Electronic Supplementary Material (ESI) for Materials Chemistry Frontiers.

Supporting Information

Structural Evolution of BCN Systems from Graphene Oxide Towards Electrocatalytically Active Atomic Layers

Sumit Bawari[1], Kshama Sharma[1], Golap Kalita[2], P. K. Madhu[1], Tharangattu N. Narayanan[1],* and Jagannath Mondal[1]*

1. Tata Institute of Fundamental Research - Hyderabad, Sy. No. 36/P Serilingampally Mandal, Gopanapally Village, Hyderabad - 500107, India.

2. Department of Physical Science and Engineering, Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya 466-8555, Japan

(tnn@tifrh.res.in (T.N.N.) and jmondal@tifrh.res.in (J.M.)

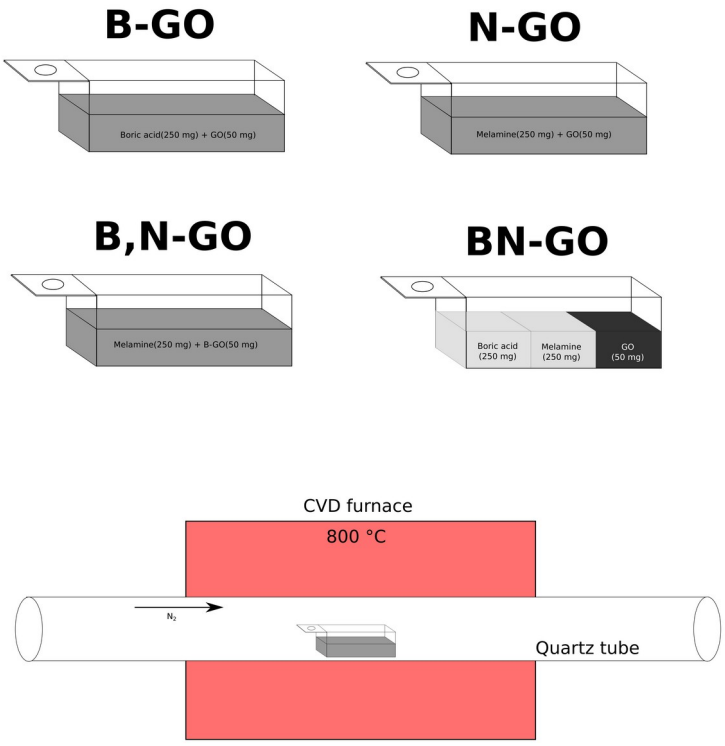

Figure S1: Schematic describing the annealing process, for B-GO, N-GO, B,N-GO and BN-GO.

