Supplementary Information for

Achieving High Energy Density in a 4.5 V All Nitrogen-Doped Graphene Based Lithium-Ion Capacitors

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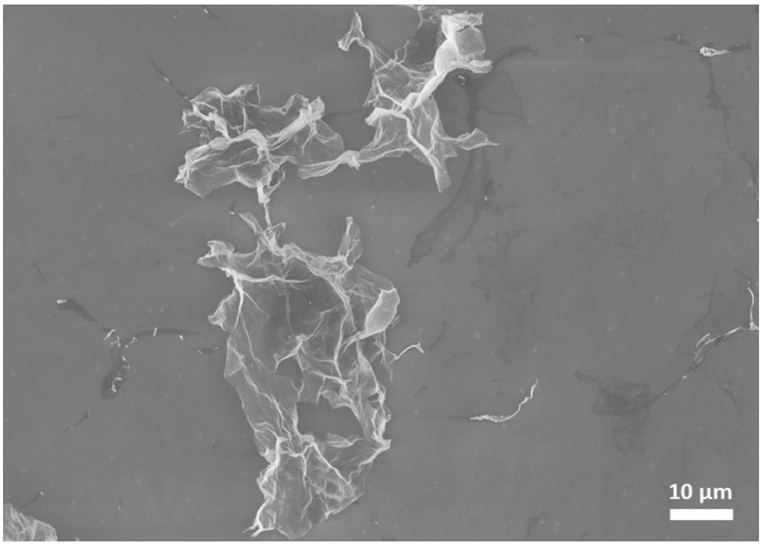

Figure S1. SEM image of the GO nanosheets.

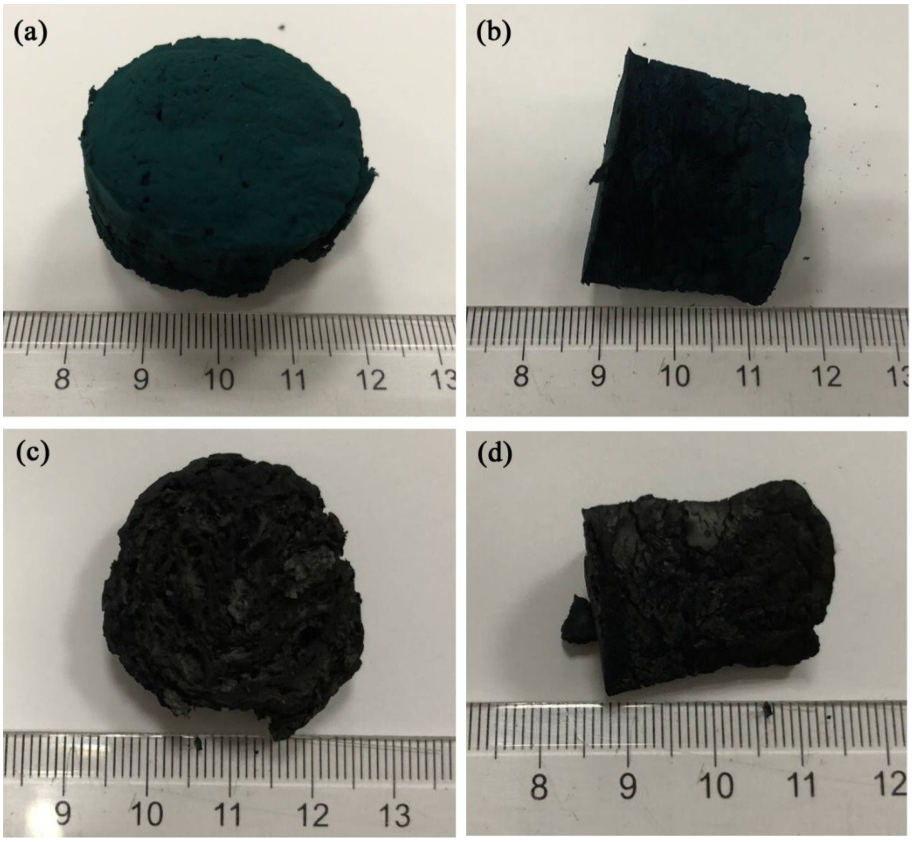

Figure S2. Optical images of GO/PANi aerogels (a, b), A-N-GS aerogel (c) and N-GS aerogel (d).

