

## Electronic Supplementary Information

### Three-dimensional Hierarchical Prussian blue Composed of Ultrathin Nanosheets: Enhanced hetero-catalytic and adsorption properties

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#### Synthesis of hierarchical PB composed by ultrathin nanosheets (HPBN):

In a typical procedure, 0.1 mmol  $K_3Fe(CN)_6$  and 5 mL 36% hydrochloric acid (HCl) were dissolved into 5 mL water and 25 mL N,N-dimethylformamide (DMF) mixed solvents. The obtained solution were transferred into a 45 mL Teflon-lined stainless-steel autoclave, sealed and maintained at 80 °C for 24 h. After the solution was cooled to room temperature, the obtained deep blue solid was collected by centrifugation, washed several times with water and ethanol, and then dried in a vacuum oven at 40 °C for 12 h. For the fabrication of cubic PB, the volume ratio of water/HCl/DMF were changed into 15/5/15 while other parameters were kept the same as those of HPBN.

#### Degradation of methylene blue (MB):

In a typical procedure, 5 mg of HPBN were added to 10 mL 300 mg/L MB solution and then shaken in the dark for 30 min in a constant temperature incubator shaker at 25 °C, which ensured the adsorption-desorption equilibrium of MB on the sample. Then the degradation reaction was initiated by adding 1 mL  $H_2O_2$  (30 wt%) into the mixed solution. At the given interval, 0.2 mL of solution was taken from the conical flask and diluted to a certain degree, and then centrifuged. The concentration of MB in the supernatant was analyzed using a UV-2802S Spectrophotometer made by UNIC at a wavelength of 665 nm. Control experiments were carried out with cubic PB microcrystals and irregular commercial PB nanoparticles from alladin.

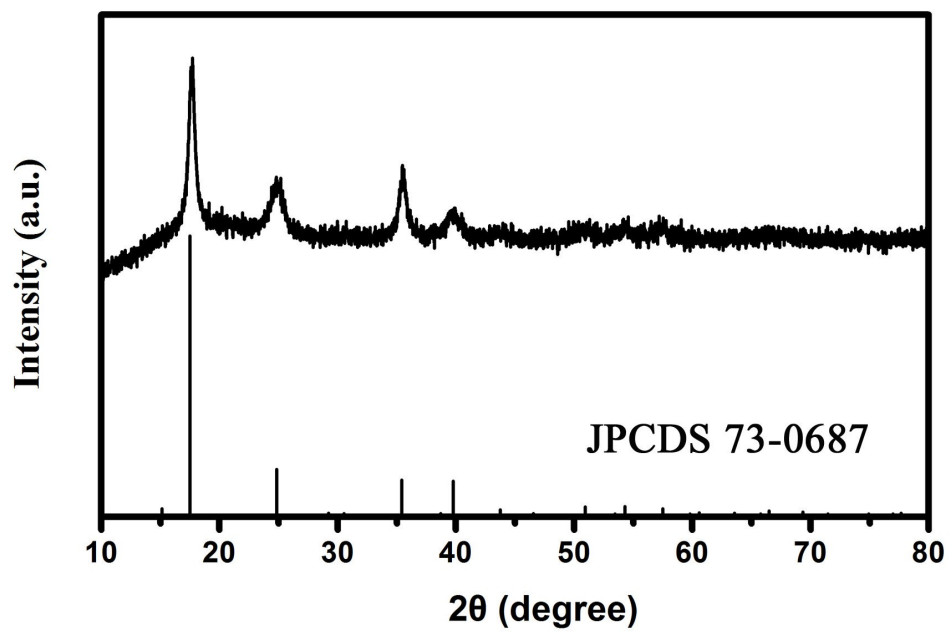
#### Adsorption of $Cs^+$ :

The HPBN were tested for  $Cs^+$  sorption performance in batch mode by dispersing 5 mg of the samples in 10 ml of  $Cs^+$  solutions prepared from CsCl at various  $Cs^+$  concentrations, namely, 100, 200, 300, 400, 500, 600 ppm. After 24 h of contact time where equilibrium was reached, the HPBN were removed from the solution by centrifugation. The concentrations of  $Cs^+$  in the solution were quantified with an atomic absorption spectrophotometer (AANALYST 800, PE). The adsorption of each sample was tested in triplicate, and average was used to evaluate the

