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Electronic Supporting Information

A Facile and Scalable Synthesis of Sulfur, Selenium and Nitrogen co-doped Hard Carbon Anode for High Performance Na- and K-ion Batteries

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Kinetics and quantitative analysis

Generally, the relationship between the peak current and the scan rate is in harmony with a power law, according to Equation (S1):[1, 2]

$$i(\mathrm{V})=\mathrm{a}v^{\mathrm{b}} \tag{S1}$$

where i represents peak current, ν represents scan rate, a and b are adjustable constants. By sketching the relationship of log(i) against log(ν), the value of b can be calculated. When the value of b equals 0.5, it indicates an ideal faradaic intercalation process, while the value gets close to 1, it presents a surficial charge storage procedure without diffusion process.

Moreover, the capacity contribution can be further quantitatively separated into capacitive effects (k_1v) and diffusion-controlled reactions ($k_2v^{1/2}$), according to the Equation (S2):[1, 2]

$$i(\mathrm{V})=k_1v+k_2v^{1/2} \tag{S2}$$

The current response i at a fixed potential can be regarded as the combination of surface-controlled and diffusion-dependent processes, which are proportional to v and $v^{1/2}$. By counting k_1 and k_2 at fixed potentials, the proportion of the current from each contribution can be quantified into the two processes.

The diffusion coefficient (D) of electrode is calculated from the galvanostatic intermittent titration (GITT) potential profiles using Fick's second law with the following equation(S3):

$$D=\frac{4}{\pi\tau}\left(\frac{m_BV_M}{M_BS}\right)^2\left(\frac{\Delta E_S}{\Delta E_\tau}\right)^2 \tag{S3}$$

where τ is the duration of the current impulse; m_B is the mass load of the electrode material; S represents the geometric area of the electrode; ΔE_S is the quasi-thermodynamic equilibrium potential difference between before and after the current pulse; ΔE_τ represents the potential difference during the current pulse; V_M is the molar volume of the materials; and M_B is the molar mass of carbon. The relative value of M_B/V_M can be calculated from the density of materials, which is calculated with Equation (S4)

$$\rho = \frac{1}{V_{total} + \frac{1}{\rho_{carbon}}} \tag{S4}$$

where ρ (g cm^{-3}) is the density of materials, V_{total} (cm^3 g^{-1}) is the total pore volume analyzed from the N_2 isotherm, and ρ_{carbon} is the true density of carbon (2 g cm^{-3}).

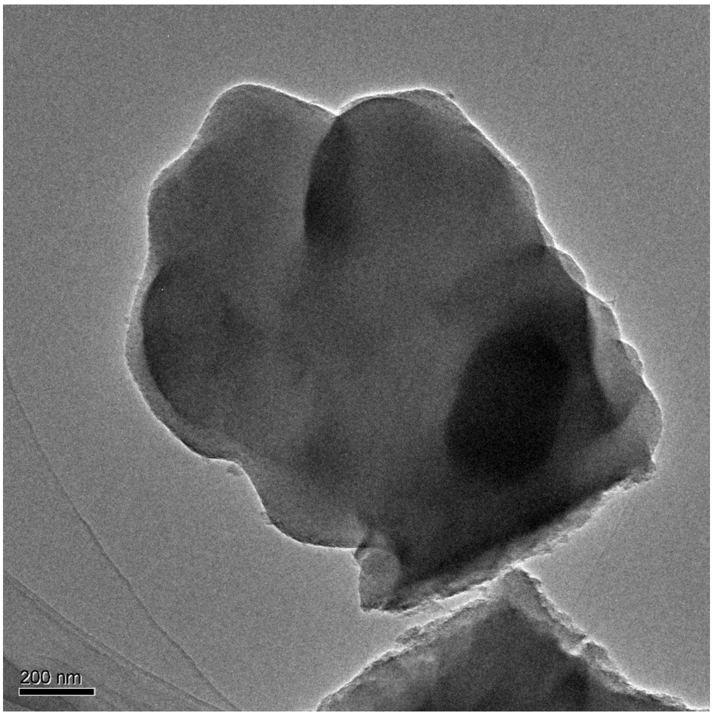

Figure S1 The TEM image of CPAN.

