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水素吸着 Pd(110)再構成表面における水素原子吸収過程

Hydrogen atom absorption in hydrogen pre-covered Pd(110) surface

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Hydrogen interaction with metal surfaces is a fundamental concept in the field of surface science and is an important field for the realization of sustainable hydrogen-based technologies. The absorption of hydrogen in the surfaces in particular is a very crucial process for hydrogen storage and permeation applications. Pd and Pd–based surfaces are popular materials for hydrogen technology related applications due to their capability to absorb large volumetric quantity of hydrogen [1]. In this present study, the absorption of H atom in Pd(110) was investigated by employing density functional theory based calculations. Being the least stable low-index surface of the Pd, the (110) surface is known to reconstruct when dosed by large amount of H [2]. Although the presence of H in the subsurface of the reconstructed surface is observed experimentally [3], the absorption mechanism is not yet theoretically established.

Hydrogen atom absorption from the surface to the subsurface of unreconstructed Pd(110) is found to be accompanied by large activation barrier (~0.30 eV) [4]. This suggests the possibility of H-trapping in the surface or near-surface region at low H coverage regime. On the other hand, the absorption of H atom in H-covered Pd(110) (1×2) missing-row surface is found to be non-activated when assisted by incoming H atom from vacuum [5]. In particular, neither the monoatomic absorption of H nor the assistance from the initially adsorbed H atom explains the experimentally observed presence of subsurface H in Pd(110) at high H coverage. Aside from the non-activated absorption of H in the H-covered surface, the present results also revealed that the dissociation of H_2 is the rate-limiting process which completely reverses the behavior of H in a clean Pd(110) surface [4,6]. The findings of this work offer the challenge to experimentally confirm the two possible dissociation/ absorption channels through the ridge and trough sites of the missing-row surface. Consequently, it would also be interesting to observe and exploit the performance of the missing-row surface toward catalytic reactions.

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