

Supporting Material

Enhancement of supercapacitive properties of laser deposited graphene-based electrodes through carbon nanotube loading and nitrogen doping

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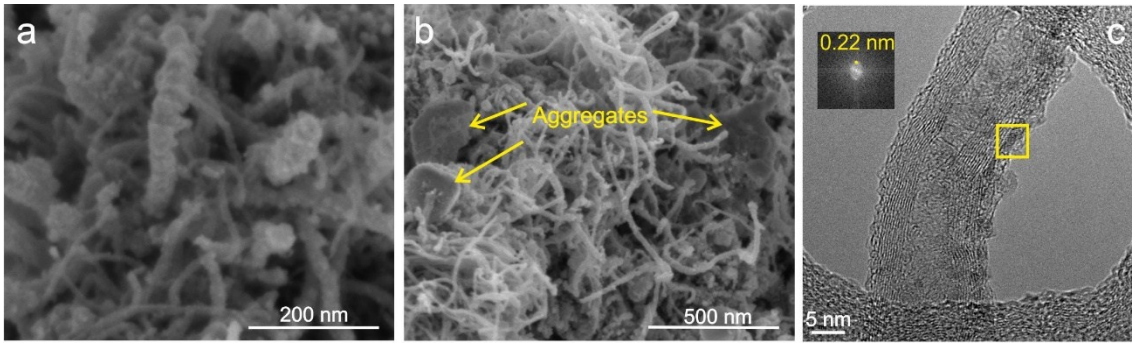


Figure S1. High-resolution (a, b) SEM and (c) TEM images of sample GO-CNT-NiO (0/1/5). Inset in (c): FFT of the selected crystallite (square in c).

