WILDE LAB.

[Understanding & Control of Hydrogen Absorption]

Department of Fundamental Engineering

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Nanoscale Surface Physical Chemistry

Graduate School of Engineering, Department of Applied Physics

How does hydrogen enter through surfaces into metals?

Nanoscale Analysis of Hydrogen Penetration, Diffusion, and Reactions at Surfaces and Interfaces

Hydrogen absorption into bulk metals and nanoparticles plays a key role in the industrial storage and purification of H_2 for clean energy applications such as fuel cells and also in hydrogenation catalysis. This research clarifies the microscopic reaction routes that connect the hydrogen states in the H_2 gas, adsorbed on the surface, and in the interior of H-absorbing metals such as palladium (Pd). Atomic level insight into the surface penetration process is obtained by using isotopic labeling (H, D) and well-characterized model surfaces of pure and modified (alloyed) Pd single crystals. The fundamental understanding of the absorption mechanism paves the way for the controlled design of novel highly efficient hydrogen storage materials and selective hydrogenation catalysts.

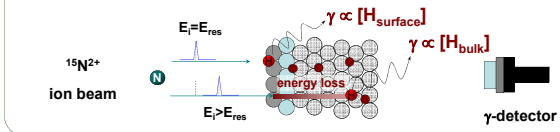
◆ Experimental Techniques & Key Information

- ✓ *Nuclear Reaction Analysis (NRA)*: Non-destructive・Quantitative・High-resolution - Hydrogen Depth Profiling ・Visualization of H-breathing by metallic nanocrystals and hydride nucleation beneath surfaces
- ✓ *Thermal Desorption Spectroscopy (TDS)*: Bonding stability evaluation of hydrogen species・Hydrogen absorption kinetics・Gas/surface/subsurface-H exchange mechanism・Kinetic isotope effects

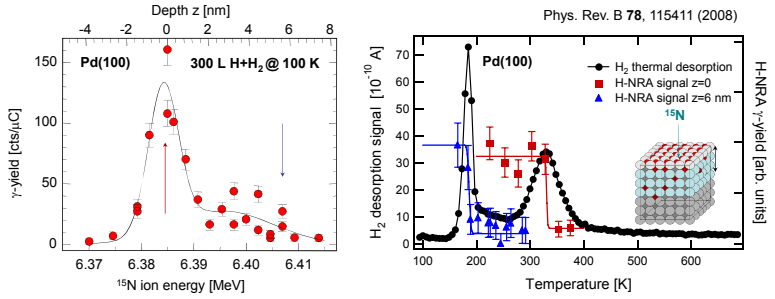
◆ Latest Research Topics

- ✓ *Hydrogen storage* → H-Absorption/Release Mechanism
- ✓ *Hydrogenation Catalysis* → Reactivity of 'Subsurface-H'
- ✓ *MOS Devices* → Relation of H-Impurities and Reliability

Principle of NRA $^{15}N(6.385\ MeV) + {}^{1}H \rightarrow {}^{12}C + \alpha + \gamma(4.43\ MeV)$

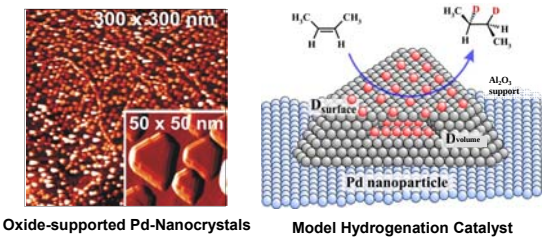


Identification of *surface-adsorbed* and *'subsurface'-absorbed* hydrogen (NRA+TDS)



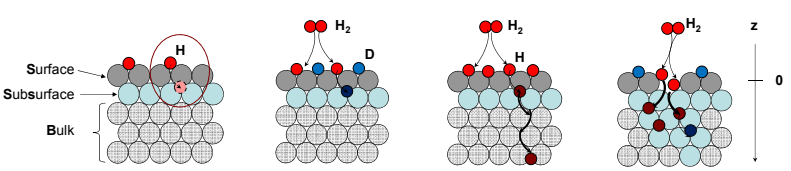
Relation of hydrogen absorption in Pd-nanocrystals and catalysis

Angew. Chem. Int. Ed. **47**, 9289 (2008); Phys. Rev. B **77**, 113412 (2008)



NRA proves directly the necessity of **nanocrystal-absorbed hydrogen** for **catalytic olefin hydrogenation.**

Atomistic mechanism of surface penetration and hydride nucleation in Pd crystals



Spontaneous $H_s \rightarrow H_{ss}$ transitions without H_2: Rare events!

Absorption requires H_2. *Concerted* gas/surface H exchange and penetration.

Slow penetration at regular terrace sites: → Bulk-dissolved H.

Fast penetration at defects: → Localized subsurface hydride nucleation.