Electronic Supplementary Information

Surface modification of co-doped reduced graphene oxide through alkanolamine functionalization for enhanced electrochemical performance

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Experimental Section

Methodology

For Raman measurements, a pinch of dried powdered sample was placed on a precleaned glass substrate beneath a 50× objective lens. All Raman spectra were recorded at a low laser power of 1 mW for an exposure time of 30 s in order to avoid any thermal damage to the samples. For FESEM analysis, a pinch of powdered sample was placed on the carbon tape, which is fixed on an aluminum stub. The surface of the samples was sputtered with platinum in order to make them conducting, and then the images were captured by applying an acceleration voltage of 15 - 20 kV. Samples for TEM analysis were prepared by dispersing a small amount of powdered sample in ethanol by ultrasonication for approximately 15-20 min, and then the copper grid was immersed in the dispersed solution for 1 s followed by its drying. For N_2 adsorption-desorption measurements, the samples were degassed at 180 °C for approximately 12 h under vacuum prior to analysis. The specific surface area was analyzed using the Brunauer–Emmett–Teller (BET) equation employing NovaWin software. pore size of the samples was determined by applying Barrett-Joyner-Halenda (BJH) method to the desorption branch of the N_2 sorption isotherm. Total pore volume was obtained at the relative pressure of $P/P_0 = 0.99$. The obtained dried powder samples were employed as such for the XRD, FTIR, XPS and BET measurements.

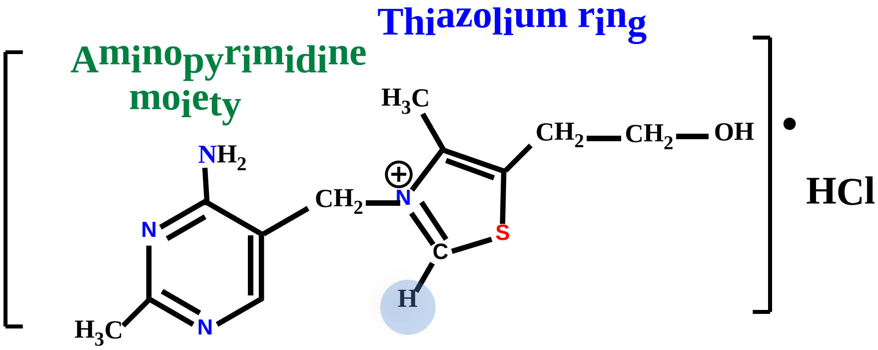
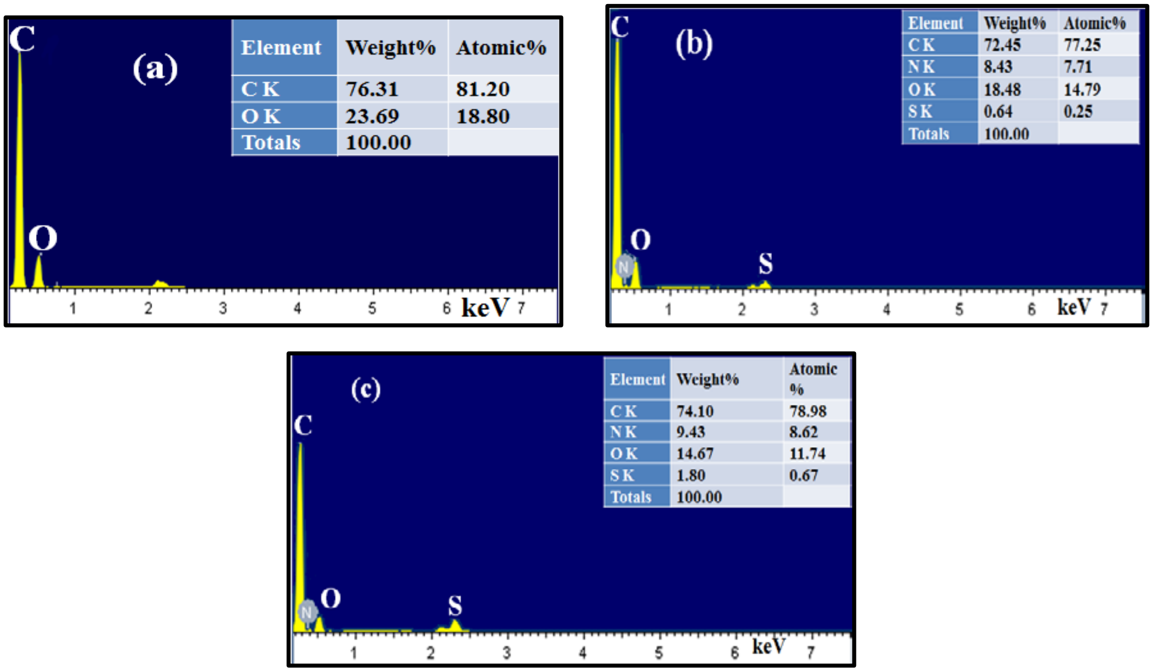