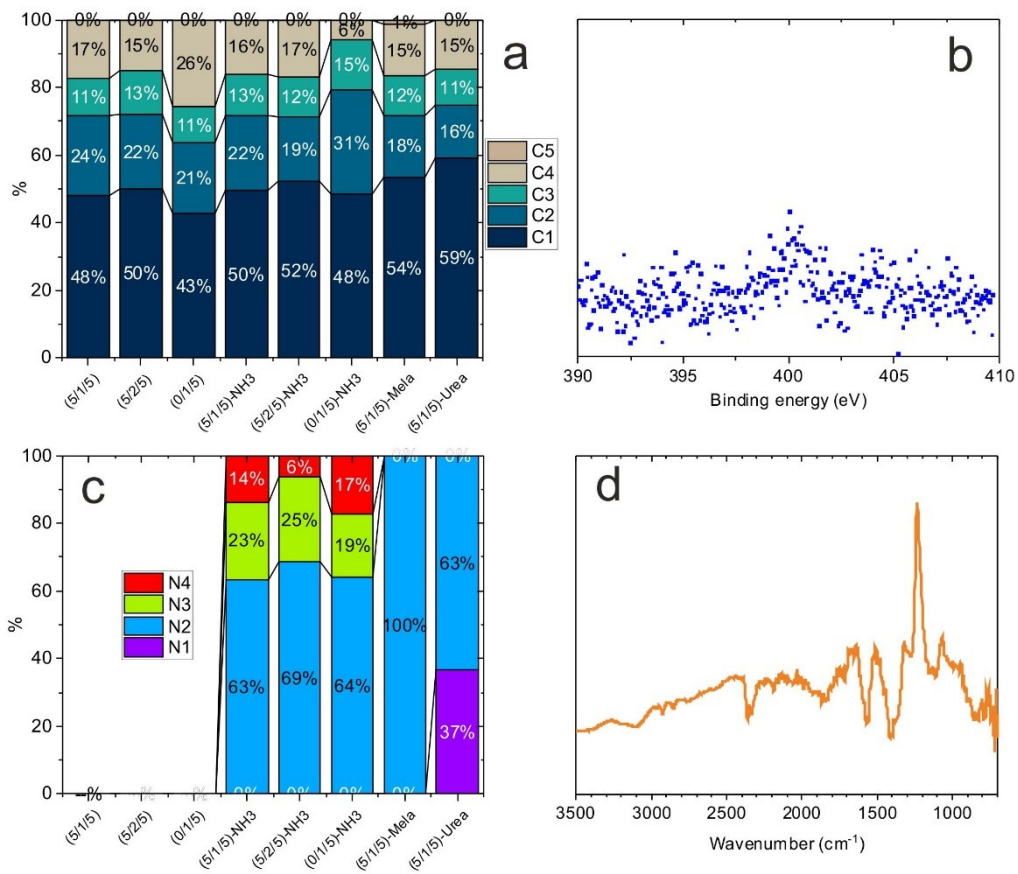


Figure S2. High-resolution XPS and FTIRM results of GO-CNT-NiO samples. (a) Integrated area percentage of C1s components, (b) N1s signal of (5/1/5), (c) integrated area percentage of N1s components. (d) Characteristic FTIRM spectrum of (0/1/5)-ammonia sample.

GO	Assignment
800	C-O-C or C-H
890	C-O-C
980	C=C
1150	C-OH
1550	C-C
1640	C=C
2900	OH
3200	OH

CNT	Assignment
1240	ν (C-O), C-OH
1560	C-C

Urea	Assignment
1150	ν_2 (or $2\nu_L$) amorphous
1450	C-N , CN ₂
1600	C-N, CN ₂ , N-H
1680	O+NH, CO
3250	NH
3330	ν_{as} NH ₂
3420	ν_{as} NH ₂

Melamine	Assignment
800	$\delta_{oopring}$; $\delta_{twistNH_2}$
1020	δ_{rockNH_2} ; $\nu_{C-N(H_2)}$; δ_{ipring}
1200	δ_{rockNH_2}
1420	$\nu_{C-N(H_2)}$; δ_{ipring}
1530	$\delta_{scissNH_2}$; δ_{rockNH_2} ; δ_{ipring}
1620	$\delta_{scissNH_2}$
3100	ν_s NH ₂
3320	ν_{as} NH ₂
3410	ν_{as} NH ₂
3460	ν_{as} NH ₂

Table S1. Assignments of bands appearing in GO, CNT, urea and melamine raw materials.

GO-CNT-NiO	Assignment
890	C-O-C
1010	C=C
1320	C-OH / C-O-C
1530	C-N, CN ₂ , N-H
1680	C=O

NH ₃	Assignment
750	γ CO, γ NH ₂ +CO out-of-phase
950	Second vibrational mode of Ammonia
1090	v ₂ (or 2v _L) amorphous
1260	C-OH, C-O-C
1350	C-OH, C-O-C, v ₂ + v _L Crystalline
1510	C-N, CN ₂ , N-H
1700	C=O
1790	H-N-H scissoring
3100	ν s NH ₂
3530	N-H symmetric stretching

Urea	Assignment
770	γ CO, γ NH ₂ +CO out-of-phase
810	C-O-C or C-H
890	ν (C-N), CN ₂
1100	C-O
1210	NH
1480	C-N, CN ₂ , N-H
1600	C-O and NH ₂
1700	O+NH, CO

Melamine	Assignment
790	δ _{oopring} ; δ _{twistNH₂}
1100	C-O
1240	δ _{rockNH₂}
1490	C-N, CN ₂
1580	N-H
1750	C=O

Table S2. Assignments of bands appearing in GO-CNT-NiO (5/1/5) samples.

NH ₃	Assignment
750	γ CO, γ NH ₂ +CO out-of-phase
930	C=C
1070	ν_2 (or $2\nu_L$) amorphous
1240	C-OH , C-O-C
1320	C-OH, C-O-C , ν_2 + ν_L Crystalline
1520	C-N, CN ₂ , N-H
1680	C=O
1760	H-N-H scissoring
2030	C=C=N stretching
2640	C-H aldehyde
2880	N-H stretching
3100	ν_s NH ₂
3430	NH

Table S3. Assignments of bands appearing in GO-CNT-NiO (0/1/5)-ammonia sample.

	ESR (Ω)	CPE - n	R (k Ω)
(5/0/5)	24	0.93	50
(5/1/5)	23	0.94	54
(5/2/5)	32	0.93	97
(5/1/5)-NH ₃	23	0.93	> 1000
(5/1/5)-Urea	25	0.95	63
(5/1/5)-Mela.	37	0.94	260

Table S4. Equivalent circuit parameters obtained from regression of EIS data of GO-CNT-NiO electrodes.

