

## Supplementary Material

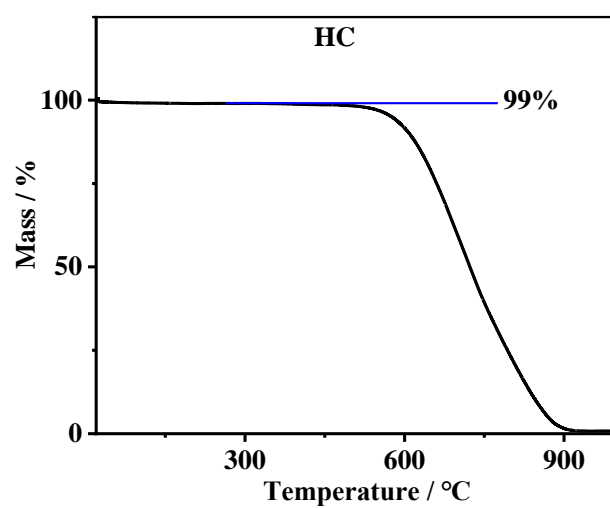
### Tailoring hard carbon interfaces in carbonate-based electrolytes for sodium-ion hybrid capacitors

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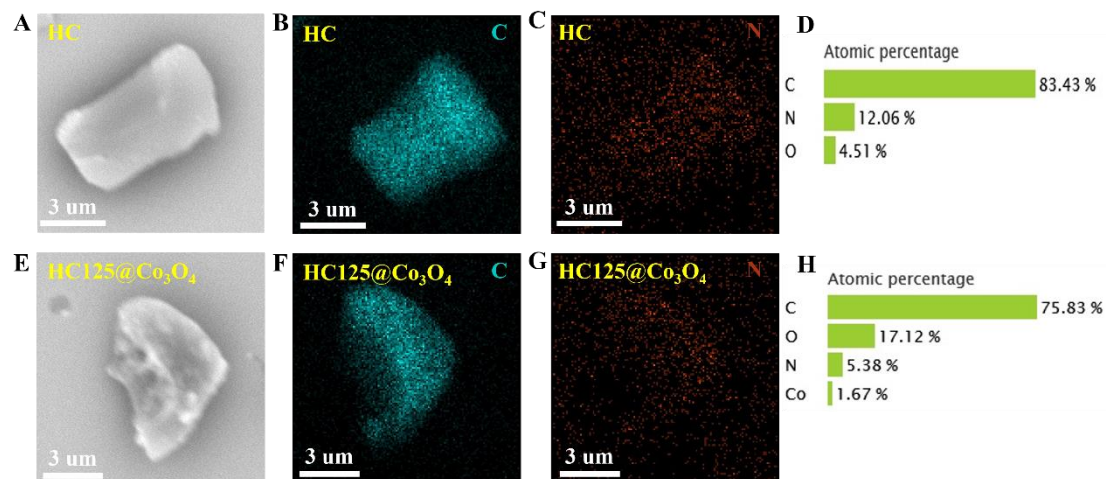
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**Figure S1.** TG of the HC.



**Figure S2.** SEM, corresponding mapping of C and N element, and atomic percentage of HC (A-D) and HC125@Co<sub>3</sub>O<sub>4</sub> (E-H).

