Nucleotide(s)-mediated simultaneous N, P co-doped reduced graphene oxide (N, P-rGO) porous nanohybrids as a high-performance electrode material for designing sustainable binder-free high-voltage (2.8 V) aqueous symmetric supercapacitor and electrochemical sensor

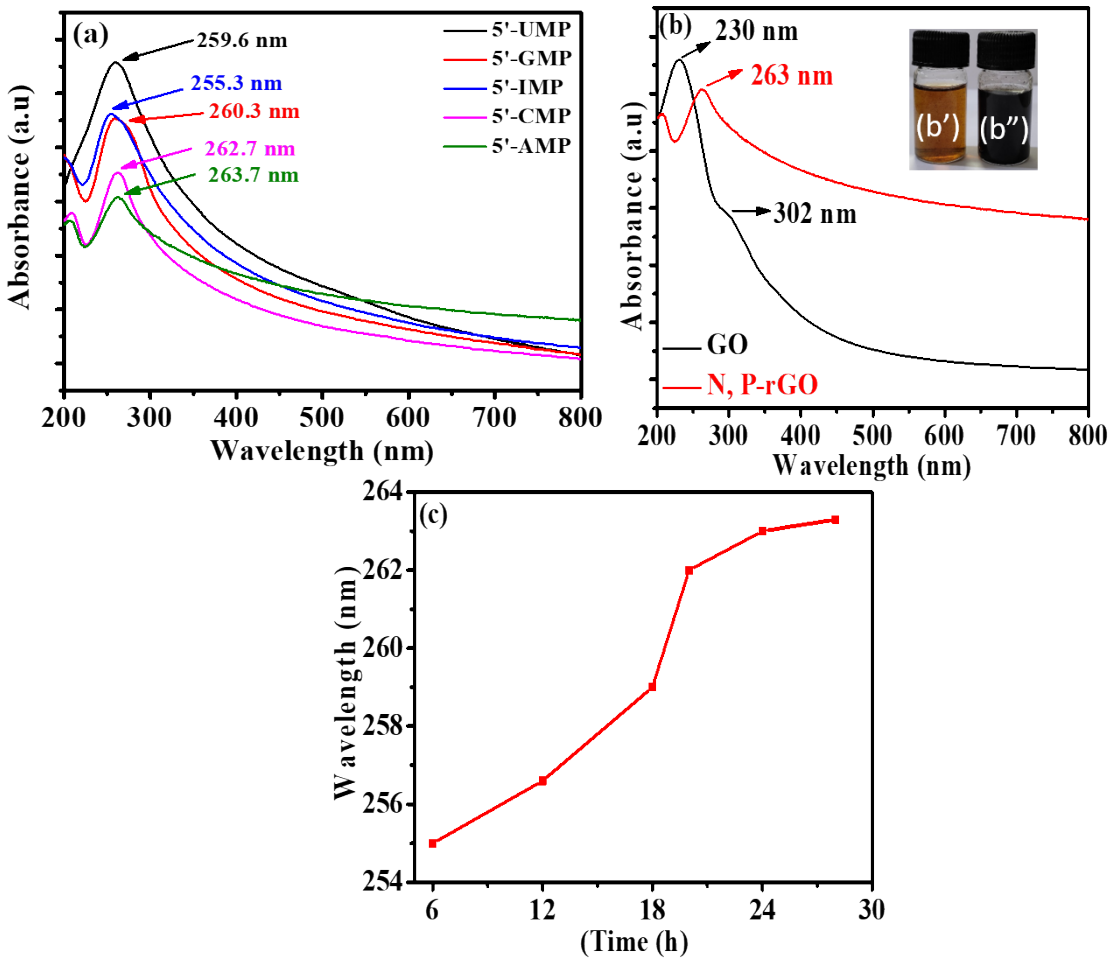


Fig. S1. Optical spectra of different nucleotide(s)-mediated reduction of GO (a); Optical spectra of GO and 5′-AMP-mediated-N, P-rGO (inset) digital photograph of GO (b') and N, P-rGO (b") (b); maximum absorption of N, P-rGO with time (c).

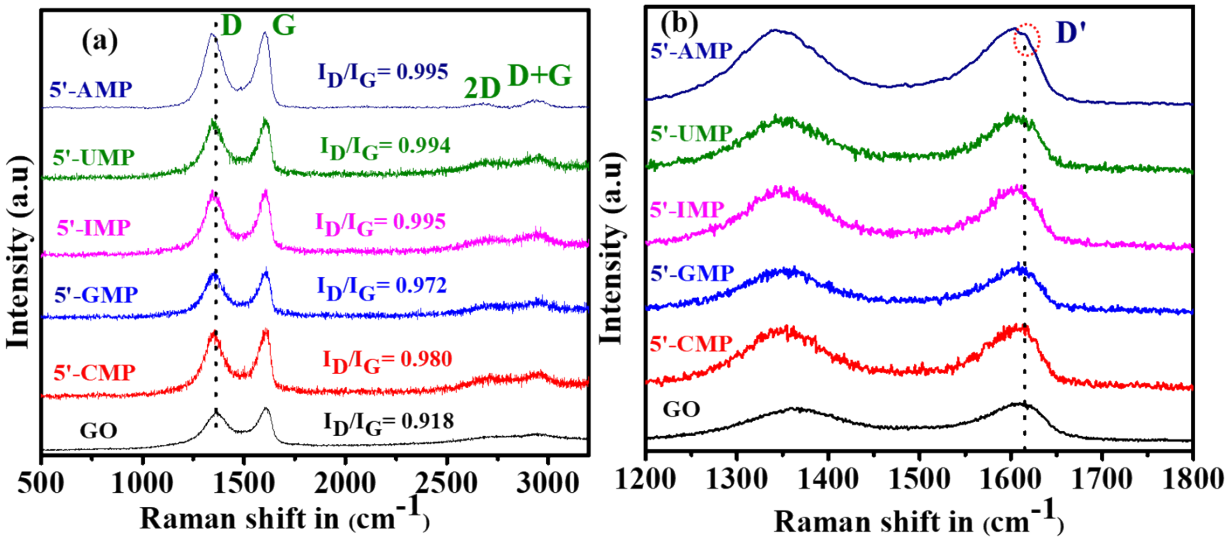


Fig. S2. Raman spectral comparison of different nucleotide(s) (5′-AMP, 5′-UMP, 5′-IMP, 5′-GMP and 5′-CMP)-mediated N, P- rGO in the wavenumber range of 500-3500 cm^{-1}; Raman spectra of GO (a); Raman spectra of these samples extended in the wavenumber range of 1200-1800 cm^{-1} (a').

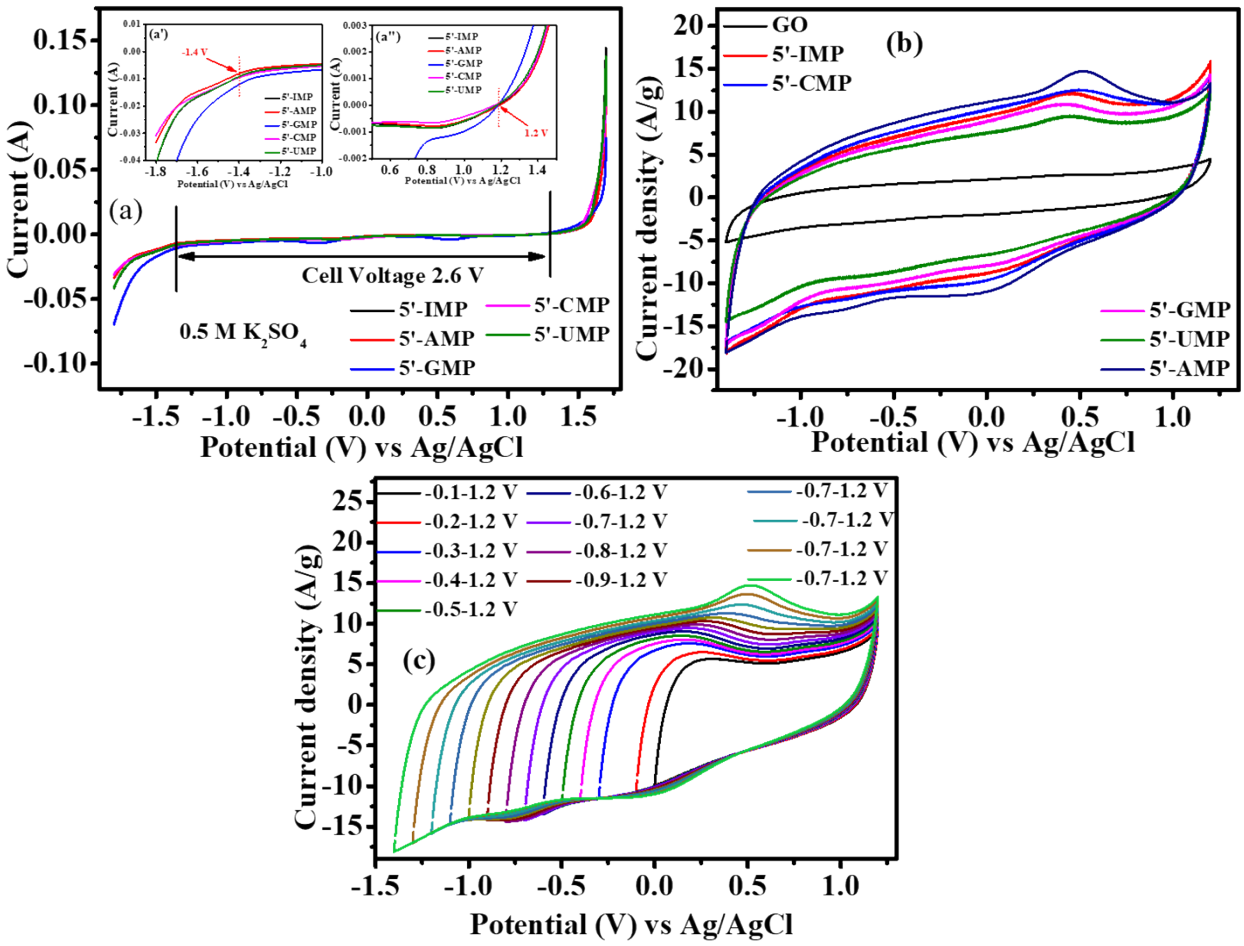


Fig. S3. Linear sweep voltammetry (LSV) of three electrode system for different nucleotide(s)-mediated N, P-rGO in 0.5 m K_2SO_4 (a), extended LSV Voltammograms in different potential window of these samples - Inset (a') (a''). A comparison of CVs of different nucleotide(s) (5′-AMP, 5′-UMP, 5′-IMP, 5′-GMP and 5′-CMP)-mediated N, P-rGO in the potential range of -1.4 to 1.2 V with a GO (b); CV plots of 5′-AMP mediated N, P-rGO as a function of variation in cathodic potential ranging from: -1.4 to 0 V (c)

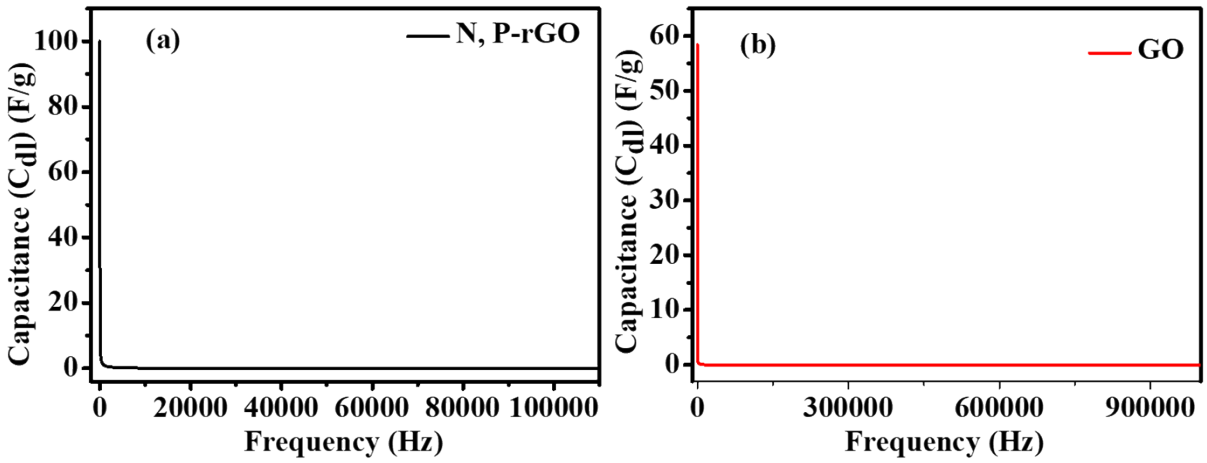


Fig. S4. Capacitance (C_{dl}) vs Frequency curve at frequency range of 0.01 H_Z to 10^6 Hz obtained using the Nyquist plot.