Fig. S1 Structure of thiamine hydrochloride.



(a)

Element	Weight%	Atomic%
C K	76.31	81.20
O K	23.69	18.80
Totals	100.00	

(b)

Element	Weight%	Atomic%
C K	72.45	77.25
N K	8.43	7.71
O K	18.48	14.79
S K	0.64	0.25
Totals	100.00	

(c)

Element	Weight%	Atomic %
C K	74.10	78.98
N K	9.43	8.62
O K	14.67	11.74
S K	1.80	0.67
Totals	100.00	

Fig. S2 EDAX spectra of rGO (a) NSrGO (b) and TEA-NSrGO (c) along with their elemental composition given in inset.

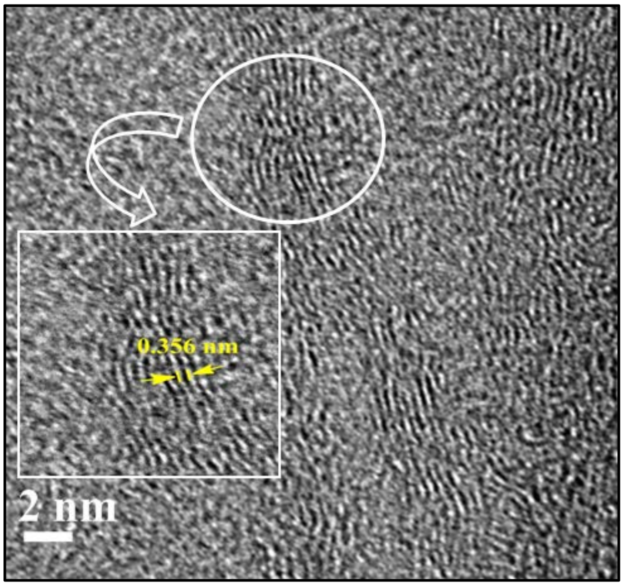


Fig. S3 HRTEM image of NSrGO.