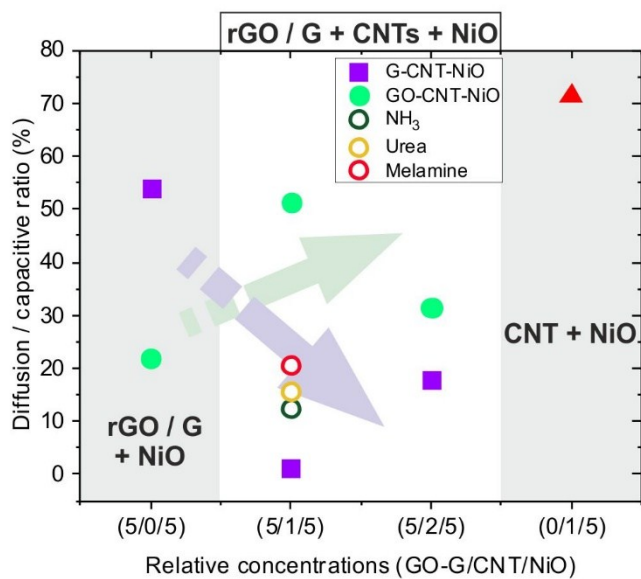


Figure S3. Ratio between integrated areas of diffusion and capacitive components calculated from cyclic voltammetry measurements of GO-CNT-NiO and G-CNT-NiO samples. Ammonia, urea and melamine samples were obtained with GO-CNT-NiO targets with a relative concentration of (5/1/5). Triangular symbol belongs to the sample without graphene oxide.

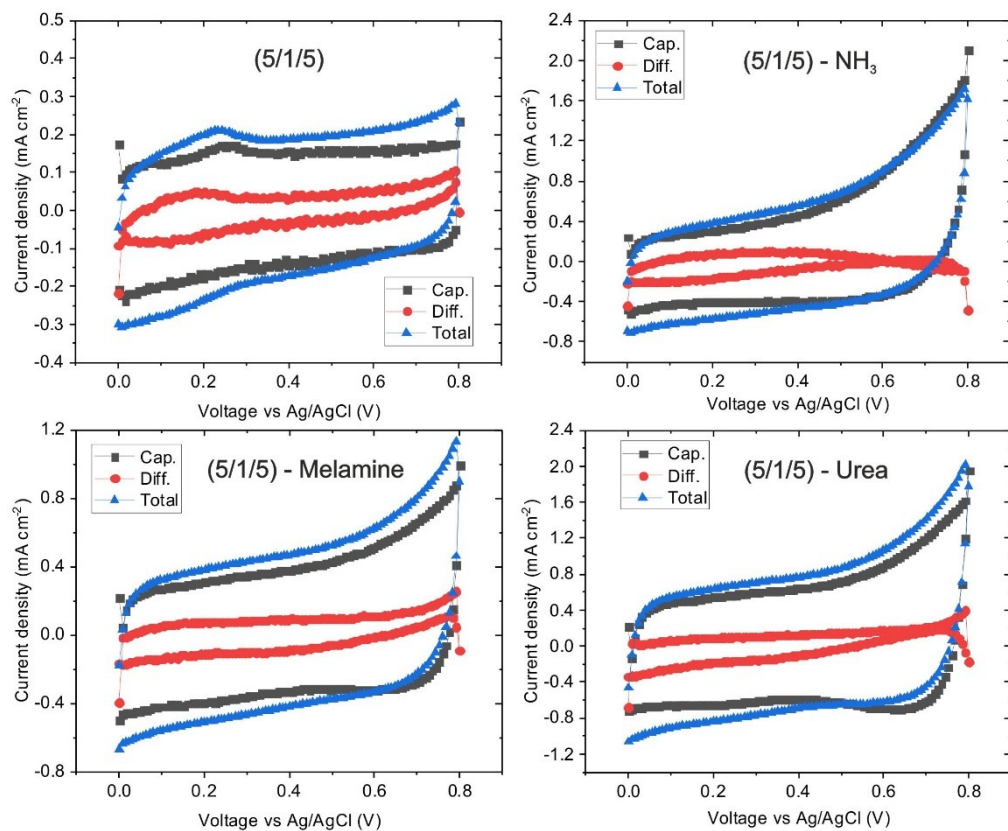


Figure S4. Cyclic voltammetry curves of GO-CNT-NiO (5/1/5) and the equivalent ones obtained with ammonia / melamine / urea taken with 100 mV s⁻¹. Calculated capacitive and diffusion components are also depicted.