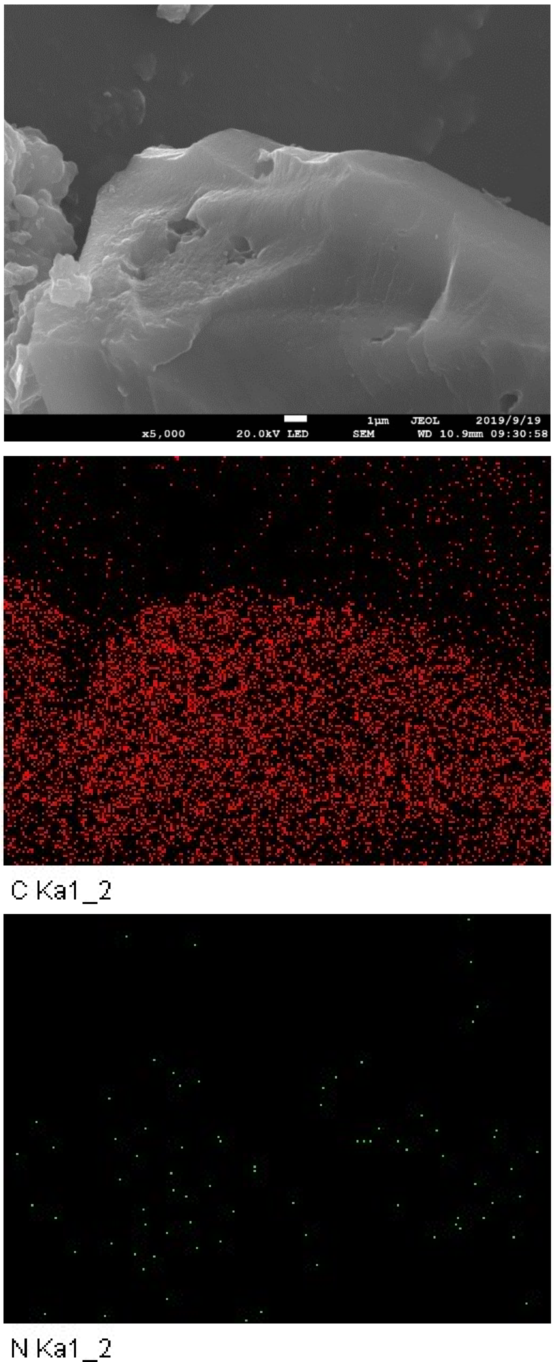

Figure S2 The EDS mapping of CPAN.

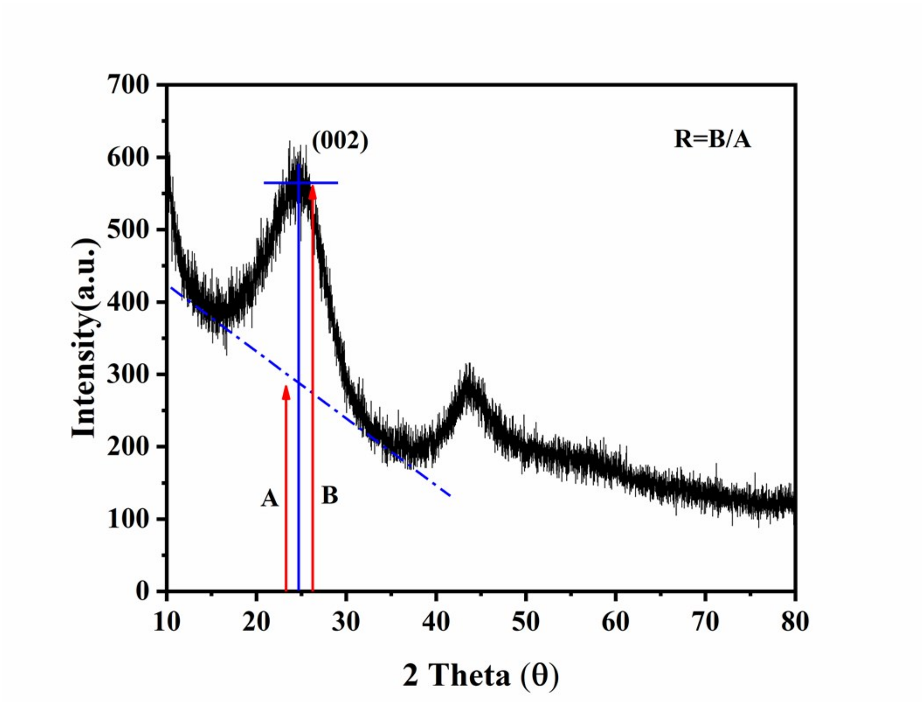

Figure S3 Calculation of the R value. Illustration of the calculation of the empirical R value according to the study of Dahn et al[3]. The XRD pattern of SSHC and CPAN are used as an example.