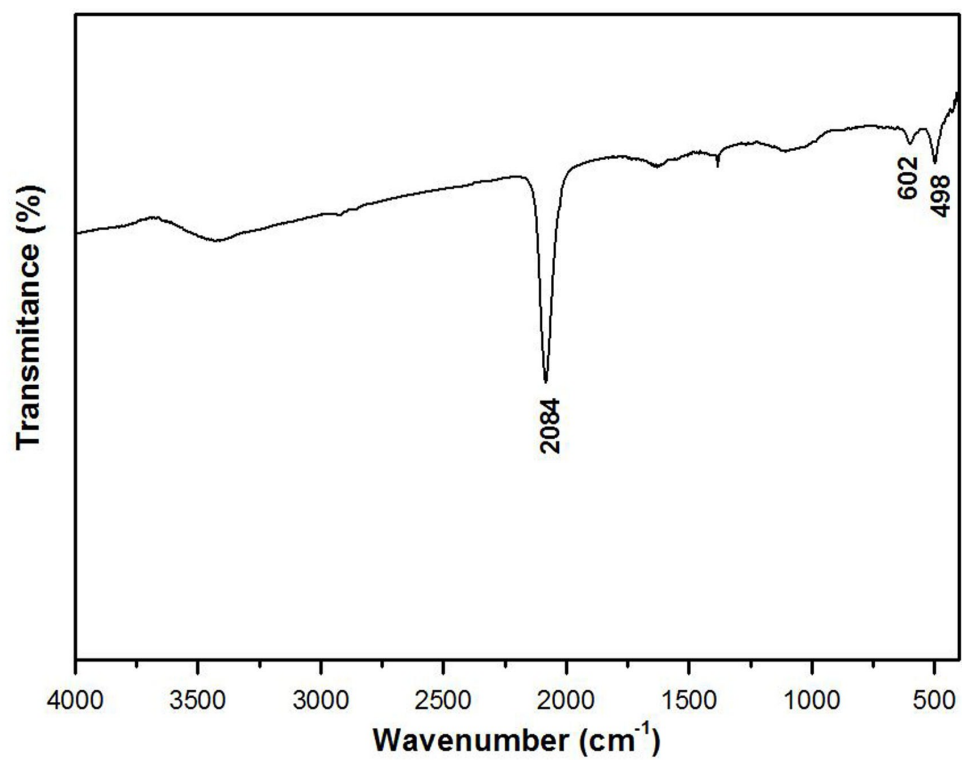
adsorbent capabilities. Control experiments were carried out with cubic PB microcrystals and irregular commercial PB nanoparticles from alladin