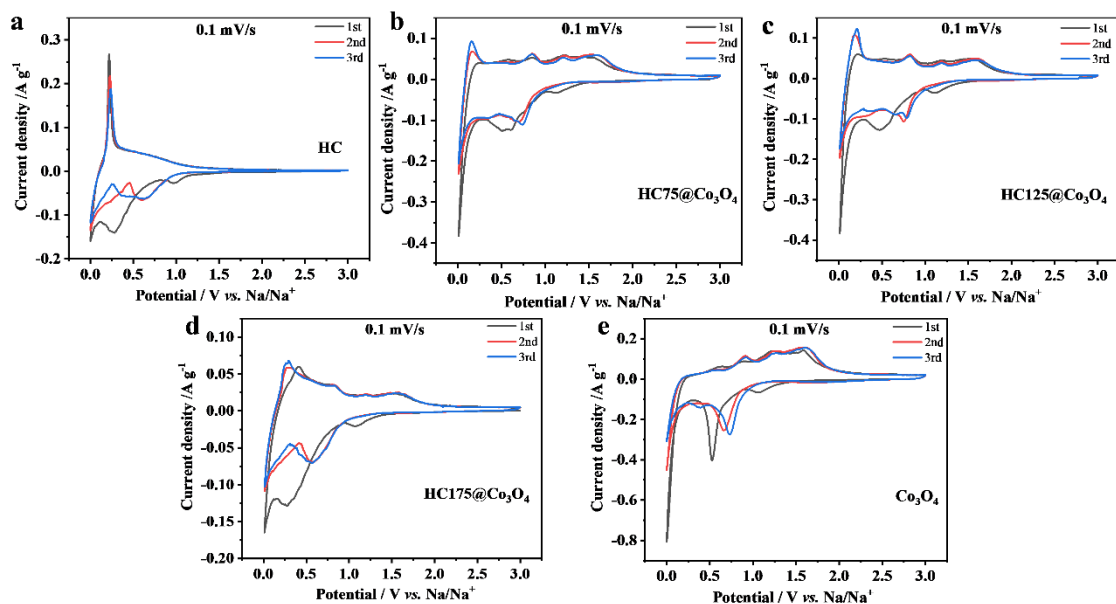


Figure S3 The initial three CV plots at  $0.1 \text{ mV s}^{-1}$  for (a) HC, (b)  $\text{HC75@Co}_3\text{O}_4$ , (c)  $\text{HC125@Co}_3\text{O}_4$ , (d)  $\text{HC175@Co}_3\text{O}_4$ , and (e)  $\text{Co}_3\text{O}_4$ .

