Electronic Supplementary Information for

Redox-Mediated 3D Graphene based nanoscoop design for Ultracapacitor Applications

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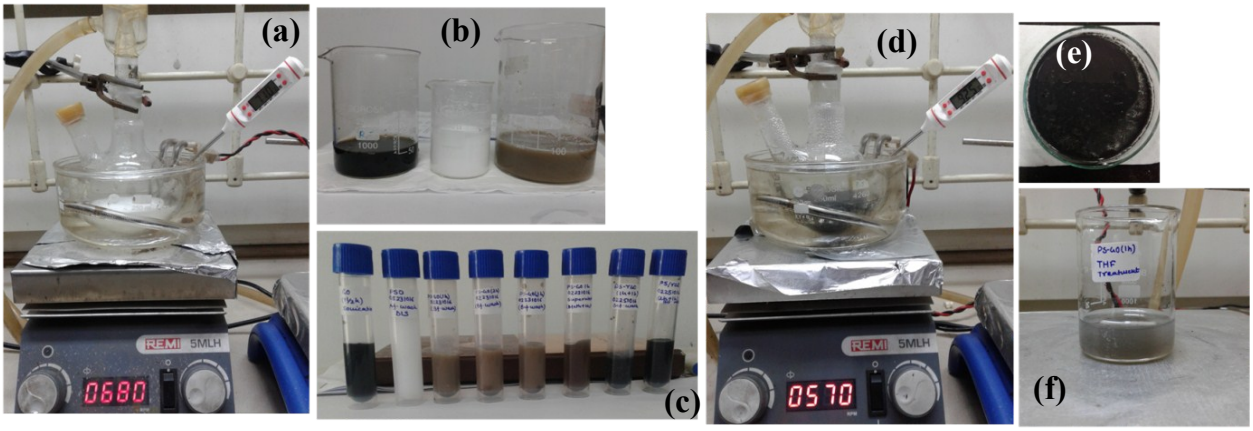
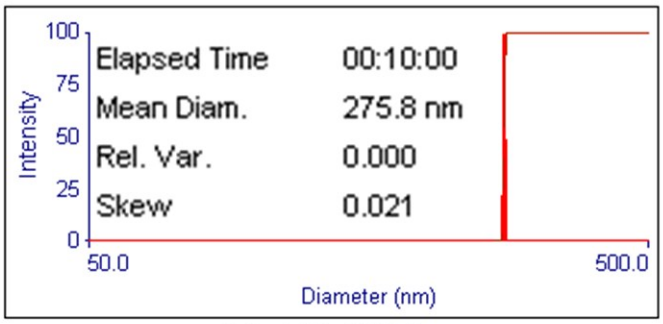


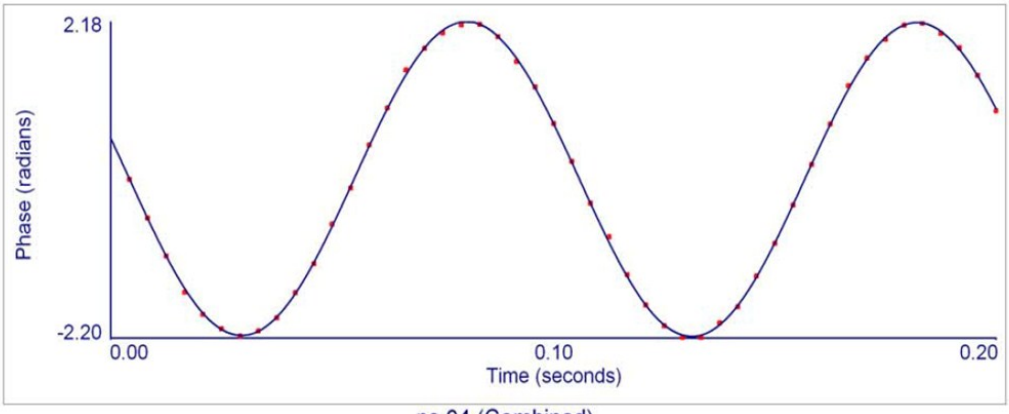
Fig. S1: Photos of setup for synthesis of polystyrene beads (a) GO suspension and PS colloidal solution (b) sample vial with PS-GO and PS-rGO solution with different reaction time (c) synthesis of PS-rGO composites (d) obtained PS-rGO powder product (e) treatment of PS-rGO powder with THF



