

Activity, Selectivity, and Stability of Subnanometer and Nanoscale Clusters

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In this two-part talk, computational studies will first be reported on the effect of size, composition, and support on the catalytic activity of a series of subnanometer clusters using density functional theory. Understanding these effects is instrumental for addressing fundamental aspects of catalysis that may lead to the development of new classes of catalysts. Our effort, which is integrated with experimental synthesis, characterization, and catalytic chemistry programs at Argonne, has resulted in the development of structure-property relationships on subnanometer pure metal clusters. Several examples will be given, including 1) activation of the C=C bond in propylene to form propylene oxide on alumina-supported Ag trimers and larger Ag aggregates, 2) oxidative dehydrogenation of propane to form propylene on alumina-supported Pt clusters, 3) methanol decomposition of alumina-supported Pd clusters. We will discuss how comparative studies of the clusters can be used to quantitatively determine the key reactivity descriptors for specific reactions. Initial results have already suggested, for example, that alloys of Pd and Cu might offer a compromise between the excessive CO binding that plagues Pd clusters and the sluggish kinetics of the Cu clusters.

The second part of the talk will focus on stability considerations associated with bimetallic transition metal alloys. Emphasis will be placed on the identification of stability descriptors relevant to realistic catalytic environments and on the efficient evaluation of these descriptors for a variety of alloy structures.