

## Supplementary Materials

# Graphene/Reduced Graphene Oxide-Carbon Nanotubes Composite Electrodes: from Capacitive to Battery-type Behaviour

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**Table S1.** Published details of capacitive G/rGO-CNT single electrodes measured in three-electrode configuration.

Electrode materials	Substrate	Processing details	Electrolyte	Capacitance	Ref.
rGO,CNT,PVDF	C-cloth	80%rGO+10%CNT+10%PVDF,180 °C	Na <sub>2</sub> SO <sub>4</sub>	129 F/g, 0.1 A/g	[1]
rGO,CNT	FS	VF, hydrozine-gas	KOH	265 F/g, 0.1 A/g	[2]
rGO,CNT,PDAA,CB,PTFE	Ni foam	Urea, KMnO <sub>4</sub> , 80 °C	Na <sub>2</sub> SO <sub>4</sub>	80 F/g, 0.2 A/g	[3]
rGO,CNT,AB,PTFE	Ni foam	H <sub>6</sub> N <sub>2</sub> O, 95°C, KMnO <sub>4</sub>	Na <sub>2</sub> SO <sub>4</sub>	91 F/g, 0.25 A/g	[4]
rGO,CNT	Ni foam	dried, 250 °C, pressed	KOH	200 F/g, 0.25 A/g	[5]
rGO,CNT	GCE	urea	KOH	10 F/g, 0.5 A/g	[6]
rGO,CNT	Ni foam	VF, dried, 250 °C, pressed	Na <sub>2</sub> SO <sub>4</sub>	151 F/g, 0.5 A/g	[7]
rGO,CNT,CB,PTFE	Ni foam	chem. oxidation, 200 °C	Na <sub>2</sub> SO <sub>4</sub>	202 F/g, 0.5 A/g	[8]
rGO,CNT,MC,Naf	C-paper	HT, 180 °C, aerogel 80%AM+15%MC+5% Naf	H <sub>2</sub> SO <sub>4</sub>	335 F/g, 0.5 A/g	[9]
G,CNT	Cu foil	CNT-G	Na <sub>2</sub> SO <sub>4</sub>	42 F/g, 1 A/g	[10]
rGO,CNT	GCE	H <sub>6</sub> N <sub>2</sub> O, 95 °C, in-situ polymerization	H <sub>2</sub> SO <sub>4</sub>	53 F/g, 1 A/g	[11]
rGO,CNT	fiber	vit. C, 80 °C, inject	Na <sub>2</sub> SO <sub>4</sub>	60 F/g, 1 A/g	[12]
rGO,CNT	st. steel	aerogel, 800 °C, urea, 70 °C	KOH	149 F/g, 1 A/g	[13]
rGO,CNT,pp-AC	FS	600 °C, VF 10%rGO+10%CNT+80%pp-AC	KOH	214 F/g, 1 A/g	[14]
rGO,CNT,SDBS,PVA,AB,PTFE	Ni foam	H <sub>6</sub> N <sub>2</sub> O, coating 85%AM+10%AB+5%PTFE	KOH	375 F/g, 1 A/g	[15]
rGO,CNT	Ni foam	VF, 300 °C, pressed	KOH	194 F/g, 2 A/g	[16]
rGO,CNT	ITO	LbL, into PEI-rGO/CNT-COOH, 150°C	H <sub>2</sub> SO <sub>4</sub>	120 F/g, 1 mV/s	[17]
rGO,CNT,AB,PTFE	Ni foam	400 °C, 800 °C, KMnO <sub>4</sub>	Na <sub>2</sub> SO <sub>4</sub>	35 F/g, 5 mV/s	[18]
rGO,CNT	Gr-paper	MW, 150°C, EPD	KCl	87 F/g, 5 mV/s	[19]
rGO,CNT	Ti foil	200 °C	Na <sub>2</sub> SO <sub>4</sub>	272 F/g, 5 mV/s	[20]
CNT,Gr	Ni foil	VA CNT on/from HOPG	KOH	110 F/g, 10 mV/s	[21]
GO,CNT, Gr.powder, PVDF	Gr.-plates	H <sub>6</sub> N <sub>2</sub> O 80%AM+10%Gr.powder+10%PVDF	Na <sub>2</sub> SO <sub>4</sub>	150 F/g, 20 mV/s	[22]
rGO,CNT	ITO	LbL, transfer	H <sub>2</sub> SO <sub>4</sub>	157 F/g, 50 mV/s	[23]
rGO,CNT	Si	H <sub>6</sub> N <sub>2</sub> O	KOH	194 F/g, 50 mV/s	[24]
rGO,CNT,AB,PVDF	Ti plate	100 °C, H <sub>6</sub> N <sub>2</sub> O 40%rGO+40%CNT+10%AB+10%PVDF	H <sub>2</sub> SO <sub>4</sub>	250 F/g, 50 mV/s	[25]
rGO,CNT	GCE	110 °C	KOH	132 F/g, 100 mV/s	[26]
rGO,CNT	FS	VF, H <sub>6</sub> N <sub>2</sub> O, NH <sub>4</sub> OH	KOH	294 F/cm <sup>3</sup> , 5 mV/s	[27]
rGO,CNT	fiber	vit. C, 90 °C	LiCl	10.8 F/cm <sup>3</sup> , 10 mV/s	[28]
rGO,CNT	fiber	vit. C, 200 °C	H <sub>2</sub> SO <sub>4</sub> -PVA	36.7 F/cm <sup>3</sup> , 1 A/cm <sup>3</sup>	[29]