Multimodal Size Distribution

d(nm)	G(d)	C(d)	d(nm)	G(d)	C(d)	d(nm)	G(d)	C(d)
266.7	0	0	276.4	88	82	286.4	0	100
267.6	0	0	277.3	49	97	287.3	0	100
268.5	0	0	278.2	9	100	288.2	0	100
269.3	0	0	279.1	0	100	289.2	0	100
270.2	0	0	280.0	0	100	290.1	0	100
271.1	0	0	280.9	0	100	291.0	0	100
272.0	0	0	281.8	0	100	292.0	0	100
272.8	0	0	282.7	0	100	292.9	0	100
273.7	21	6	283.6	0	100	293.9	0	100
274.6	61	25	284.5	0	100	294.8	0	100
275.5	100	55	285.5	0	100	295.8	0	100

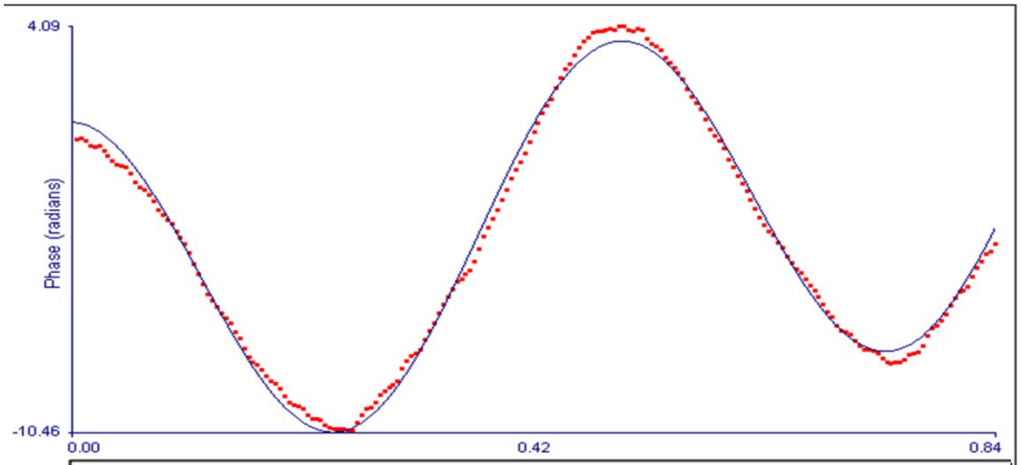
Fig. S2: Z-average particle size of polystyrene beads



ps 04 (Combined)

Run	Mobility	Zeta Potential (mV)	Rel. Residual
1	-3.00	-56.86	0.0123
2	-3.05	-57.79	0.0137
3	-2.97	-56.41	0.0126
4	-3.05	-57.91	0.0140
5	-2.96	-56.23	0.0127
6	-3.03	-57.43	0.0125
7	-2.99	-56.62	0.0144
8	-3.01	-57.05	0.0137
9	-2.97	-56.38	0.0115
10	-3.03	-57.53	0.0114
Mean	-3.01	-57.02	0.0129
Std. Error	0.01	0.19	0.0003
Combined	-3.01	-57.02	0.0036

Fig. S3: Zeta potential measurement of polystyrene colloidal particle (PS)



Run	Mobility	Zeta Potential (mV)	Rel. Residual
1	-2.86	-36.65	0.0390
2	-2.55	-32.60	0.0410
3	-2.76	-35.33	0.0346
4	-2.85	-36.49	0.0396
5	-2.95	-37.74	0.0358
6	-2.93	-37.49	0.0323
7	-3.06	-39.15	0.0177
8	-3.50	-44.84	0.0270
9	-2.92	-37.41	0.0169
10	-3.22	-41.23	0.0159
Mean	-2.96	-37.89	0.0300
Std. Error	0.08	1.05	0.0031
Combined	-2.95	-37.76	0.0184

Fig. S4: Zeta potential measurement of Graphene oxide (GO)