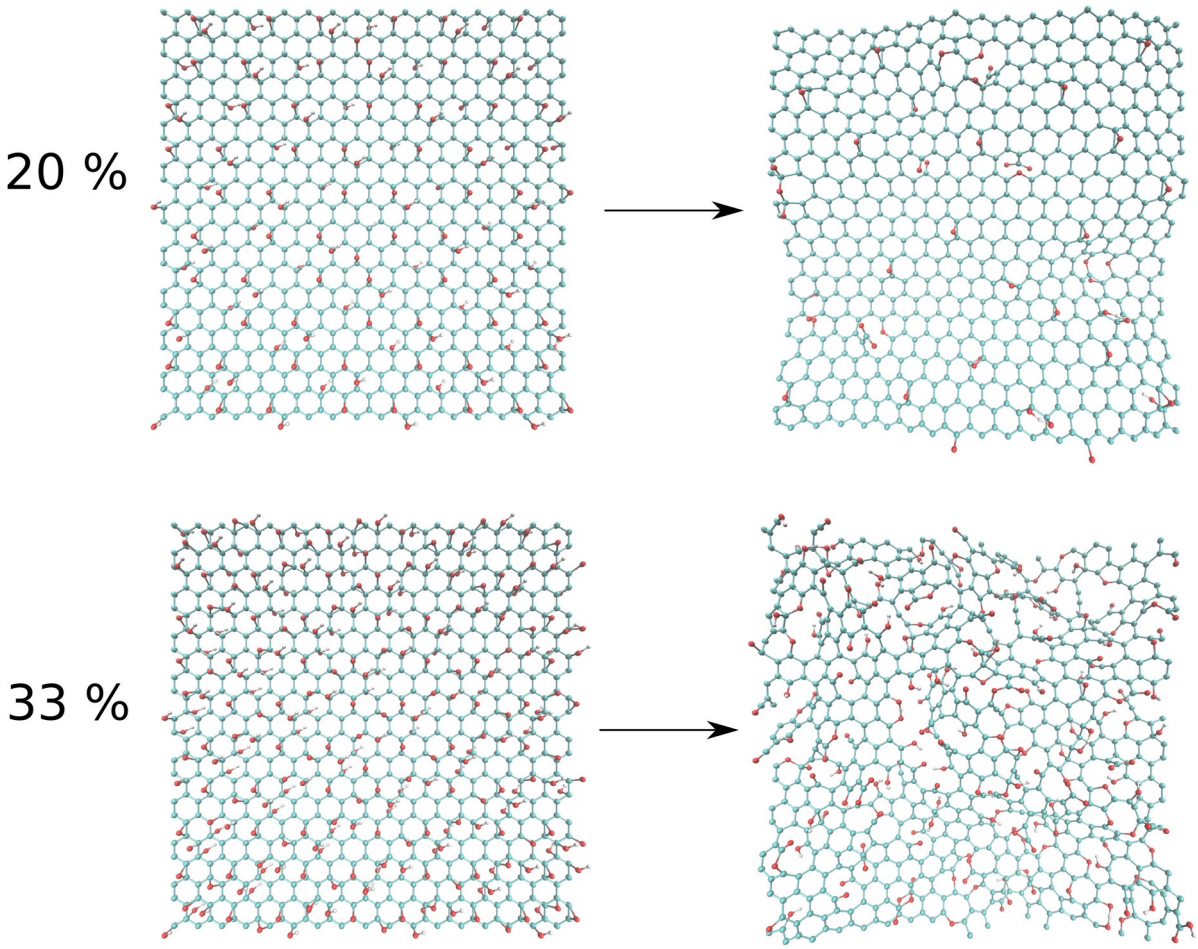

Figure S2: Initial and final structures of the MD simulation for GO, 20% and 33% O/C.

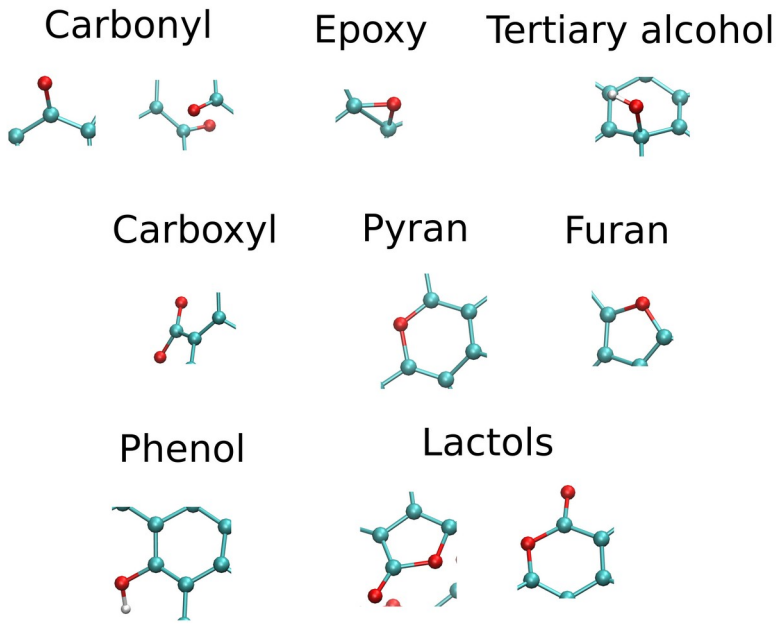

Figure S3: Different oxygen functionalities in GO generated after annealing.

