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Plasmon-enhanced gold catalysis: towards understanding the processes involved in the selective photo-oxidation of carbohydrates using Au/CeO2 photocatalyst

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PURPOSE OF THE ABSTRACT

Carbohydrates are the most abundant materials from biomass, mostly as lignocellulose (75% of plant biomass). Owing to their polyfunctionality, they pave the way to a wide range of derivative products that can be incorporated into cosmetic, detergent or pharmaceutical formulations. However, most of chemical transformations of carbohydrates do not entirely satisfy green chemistry principles because of multi-steps protocols (involving protection/deprotection stages), use of hazardous chemicals, and energy/time consumption. The promotion of greener methodologies for the modification of free carbohydrates is thus becoming increasingly desirable.

Heterogeneous photocatalytic transformations represent an ideal green methodology to modify free carbohydrates as long as high conversion yield and selective reactions under realistic light radiations can be reached. Our group proposed a fast (ie. < 10 minutes) and efficient photocatalytic procedure leading, under realistic A.M. 1.5G light conditions (sunlight illumination), to quantitatively and selectively oxidation of free mono- and oligosaccharides into corresponding sodium aldonates by means of Au/CeO2 photo-catalyst.[1] The plasmonic contribution of gold NPs in the photoinduced processes driving the reaction is not clear so far. For example, when O2 is used as oxidant, as frequently in literature, we demonstrated that the contribution of LSPR is totally annihilated, and only thermal activation induced by NIR radiation is responsible of the enhancement of the reaction under A.M. 1.5G illumination conditions.[2] Thus, understanding mechanisms involved in the reaction is essential, not only for the fundamental knowledge but also for the development of new photocatalysts, allowing to selectively convert other hydroxyl functions of the sugar.

In this communication, a particular emphasis will be given on the mechanism to explain the fast process, the versatility to different carbohydrates and the selectivity of this photo-oxidation reaction. We will discuss the influence of several parameters on the photo-oxidation of a model molecule: glucose. Based on this study, time-resolved spectroscopies and electron spin resonance experiments recorded under illumination, a plausible mechanism will be proposed. Moreover, we will discuss about the kinetic competition existing between surface catalysis and photocatalysis activity.

FIGURES

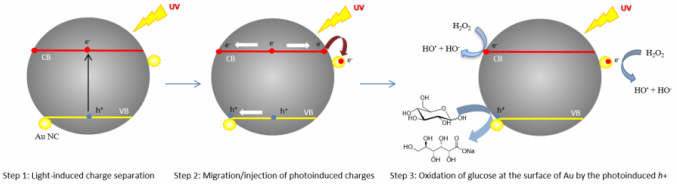

FIGURE 1

Figure 1

Proposed mechanism for the selective photo-oxidation of carbohydrates using Au/CeO2 photocatalyst

FIGURE 2

KEYWORDS

gold catalysis | conversion of free carbohydrates | photocatalysis

BIBLIOGRAPHY

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