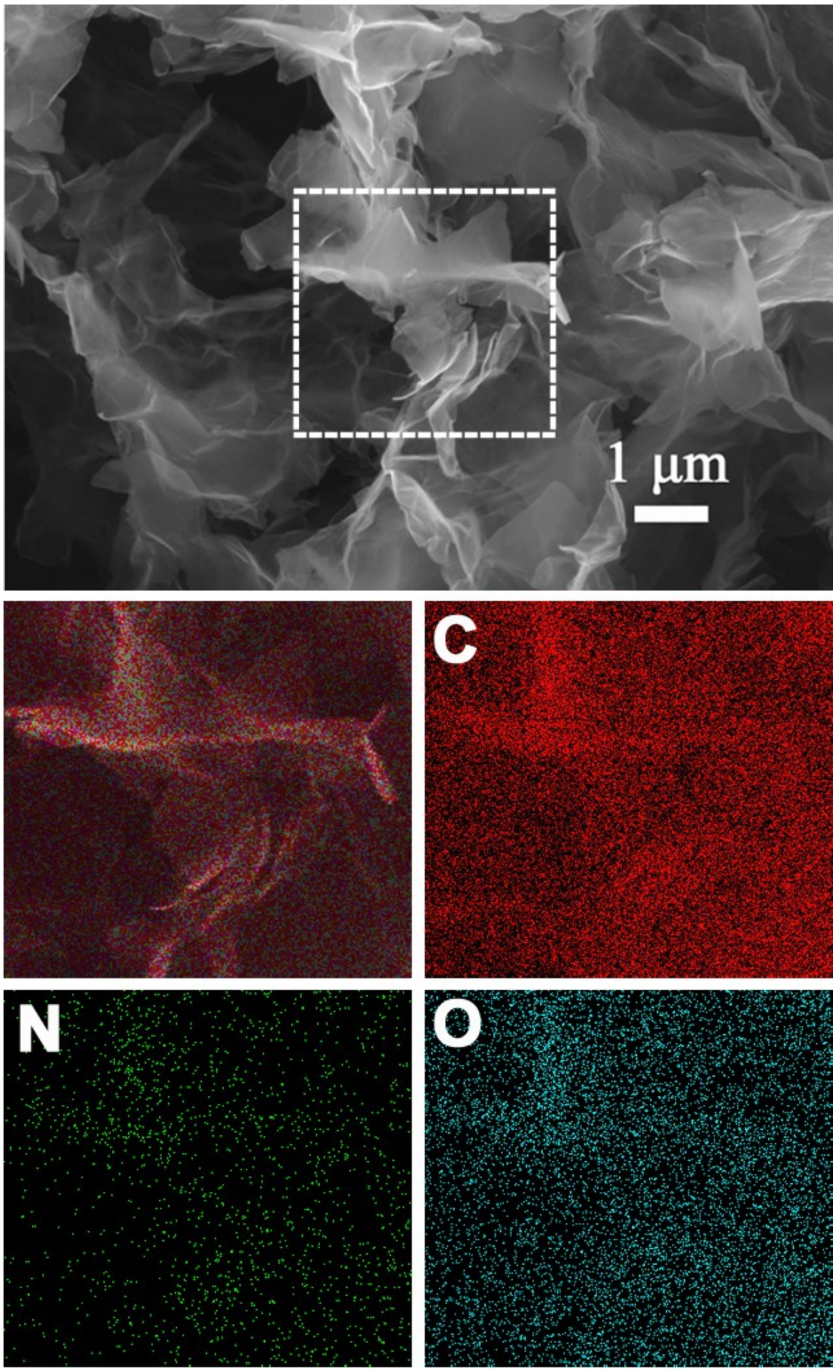

Figure S3. SEM images of GO/PANi aerogels and the corresponding EDS mapping.