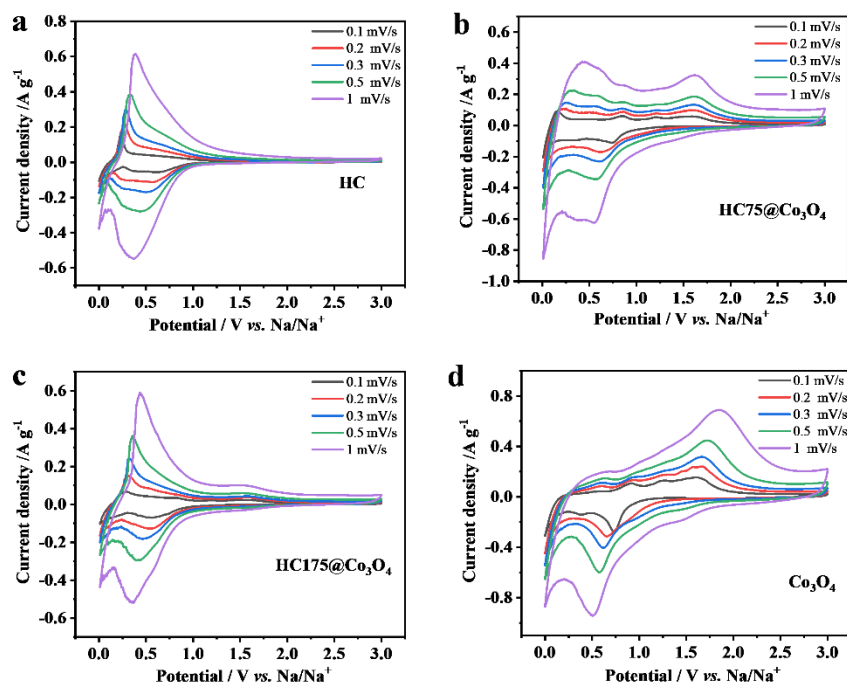
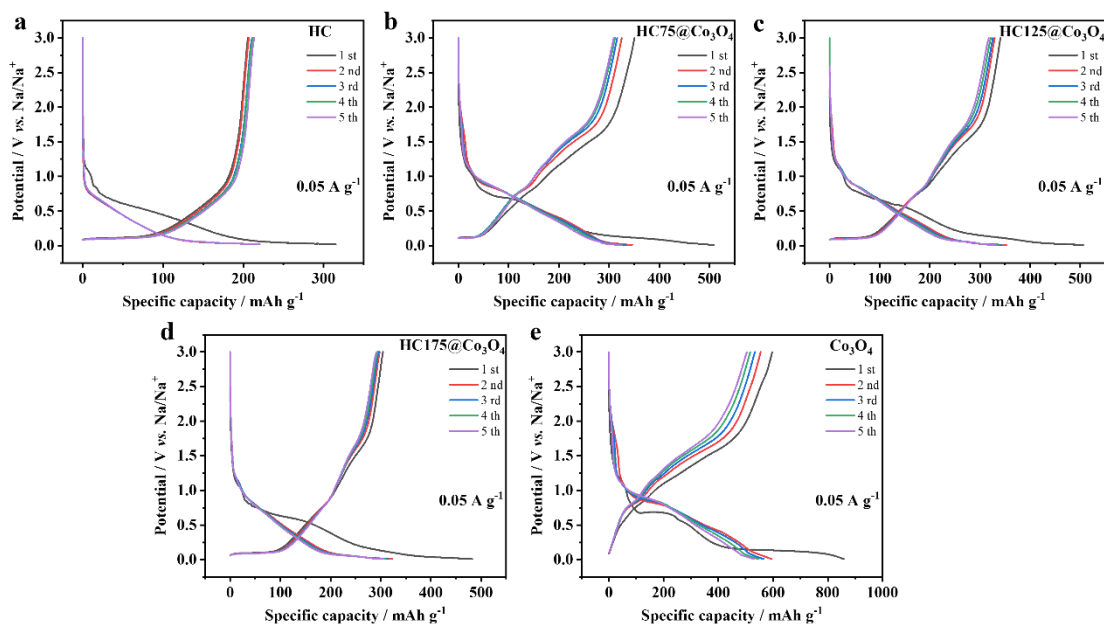
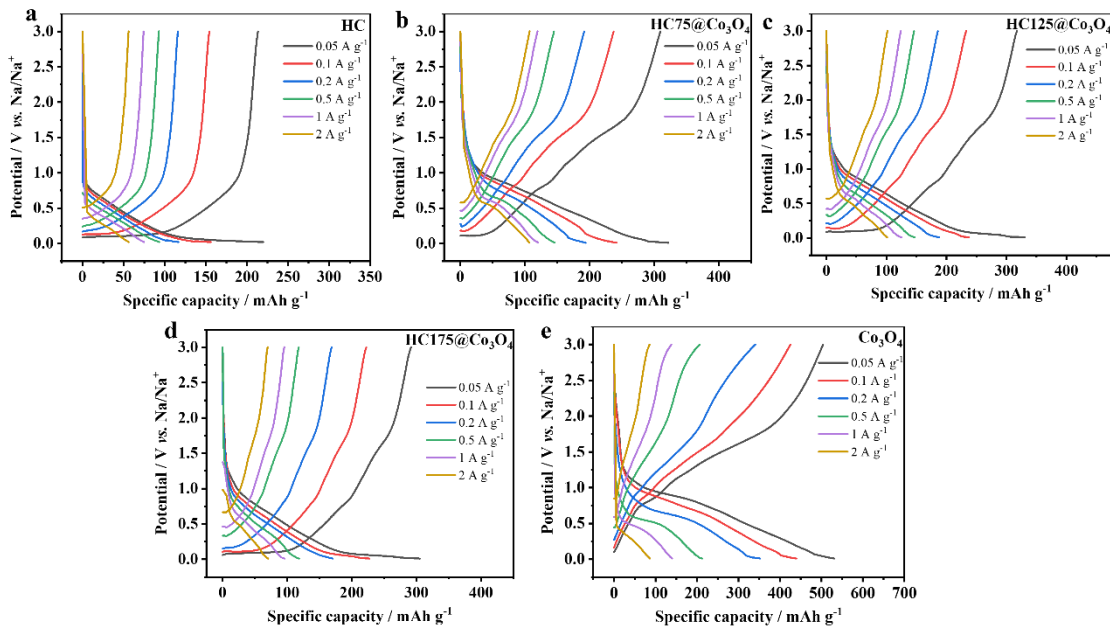


