Science Advances

AAAS

advances.sciencemag.org/cgi/content/full/2/1/e1501038/DC1

Supplementary Materials for

Environmentally-friendly aqueous Li (or Na)-ion battery with fast electrode kinetics and super-long life

Xiaoli Dong, Long Chen, Jingyuan Liu, Servane Haller, Yonggang Wang, Yongyao Xia

Published 22 January 2016, *Sci. Adv.* **2**, e1501038 (2016)
DOI: 10.1126/sciadv.1501038

The PDF file includes:

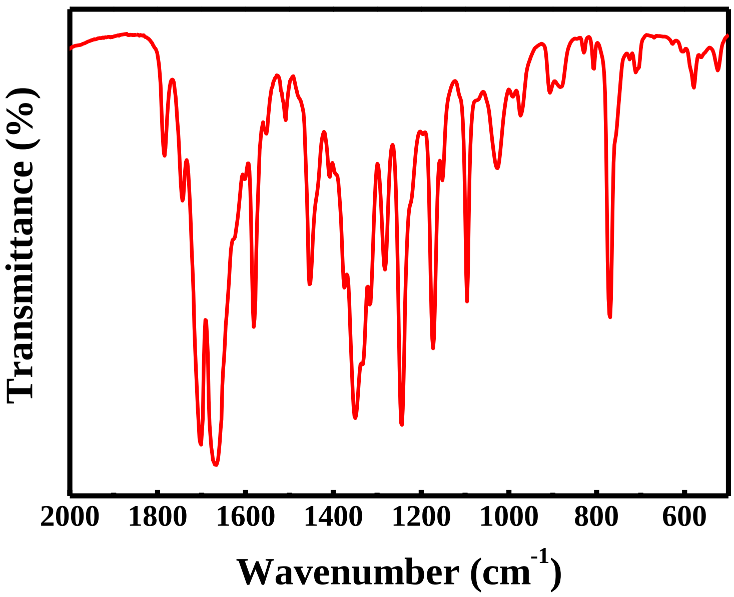

Fig. S1. FT-IR spectrum of as-prepared PNTCDA.

Discussion of Fig. S1: The Fourier transform infrared spectroscopy (FT-IR) spectrum of as-prepared PNTCDA was characterized with a NICOLET 6700 FT-IR Spectrometer using KBr pellets. As shown in **Fig. S1**, the absorption bands at 1350 cm^{-1} and 1581 cm^{-1} can be assigned to the vibration of imide C-N and naphthalene, respectively. The sharp absorption peaks at 1702 cm^{-1}, 1670 cm^{-1} and 771 cm^{-1} can be attributed to the vibration of the imide C=O bond. From the FT-IR spectrum, it can be detected that the characteristic absorption bands of the imide group are consistent with previous reports *(24,33-35)*, which thus confirms the successful preparation of PNTCDA.