**Table S2.** Published details of N-doped G/rGO-CNT single electrodes measured in three-electrode configuration.

Electrode materials	Substrate	Processing details	Electrolyte	Capacitance after (before) N doping	Ref.
rGO,CNT,PDA,AB,PTFE	Ni foam	HT, 180 °C, coating 80%AM+10%PVDF+10%AB	KOH	176 (-) F/g, 0.5 A/g	[30]
rGO,CNT,LS	GCE	urea, LS, 800 °C	KOH	246 (10) F/g, 0.5 A/g	[6]

**Table S3.** Published details of single G/rGO-CNT electrodes modified by PPy measured in three-electrode configuration.

Electrode materials	Sub- strate	Processing details	Electrolyte	Capacitance with (without) PPy	Ref.
rGO,CNT,PPy	fiber	vit. C, PPy, twisted into springs, coated by	LiCl	25.9 (10.8) F/cm <sup>3</sup> , 0.01 V/s	[28]
rGO,CNT,PPy	FS	VF, H <sub>6</sub> N <sub>2</sub> O, (NH <sub>4</sub> ) <sub>2</sub> S <sub>2</sub> O <sub>8</sub> , 95 °C	KCl	211 (-) F/g, 0.2 A/g	[31]
rGO,CNT,PPy,PTFE	Gr.-sub	H <sub>6</sub> N <sub>2</sub> O 80%AM+15%CB+5%PTFE	KCl	361 (-) F/g, 0.2 A/g	[32]
G,CNT,PPy	FS	el.-chem. polymerization 0.05%G+0.05%CNT+99.9%PPy	H <sub>2</sub> SO <sub>4</sub>	453 (-) F/g, 5 mV/s	[33]

**Table S4.** Published details of G/rGO-CNT single electrodes modified by PANI measured in three-electrode configuration.

Electrode materials	Substrate	Processing details	Electrolyte	Capacitance with (without) PANI	Ref.
rGO,CNT,PANI	FS	VF, 200 °C	H <sub>2</sub> SO <sub>4</sub>	257 (-) F/g, 0.2 A/g	[34]
rGO,PANI,CNT	FS	VF, 250 °C	H <sub>2</sub> SO <sub>4</sub>	638 (-) F/g, 0.5 A/g	[35]
rGO,CNT,PANI,MC,Naf	C-paper	HT, 180 °C, aerogel. 80%AM+15%MC+5% Naf	H <sub>2</sub> SO <sub>4</sub>	987 (335) F/g, 0.5 A/g	[9]
rGO,CNT,PANI	GCE	H <sub>6</sub> N <sub>2</sub> O, 95 °C	H <sub>2</sub> SO <sub>4</sub>	359 (53) F/g, 1 A/g	[11]
rGO,CNT,PANI,CB,PTFE	Ni foam	H <sub>6</sub> N <sub>2</sub> O, 100 °C 75%AM+20%CB+5%PTFE	KOH	1035 (-) F/g, 1 mV/s	[36]
rGO,CNT,PANI	fiber	vit. C, 200 °C, PANI onto fiber by cycling voltammetry process	H <sub>2</sub> SO <sub>4</sub> -PVA	193 (36.7) F/cm <sup>3</sup> , 1 A/cm <sup>3</sup>	[29]

**Table S5.** Published details of G/rGO-CNT-based single electrodes prepared with metal catalysts measured in three-electrode configuration.

