Hydrogen Transportation Mechanism Across Metal Surfaces
Revealed Through Thermal Desorption and Nuclear Reaction Analysis –
From Hydrogen Absorption to Hydrogenation Catalysis

Markus Wilde, Satoshi Ohno, Katsuyuki Fukutani
Institute of Industrial Science, The University of Tokyo, Japan

The transportation of hydrogen (H) between gas phase H\(_2\) and the interior of metals is crucial for H storage in metal hydrides, for H\(_2\) purification, as well as for industrial olefin hydrogenation over palladium (Pd) catalysts, where Pd-dissolved hydrogen has been identified as an indispensable ingredient for catalytic reactivity toward hydrogen attachment to the unsaturated C=C double bond [1, 2]. In this introductory talk, I will explain how we elucidate experimentally the atomic-level transportation processes that occur at Pd surfaces during H\(_2\) absorption, H\(_2\) desorption, and in the catalytic hydrogenation of 2-butene through a combination of H depth profiling via \(^1\)H\(^{(15}\text{N}, \text{□□})\(^{12}\text{C}\) nuclear reaction analysis (NRA) [3] and thermal desorption spectroscopy (TDS) with isotope-labeled (H, D) surface hydrogen. Choosing a Pd(110) single crystal as a structurally well-defined model system, we demonstrate first that H\(_2\) absorption proceeds along two different microscopic pathways attributable to H penetration at surface defects and at regular terrace sites [4]. At low temperatures (<145 K), these pathways produce Pd-absorbed H states with distinctly different depth distributions. Moreover, the H\(_2\) ingress into Pd(110) is found to involve a strikingly small activation energy, which suggests that energetic surface H species are important intermediates in the absorption mechanism. Investigating finally the role of Pd-absorbed H in the olefin hydrogenation catalysis reveals that whenever Pd-dissolved H resurfaces (even from the deep metal interior) onto the Pd surface – which is H-saturated under the reaction conditions - it produces reactive surface H species that can hydrogenate the co-adsorbed olefin molecule [2]. Our results invite the hypothesis that the reactive surface H species in the absorption and catalytic hydrogenation process are the same.