

# Supporting Information

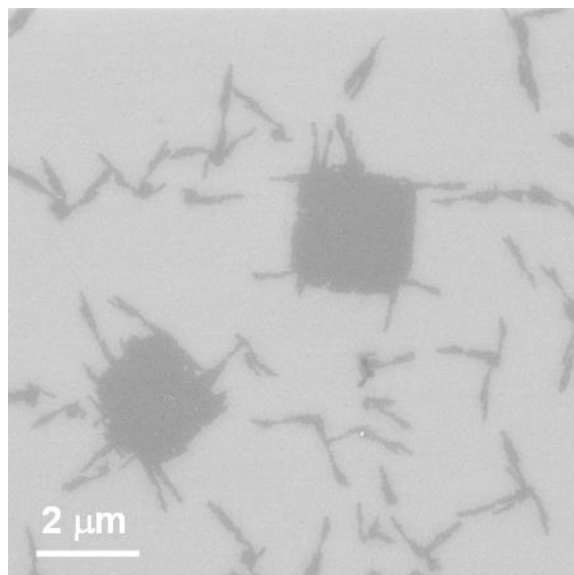
## **Ionic Layer Epitaxy of Nanometer-thick Palladium Nanosheets with Enhanced Electrocatalytic Properties**

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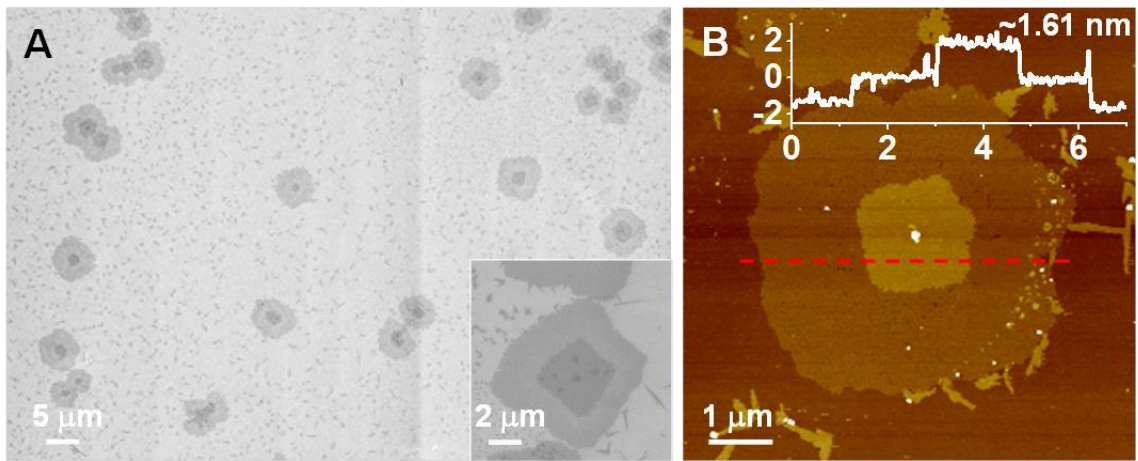
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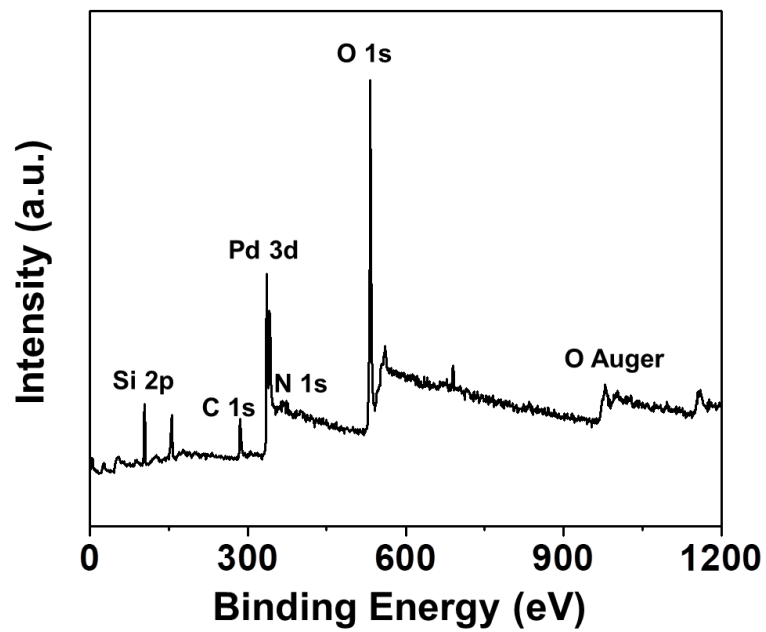
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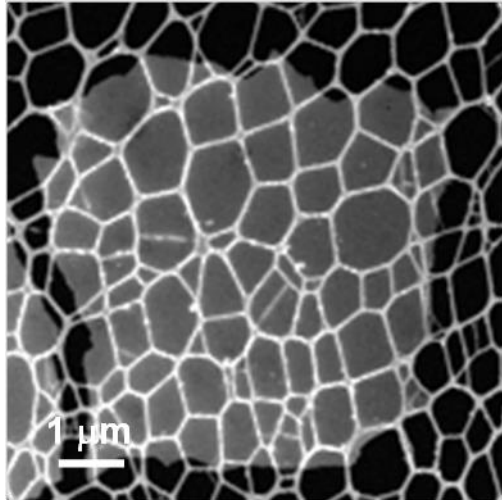
**Figure S1.** Enlarged SEM image showing the Pd nanosheets with the spikes extending out at the corners.