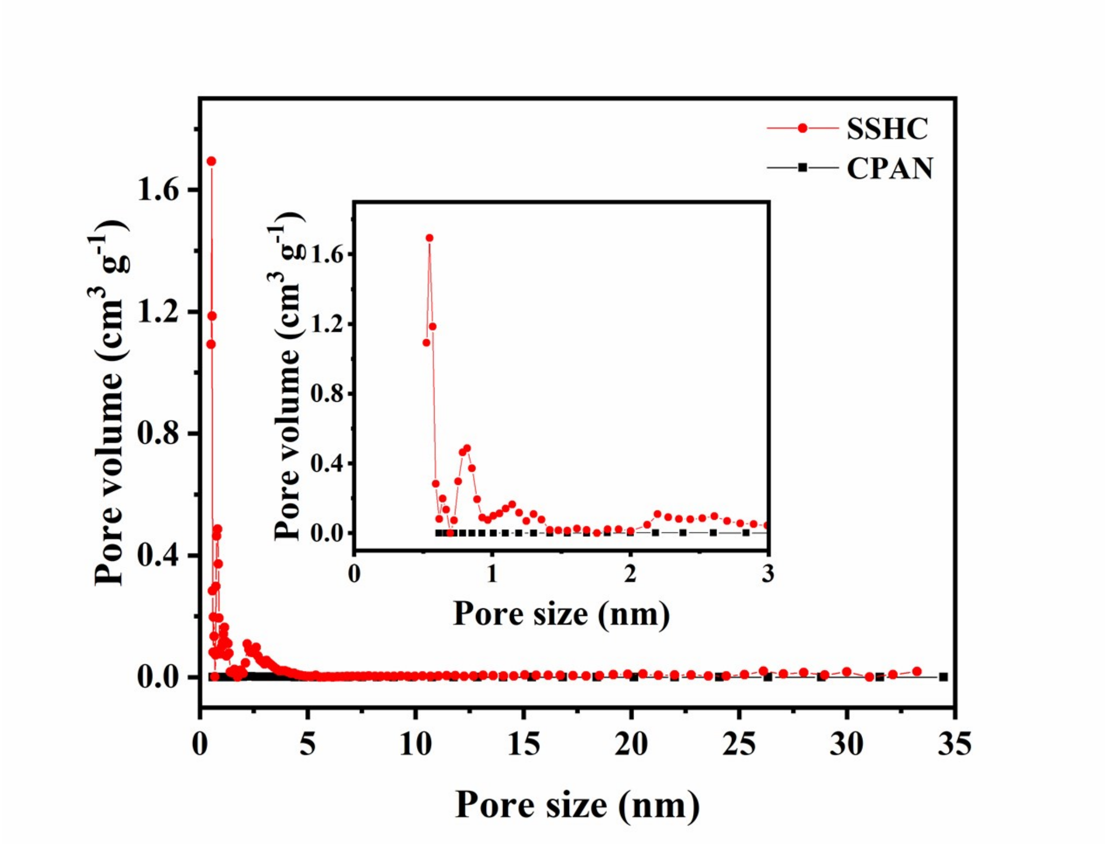

Figure S4 Pore size distribution of the CPAN and SSHC.

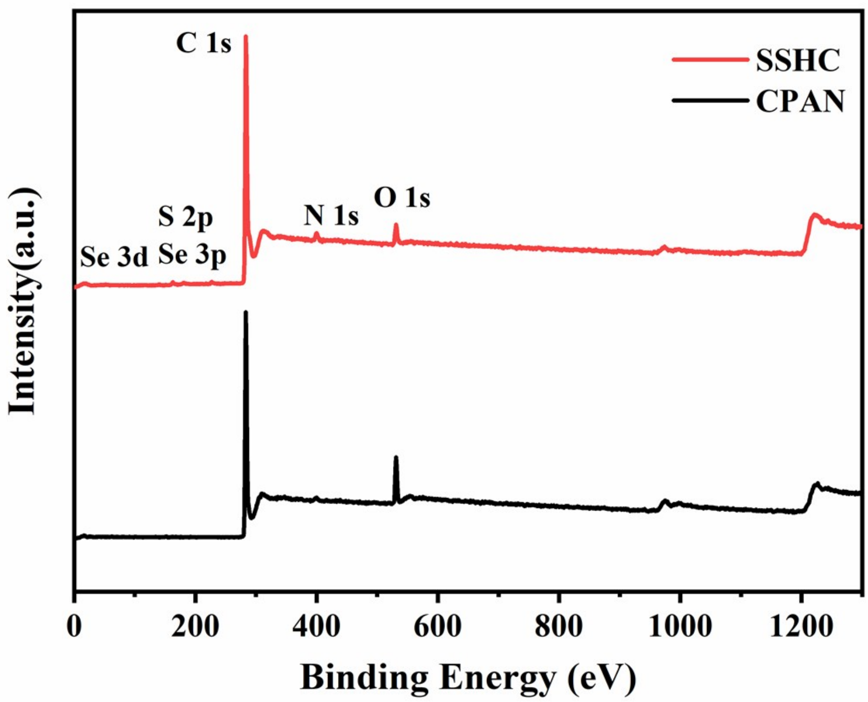

Figure S5 XPS survey spectra of the CPAN and SSHC.