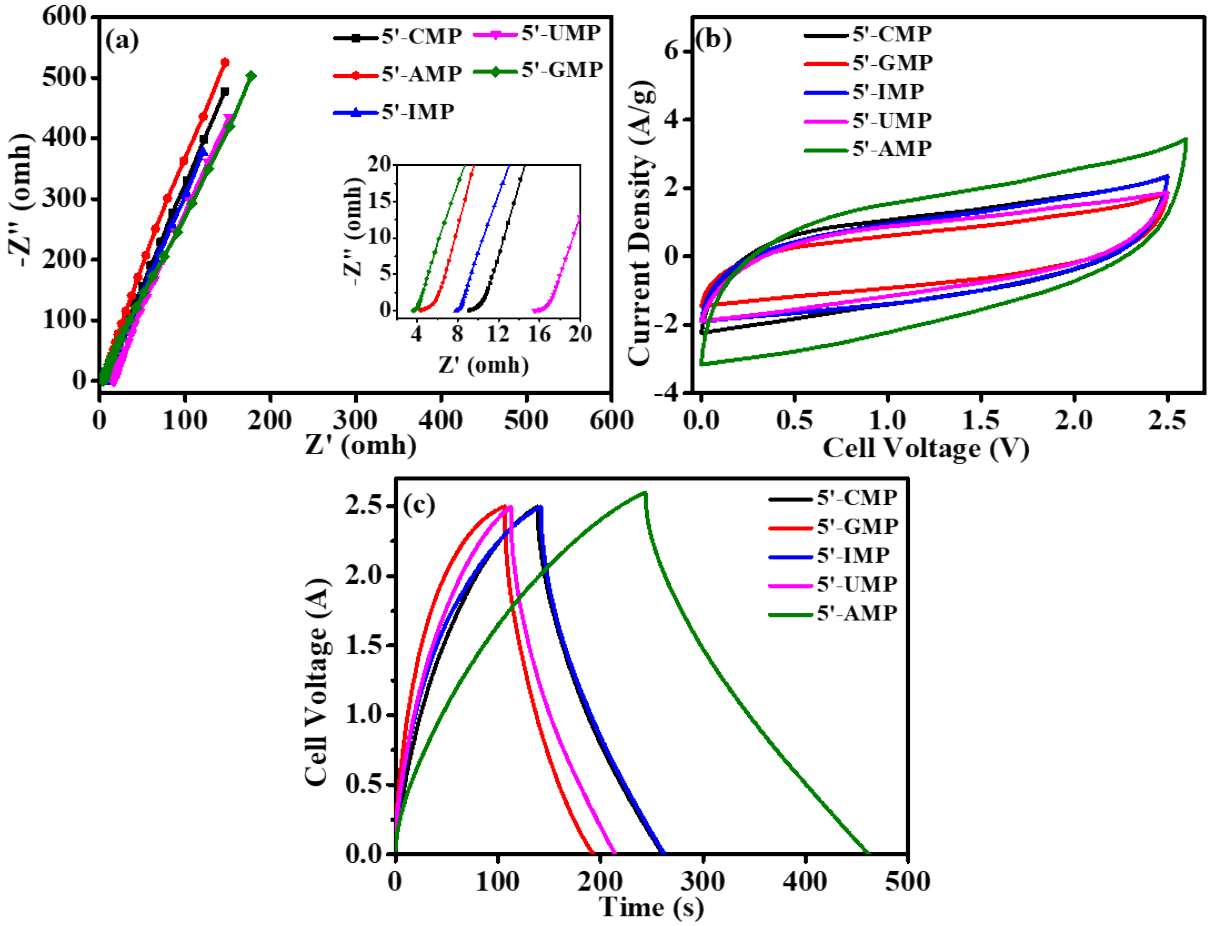


Fig. S5. Impedance curves, inset: extended curves between 0-3 Ohm (a); CV measurements for SSC constructed from nucleotides 5′-CMP/ 5′-AMP/ 5′-GMP/ 5′-UMP and 5′-IMP-mediated N, P-rGO in 0.5 m K_2SO_4 (b). CV voltammograms recorded at 100 mV/s scan rate GCD curves at 1 A/g current density (c).

Table S1. Electrochemical data for SSC, constructed from nucleotides 5′-CMP/5′-AMP/ 5′-GMP/ 5′-UMP/ 5′-IMP-mediated N, P-rGO, recorded using 0.5 m K_2SO_4 as an electrolyte.

S. No.	Nucleotide	Electrolyte	Cell Voltage (V)	Capacitance (F g^{-1})	Energy density (W h kg^{-1})	Power density (W kg^{-1})	Coulombic efficiency
1	5′-GMP	0.5 m K_2SO_4	2.5	17.4	15.1	624.8	81.0
2	5′-CMP	0.5 m K_2SO_4	2.5	24.5	24.2	624.8	88.4
3	5′-AMP	0.5 m K_2SO_4	2.6	36.0	33.9	644.9	90.0
4	5′-UMP	0.5 m K_2SO_4	2.5	20.3	17.6	624.8	90.3
5	5′-IMP	0.5 m K_2SO_4	2.5	24.2	21.0	624.8	85.4

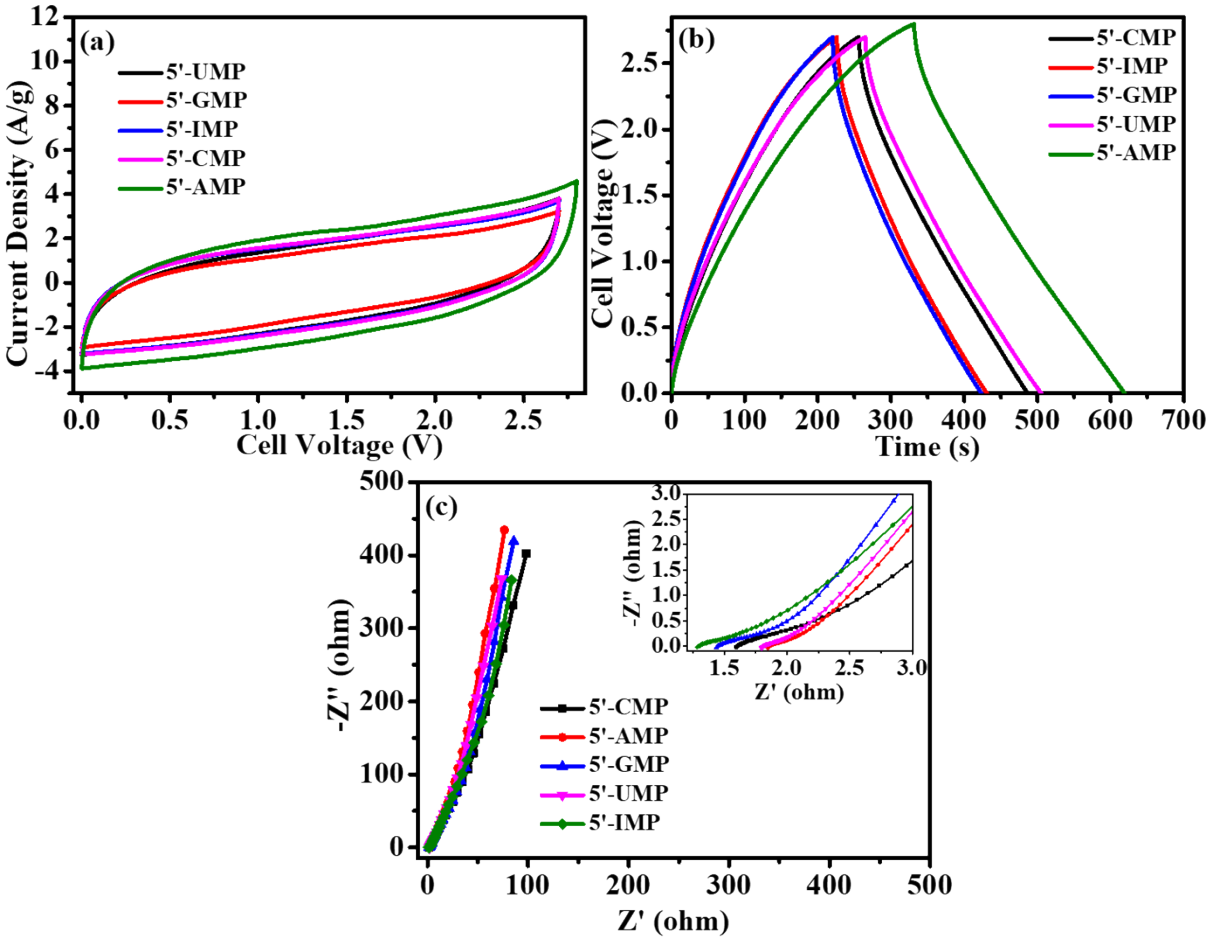


Fig. S6. CV measurements for SSC constructed from nucleotides 5′-CMP/ 5′-AMP/ 5′-GMP/ 5′-UMP/ 5′-IMP-mediated N, P-rGO in 17 m $NaClO_4$: CV voltammograms recorded at 100 mV/s scan rate (a); GCD curves at 1 A/g current density (b). Impedance curves, inset: extended curves between 0-3 Ohm (c).

Table S2. Electrochemical data for SSC, constructed from nucleotides 5′-CMP/ 5′-AMP 5′-GMP/ 5′-UMP/ 5′-IMP-mediated N, P-rGO, recorded using 17 m $NaClO_4$ as an electrolyte.

S. No.	Nucleotides	Electrolyte	Cell Voltage (V)	Capacitance (F g^{-1})	Energy Density (W h kg^{-1})	Power Density (W kg^{-1})	Coulombic efficiency
1	5′-GMP	17 m $NaClO_4$	2.7	37.64	38.11	674.58	92.4
2	5′-CMP	17 m $NaClO_4$	2.7	42.70	43.23	674.56	90.2
3	5′-AMP	17 m $NaClO_4$	2.8	51.35	55.92	699.67	87.0
4	5′-UMP	17 m $NaClO_4$	2.7	44.44	44.99	674.58	90.6
5	5′-IMP	17 m $NaClO_4$	2.7	37.96	38.43	674.53	90.8

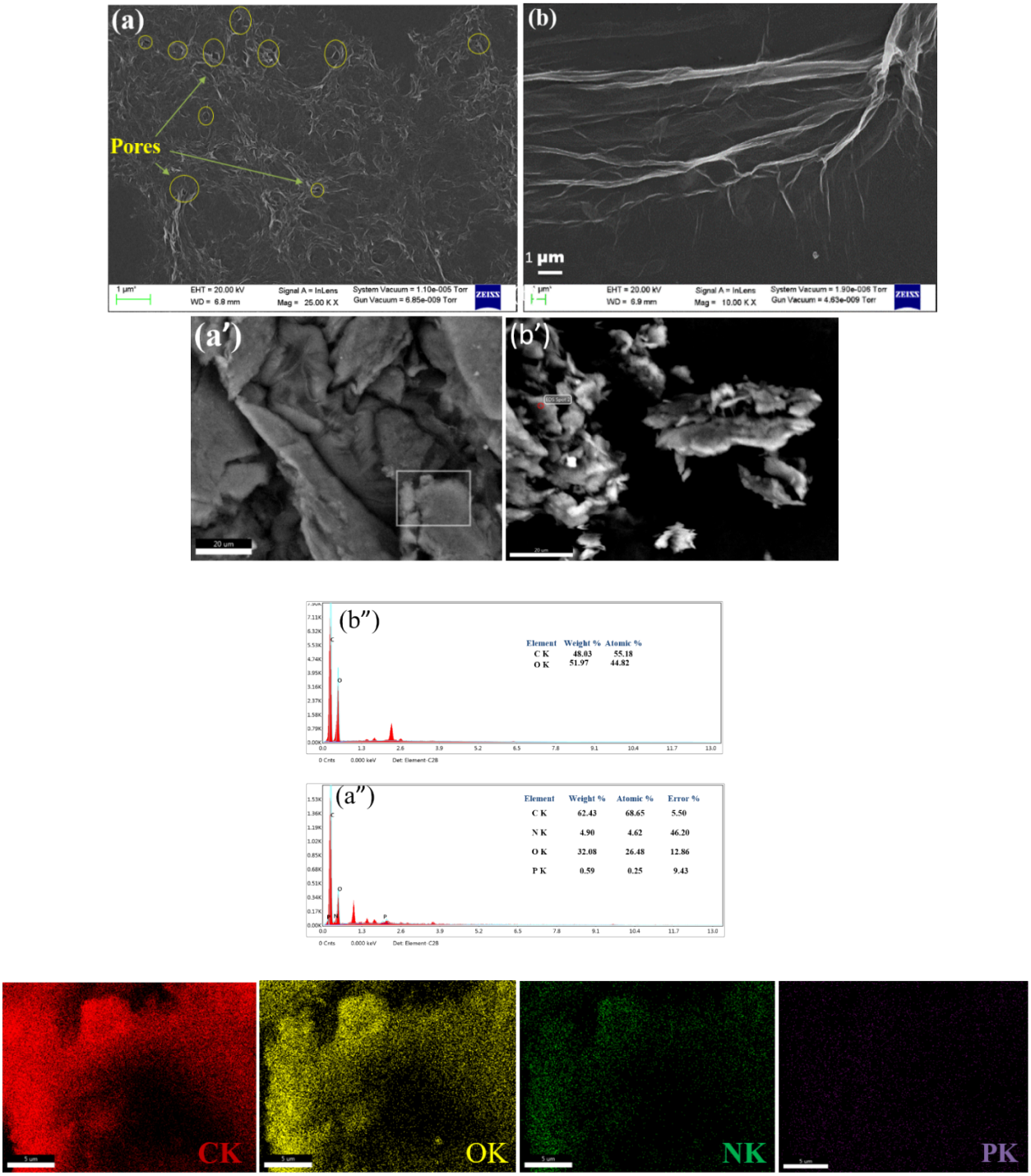


Fig. S7. SEM images of solid sample N, P-rGO (a) and GO (b), EDAX analysis and elemental composition of N, P-rGO (a′) and GO (b″), respectively; elemental mapping of N, P-rGO.