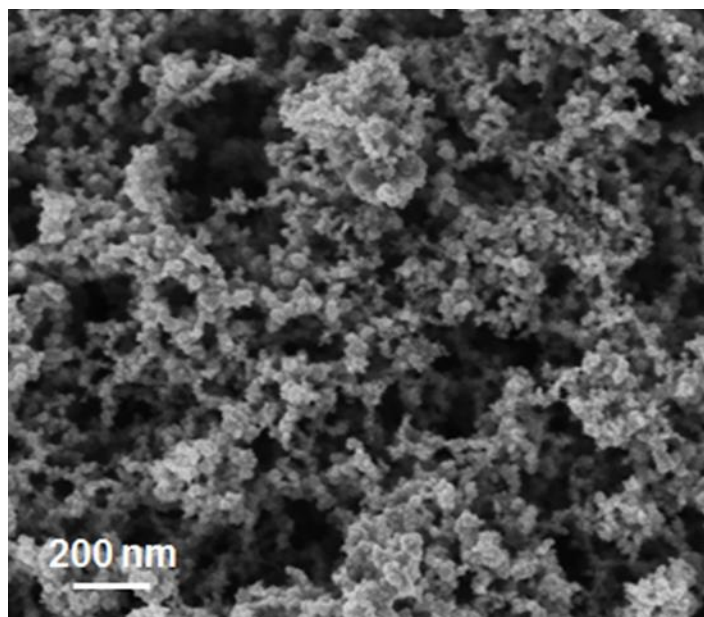
**Figure S2.** Pd nanosheets with a terraced morphology collected at a longer reaction time. (A) Large scale SEM image. Inset is a zoomed-in image of Pd nanosheets. (B) AFM topography image. Inset is the height profile along the red dashed line.



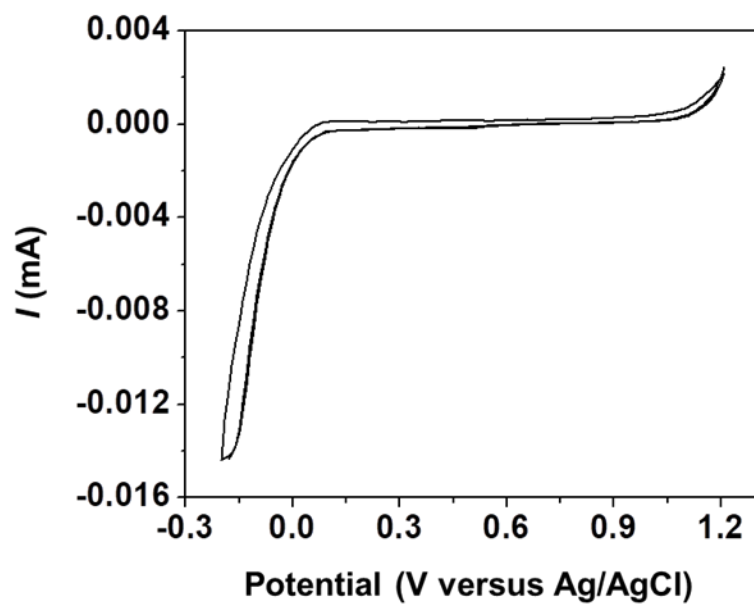
**Figure S3.** A full wavelength survey scan XPS spectrum of Pd nanosheets after being annealed at 200 °C for 2 hours.



