Person to contact for this submission: Takashi Harumoto
(harumoto.t.aa@m.titech.ac.jp; phone: +81-3-5734-3145; fax: +81-3-5734-3145)

Supplementary Information

X-ray diffraction study on (111)-textured palladium thin film under hydrogen loading and unloading: film structure evolution and its mechanism

Takashi Harumoto[a]*, Yuichi Suzuki[b], Ji Shi[a] and Yoshio Nakamura[a]

[a]Department of Materials Science and Engineering, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro-ku, Tokyo, 152-8552, Japan, and [b]Materials Analysis Division, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro-ku, Tokyo, 152-8552, Japan

Correspondence email: harumoto.t.aa@m.titech.ac.jp

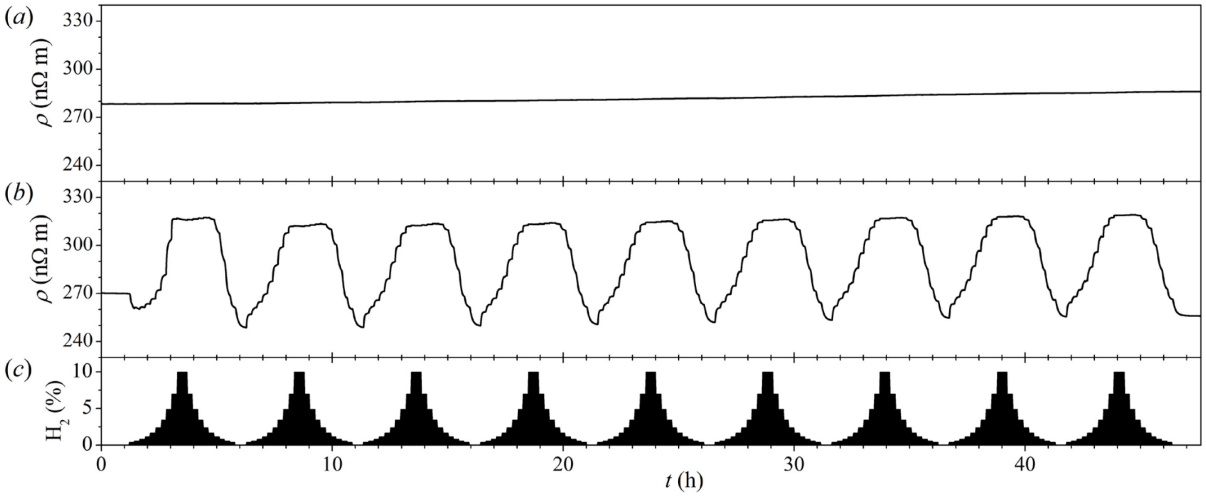

Figure S1 Electrical resistivity (ρ) of (*a*) the APA underlayer/substrate (*i.e.* AlN(20 nm)/Pd(8 nm)/AlN(20 nm)/substrate) and (*b*) Pd(8 nm)/AlN(20 nm)/substrate during the same hydrogen loading cycles as the *in situ* diffraction measurement (*c*). The resistivity of the films was measured using the van der Pauw method.

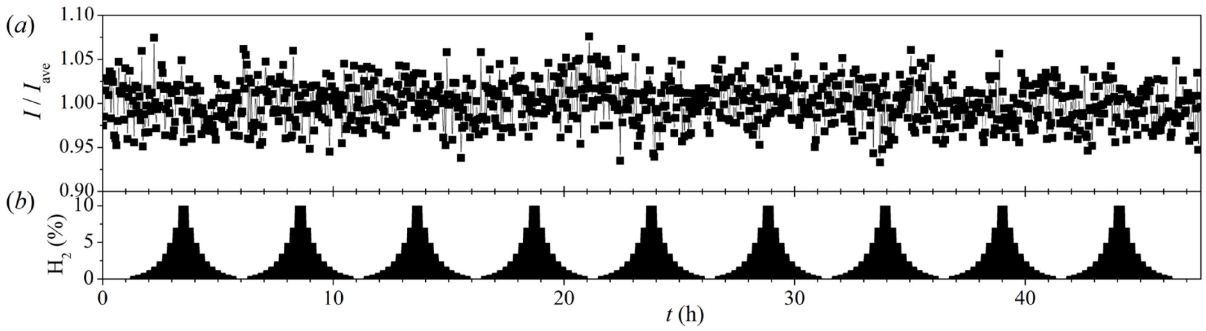

Figure S2 Intensity stability of the diffractometer during the *in situ* measurement (*a*). *I* represents the background level of the diffraction line profile at $2\theta = 50-55°$ where there is no peak except for the background derived from the amorphous fused silica glass substrate. I_{ave} denotes the average of *I* for 47.6 h. (*b*) shows the profile of the introduced hydrogen gas.

Table S1 Details of the diffraction elastic constants employed for the diffraction stress analysis. The error bar for each plot point was calculated considering the accuracy of the diffraction angle (2θ), the standard error of the peak fitting and the lower/upper limits of the diffraction elastic constants shown in this table.

	Lower limit (TPa^{-1})	Most probable value (TPa^{-1})	Upper limit (TPa^{-1})
S_1^{111} for α phase	−1.991	−1.754	−1.754
	(S_1^{111} for $PdH_{0.66}$)	(S_1^{111} for pure Pd)	(S_1^{111} for pure Pd)
$\frac{1}{2}S_2^{111}$ for α phase	7.018	7.018	7.880
	($\frac{1}{2}S_2^{111}$ for pure Pd)	($\frac{1}{2}S_2^{111}$ for pure Pd)	($\frac{1}{2}S_2^{111}$ for $PdH_{0.66}$)
S_1^{111} for β phase	−1.991	−1.991	−1.754
	(S_1^{111} for $PdH_{0.66}$)	(S_1^{111} for $PdH_{0.66}$)	(S_1^{111} for pure Pd)
$\frac{1}{2}S_2^{111}$ for β phase	7.018	7.880	7.880
	($\frac{1}{2}S_2^{111}$ for pure Pd)	($\frac{1}{2}S_2^{111}$ for $PdH_{0.66}$)	($\frac{1}{2}S_2^{111}$ for $PdH_{0.66}$)