One-Step Regulation of Pore Evolution in Hard Carbon from Open to Closed for High Rate and High Plateau Capacity Sodium-Ion Storage

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Correlation formula

d_{002} is calculated from calculated by the Bragg's equation:

$$d_{(002)}(nm) = \frac{\lambda}{2sin\,\theta} \tag{S1}$$

The SAXS fitted model is as follows:

$$I(Q) = \frac{A}{Q^4} + \frac{Ba_1^4}{(1 + a_1^2Q^2)^2} + D \tag{S2}$$

where A and B are proportional to the total surface areas of the large and small pores, respectively. Q is the scattering vector and D is a constant background term. The diameters of closed nanopores can be obtained from a1 value ($R=2*a_1*10^{1/2}$).

Calculation of apparent Na^+ diffusion coefficients through GITT results

The calculation formula is as follows.

$$D_{Na^+} = \frac{4}{\pi\tau}\left(\frac{m_BV_m}{M_BS}\right)^2\left(\frac{\Delta E_S}{\Delta E_\tau}\right)^2 \tag{S3}$$

Here, τ, m_B, M_B, V_m and S are the pulse duration, the mass of the electrode active material, molar mass, molar volume and effective contact area, respectively. In addition, ΔE_s is the voltage change caused by the pulse, and ΔE_t is the voltage change in a constant current charging (discharging) process.

The *b* value can be estimated based on the formula:

$$i = av^b \tag{S4}$$

Here i is the peak current, ν is the scan rate, a and b are constants.

Quantitative the contribution of the capacitive-controlled and diffusion-controlled process by the formula:

$$i(V) = k_1v + k_2v^{1/2} \tag{S5}$$

Here, k_1v and $k_2v^{1/2}$ correspond to the capacitive-controlled and diffusion-controlled capacity contribution, respectively.

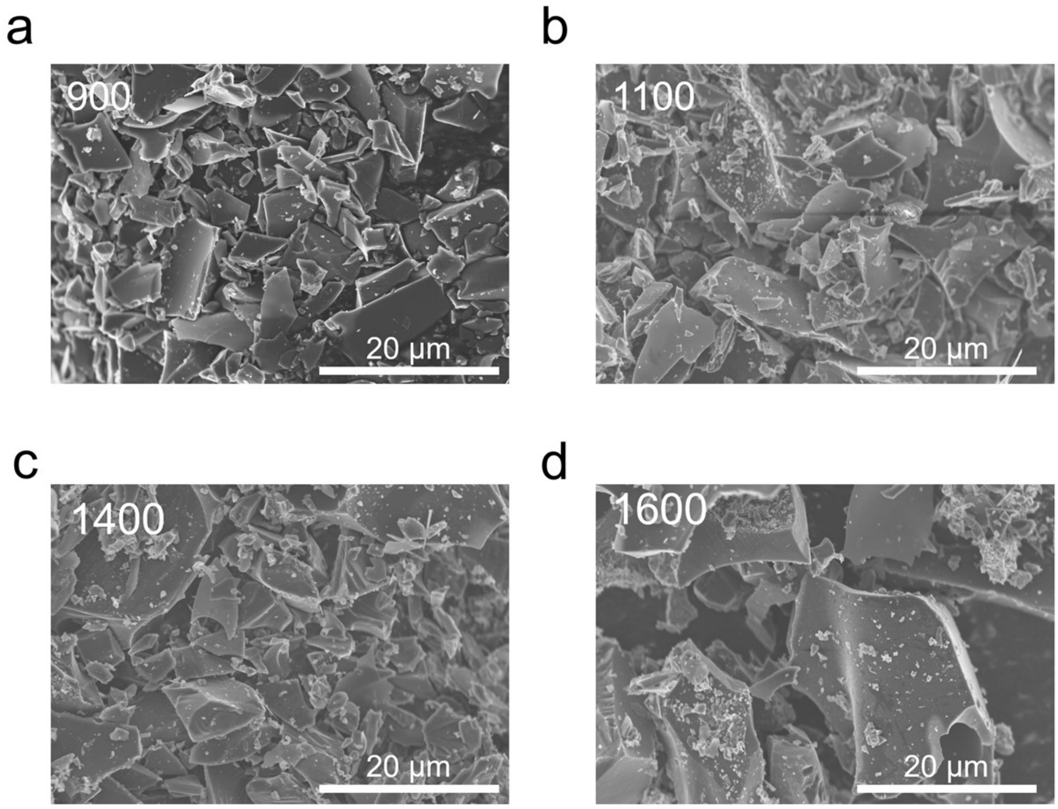

Figure S1 SEM image of the (a) 900, (b) 1100, (b) 1400, and (d) 1600 materials.