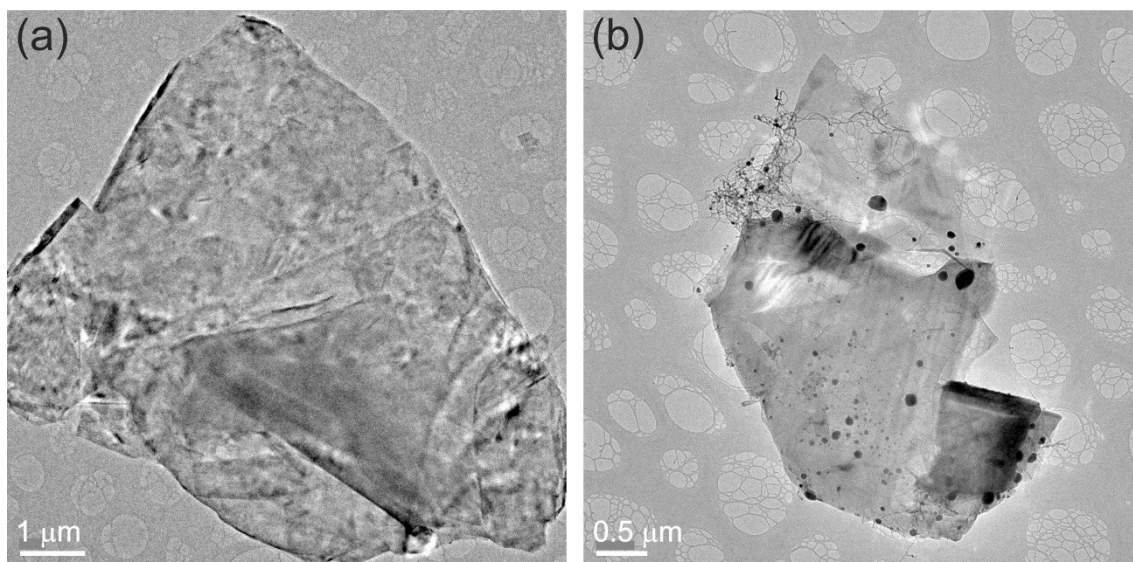


Figure S5. TEM images of (a) graphene sheet from the precursor powder, (b) graphene sheet from G-CNT-NiO (5/2/5) sample.

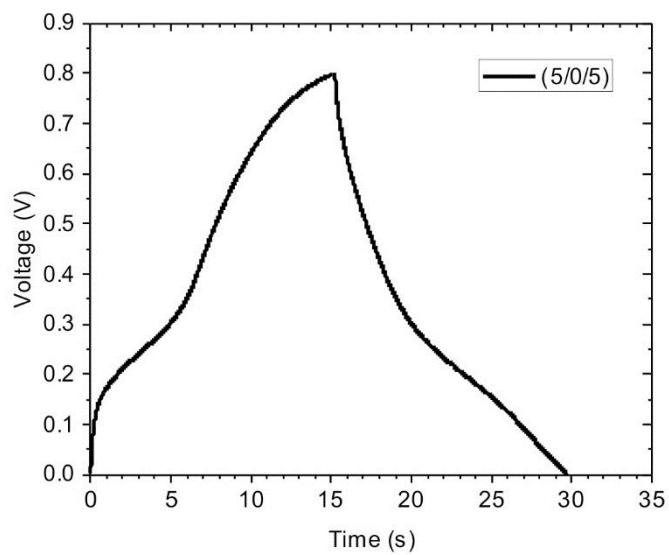


Figure S6. Galvanostatic charge-discharge cycle of G-CNT-NiO (5/0/5) electrode at applied current of $50 \mu\text{A cm}^{-2}$.