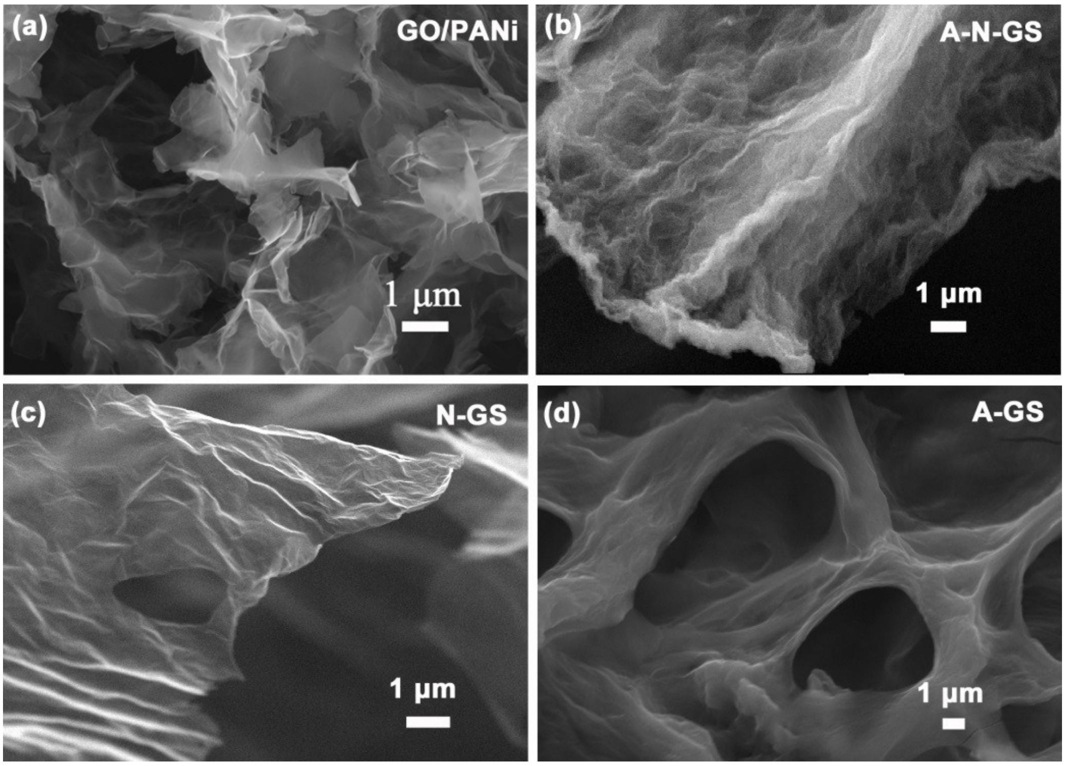

Figure S4. SEM images of (a) GO/PANi, (b) A-N-GS, (c) N-GS and (d) A-GS.

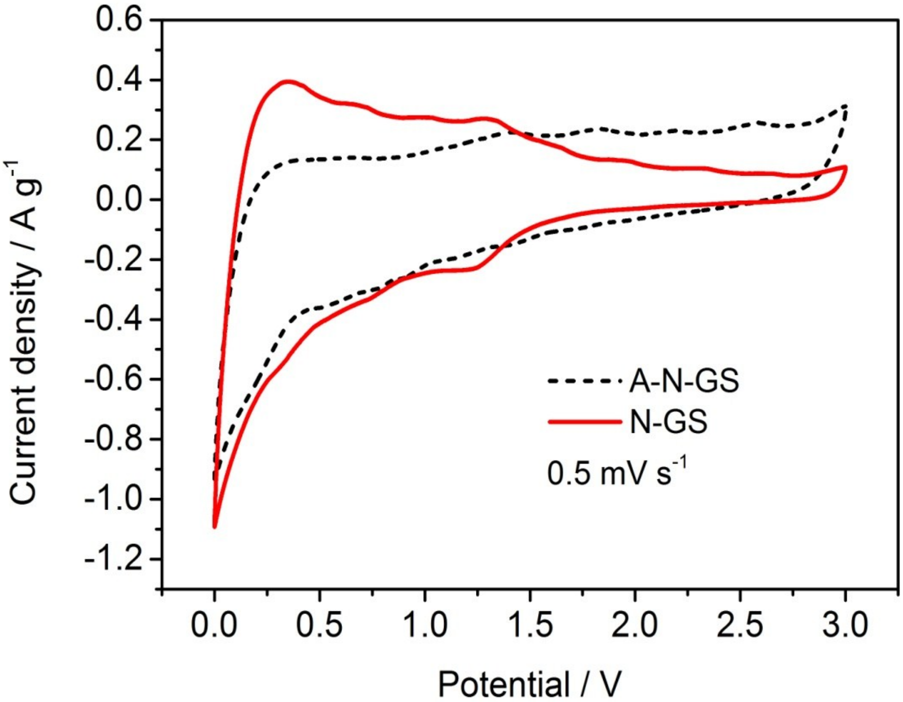

Figure S5. CV curves of A-N-GS and N-GS anodes at a scan rate of 0.5 mV/s.