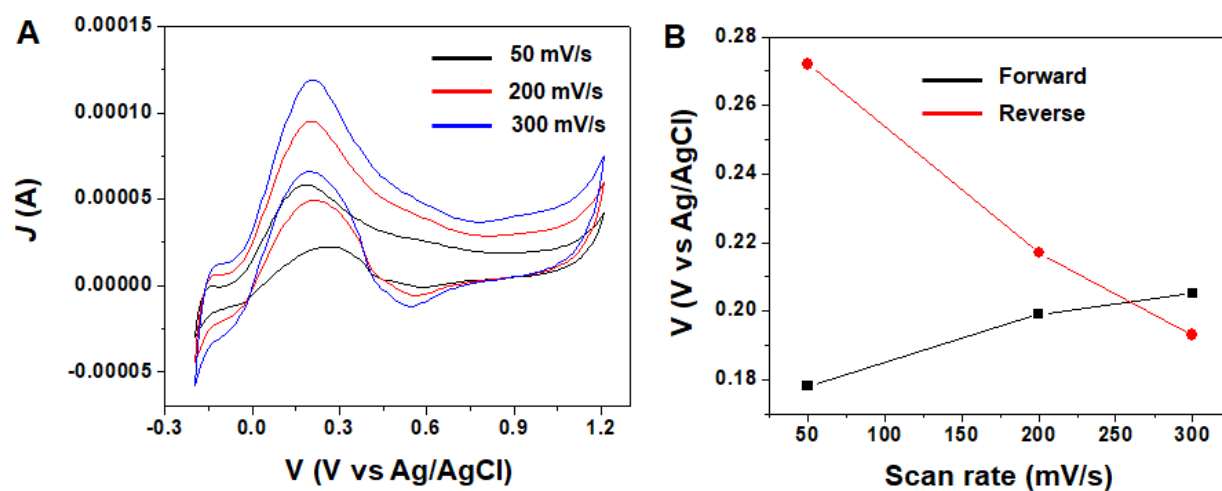
**Figure S4.** Low magnification TEM image of one Pd nanosheet on the holey carbon TEM grid.