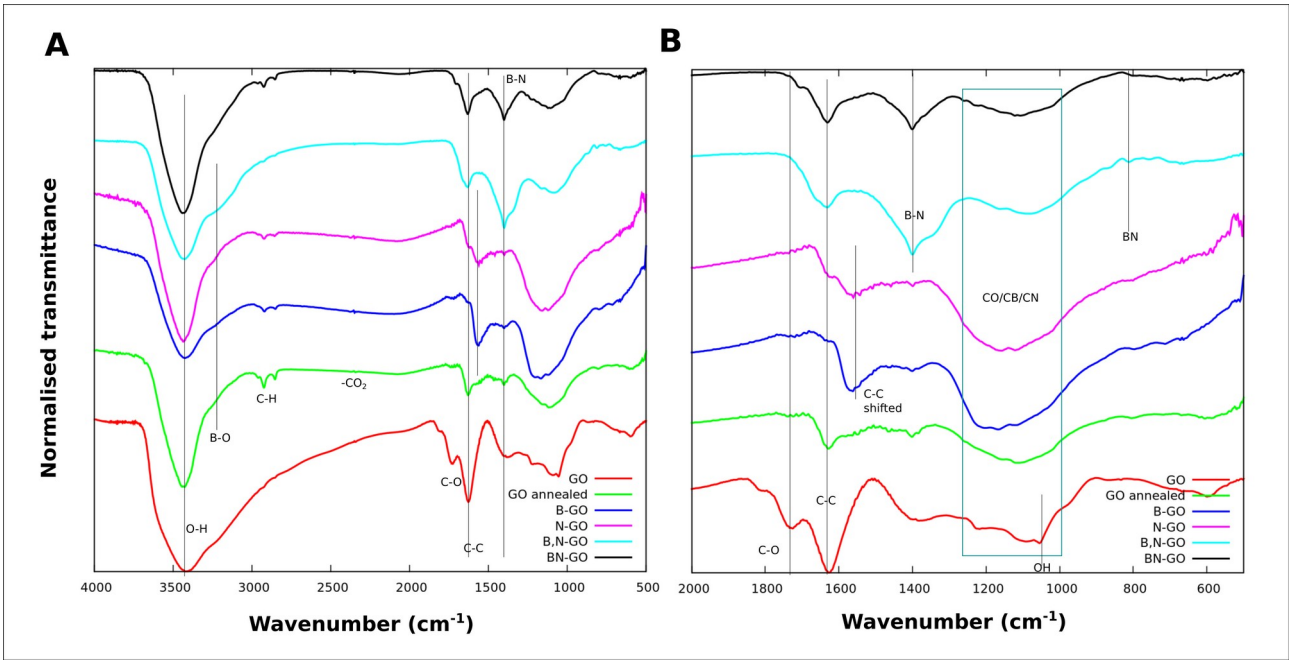

Figure S4: FTIR spectra for GO, annealed GO, B-GO, N-GO, B,N-GO and BN-GO, with certain peaks highlighted.