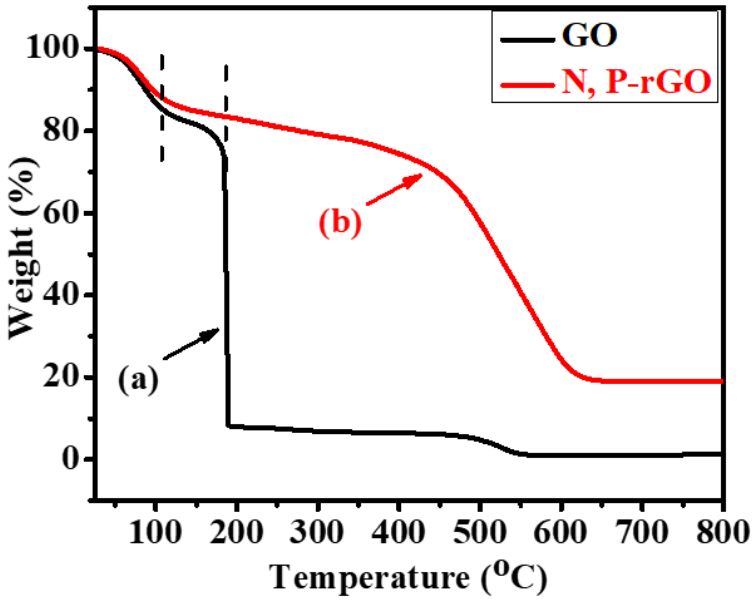


Fig. S8. Thermogravimetric analysis of GO (a) and N, P-rGO (b).

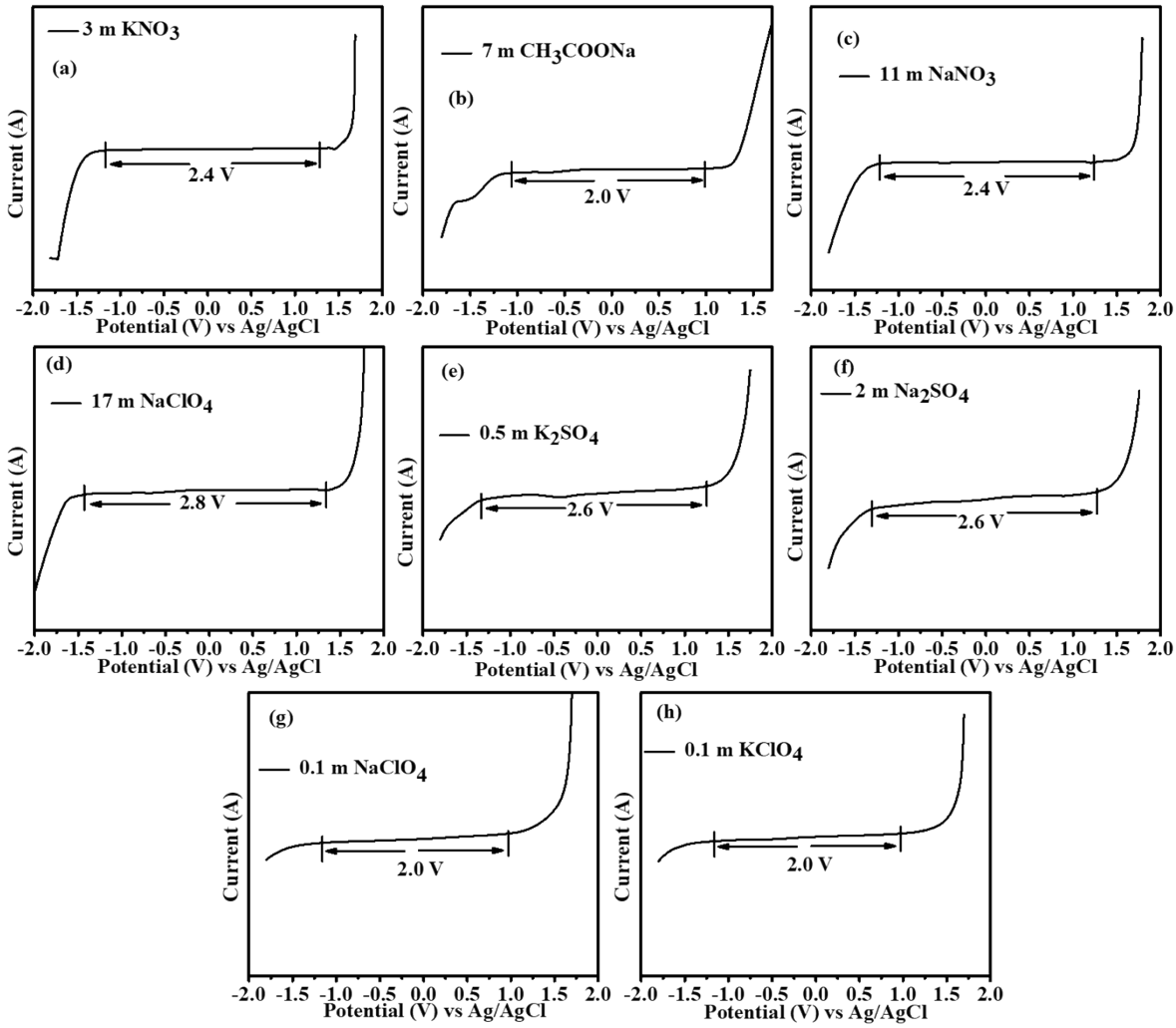


Fig. S9. Linear sweep voltammetry (LSV) measurements for estimating electrode potential window of N, P-rGO in different electrolytes performed at a scanning speed of 10 mV/ s using three-electrode set-up.

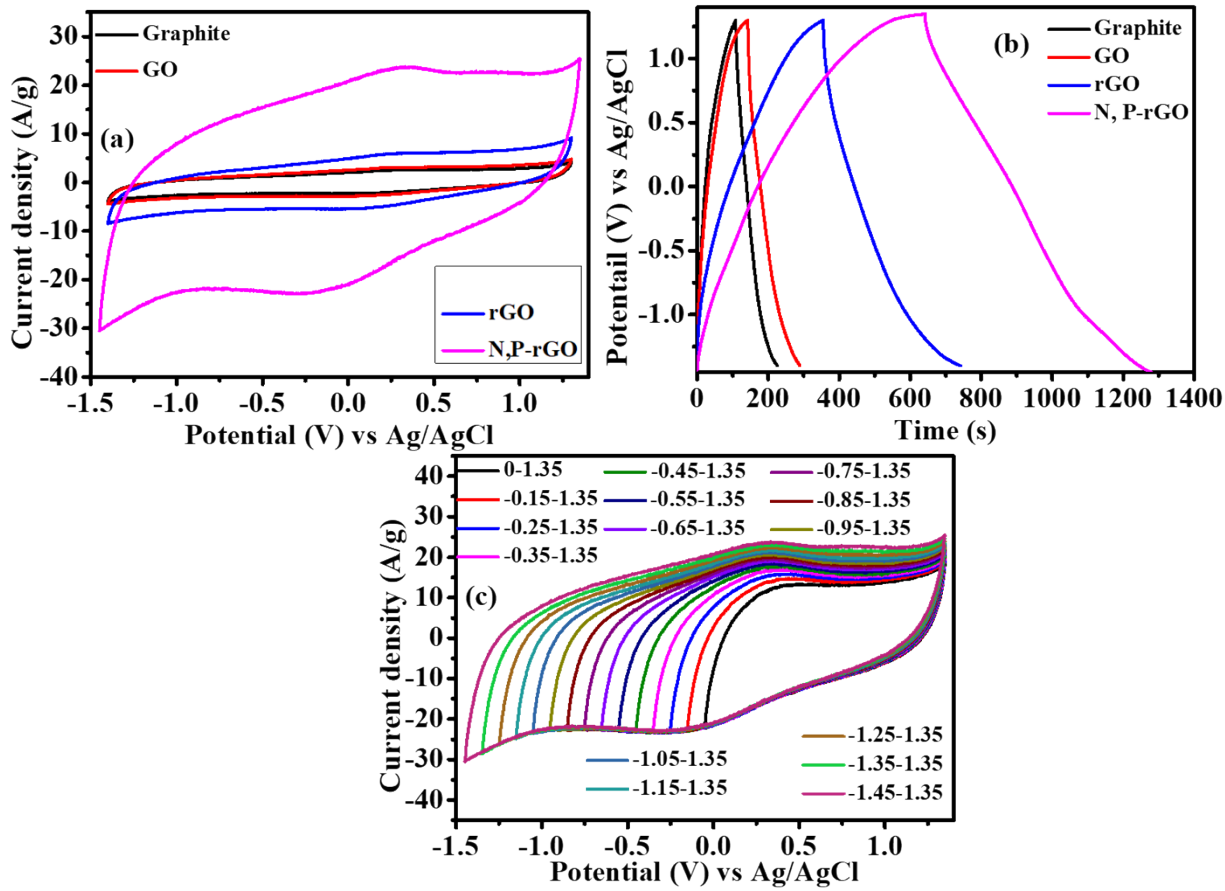


Fig. S10. Cyclic voltammograms recorded for bare graphite, GO, rGO and N, P-rGO (a) and their GCD at 1 A/g (b) with their C_s values (F g^{-1}) calculated to be: 43.8, 54.7, 144.1 and 227.5 respectively. Cyclic voltammograms recorded by varying reduction potential from 0 to -1.5 V in 17 m $NaClO_4$ (c).

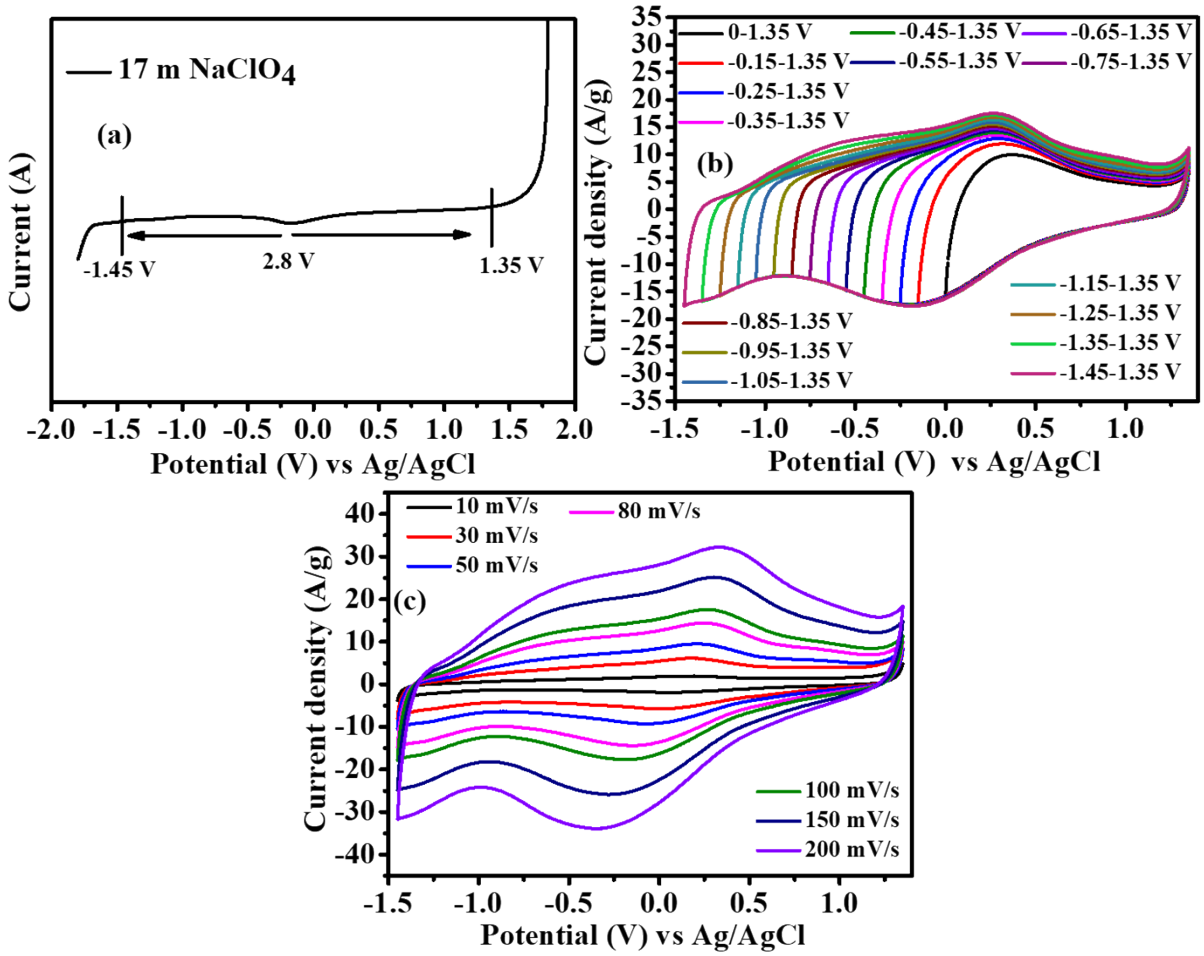


Fig. S11. Linear sweep voltammetry (LSV) of three-electrode system were recorded at a scan rate of 10 mV/ s in the used electrolytes 17 m $NaClO_4$ on a GCE as working electrode (a). Cyclic voltammograms recorded by varying reduction potential from 0 to -1.45 V in 17 m $NaClO_4$ (b) and CV recorded varied scan rate 10 mV/s to 200 mV/s (c).

