

Supplemental Information

**Insight into the structural evolution mechanism and potassium storage performance  
of expanded graphitic onion-like carbon**

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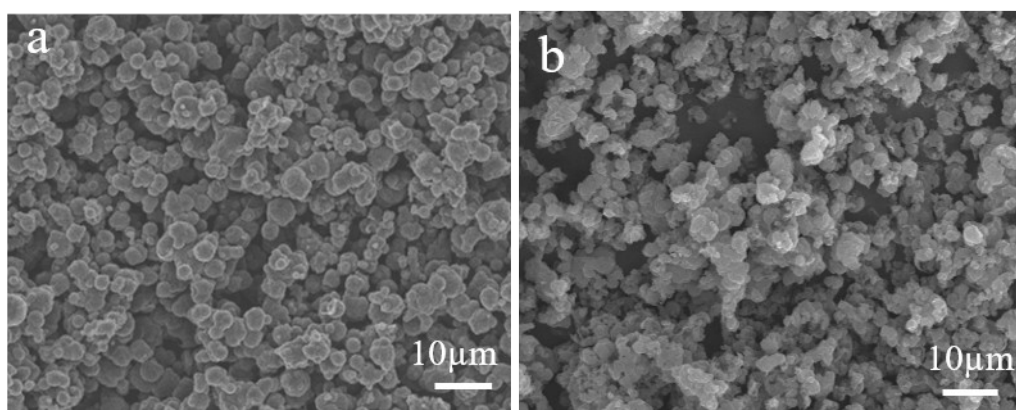


Fig. S1. SEM micrographs of (a) GOC and (b) O-GOC.

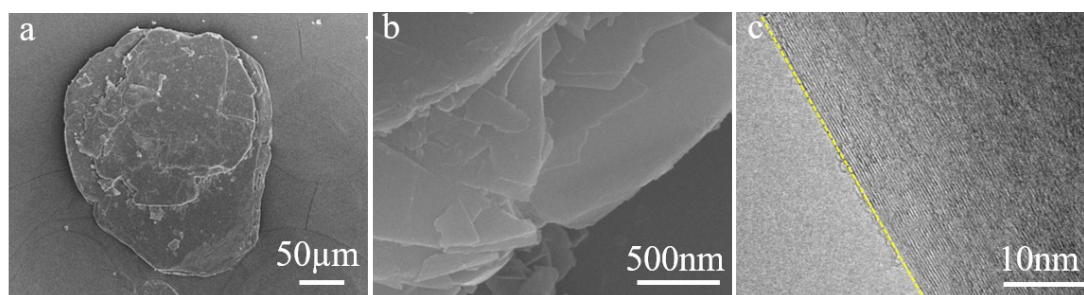


Fig. S2. (a, b) SEM images and (c) TEM image of NFG.

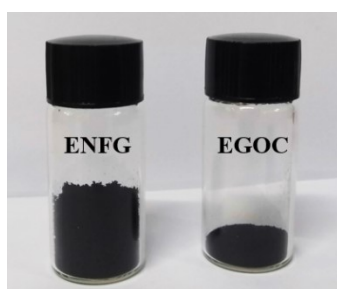


Fig. S3. Photograph of ENFG and EGOc with the same weight of 20 mg.

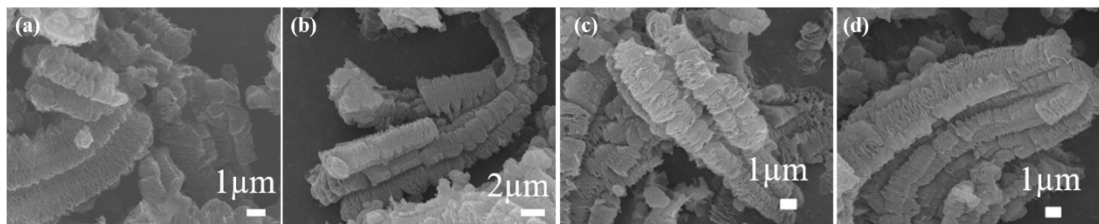


Fig. S4. Morphology and structure characterizations. SEM images of the (a) EGO C200, (b) EGO C300, (c) EGO C400 and (d) EGO C500.

