Supporting information:

Hetero-chelation boosting sodium storage in π -d conjugated coordination polymers

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Materials

All commercially available chemical reagents and solvents were purchased from Energy Chemical or Sinopharm Chemical Reagent Co., Ltd, and without additional purification.

Synthesis of Ni-DTA

In a 250 mL three-neck flask, 1 mmol of dithiooxamidato were dissolved in 50 mL degassed DMF. The flask was put into ice bath and the oxygen was eliminated with Ar bubbling. 1 mmol NiCl₂ were dissolved in 50 mL of degassed water and the solution were added drop wise to the aforementioned solution. Then triethylamine (0.4 mL) were added drop wise. Upon addition of the base, the dark brown precipitates were formed. The solution was stirred for another 72 hours at room temperature. The black solid was collected through centrifugation, washed with DMF, water, acetone and ethanol. The resultant black powder was dried at 70 °C under vacuum for 12 h. Anal. Calcd for [Ni(C₂H₂S₂N₂)] (%) by using infinite polymerization degree: C, 13.58 ; H, 1.14; Ni, 33.18; N, 15.84; S, 36.26. Found (%): C, 13.12; H, 2.19; Ni, 32.39; N, 16.06; S, 36.19.

Synthesis of Ni-DABDT

In a 250 mL three-neck flask, 1 mmol of 2,5-diamino-1,4-dithiolbenzene were dissolved in 50 mL degassed DMF. The flask was put into ice bath and oxygen was eliminated with Ar bubbling. 1 mmol NiCl₂ were dissolved in 50 mL degased water and the solution were added drop wise to the aforementioned solution. Then a small bottle with saturated NH₄OH were added in the flask. The NH₃ gas will slowly diffuse into the mixed solution. The mixed solution were stirred for 72 hours at room temperature. The black solid was collected through centrifugation, washed with DMF, water, and ethanol. The resultant black powder was dried at 70 °C under vacuum for 12 h. Anal. Calcd for [Ni(C₆H₄S₂N₂)] (%) by using infinite polymerization degree: C, 31.76; H, 1.78; Ni, 25.86; N, 12.35; S, 28.26 . Found (%): C, 31.19 ; H, 2.17; Ni, 26.19; N, 12.81; S, 27.58.

Materials characterization

Elemental analyses including C, H, N and S were measured by a Vario Micro Cube Elemental Analyzer. The structure characterizations and morphology were obtained by scanning electron microscope (ZEISS Gemini 300) and transmission electron microscope (JEOL 2100). The nickel contents were recorded in Agilent ICP-OES 730. Powder XRD diffraction patterns were measured with a Cu-Ka X-ray radiation source ($\lambda = 0.154056$ nm) incident radiation by an X'Pert3 Powder instrument. The FT-IR spectra were recorded in the range 400-4000 cm⁻¹ on Bruker ALPHA spectrometer. XPS measurements were carried out on a Thermo Fisher EscaLab 250Xi using a monochromic Al X-ray source (hv = 1486.6 eV). EPR spectra were acquired on Bruker EMS nano spectrometer at room temperature. Microwave frequency was set at 9.588 GHz (X-band) with a power of 19 mW.

Electrochemical measurement

The Ni-DABDT and Ni-DTA electrode were obtained by mixing the active material with super P as conductive additives and sodium carboxymethyl cellulose (CMC) as the binder in a weight ratio of 6:3:1. The slurry was coated onto copper foil and dried at 70 °C in vacuum overnight. The mass loading of the active material is $1\sim2$ mg cm⁻². The 2032 coin cells were assembled in glove box with Ar atmosphere and 1 M NaPF₆ in DME was used as the electrolyte. GCD charge/discharge curves were performed in potential range of 0.01-2.8 V on LAND CT2001A battery test system (Wuhan, China) at room temperature. Bio-logic VMP3 was used for cyclic voltammetry measurements, galvanostatic intermittent titration technique measurements and electrochemical impedance spectrum measurements. The electrodes for XPS and FT-IR measurements were prepared by mixing the active material with super P and polyvinylidene fluoride (PVDF) in a weight ratio of 7:2:1. For the construction of full cells, the Na₃V₂(PO₄)₃ (NVP) cathode and Ni-DTA (or Ni-DABTD) anode materials were precycled in half cells, and assembled in 2032 coin cells with the cathode to anode mass ratio of ≈ 6 . The current density and capacity were calculated on the mass of anode.

