Spectro-Electrochemical Characterization on H/D-Pd Monolayers Coated Au Single-Crystal Electrodes and Nanoparticles

Yao-Hui Wang[1], Ru-Yu Zhou[1], Yu-Lin Sun[1], Sai Duan[2], Yue-Jiao Zhang[1], Jin-Chao Dong[1], Jian-Feng Li[1]*, Zhong-Qun Tian[1]*

[1]State Key Lab of Physical Chemistry of Solid Surfaces; Fujian Science & Technology Innovation Lab for Energy Materials of China; Department of Chemistry, College of Chemistry and Chemical Engineering, Xiamen University; Xiamen 361005, China.
[2]Department of Chemistry, Fudan University; Shanghai 200433, China.
E-mail: zqtian@xmu.edu.cn, li@xmu.edu.cn

Adsorption/absorption/reaction of hydrogen and deuterium on/in palladium (H/D-Pd) are important in both fundamental research and application, especially for hydrogen energy and other unexplored energies. The behaviour of H/D in subsurface of Pd metal is distinctively different from that at the surface and in the bulk phase. It could be possible to have overloading of H/D in the subsurface and create some unique lattices and configurations with input energy pulse. It's therefore highly desirable to comprehensively understand the H/D-Pd subsurface systems on many aspects. However, it's not easy to isolate the subsurface experimentally and avoid large discrepancies between the experiment and theoretical calculation. Recently, we have tried to systematically study H/D-Pd monolayer(s) coated on structurally well-defined Au single-crystal electrodes and nanoparticles. Two methods, *i.e.*, indirectly redox replacement of Cu UPD layer and directly electrochemical deposition of Pd, were developed to well control the Pd atomistic layer from 1 to 3, 5 monolayers over Au(111), (100) and (110) facets and Au nanoparticles of ~ 50 nm in diameter, respectively. The Pd layer, crystal facet, applied potential and electrolyte dependences were studied using cyclic voltammetry and in-situ Raman spectroscopy. The metal surfaces and subsurface of various monolayers were modelled with the first-principles computations using the Vienna ab initio simulation package (VASP), which will be discussed in details.