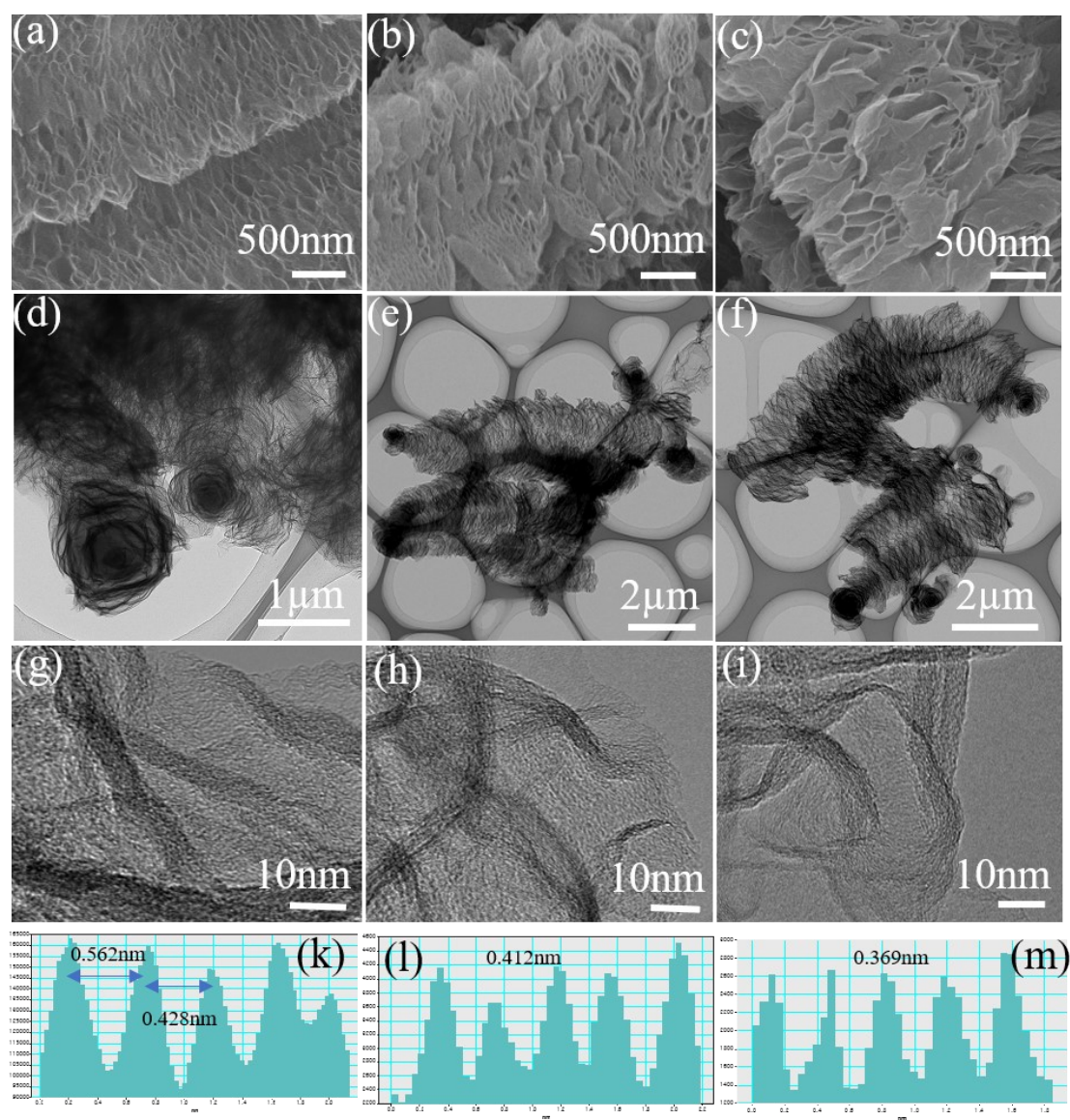


Fig. S5 (a-c) SEM images, (d-f) TEM images, (g-i) HRTEM images and (k-m) the calculated interlayer spacing of EGO C200, EGO C300 and EGO C500.

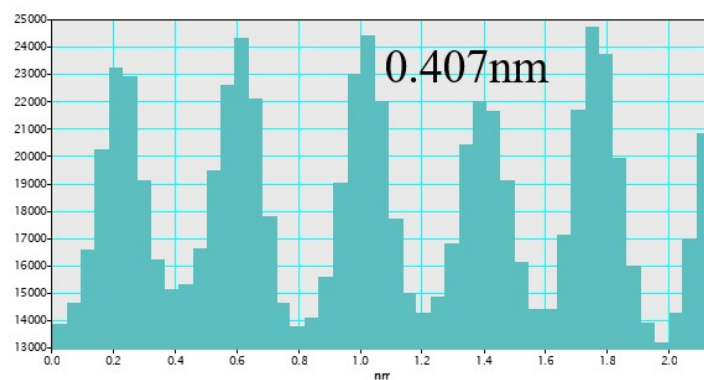


Figure. S6 The calculated interlayer spacing of Figure 2f.