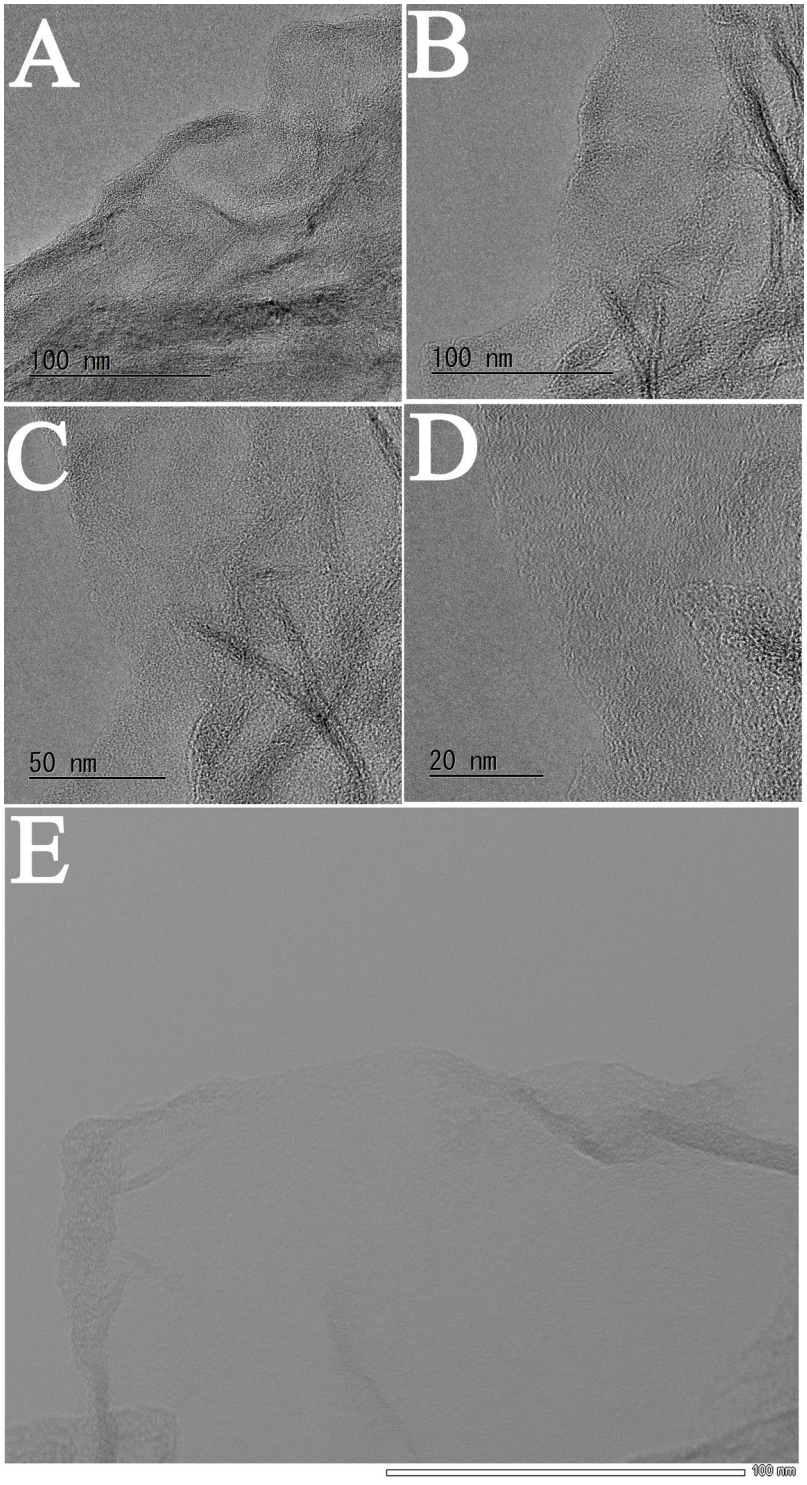

Figure S5: Bright field TEM images of the atomic layers of BN-GO.