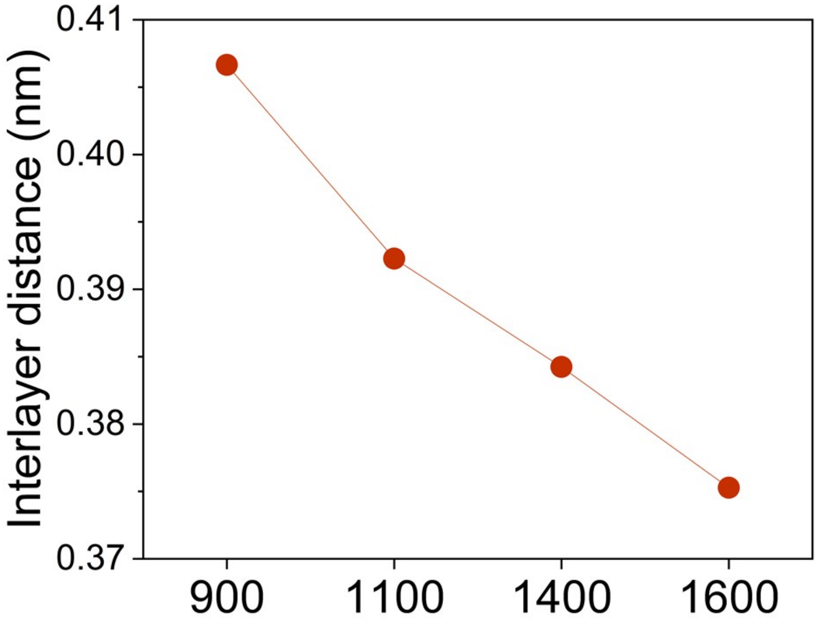

Figure S2 d_{002} of samples.

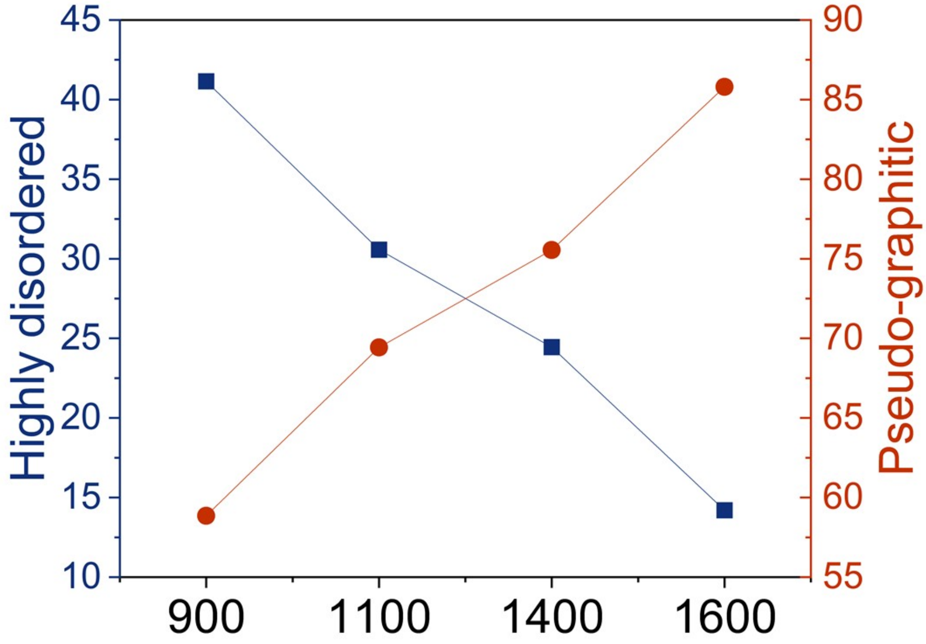

Figure S3 The area percentages of the two regions after fitting.