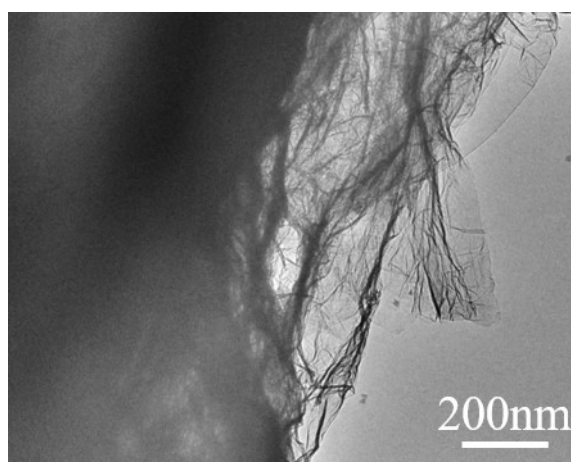


Fig. S7 TEM images of ENFG400 without ultrasound treatment.

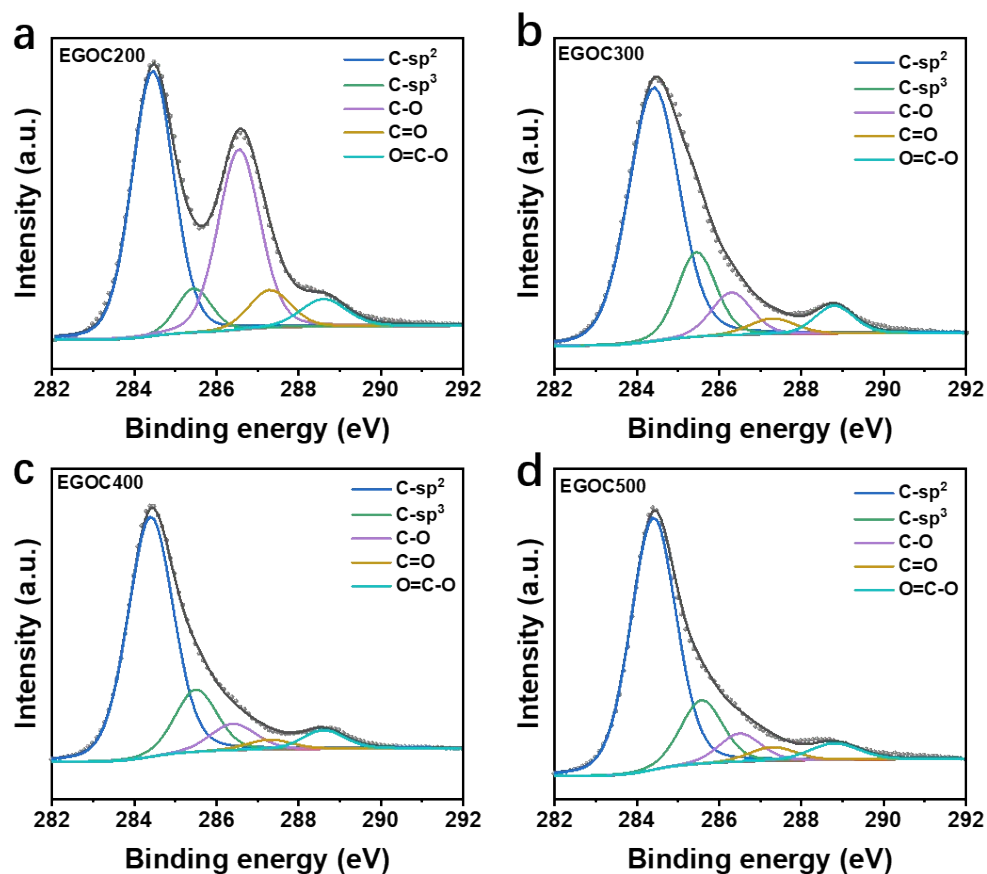


Fig. S8 High-resolution C1s spectra of (a) EGOX200, (b) EGOX300, (c) EGOX400 and (d) EGOX500.

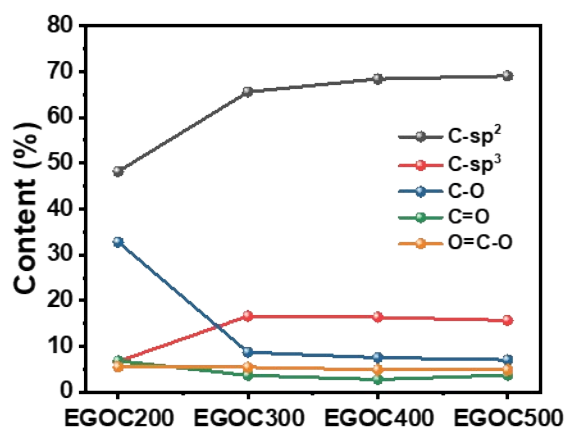


Fig. S9 Variation of the C-sp<sup>2</sup>, C-sp<sup>3</sup>, C-O, C=O and O=C-O values of EGOX200, EGOX300, EGOX400 and EGOX500.