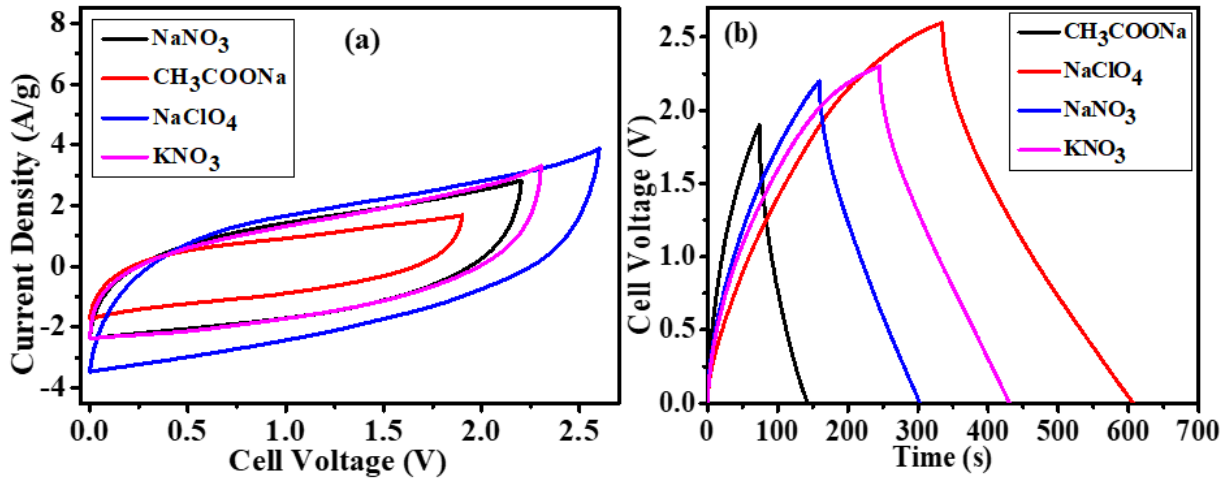


Fig. S12. Cyclic voltammetry measurements performed using SSC, designed from 5′-AMP mediated- N, P-rGO as an electrode material: recorded in 3 m each KNO_3, CH_3COONa, $NaNO_3$, $NaClO_4$ electrolytes at 100 mV/s scan rate (a); GCD curves recorded at 1 A/g current density in these electrolytes (b).

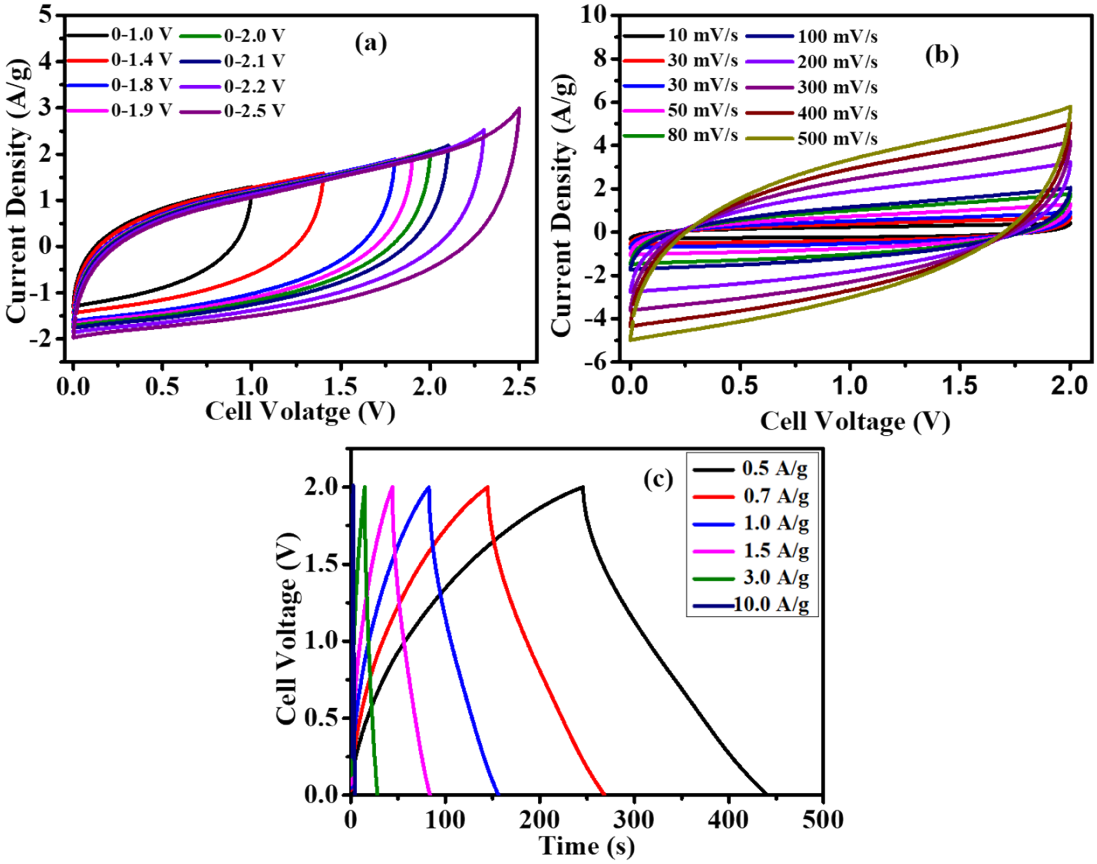


Fig. S13. Cyclic voltammetry measurements performed using SSC, designed from N, P-rGO as an electrode material in 7 m CH_3COONa electrolyte: in varied range of cell voltages (1-2.5 V) at a scan speed of 100 mV/s (a), at different scan rates from 10-500 mV/s for cell voltage of 2.0 V (b); GCD curves recorded at different current density from 0.7 -10 A/g (c).

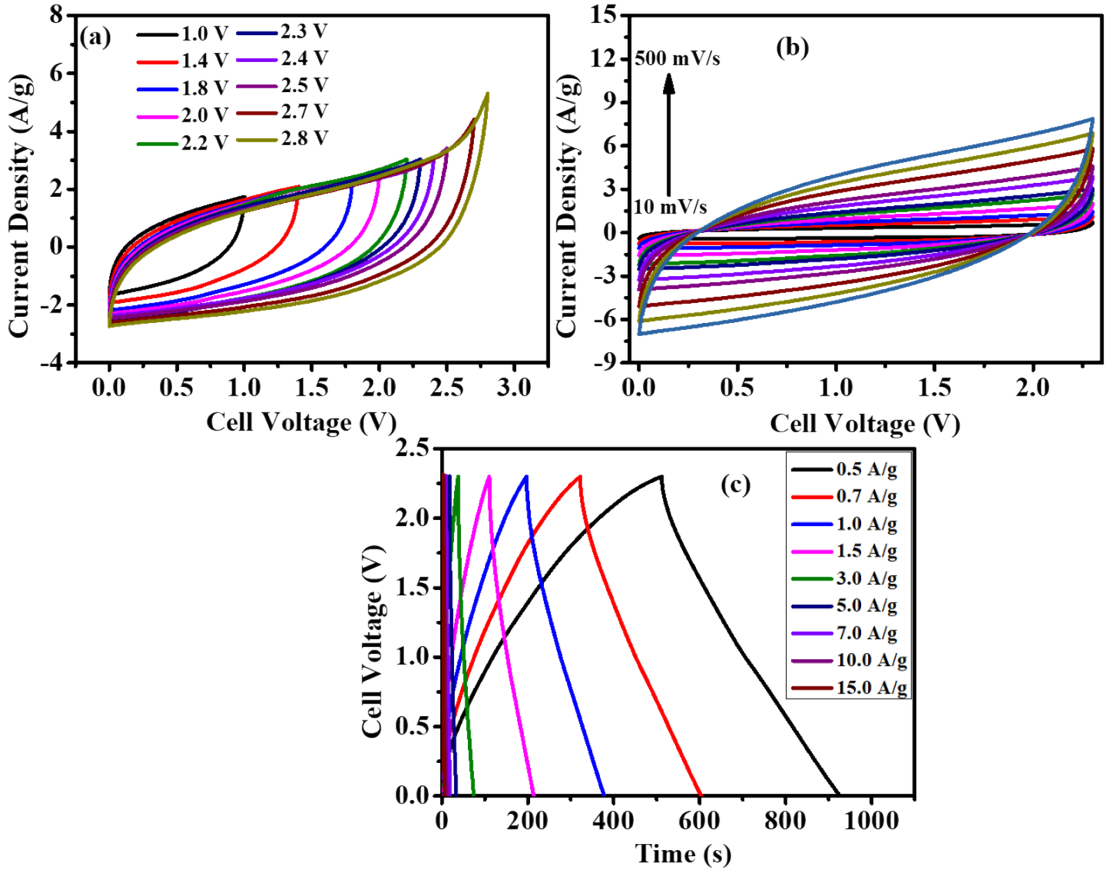


Fig. S14. CV measurements made for the SSC (using N, P-rGO as an electrode material) in the 11m $NaNO_3$ electrolyte: for a varied range of cell voltages (1-2.8 V) at a scan speed of 100 mV/s (a), at different scan rates from 10-500 mV/s (b); GCD for 2.3 V at different current density from 0.7 -15 A/g (c).

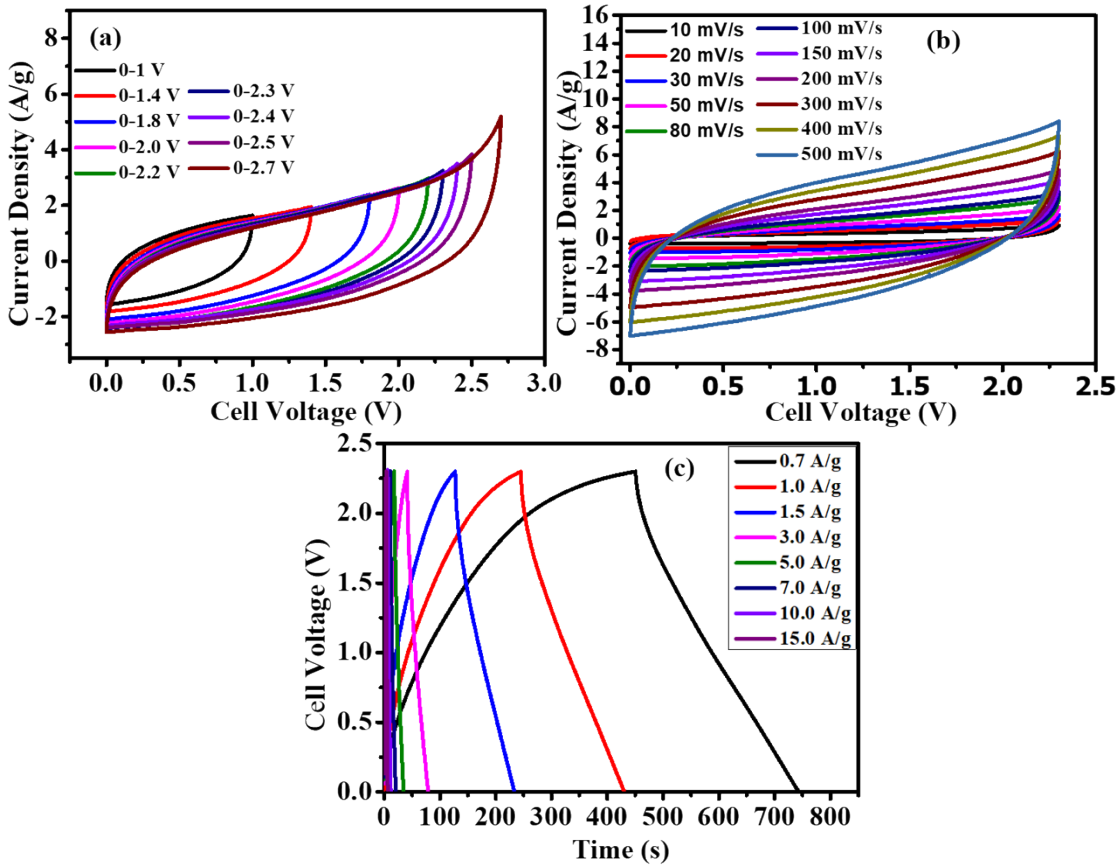


Fig. S15. CV measurements made for the SSC (using N, P-rGO as an electrode material) in the 3 m KNO_3 electrolyte: for a varied range of cell voltages (1-2.7 V) at a scan speed of 100 mV/s (a), at different scan rates from 10-500 mV/s (b); GCD for 2.3 V at different current density from 0.7 -15 A/g (c).