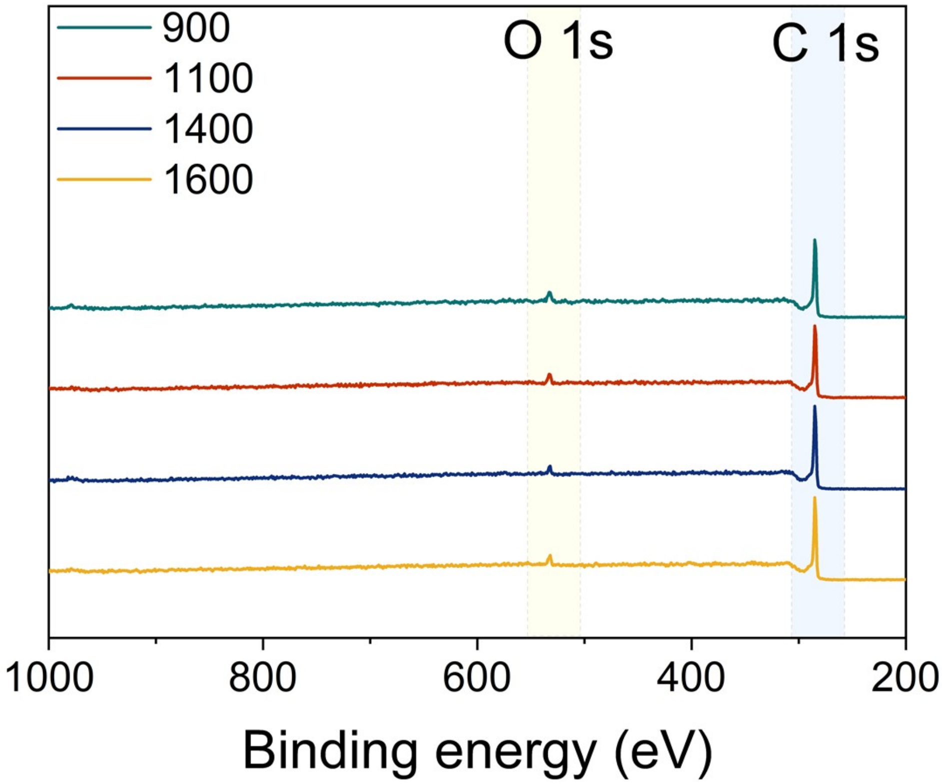

Figure S4 XPS whole spectra of the 900, 1100, 1400, and 1600 materials.

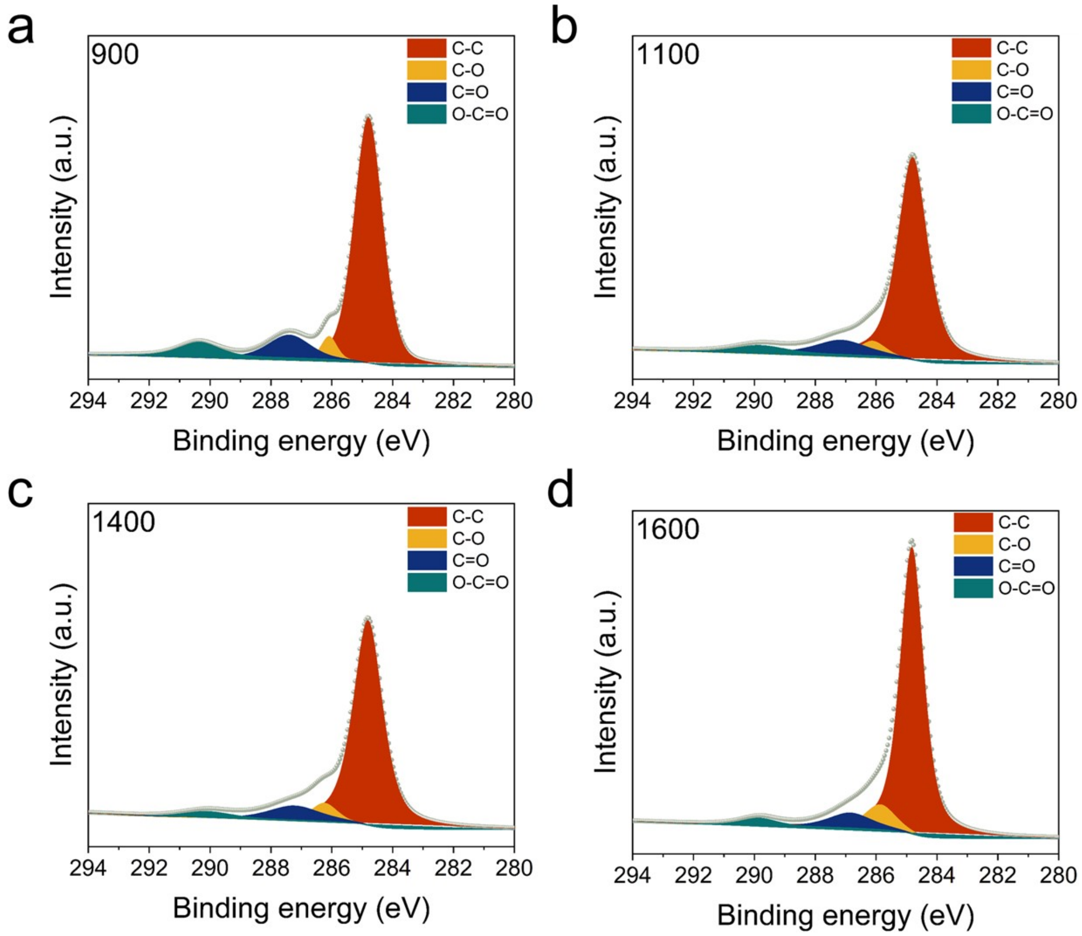

Figure S5 C 1s XPS high-resolution spectra of samples.