Titration methods

The electrode after discharged was cleaned with anhydrous DMC and sealed in a Arfilled tube. Then, the deoxygenated dilute sulfuric acid (2 mL, 1 mol/L) was injected into the tube, and the reaction was fully carried out by shaking the tube. The produced gas was checked by GC, and the hydrogen content was calculated by comparing the results with the hydrogen standard sample, and then the volume of hydrogen produced was obtained. For every molecule of hydrogen produced, two electrons need to be transferred, therfore the number of electrons transferred by the active material can be calculated from the amount of hydrogen. The detail computational equation can be expressed as below:

$$n = 2 \times \frac{PV}{RT(\frac{m}{M})} \times \frac{CS_1}{S_2}$$

Where n is electron transfer number per repeating unit; P is atmosphere pressure; V is the volume of gas (the closed tube); C is concentration of hydrogen in standard gas sample; R is the ideal gas constant: 8.314 J K⁻¹ mol⁻¹; T is the temperature: 298.15 K; m and M is the mass and molar molecular weight of active material; S_1 is the integral area of hydrogen produced by the active material; S_2 is the integral area of hydrogen standard sample.

Details of GITT measurement

The cells were charged and discharged at constant current density of 0.2 A g⁻¹ for 5 min ($E\tau$) and then rest for 30 min to reach the steady-state voltage (Es). The sodium ion diffusivity D^{GITT} can be obtained via the following formula:

 $D^{\text{GITT}} = \frac{4}{\pi\tau} \left(\frac{m_B V_M}{M_B S}\right)^2 \left(\frac{\Delta E s}{\Delta E \tau}\right)^2$, Where τ is the constant current pulse time, m_B, V_M, S, and M_B are the mass loading, molar volume, electrode–electrolyte interface area, and molar mass of the CCPs respectively. The V_M were obtained by calculated the mass and the total volume of pressed Ni-DABDT and Ni-DTA piece. Δ Es is the voltage difference during the open circuit period, and Δ Et is the total change of cell voltage during a constant current pulse excluding the IR drop.

Details of theoretical calculations

All the calculations were performed with CASTEP¹, as implemented in Material Studios software². The DFT was applied to geometry optimizations at the level of GGA/PBE³. The ultra-soft pseudopotential was used to describe the interaction between valence electrons and ionic core. The cutoff energy for the plane wave expansion was 500.0 eV. Furthermore, Hubbard U correction^{4, 5} in the form of GGA+U was applied to compute on Ni (U = 4.0 eV, J = 0 eV) surfaces. The spin polarization calculations performed for all simulations with the consideration of

empirical dispersions of Grimme (DFT-D3) for the long-range van der Waals (vdW) interactions.⁶ In order to maintain the same computational accuracy for all CCPs, the k-point mesh was sampled using a $9 \times 2 \times 4$ Γ -centered grid for Ni-TTO, $4 \times 2 \times 3$ for Ni-BTA, $10 \times 2 \times 4$ for Ni-DTA and $3 \times 2 \times 3$ for Ni-DABDT, respectively.⁷

The binding energy per Na ion, E_{bind} , of the Ni-CCP-nNa complex was calculated by using the following equation:

 $E_{bind} = (E_{CCP-nNa} - E_{CCP} - E_{nNa})/n$

where $E_{CCP-nNa}$ is the energy of the complex with n Na ions inserted into the CCP, and E_{CCP} is the energy of pristine (Ni-DABDT and Ni-DTA). The value of n (n = 1 to 5) represents the number of Na ions.