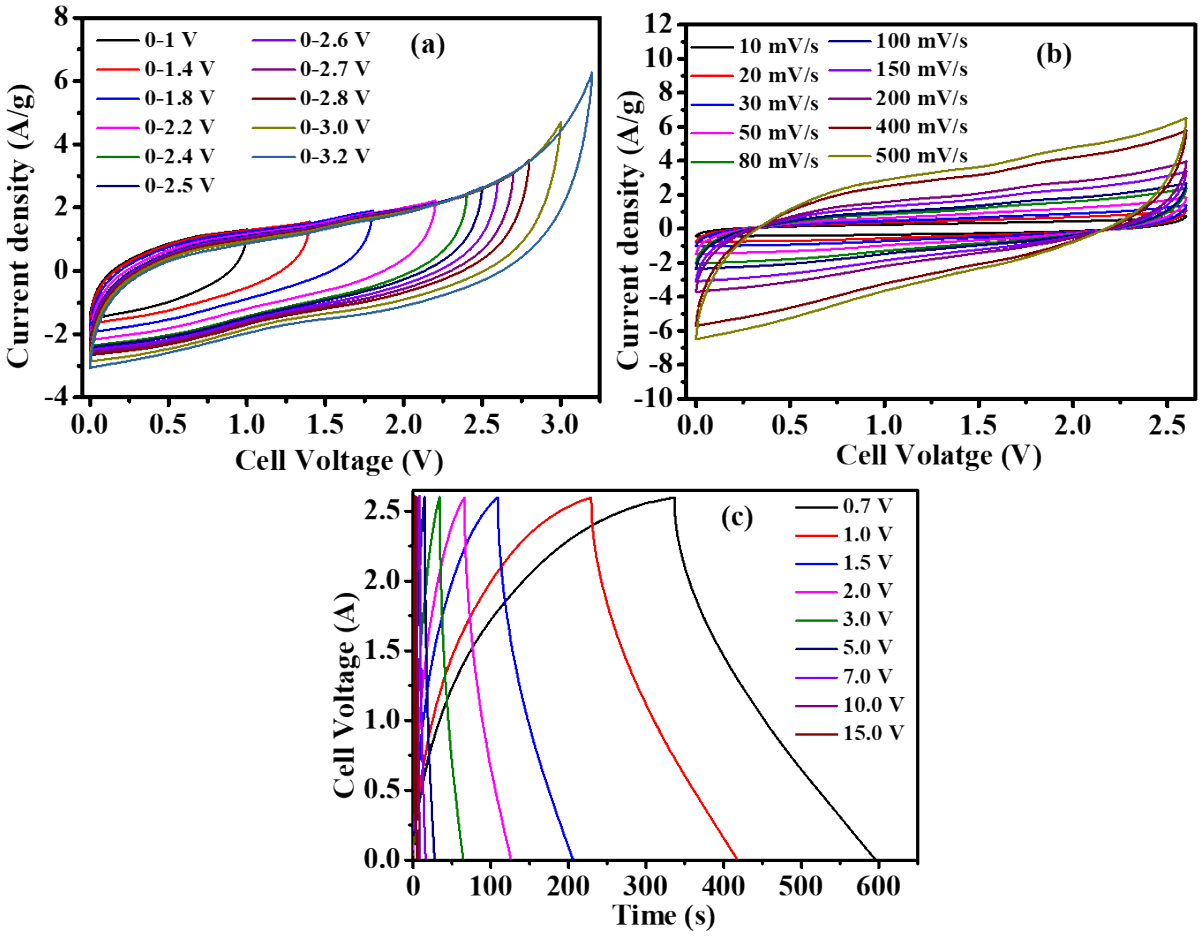


Fig. S16. CV measurements made for the SSC (using N, P-rGO as an electrode material) in the 0.5 m K_2SO_4 electrolyte: for a varied range of cell voltages (1-3.2 V) at a scan speed of 100 mV/s (a), at different scan rates from 10-500 mV/s (b); GCD for 2.6 V at different current density from 0.7 -15 A/g (c).

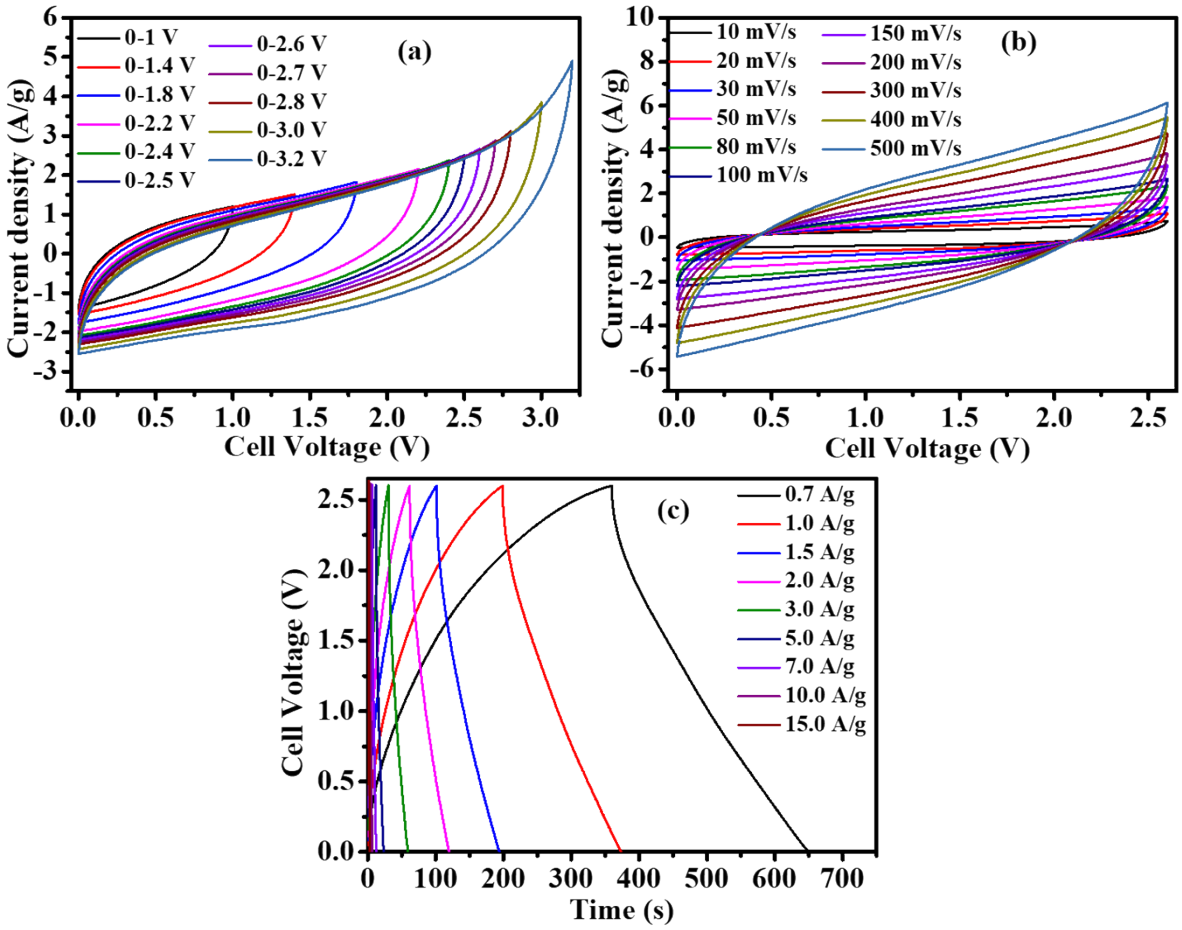


Fig. S17. CV measurements made for the SSC (using N, P-rGO as an electrode material) in the 2 m Na_2SO_4 electrolyte: for a varied range of cell voltages (1-3.2 V) at a scan speed of 100 mV/s (a), at different scan rates from 10-500 mV/s (b); GCD for 2.6 V at different current density from 0.7 -15 A/g (c).

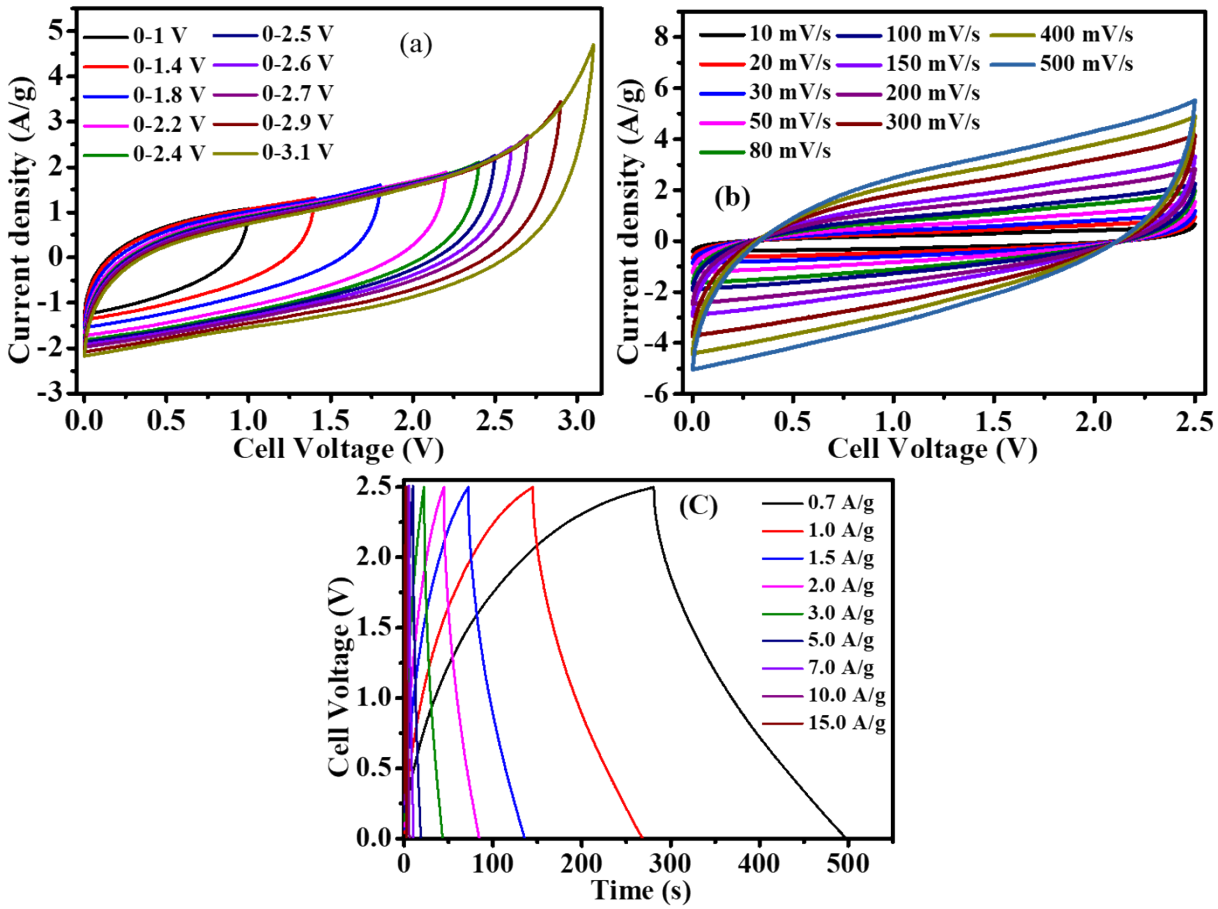


Fig. S18. CV measurements made for the SSC (using N, P-rGO as an electrode material) in the 0.5 m Na_2SO_4 electrolyte: for a varied range of cell voltages (1-3.1 V) at a scan speed of 100 mV/s (a), at different scan rates from 10-500 mV/s (b); GCD for 2.5 V at different current density from 0.7 -15 A/g (c).

