Density Functional Theory Studies on Graphene/h-Boron Nitride Hybrid Nanosheets for Supercapacitor Electrode Applications

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Supplementary information

Table S1 Bader Charge analysis of optimized nanosheets.

Structure	C – charge	B – Charge	N – charge
C32	C1: 0.134		
	C2: -0.237		
	C3: 0.004		
	C4: -0.001		
	C5: 0.004		
	C6: -0.001		
	C7: 0.110		
	C8: -0.002		
	C9: -0.092		
	C10: 0.108		
	C11: 0.000		
	C12: -0.011		
	C13: 0.001		
	C14: 0.001		
	C15: 0.002		
	C16: 0.001		
	C17: -0.104		
	C18: 0.106		
	C19: 0.002		
	C20: 0.001		
	C21: 0.005		

	C22: -0.002		
	C23: 0.005		
	C24: -0.001		
	C25: 0.105		
	C26: -0.102		
	C27: 0.004		
	C28: -0.001		
	C29: 0.004		
	C30: -0.002		
	C31: 0.004		
	C32: -0.002		
C24B4N4	C9: 0.098	B1: 2.319	N1: -2.235
	C10: 0.072	B2: 2.617	N2: -1.887
	C12: -0.029	B3: 2.015	N3: -2.600
	C18: -0.015	B4: 2.316	N4: -2.238
	C21: 0.087		
	C22: 0.075		
C20B6N6	C11: 0.143	B1: 2.331	N1: -2.218
	C12: 0.004	B2: 2.609	N2: -2.239
	C14: -0.012	B3: 2.626	N3: -1.963
	C18: 0.118	B4: 2.032	N4: -2.585
	C19: -0.107	B5: 2.297	N5: -2.582
	C20: 0.105	B6: 2.301	N6: -2.332
C16B8N8	C1: -0.852	B1: 1.355	N1: -3.587
	C3: -0.853	B2: 1.355	N2: -3.588

	C4: -0.837	B3: 1.355	N3: -3.587
	C5: -0.853	B4: 1.355	N4: -3.587
	C6: -0.857	B5: 1.619	N5: -3.382
	C7: -0.853	B6: 1.623	N6: -3.386
	C8: -0.857	B7: 1.620	N7: -3.383
		B8: 1.620	N8: -3.383
C8B12N12	C3: -0.852	B1: 2.325	N1: -2.650
	C4: -0.853	B2: 2.665	N2: -2.277
	C5: -0.837	B3: 2.318	N3: -2.598
	C6: -0.853	B4: 2.644	N4: -2.277
	C7: -0.857	B5: 2.649	N5: -2.274
	C8: -0.853	B6: 2.664	N6: -2.275
		B7: 2.707	N7: -2.641
		B8: 2.694	N8: -2.673
		B9: 2.317	N9: -2.605
		B10: 2.324	N10: -2.649
		B11: 2.704	N11: -2.712
		B12: 2.703	N12: -2.639
C4B14N14	C1: -0.512	B1: 2.323	N1: -2.661
	C2: 0.230	B2: 2.633	N2: -2.676
	C3: -0.367	B3: 2.674	N3: -2.396
	C4: 0.439	B4: 2.461	N4: -2.611
		B5: 2.632	N5: -2.629
		B6: 2.665	N6: -2.386
		B7: 2.653	N7: -2.285

	B8: 2.705	N8: -2.449
	B9: 2.718	N9: -2.657
	B10: 2.707	N10: -2.688
	B11: 2.321	N11: -2.650
	B12: 2.701	N12: -2.716
	B13: 2.718	N13: -2.712
	B14: 2.715	N14: -2.651
B16N16	B1: 1.659	N1: -3.656
	B2: 1.659	N2: -3.655
	B3: 1.659	N3: -3.655
	B4: 1.659	N4: -3.655
	B5: 1.658	N5: -3.655
	B6: 1.659	N6: -3.650
	B7: 1.659	N7: -3.655
	B8: 1.659	N8: -3.655
	B9: 1.658	N9: -3.655
	B10: 1.654	N10: -3.655
	B11: 1.659	N11: -3.655
	B12: 1.689	N12: -3.655
	B13: 1.689	N13: -3.655
	B14: 1.689	N14: -3.655
	B15: 1.689	N15: -3.655
	B16: 1.689	N16: -3.655

Method for calculating the effective mass

The effective mass of the electron and hole is calculated by fitting a parabolic curve to the *E* – *K* diagram (Band structure) obtained from DFT calculation.

