Effects of Au Addition onto Pt/MFI Catalysts on Partial Oxidation of Bioethanol to Acetaldehyde

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Acetaldehyde synthesis via partial oxidation of bioethanol is a green process that may replace Wacker process depending on fossil resources. We have found that Pt/MFI catalysts adding a small amount of Au provided higher acetaldehyde yield than those of Pt/MFI and Au/MFI catalysts¹). In this study, in order to gain more insights about the coexistence effect of Pt and Au, optimization of preparation condition for highest performance catalyst, Au(0.05)-Pt(0.95)/MFI zeolites, in previous report¹) catalysts was attempted, and effects of pretreatment on the electronic state of precious metals and the reaction behavior were studied. Furthermore, the influence of impurities in real bioethanol on the reaction was investigated.

Pt-Au/MFI were prepared by impregnation of HAuCl₄ and H₂PtCl₆ aqua solutions on MFI (Si/Al = 45). Pt and Au contents supported on MFI were 0.95wt% and 0.05wt%, respectively. To change the sates of Pt-Au cluster, Pt-Au/MFI(n) were calcinated at different temperature (n = 773K-1073K). EtOH and BioET (ethanol content 10wt%) used as reactant solutions were prepared by dilution of reagent ethanol and real bioethanol (Sanwa Starch, ethanol 74wt%) with distilled water. Reaction tests were carried out at 533-583K at 0.05mL/min of the reactant solution and 12.4 mL/min of air with 0.1 g of catalyst.

Acetaldehyde yields on Pt-Au/MFI(n) catalysts at 533K were shown in Fig.1. The yield was strongly depending on calcination temperature, and the highest yield achieved with Pt-Au/MFI(873). Surface areas of four catalysts were nearly equal to 350 m²/g, but Pt particle sizes of Pt-Au/MFI(873) and Pt-Au/MFI(773) were smaller than those of Pt-Au/MFI(973) and Pt-Au/MFI(1073). The higher calcination temperature than 973K leaded to sintering of Pt particles, which decreased dehydrogenation activity. On the other hand, Pt-Au(773) is less active than Pt-Au(873), and the activity may decrease for reasons other than sintering.

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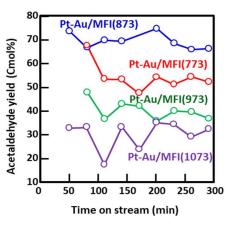


Fig.1 Acetaldehyde yields on Pt-Au/MFI catalysts at 533K for EtOH as a function of time on stream