All optimized structures of Ni-DABDT or Ni-DTA with different Na ions insertion content, n, were given in Fig. S28 or Fig. S29 and the corresponding binding energy were given in Table S4.

Although the simulated structures of Ni-DABDT or Ni-DTA adopted herringbone structures, two possible positions for the hosting of Na ions were calculated: close to the lateral position (bay position) of the molecular plane (called a position) or close to the top/bottom of the molecular plane (b position, more close to the π electron cloud). Different k-points were adopted for Ni-DABDT and Ni-DTA, according to the different cell parameters. It was found that the Ni-DABDT and Ni-DTA had stronger binding interactions with the inserted Na ions than those of Ni-BTA and Ni-TTO. The co-existence of nitrogen and sulfur atom in Ni-DABDT and Ni-DTA hare capable of accommodating more Na ions than Ni-BTA and Ni-TTO during the charge-discharge process.

The calculations of ESP were performed with the Gaussian 16 package suite.⁸ Structures were optimized by using DFT with the B3LYP functional^{9, 10}, and the 6-31+G(d,p) basis set was employed for all atoms¹¹⁻¹⁴. Frequency calculations at the same level of theory were also performed to identify all the optimized structures were at a local minimum on the potential energy surface. The optimized structures were then used in ESP calculations. The ESP map analyses were performed with the Multiwfn 3.8 program¹⁵ and the isosurface maps were drawn using Visual Molecular Dynamics (VMD) software¹⁶.



Fig. S1. XPS spectra of the obtained Ni-DTA powder. a) Full spectra, indicating the presence of C, N, S and Ni. b) S_{2p} spectra. c) N_{1s} spectra. d) Ni_{2p} spectra. The coexistence of high valence and low valence in N_{1s} and S_{2p} spectra can be assigned to the coexistence of single bonds and double bonds, indicating the resonance states of Ni-DTA, which are coincident with the previously proposed ideal structures of CCPs.



Fig. S2. XPS spectra of the obtained Ni-DABDT powder. a) Full spectra, indicating the presence of C, N, S and Ni. b) S_{2p} spectra. c) N_{1s} spectra. d) Ni_{2p} spectra. The coexistence of high valence and low valence in N_{1s} and S_{2p} spectra can be assigned to the coexistence of single bonds and double bonds, indicating the resonance states of Ni-DABDT, which are coincident with the previously proposed ideal structures of CCPs.



Fig. S3. a) FT-IR spectra of the Ni-DTA, showing the presence of N-H, C=N, C=C, C-N, C-C and C-S. b) EPR spectra of the Ni-DTA.



Fig. S4. a) FT-IR spectra of the Ni-DABDT, showing the presence of N-H, C=N, C=C, C-N, C-C and C-S. b) EPR spectra of the Ni-DABDT.



Fig. S5. (a-b) SEM images and (c-d) XRD patterns of Ni-DTA (a,c) and Ni-DABDT (b,d).



Fig. S6. a) HAADF-STEM image and corresponding EDS mapping for the Ni-DTA: b) C, c) N, d) S, e) Ni, f) total elemental mappings of C, N, S, and Ni. The results showed that the elements are homogenously distributed in the Ni-DTA samples.



Fig. S7. a) HAADF-STEM image and corresponding EDS mapping for the Ni-DABDT: b) C, c) N, d) S, e) Ni, f) total elemental mappings of C, N, S, and Ni. The results showed that the elements are homogenously distributed in the Ni-DBDT samples.



Fig. S8. N₂ adsorption and desorption isotherm profile at 77 K. a) the Ni-DABDT; c) the Ni-DTA; the pore distribution b) the Ni-DABDT; d) the Ni-DTA.



Fig. S9. The TGA curves of Ni-DTA and Ni-DABDT.



