

Supporting Information

Oxidative Unzipping of Stacked Nitrogen-Doped Carbon Nanotube Cups

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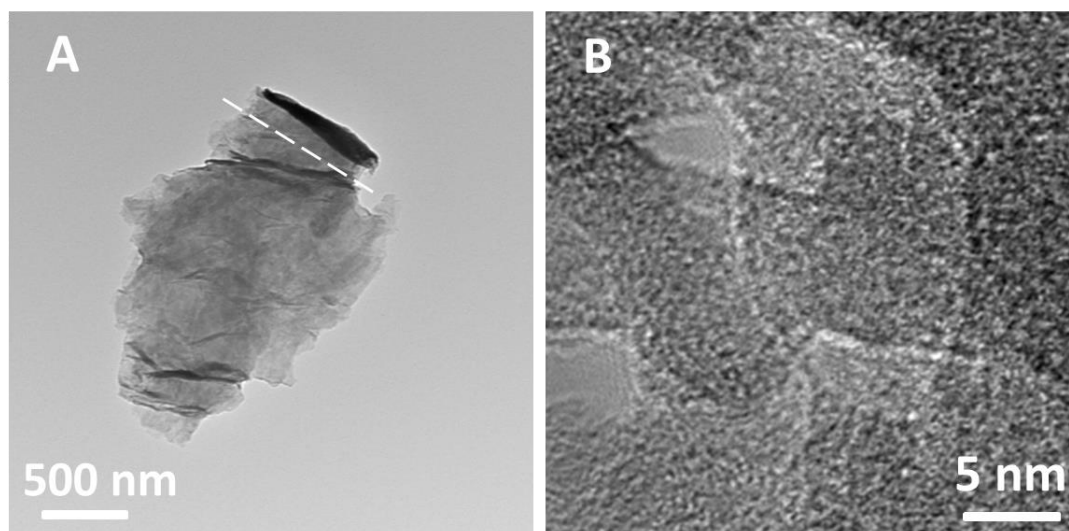


Figure S1. (A) TEM image showing an individual separated NCNC after undergoing the unzipping process resulting in GNSs. The unzipping process longitudinally opened the graphitic surface along the cup axis (indicated by the white dotted line). (B) High-resolution TEM image showing multiple graphitic layers on a large graphene flake.