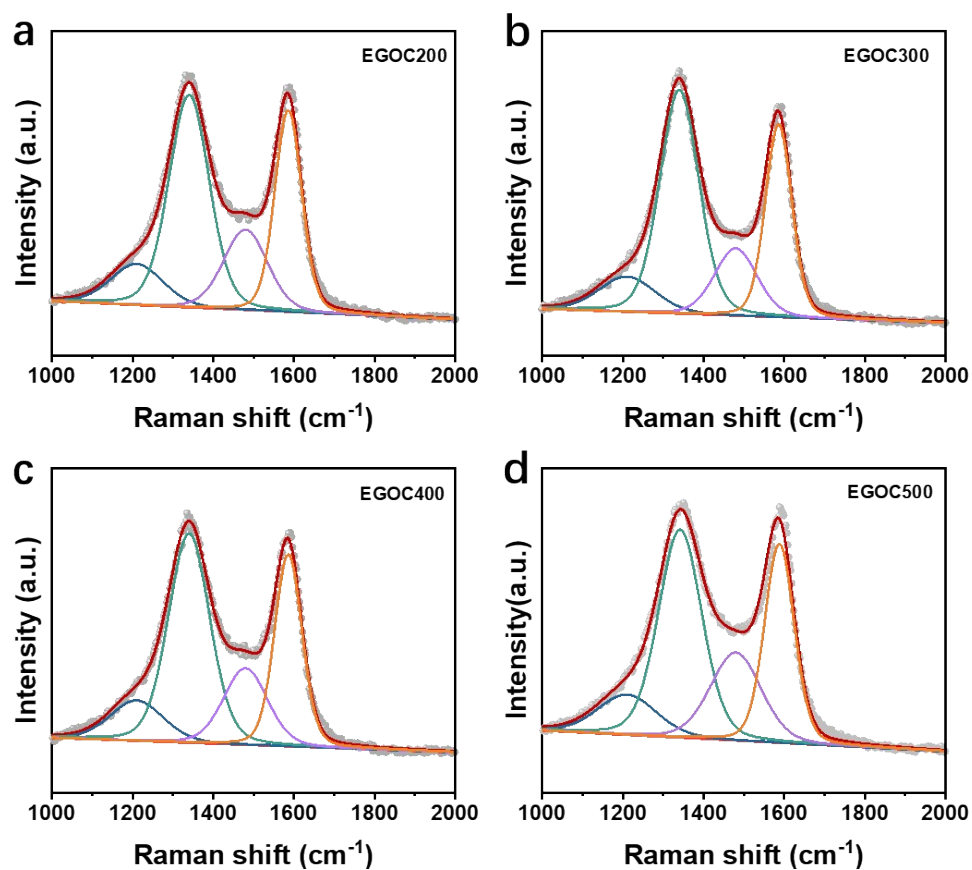


Fig. S10 Raman spectra of (a) EGOx200, (b) EGOx300, (c) EGOx400 and (d) EGOx500.

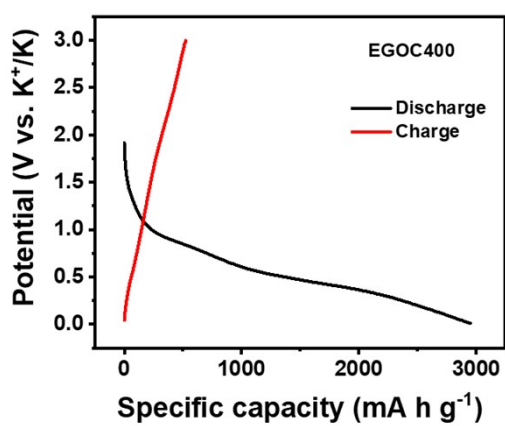


Fig. S11 Galvanostatic charge discharge curves for the first cycle of EGOx400.

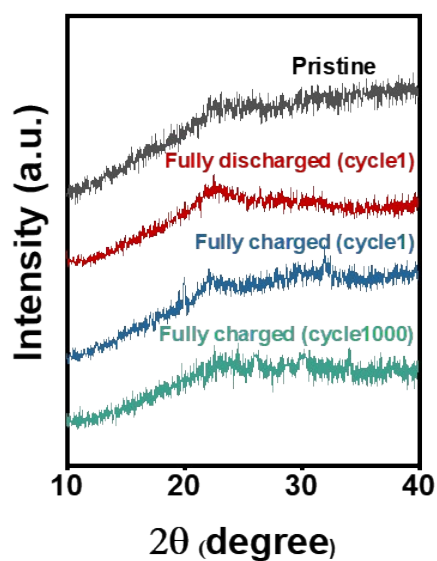


Fig. S12 The XRD of EGOc400 during potassiation/depotassiation process at different states: pristine, potassiated to 0.01 V in the first cycle, depotassiated to 3.0 V in the first cycle and fully charged after 1000 cycles.

