One-pot two-steps hybrid catalysis for 5-HMF valorization

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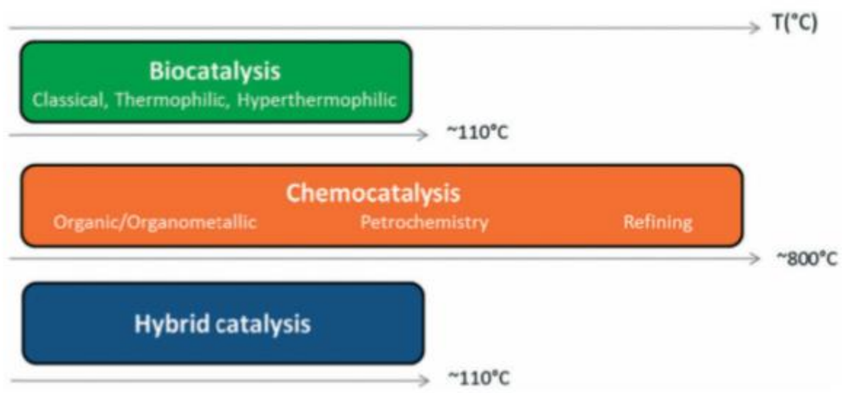
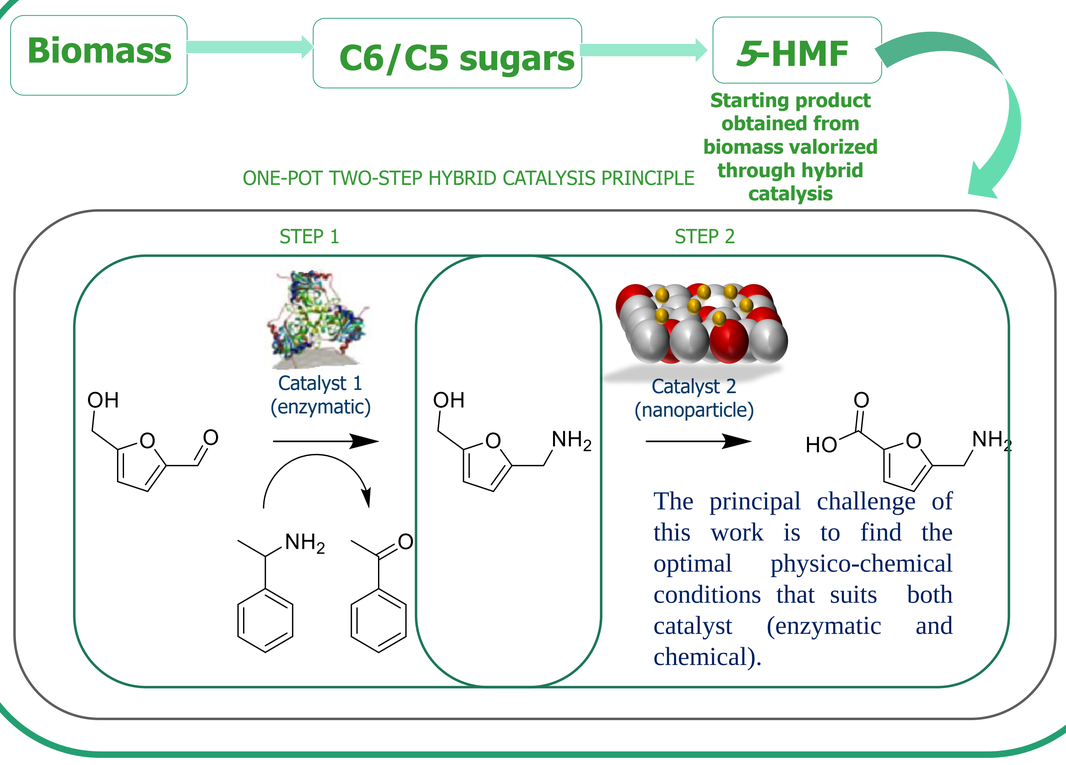
Introduction to hybrid catalysis

Enzymatically catalyzed reactions are highly selective and work very well under mild conditions (pH, buffer, temperature). However, higher scale extrapolation is limited to a certain class of stable and easily recyclable enzymes. Homogeneously and/or heterogeneously catalyzed reactions are often limited by the multiphasic nature of catalytic systems and/or variations in the distribution of active sites. For these reasons, the development of highly selective and efficient hybrid catalysts is especially needed, and it is now well admitted that disruptive innovation is highly desirable in process engineering, (bio)catalyst design and materials science. Optimal integration both types of catalyst is the key for efficient global implementation of sustainable processes in the hybrid catalysis field.