### **Characterization:**

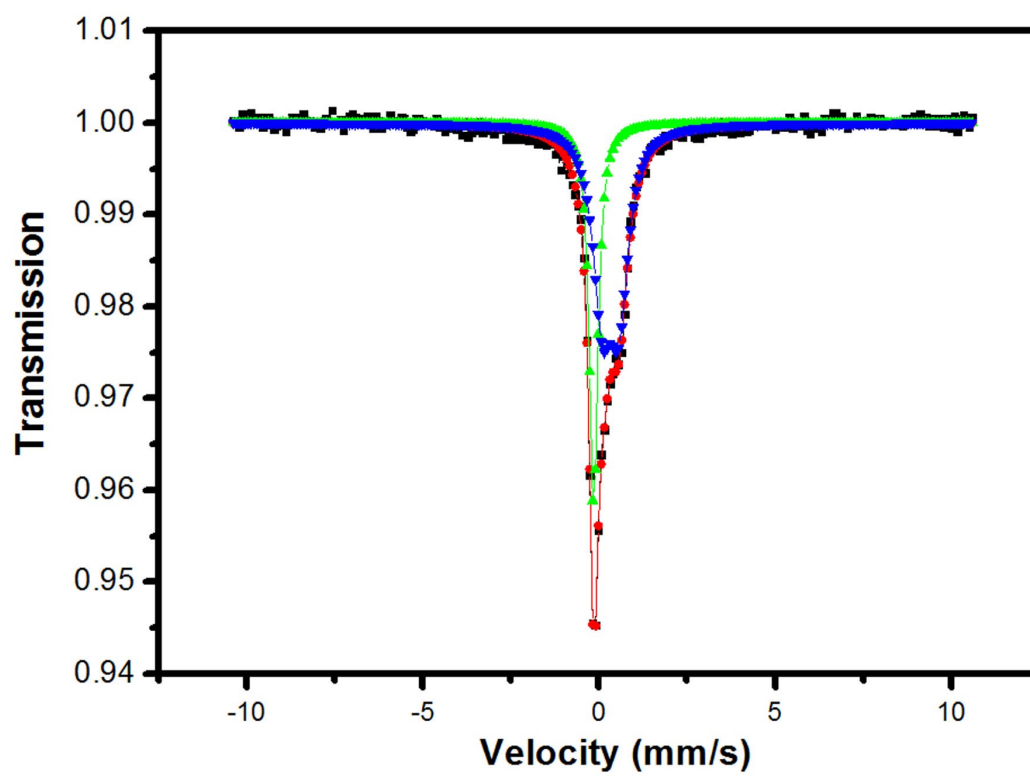
The phase compositions of the prepared samples were monitored by X-ray diffraction (XRD) using a Rigaku RINT 2500X diffractometer with Cu-K $\alpha$  radiation and conventional  $\theta$ -2 $\theta$  geometry. The morphologies of the products were determined on a field emission scanning electron microscope (FESEM, Hitachi S-4800), and a transmission electron microscope (TEM, JEOL JEM--2100F). Fourier-transform infrared (FT-IR) spectra were measured in wavenumber ranging from 400 to 4000 cm<sup>-1</sup> using Nicolet Nexus 670 FT-IR spectrophotometer. The Mössbauer spectrum was measured using a Mössbauer spectrometer in a constant acceleration mode with a 25 mCi <sup>57</sup>Co(Pd) source, and the spectrum was calibrated by a 25 $\mu$ m  $\alpha$ -Fe foil at room temperature. The Mössbauer parameters were fitted by a standard least-squares fitting program. Nitrogen sorption measurements were carried out by using an ASAP-2000 surface area analyzer at 77 K. The atomic ratio of K/Fe in PB was examined by inductively coupled plasma analysis (ICP, iCAP 6300, Thermo Fisher Scientific Inc.)



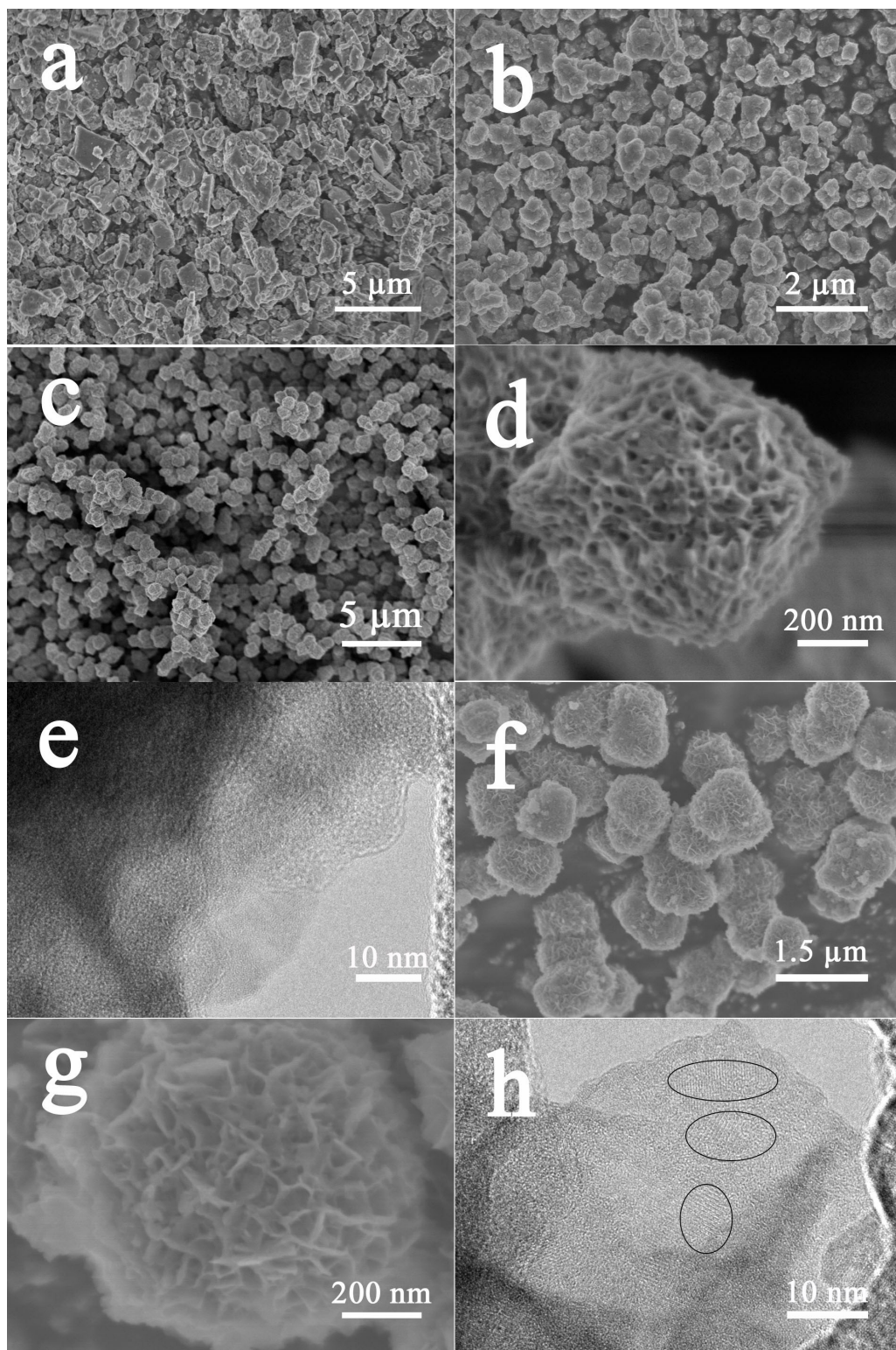
**Fig. S1** XRD pattern of HPBN prepared following a typical procedure.



