

Supplementary information

Complementary behaviour of EDL and HER activity in functionalized graphene nanoplatelets

Anand B. Puthirath^{1*}, Sharmila Shirodkar¹, Minfei Fei¹, Abhijit Baburaj¹, Keiko Kato¹, Sreehari K Saju¹,
Ranjith Prasannachandran^{1,2}, Nithya Chakingal¹, Robert Vajtai^{1,3}, Boris Yakobson¹ and Pulickel M. Ajayan^{1*}

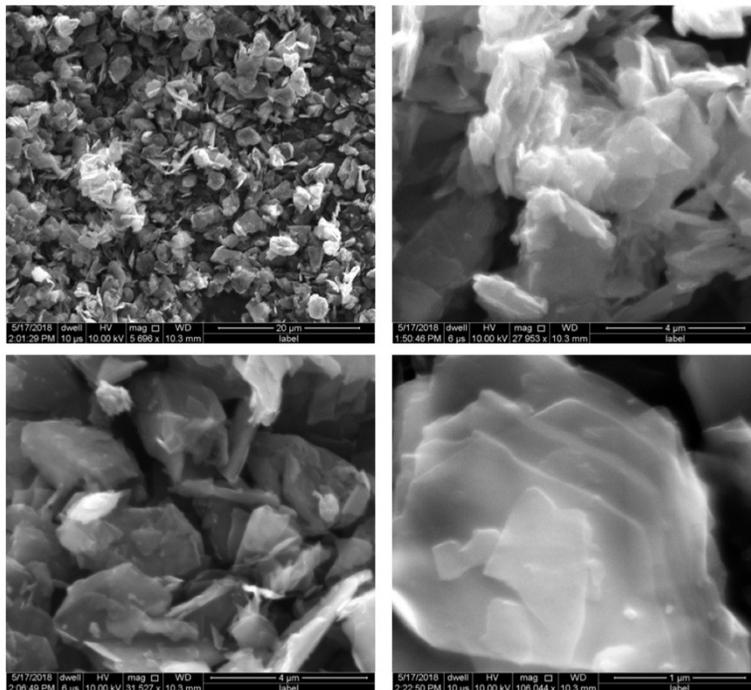
¹Department of Materials Science and NanoEngineering, Rice University, Houston, TX 77005, USA

²School of Physics, Indian Institute of Science Education and Research Thiruvananthapuram, Kerala 695551, India

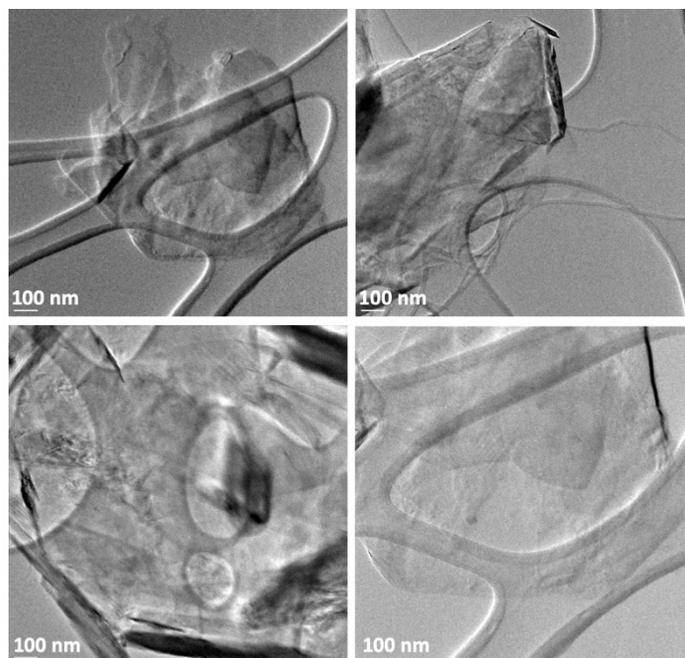
³Interdisciplinary Excellence Centre, Department of Applied and Environmental Chemistry, University of Szeged, Hungary

