MAR15-2014-020480

Abstract for an Invited Paper for the MAR15 Meeting of the American Physical Society

Cluster size matters: Size-driven performance of subnanometer clusters in catalysis, electrocatalysis and Li-air batteries STEFAN VAJDA, Argonne National Laboratory

This paper discusses the strongly size-dependent performance of subnanometer cluster based catalysts in 1) heterogeneous catalysis, 2) electrocatalysis and 3) Li-air batteries. The experimental studies are based on I. fabrication of ultrasmall clusters with atomic precision control of particle size and their deposition on oxide and carbon based supports; II. test of performance, III. in situ and ex situ X-ray characterization of cluster size, shape and oxidation state; and IV.electron microscopies. Heterogeneous catalysis. The pronounced effect of cluster size and support on the performance of the catalyst (catalyst activity and the yield of C_n products) will be illustrated on the example of nickel and cobalt clusters in Fischer-Tropsch reaction. *Electrocatalysis*. The study of the oxygen evolution reaction (OER) on size-selected palladium clusters supported on ultrananocrystalline diamond show pronounced size effects. While Pd_4 clusters show no reaction, Pd_6 and Pd_{17} clusters are among the most active catalysts known (in in terms of turnover rate per Pd atom). The system (soft-landed Pd₄, Pd₆, or Pd₁₇ clusters on an UNCD Si coated electrode) shows stable electrochemical potentials over several cycles, and the characterization of the electrodes show no evidence for evolution or dissolution of either the support Theoretical calculations suggest that this striking difference may be a demonstration that bridging Pd-Pd sites, which are only present in three-dimensional clusters, are active for the oxygen evolution reaction in Pd_6O_6 . Li-air batteries. The studies show that sub-nm silver clusters have dramatic size-dependent effect on the lowering of the overpotential, charge capacity, morphology of the discharge products, as well as on the morphology of the nm size building blocks of the discharge products. The results suggest that by precise control of the active surface sites on the cathode, the performance of Li-air cells can be significantly improved