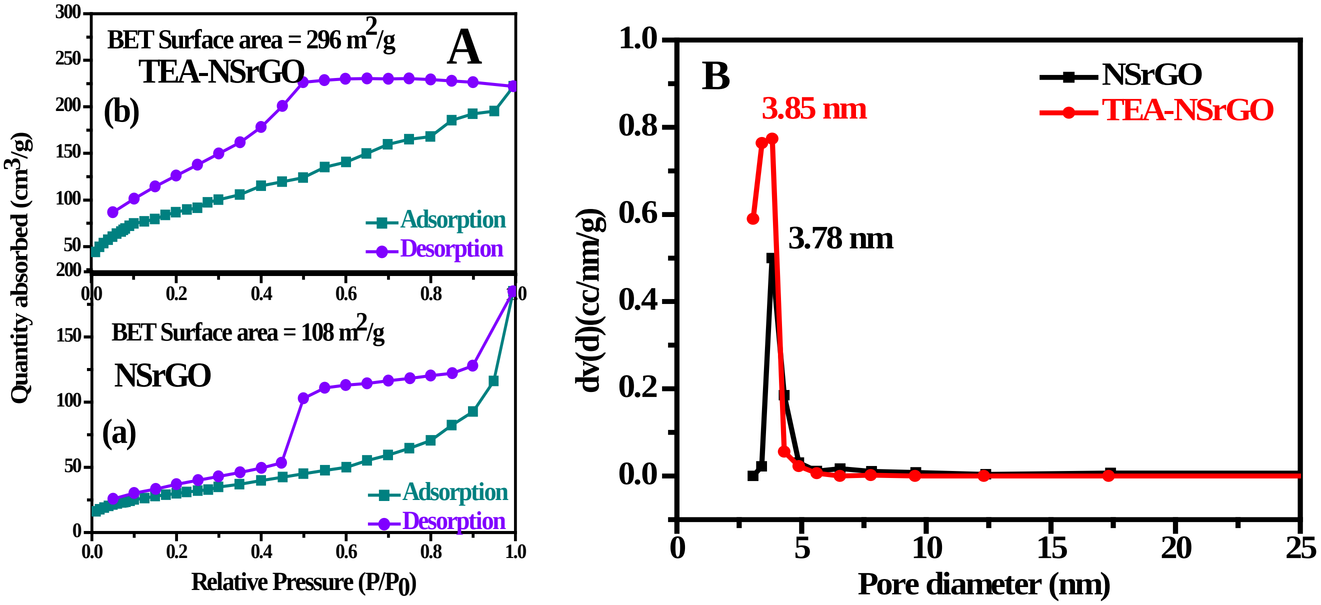


Fig. S4 Nitrogen sorption isotherms of NSrGO (a) and TEA-NSrGO (b) – (Panel A). The pore size distribution curves of: NSrGO and TEA-NSrGO obtained by employing BJH method – (Panel B).

