Ethanol Conversion to Butanol over Small Coinage Metal Clusters: An Experimental and Computational Study

We illustrate the mechanism for the C–O, C–H and O–H bond activation in the coupling of ethanol to butanol over small coinage clusters (copper, silver and gold). It is found that charge transfer interactions between the clusters and the alcohol initiate their reactions allowing a chemisorption step. The binding energy is calculated, whereby ethanol adsorbs very strongly on Au in comparison to Ag and Cu. The nature of bonding is investigated using natural bond orbital (NBO) analysis and quantum theory of atoms-in-molecules (QTAIM). The reactive intermediates, activated complexes, transition states, and bond breaking on icosahedral $Au_{13,}$ Cu_{13}, Ag_{13} and also triangular $Au_{3,}$ Cu_3, Ag_3 have been calculated alongside the cycle kinetics. Furthermore, high resolution mass spectroscopy has been used to study the ethanol coupling reactions over small Au cluster catalysts. The observation of the coupling products concurs with the kinetic- and thermodynamic- allowed reaction pathway of Guerbet coupling of ethanol. The highest selectivity for butanol (61%) is obtained after a reaction time of 2 h while the highest ethanol conversion (91%) is obtained after a reaction time of 5 h.

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