**Figure S5.** SEM image of the commercial Pd black sample with an average size of ~40 nm.

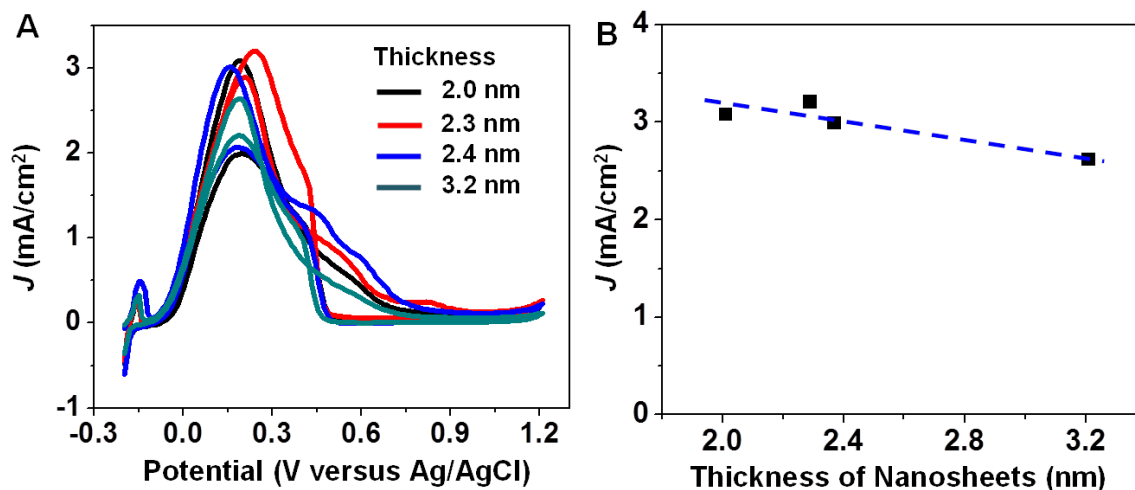


**Figure S6.** IV curve of the substrate without Pd nanosheets.

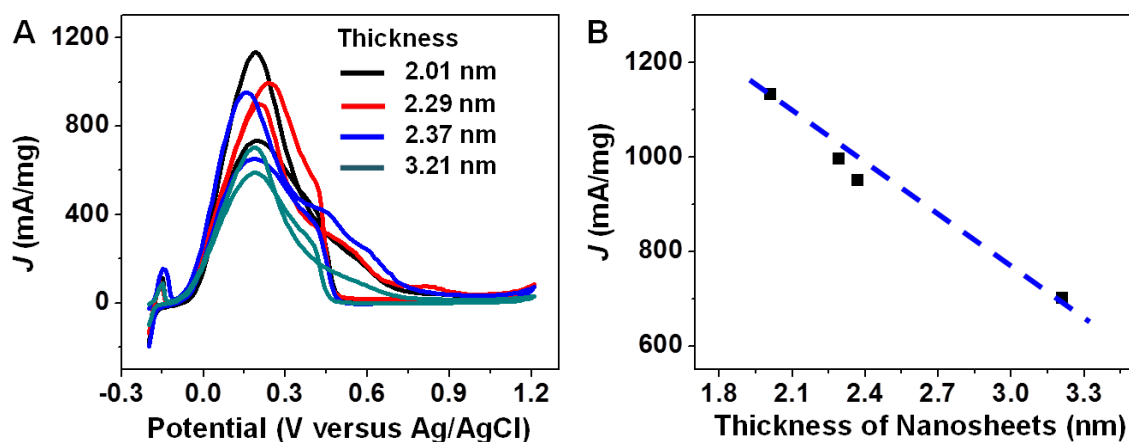


**Figure S7.** (A) J-V curves of Pd nanosheets measured at different scan rate. (B) The oxidation peak in the reverse scan shifted toward the negative direction, and the reduction peak in the forward scan shifted toward the positive direction as the scanning rate increased from 50 mV/s to 500 mV/s.