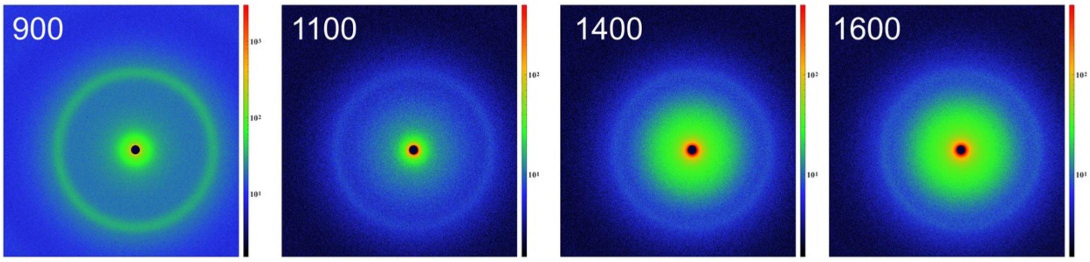

Figure S6 2D SAXS profiles of samples.

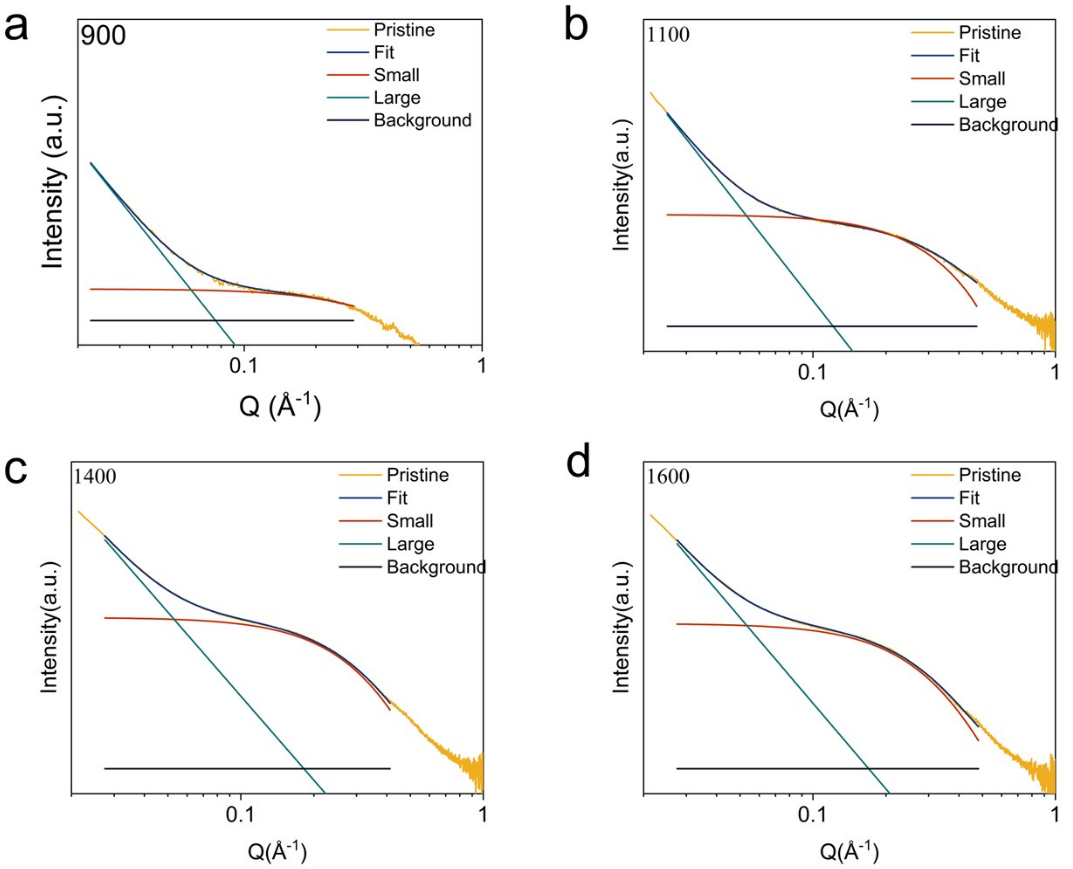

Figure S7 Fitted SAXS patterns of (a) 900, (b) 1100, (b) 1400, and (d) 1600 samples.

