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First Principles Studies of Pt Electrode in PEM Fuel Cell

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Pt and its alloys are used as electrocatalysts in proton-exchange membrane (PEM) fuel cells. However, existing fuel-cell technologies still have many drawbacks, such as low catalytic activity for oxygen reduction reaction (ORR), dissolution of Pt atoms and coarsening of Pt nanocrystals on electrodes. ORR on cathode is a complex multi-electron transfer process and its reaction mechanism is still unclear partially because of the difficulties of direct investigation on its reaction intermediates, such as O_2*, OOH*, O* and OH*(* means adsorbed state). We analyze the charge states of all ORR intermediates adsorbed on Pt (111) surface based on first principles calculation and find all of them are in near-neutral states, which enables one to greatly simplify the analysis and modeling of ORR electrocatalysis. Beside surface adsorbates, we also investigate initial surface oxide formation on Pt electrode during anodic polarization, which induces the dissolution of Pt atoms during cathodic polarization. On Pt surfaces we find oxide structures whose equilibrium formation potentials satisfy with experimental values. Finally we study the diffusion process of Pt adatoms on Pt (111) surface, which could be the rate limiting step for coarsening of platinum nanocrystals. Pt adatom diffusion mechanisms and activation barriers are found to be quite different with variant surface adsorbates (H* or O*), providing possible method to impede the coarsening of Pt particles.