In general, effective mass is given as, $$m^* = \frac{\hbar^2}{\frac{d^2E}{dK^2}}$$

Also, the dispersion relation *E(K)* compensates for the internal force due to the crystal, which allows us to use the classical concepts for the electron as long as its mass is taken as m^*. So, from the *E* – *K* diagram, the $\frac{d^2E}{dK^2}$ is obtained by fitting a parabolic curve at the required high symmetry point near the Fermi region. In particular, valence band maximum (VBM) and conduction band minimum (CBM) are considered for our calculation.

The parabolic curve fit in terms of *E* – *K* is written as,

$$E = A_o K^2 + A_1 K + A_2$$

On differentiating E concerning K twice, we get, $\frac{d^2E}{dK^2} = 2A_o$. Therefore, the effective mass in terms of parabolic curve fit is given as, $m^* = \frac{\hbar^2}{2A_o}$.

The coefficient A_o is obtained by fitting the curves, and just from this, we have calculated the effective mass of electrons and holes at VBM and CBM, respectively.

Dimension of A_o:

We know, $D[E] = eV$, $D[K] = \frac{1}{\text{Å}}$ as $K = \frac{2\pi}{\lambda}$ and $\frac{d^2E}{dK^2} = 2A_o$

So, $D[A_o] = eV(\text{Å})^2 = 1.6 \times 10^{-39} Jm^2$

From which, we can obtain effective mass in terms of m_e

$$m^* = \frac{\hbar^2}{2A_o} = \frac{3.445 \times 10^{-30}}{A_o} kg = \frac{3.786}{A_o} m_e$$

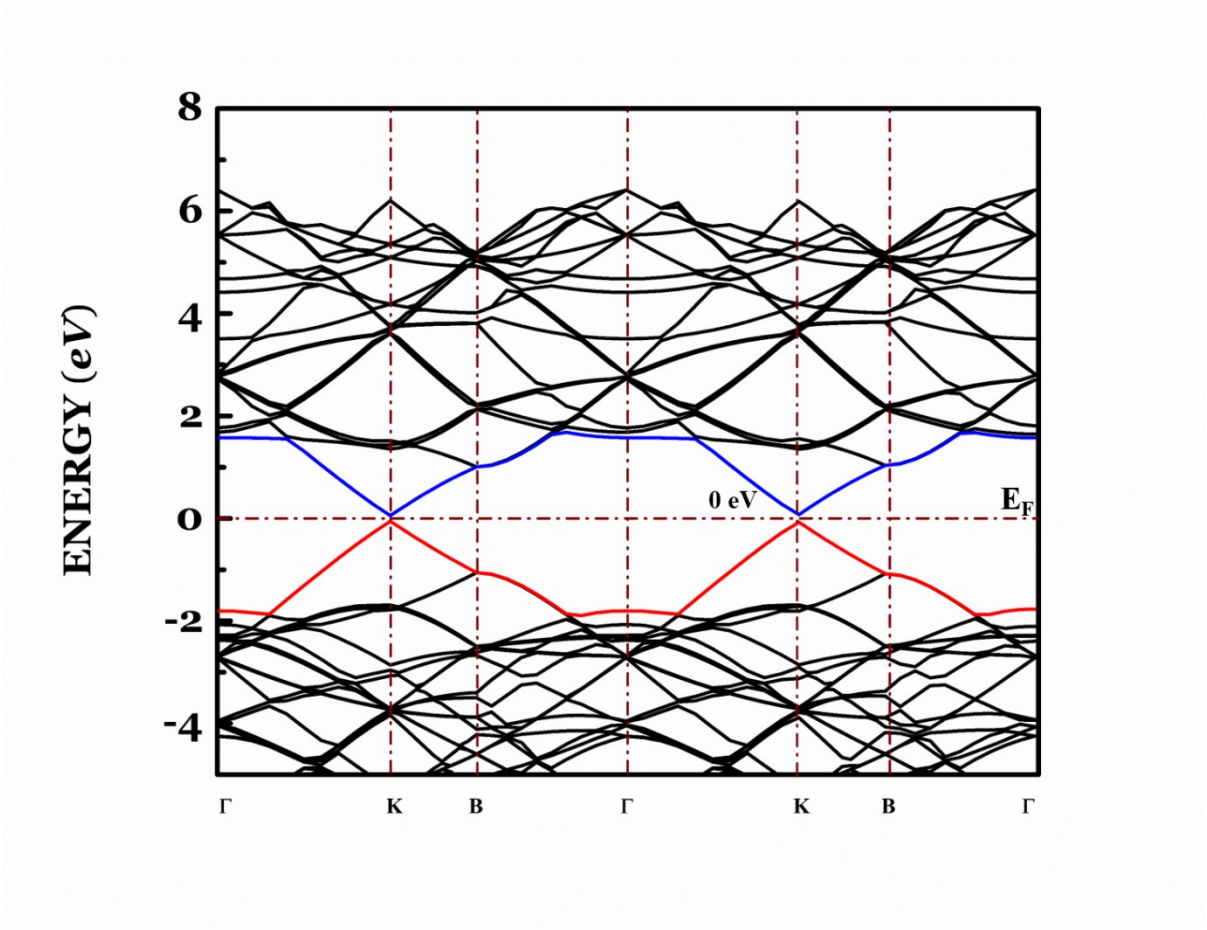
Table S2 Excessive surface charge density (Σ) of optimized nanosheets.