Fig. S10. The conductivity of the Ni-DABDT and Ni-DTA at room temperature, which were measured by using pressed pellets: a) the diameter, b) the thickness and c) the Current-Voltage relationship of the pressed pellets Ni-DABDT; d) the diameter, e) the thickness and f) the Current-Voltage relationship of the pressed Ni-DTA pellets.



Fig. S11. a) The optical image of the discharged Ni-TTO electrodes (at 0.01 V) in the methanol solution (containing 0.1% NH₄OAc); and b) the corresponding solution after removing the electrodes by filtration. The yellow solution confirmed the decomposition of the Ni-TTO electrodes. c) Typical ESI-MS spectra (in positive mode in the solution of (MeOH, 0.1wt% of NH₄OAc)) of the Ni-TTO electrodes after being discharged: regionally amplified spectra showing the presence of Na₄-ETT.



Fig. S12. CV curves of Ni-DTA electrodes ranging from 0.01 to 2.8 V (vs. Na/Na⁺) at a scan rate of 0.2 mV s⁻¹.



Fig. S13. Cycle performance of Ni-DTA at different voltage ranges: (a) $0.01 \sim 2.8$ V and (b) $1.0 \sim 2.8$ V. The activation process of Ni-DTA in the initial cycles may be caused by the large amount of sodium ions embedded in the structures during the discharge process ($0.01 \sim 2.8$ V). On the other hand, the activation process of Ni-DTA was not obvious when the voltage range was from 1.0 to 2.8 V.



Fig. S14. High resolution transmission electron microscopy (HRTEM) images of (a, c) Ni-DTA and Ni-DABDT powders, and (b, d) Ni-DTA and Ni-DABDT electrodes after discharged. An obvious amorphous SEI layer was formed on the surface of Ni-DTA and Ni-DABDT electrodes. What's more, the interfacial distance increased slightly after discharging due to the insertion of Na-ions.



Fig. S15. The integral areas for the redox peaks of Ni-DTA electrodes ranging from 0.01 to 2.8 V (vs. Na/Na⁺) at a scan rate of 0.2 mV s⁻¹. The first reduction peak of Ni-DTA is at about 1.5 V, and the corresponding integral area is 0.304 mA V. After that, a broad reduction peak was observed below 1 V, corresponding to an integral area of 0.593 mA V. The ratio of the integral areas for the two peaks is around 1:2, which is quite close to the proposed succesive one- and two-electron process. For the charging process, although the oxidation peaks appear at high voltages, the integral areas of two oxidation peaks are close to their corresponding reduction peaks (e.g. 0.485 vs. 0.593 mA V, and 0.409 vs. 0.304 mA V). The difference between the reduction peak and the oxidation for each pairs of redox peaks should be ascribed to the overlap between the two oxidation peaks, which make it difficult to distinguish. Moreover, the total integral area of the two reduction peaks (0.895 mA V), corresponding to a three-electron transfer.



Fig. S16. The electrochemical performance of the conductive additives (Super P). a) Cyclic volumetric curves of Super P in the range of 0.01-2.8 V at a scan rate of 0.5 mV s⁻¹; b) the cycling performance of Super P at a current density of 0.2 A g⁻¹; c) long cycling stability of Super P at a current density of 2 A g⁻¹; d) the rate performance of Super P at various current densities from 0.2 to 5 A g⁻¹.



Fig. S17. a) The optical image of the discharged Ni-DTA electrodes (at 0.01 V) in the methanol solution (containing 0.1% NH₄OAc); and b) the corresponding solution after removing the electrodes by filtration.

