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Unlocking the potential of a sleeping giant: lignins as sustainable raw materials for renewable fuels, chemicals and materials

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Lignin, one of the three main components of non-edible, lignocellulosic biomass, is a renewable feedstock with great potential. Given its high abundance, constituting up to one-third of the dry weight of the biomass, the efficient valorisation of this aromatic biopolymer is of paramount importance for the economic viability of current and future biorefinery operations. Currently, the pulp and paper industry liberates approximately 50 million tons of degraded lignins as part of the 130 million tons of kraft pulp produced annually. Notably, the first commercial scale plants for the production of cellulosic ethanol are also starting to significantly contribute to further lignin generation. The rapid growth of this new industry is evidenced by the production of 125 million liters of cellulosic ethanol in the United States in 2014, meeting the volume mandated by the Renewable Fuel Standard for the first time. For 2016, the annual volume standard, as set by the Environmental Protection Agency, should soar to 780 million liters per year. Importantly, for every liter of cellulosic ethanol produced, about 0.5–1.5 kg of lignin will be co-generated depending on the type of lignocellulose employed in the process. Taking into account that about 40% of the lignin-rich residue would suffice to cover the heat and power demand for bioethanol production (including biomass pretreatment and ethanol distillation), it is clear that cellulolytic enzyme lignins will add to the already enormous pile of underutilised, technical lignins from kraft mills.

The potential of lignin lies in its structure, since lignin is the most abundant, renewable resource of aromatics on Earth. Worth mentioning here is the fact that the term 'lignin' or 'native (proto)lignin' refers to a class of phenolic biopolymers that occur in plant cell walls together with cellulose and hemicelluloses, rather than to a well-defined, regular biopolymer, such as cellulose. Despite the potential of lignins, their complex and random structures pose serious challenges for their efficient valorisation. Adding to the complexity, the structure of native (proto)lignin (*i.e.* the polymer occurring in the plant cell wall) depends strongly on the plant species and shows local and seasonal variation. Importantly, the pretreatment or pulping processes performed on lignocellulose cause extensive changes in the original lignin structure and, depending on the severity of this treatment, the isolated technical lignin will possess quite a different chemical connectivity than that of native lignins for which most of the structural representations are drawn. Fortunately, nowadays much is known about the structural features of native and, to a lesser extent, technical lignins. Such structural information constitutes a very useful guide to the design of valorisation strategies.

Having mentioned these challenges, it is very exciting to see that the scientific community interested in lignin applications and catalytic conversions has rapidly expanded in the last decade or so. Undoubtedly, after all the pioneering work done in the second half of the last century, a new wave now again seems to be emerging, bringing multifarious advances in lignin research.

The scientific challenge of coping with lignin's chemical complexity requires a multifaceted, translational approach, which involves the development of new analytical tools for structure characterisation of substrates and products, the search for new materials applications for lignins, and the development of mild and selective catalytic depolymerisation strategies, as well as harsher methods to deal with the recalcitrant technical lignins. For the depolymerisation strategies, detailed insight into the chemistry of the cleavage of certain lignin linkages can be obtained from model compound studies, while the application of new and known conversion strategies to real lignins urges us to deal with the challenges posed by the structural heterogeneity of lignin, its propensity to recondense, and the effect of any impurities contained in the stream.

This issue collects around 20 contributions from various fields of lignin valorisation. The collected articles present original research papers as well

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as critical reviews on topics ranging from lignin analytics, the use of lignin in new functional materials, new methods for the isolation of lignins, and homogeneous, heterogeneous and biocatalytic pathways for the production of chemicals and fuel components. Given the current flurry of activity in this field of research, this themed issue seems very timely and aims to represent advances made in the several multidisciplinary approaches that are required to tackle the lignin challenge. Only then will we be able to unlock the real potential of this sleeping giant.