Electrode materials	Substrate	Processing details	Electrolyte	Capacitance in F/g	Ref.
G,CNT,Co,Mo	Gr-paper	VA G by plasma-assisted method from CNT by CVD with Co/Mo	Na <sub>2</sub> SO <sub>4</sub>	278, 10 mV/s	[37]
rGO,CNT,Co,Fe/Al <sub>2</sub> O <sub>3</sub> ,CB,PTFE	Ni foam	GO with Co(NO <sub>3</sub> ) <sub>2</sub> ·6H <sub>2</sub> O and urea, CNT by CVD with Fe/Al <sub>2</sub> O <sub>3</sub> , 750 °C 75%AM+20%CB+5%PTFE	KOH	385, 10 mV/s	[38]
G,CNT,Au	Ni foam	Au on CNT, core-shell G on CNT by powder pressed	KOH-CMC	373, 1 mV/s	[39]

**Table S6.** Published details of G/rGO-CNT-based single electrodes modified by MnO<sub>2</sub> measured in three-electrode configuration.

Electrode materials	Substrate	Processing details	Electrolyte	Capacitance with (without) MnO <sub>2</sub>	Ref.
rGO,CNT,MnO <sub>2</sub> ,PDAA,CB,PTFE	Ni foam	Urea, KMnO <sub>4</sub> , 80 °C	Na <sub>2</sub> SO <sub>4</sub>	193 (80) F/g, 0.2 A/g	[3]
rGO,CNT,MnO <sub>2</sub> ,AB,PTFE	Ni foam	H <sub>6</sub> N <sub>2</sub> O, 95 °C, KMnO <sub>4</sub>	Na <sub>2</sub> SO <sub>4</sub>	126 (91) F/g, 0.25 A/g	[4]
rGO,CNT,MnO <sub>2</sub> ,AB,PTFE	Ni foam	HT,150 °C, KMnO <sub>4</sub>	Na <sub>2</sub> SO <sub>4</sub>	336 (-) F/g, 0.5 A/g	[40]
rGO,CNT,MnO <sub>2</sub> ,AB,PTFE	Ni foam	400 °C, 800 °C, KMnO <sub>4</sub>	Na <sub>2</sub> SO <sub>4</sub>	133 (35) F/g, 5 mV/s	[18]
rGO,CNT,MnO <sub>2</sub>	FS	VF, H <sub>6</sub> N <sub>2</sub> O, NH <sub>4</sub> OH, KMnO <sub>4</sub>	Na <sub>2</sub> SO <sub>4</sub>	372 (-) F/g, 10 mV/s	[41]
rGO,CNT,MnO <sub>2</sub> ,Gr.powder,PV	Gr. paper	H <sub>6</sub> N <sub>2</sub> O, 95 °C, KMnO <sub>4</sub>	Na <sub>2</sub> SO <sub>4</sub>	367 (150) F/g, 20 mV/s	[22]
G,CNT,MnO <sub>2</sub>	Cu foil	LbL, CVD, 1000 °C thermal decomposition, 410 °C	Na <sub>2</sub> SO <sub>4</sub>	365 (42) F/g, 1 A/g	[10]

**Table S7.** Published details of G/rGO-CNT-based single electrodes modified by other metal oxides measured in three-electrode configuration.

Electrode materials	Substrate	Processing details	Electrolyte	Capacitance, F/g	Ref.
rGO,CNT,CeI <sub>3</sub> ,Co <sub>3</sub> O <sub>4</sub> ,SnO <sub>2</sub> ,AB,PTFE	Ni foam	NH <sub>4</sub> OH, HCl, CoCl <sub>2</sub> ·6H <sub>2</sub> O, SnCl <sub>2</sub> ·2H <sub>2</sub> O, urea,150 °C, coating	KOH	215, 0.2 A/g	[42]
rGO,CNT,Fe <sub>2</sub> O <sub>3</sub> ,CB,PTFE	Ni foam	70%AM+20%AB+10%PTFE 75%AM+20%CB+5%PTFE	KOH	657, 1 A/g	[43]
G,CNT,LiMn <sub>2</sub> O <sub>4</sub> ,AB,PTFE	Ni foam	85%AM+10%AB+5%PTFE	Li <sub>2</sub> SO <sub>4</sub>	676, 0.2 mV/s	[44]

**Table S8.** Published details of G/rGO-CNT-based single electrodes modified by Ni(OH)<sub>2</sub> measured in three-electrode configuration.