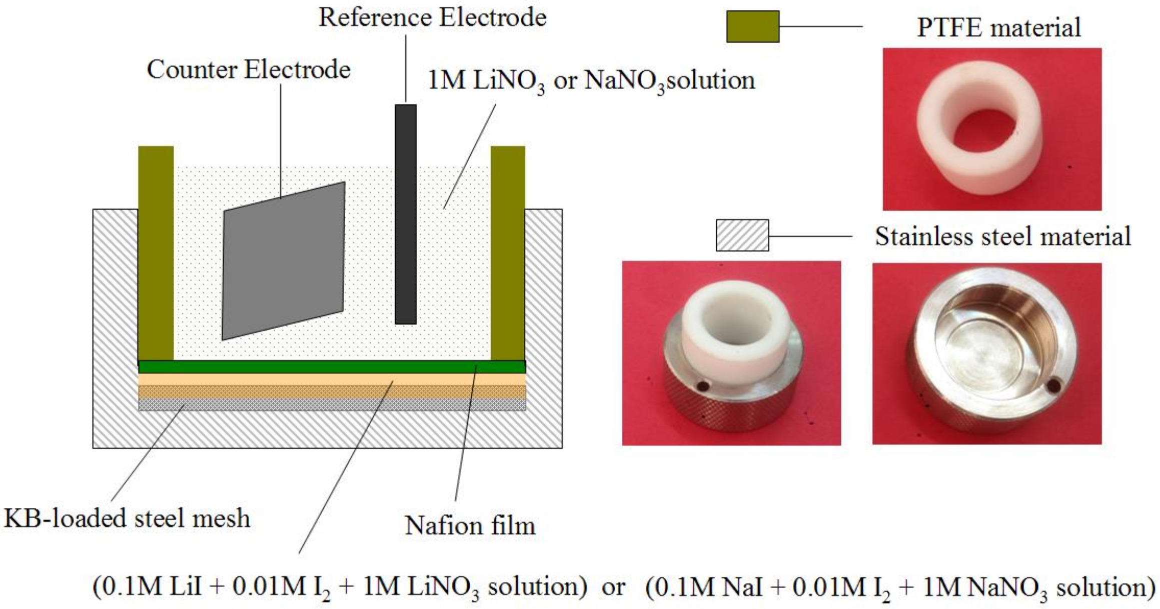

Fig. S2. Schematic illustration of the cell for aqueous electrode investigation with a three-electrode system.

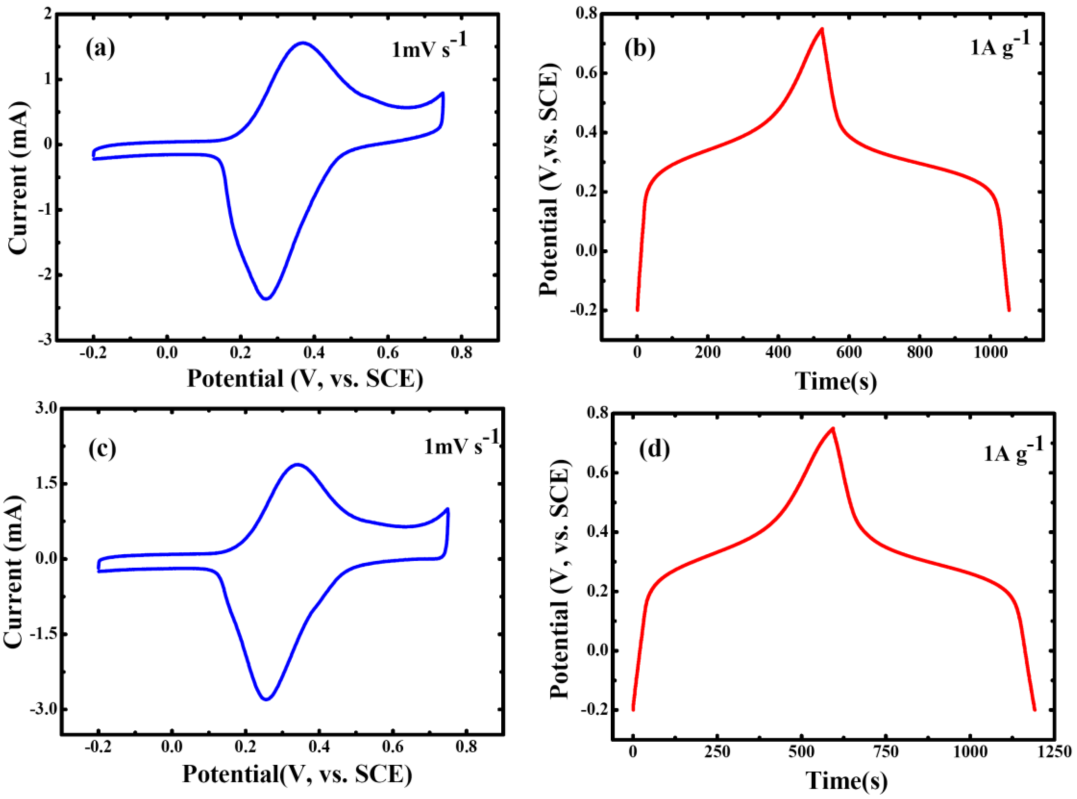

Fig. S3. Electrochemical behavior of I^{-}/I_3^{-}-based liquid electrode. (a) CV curve at a sweep rate of 1mV s^{-1} and (b) Galvanostatic charge/discharge curve of 0.1M LiI + 0.01M I_2 + 1M $LiNO_3$ solution. (c) CV curve at a sweep rate of 1mV s^{-1} and (d) Galvanostatic charge/discharge curve of 0.1M NaI + 0.01M I_2 + 1M $NaNO_3$ solution. In this experiment, carbon (KB)-loaded steel mesh was used as current collector. A SCE and activated carbon film electrode were used as reference and counter electrodes, respectively. It should be noted that liquid electrode and the reference/ counter electrode are separated by Nafion film through a special three-electrode cell (see **Fig. S2** for detail).