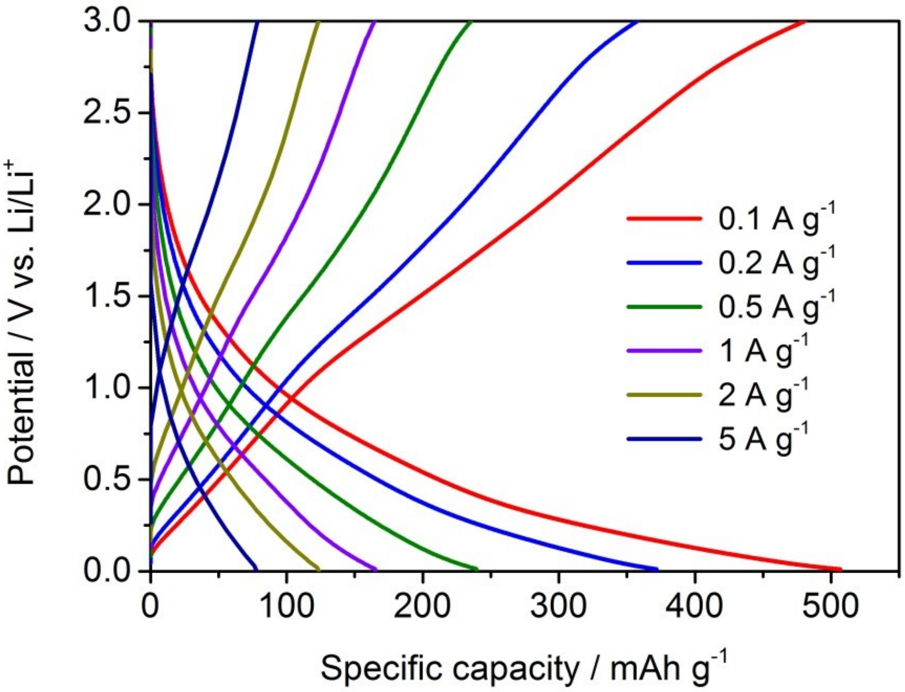

Figure S6. Charge-discharge curves of A-N-GS anodes at different current densities.

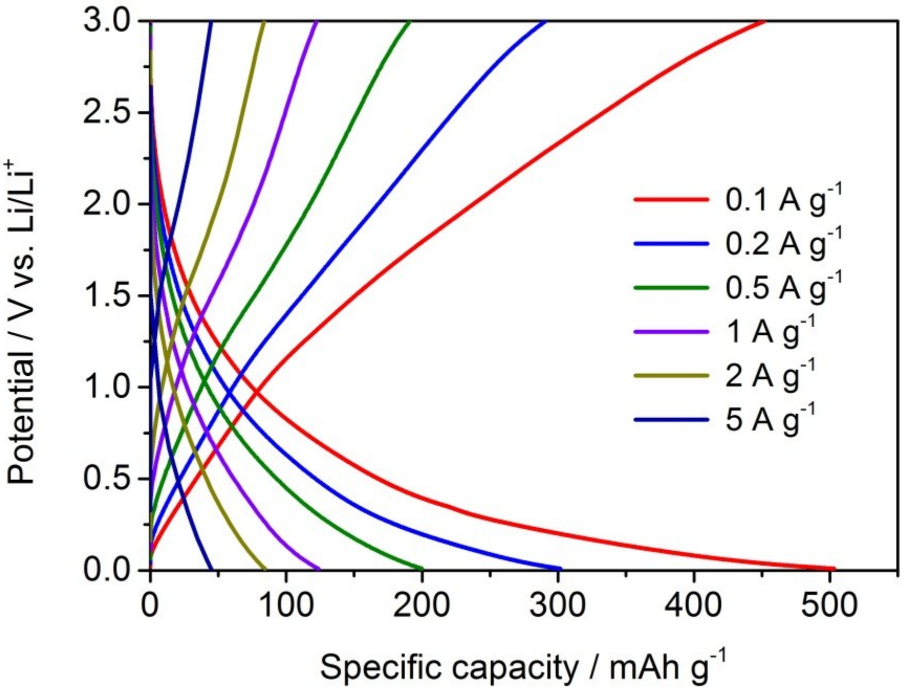

Figure S7. Charge-discharge curves of A- GS anodes at different current densities.