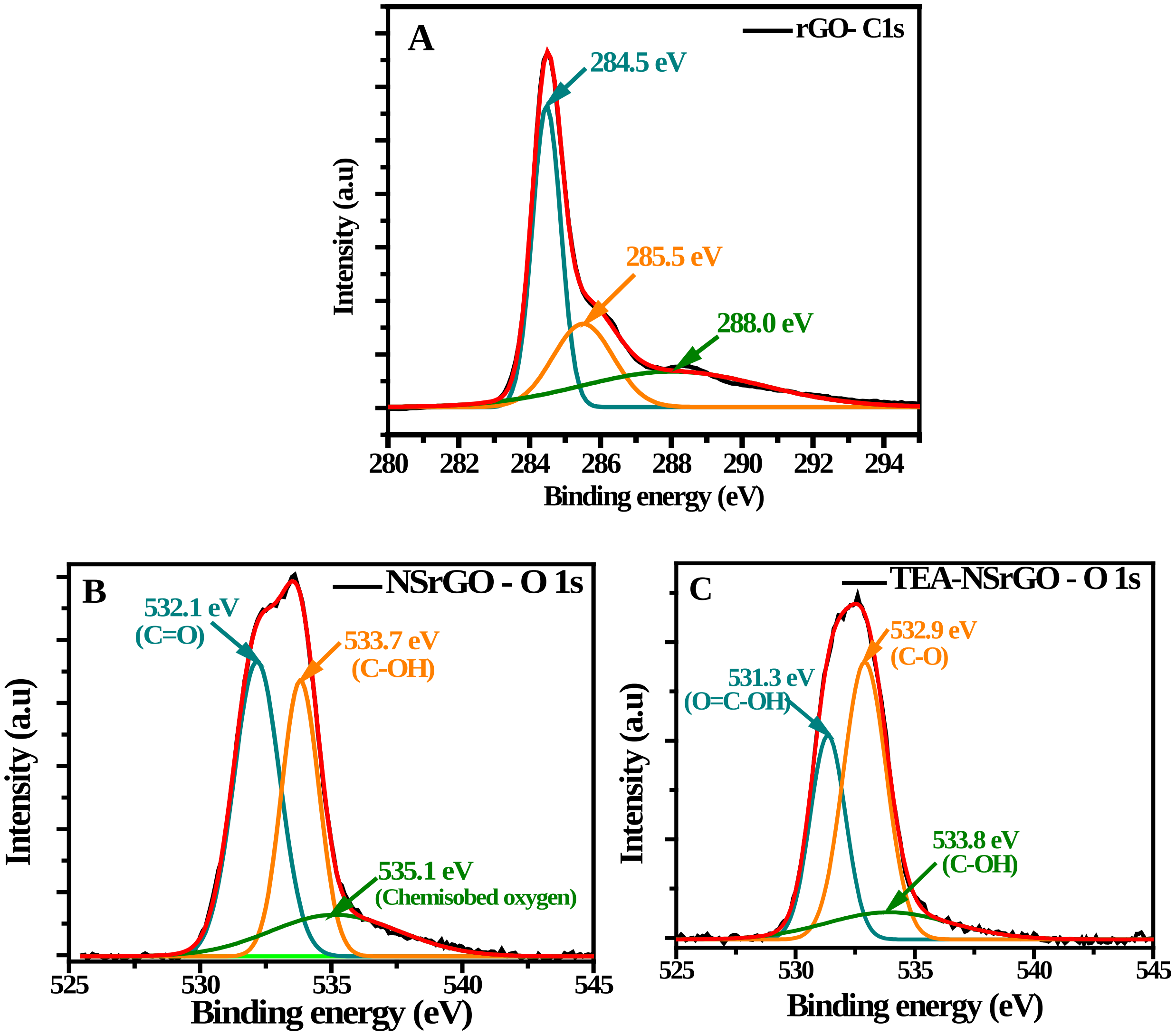


Fig. S5 High resolution C 1s spectrum of reduced graphene oxide (rGO) – (panel A). High resolution O 1s spectra of NSrGO - (Panel B) and TEA-NSrGO – (Panel C).

The O1s spectrum of NSrGO shows three different components of carbon at binding energies (eV) of 532.1, 533.7 and 535. 1 which have been attributed to carbonyl (C=O), hydroxyl (C-OH) and chemisorbed oxygen, respectively. In case of TEA-NSrGO, the O 1s spectrum also shows three different peaks at binding energies (eV) of 531.3, 532.9 and 533.8 which have been assigned to carboxyl (O=C-OH), carbon single bonded to oxygen (C-O) and hydroxyl (C-OH), respectively [S1]. The close examination of the deconvoluted O 1s spectrum of TEA-NSrGO clearly reveals the high intensity of peak corresponding to carbon single bonded to oxygen (C-O) suggesting the functionalization of hydrophilic TEA molecules on the surface of NSrGO.

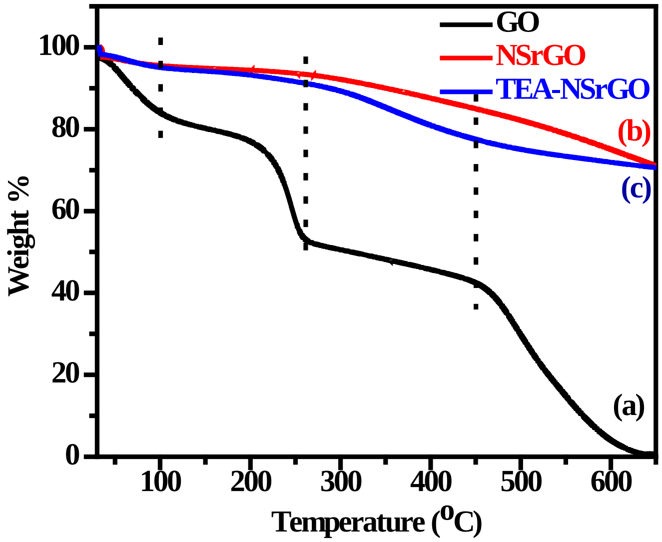


Fig. S6 TGA curves of GO (a), NSrGO (b) and TEA-NSrGO (c).

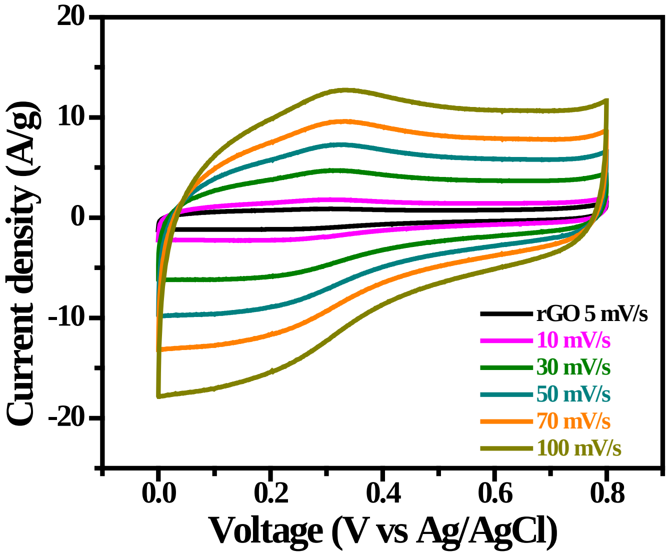


Fig. S7 CV curves of rGO at different scan rates varying from 5 – 100 mV/s.