Email: ajayan@rice.edu, anandputhirath@gmail.com

S1: SEM & TEM Analysis

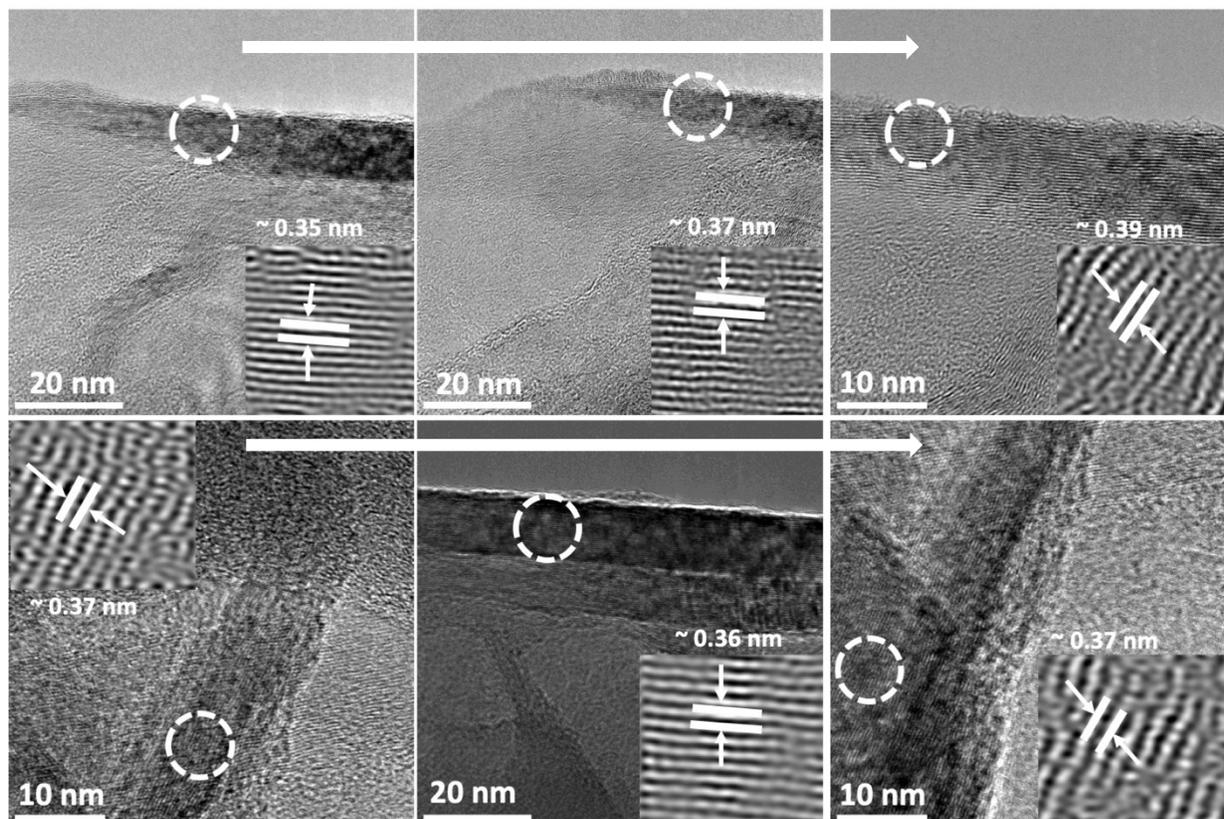


Low- and high-resolution images of pristine graphene nanoplatelets (powder form)