CCPs	Chemical structure	Potential windows (V)	Reversible capacity at low rate (mAh g ⁻¹) (Current (mA g ⁻¹), cvcles)	High rate reversible capacity (mAh g ⁻¹) (Current (mA g ⁻¹), cvcles)	Mass loading (mg/cm ²)	Ref.
Со-НАВ (Со-N ₄)	2D	0.5-3.0	190 (50, 50)	185 (4000, 150)	1~1.5	17
Ni-BTA (Ni-N ₄)	1D	0.01-2.5	~450 (100, 90)	250 (10000, 2000)	~1	18
Ni- TABQ (Ni-N ₄)	1D	0.2-3.0	450 (200, 100)	330 (2000, 450)		19
Ni-TTO (Ni-S ₄)	1D	1.2-3.2	130 (100, 100)	104 (1000, 1000)	~2	20
Ni-	1D	0.01-2.5	~340 (100, 100)	~250 (1000, 500)	1~1.5	
(Ni-O ₄)	ID	0.5-2.3	~230 (100, 100)	~200 (1000, 200)	1~1.5	
Mn- DHBQ (Mn-O ₄)	1D	0.01-2.5	~250 (100, 100)	~110 (1000, 500)	1~1.5	21
Zn- DHBQ (Zn-O ₄)	1D	0.01-2.5	~180 (100, 100)	~100 (1000, 500)	1~1.5	
Cu- HHTP (Cu-O ₄)	2D	0.01-2.4	~221 (100, 200)	76 (2000, -)	~0.4	22
Cu- HHTP (Cu-O ₄)	2D	1.0-3.6	~150 (100, 200)	~130 (1000, 500)	0.7~1	23
Zn- HHTP (Zn-O ₄)	2D	1.0-3.5	~150 (100, 200)	~100 (1000, 1000)	0.7~1	
Ni-DTA (Ni- S ₂ N ₂)	1D	0.01-2.8	492 (200, 100)	382 (2000, 800)	1~2	This work
$\begin{array}{c c} \hline \mathbf{Ni-} \\ \mathbf{DABDT} \\ (\mathbf{Ni-} \\ \mathbf{S_2N_2}) \end{array}$	1D	0.01-2.8	517 (200, 100)	334 (2000, 1000)	1~2	This work

Table S1. Electrochemical performance of reported CCPs in sodium-ion batteries.The secondary building units are shown after the abbreviated name of these CCPs

Positive mode		Negative mode		
cation	u	anion	u	
${Na_3[DTA]}^+$	188.95	${Na_2 [DTA]}^-$	165.96	
$\{ Na_3 [DTA] + NaOAc \}^+$	270.95	${Na_2 [DTA] + NaOAc}^-$	247.96	
$\{ Na_3 [DTA] + 2 NaOAc \}^+$	352.96	${Na_2 [DTA] + 2NaOAc}^-$	329.97	
$\{ Na_3 [DTA] + 3 NaOAc \}^+$	434.96	${Na_2 [DTA] + 3NaOAc}^-$	411.97	
$\{ Na_3[DTA] + 4 NaOAc \}^+$	516.96	${Na_2 [DTA + 4NaOAc}^-$	493.97	
$\{ Na_3 [DTA] + 5 NaOAc \}^+$	598.97	${Na_2 [DTA] + 5NaOAc}^-$	575.98	

Table S2: Possible ESI-MS signals (in positive or negative mode) for Ni-DTAfragments after being discharged.

Table S3: Possible ESI-MS signals (in positive or negative mode) for sulfide spices.

Positive mode		Negative mode		
cation	u	anion	u	
${Na_3S}^+$	100.94	{NaS}-	54.96	
$\{ Na_3S + NaOAc \}^+$	182.94	${NaS + NaOAc}^-$	136.96	
$\{ Na_3S + 2 NaOAc \}^+$	264.94	${NaS + 2NaOAc}^-$	218.96	
$\{ Na_3S + 3 NaOAc \}^+$	346.95	{NaS + 3NaOAc}-	300.97	
$\{ Na_3S + 4 NaOAc \}^+$	428.95	${NaS + 4NaOAc}^{-}$	382.97	
$\{ Na_3S + 5 NaOAc \}^+$	510.95	{NaS + 5NaOAc}-	464.97	



Fig. S18. ESI-MS spectra (in positive mode in the solution of (MeOH, 0.1wt% of NH₄OAc)) of the Ni-DTA electrodes after being discharged.