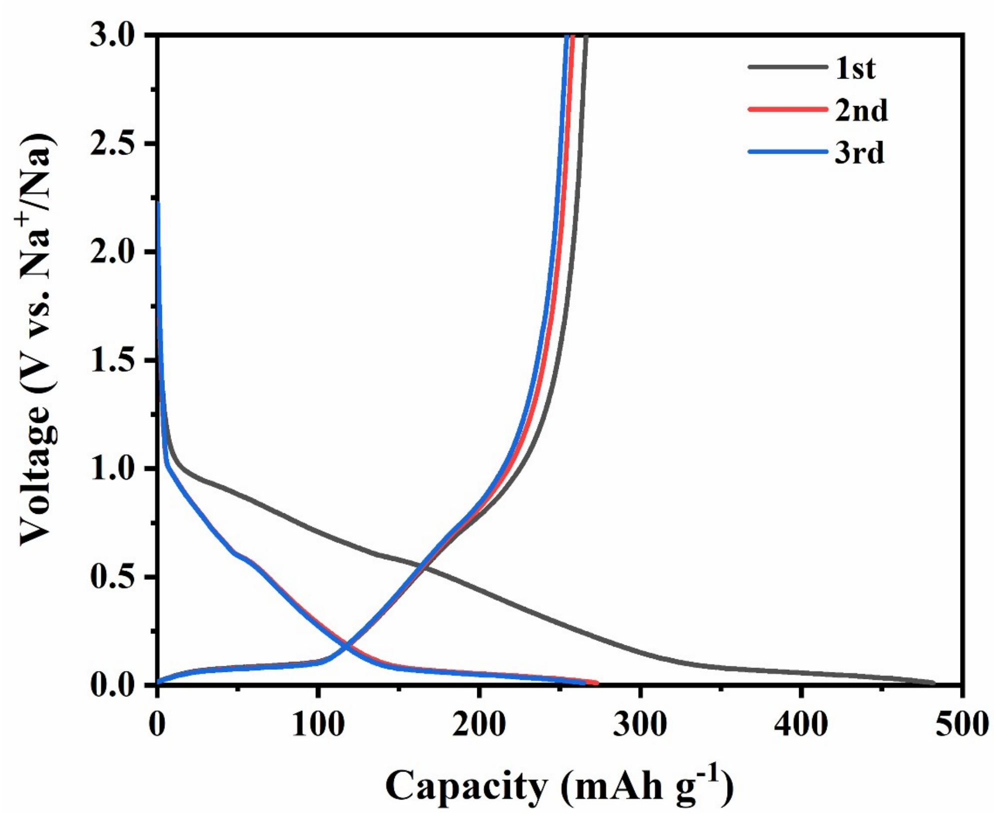

Figure S6 Electrochemical evaluation of the SSHC electrode in NIBs: Charge/discharge curves at a current density of 0.1 A g^{-1}.