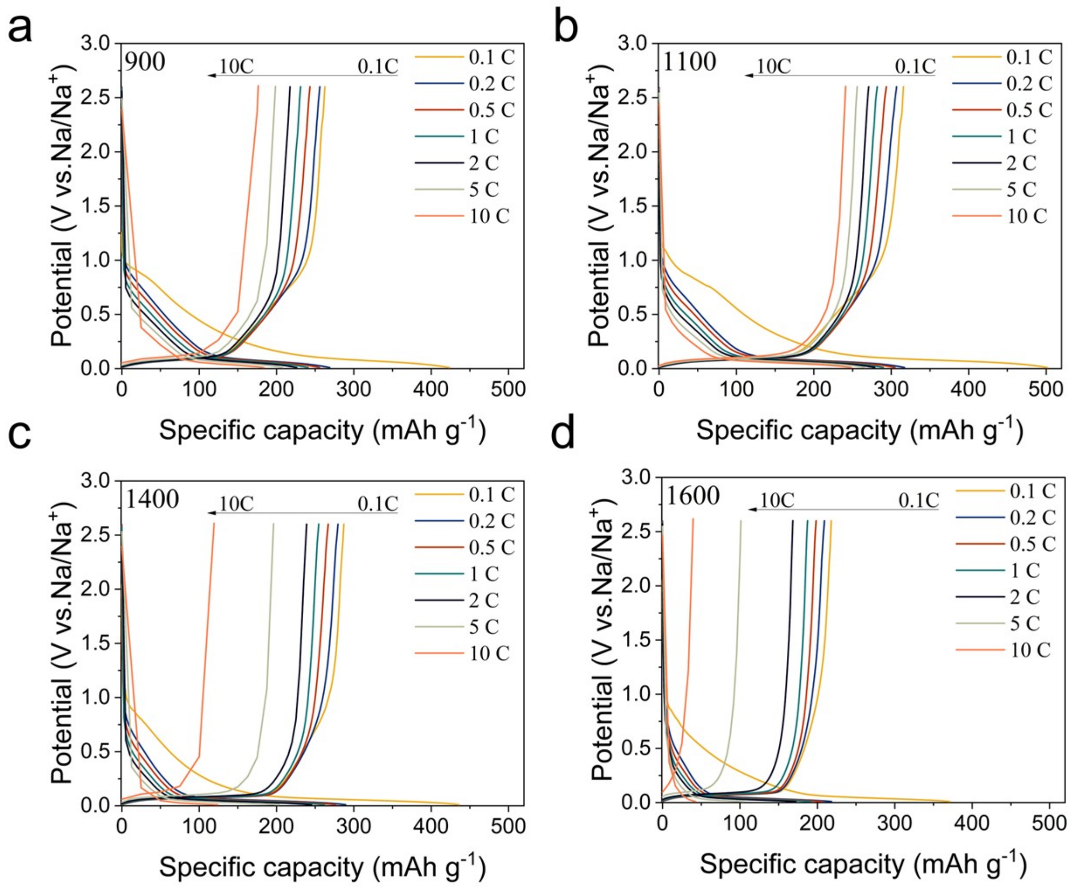

Figure S8 GCD curves at different current density of (a) 900, (b) 1100, (b) 1400, and (d) 1600 electrodes.

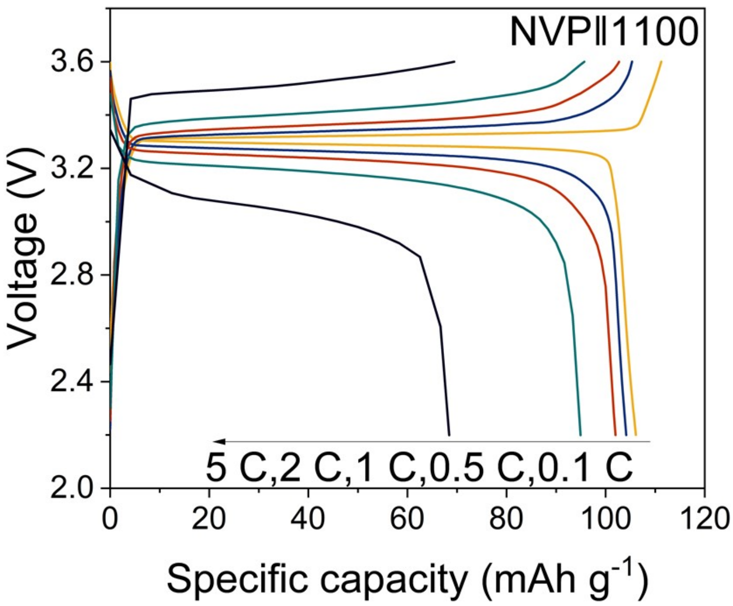

Figure S9 GCD curve of NVP‖1100 full cell at different rate.