Structure	Σ at -1ev ($\mu C\ cm^{-2}$)	Σ at +1ev ($\mu C\ cm^{-2}$)
C32	-1.110	1.200
C24B4N4	-4.239	0.132
C20B6N6	-5.349	0.311
C16B8N8	-3.591	2.204
C8B12N12	-3.519	0.078
C4B14N14	-5.790	0.183
B16N16	-17.583	0.131

Table S3 Quantum capacitance (C_q) of optimized nanosheets.

Structure	Potential (eV)	C_q ($\mu F\ cm^{-2}$)
C32	1.00	2.629
C24B4N4	-0.53	9.128
C20B6N6	-0.64	14.950
C16B8N8	0.97	16.655
C8B12N12	-0.28	11.015
C4B14N14	-0.99	22.518
B16N16	-0.65	31.539

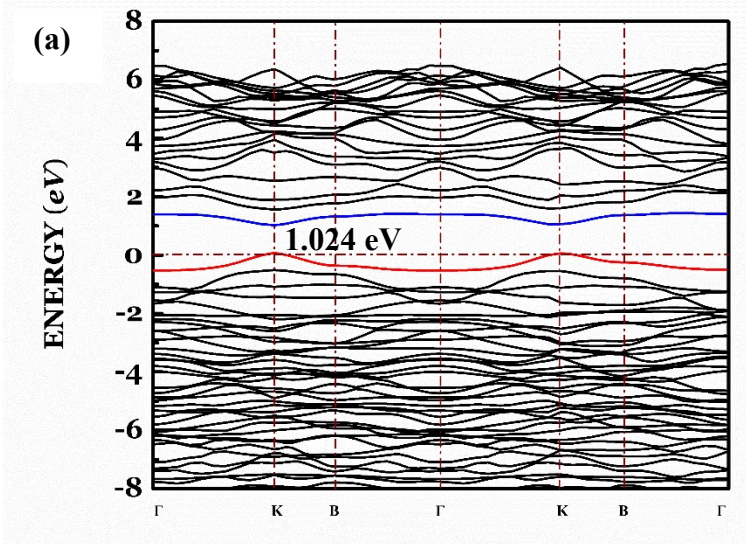
Fig. S1 Band structure of pristine graphene

