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Transient FTIR spectroscopy for probing reaction pathways on Au catalysts¹

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Au is now well known to be an active catalyst if the Au particles are sufficiently small, less than about 5 nm. The causes for this structure sensitivity are now beginning to be better understood. Computational modeling and measurements of size dependence on a single catalyst are consistent with activity at sites with low coordination numbers, due in part to flexibility of adsorbate geometry in these sites. Although small size and low coordinate sites are important in catalyzing, e.g. the CO oxidation reaction, there appear to be other factors which control the observed activity as demonstrated by catalyst deactivation and unusual temperature dependence. We have performed studies of CO oxidation over Au/TiO₂, Au/SiO₂, Au/ZnO/TiO₂ and Au/FePO₄ catalysts to explore reaction pathways and the causes for activation and deactivation. Three different reactor systems, a fast gas transient FTIR spectrometer, a slower transient DRIFTS cell and a steady state plug flow reactor have been used to correlate activity with surface species. Using this *operando* approach the elementary steps in the CO oxidation reaction have been explored. Striking differences between the supports are found. The effect of various pre-treatments, the evolution of the surface species during “steady state” reaction and the role of carbonate, oxygen storage, water, hydroxyl upon catalyst activation and deactivation have been explored and will be described. Reaction pathways and mechanisms will be proposed and compared for the different catalysts.

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