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Efficient transfer hydrogenation of alkyl levulinates to gamma-valerolactone catalyzed by simple Zr/Co-TiO₂ bimetallic oxide

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PURPOSE OF THE ABSTRACT

Facing the depletion crisis of fossil resources and serious environmental pollution, more and more attentions focused on renewable resources valorization process [1]. gamma-valerolactone (GVL) as biomass downstream derivatives could be directly employed as raw materials to prepare high calorific value liquid hydrocarbon fuels and fuel additives through ring-opening hydrogenation reactions [2]. Thus, strategies for the production of GVL using catalytic transfer hydrogenation (CTH) from ethyl levulinate (EL) attracted dramatic interest due to the inexpensive, environmentally friendly nature and safer properties of short-chain alcohols as H-donor [3].

Latest work involved the preparation of TiO₂, 5%-10wt% Zr/TiO₂, 5% Co/TiO₂, as well as 5% Zr/Co-TiO₂ via a facile sol-gel hydrothermal method [4], further employed in the CTH process starting from EL to GVL using 2-ProH. Obtained catalysts were characterized by XRD, XPS, SEM, HR-TEM, FT-IR, ICP-OES, NH₃/CO₂-TPD, Pyridine-IR, H₂-TPR, and N₂ adsorption and desorption measurements. 5% Zr/Co-TiO₂ exhibited the highest catalytic performance with 95% EL conversion and 88% GVL yield under optimum condition (0.2 M EL, 15 mL 2-ProH, 190 °C, 11 h). The catalysts could still maintain the anatase TiO₂ type after Zr and Co doping. 5% Zr/Co-TiO₂ exhibited highest surface area and L/B acid ratio, Zr and Co elements showed a synergetic effect during CTH process. Co³⁺ and Co²⁺ were the active species, with the incorporation of Co onto Ti-Zr-O support increasing acidic and basic sites content. In addition, 5% Zr/Co-TiO₂ exhibited high stability in 3 times recycling experiments. A full reactivation of the spent catalyst was possible after 300 °C calcination in 2 h. This contribution no doubt paved the way for the biomass downstream derivatives valorization process.

FIGURES

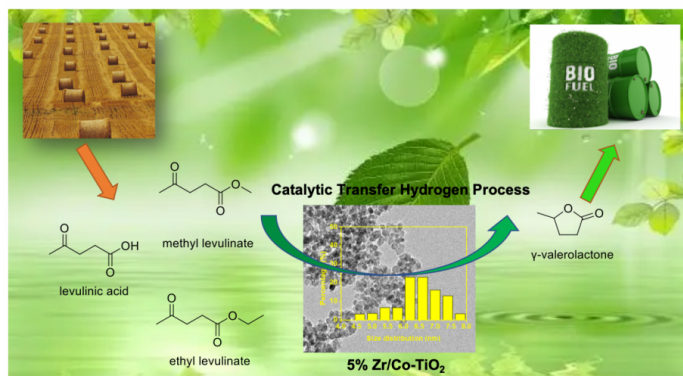


FIGURE 1

Scheme 1

Production of γ -valerolactone from levulinic derivatives via Catalytic Transfer Hydrogen process in the presence of 5% Zr/Co-TiO₂

FIGURE 2

KEYWORDS

Catalytic Transfer Hydrogen | γ -valerolactone | heterogeneous catalysis | bimetallic oxide

BIBLIOGRAPHY