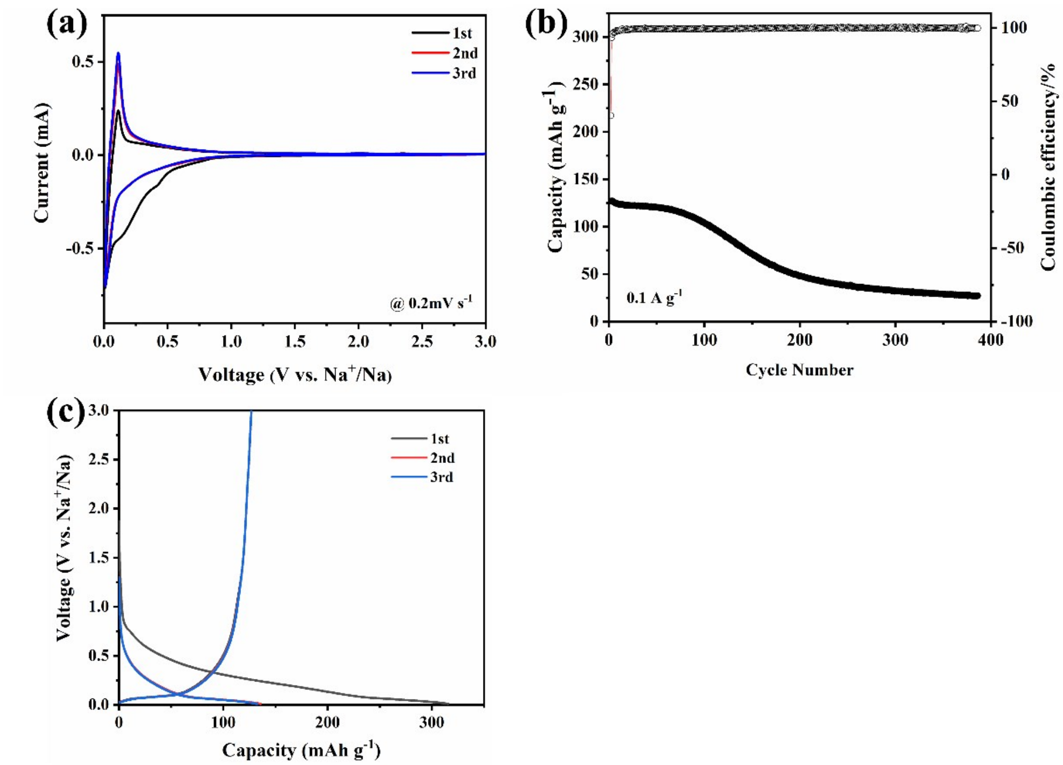

Figure S7 Sodium storage behaviors of CPAN. (a) First third consecutive CVs of the electrode made from CPAN in a voltage range of 0.01–3.0 V versus Na^+/Na at a scan rate of 0.2 mV s^{-1}. (b) Cycling performance of CPAN electrodes at 0.1 A g^{-1}. (c) First third Charge/discharge curves at a current density of 0.1 A g^{-1}.

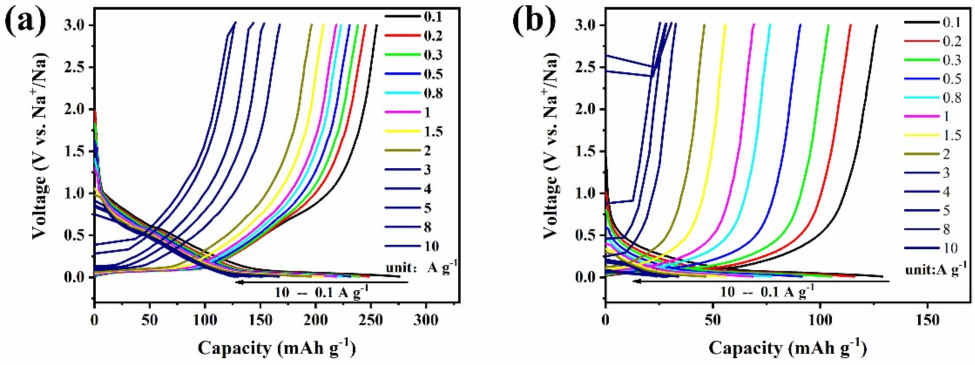

Figure S8 Charge/discharge profiles of (a) SSHC and (b) CPAN at different rates.