Strategy



Experimental

Thermo scientific caroussel (96 reactions)

Reactions system :

- 12 or 96 reactions system
- Susbtrate- *5*-HMF
- Amino donor- MBA, Alanine
- H_2O_2

Radlays caroussel (12 reactors)

Characterization and Analysis:

- *ICP, BET*
- XRD
- TEM
- HPLC
- NMR

Nanoparticles characterization

The Au-based catalysts were prepared using the two-step sol immobilization method where gold nanoparticles of similar size are first prepared and then immobilized in MgO through a reproducible method. PVA-stabilized Au NPs of average 3 nm, as determined by TEM (Figure 1A), had their size preserved after immobilization on different supports (Figure 1B). The Au NPs deposited on the supports were monodispersed with average size 3.6 nm ± 0.8 nm and no significant agglomeration of gold nanoparticles was detected.

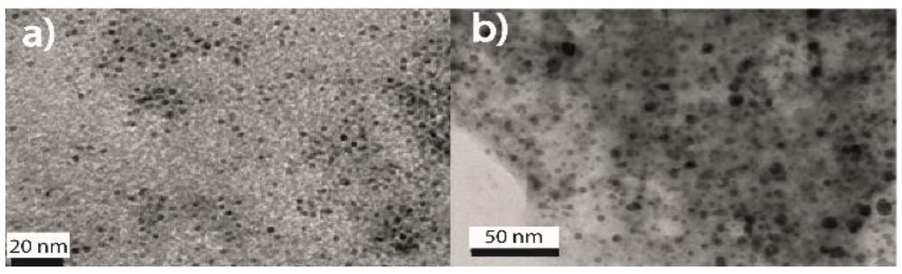
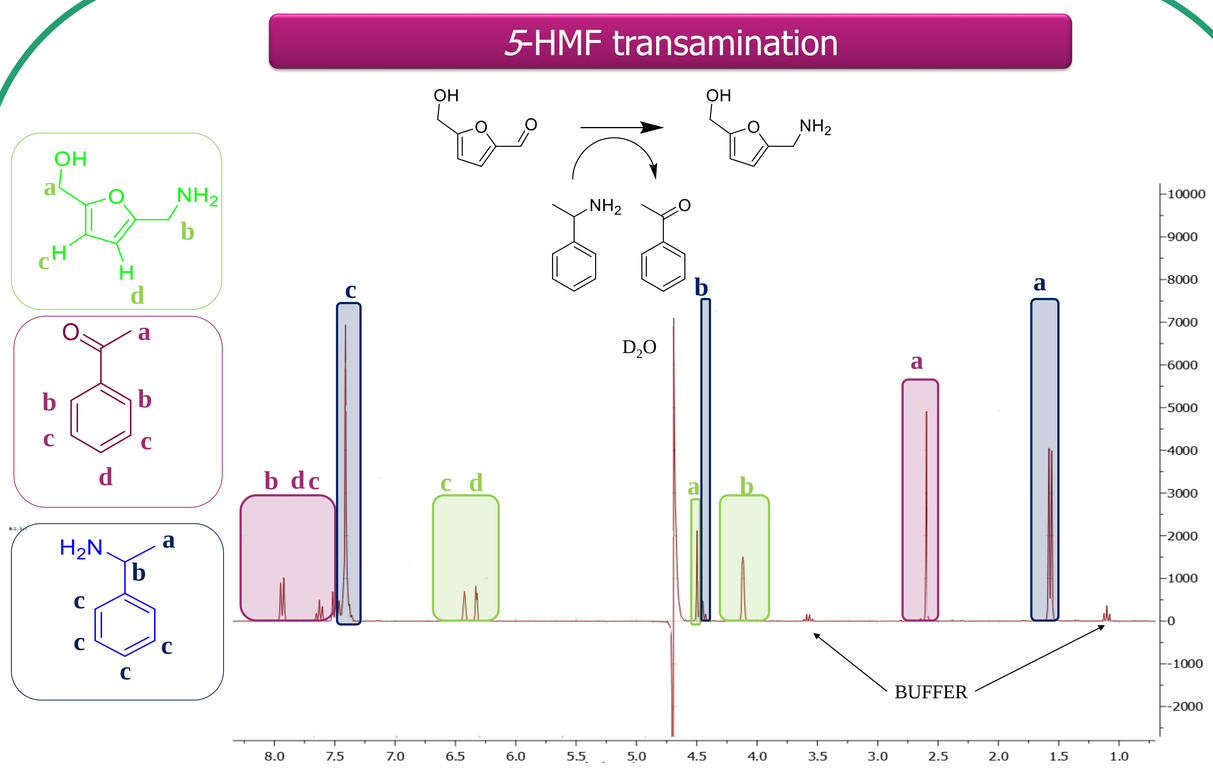