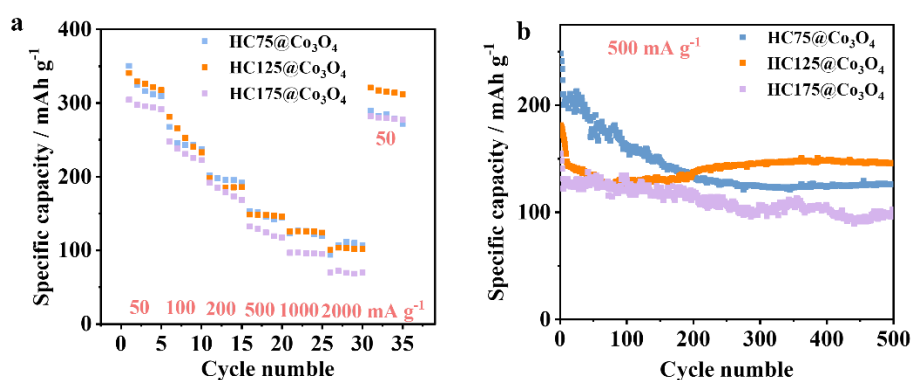
Figure S4 The CV plots at  $0.1\text{-}1 \text{ mV s}^{-1}$  for (a) HC, (b)  $\text{HC75@Co}_3\text{O}_4$ , (c)  $\text{HC175@Co}_3\text{O}_4$ , and (d)  $\text{Co}_3\text{O}_4$ .



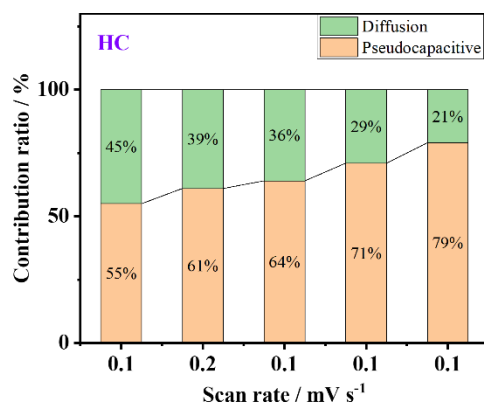
**Figure S5** GCD curves of (a) HC, (b) HC75@Co<sub>3</sub>O<sub>4</sub>, (c) HC125@Co<sub>3</sub>O<sub>4</sub>, (d) HC175@Co<sub>3</sub>O<sub>4</sub>, and (e) Co<sub>3</sub>O<sub>4</sub> electrodes for the first five cycles at 0.05 A g<sup>-1</sup>.