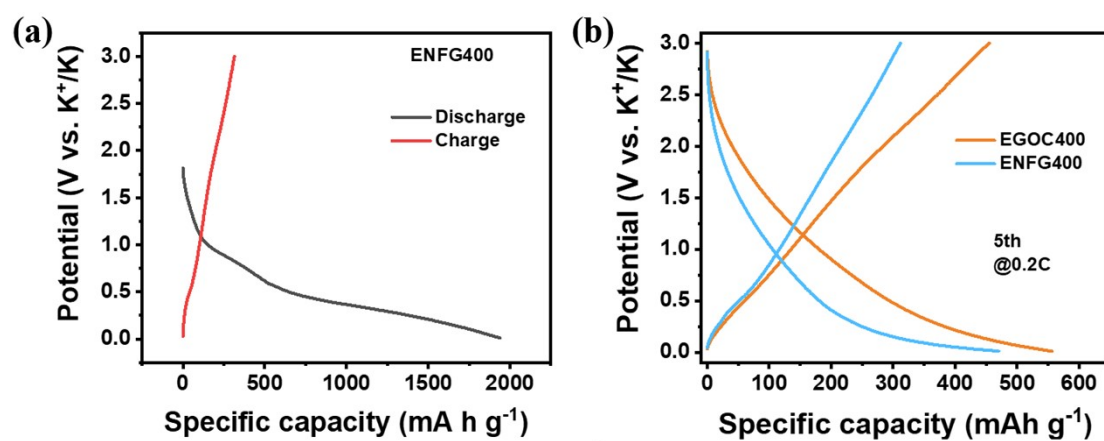


Fig. S13 Galvanostatic charge/discharge curves for (a) the first cycle of ENFG400, and (b) the fifth cycle of EGOc400 and ENFG400.

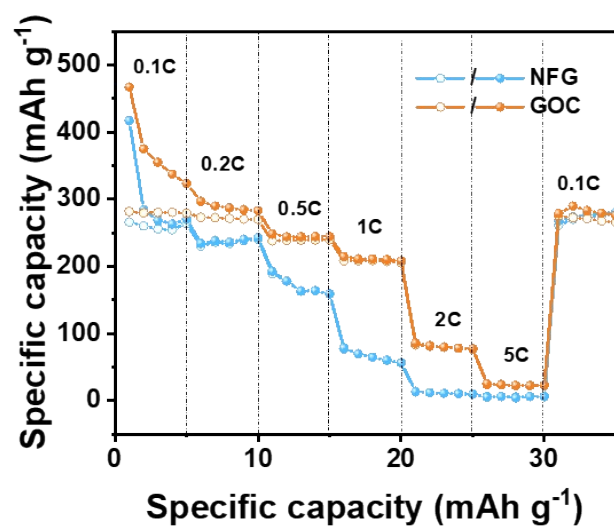


Fig. S14 The rate performance of EGOC400 and ENFG400.