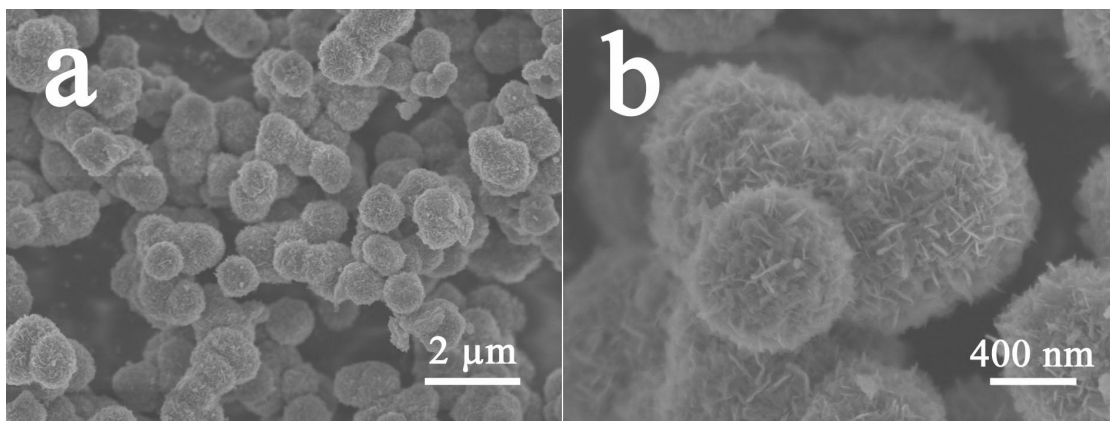
**Fig. S2** FTIR spectrum of HPBN prepared following a typical procedure.