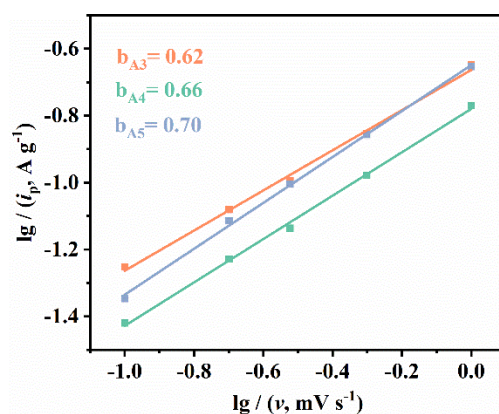
**Figure S6** GCD curves of (a) HC, (b) HC75@Co<sub>3</sub>O<sub>4</sub>, (c) HC125@Co<sub>3</sub>O<sub>4</sub>, (d) HC175@Co<sub>3</sub>O<sub>4</sub>, and (e) Co<sub>3</sub>O<sub>4</sub> electrodes for the respective 5th cycle at 0.05-2 A g<sup>-1</sup>.



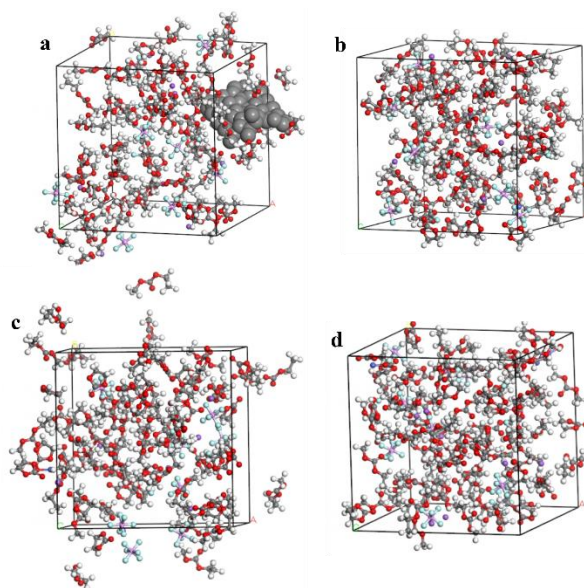
**Figure S7** The rate capability (a) and cycling performance (b) of the HC75@Co<sub>3</sub>O<sub>4</sub>, HC125@Co<sub>3</sub>O<sub>4</sub> and HC175@Co<sub>3</sub>O<sub>4</sub>.



**Figure S8** Pseudocapacitive contribution to the total current contribution at 0.1–1.0 mV s<sup>-1</sup> of HC electrodes.



**Figure S9** The relationship of  $\lg i$  vs.  $\lg v$  for A3, A4 and A5 peak in HC125@Co<sub>3</sub>O<sub>4</sub>.



**Figure S10** Snapshots of (a) Model A, (b) Model B, (c) Model C, and (d) Model D obtained by MD simulation.

**Table S1.** A comparison for the performance of the HC125@Co<sub>3</sub>O<sub>4</sub>//AC in this work with some representatively reported SICs.

SIC devices	Voltage (V)	Energy density (Wh/kg)	Power density (W/kg)	Capacitance retention/ cycles	Ref.
NHCNF-600//AC	1-4	82.1-23.4	39.9-7000	70%/4000	[S1]
EEG//AC	1-4.3	90-17	2000-17100	100%/700	[S2]
HAT550@ZTC//STC-16	1-4	61-44	100-3400	91.5%/1000	[S3]
Mo <sub>2</sub> C//AC	0-4.5	76.1-8	112-2241	83%/4000	[S4]
NHPC-800//AC/Na <sub>2</sub> C <sub>6</sub> O <sub>6</sub> - 40%	0-4	33.5	100	/	[S5]
FeSe-1000//AC	0.5-3.8	112-44.2	107.5-10750	76%/5000	[S6]
MSC//AC	0.01-4	109-49	98-5136	80%/2000	[S7]
KNZMF@//AC/KS6	0-5	126.69-14.45	730-12590	59.6%/1000	[S8]
<b>HC125@Co<sub>3</sub>O<sub>4</sub>//AC</b>	<b>0.01-3.8</b>	<b>129.5-26.5</b>	<b>583-11650</b>	<b>76%/1000</b>	<b>This work</b>

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