Discussion of Fig. S3: It can be detected from **Fig. S3** that the electrochemical behavior of I^{-}/I_3^{-} redox couple in aqueous electrolyte mainly locates in the potential window from 0 to 0.7 V vs. SCE, which is consistent with previous report *(10)*. A pair of redox peaks located in the potential of +0.35 V/+0.29 V (**Fig. S3a** and **S3c**). The corresponding charge/discharge process **(Fig. S3b** and **S3d)** is consistent with the CV investigation.

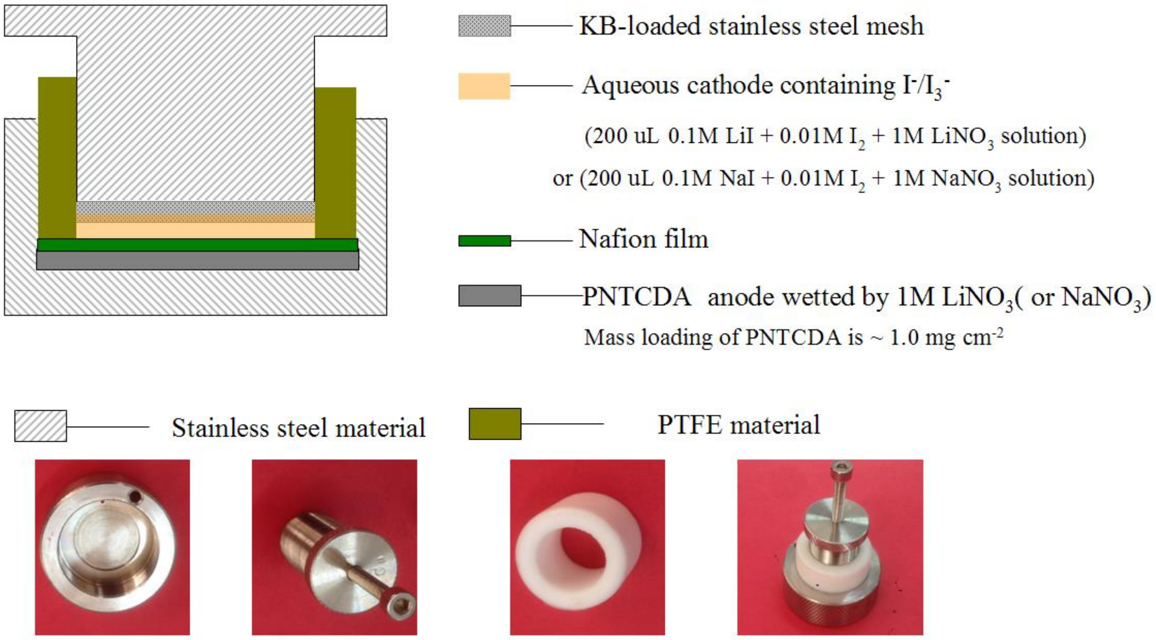

Fig. S4. Schematically showing the assembly of full cell. Detailed information and the photos of devices are given in above figure.