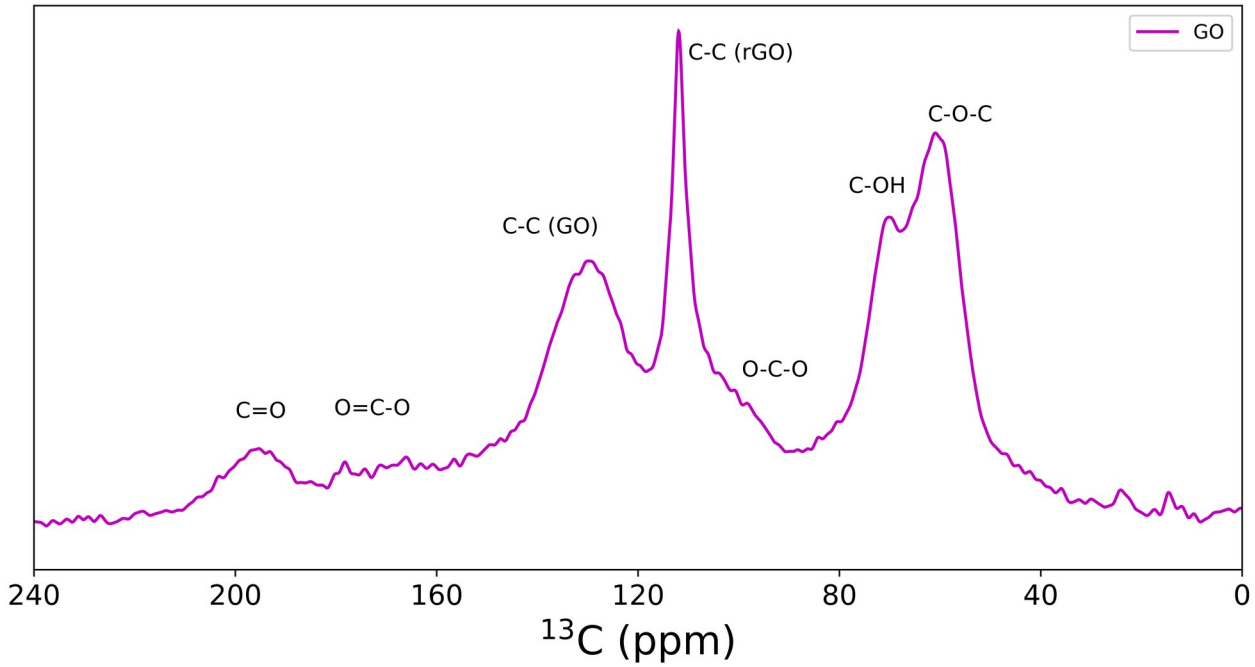

Figure S6: ^{13}C SSNMR of GO highlighting major peaks.

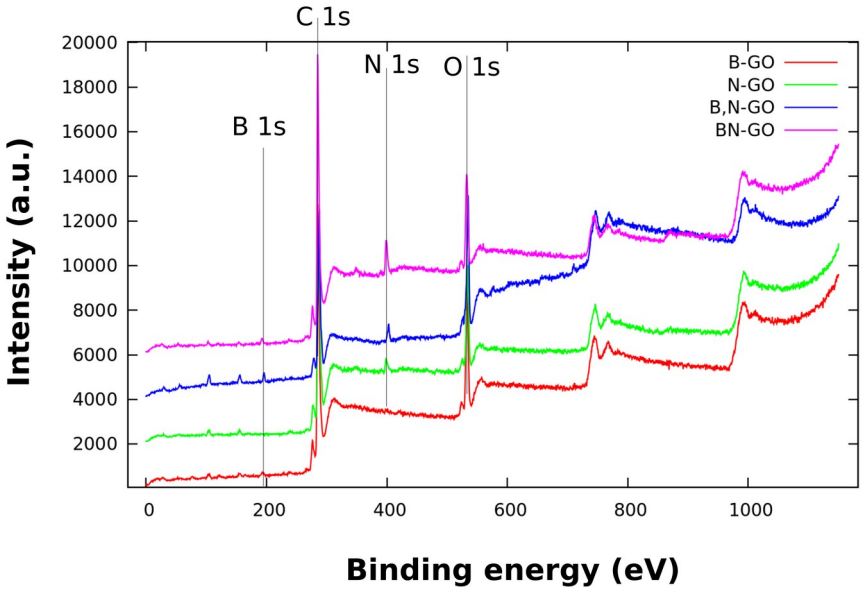

Figure S7: Survey spectrum for B-GO, N-GO, B,N-GO and BN-GO, showing clear peaks for B,C,N,O.

Table S1: Peaks of the deconvoluted XPS spectra for B,C,N,O regions for all four samples.

B 1s

Species	B-GO	B,N-GO	BN-GO
B-3C	191.27	-	191.04
B-O-2C	192.86	192.51	192.19
B-N	-	-	193.58
B-2O-C	194.67	195.18	195.68
B2O3	-	196.39	-

N 1s

Species	N-GO	B,N-GO	BN-GO
N-B	-	-	397.83
Pyridinic	398.16	-	398.91
Pyrrolic	399.74	399.15	400.32
Graphitic	401.51	401.65	402.62

N-O	-	403.06	-

C 1s

Species	B-GO	N-GO	B,N-GO	BN-GO
C-B	283.31	-	-	283.9
C=C	284.43	284.64	284.69	284.61
C-O	285.43	285.33	285.25	285.33
C=O	287.12	286.15	285.94	286.08
C-N	-	287.74	287.63	286.69
O-C=O	289.23	289.44	288.75	289.22
pi-pi*	291.43	291.83	291.66	-