Fig. S19. ESI-MS spectra (in positive mode in the solution of (MeOH, 0.1wt% of NH₄OAc)) of the Ni-DTA electrodes after being discharged. No peaks can be assigned to the DTA species (see possibilities in Table S2).



Fig. S20. ESI-MS spectra (in positive mode in the solution of (MeOH, 0.1wt% of NH₄OAc)) of the Ni-DTA electrodes after being discharged. No peaks can be assigned to the sulfide species (see possibilities in Table S3).



Fig. S21. ESI-MS spectra (in negative mode in the solution of (MeOH, 0.1wt% of NH₄OAc)) of the Ni-DTA electrodes after being discharged.



Fig. S22. ESI-MS spectra (in negative mode in the solution of (MeOH, 0.1wt% of NH₄OAc)) of the Ni-DTA electrodes after being discharged. No peaks can be assigned to the DTA species (see possibilities in Table S2).



Fig. S23. ESI-MS spectra (in negative mode in the solution of (MeOH, 0.1wt% of NH₄OAc)) of the Ni-DTA electrodes after being discharged. No peaks can be assigned to the sulfide species (see possibilities in Table S3).



Fig. S24. a) Ex-situ FT-IR spectra of the Ni-DTA electrodes at different voltages. b-d) The XPS spectra of Ni-DTA electrodes at different voltages. b) N_{1s} spectra. c) S_{2P} spectra. d) Ni_{2P} spectra. The IR spectra indicated that the whole ligand was conjugated and the entire system was reduced from more double bond characters of C=Y bonds (Y=C, S or N) to more single bond characters of C-Y bonds. The conjugation of the ligand could be further proved by the appearance of S=C=N bonds, showing the resonance of the structure and the delocalization of electrons in the conjugated system. The presence of the S=C=N peaks afterwards suggested that the whole structure was activated and the electrons were more delocalized in the system. The XPS spectra also confirmed the reversible conversion of both C=S and C=N double bond characters into C-S and C-N single bond characters in the charge-discharge process.



Fig. S25. EPR spectra of Ni-DTA electrodes during cycling. The purple line had a g-value close to 2.002, while the other lines showed a similar g-value of close to 2.001.



Fig. S26. a) CV curves and b) charge-discharge profiles of Ni-DABDT after 5 cycles, respectively. The CV curves and charge-discharge profiles showed insignificant varies after 20 cycles.



Fig. S27. a) Charge-discharge curve of NVP cathode at 0.1 C (half cells). b) Charge-discharge profile and c) cycling stability of NVP \parallel Ni-DTA full cells at 0.2 A g⁻¹. d) Charge-discharge profile and e) cycling stability of NVP \parallel Ni-DABDT full cells at 0.2 A g⁻¹. A g⁻¹.



Fig. S28. a) Ex-situ FT-IR spectra of the Ni-DABDT electrodes at different voltages. b-d) The XPS spectra of Ni-DABDT electrodes at different voltages. b) N_{1s} spectra. c) S_{2P} spectra. d) Ni_{2P} spectra. The IR spectra indicated that the whole ligand was conjugated and the entire system was reduced from more double bond characters of C=Y bonds (Y=C, S or N) to more single bond characters of C-Y bonds. The XPS spectra also confirmed the reversible conversion of both C=S and C=N double bond characters into C-S and C-N single bond characters in the charge-discharge process.



Fig. S29. EPR spectra of Ni-DABDT electrodes during cycling.



Fig. S30. Isosurface maps of the ESP for (a) Ni-TTO (isovalue 0.001 a.u.), (b) Ni-BTA (isovalue 0.001 a.u.), (c) Ni-DTA (isovalue 0.001 a.u.) and (d) Ni-DABDT (isovalue 0.001 a.u.). Red and blue colors correspond to positive and negative parts of ESP, respectively. The part selected in the dotted box is the minimum area of ESP, with which metal ions are likely to bind.