Electrode materials	Substrate	Processing details	Electrolyte	Capacitance with (without) Ni(OH) <sub>2</sub>	Ref.
rGO,CNT,Ni(OH) <sub>2</sub>	st. steel	GO-CNT aerogel, 800 °C Ni(NO) <sub>3</sub> ×6H <sub>2</sub> O, urea, 70 °C, pressed	KOH	1208 (149) F/g, 1 A/g	[13]
rGO,CNT,Ni(OH) <sub>2</sub> ,AB,PTFE	Ni foam	GO,CNT, urea, NiCl <sub>2</sub> ×6H <sub>2</sub> O, autoclave at 120 °C	KOH	1320 (-) F/g, 6 A/g	[45]
G,CNT,Ni(OH) <sub>2</sub>	Ni foam	VA CNT on/from HOPG, 1200 °C, G by pyrolysis of FePc, 1000 °C, Ni(OH) <sub>2</sub> coating by ECD	KOH	1384 (110) F/g, 5 mV/s	[21]

## Glossary

FS	freestanding	AB	acetylene black
G	graphene	AC	active carbon
GCE	glassy carbon electrode	Al <sub>2</sub> O <sub>3</sub>	aluminium oxide
Gr	graphite	AM	active material
GO	graphene oxide	Au	gold
ITO	indium tin oxide	C-paper	carbon paper
HCl	hydrochloric acid	CB	carbon black
HOPG	highly ordered pyrolytic graphite	C-cloth	carbon cloth
HT	hydrothermal method	Cell.	ethyl cellulose
H <sub>6</sub> N <sub>2</sub> O	hydrazine hydrate or hydrazine solution	chem. oxidation	chemical oxidation
H <sub>2</sub> SO <sub>4</sub>	sulfuric acid	CMC	carboxymethyl cellulose
HNO <sub>3</sub>	nitric acid	CNT	carbon nanotubes
KMnO <sub>4</sub>	potassium permanganate	CNT-COOH	carboxylic acid functionalized CNT by concentrated H <sub>2</sub> SO <sub>4</sub> /HNO <sub>3</sub>
KOH	potassium hydroxide	Co	cobalt
KCl	potassium chloride	Co <sub>3</sub> O <sub>4</sub>	cobalt oxide
LbL	layer-by-layer	CoCl <sub>2</sub> ×6H <sub>2</sub> O	cobalt chloride hexahydrate
LiCl	lithium chloride	Co(NO <sub>3</sub> ) <sub>2</sub> ×6H <sub>2</sub> O	cobaltous nitrate hexahydrate
LiClO <sub>4</sub>	lithium perchlorate	Cu	copper
LiMn <sub>2</sub> O <sub>4</sub>	lithium mangan dioxide	CVD	chemical vapor deposition
Li <sub>2</sub> SO <sub>4</sub>	lithium sulfate	ECD	electrochemical deposition
LS	lignosulfonate	EPD	electrophoretic deposition
MC	mesoporous carbon	Fe	iron
		FePc	iron phthalocyanines

<i>pp</i> -AC	porous AC fabricated by pyrolysis, pre-carbonization, and alkali activation of biomass of pomelo peels	MnO <sub>2</sub> Mo MW	manganese dioxide  microwave
PPy	polypyrrole	Na <sub>2</sub> SO <sub>4</sub>	sodium sulfate
PTFE	polytetrafluoroethylene	Naf	Nafion
PVA	polyvinyl alcohol	(NH <sub>4</sub> ) <sub>2</sub> S <sub>2</sub> O <sub>8</sub>	ammonium persulfate
PVDF	polyvinylidene fluoride	NH <sub>4</sub> OH	ammonia hydroxide or ammonia solution
rGO	reduced graphene oxide	Ni NiCl <sub>2</sub> ×6H <sub>2</sub> O Ni(NO) <sub>3</sub> ×6H <sub>2</sub> O Ni(OH) <sub>2</sub>	nickel nickel chloride hexahydrate nickel nitrate hexahydrate nickel hydroxide
Si	silicon		
SnO <sub>2</sub>	tin oxide	PANI	polyaniline
SnCl <sub>2</sub> ×2H <sub>2</sub> O		PDA	polydopamine
st. steel	stainless steel	PDAA	poly(1,5-diaminoanthraquinone)
Ti	titanium	PEI-rGO	reduced GO and poly(ethyleneimine) solution after removing by centrifugation of excess polymer and hydrazine hydrate
VA	vertically aligned		
VF	vacuum filtration		
vit. C	vitamin C		

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