Pd 系合金における水素吸着・吸蔵反応のリアルタイム観察

In-situ XPS studies of hydrogen adsorption/absorption on Pd-based alloys

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The Pd-based alloys, particularly the PdAg and PdCu alloys, have attracted many attentions recently because of their high hydrogen storage ability and hydrogen permeability [1,2]. However, the hydrogen interactions with both the surface and bulk of the alloys during the hydrogen permeation are still under debates. In the present study, the mechanisms of hydrogen adsorption and absorption processes on PdAg and PdCu alloys have been investigated by the *in-situ* ambient pressure X-ray photoelectron spectroscopy (AP-XPS) and *in-situ* UHV-XPS. The *in-situ* AP-XPS measurements were carried out under the hydrogen pressure of 1.5 Torr at 300-573 K at BL07LSU, SPring-8. The *in-situ* UHV-XPS experiments were performed under the hydrogen pressure of 5×10^{-8} Torr and 3.8×10^{-8} Torr on PdAg and PdCu alloys at 300-673 K, respectively, at BL23LSU, SPring-8 and BL13B, Photon Factory.

In the UHV-XPS measurements, Ag atoms was found to be the more reactive sites for the hydrogen adsorption than Pd atoms during the hydrogen adsorption at 300-620 K. Figs. 1 show the Pd $3d_{5/2}$ and Ag $3d_{5/2}$ spectra measured on the clean surface and after H_2 exposure at 1.5 Torr on PdAg alloy. The bulk and surface Pd components were observed from Pd $3d_{5/2}$ spectrum on the clean surface in Fig. 1(a). The surface Pd disappeared after H_2 exposure, and followed by the appearance of the Pd hydride component (bulk Pd-H) at 473 K. The intensity of bulk Pd-H component further increased when the temperature was increased from 473 K to 573 K. In Fig. 1 (f), the surface and bulk Ag component were observed from Ag $3d_{5/2}$ spectrum on the clean surface, which is similar with the Pd $3d_{5/2}$ spectrum. However, the Ag hydride component was not found after H_2 exposure. It indicates that the Pd atoms are more reactive for hydrogen absorption than Ag atoms.

The UHV- and AP-XPS measurements on the B2- and fcc/bcc- phased PdCu alloys show no strong phase structure dependence of the hydrogen adsorption, however, the activation energy for the hydrogen absorption strongly influenced by the phase structure. The detailed interactions between hydrogen and the surface and bulk during hydrogen adsorption/absorption will be discussed.

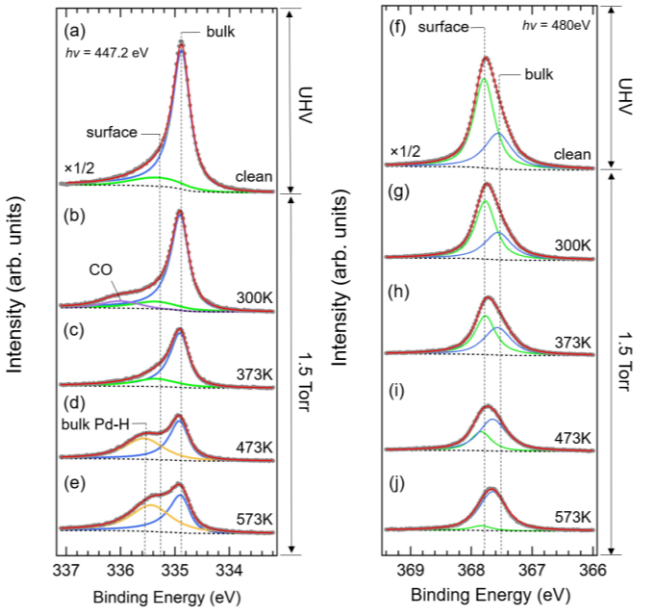

Fig. 1 Pd $3d_{5/2}$ spectra measured on (a) clean surface, and under hydrogen pressure at 1.5 Torr at (b)-(e) 300-573 K, and Ag $3d_{5/2}$ spectra measured on (f) clean surface, and under hydrogen pressure at 1.5 Torr at (g)-(j) 300-573 K on PdAg alloy.

Reference

[1] Z. Jovanovic et al., *Int. J. Hydrogen Energy* **2011**, *36*, 15364.

[2] R.J. Westerwall, et al., *Int. J. Hydrogen Energy* **2015**, *40*, 3932.