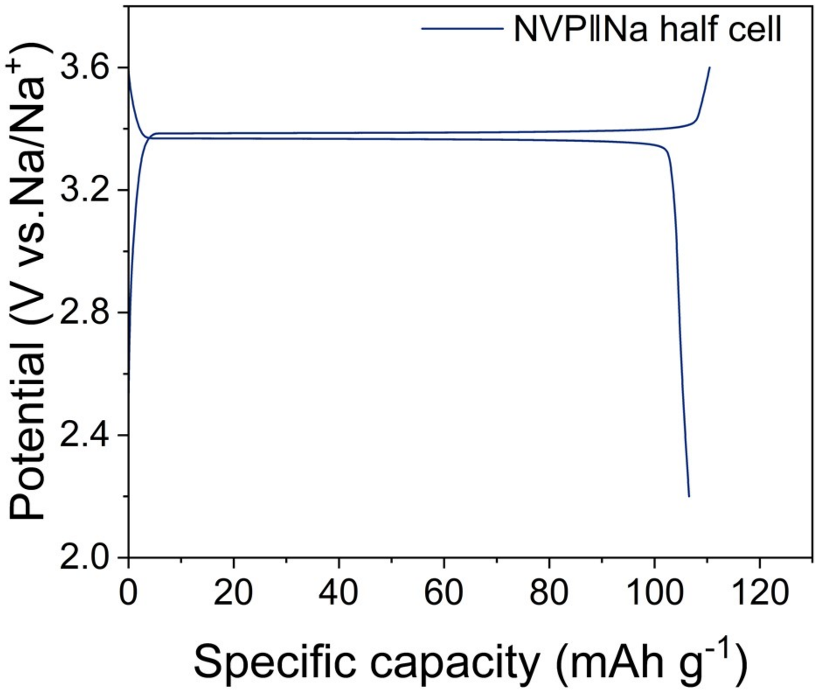

Figure S10 First GCD curve of NVP‖Na half cell.

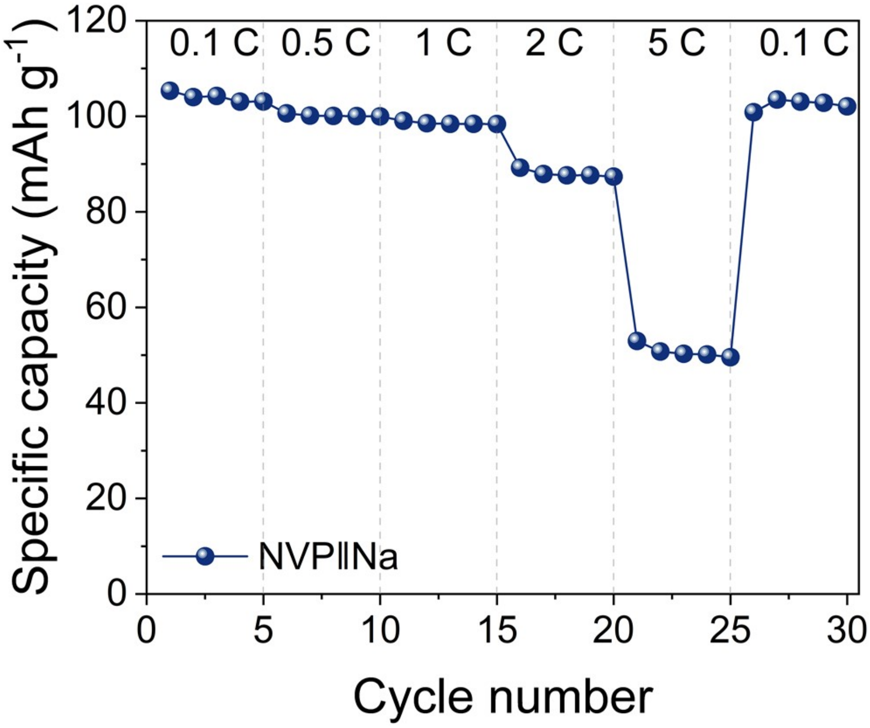

Figure S11 The rate performance of the NVP‖Na half cell.