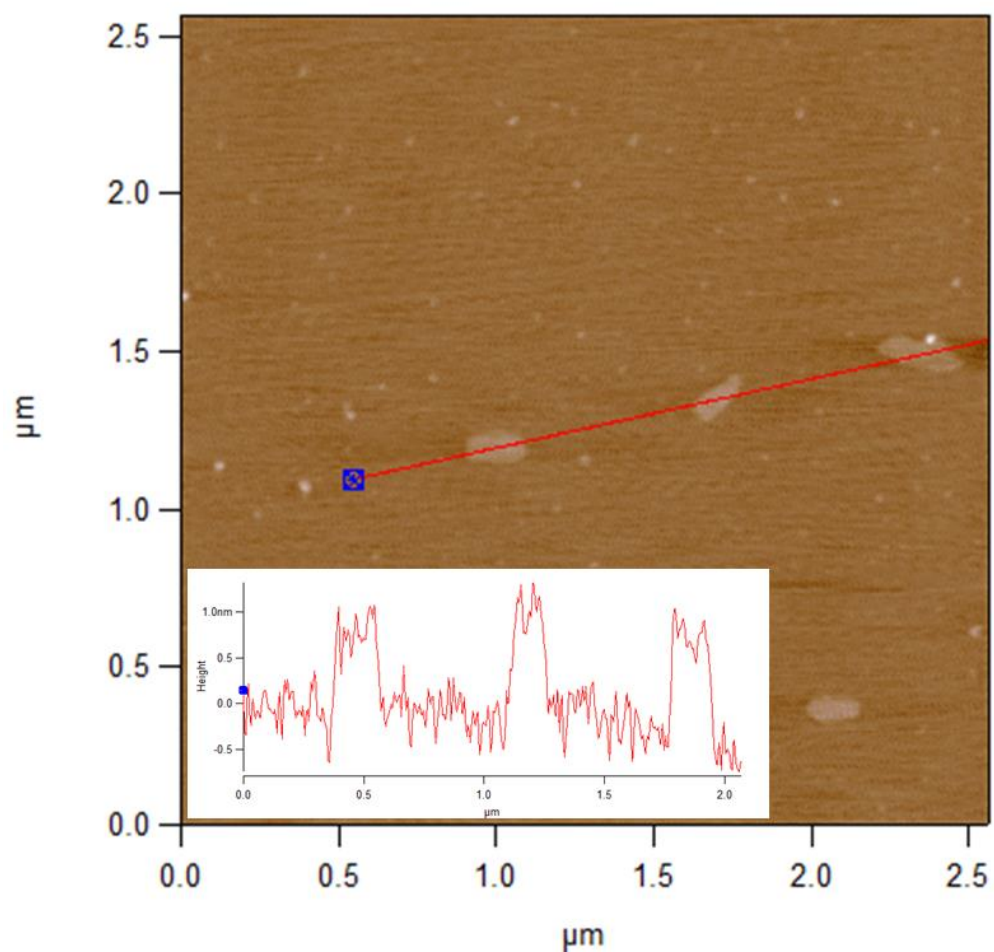


Figure S2. Atomic force microscopy (AFM) image of obtained nitrogen-doped GNSs. The red line indicates where the height profile was taken with sizes around 100 – 200 nm and heights of ~ 1 nm corresponding to single or double graphene layers.

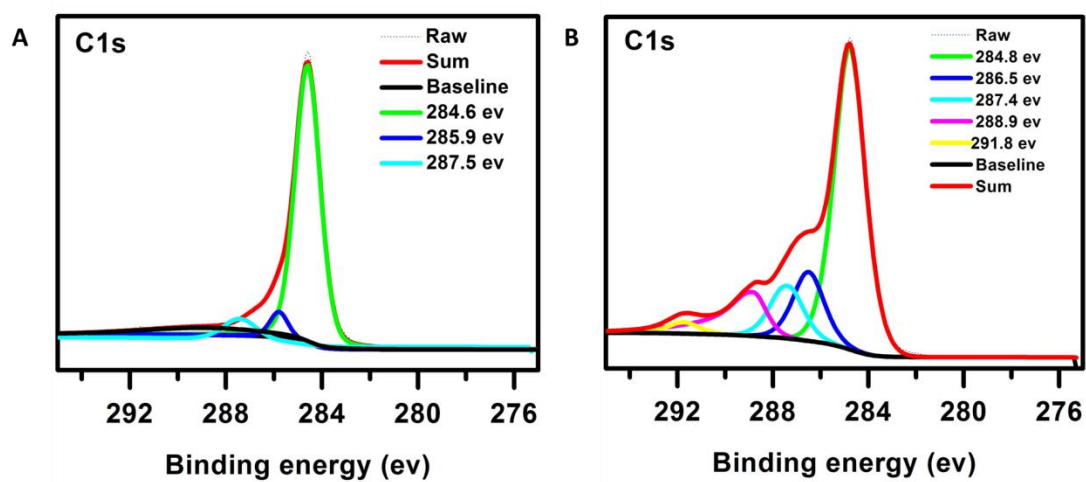


Figure S3. XPS spectra of the high-resolution C1s profiles of (A) o-NCNCs and (B) u-NCNCs. After the unzipping process, oxidized carbon functionalities containing C-O and C=O groups are revealed.