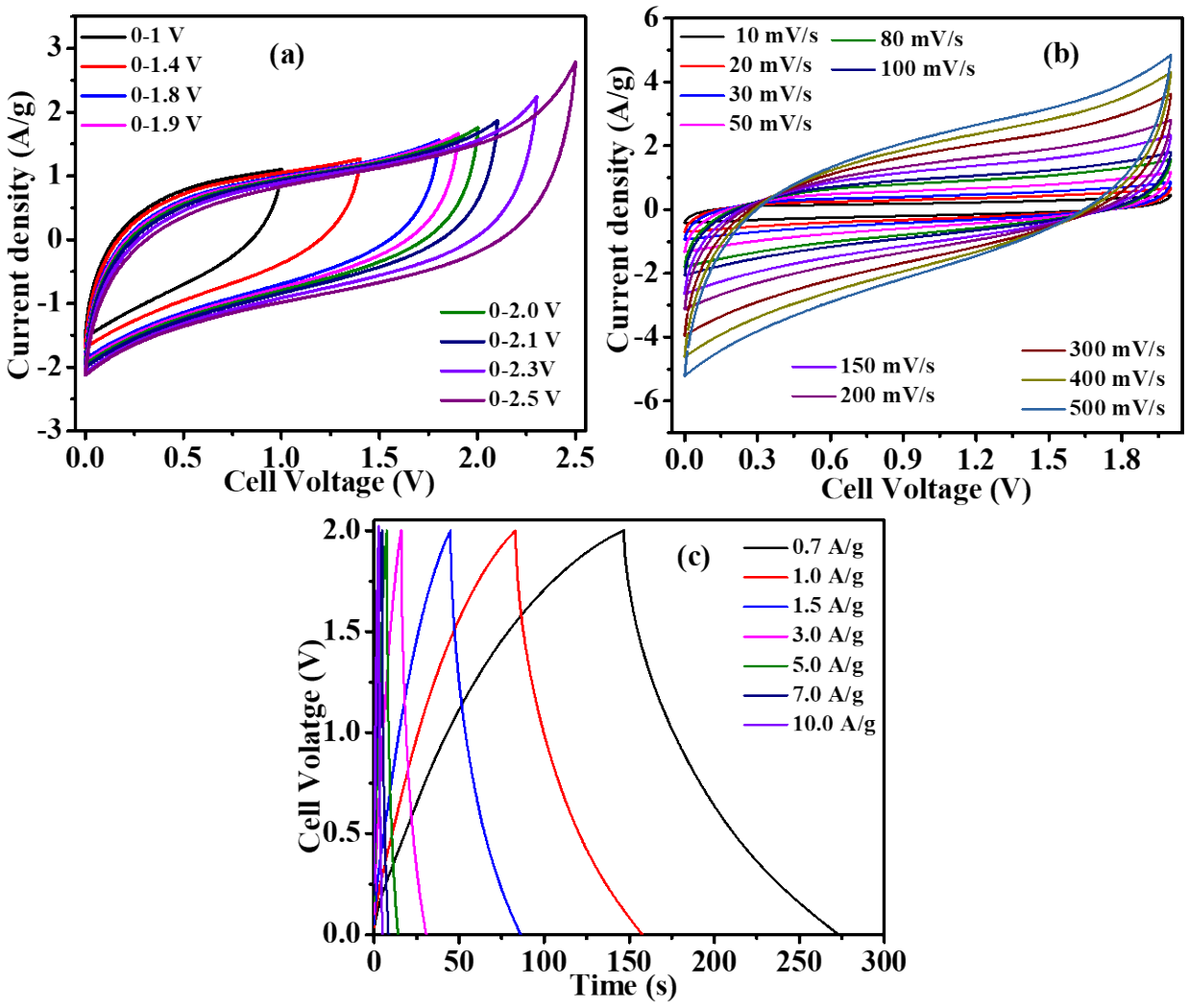


Fig. S19. CV measurements made for the SSC (using N, P-rGO as an electrode material) in the 0.1 m $KClO_4$ electrolyte: for a varied range of cell voltages (1-2.5 V) at a scan speed of 100 mV/s (a), at different scan rates from 10-500 mV/s (b); GCD for 2.0 V at different current density from 0.7 -10 A/g (c).

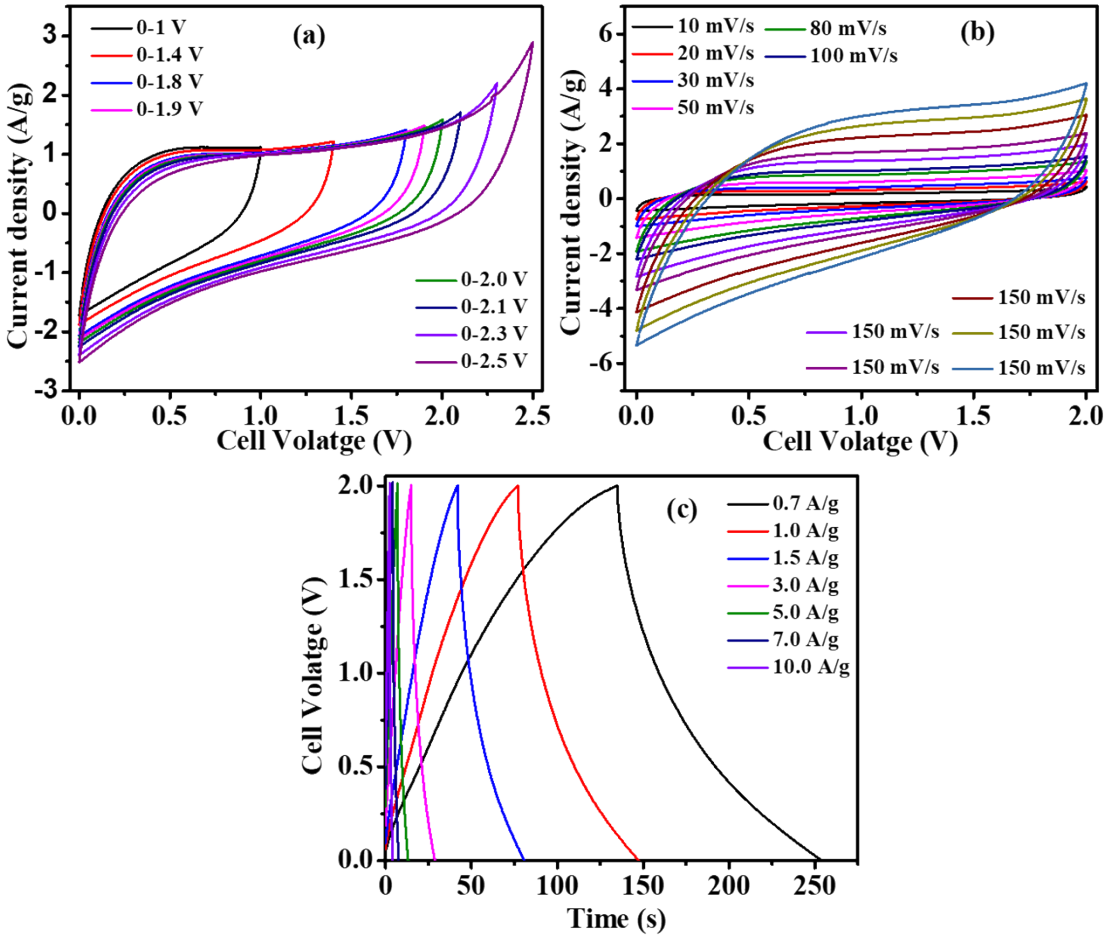


Fig. S20. CV measurements made for the SSC (using N, P-rGO as an electrode material) in the 0.1 m $NaClO_4$ electrolyte: for a varied range of cell voltages (1-2.5 V) at a scan speed of 100 mV/s (a), at different scan rates from 10-500 mV/s (b); GCD for 2.0 V at different current density from 0.7 -10 A/g (c).

Table S3. ATR-FTIR data obtained for DIW and electrolytes at their maximum solubility: CH_3COONa (7 m), $NaClO_4$ (17 m), $NaNO_3$ (11 m), KNO_3 (3 m), K_2SO_4 (0.5 m), and Na_2SO_4 (2 m).

Electrolytes	**Ice-like component (cm^{-1})**	**Ice-like liquid component (cm^{-1})**	**Monomeric water (cm^{-1})**
DIW	3235	3380	3590
K_2SO_4 (0.5 m)	3238	3382	3591
Na_2SO_4 (2 m)	3237	3378	3589
KNO_3 (3 m)	3241	3384	3590
CH_3COONa (7 m)	3327	3378	3591
$NaNO_3$ (11 m)	3249	3421	3590
$NaClO_4$ (17 m)	3246	3429	3587

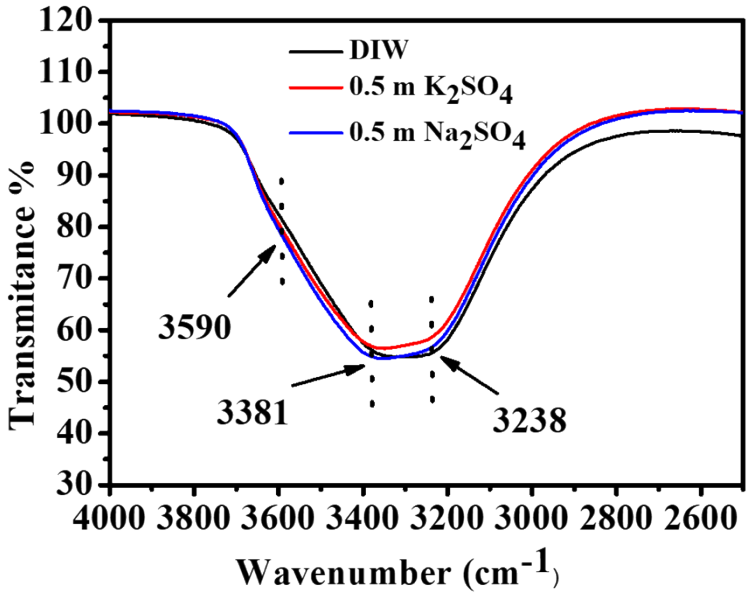


Fig. S21. Extended ATR−FTIR spectra of DIW, 0.5 m Na_2SO_4 and 0.5 m K_2SO_4.

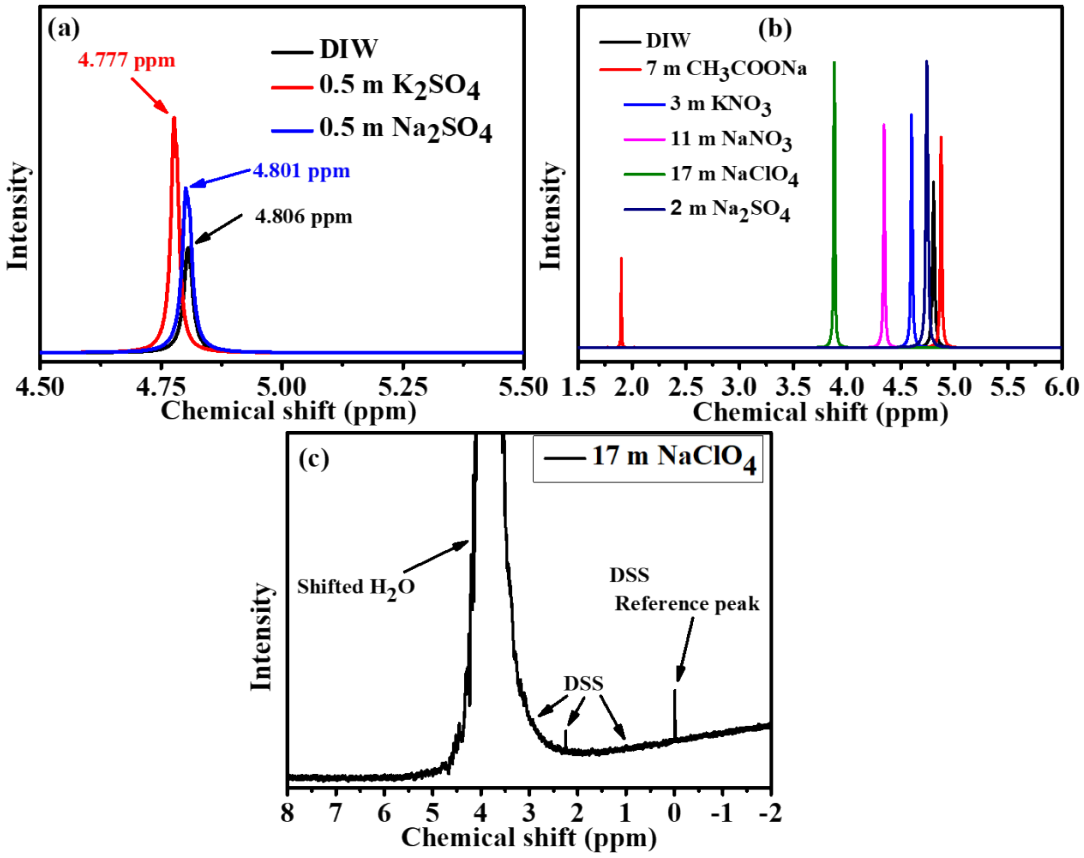
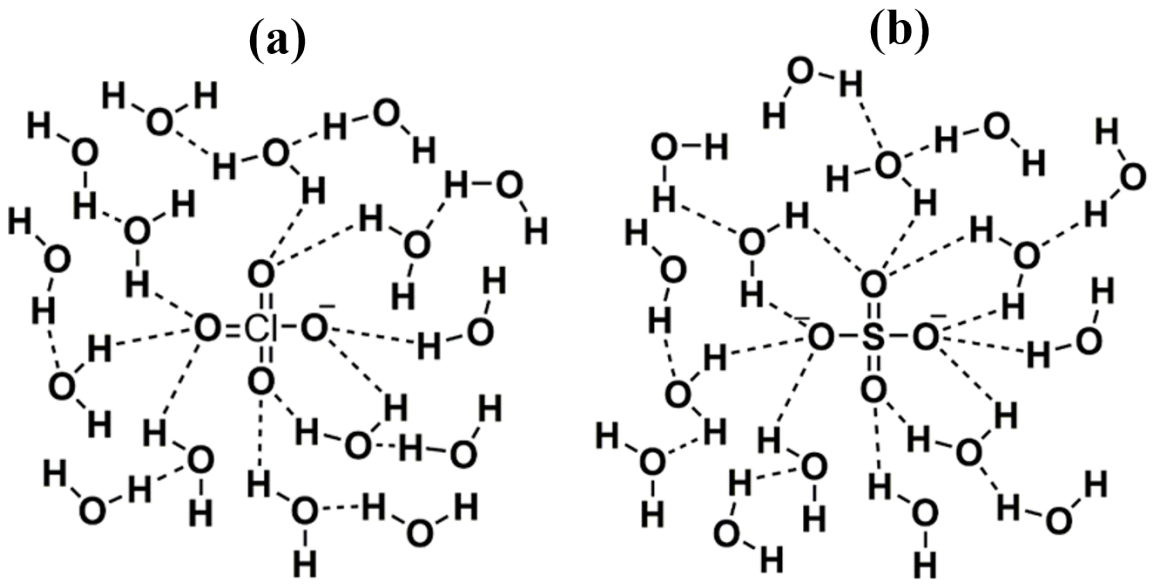