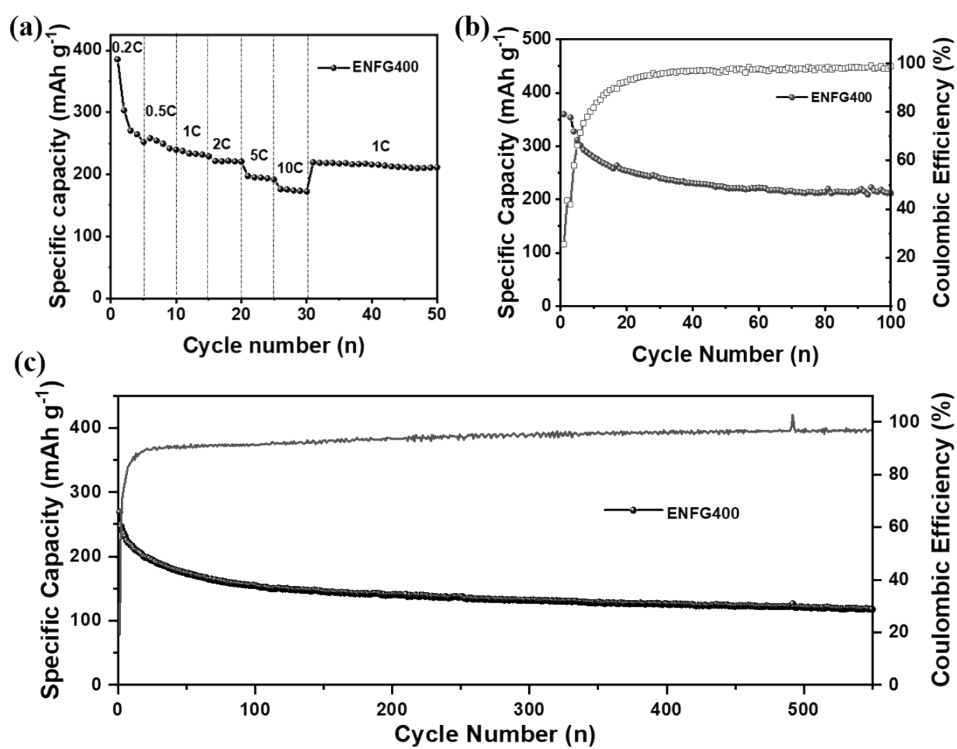


Fig. S15 (a) Rate performance of ENFG400. Cycling performance of ENFG400 at (b) 0.2C and (c) 2C.



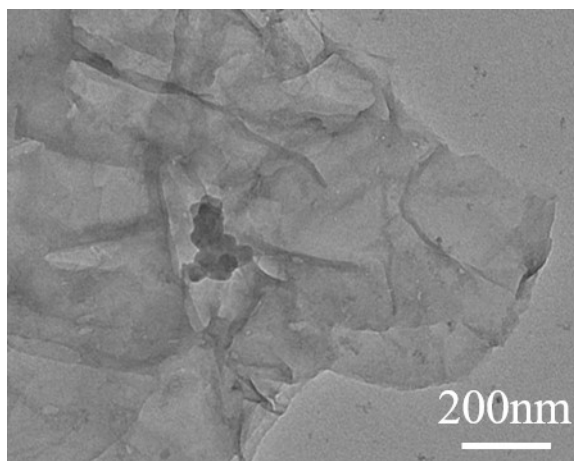


Fig. S16 TEM image of the ENFG400 after 500 cycles.

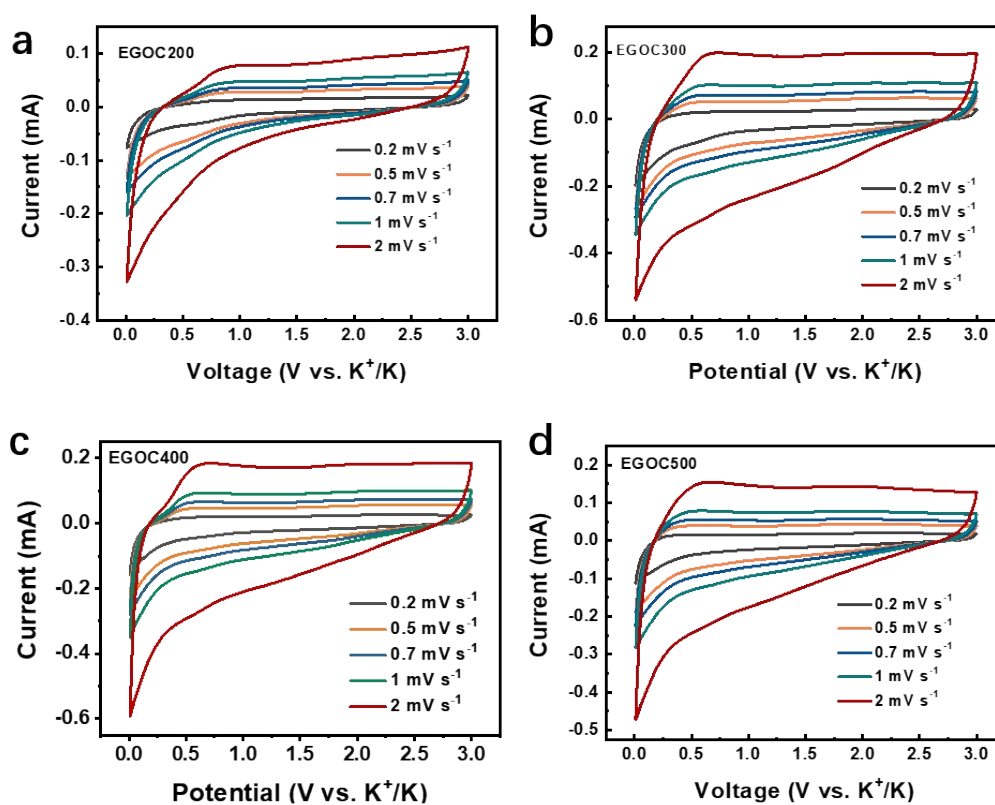


Fig. S17 CV curves at various scan rates of (a) EGO200, (b) EGO300, (c) EGO400 and (d) EGO500.