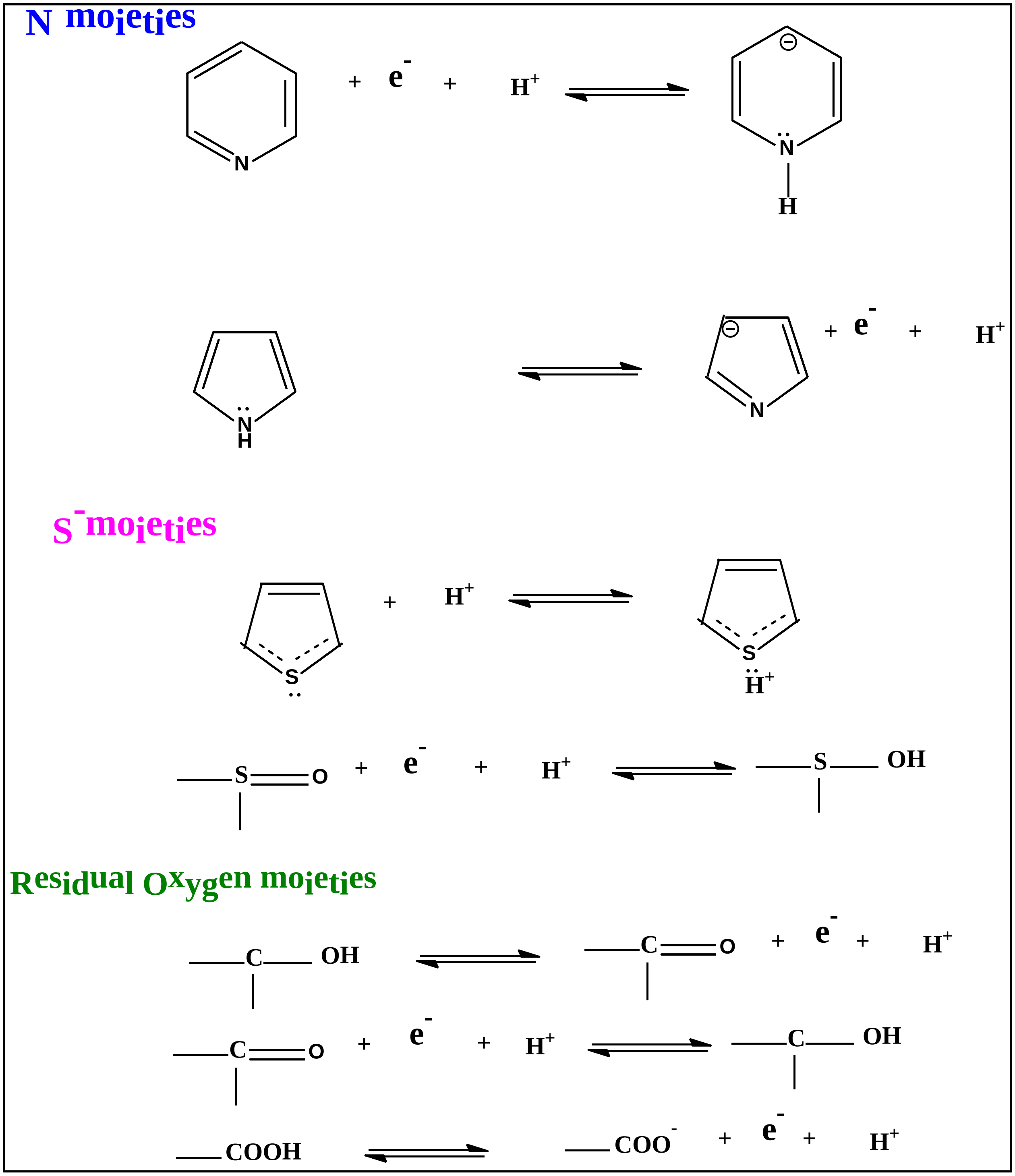


Fig. S8 The various possible bonding configurations of N, S and O in rGO, NSrGO and TEA-NSrGO which are taking part in faradaic redox reaction with H^+ of electrolyte.