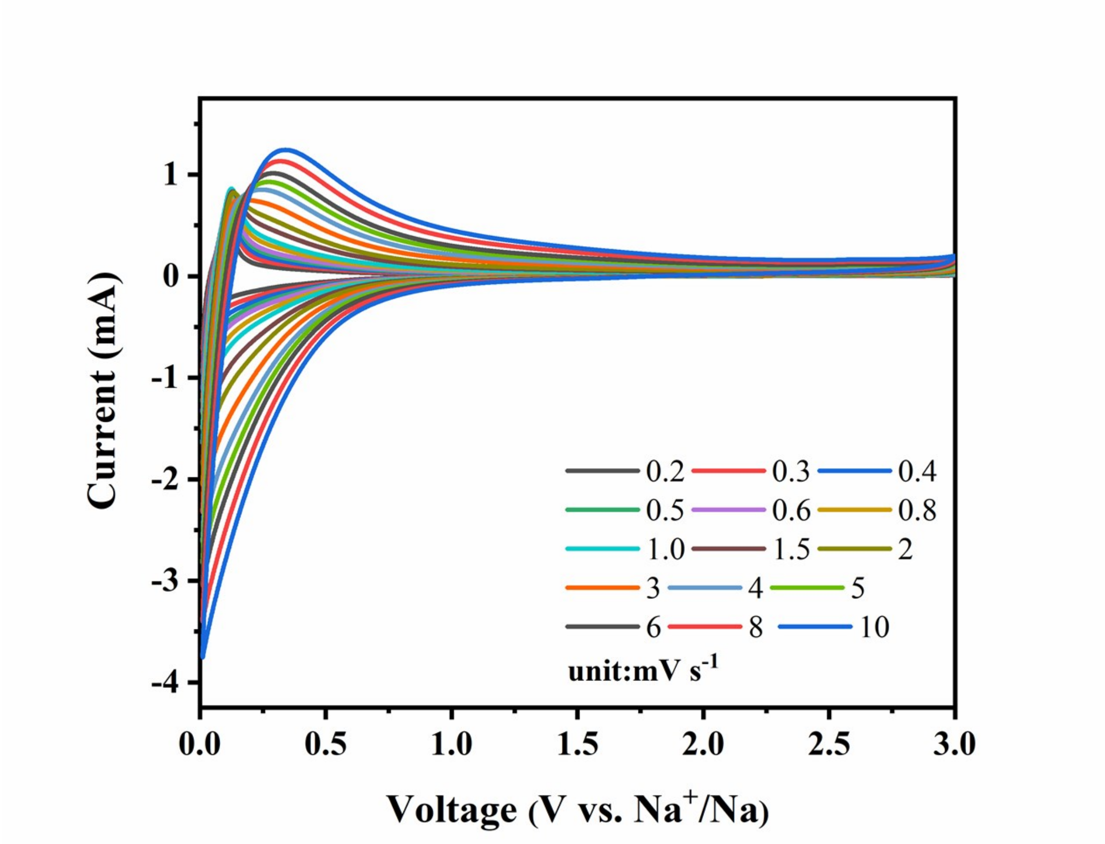

Figure S9 (a) CV curves of CPAN at the various scan rates from 0.2 to 10 mV s^{-1}.

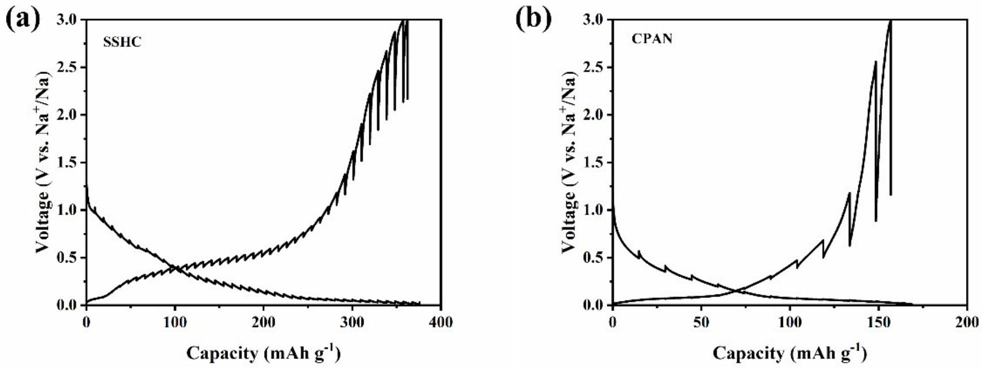

Figure S10 Study of the Na-ion diffusion coefficient of the SSHC and CPAN electrodes and the GITT profiles of the discharge/charge process.

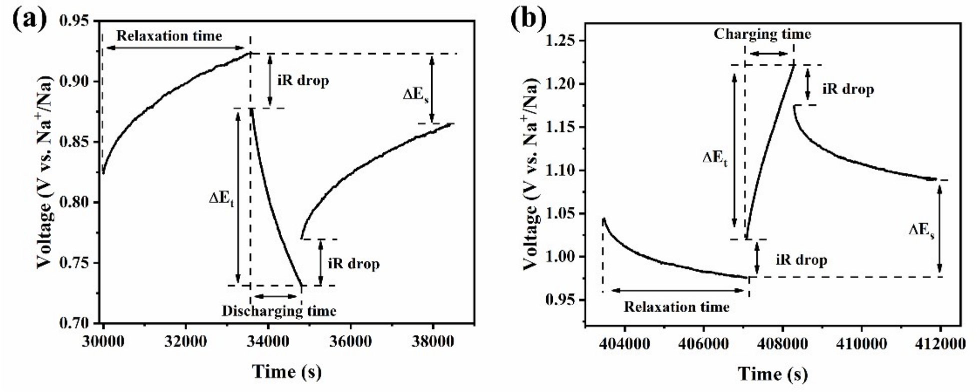

Figure S11 (a-b) Schematic of the calculation of diffusion coefficient using the GITT technique.

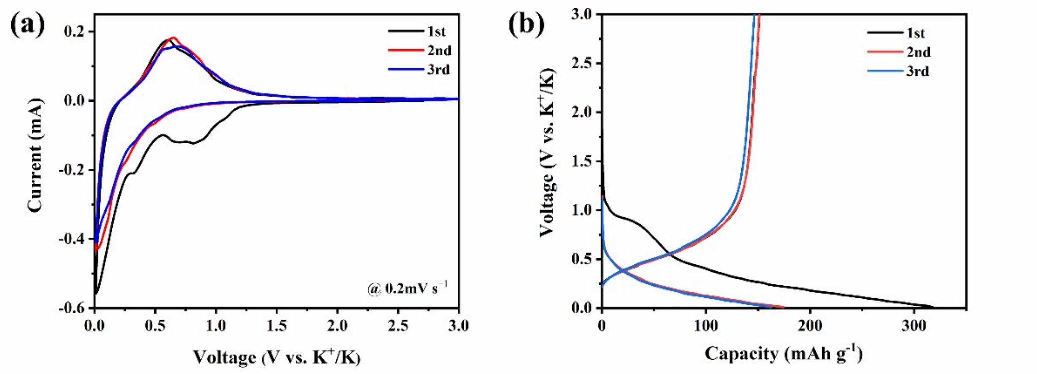

Figure S12 Electrochemical evaluation of the CPAN electrode for use in KIBs: (a) the first third CV curves of the electrode of SSHC in a voltage range of 0.01–3.0 V vs. K^+/K at a scan rate of 0.2 mV s^{-1}; (b) charge/discharge curves at a current density of 0.1 A g^{-1}.

