

## Supporting Information

# Elucidating the Unexpected Electrocatalytic Activity of Nanoscale PdO Layers on Pd Electrocatalyst Towards Ethanol Oxidation in Basic Solution

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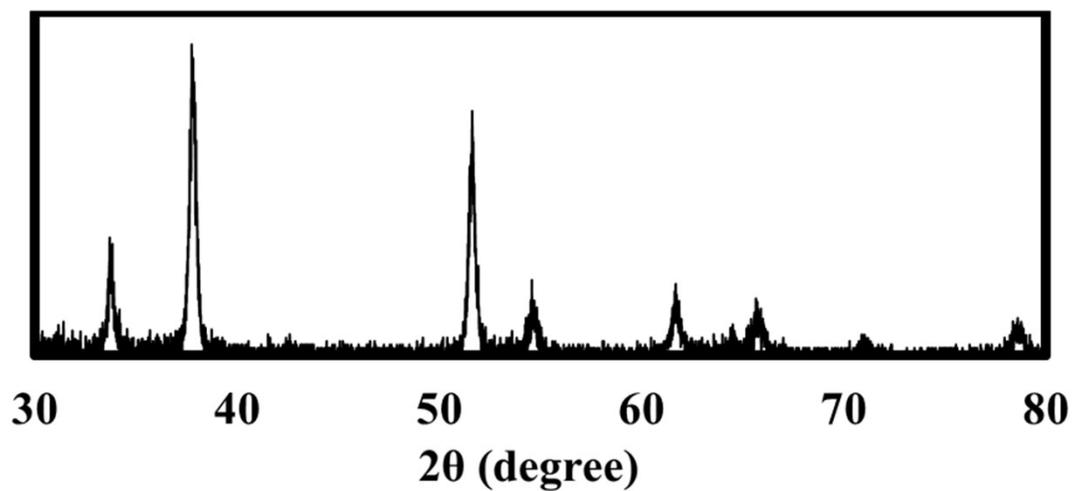


Figure S1. XRD pattern of FTO electrode.

