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Ab Initio Prediction of the Size-Dependence of Nano-scale Platinum Dissolution in Water KRISTIN PERSSON, LBNL, BYUNGCHAN HAN, GERBRAND CEDER, MIT — In low-temperature fuel cells, the mechanism behind the observed performance loss of the platinum catalyst is not well understood. Using ab initio methods, we calculate 0.5 - 2 nm diameter Pt nanoparticles with varying degrees of O and OH surface absorbates, optimized by site and particle surface structure. In fuel cells, the oxidation of the particle surface origins from the breakup of water molecules. To mimic these conditions we employ a grand canonical ensemble treatment of water as a source of O and H. Additionally, pH effects and dissolved species (from experiments) are incorporated, the latter by changing the experimental element reference state to that of calculated solids. This formalism allows us to determine the stability regions of nanoparticle Pt in equilibrium with water, as a function of particle size, potential and pH. As a result we find enhanced dissolution for the smaller Pt nano-particles, compared to the larger. Furthermore, surface passivation effects from O and OH adsorption do not significantly increase the stability of the nano-particle phases in the potential-pH region relevant for fuel cell operating conditions. Thus, we can identify size-dependent dissolution as a mechanism which will promote the growth of larger particles at the expense of smaller ones and ultimately cause a degradation in the nanoparticle Pt catalyst performance.

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