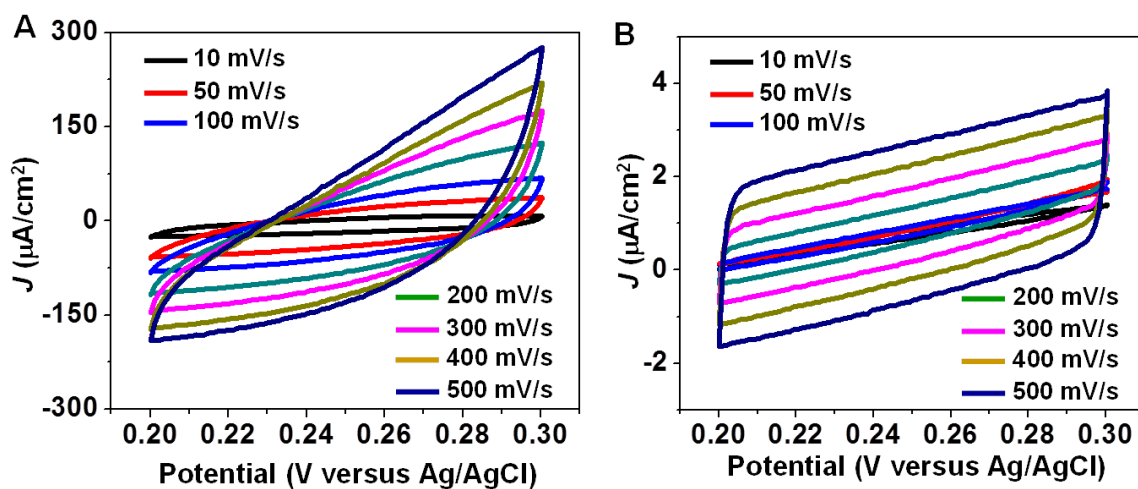




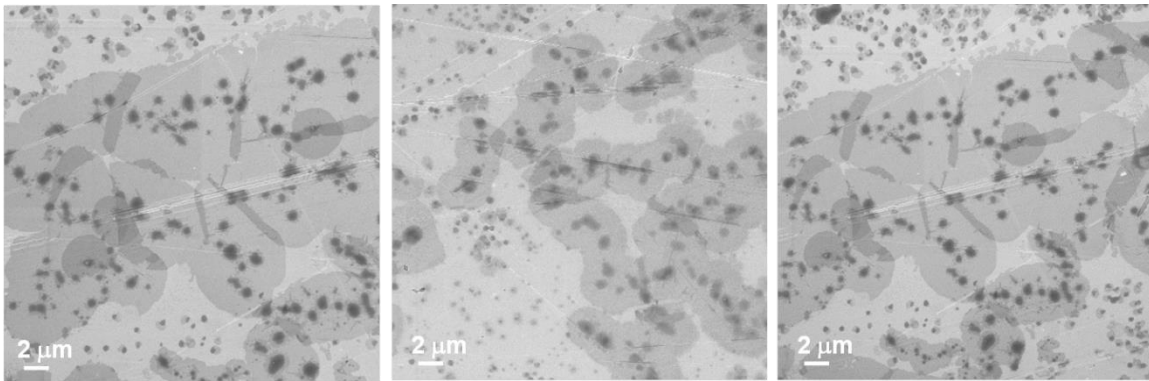
**Figure S8.** Thickness dependent measurements of the catalytic activity (area) of Pd nanosheets. (A) Current density (area) – potential curves of Pd nanosheets with the thickness of 2.01 nm, 2.29 nm, 2.37 nm and 3.21nm. (B) Plot of current density versus thickness of the nanosheets extracted from (A).



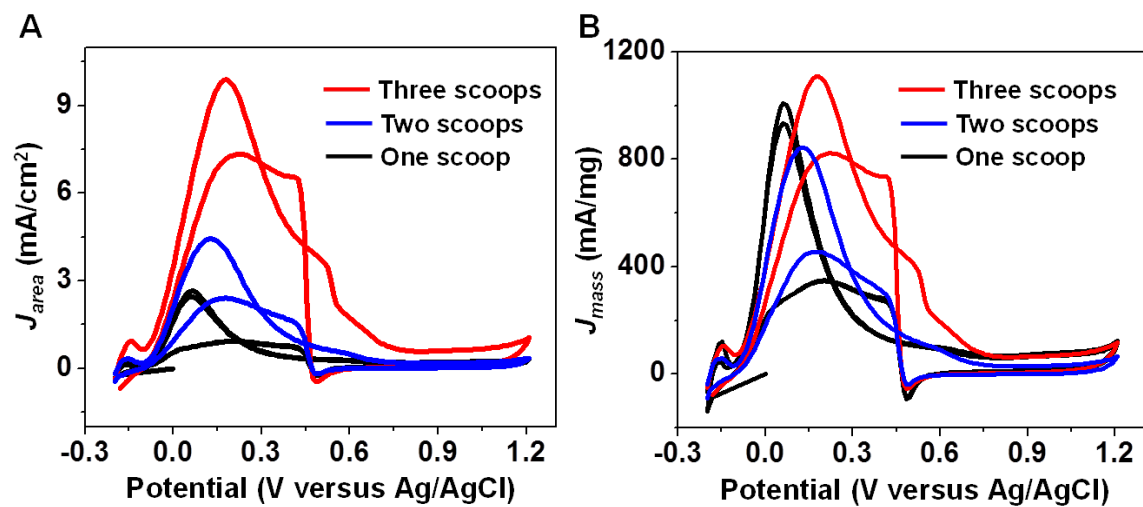
**Figure S9.** Thickness dependent measurements of the catalytic activity (mass) of Pd nanosheets. (A) Current density (mass) – potential curves of Pd nanosheets with the thickness of 2.01 nm, 2.29 nm, 2.37 nm and 3.21nm. (B) Plot of current density versus thickness of the nanosheets extracted from (A).



**Figure S10.** Cyclic voltammetry measurements of (A) the Pd nanosheets and (B) the commercial Pd black with different scan rates of 10 mV/s, 50 mV/s, 100 mV/s, 200 mV/s, 300 mV/s, 400 mV/s and 500 mV/s in order to extract the double-layer capacitance ( $C_{dl}$ ). The scan range is from 0.2 V to 0.3 V.



**Figure S11.** SEM images showing the overlap of the Pd nanosheets on the support electrode surface after multiple-time transfer.



**Figure S12.** Full scanning curves of (A) area current density and (B) mass current density – potential (versus Ag/AgCl) as increasing the loading of Pd nanosheets by multiple-time transfer.