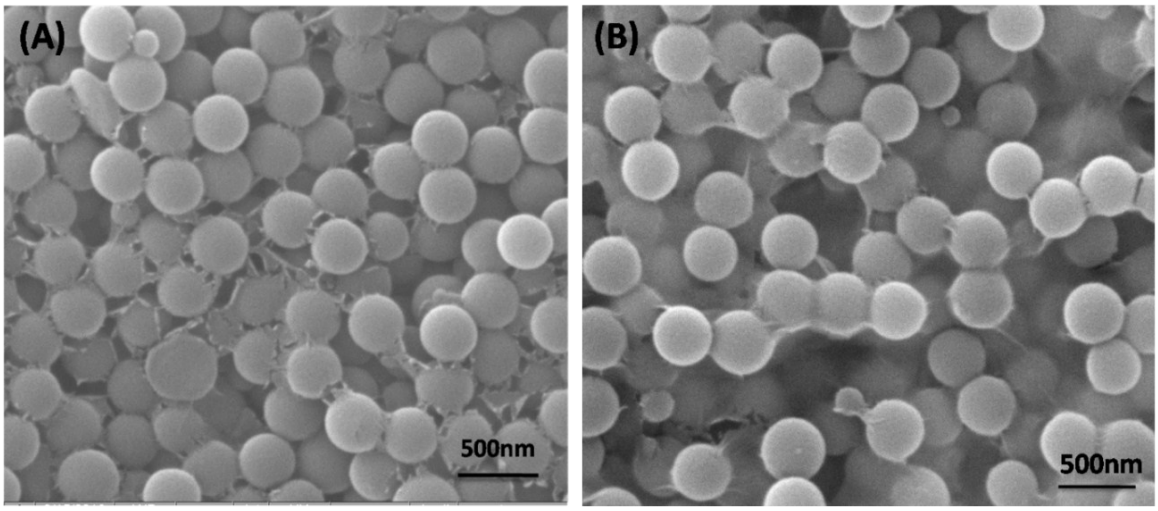


Fig. S5: SEM images of displaying PS-GO interaction in (A) 1h (B) 2h

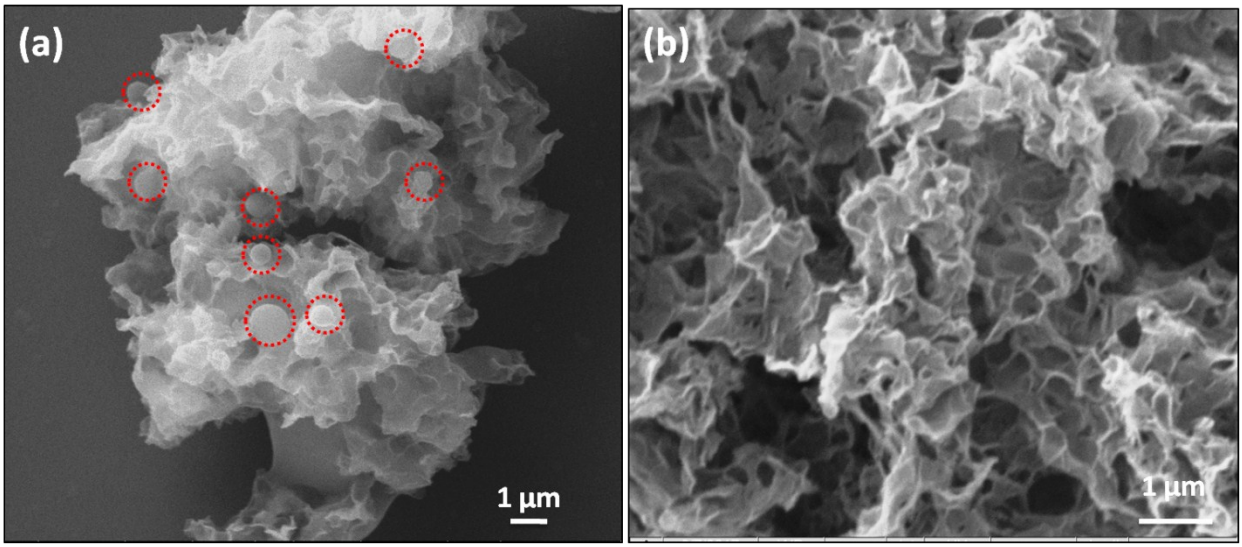


Fig. S6: SEM image of 3D graphene nanoscoop prepared after (a) THF treatment and (b) thermal treatment