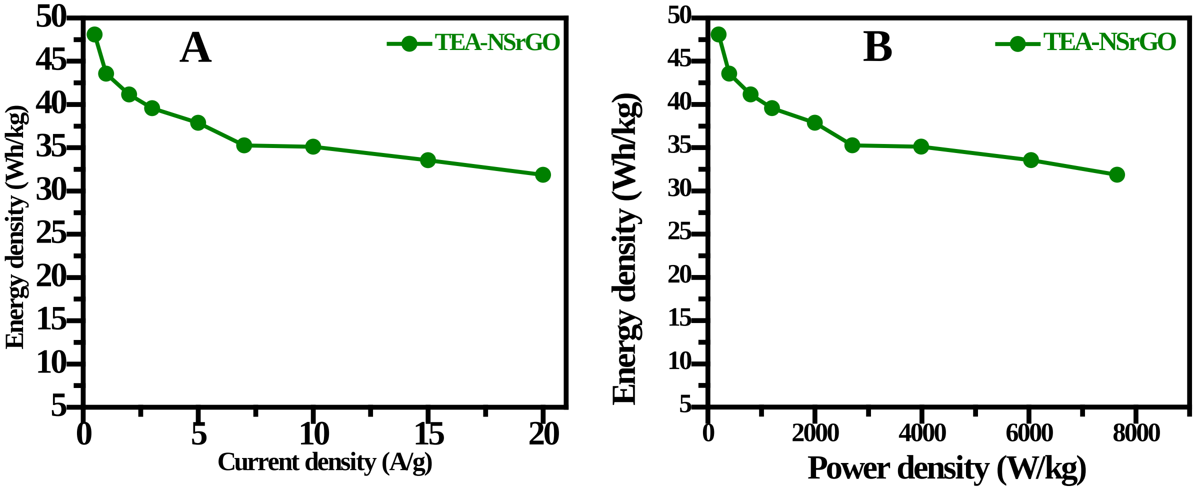


Fig. S9 Energy density variation of TEA-NSrGO electrode material as a function of current density – (panel A). Ragone plot of TEA-NSrGO – (panel B).

Table S1 Raman spectral data of GO, rGO, NSrGO and TEA-NSrGO.

Sample(s)	**D Band (cm^{-1})**	**G Band (cm^{-1})**	**I_D/I_G ratio**
GO	1355	1604	**0.86**
rGO	1359	1609	**0.89**
NSrGO	1358	1602	**0.91**
TEA-NSrGO	1354	1610	**0.96**

Table S2 BET surface area, total pore volume and pore diameter of NSrGO and TEA-NSrGO.

Sample(s)	BET Surface Area (m^2/g)	Total Pore Volume (cm^3/g)	Pore diameter (nm)
NSrGO	108	0.2861	3.78
TEA-NSrGO	296	0.3433	3.85

Table S3 Atomic percentages of C, O, N and S present in rGO, NSrGO and NSrGO-TEA and their C/O ratios.

Sample(s)	Carbon (at.%)	Oxygen (at.%)	Nitrogen (at.%)	Sulphur (at.%)	C/O ratio
rGO	83.01	16.99	-	-	4.89
NSrGO	82.13	14.08	2.95	0.84	5.83
TEA-NSrGO	80.49	12.72	5.40	1.39	6.33

Table S4 Specific capacitance (C_s) values of un-doped rGO, NSrGO and TEA-NSrGO obtained from CV and GCD cures at a scan rate of 5 mV/s and current density of 1 A/g, respectively using 1 M H_2SO_4 as an aqueous electrolyte.

Sample(s)	C_s in F/g (5 mV/s)	C_s in F/g (1 A/g)
Un-doped rGO	144	121
NSrGO	231	237
TEA-NSrGO	465	490

Supplementary Reference

S1. Y. J. Oh, J. J. Yoo, Y. Il Kim, J. K. Yoon, H. N. Yoon, J.- H. Kim and S. B. Park, *Electrochim. Acta*, 2014, **116**, 118.