BIOMASS VALORISATION

Controlled cellulose decomposition

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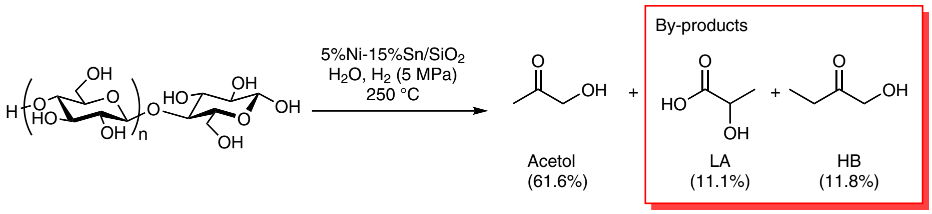

The conversion of cellulose into useful C3 chemicals holds great promise for the sustainable preparation of different molecules of interest to the pharmaceutical and chemical industry. However, depending on the desired product, limited yields and selectivity may represent an insurmountable obstacle towards commercial applications, as it is the case for acetol. Now, Ying Zhang, Longlong Ma and co-workers report a strategy to produce this chemical with significant yields via catalytic hydrothermal conversion of cellulose in the presence of hydrogen gas.

Previous investigations had revealed a promising reactivity for nickel–tin composites as catalyst for cellulose processing. Such combination of metals, in fact, can work in synergy, promoting the formation of C3 chemicals, albeit the reported yields were in the range of just 30%. In this study, the group set out to improve the efficiency of the process by systematically screening the properties of a series of catalysts based on Ni and Sn supported on SiO_2. By varying the amount of metals in the composites, they identified 5%Ni-15%Sn/SiO_2 as the catalyst featuring the best combination of hydrogenation sites and Sn-basic sites. The latter are capable of promoting glucose isomerization and retro-aldol condensation to yield C3 species. In this way, the desired acetol product could be obtained in 61% yield under optimized conditions, with lactic acid (LA) and 1-hydroxy-2-butanone (HB) as the major by-products (pictured).

Controlling the hydrogenation ability was particularly important to prevent over-reduction. Structural analysis based on X-ray diffraction and photoelectron spectroscopy suggested the formation of a Ni_3Sn_4 alloy as the crucial active species. In fact, control experiments with catalysts containing different Ni/Sn ratios confirmed that this specific alloy composition is the most effective in limiting undesired acetol over-hydrogenation. Moreover, the alloy positively influences the electronic properties of Sn species, favouring the chemistry responsible for the splitting of glucose into smaller fragments. This work showcases the importance of structure–activity relationships studies for the design of superior catalysts in the area of biomass valorization and renewable chemicals.

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