

Abstract No. argo750

Reactivity of Site-Isolated Metal Clusters: Propylidyne on Gamma-Alumina-Supported Tetrairidium

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Beamline: X11A

To contrast the reactivity of supported metal clusters with that of extended metal surfaces, we investigated the reactions of tetrairidium clusters supported on porous γ -Al₂O₃ (Ir₄/ γ -Al₂O₃) with propene and with H₂. Infrared, ¹³C NMR, and extended X-ray absorption fine structure spectroscopy were used to characterize the ligands formed on the clusters. Propene adsorption on Ir₄/ γ -Al₂O₃ at 298 K gave stable cluster-bound γ ₃-propylidyne. Propene adsorbed on Ir₄/ γ -Al₂O₃ at 138 K reacted at approximately 219 K to form a stable, highly dehydrogenated, cluster-bound hydrocarbon species approximated as C_xH_y (such as, for example C₃H₂ or C₂H). H₂ reacted with Ir₄/ γ -Al₂O₃ at 298 K, forming ligands (likely hydrides), which prevented subsequent reaction of the clusters with propene to form propylidyne. Propylidyne on Ir₄ was stable in helium or H₂ as the sample was heated to 523 K, whereupon it reacted with oxygen of the support to give CO. Propylidyne on Ir₄ did not undergo isotopic exchange in the presence of D₂ at 298 K. In contrast, the literature shows that propylidyne chemisorbed on extended metal surfaces is hydrogenated in the presence of H₂ (or D₂) and exchanges hydrogen with gaseous D₂ at room temperature; in the absence of H₂, it decomposes thermally to give hydrocarbon fragments at temperatures much less than 523 K. The striking difference in reactivities of propylidyne on clusters and propylidyne on extended metal surfaces implies the requirement of ensembles of more than the three metal surface atoms bonded to propylidyne in the surfaces reactions. The results highlight the unique reactivity of small site-isolated metal clusters.