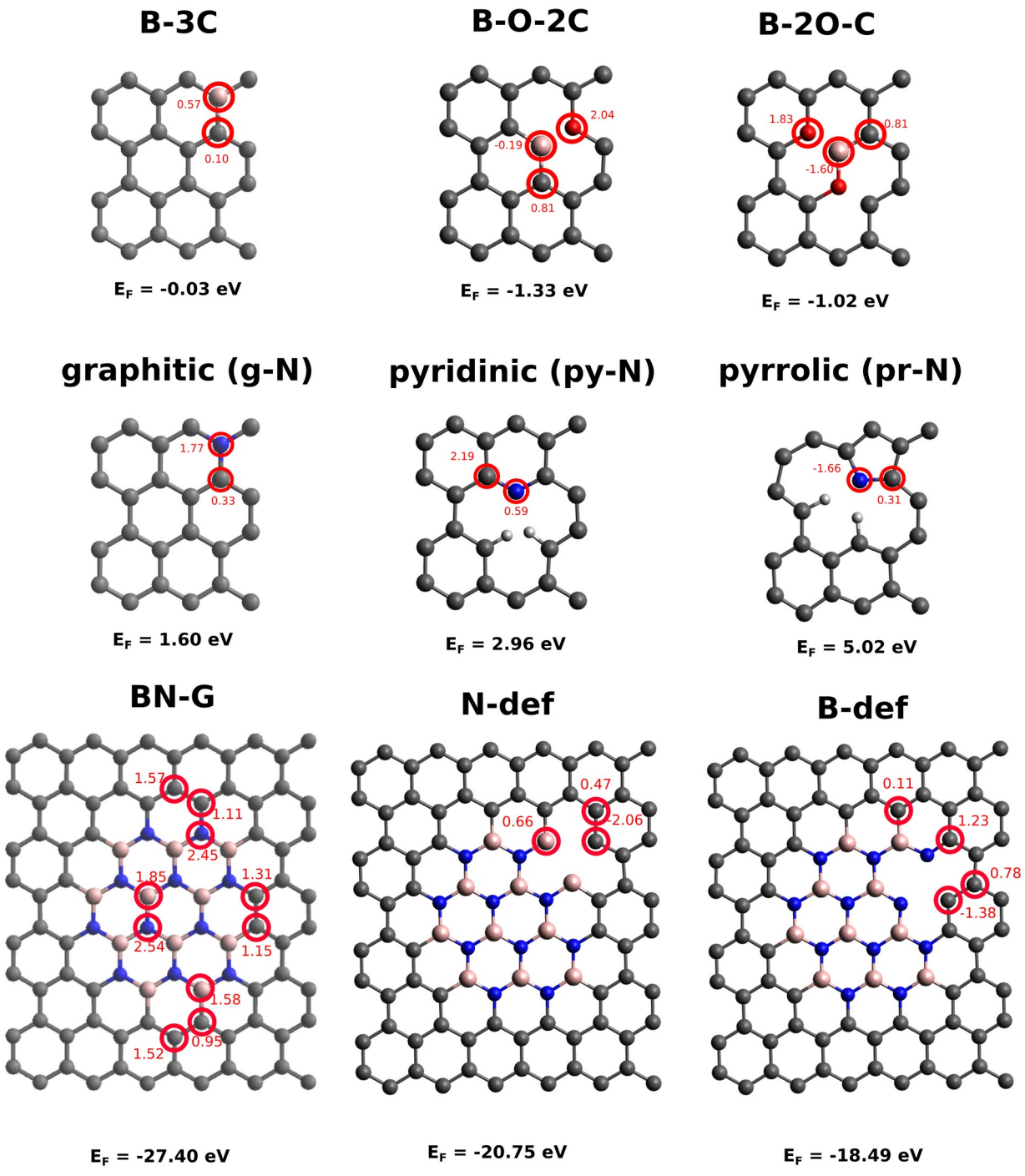

Figure S8: Different simulated structures for B-GO, N-GO and BN-GO, with formation energies(E_F) and ΔG_H values mentioned.

