

Investigating the effect of the defectiveness of spinel structure in ZnAl mixed oxide catalysts on the glycerol carbonation with urea

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In this study, we investigated how the defectiveness of ZnAl_2O_4 lattice structure influenced the catalytic performance in glycerol carbonylation with urea. The defectiveness in the catalysts was examined with three types of catalyst preparation: physical mixing of coprecipitated ZnO and ZnAl_2O_4 (p-ZnAlO), impregnation of ZnO onto ZnAl_2O_4 (i-ZnAlO), and citric acid-templated ZnAlO (c-ZnAlO). Zn atoms could substitute in the spinel structure to form inverse spinel structure of ZnAl_2O_4 lattice, producing the defectiveness in the ZnAl_2O_4 lattice structure with the order of p-ZnAlO < i-ZnAlO < c-ZnAlO. The order of defectiveness in the different types of catalysts was strongly related to not only the catalyst acidity but also the existence of Zn NCO complex on the solid surface, which enhanced the glycerol carbonate (GC) production with a high GC yield. The Zn NCO complex on solid surface played an important role as a Zn-containing intermediated with which glycerol reacted with urea not Zn to produce GC while the Zn NCO complex in liquid phase facilitated either the reaction between Zn and glycerol to form Zn glycerolate or the GC production by the reaction between glycerol and urea.