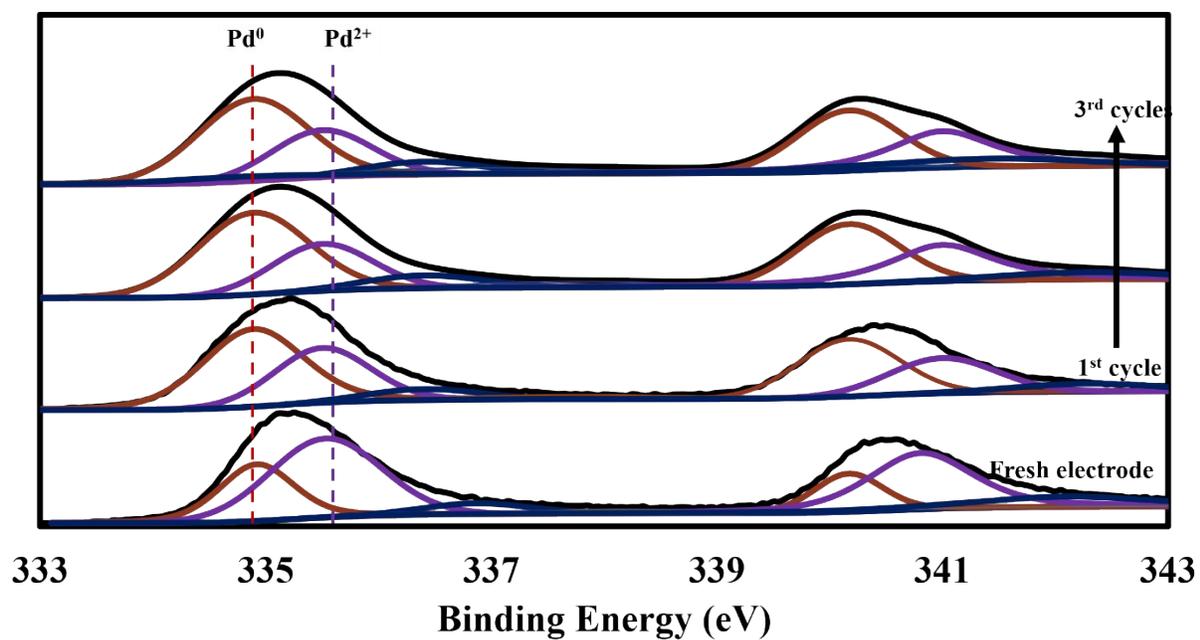


Figure S2. Pd 3d XPS profiles of the as-electrodeposited palladium on FTO as a function of sputtering time (1 min/cycle) using Ar ion beam energy of 3 kV.

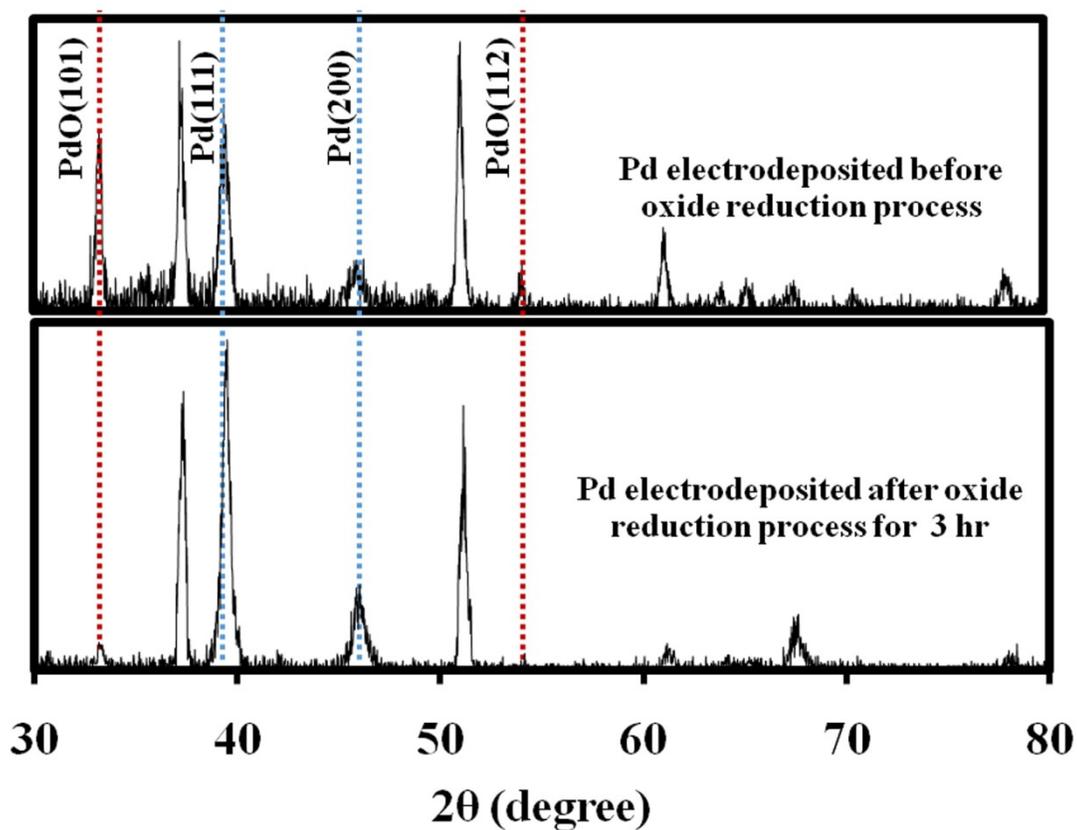


Figure S3. XRD patterns of the electrodeposited Pd before and after oxide reduction process for 3h.