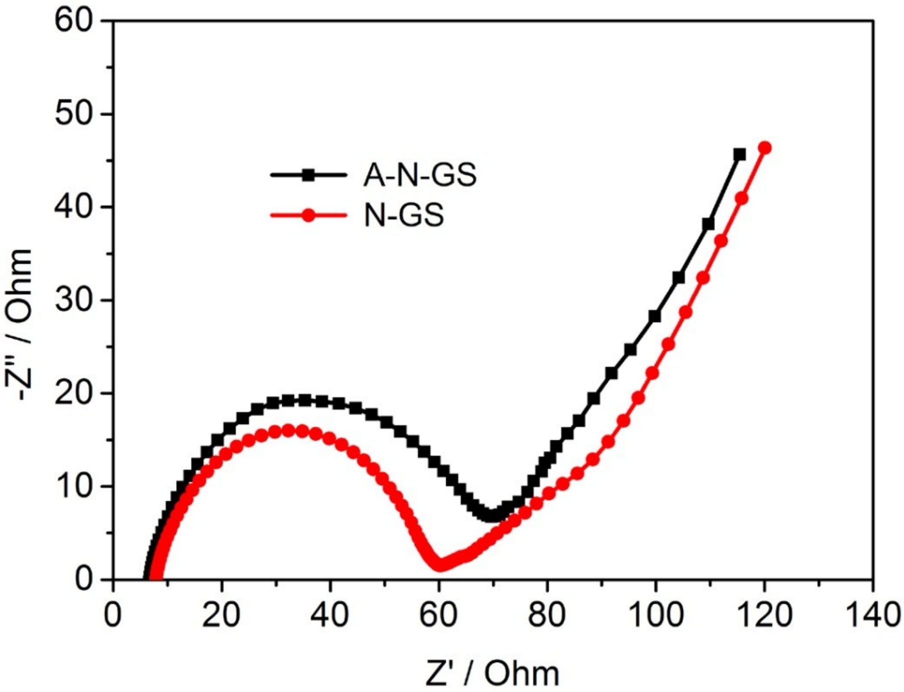

Figure S8. Nyquist plots of A-N-GS and N-GS.

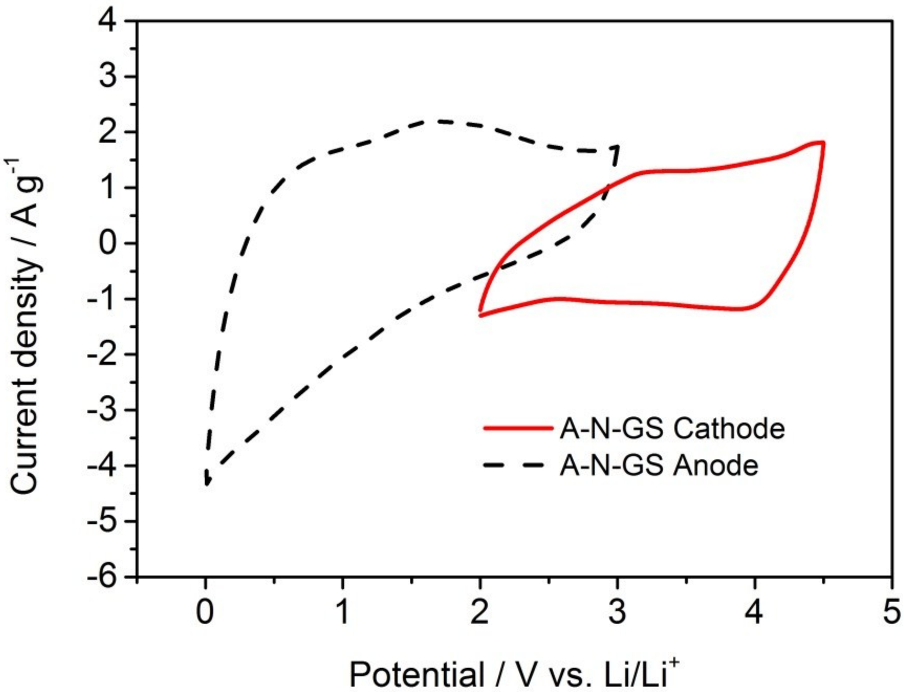

Figure S9. CV curves of A-N-GS cathode and A-N-GS anode at a scan rate of 10 mV/s.

