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Development of grafted acid-base catalysts for biomass related transformations

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In recent years there is a growing need to develop greener processes, where the production of noxious substances can be avoided. The utilization of renewable resources such as biomass is a promising route for developing sustainable processes.

Aldol condensation is an important reaction in green chemistry, enabling the upgrade of biomass-derived molecules to fuels and chemicals. Performing this reaction in the Liquid phase using heterogeneous catalysts is more sustainable as opposed to using homogeneous mineral bases. Therefore, the development of reusable and highly active solid catalysts for liquid reactions is drawing increasing interest.

Cooperative interactions between acid and base active sites can produce a more active catalyst for the aldol reaction, which in turn can facilitate running the reaction under milder conditions. However, making a synthetic heterogeneous catalyst that utilizes cooperative interactions between active sites is a great challenge. Cooperative catalytic interactions are highly dependent on the coexistence of both the acid and base functionalities in sufficient proximity and correct orientation. Without the correct positioning of both functions, a significant reduction in catalytic activity is observed. This is where the flexibility of polymers can be leveraged towards tuning the right molecular level conformation. However, the main drawback of such flexible supports is their structure collapse, which limits the accessibility of the reactants to the active sites. This can be solved by partial crosslinking of the polymer, which stabilizes the structure.

In this work, a hybrid metal-polymer system is used to study the critical parameters involved in inducing cooperative catalytic interactions in the probe Henry reaction. To incorporate acid and base active sites, metal sites are grafted to the hydroxy groups of chitosan (CS) polymer, which is inherently functionalized with basic amine sites. The grafting of the metal to CS stabilizes the structure as well as introduces Lewis acid sites.

The observed results show that the synthesis conditions have a significant effect on the 3D structure of the obtained materials, which in turn affect the reaction activity and selectivity. Combining several spectroscopic techniques with kinetic measurements allowed us to determine the effect of the material structure on the catalytic activity.