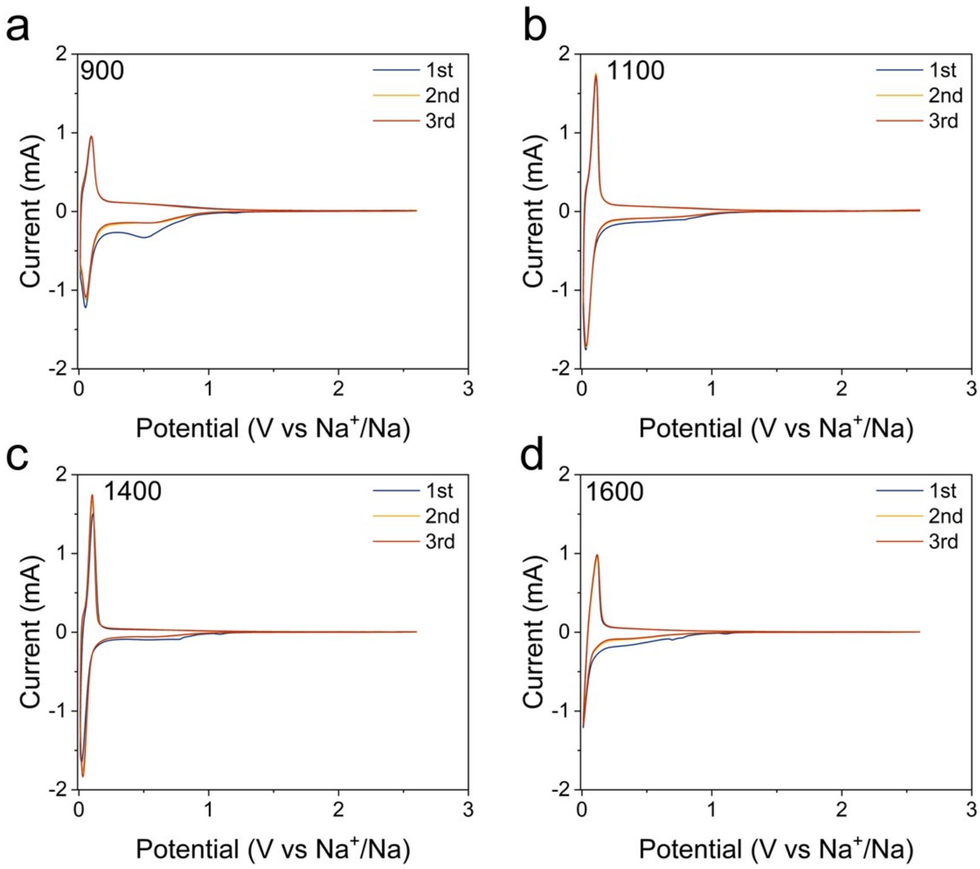

Figure S12 CV curves under 0.1 mV s^{-1} for (a) 900, (b) 1100, (b) 1400, and (d) 1600 anodes.

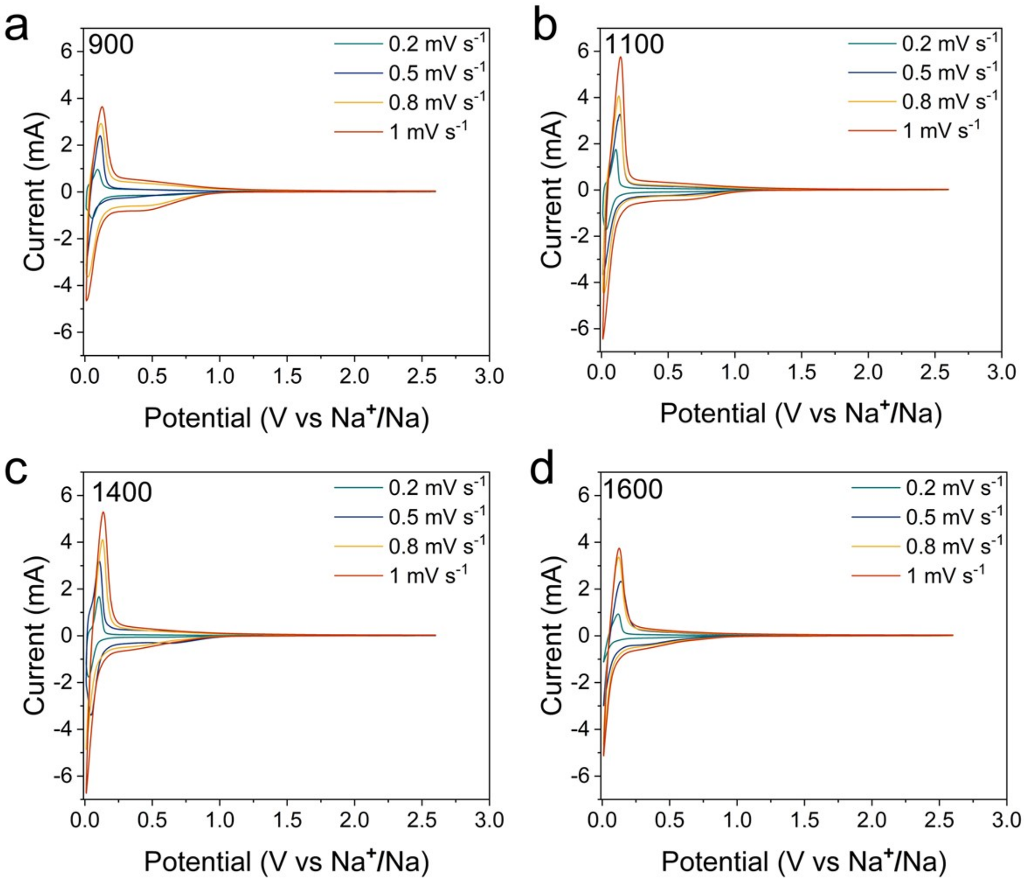

Figure S13 CV curves under different scan rates from 0.1 to 1.0 mV s^{-1} of (a) 900, (b) 1100, (b) 1400, and (d) 1600 anodes.

