Supplementary Information

Lithium-rich disordered rock salt $Li_{1.25}V_{0.5}Nb_{0.25}O_2$ as the anode material for lithium-ion capacitors

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Experimental section

Materials preparation

All chemicals are of analytical grade and are used directly without any purification. The following reagents were purchased from Aladdin Chemicals: Li_2CO_3 (AR, ≥99.0%), Nb_2O_5 (AR, ≥99.9%), V_2O_3 (, AR, ≥99.0%) and 1-Methyl-2-pyrrolidinone (NMP, AR, ≥99.9%). Polyvinylidene fluoride (PVDF), acetylene black and Li foil are purchased from MTI corporation. The electrolyte,1 M $LiPF_6$ in EC: DMC: EMC=1:1 vol%, was purchased from DoChem of China.

$Li_{1.25}V_{0.5}Nb_{0.25}O_2$ (named as LVNO) was synthesized by solid phase method. Precursor powders with a total weight of around 2 g were put into an 80 ml agate jar (the powder-to-ball weight ratio of was 1:20) and mixed in air at room temperature under 500 rpm for 10 h (YXQM-1L, MITR). Then, the mixture was sintered in an argon atmosphere at 950℃ for 10 h with a heating rate of 5℃ min^{-1}.

Using polyphenylene derived porous carbon (PDPC) as cathode material for LICs. The PDPC was synthesized according to our previous report. The 80 wt% PDPC, 10 wt% acetylene black and 10 wt% polytetrafluoroethylene (PTFE) were mixed and then were rolled into thin sheets. After heated at 120 ℃ for 10 h, it was stored in a desiccator.

Materials characterizations

The morphology of the samples was characterized by scanning electron microscopy (SEM, JSM-7800F, Hitachi). X-ray diffraction (XRD, D/Max-2400, Rigaku, Japan) was performed using Cu Kα radiation with a scanning range of 10° to 80°. Detailed morphological and structural characterization was conducted using transmission electron microscopy (TEM, JEM-F200, JEOL, Japan).

Battery Assembly and Testing

For the DRX-LVNO anode fabrication, the as-prepared LVNO, acetylene black, and PVDF with a 6:3:1 mass ratio is mixed in NMP using a mortar. The well-mixed slurry was uniformly coated onto a copper foil and heated at 80℃ for 8 h in a vacuum oven. The film was punched into 14 mm-diameter circular electrodes with an active material mass loading of approximately 1 mg. For the lithium half-cell assembly, the electrodes were assembled in 2032-type coin cells with Li foil, a Celgard 2400 separator, and 1 M $LiPF_6$ in EC: DMC: EMC (1:1:1 by volume) within an

Ar-filled glovebox. The O_2 and H_2O levels inside the box were stably maintained below 0.01 ppm.

For the fabrication of LICs, the anode is pre-lithiated, which the anode is charged and discharged multiple times and finally discharged to 0.01 V at a current density of 0.1 A g^{-1} in a half-cell, and then the anode electrode is dismantled in the glove box. LICs were assembled with the pre-lithium anode and the PDPC cathode. Anode and cathode active materials mass ratio is 1:1.

Electrochemical Measurements: Cyclic voltammetry (CV) test, galvanostatic charge/discharge measurements, and electrochemical impedance spectroscopy (EIS) were recorded by a CHI760E (Shanghai China) and IviumStat (Ivium Technologies BV, The Netherands). Life-span tests for half-cell and hybrid cells were conducted by a battery test system (Land CT2001A model, Wuhan Land Electronics, Ltd.). All electrochemical tests were carried out in a thermostatic chamber. The temperature was controlled at 30 ± 1 °C throughout the entire testing process.

The energy density and power density of the full cells were calculated based on the following equations:

