Electronic Supplementary Information

Comparison of the Electrochemical Performance of Iron Hexacyanoferrate with High and Low Quality as Cathode Materials for Aqueous Sodium-Ion Batteries

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1. Experimental Section

1.1. Materials preparation

All of the reagents were of analytical purity grade and used without further purification.

Synthesis of HQ-PB nanocubes: In a typical synthesis, 3.8 g of polyvinylpyrrolidone (PVP, K30) and 110 mg of $K_4[Fe(CN)_6] \cdot 3H_2O$ were dissolved in 50.0 mL distilled water under magnetic stirring. Then 0.45 mL of concentrated hydrochloric acid was added into the above solution. After stirring for 10 min, the resulting solution was transferred to a Teflon-lined stainless steel autoclave and kept at 80 °C for 12 h. The obtained blue products were collected by centrifugation, washed with distilled water and ethanol several times, and finally dried for further use.

Synthesis of LQ-PB nanoparticles: In a typical synthesis, 1.0 g of PVP and 120 mg of K_3 [Fe(CN)₆] were dissolved in ethanol/distilled water (30.0 mL, 1/2 v/v) under magnetic stirring. Then 0.4 mL of concentrated hydrochloric acid was added into the above solution. After stirring for 10 min, the resulting solution was transferred to a Teflon-lined stainless steel autoclave and kept at 80 °C for 12 h. The obtained blue

products were collected by centrifugation, washed with distilled water and ethanol several times, and finally dried for further use.

1.2. Materials Characterization.

Powder X-ray diffraction (XRD) patterns of the as-prepared products were collected using an X-ray diffractometer (Rigaku Ultima IV) with Cu K α irradiation. The morphology and crystal structure of the as-prepared products were examined by a field-emission scanning electron microscopy (SEM, ZEISS SUPRA 55) and a transmission electron microscopy (TEM, JEOL JEM 2100) with an acceleration voltage of 200 kV. The water contents in the two PB materials were characterized by a thermal analyzer (TGA, SDT Q600, USA) under nitrogen atmosphere at a scan rate of 10 °C min⁻¹.

1.3. Electrochemical Measurements.

The electrochemical measurements were carried out in a three–electrode electrochemical cell containing 1 M Na₂SO₄ aqueous solution as the electrolyte. Electrolyte was acidified to pH = 2 by adding dilute nitric acid. The electrochemical measurements were conducted with a CHI660E electrochemical workstation. A standard calomel electrode (SCE) was used as the reference electrode and a Pt foil as the counter electrode, and all the experiments were done at ambient temperature. The active materials were mixed with carbon black and polyvinylidene difluoride (PVDF, Aldrich) in a weight ratio of 7 : 2: 1, which were dispersed in N-methyl-2-pyrrolidone with constant stirring for 8 h to form a homogeneous slurry. The well-mixed slurry was then spread onto a carbon cloth and dried at 100 °C in a vacuum oven for 12 h. The mass loading of the active material is about 4.0–5.0 mg.



Fig. S1 The XRD pattern of the LQ-PB NPs.



Figure S2. The particle size distributions of the (A) HQ PB NCs and (B) LQ PB NPs.



Fig. S3 The SEM image of the LQ-PB NPs.



Fig. S4 The TEM image of the LQ-PB NPs.



Fig. S5 The EDX pattern of the HQ-PB NCs.



Figure S6. TG curves of the as-prepared HQ-PB NCs and LQ-PB NPs.



Fig. S7 CV curve of the LQ-PB NPs recorded at a scan rate of 2 mV s⁻¹.



Figure S8. SEM images of the HQ-PB NCs and LQ-PB NPs after cycling test.