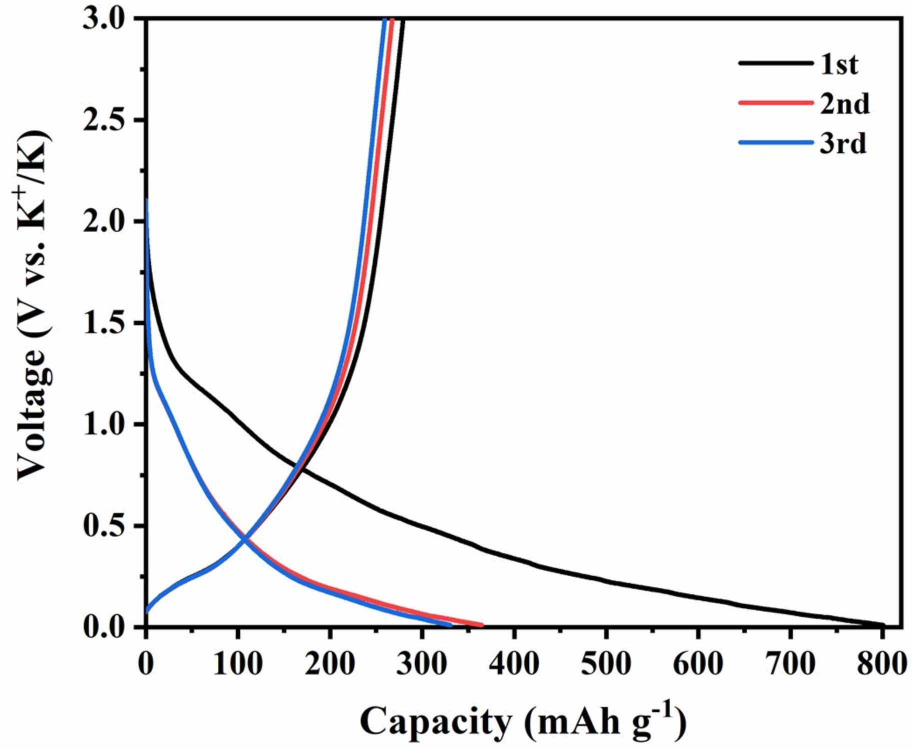

Figure S13 Electrochemical evaluation of the SSHC electrode in KIBs: Charge/discharge curves at a current density of 0.1 A g^{-1}.

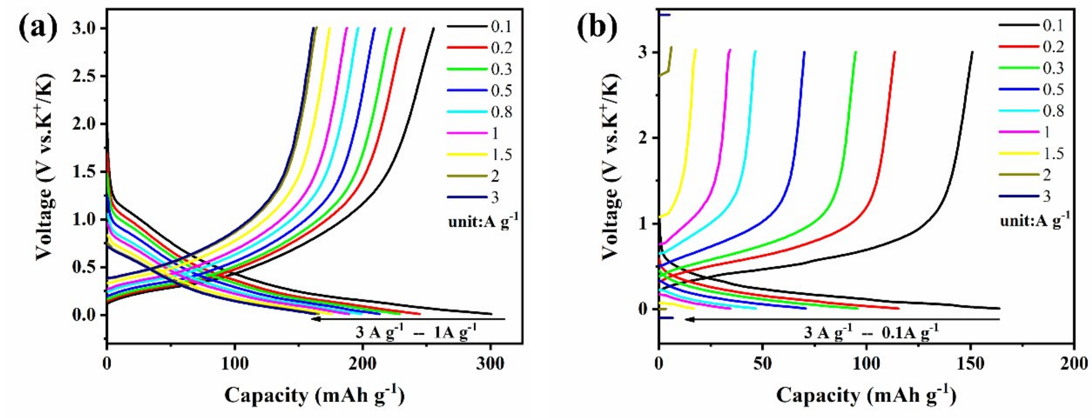

Figure S14 Charge/discharge profiles of (a) SSHC and (b) CPAN at different current densities.