$$E = \int_{t1}^{t2} IVdt$$

$$P = E/t$$

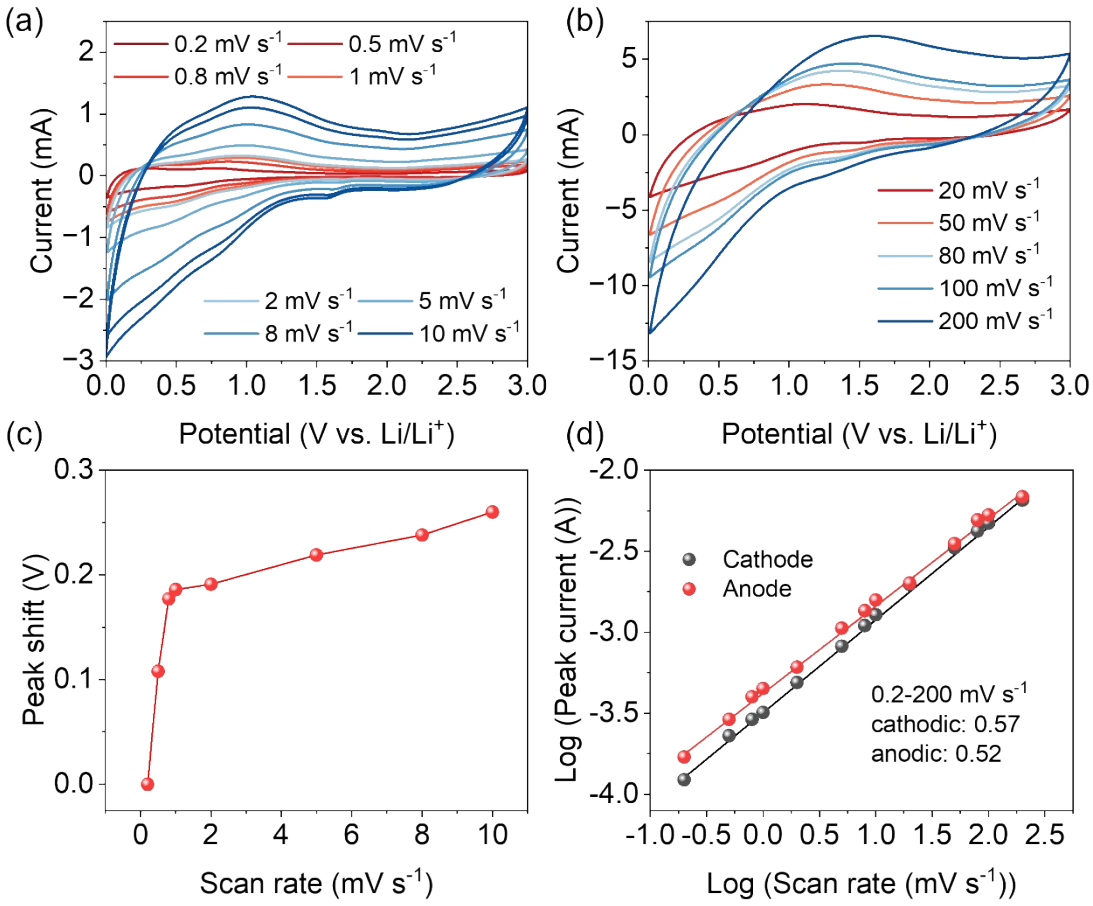

Fig. S1. (a, b) CV curves of LVNO electrode with different scan rates ranging from 0.2 to 200 mV s^{-1}. (c) The variation of cathodic peak voltages with the different scan rates ranging from 0.2 to 10 mV s^{-1}. (d) b-values determination of the anodic and cathodic peak currents.