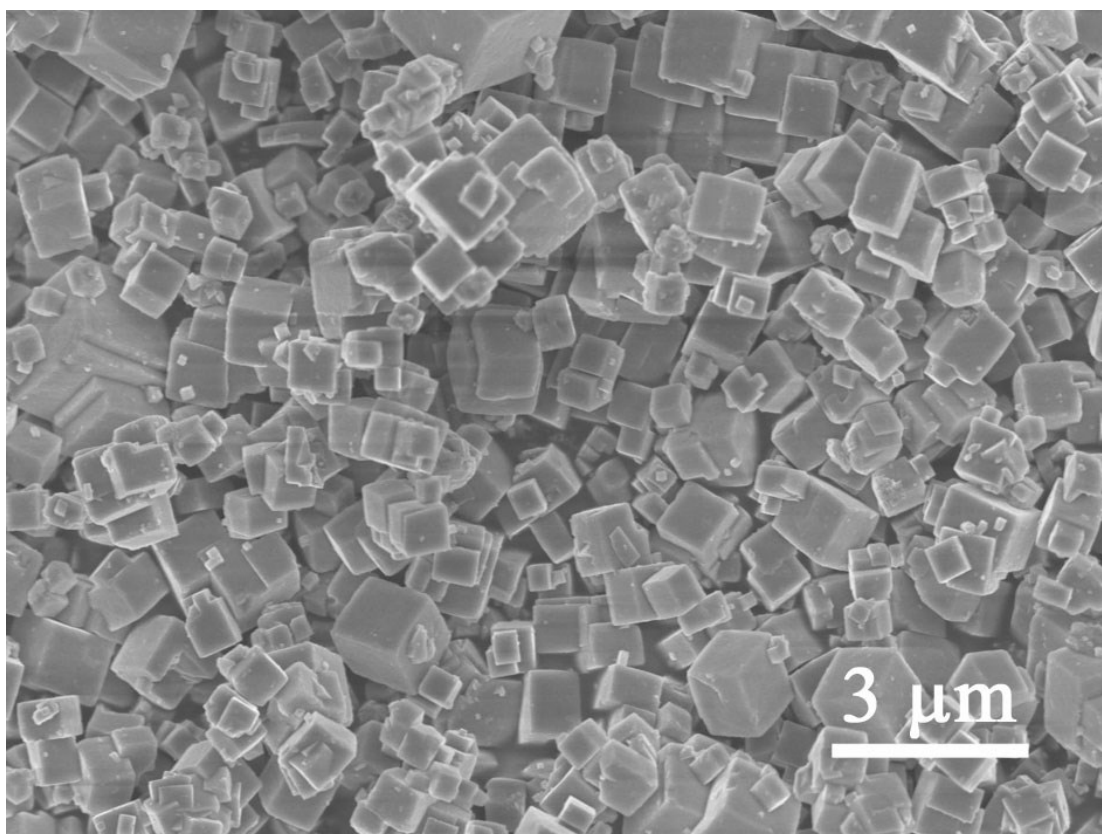
**Fig. S3** Mössbauer spectrum of HPBN prepared following a typical procedure.



**Fig. S4** FESEM images of products obtained at different reaction times (a) 1.5 h, (b) 2h, (c) and (d) 3h, (f) and (g) 9 h. High magnification TEM pictures of products obtained at (e) 3h and (h) 9h.