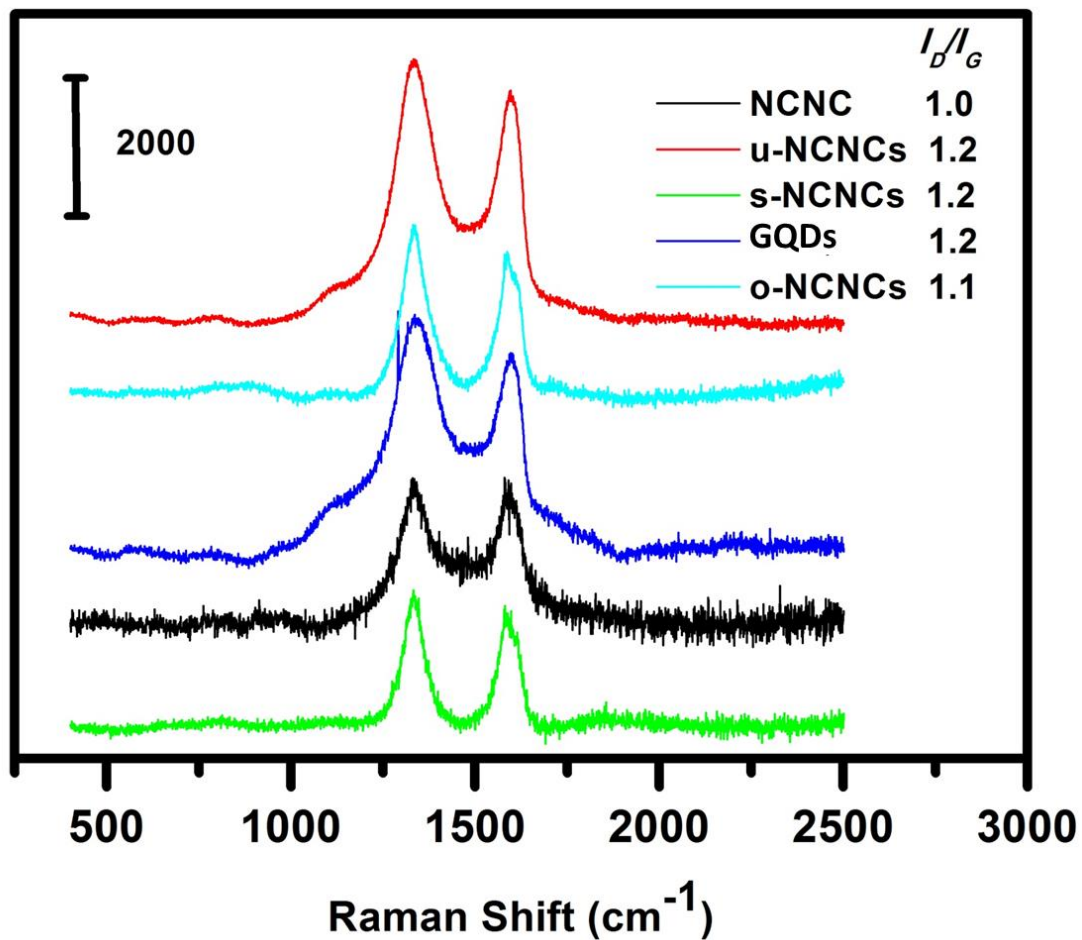


Figure S4. Raman spectra of pristine (as-CVD-synthesized) NCNCs (black), o-NCNCs (blue), and u-NCNCs (red), s-NCNCs (green), and N-doped GQDs (cyan). The oxidation of NCNCs causes an increase of the I_D/I_G ratio from 1.0 to 1.1, indicating the formation of a large amount of defects. The separation and unzipping processes further increase the amount of defects and the I_D/I_G ratio goes to 1.2.

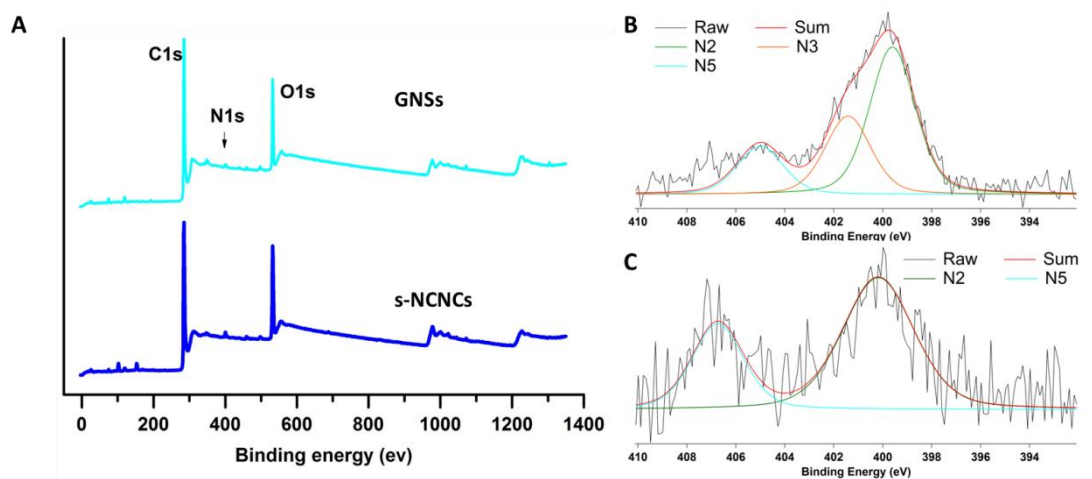


Figure S5. (A) XPS survey scan spectra of s-NCNCs (blue) and N-doped GNSs after unzipping of s-NCNCs. High-resolution XPS peak profile of (B) s-NCNCs and (C) completely unzipped GNSs.