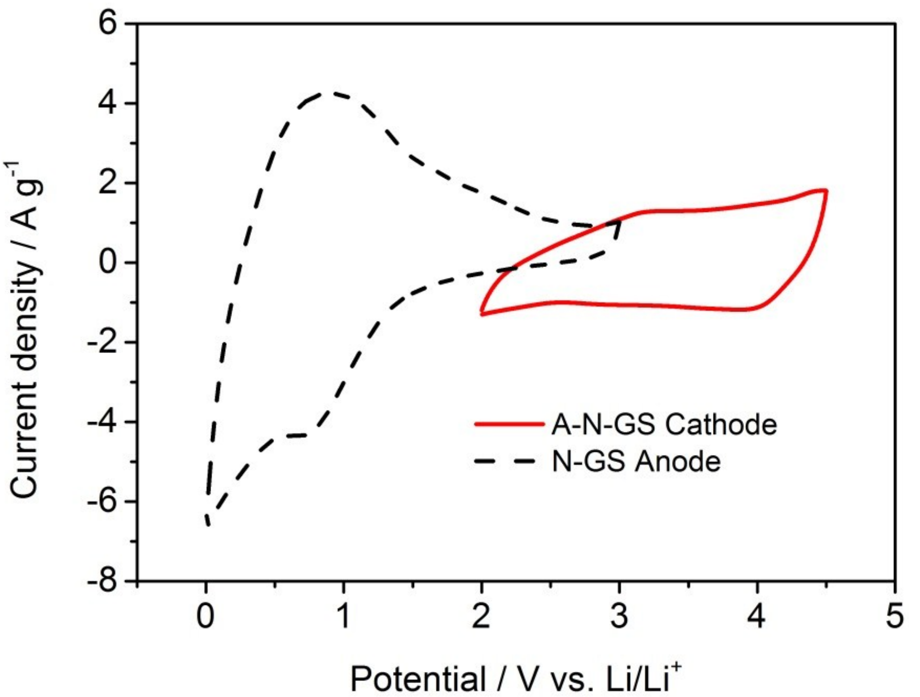

Figure S10. CV curves of A-N-GS cathode and N-GS anode at a scan rate of 10 mV/s.

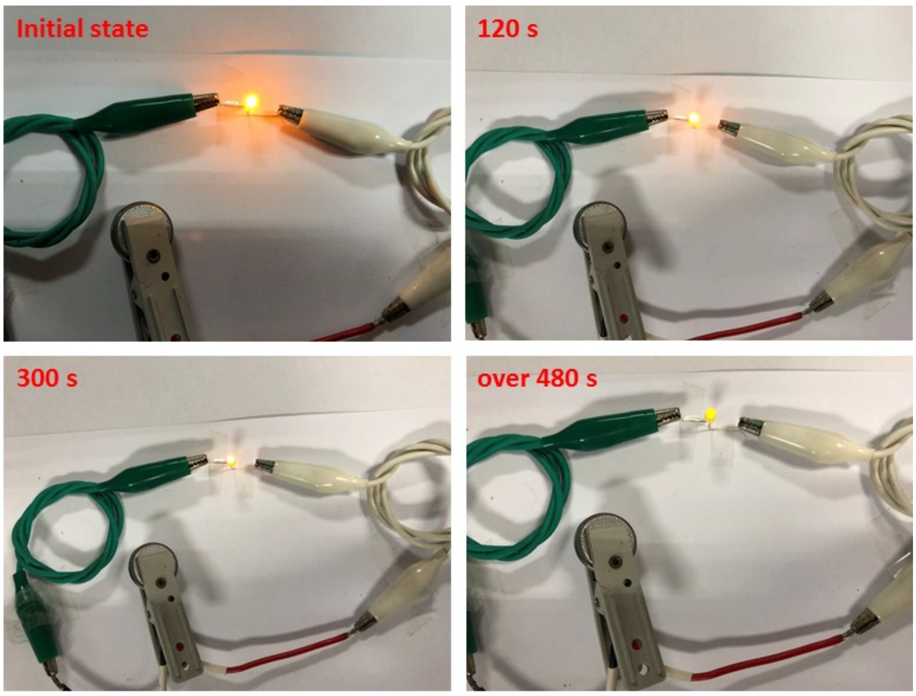

Figure S11. Time-dependent optical images of yellow LED powered by a single 4.5 V A-N-GS//N-GS LIC device. Also, a supplementary movie clearly shows that a single LIC could light a LED more than 8 minutes.