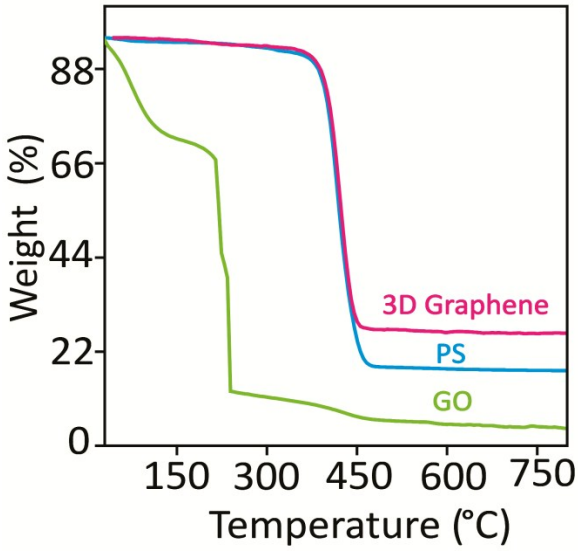


Fig.S7: TG of GO, PS and 3D Graphene.

As shown in Fig.S6, TG curve of GO shows significant weight losing events. The first event at around 110°C is related to the absorbed water while the second significant loss around 250°C is attributed to the decomposition of oxygen-containing functional group, yeiding CO, CO_2 and steam. The PS spheres start losing weight at 350°C and were burned completely at around 450°C. However, as expected, the PS and graphene shell began to decompose significantly at a temperature of about 450°C. Therefore, to retained spherical shell, heat treatment at a temperature of 420°C under heating condition (air,heating rate 5°C per min, held at 420°C for 2 h) was required.

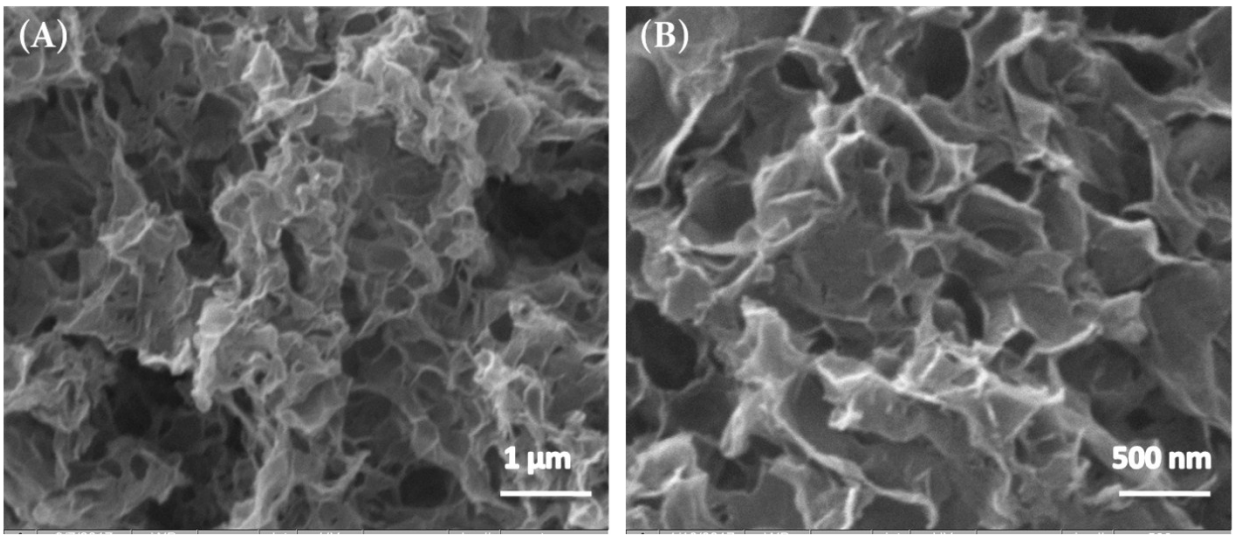


Fig. S8: SEM image of 3D graphene showing the effect of increasing heat rate with curvature during thermal treatment (A) 5˚C/ min (B) 30˚C/min