Table S1. Survey XPS with average atomic % and standard deviation of all materials

	o-NCNCs	u-NCNCs	s-NCNCs	GNSs
C1s	88.11 ± 0.75	77.83 ± 1.45	84.93 ± 0.70	93.56 ± 1.26
O1s	11.04 ± 0.96	21.52 ± 1.55	13.48 ± 0.15	5.70 ± 0.97
N1s	0.86 ± 0.43	0.65 ± 0.13	1.59 ± 0.69	0.74 ± 0.34

The increase in nitrogen content from o-NCNCs to s-NCNCs is hypothesized to be due to the removal of amorphous carbon thus exposing more exterior nitrogen functionalities previously inaccessible due to the short depth penetration afforded by XPS analysis (~10 nm).

Table S2. N1s high resolution XPS and percent of nitrogen functionalities of all materials

	Electron Volts (eV)	o-NCNCs	u-NCNCs	s-NCNCs	GNSs
N1 (Pyridinic)	398.9 – 399.5	31%	7%	-	-
N2 (Pyrolic/amine)	400.0 – 400.3	17%	44%	54%	68%
N3 (Graphitic)	401.0 – 401.6	35%	-	28%	-
N4 (Valley)	402.3	-	36%	-	-
N5 (Oxidized)	405.0 – 406.0	17%	13%	18%	32%

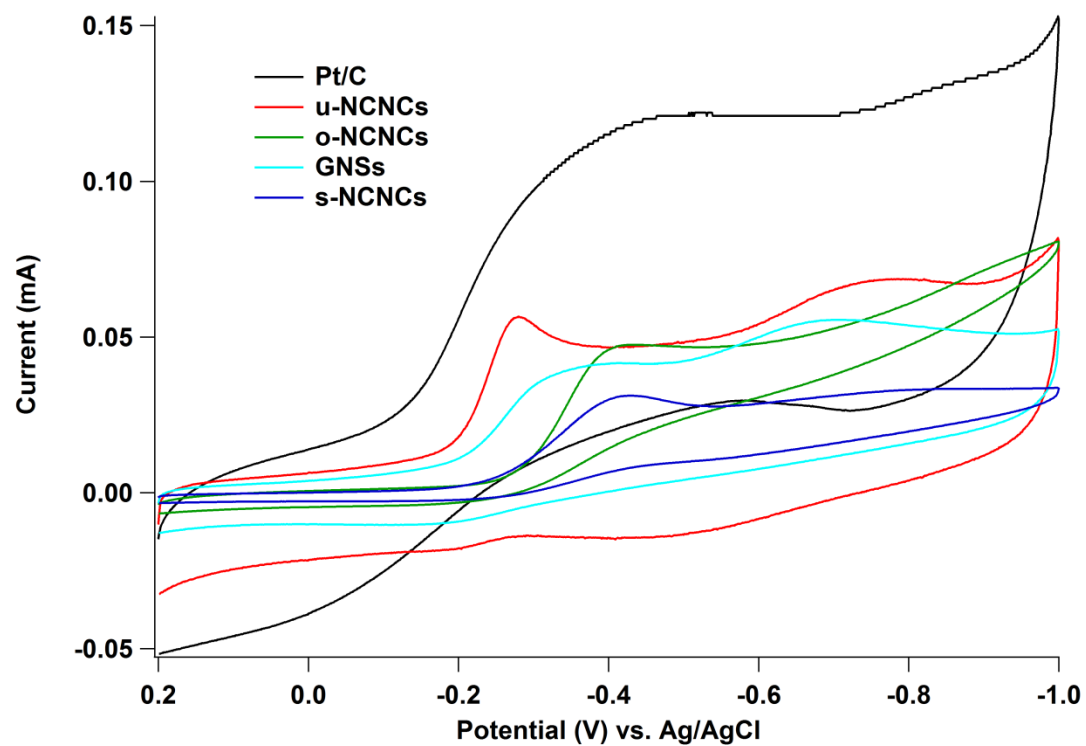


Figure S6. Cyclic voltammetry (CV) of Pt/C (black), u-NCNCs (red), o-NCNCs (green), N-doped GNSs (cyan), and s-NCNCs (blue) catalysts in O_2 -saturated 0.1M KOH.