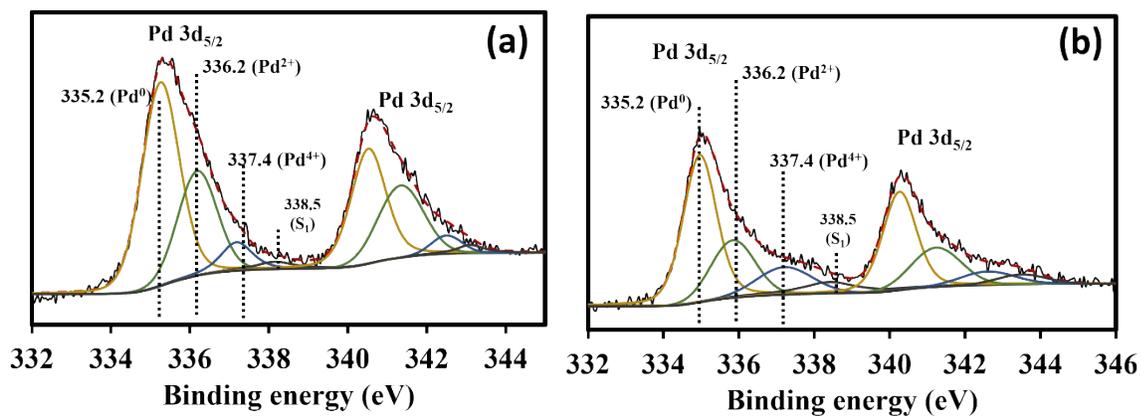
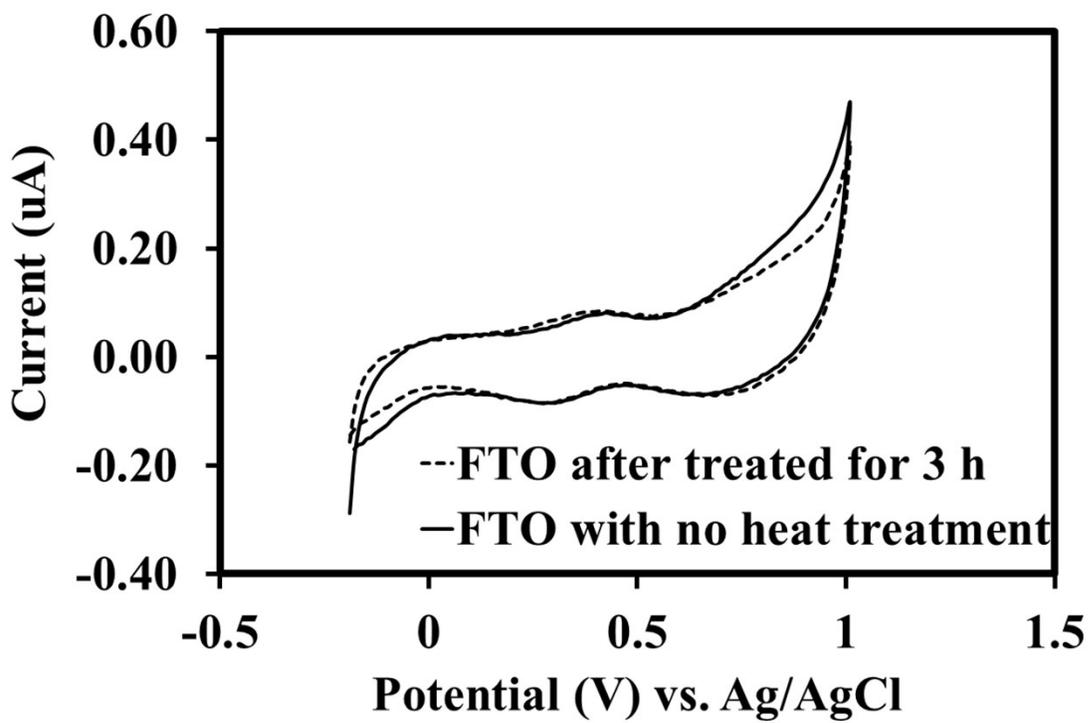
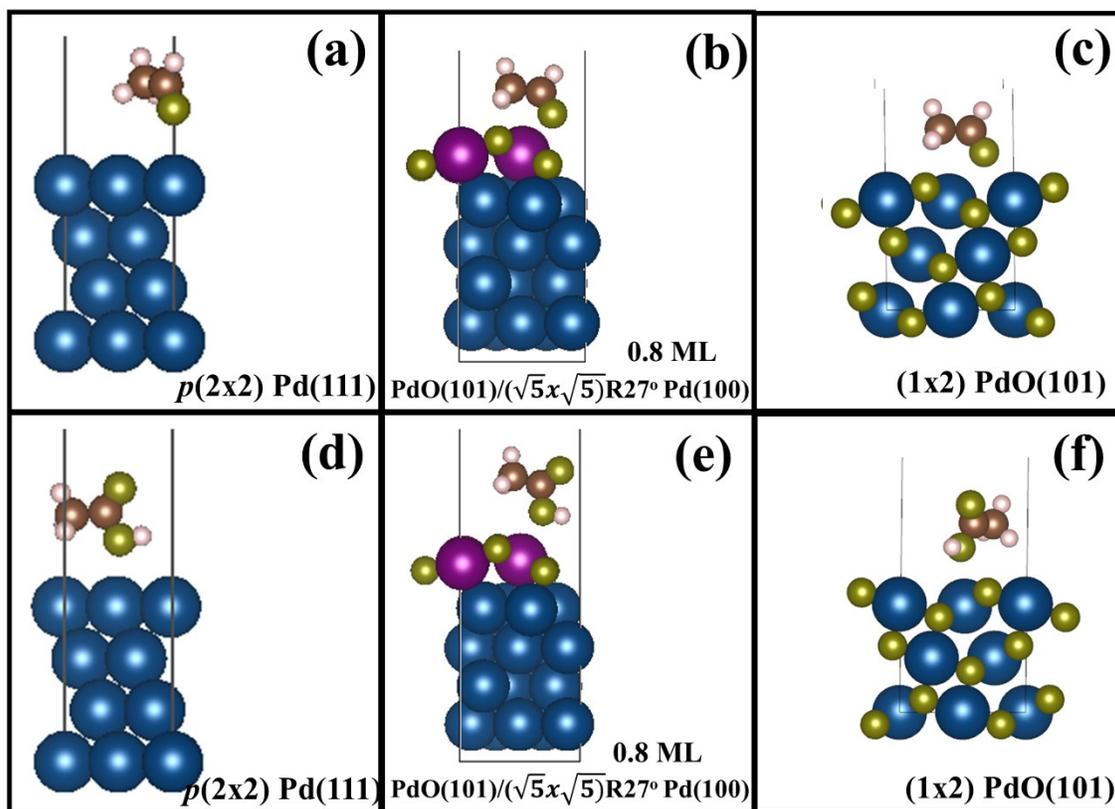


Figure S4 XPS spectra of the as-prepared Pd after the oxide reduction process for 10min (a), and (b).



**Figure S5.** CVs of FTO with and without H<sub>2</sub> treatment at 100°C for 3 hours in 1M NaOH at a scan rate of 50 mV/s.



**Figure S6.** Top and side views of the atomic geometry of the most stable acetaldehyde (a, and b), and acetic acid (d and e) adsorption on the (a, d)  $p(2 \times 2)$  Pd(111) unit cell, (b, e)  $\text{PdO}(101)/(\sqrt{5} \times \sqrt{5})R27^\circ \text{Pd}(100)$  and (c, f)  $(1 \times 2)$  PdO(101).