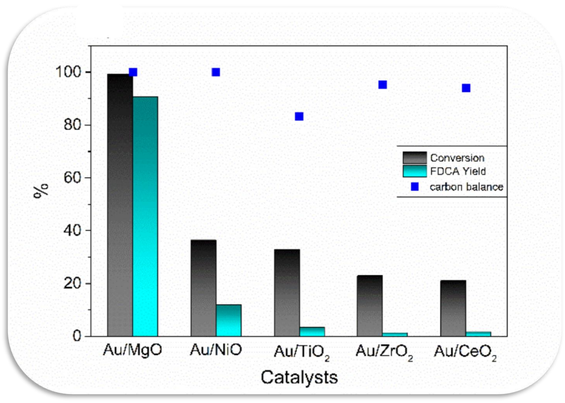
Figure 1. TEM images of the (a) unsupported gold NPs, (b) Au/MgO.

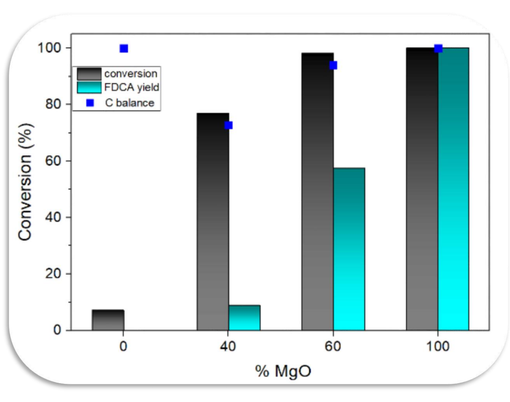
Results

5-HMF transamination



5-HMF oxidation





- Nanoparticles works very well on 5-HMF and successfully oxidize both functions (aldehyde and alcohol) into carboxylic acid. We assume that the presence of the amino function (after the first step in the hybrid catalysis process) will not disturb nanoparticles for the oxydation of the alcohol function.

Perspective

- The study confirmed the advantages of Au-based catalysts for the selective oxidation of HMF under the conditions used for the enzymatic part of hybrid catalysis .
- The enzymatic study confirmed that the transaminase we use is capable of transamination on the *5*-HMF at different pH (free end immobilised).
- The aim of this project is to perform this reaction in "one-pot one–step" with the transamination of the aldehyde function of 5-HMF and the oxidation of alcohol function into a carboxylic acid in order to obtain a recoverable product from the lignocellulosic biomass.
- This work could involve on the research of a new thermostable enzyme.
- It could also be based on the research of new chemical catalyst which are not disturbed by the presence of an amino function.

1. Isikgor FH, Becer CR. Lignocellulosic biomass: a sustainable platform for the production of bio-based chemicals and polymers. Polym Chem. 2015;6(25):4497-559.
2. Dumeignil F, Guehl M, Gimbernat A, Capron M, Ferreira NL, Froidevaux R, et al. From sequential chemoenzymatic synthesis to integrated hybrid catalysis: taking the best of both worlds to open up the scope of possibilities for a sustainable future. Catal Sci Technol. 2018;8(22):5708-34.