Table S4. The binding energy of Ni-DABDT/DTA-nNa with different Na content n.k-point: 3×2×3(Ni-DABDT), 4×2×2 (Ni-DTA)

unit	n	position	E_{bind}/eV
	1	a	-2.08
	1	b	-2.3
	2	a	-1.76
	2	b	-1.36
N: DADDT nNo	2	а	-0.69
INI-DADD I-IIINa	J	b	-0.81
	4	a	-0.20
	4	b	-0.09
	5	a	0.08
		b	0.03
	1	a	-1.74
	1	b	-1.36
Ni-DTA-nNa	2	a	-0.68
	2	b	-0.99
	2	a	-0.24
	3	b	-0.28
		a	-0.04
	4	b	0.12
	E	a	0.51
	5	b	0.47



Fig. S31. The binding energy of Ni-CCP-nNa. Inset: the most possible structures Ni-DABDT and Ni-DTA after insertion of Na ions. Although the binding energy was also negative for Ni-DTA with acceptance of four Na ions, the structure of Ni-DTA showed significant distortion.



Fig. S32. Simplified structures (top view) to demonstrate the most possible sites of inserted Na ions for Ni-DTA and Ni-DABDT. Ni-BTA was also shown for comparison. The co-existence of S and N as chelating atoms enabled the storage of more electrons, which could also be verified by the variation of C=C/C-C bonds during cycling.



Fig. S33. The electrochemical impedance spectra of the Ni-DABDT electrode, the 1st, 2^{nd} , 25^{th} , 100^{th} cycles were selected as representatives. After the first discharge, the electrode showed a resistance of 13 Ω , and after the second cycles the resistance decreased to 2 Ω , which kept almost unchanged after 100 cycles.



Fig. S34. The electrochemical impedance spectra of the Ni-DTA electrode, the 1st, 2nd, 25th, 100th cycles were selected as representatives. A resistance of 3 Ω was recorded after the first discharge. After the second cycles, the resistance decreased to 1.5 Ω , which slightly increased in the following cycles to 5 Ω after 25 cycles and 16 Ω after 100 cycles.



Fig. S35. a) CV curves of Ni-DABDT electrodes at different scan rates from 0.1 to 2 mV s⁻¹. b) The log relationship between the absolute value of the peak current and the scan rate. c) GITT potential response curve with time for one typical step. d) The plot of Na-ion diffusivity calculated from GITT methods versus potential.

Table S5. Slopes of each peak in the log i vs. log v plots in Fig. S35b.

Peak	A1	A2	C1	C2
Slope	0.74	0.82	0.81	0.75



Fig. S36. a) CV curves of Ni-DTA electrodes at different scan rates from 0.1 to 2 mV s⁻¹. The CV curves of Ni-DTA were obtained after activation for 20 cycles at slow rate of 0.2 mA g⁻¹. b) The log relationship between the absolute value of the peak current and the scan rate. c) GITT potential response curve with time for one typical step. d) The plot of Na-ion diffusivity calculated from GITT methods versus potential.

Table S6. Slopes of each peak in the log i vs. log v plots in Fig. S36b.

Peak	A1	A2	C1	C2
Slope	0.95	0.82	0.80	1.00



Fig. S37. a) One full GITT measurement for Ni-DABDT. b) The mass, diameter and thickness of the Ni-DABDT pellets prepared by isostatic pressing, for calculating the density (1.338 g cm⁻³) and V_M . The calculated volumetric capacity of Ni-DABDT (474 mAh g⁻¹) was 634.2 mAh cm⁻³.



Fig. S38. a) One full GITT measurement for Ni-DTA. b) The mass, diameter and thickness of the Ni-DTA pellets prepared by isostatic pressing, for calculating the density (1.508 g cm⁻³) and V_M . The calculated volumetric capacities of Ni-DTA (456 mAh g⁻¹) is 687.6 mAh cm⁻³.

References

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