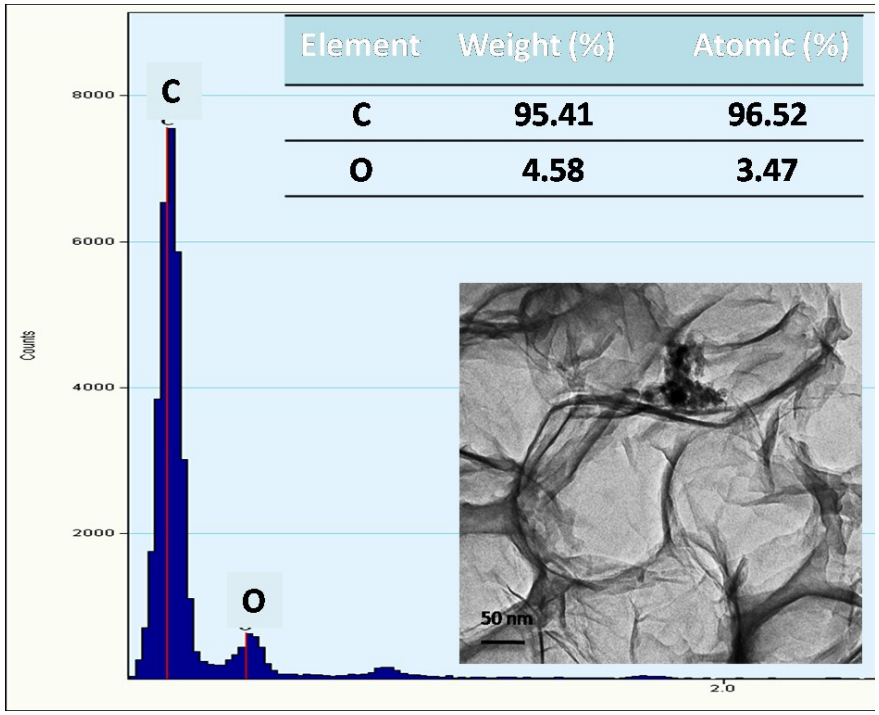


Fig. S9: EDS Spectra of 3D graphene representing atomic percentage of elements present

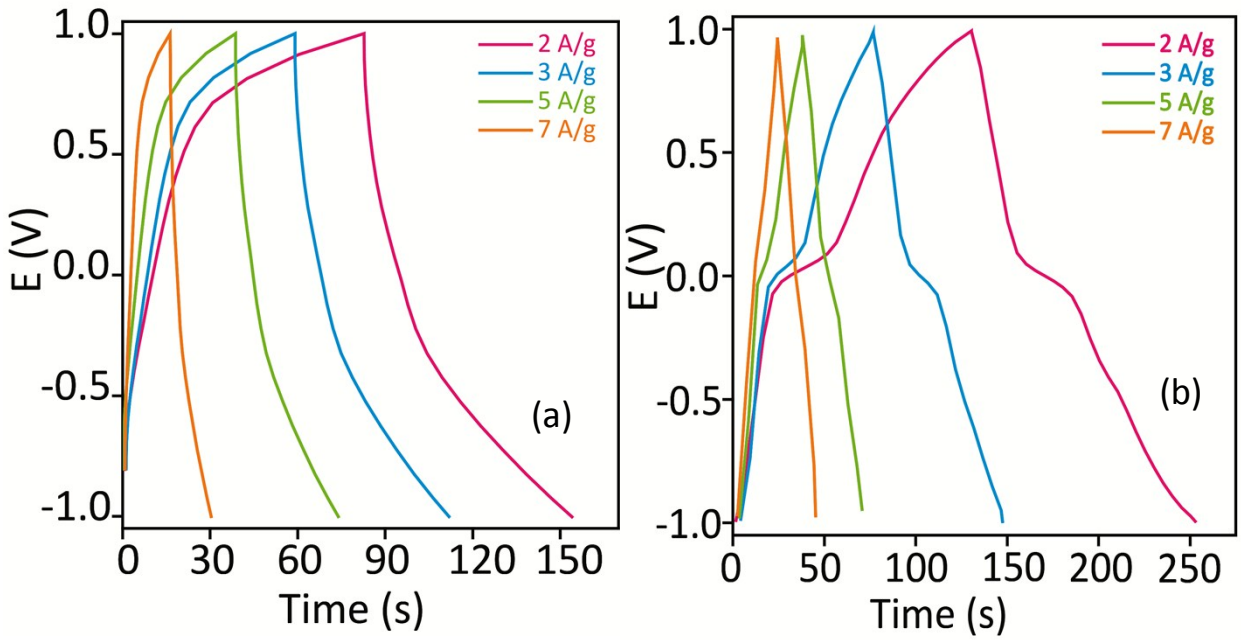


Fig. S10: Charge-discharge curve of rGO at different current densities in (a) 1M KOH (b) 1M KOH + 0.08 M $K_3Fe(CN)_6$ electrolytes