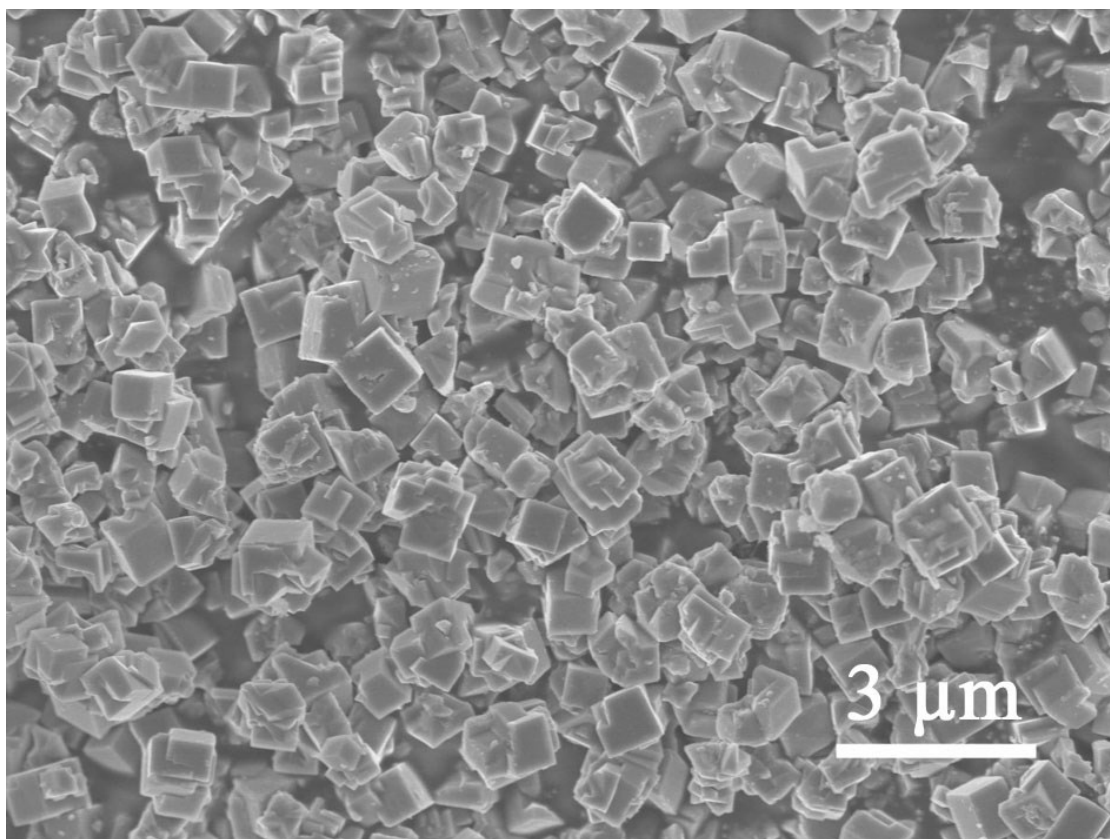


**Fig. S5** Low and high-magnification FESEM images of these as-prepared products obtained by changing dimethylformamide into dimethylacetamide.