Table S2:

Weighting of the ΔG_H of all four systems, depending upon the percentages of that functionality found in MD simulations.

System	%MD	ΔG_H (eV)	Weighted ΔG_H (eV)
B-GO			0.74
B-3C	10.31	0.10	
B-O-2C	35.05	0.81	
B-2O-C	54.64	0.81	
N-GO			0.51
g-N	32.45	0.33	
py-N	67.55	0.59	
B,N-GO			0.58
B-3C	5.36	0.10	
B-O-2C	14.29	0.81	
B-2O-C	14.29	0.81	
g-N	30.36	0.33	
py-N	35.71	0.59	
BN-GO			0.29
B-def	50	0.11	
N-def	50	0.47	

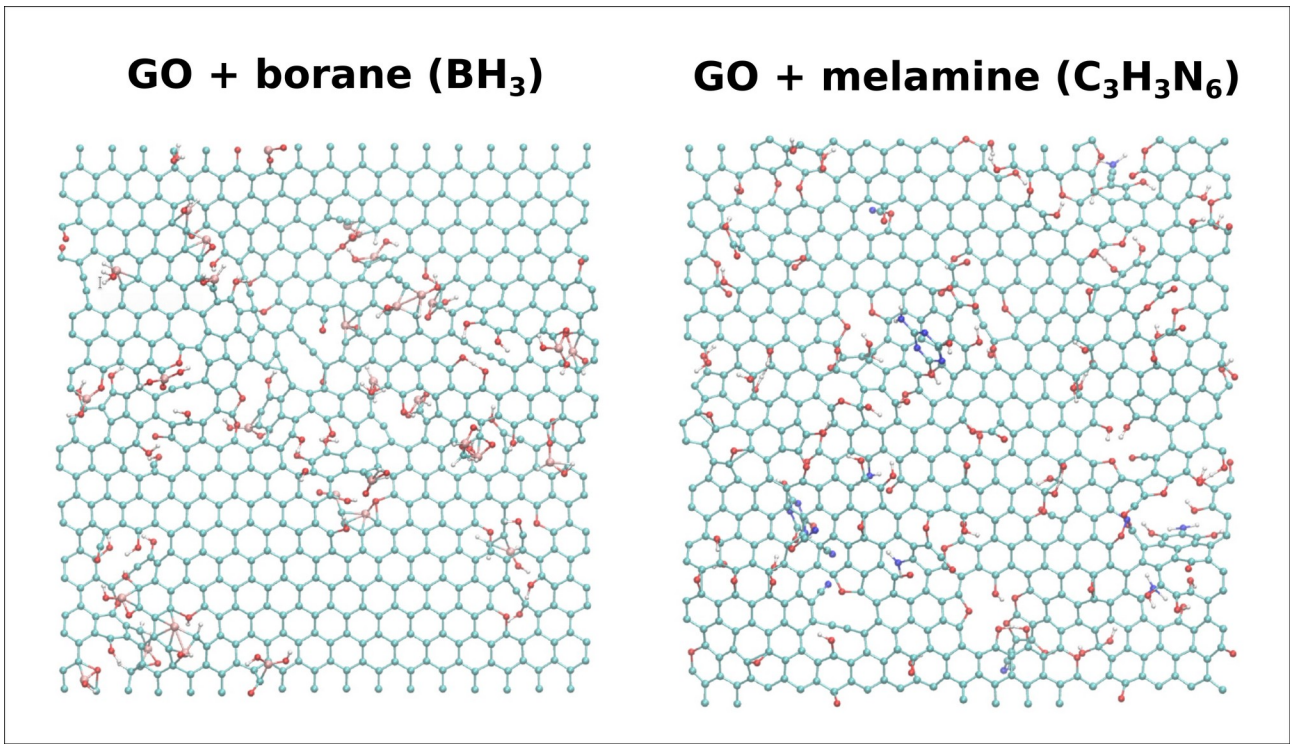

Figure S9: The structure of doped GO structures, annealed with borane and melamine, for B and N doping, respectively.