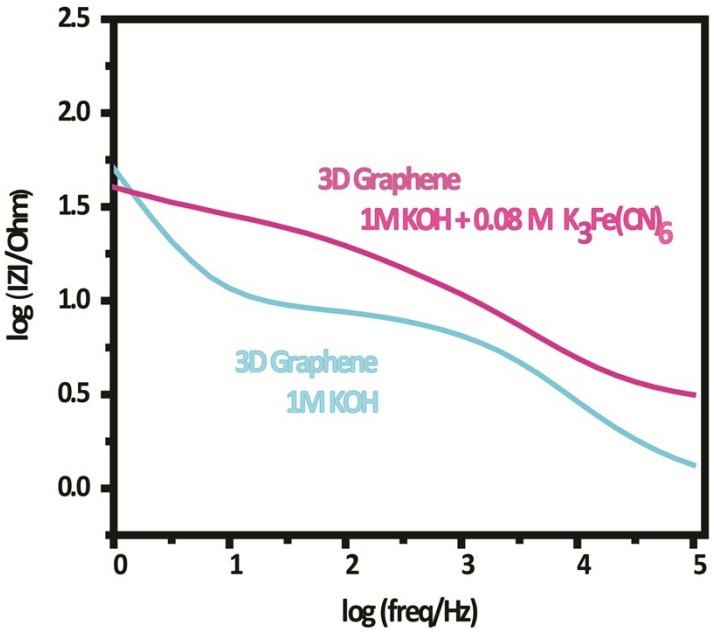


Fig. S11: Bode Plot of 3D Graphene in 1M KOH and 1M KOH+ 0.08M $K_3Fe(CN)_6$

As shown in Fig S10, Bode plot of 3D graphene in different electrolyte system. Bode plot automatically reflects the system dynamics with respect to the frequency. Bode plot shows that irrespective of the frequency the modulus (|Z|) is always higher for the redox mediated 3D graphene than that of 3D graphene. The difference in |Z| between these two systems at any given frequency implies that it is the actual contribution of the redox system towards the total impedance and that it is largely an imaginary component of impedance.

Supplementary Table

Table S1. Performance of various carbon based electrode and redox electrolyte systems.

Electrode material	Redox additive electrolyte	Specific capacitance(F/g)	Ref.
Co–Al LDH	KOH + $K_4Fe(CN)_6$	317 at 2 A/g	S1
	KOH + $K_3Fe(CN)_6$	712 at 2 A /g	S1
CuS	KOH + Na_2S + S	2175 at 15 A /g	S2
Activated carbon	H_2SO_4 + KI	912 at 2 mA/cm^2	S3
	H_2SO_4 + KBr	572 at 2 mA/cm^2	S3
Activated carbon	H_2SO_4 + VOSO4	630.6 at 1 mA/cm^2	S4
Activated carbon	H_2SO_4 + hydroquinone	901 at 2.65 mA/ cm^2	S5
Activated carbon	H_2SO_4 + hydroquinone	220 at 2.65 mA/cm^2	S6
Polyaniline - graphene	H_2SO_4 + hydroquinone	553 at 1 A/g	S7
MnO_2	KOH + p-phenylenediamine	325.24 at 1 A/g	S8
Activated carbon	KOH + p-phenylenediamine	605.22 at 1 A/g	S9
SWCNT	KOH + p-phenylenediamine	162.66 at 1 A/g	S10
MWCNTs	KOH + m-phenylenediamine	78 at 0.5 A/g	S11
MWCNTs	H_2SO_4 + indigo carmine	50 at 0.88 mA/cm^2	S12
Activated carbon	H_2SO_4 + methylene blue	23 F/g	S13
Activated carbon	H_2SO_4 + lignosulfonates	178 at 0.1 A/g	S14
3D graphene	KOH + $K_3Fe(CN)_6$	1830 F/g at 2 A/g 2091 F/g at 5 mV/s	Present work

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