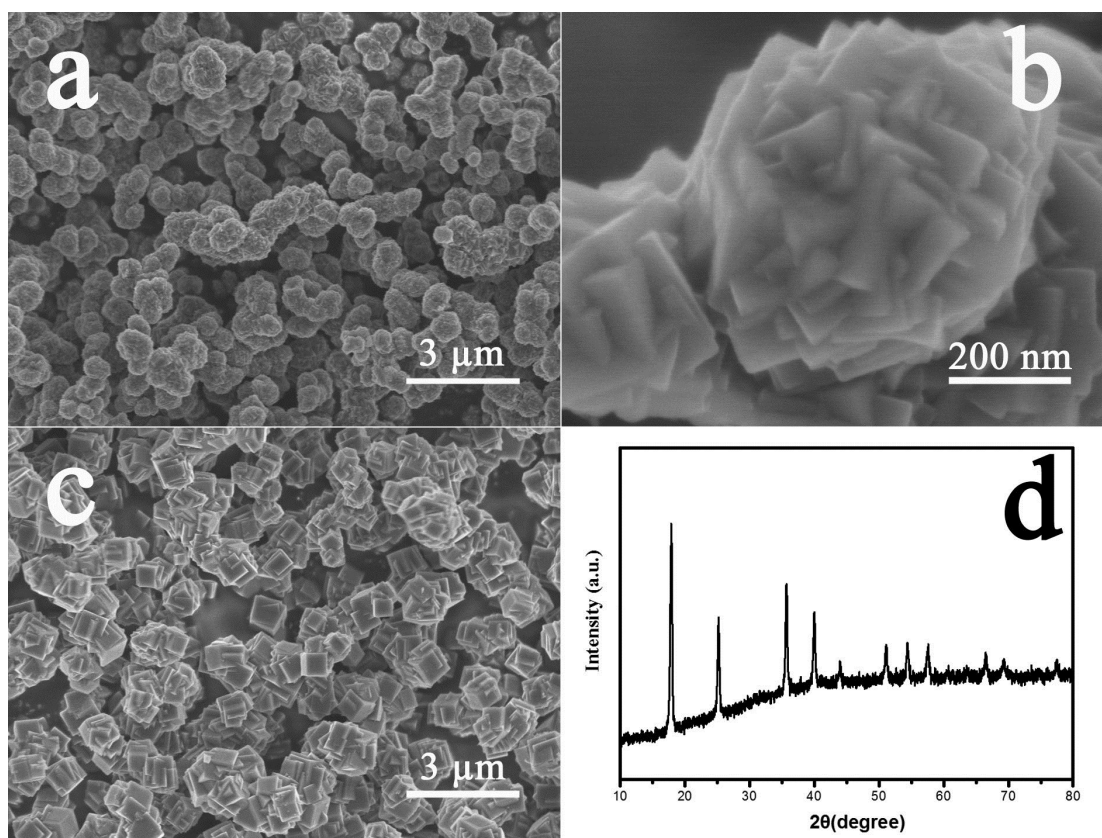


**Fig. S6** FESEM image of these as-prepared products obtained by exchanged dimethylformamide into ethanol.

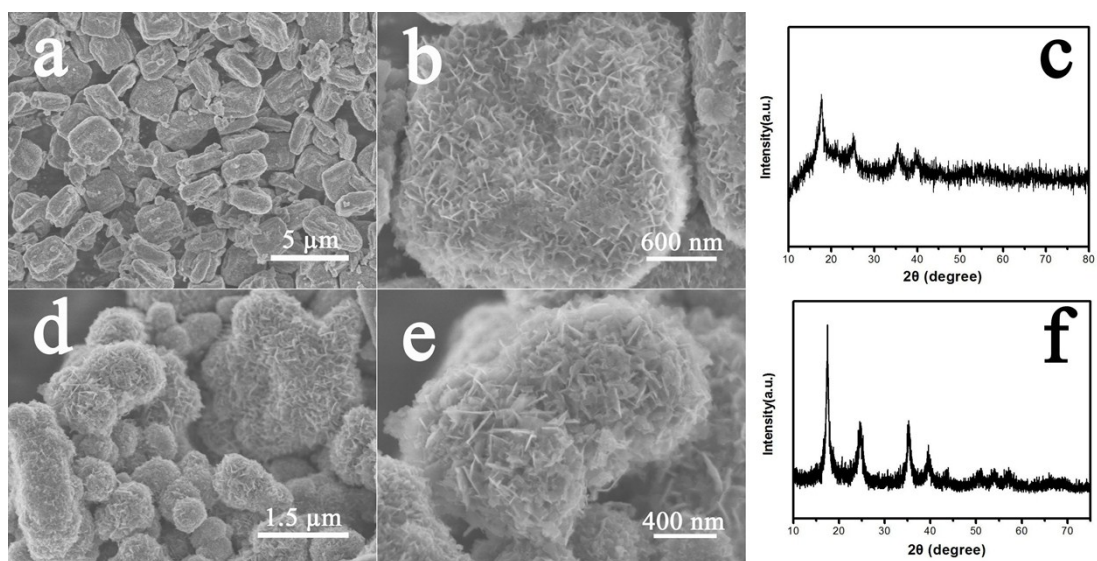




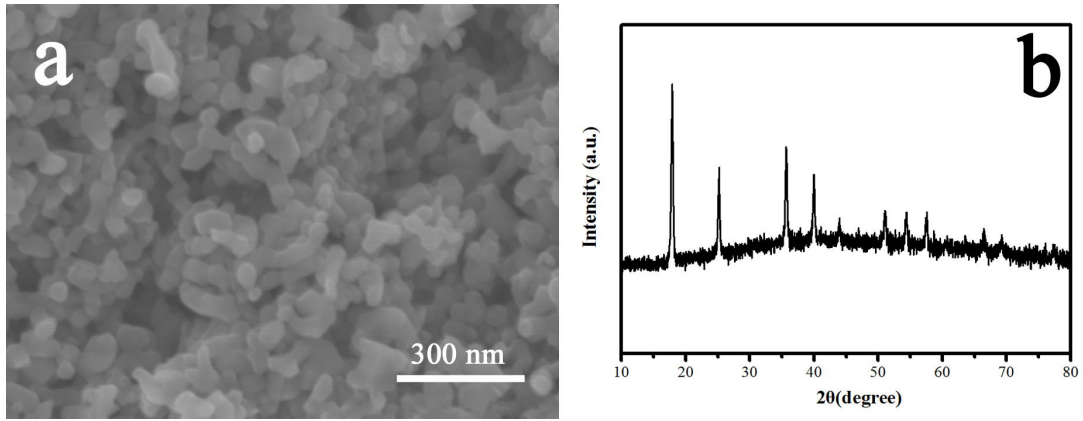
**Fig. S7** FESEM image of these as-prepared products obtained by exchanging dimethylformamide into formamide.



**Fig. S8** FESEM pictures of the products obtained with different water/DMF ratio. (a) and (b) 10/20, (c) 15/15 and (d) XRD pattern of the product obtained at water/DMF ratio of 15/15 .



**Fig. S9** FESEM images of products obtained by using sodium ferrocyanide (a) and (b), and (c) corresponding XRD pattern; FESEM images of products obtained by potassium ferrocyanide (d) and (e), and (f) corresponding XRD pattern.



**Fig. S10** (a) FESEM image and (b) XRD pattern of commercial PB nanoparticles.