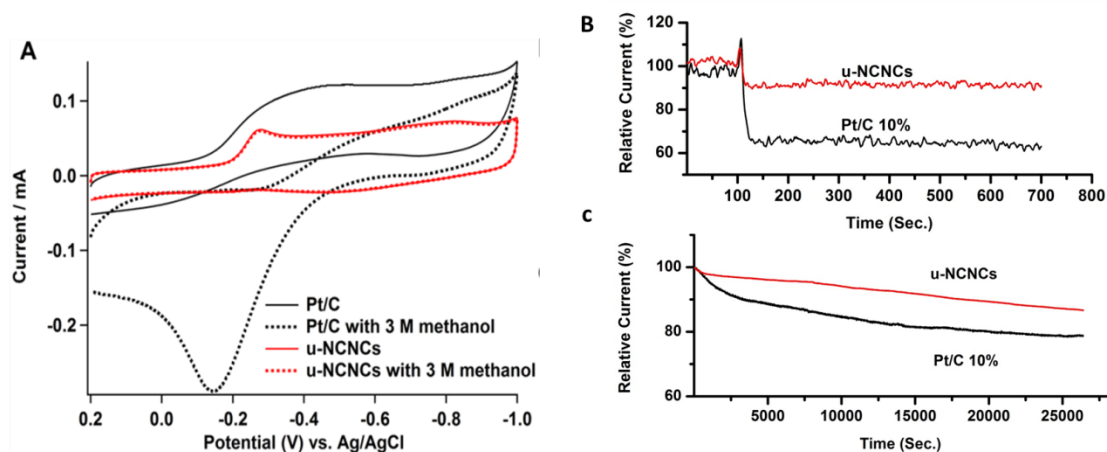


Figure S7. (A) CV curves of Pt/C (black) and u-NCNCs (red) in O₂-saturated 0.1M KOH with (dotted) or without (solid) 3 M Methanol. (B) CO poisoning effect tests for Pt/C (black) and u-NCNCs (red). 10% CO was introduced in a 120 sccm flow of O₂ at 100 s where the initial drop in current for u-NCNCs is due to the decrease of oxygen partial pressure. (C) I-t chronoamperometric response of commercial 10% Pt/C (black) and u-NCNCs (red) in O₂-saturated 0.1M KOH at -0.3 V with a rotating speed of 1400 rpm.

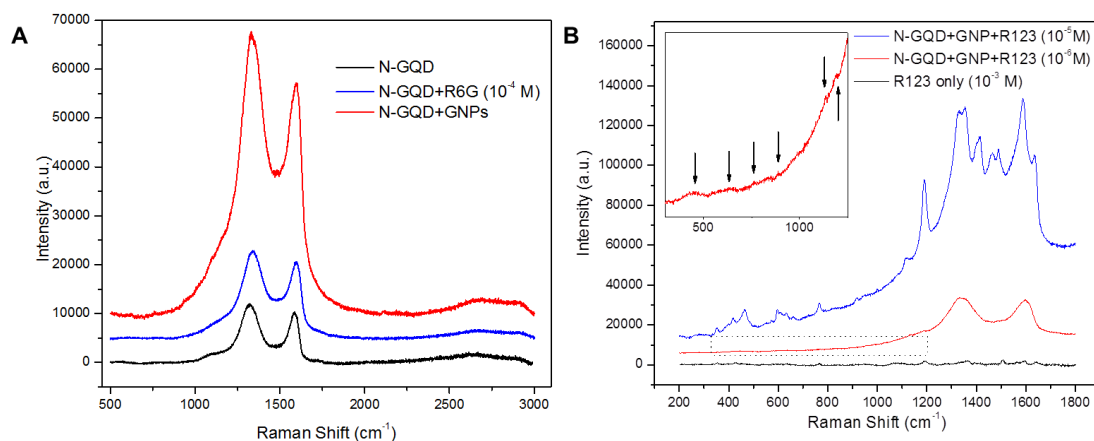


Figure S8. (A) Regular Raman spectrum of GNSs (back), as compared to the SER spectrum of GNS functionalized with GNPs (red). The Raman spectrum of GNS mixed with 10^{-4} M of Rhodamine 6G without GNP functionalization; (B) SER spectra of GNP-functionalized GNSs sensing Rhodamine 123 at different concentrations. The inset shows the magnified spectrum of the dashed block region.