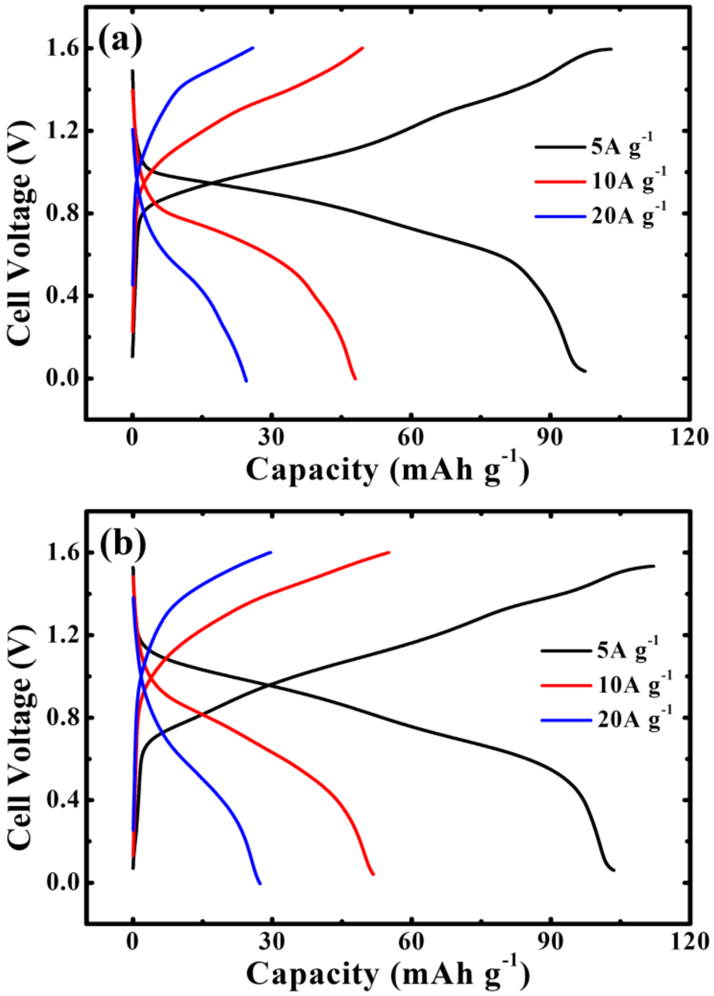

Fig. S5. Rate performance of a full cell using a high-concentration aqueous cathode (5 M LiI or NaI) and the PNTCDA anode with higher mass loading of active material (5mg cm^{-2}). (a) Aqueous Li-ion battery using LiI aqueous cathode. (b) Aqueous Na-ion battery using NaI aqueous cathode. In this experiment, 200 μL LiI solution (5M LiI (or 5M NaI) + 0.1 M I_2) was used as aqueous cathode. 5 M $LiNO_3$ (or 5M $NaNO_3$) wetted PNTCDA electrode was used as anode. The cathode and anode were separated by a Nafion film, according to Fig. S4.

Discussion of Fig. S5: As shown in **Fig. S5**, when measured with a current density of 5A g^{-1} (equals to 27C), the full cell still displays a specific capacity of 97 mAh g^{-1} (aqueous Li-ion battery; **Fig. S5a**) or 103 mAh g^{-1} (aqueous Na-ion battery; **Fig. S5b**), which is close to the data (126 mAh g^{-1} or 120 mAh g^{-1}; Fig. 5a or Fig. 6a in main text) achieved with low concentration aqueous cathode (0.1 M LiI or NaI) and PNTCDA anode with low mass loading of 1 mg cm^{-2}. However, when the applied current density is further increased to 10A g^{-1} and 20 A g^{-1}, the achieved capacity reduces greatly. The lower rate performance arises from the viscosity of high concentration aqueous cathode and the resistance of PNTCDA anode with high mass loading. This issue should be further investigated in future investigation.

Calculation of energy density

Based on the electrode reactions shown in **Fig. 1b**, the active materials in the aqueous full cell include LiI (or NaI) and PNTCDA. To calculate the energy density of the full cell, the solubility in water of LiI should also be taken into consideration. Therefore, the energy density of the aqueous full cell based on total weight of the aqueous cathode and the PNTCDA anode can be estimated according to the following equation:

$$E = \frac{Q \times m_{anode} \times V}{M_{cathode+anode}} = \frac{Q \times m_{PNTCDA} \times V}{(m_{LiI/NaI} + m_{H_2O}) + m_{PNTCDA}} \qquad (2)$$

Herein, E is the energy density (Wh kg^{-1}), Q is the theoretical capacity of the anode material PNTCDA (183 Ah kg^{-1}), V is the average voltage of the full cell (0.9V), $M_{cathode+anode}$ is the total mass of the PNTCDA anode (m_{PNTCDA}) and aqueous cathode including the mass of LiI (m_{LiI}) and water (m_{H2O}). According to the electrode reactions shown in **Fig. 1b**, theoretically the mass ratio of LiI: PNTCDA can be 0.2677kg (2 mol×0.13385kg/mol) : 0.292 kg (1mol×0. 292kg/mol). Considering the high solubility of LiI in water (~11.3mol L^{-1}), the mass of water for 0.2677 kg LiI dissolution is 0.177 kg. Thus, the calculated energy density can reach 65.3 Wh kg^{-1} for the new type aqueous Li-ion battery. Similarly, the energy density of the new type aqueous Na-ion battery based on NaI and PNTCDA can be calculated to be 63.8 Wh kg^{-1}, based on the solubility of NaI in water (~12.3mol L^{-1}).