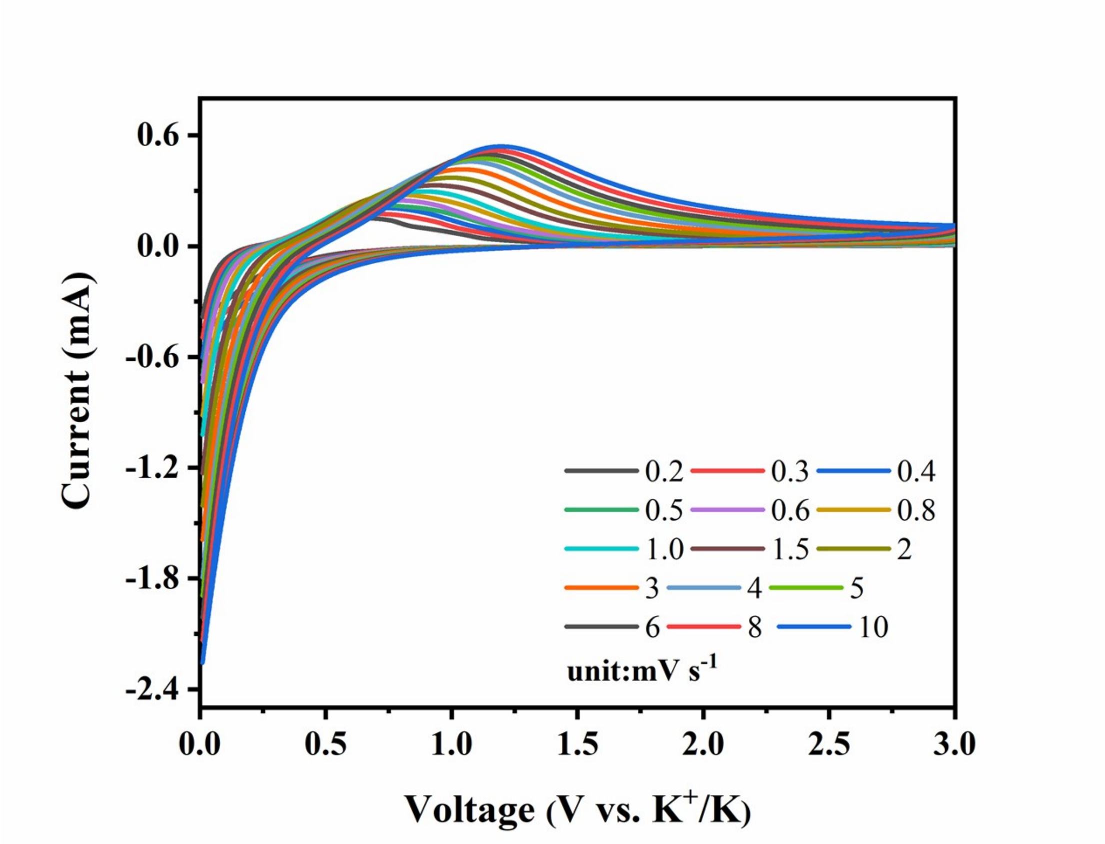

Figure S15 CV curves of CPAN in 1 M KPF_6 in ethylene carbonate and diethyl carbonate (EC: DEC = 1:1 v/v) electrolyte at the various scan rates from 0.2 to 10 mV s^{-1}.

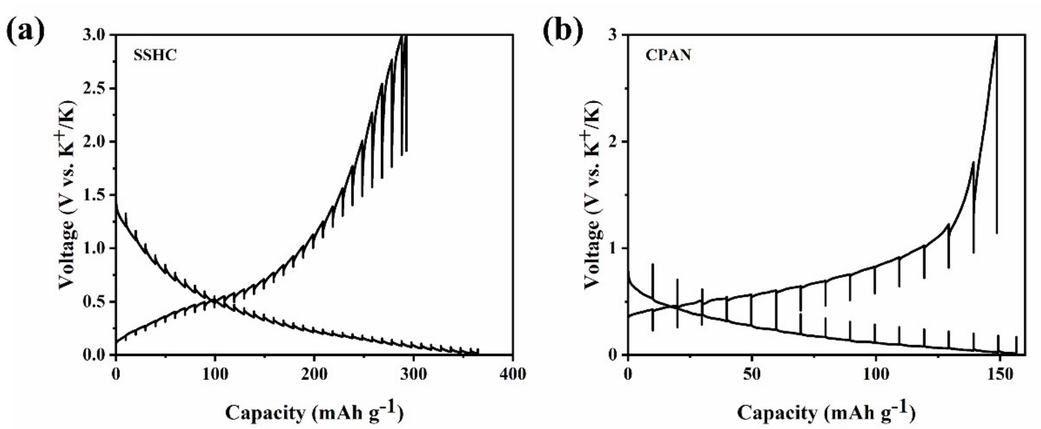

Figure S16 Study of the K-ion diffusion coefficient of the SSHC and CPAN electrodes and the GITT profiles of the discharge/charge process.

References

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2 T. Brezesinski, J. Wang, S. H. Tolbert and B. Dunn, *Nat. Mater.*, 2010, **9**, 146.

3 Y. Liu, J. S. Xue, T. Zheng and J. R. Dahn, *Carbon*, 1996, **34**, 193-200.