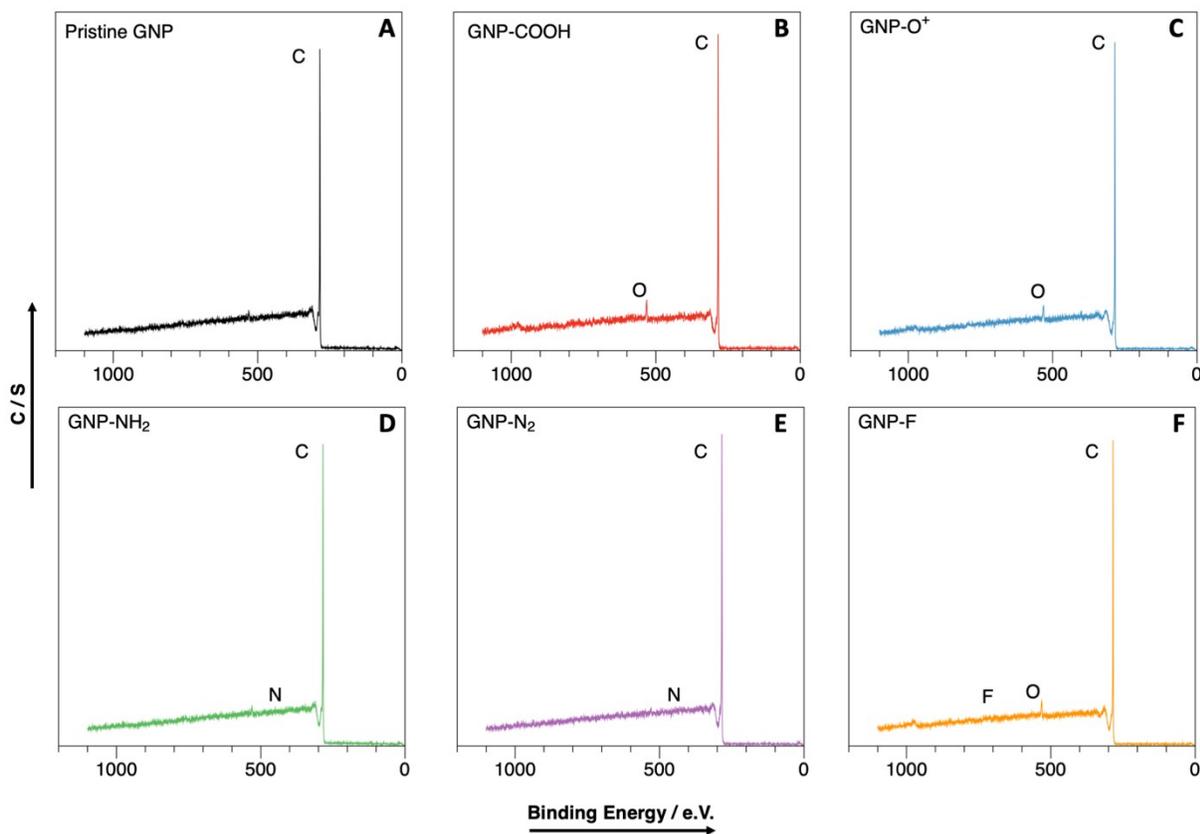
Low- and high-resolution images of COOH-functionalised (top left) and pristine graphene nanoplatelets (rest) (dispersion)

S2. TEM Analysis



High resolution TEM images of GNPs and functionalised GNPs. Starting from top left to bottom right; Pristine GNP, GNP-COOH, HNP-O⁺ GNP-NH₂, GNP-N₂, GNP-F.

