Supplementary Information

## Controllable Assembly of Pd Nanosheets: A Solution for 2D Materials Storage

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Figure S1. Low magnified TEM micrograph of Pd nanosheet stacks.



Figure S2. Thickness distribution of the Pd nanosheets.



**Figure S3.** TEM images of (a) original monodisperse, (b) assembled, (c) disassembled and (d) re-assembled Pd nanosheets. The average diameter of the Pd nanosheets is 10 nm.



Figure S4. TEM images of (a) original monodisperse, (b) assembled, (c) disassembled

and (d) re-assembled Pd nanosheets. The average diameter of the Pd nanosheets is 15 nm.



**Figure S5.** TEM images of Pd nanosheets synthesized with the amount of CTAB: is (a) 30, (b) 64, and (c) 110 mg, respectively. The reaction time of the three batches is 2 h.



Figure S6. Illustration of the two assembled Pd nanosheets.



**Figure S7.** TEM images of Pd nanosheets that broken into nanoparticles (a) uncompletely and (b) completely.



**Figure S8.** (a) Optical photograph and (b) UV-vis extinction spectra of assembled and disassembled Pd nanosheets before and after aging, respectively.



**Figure S9**. Cyclic voltammetry curve of MOR catalyzed by Pd black in Ar-purged 0.5 M NaOH and 0.5 M methanol solution at a sweep rate of 50 mV/s in the range of 0.05 V to 1.2 V vs. RHE.

## Materials 1

**Calculation for the Distance between Neighboring Nanosheets.** The circular Pd nanosheets we got in experiments, with the average diameter D, D=20.50 nm, are presented with ellipses in TEM images, for their slanting position under projection. As shown in Supplementary Fig. 6, the vertical length of ellipses along the extension of nanowires, is D, which remains the same after projection, while the transverse length L (L=13.46 nm) (minor axis of ellipses) diminishes after projection. Thus we can calculate the angle of roll  $\theta$ , which should be:

$$\theta = \cos^{-1}\frac{L}{D}$$

Therefore, through the transverse distance  $D_{\text{transverse}}$  ( $D_{\text{transverse}} = 7.32$  nm) between neighboring sheets and  $\theta$ , we can get the distance between neighboring sheets d:  $d = D_{\text{transverse}} \cdot \sin \theta$ 

d is calculated to be 5.524 nm considering that  $\theta$  is and D<sub>transverse</sub> is 7.32 nm.

Paradies et al., have declared that the average C-C bond lengths and C-C-C bond angles in the CTAB molecules are 0.152 nm and 110-112°, respectively.<sup>1</sup> According to this, we can calculate the length of a CTAB molecule is about 1.75 nm, indicating there are two layer CTAB molecules between two nanosheets.

## **Reference:**

1. H. H. PARADIES, S. F. CLANCY, Rigaku J. 17 (2000), 20-34.