Fig. S22. ^{1}H NMR spectra in extended chemical shift range for: DIW, 0.5 m Na_2SO_4 and 0.5 m K_2SO_4 (a); DIW, 7 m CH_3COONa (satd.), 3 m KNO_3, 11 m $NaNO_3$, 17 m $NaClO_4$, and 2 m Na_2SO_4 (b); 17 m $NaClO_4$ to show the DSS reference peaks.



Scheme S1. The representation of $ClO_4^-(H_2O)_{16}$ cluster (a); and $SO_4^{2-}(H_2O)_{16}$ (b)**.**

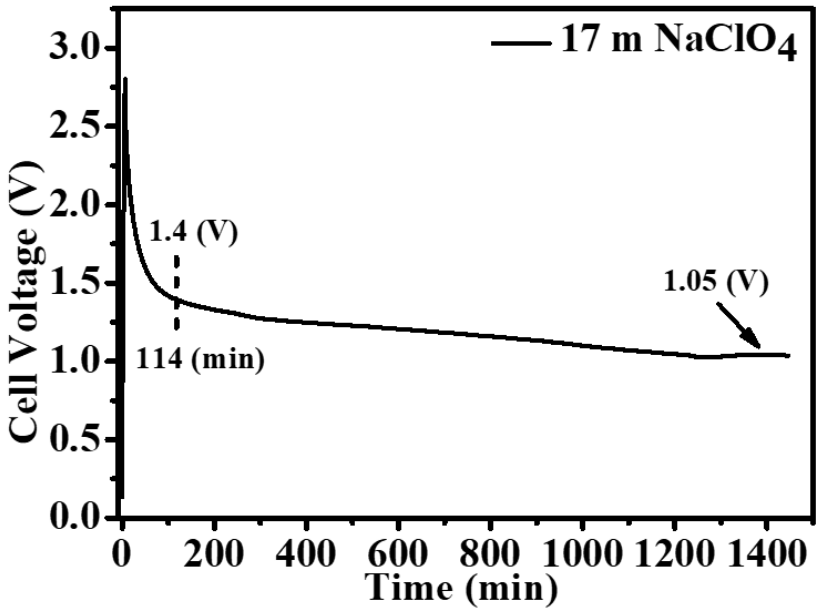


Fig. S23. Self-discharging of N, P-rGO in 17 m $NaClO_4$ electrolyte at a current density of 1 A/g.

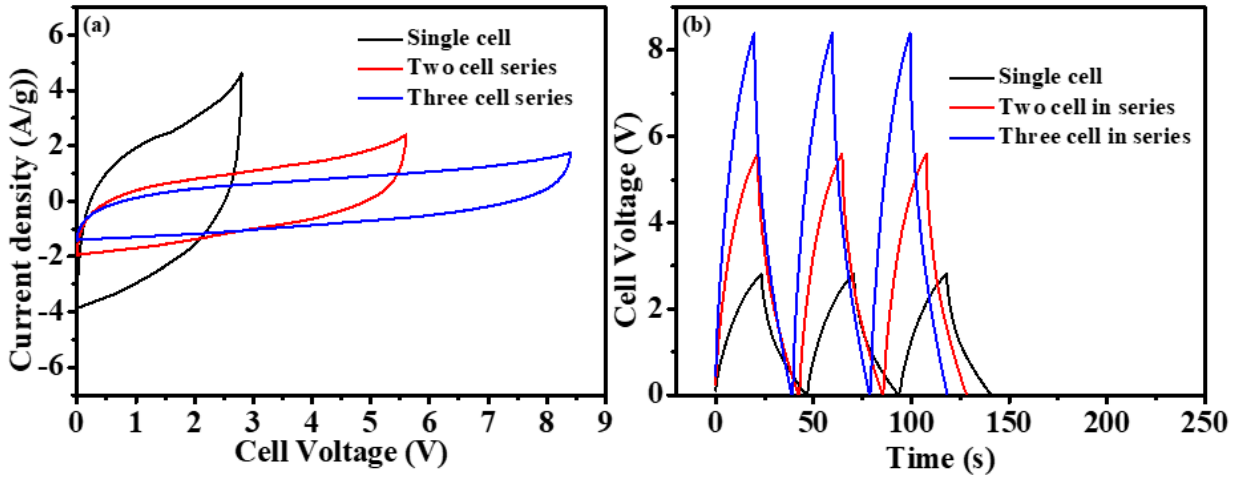


Fig. S24. Cyclic voltammograms recorded for N, P-rGO for single cell and by constructing tandem device by joining two and three SSC in series in 17 m $NaClO_4$ electrolyte at a scan rate of 100 mV/s (b); GCD curves recoded for the single cell and tandem device(s) in 17 m $NaClO_4$ at a current density of 5 A/g.

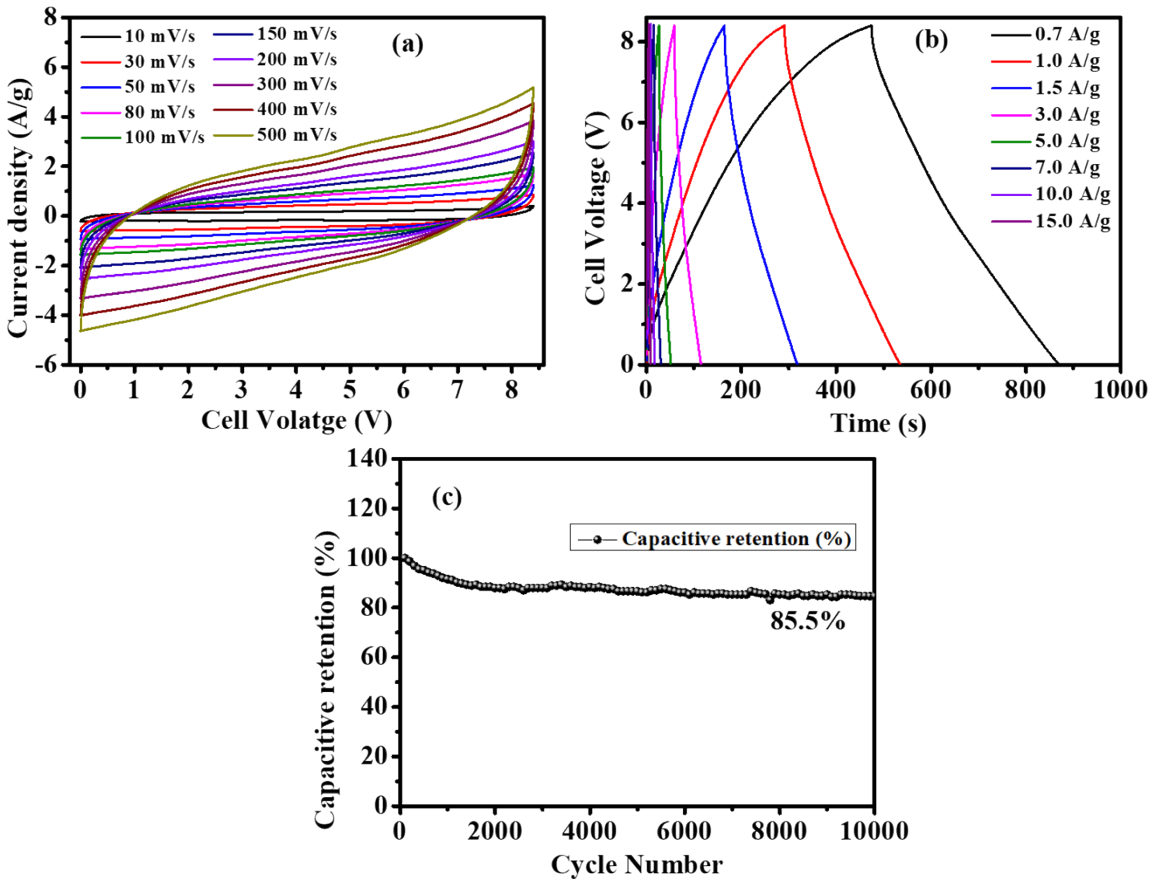


Fig. S25. CV curves of tandem device constructed by joining three SSC in series of N, P-rGO in 17 m $NaClO_4$ electrolyte recorded at scan rates varying from10- 500 mV/s (a); GCD curves of tandem device in 17 m $NaClO_4$ at varied current densities of 0.7 - 15 A/g (b); Cyclic stability of a tandem device of the as-fabricated symmetric cell at 5 A/g (c).

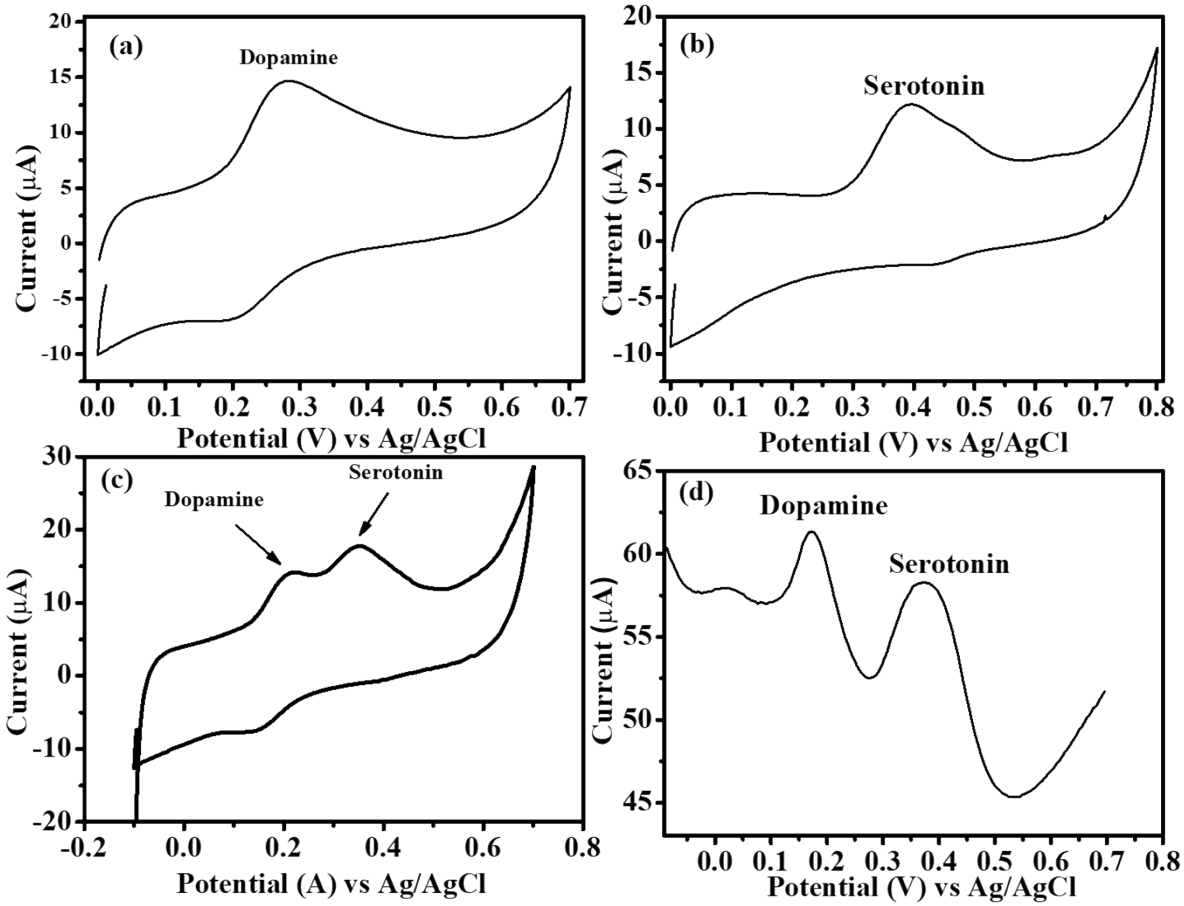


Fig. S26. CV curves of 50 μM Dopamine (a), 50 μM Serotonin (b) CV and DPV Simultaneous detection of Dopamine and serotonin respectively in 7.0 pH phosphate buffer (0.1 M).