S3: XPS Survey spectrum of pristine and functionalized GNPS

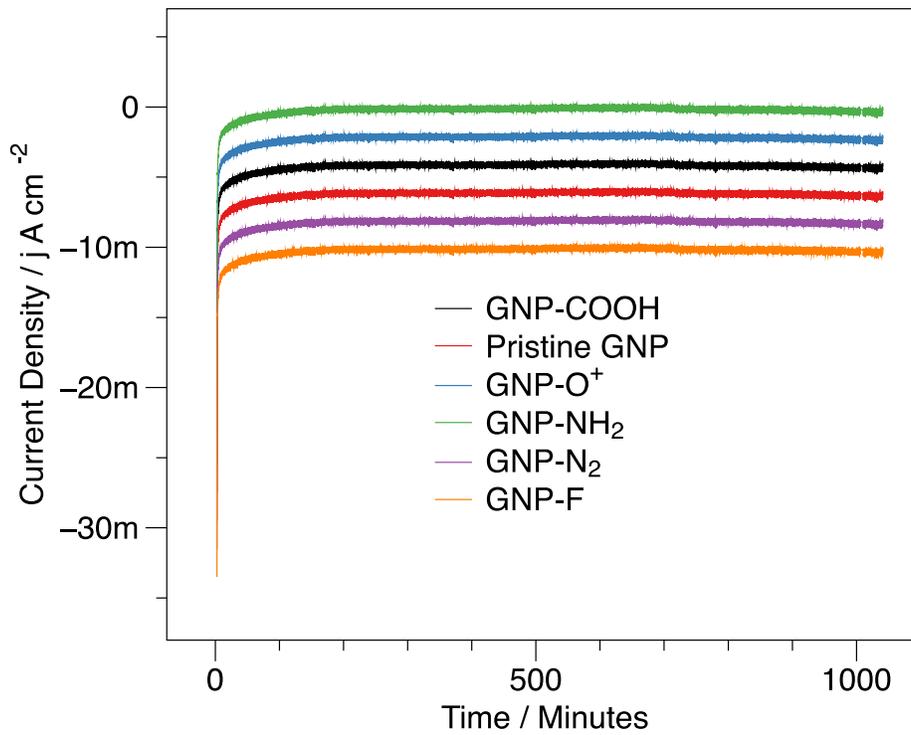


XPS Survey spectrum of pure and functionalised GNPs (passing energy 124 eV)

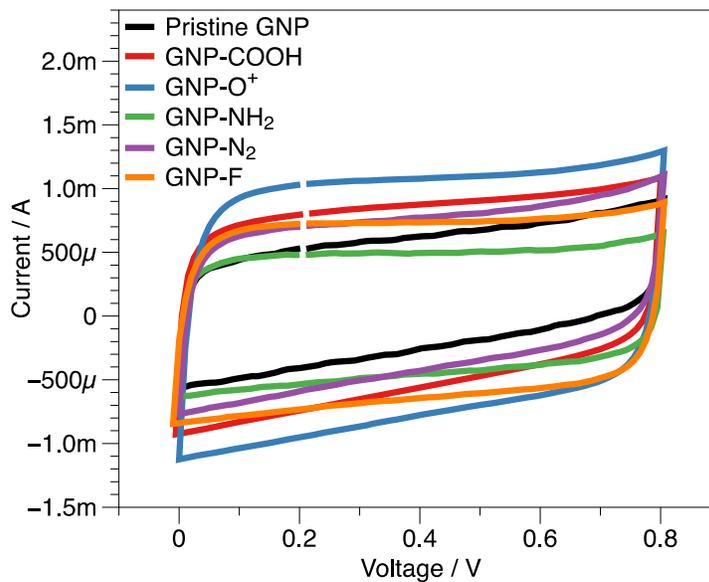
Samples	Surface Atomic Score Levels (At %)				% Functionalization
	C	O	N	F	
Pristine GNP	~98.58	~1.42	--	--	~1.5
GNP-COOH	~96.40	~3.60	--	--	~4
GNP-O ⁺	~96.10	~3.90	--	--	~4
GNP-NH ₂	~95.5	~3.15	~1.3	--	~4.5
GNP-N ₂	~93.5%	~2.0%	~4.5%	--	~5
GNP-F	~95.05	~2.47	--	~2.48%	~5

Atomic percentage and percentage of functionalisation derived from XPS survey scans. Percentage of functionalization is around 5 for all the samples, and hence we have simulated each system accordingly.