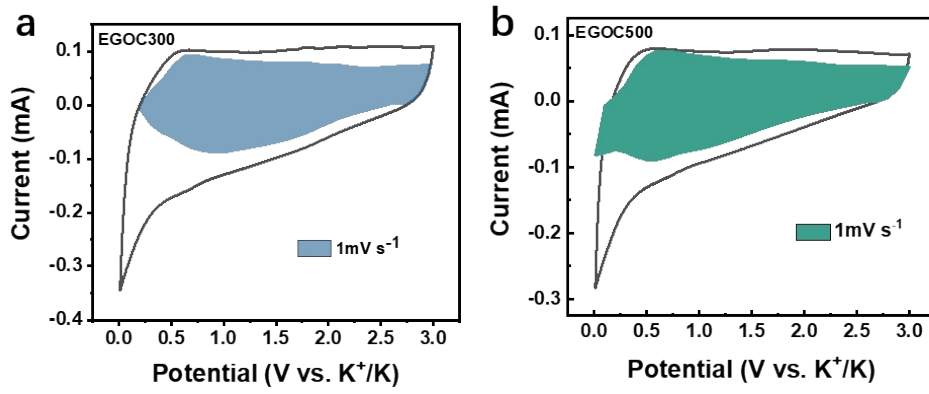


Fig. S18 The capacitive contribution at 1 mV s<sup>-1</sup> of (a) EGO300 and (b) EGO500.

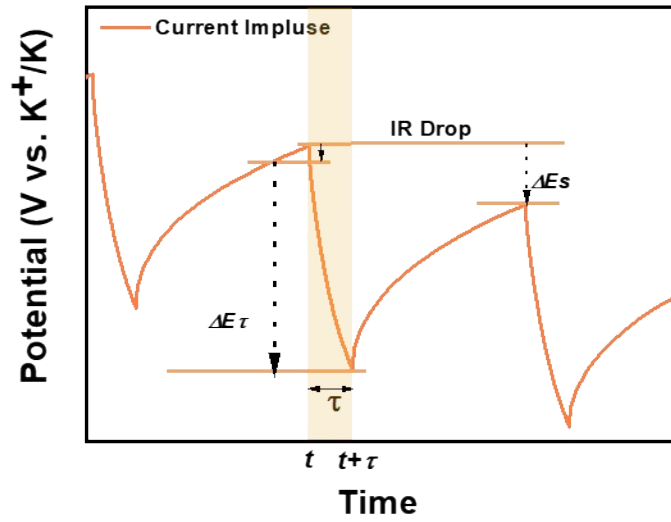


Fig. S19 Determination of  $\Delta E\tau$  and  $\Delta E_s$  from the measured GITT profiles.

The K<sup>+</sup> diffusion coefficient in EGO electrodes can be calculated by solving Fick's second law using the following equation:

$$D = \frac{4}{\pi\tau} \left( \frac{m_B V_m}{M_B S} \right)^2 \left( \frac{\Delta E_s}{\Delta E_\tau} \right)^2$$

where  $\tau$  is the relaxation time,  $m_B$  is electrode active mass,  $M_B$  is the molar mass of the electrode material for carbon,  $V_M$  is the molar volume of carbon,  $S$  is the

geometric area of the electrode,  $\Delta E_s$  is the steady-state potential charge by the current pulse and  $\Delta E_\tau$  is the potential charge during the constant current pulse.

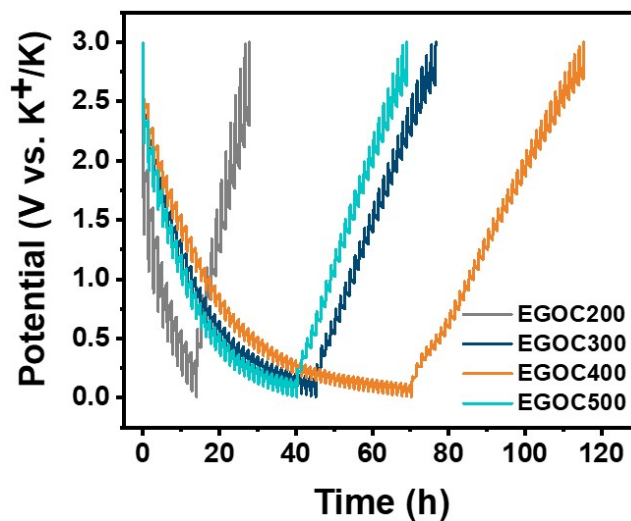


Fig. S20 GITT profiles at various potassiation/depotassiation voltages in first cycle for EGO C200, EGO C300, EGO C400 and EGO C500.