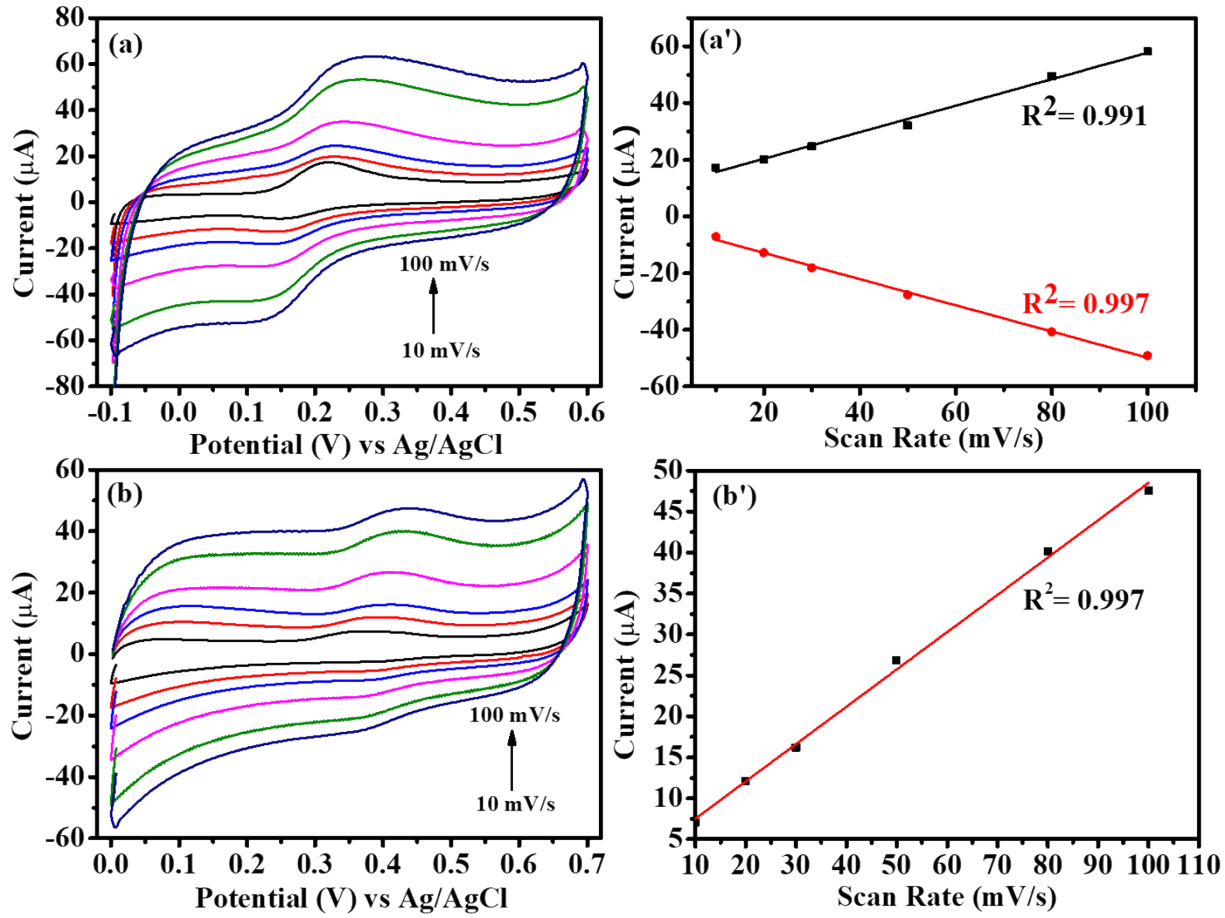
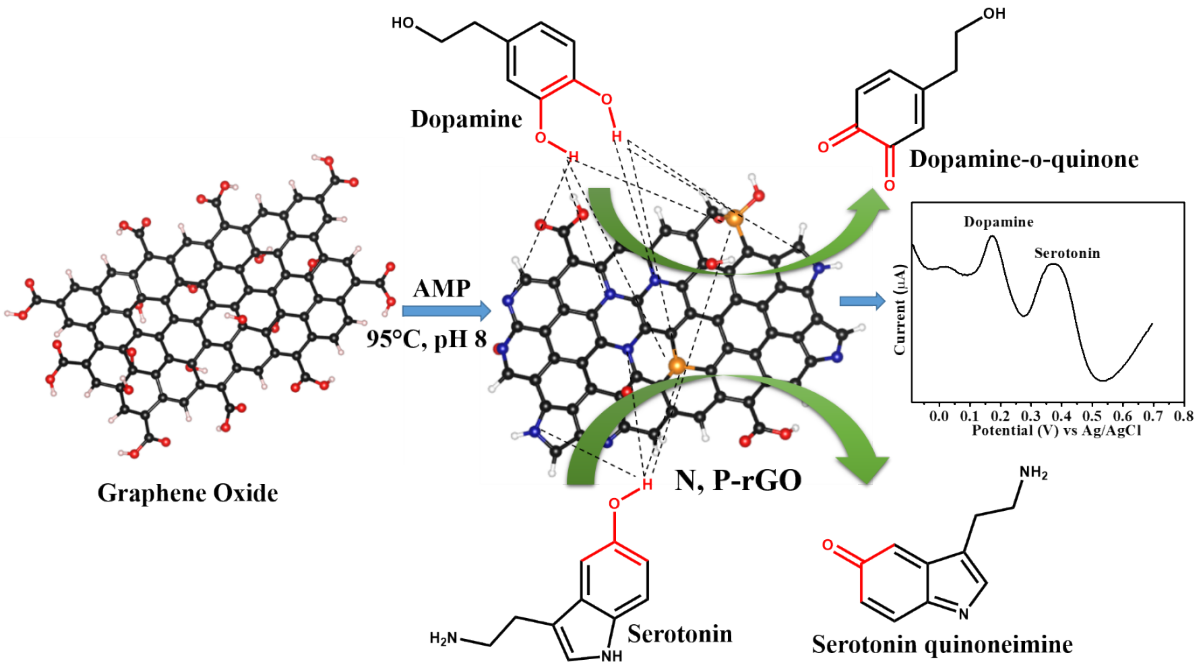
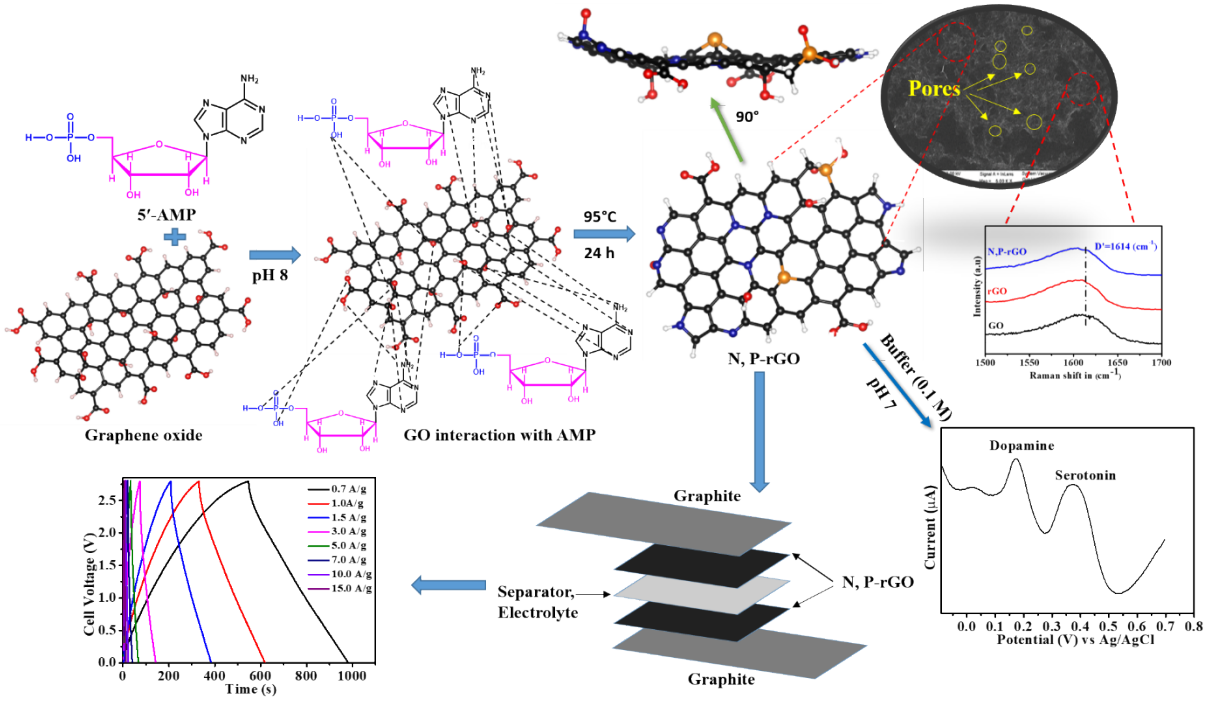


Fig. S27. CV curves of 50 μM Dopamine (a), 30 μM Serotonin (b) and corresponding current vs concentration plot (a′) (b′) respectively in 7.0 pH phosphate buffer (0.1 M).



Black = Carbon, Red = Oxygen, Blue = Nitrogen, Yellow = Phosphorous and White = Hydrogen

Scheme S2. Schematic representation of simultaneous sensing of Dopamine and Serotonin at the interface of N-P-rGO electrode.



Black = Carbon, Red = Oxygen, Blue = Nitrogen, Yellow = Phosphorous and White = Hydrogen

Scheme S3. Schematic illustration representing the synthesis of 5′-AMP-mediated porous N, P-rGO nanohybrids and their use for investigating different electrochemical aspects.

Table S4. A comparison of the present work with the literature reports with respect to supercapacitor behavior.

Materials	Conducting agent/ Binder	Weight	Voltage Range (V)	Electrolyte	Energy Density (W h kg^{-1})	Power Density (W kg^{-1})	Cycle stability (current density)	Ref.
N, P-co-doped porous carbon	acetylene black/ *PVDF	2 mg	1	6 M KOH	26.289 (0.5 Ag^{-1})	Maximum 3694.084	10000, 94.2% (10 A g^{-1})	10
N/P-co-doped Porous Carbon-Coated Graphene (KNPG)	Super P/ *PTFE	1.5 mg/ electrode	1.4	6 KOH	9.1	350	20000, 84.6% (5 A g^{-1})	11
Nitrogen, Phosphorus-co-doped Carbon (NPC-800-2)	carbon black/ PTEF	2 mg	1.8	1 M Na_2SO_4	21.5	250	100000, (2 A g^{-1})	12
N, P-CQDs/rGO	acetylene black/ PTFE	Coin used	1.3	6 M KOH	15.69	325	10000, 85.5% (5 A g^{-1})	13
N/P-G-3	carbon black/ PVDF	1.5x1 cm^{-1} thin film	1.6	1 M H_2SO_4	11.33	571	10000, 94% (5 A g^{-1})	14
(NP-rGO)	acetylene black /PVDF	3 mg	1	6 M KOH	22.3	500	10000, 94.63%	15
N, P-co-doped hierarchical porous carbon	-/ PTFE	coin-cells	1	1 M Na_2SO_4	21	50	10000, 95%	16
Tremella-like nitrogen and Phosphorus-co-doped graphene	acetylene black/ PTFE	1.25 mg	2	2 M Li_2SO_4	34.7	500	20000, 91.4% (5 A g^{-1})	17
NPHCMs-65-800	acetylene black/ PTFE	3 mg	1	KOH/PVA gel	6.4	0.1 k	5000, 91% (0.5 A g^{-1})	18
NP-PCNs-100	-	0.192 mg	1.6	1 M Na_2SO_4	17.04	400	10000, 90% (10 A g^{-1})	19
1NPHC-850//1NPHC-850	Super P/ PTFE	2 mg cm^{-1}	1.6	1 M Na_2SO_4	10.61	400	10000, 86.3% (5 A g^{-1})	20
N/P-co-doped graphene (NPG)	acetylene black (PTFE)	3 mg	1.3	6 KOH	8.17 (0.25Ag^{-1})	162	20000, 86.5% (10 A g^{-1})	21
5′-AMP-mediated-N, P co-doped rGO	-	2.2 mg	2.8	17 m $NaClO_4$	59.2	489.6	10000, 115.2% (10 A g^{-1})	This work
5′-AMP-mediated N, P co-doped rGO	-	5.2 mg	2.8	17 m $NaClO_4$	26.4	819.7	10000, 93.6% (1 A g^{-1})	This work

*PVDF: -Polyvinylidene fluoride *PTFE: -Polytetrafluoroethylene