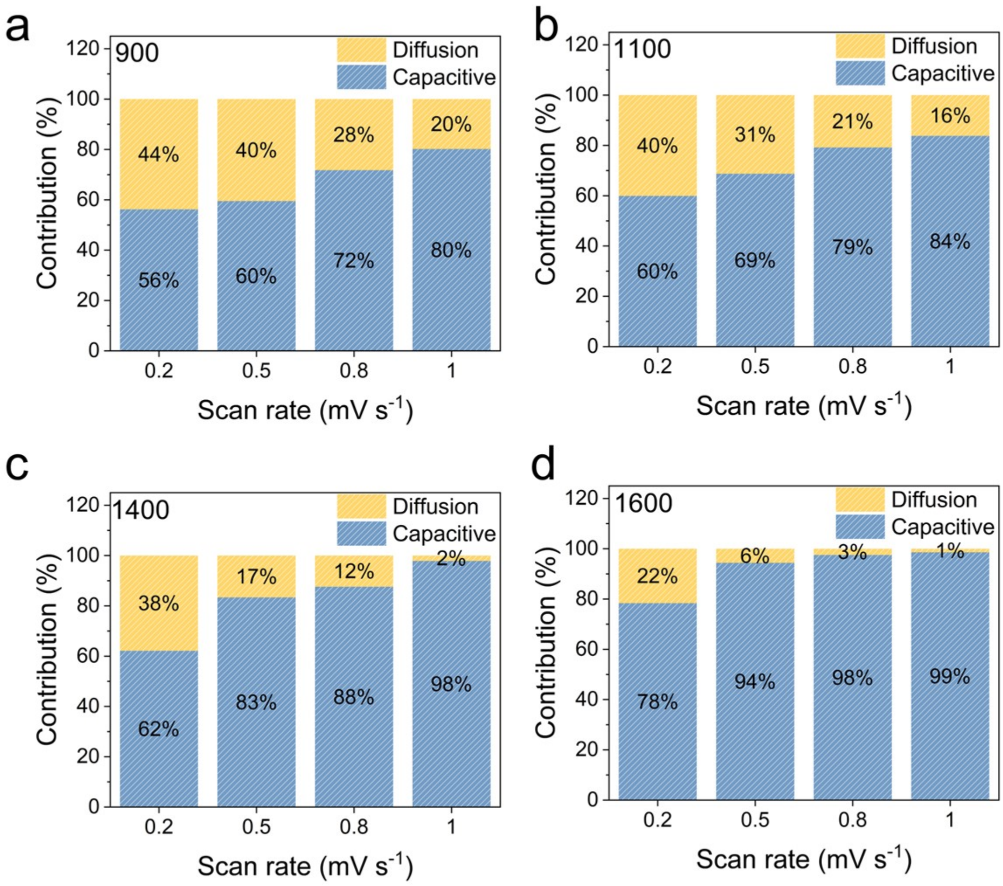

Figure S14 Capacitive capacity contribution at different scan rates of (a) 900, (b) 1100, (b) 1400, and (d) 1600 anodes.

Sample	2θ [°]	d_{002}(nm)	Highly disordered (2θ [°]=21.12)	Pseudo-graphitic (2θ [°]=23.98)
900	21.83	0.4067	41.15%	58.85%
1100	22.64	0.3923	30.57%	69.43%
1400	23.12	0.3842	24.44%	75.56%
1600	23.68	0.3753	14.20%	85.80%

Table S1 Physical parameters of different samples from the XRD spectra.

Sample	N_2-BET surface area (m^2 g^{-1})	N_2-Total pore volume (cm^3 g^{-1})	CO_2-BET surface area (m^2 g^{-1})	CO_2-Total pore volume (cm^3 g^{-1})
900	556.91	0.22	502.67	0.237
1100	8.13	0.024	465.41	0.211
1400	6.82	0.0049	110.94	0.0851
1600	1.23	0.0016	88.35	0.0582

Table S2 Physical parameters of different samples from the XRD spectra.

Table S3 C, O percentage from XPS analysis

Sample	C (%)	O (%)
900	93.95	6.05
1100	94.38	5.62
1400	94.73	5.27
1600	95.75	4.25

Table S4 The proportion of C 1s element in the XPS fitting curve in different materials.

Sample	C-C	C-O	C=O	O-C=O
900	78.25	3.97	10.59	7.19
1100	79.61	4.73	10.22	5.44
1400	79.67	5.68	9.90	4.75
1600	80.40	8.33	7.85	3.42

Table S5 The proportion of O 1s element in the XPS fitting curve in different materials.

Sample	C-O	C=O
900	9.84	90.16
1100	10.54	89.46
1400	19.66	80.34
1600	33.82	66.18

Table S6 The proportion of fitting peak in the Raman fitting curve in samples.

Sample	A_{D1}/A_G	A_{D3}/A_G
900	3.247	2.14
1100	2.14	1.54
1400	2.00	0.94889
1600	1.798	0.899

Table S7 The pore structure of samples.

Sample	R (nm) Average diameter obtained from SAXS fitting
900	0.668
1100	2.307
1400	3.196

1600	3.409