S4: Current Stability check curves of the samples at corresponding overpotentials (~18 hrs)



S5: Cyclic Voltammograms of supercapacitors based on pristine and functionalised GNPs

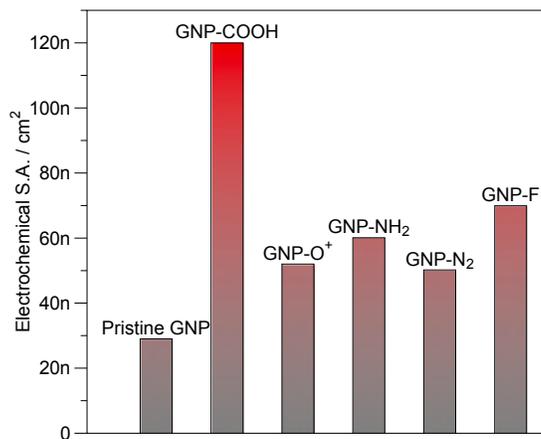
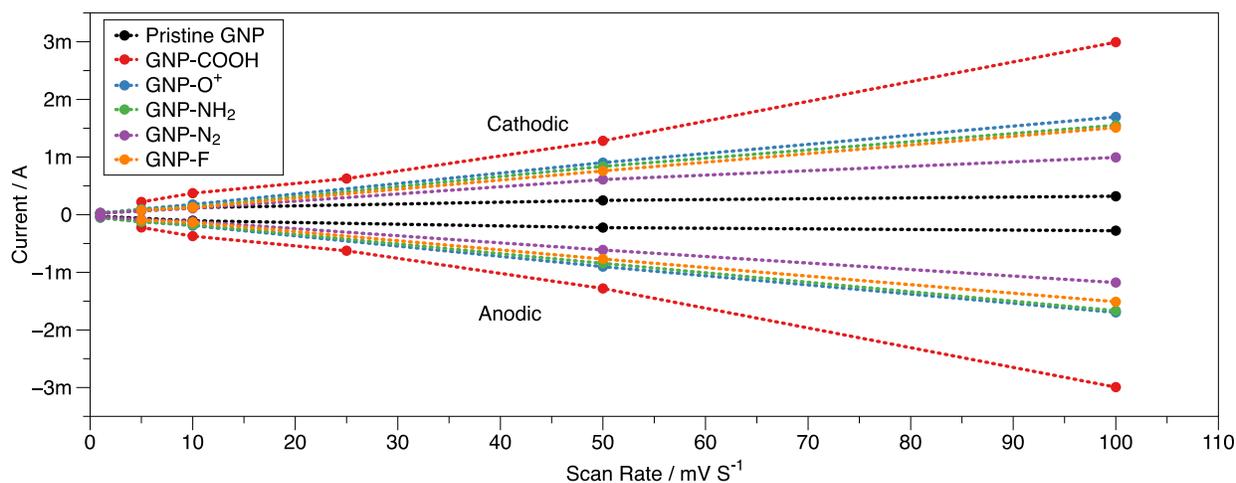


Cyclic Voltammetry Curves (in one frame) of supercapacitor devices made of Pristine GNP and functionalized GNPs

S6: Electrochemical Surface Area of pristine and functionalised GNPs

The electrochemically active surface area (ECSA) of pristine GNPs and functionalized GNPs were estimated by determining the double-layer capacitance of the system from CV (non-faradaic region) measurements. The CV measurements were carried out in the double layer region at various scan rates ranging from 1 mV/s to 100 mV/s. The plot of anodic peak current (i_a) or cathodic peak current (i_c) vs. scan rate will yield a straight line with a slope vale of C_{dl} . The ECSA of the catalyst can be calculated by dividing C_{dl} by the specific capacitance (C_s).⁽¹⁾

$$ECSA = \frac{C_{dl}}{C_s}$$



References:

1. Prasannachandran R, Vineesh TV, Anil A, Krishna BM, Shajjumon MM. Functionalized Phosphorene Quantum Dots as Efficient Electrocatalyst for Oxygen Evolution Reaction. *ACS Nano*. 2018;12(11):11511-9.