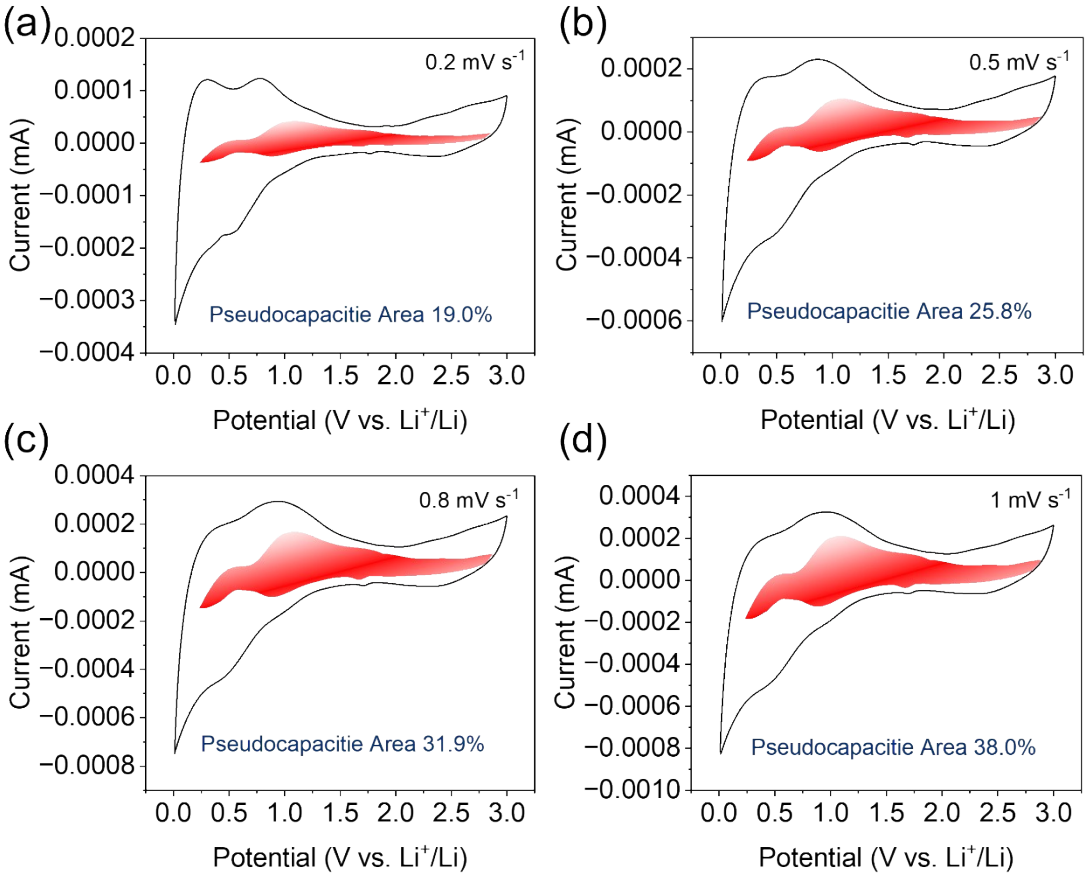

Fig. S2. Capacitive contribution (red region) to the total charge storage at 0.2 mV s^{-1} (a), 0.5 mV s^{-1} (b), 0.8 mV s^{-1} (c), 1 mV s^{-1} (d).

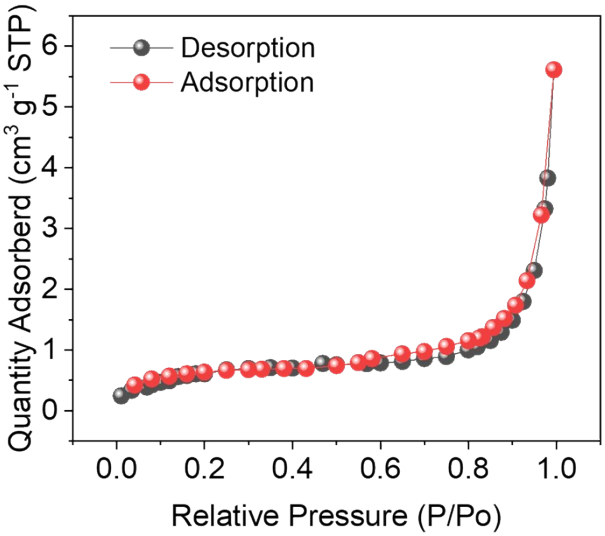

Fig. S3. N_2 adsorption–desorption isotherms of DRX-LVNO.

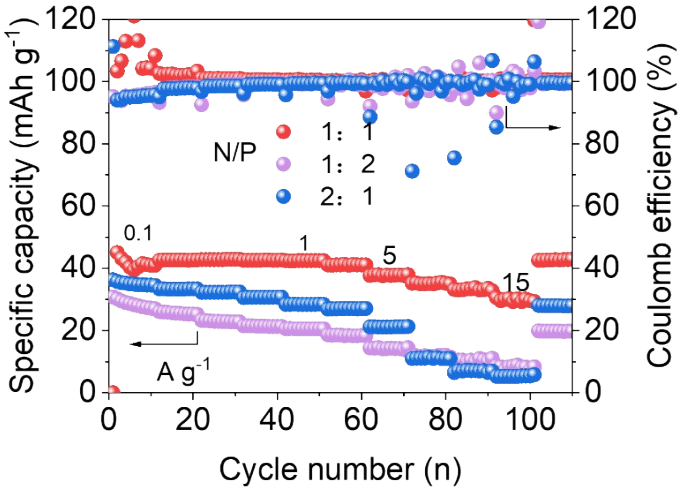

Fig. S4. The electrochemical performance of as-fabricated LIC with the different anode/cathode mass ratios.

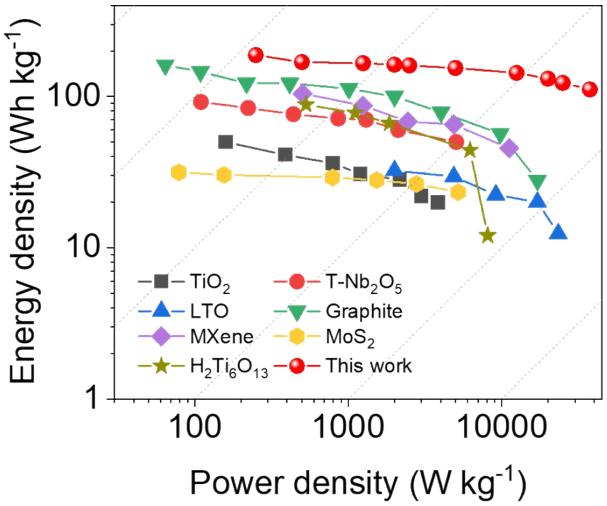

Fig. S5. Ragone plots showing energy and power densities versus other reports.