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NEW INSIGHTS ON THE CATALYTIC REDUCTIVE AMINATION OF HYDROXYACETONE AMINATION OVER $\text{RuW}_x\text{C}/\text{AC}$ CATALYST

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Summary: The production of chemicals and liquid fuels from renewable and non-edible lignocellulosic biomass has been considered as a promising way to reduce our dependence to fossil resources as well as to reduce CO_2 release. Especially, nitrogen-containing molecules, particularly primary amines, are broadly used for the synthesis of pharmaceuticals, polymers, surfactants, agrochemicals, and dyes^[1]. Owing to the high O/C ratio (~1/1) in biomass feedstocks^[2], the production of oxygenates from biomass is rather straightforward and has been largely studied. However, the further production of valuable nitrogen-containing products is far less evident due to the deficit of efficient amination strategies of oxygenates. One way is the amination of aldehydes and ketones to primary amines, employing ammonia as the nitrogen source^[3]. Due to the development of biorefining, renewable aldehydes and ketones including glycolaldehyde, glyceraldehyde, hydroxyacetone, and aromatic compounds are nowadays available at large scales, opening new opportunities to produce nitrogen-containing compounds^[4]. For example, Liang *et al.* reported the use of partly reduced Ru/ZrO_2 for the reductive amination of different biomass-based aldehydes/ketones in aqueous ammonia^[5]. Despite this encouraging development, effective heterogeneous catalytic systems that allow the amination reaction to take place under milder conditions ($T < 100\text{ }^\circ\text{C}$, $P < 50\text{ bar}$, aqueous phase, and without additives) with high amines' yields are still lacking. In particular, the production of large-market amino alcohols from hydroxyacetone hasn't been reported so far.

Herein, we report the preparation of a highly efficient and robust catalyst, $\text{RuW}_x\text{C}/\text{AC}$, for the reductive amination of hydroxyacetone (Figure 1). By varying several process parameters including time, temperature, the nature of nitrogen source, and pressure, up to 60 mol.% amines' yield has been obtained. The promoting effect of tungsten carbide nanoparticles has been particularly investigated. Finally, a kinetic study has been conducted and will be discussed.

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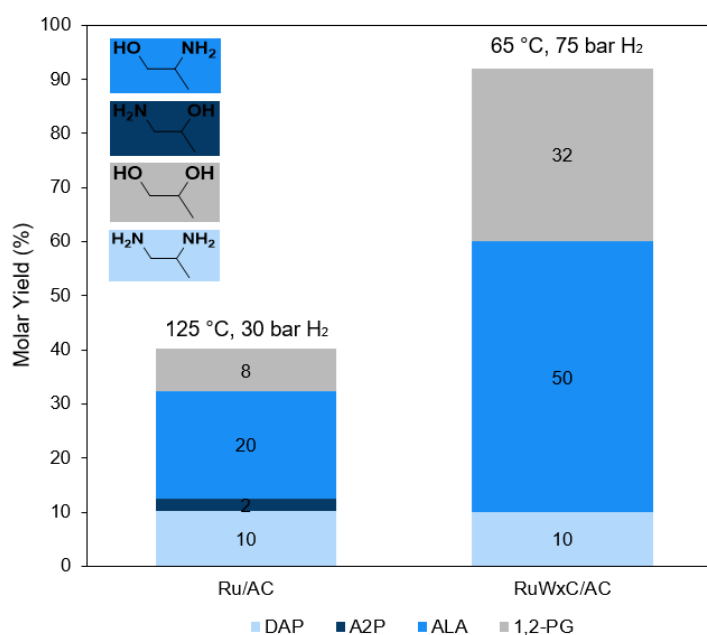


Fig. 1 Reductive amination of 10 g hydroxyacetone with 100 mL NH_3 (28 wt.%) and 1 g catalyst for 1 h.