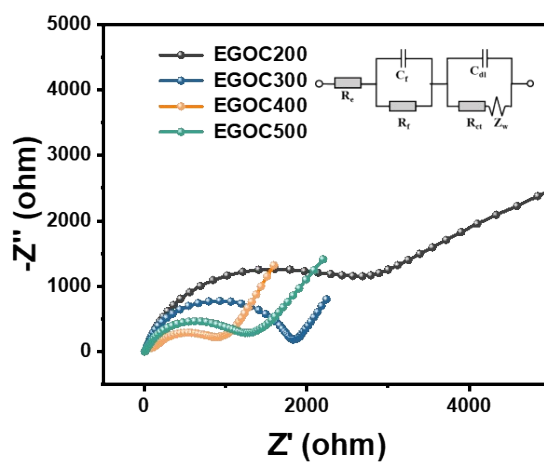


Fig. S21 EIS plots of EGO C200, EGO C300, EGO C400 and EGO C500 at open-circuit voltage.

The energy density and power density of PIC were calculated using the following equations:

$$E = \int_{t_1}^{t_2} UI/m dt$$

$$P = \frac{E}{t} \times 3600$$

where  $I$ ,  $U$ ,  $m$ ,  $t_1$ , and  $t_2$  are assigned to the discharge current (A), operating voltage (V), total mass of active materials (g), start and end times of the discharge (s), respectively, and  $t$  is  $t_1 - t_2$ .

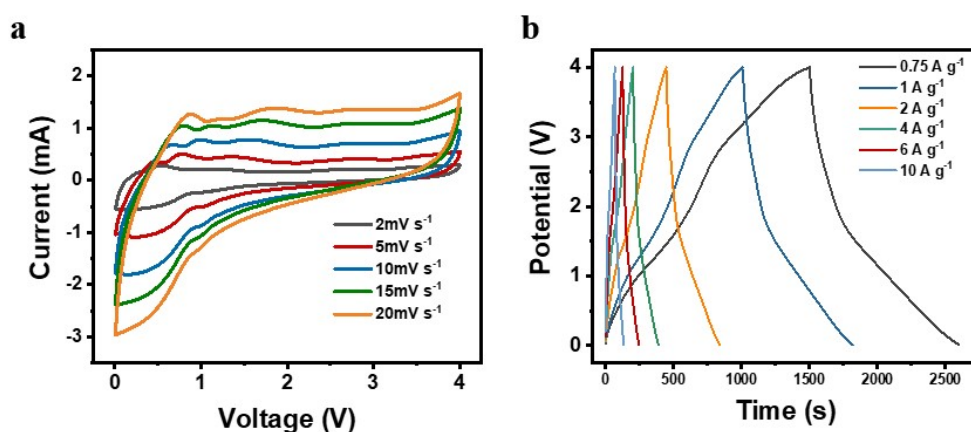


Fig. S22 (a) The CV curves at different scan rates and (b) galvanostatic charge–discharge profiles at various current densities of the (-) REGOC//AC (+) PIC.

Table S1. The structural parameters of GOC and NFG.

Samples	d <sub>002</sub> (nm)	L <sub>a</sub> (nm)	L <sub>c</sub> (nm)	G (%)
GOC	0.3384	52.34	24.02	65.60
NFG	0.3361	91.27	31.06	91.83

Table S2. The C and O contents of the as-prepared EGOc at different temperature by XPS analysis.

Samples	C (at%)	O at%	C-SP <sup>2</sup> (%)
EGOC200	71.07	28.93	48.18
EGOC300	83.74	16.26	65.55
EGOC400	85.19	14.81	68.38
EGOC500	86.09	13.91	69.03

Table S3. Textual parameters of EGOc.

sample	<sup>a</sup> S <sub>BET</sub> m <sup>3</sup> g <sup>-1</sup>	<sup>b</sup> V<2nm m <sup>3</sup> g <sup>-1</sup>	<sup>c</sup> V<50nm m <sup>3</sup> g <sup>-1</sup>	<sup>d</sup> V>50nm m <sup>3</sup> g <sup>-1</sup>	<sup>e</sup> V <sub>DFT</sub> m <sup>3</sup> g <sup>-1</sup>
EGOC200	103.34	0.165	0.148	0.259	0.572
EGOC300	369.17	0.44	0.894	0.413	1.351
EGOC400	573.40	0.067	1.37	1.509	2.496
EGOC500	449.09	0.054	1.128	0.466	1.648

a: The BET specific surface area; b: Pore volume of micropores calculated by DFT; c: Pore volume of  $2\text{ nm} < d < 10\text{ nm}$  calculated by DFT; d: Pore volume of  $10\text{ nm} < d < 50\text{ nm}$  calculated by DFT; e: Pore volume of macropores calculated by DFT; f: The total DFT pore volume.

Table S4 Kinetics parameters of PCNSs

sample	$R_e$	$R_f$	$R_{ct}$
EGOC200	6.12	18.8	298.3
EGOC300	5.74	13.47	164.1
EGOC400	5.15	11.65	147.54
EGOC500	5.66	12.89	157.7