chemical engineering and biology were covered at the symposium and are well represented in this themed issue. Indeed, the 19 different contributions from various leaders in the field, highlight the breath of the challenges faced in this area and the powerful solutions that can be achieved with catalysis. The contributions focus on a variety of important topics that range from developing new catalytic routes, and designing non-noble metal alternatives, to the understanding of the relation between catalyst structure and molecular activation with state-of-the-art theoretical, spectroscopic, and reactivity approaches. The many different reactions, types of catalysts and methods surely make this an exciting and timely issue.