Table S1. Detailed surface and pore parameters of the graphene samples.

Sample	S_{BET} (m^2/g)	V_{tot}(cm^3/g)	V_{mic} (cm^3/g)	V_{mes}(cm^3/g)	V_{mac}(cm^3/g)
A-N-G	1086.2	0.503	0.134	0.36	0.009
A-GS	809.2	0.410	0.125	0.259	0.026
N-GS	201.1	0.506	0.017	0.15	0.338

Table S2. Summary of the electrochemical performance of high voltage (>4.0 V) LICs based on different active materials.

References	**LIC configuration (anode // cathode)**	**voltage window**	**max energy (Wh/kg) @ power (W/kg)**	**energy (Wh/kg) @ max power (W/kg)**	**capacity retention**
[1]	Si/C // porous activated carbon	2.0~4.5 V	257 @ 867	147 @ 29893	79.2% over 15000 cycles
[2]	Si/C // rice husks activated carbon	2.0~4.0 V	227 @ 1146	~181 @32595	N.A.
[3]	B-doped Si/SiO_2/C // activated carbon	2.0~4.5 V	128 @ 1229	89 @ 9704	70% over 6000 cycles
[4]	Si/C // Biomass-derived porous carbon	2.0~4.5 V	159 @ 945	99 @ 31235	80% over 8000 cycles
[5]	Li_3VO_4/N-doped carbon nanowires // activated carbon	1.0~4.0 V	136.4 @ 532	24.4 @ 11020	~87% over 1500 cycles
[6]	$BiVO_4$ // reduced graphene oxide	0.0~4.0 V	152 @ 384	42 @ 3861	81% over 6000 cycles
[7]	SnO_2-C // activated carbon (YP47)	0.0~4.0 V	110 @ 173	~48.5@ 2960	80% over 2000 cycles
[8]	$MnFe_2O_4$/C // 3D amorphous carbon	0.0~4.0 V	157 @ 200	58 @ 20000	86.5% over 6000 cycles
[9]	$ZnMn_2O_4$-graphene nanosheets // N-doped carbon nanosheets	1.0~4.0 V	202.8 @ 180	98 @ 21000	86.6% over 6000 cycles
[10]	Fe_3O_4 // activated carbon nanofiber	1.0~4.0 V	124.6 @ 93.8	103.7 @ 4687.5	85.8% over 1000 cycles

[11]	Co_3ZnC@N-doped porous carbon // heteroatom-doped microporous carbon	1.0~4.5 V	141.4 @ 275	15.2 @ 10300	~80% over 1000 cycles
[12]	3D TiC // porous N-doped carbon	0.0~4.5 v	101.5 @ 450	23.4 @ 67500	~82% over 5000 cycles
[13]	3D VN-rGO // porous carbon nanorods	0.0~4.0 V	162 @ 200	64 @ 10000	83% over 1000 cycles
[14]	B,N-codoped carbon nanofiber (BNC) // BNC	0.0~4.5 V	220 @ 225	104 @22500	81% over 5000 cycles
[15]	N-doped porous carbon microspheres (NPCM) // activated NPCM	2.0~4.0 V	95.08 @ 300	48.2 @ 15000	80.1% over 5000 cycles
[16]	Graphene-CNT // graphene-CNT	0.01~4.3 V	~120 @ ~110	29 @ ~20500	89% over 10000 cycles
[17]	Porous carbon nanofiber (PCNF) // PCNF	1.0~4.0 V	106 @ 242	53 @ 30000	91% over 2000 cycles
This work	**N-GS // 3D A-N-GS (binder-free)**	**0.0~4.5 V**	**187.9 @ 2250**	**111.4 @ 11250**	**93.5% over 3000 cycles**

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