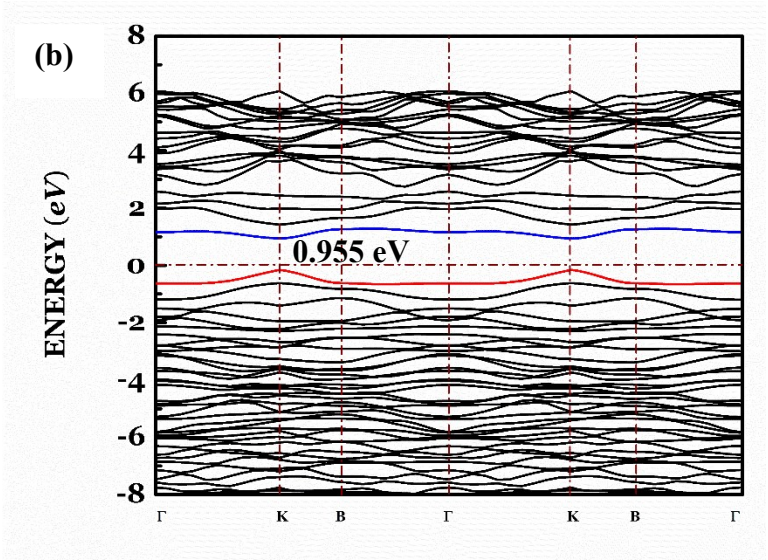


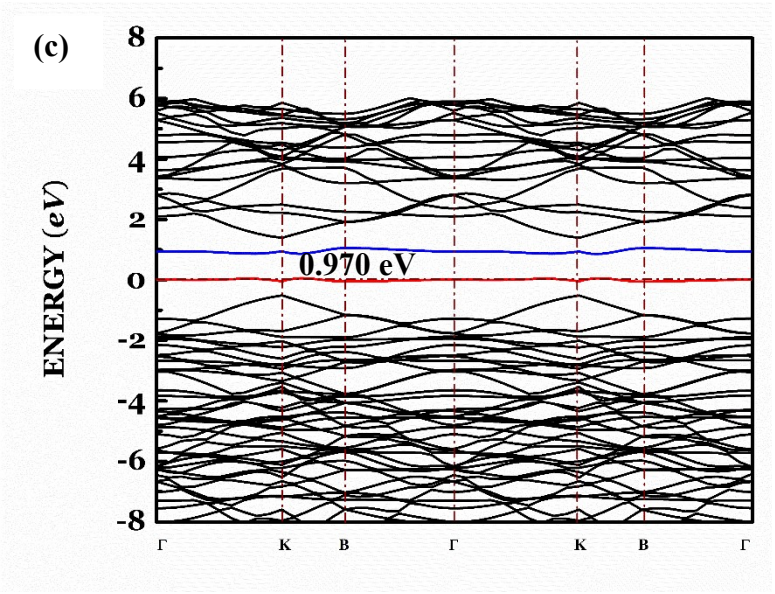
Conduction Band Minimum (CBM)

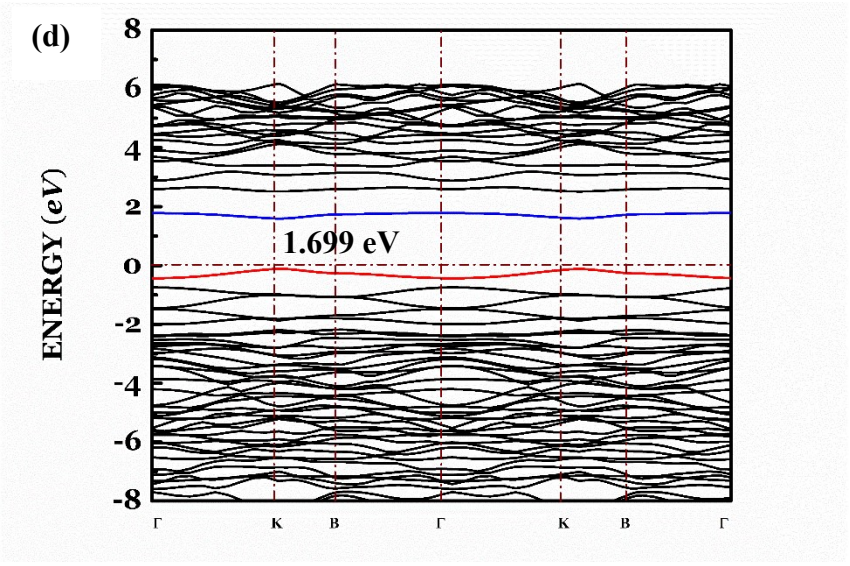
Valence Band Maximum (VBM)

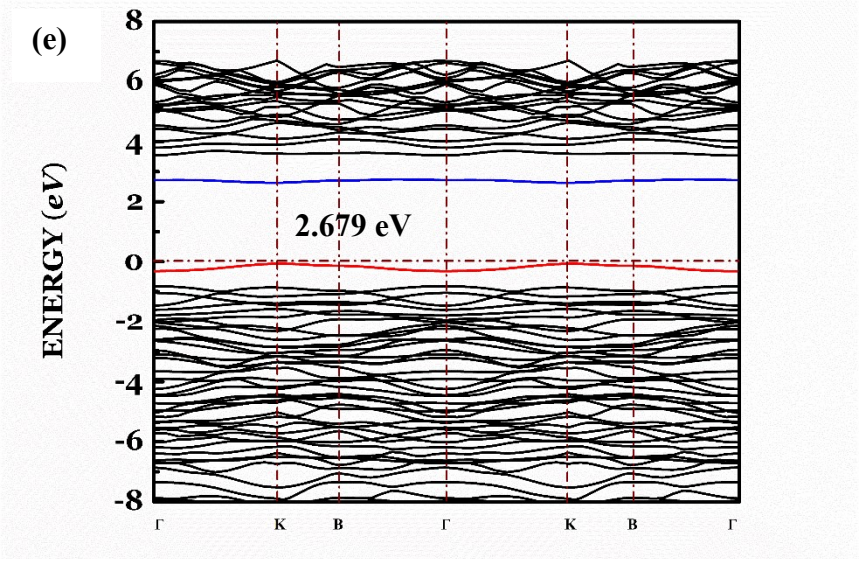
Fig. S2 Band structure of (a) C24B4N4, (b) C20B6N6, (c) C16B8N8, (d) C8B12N12, (e) C4B14N14, and (f) B16N16 nanosheets.

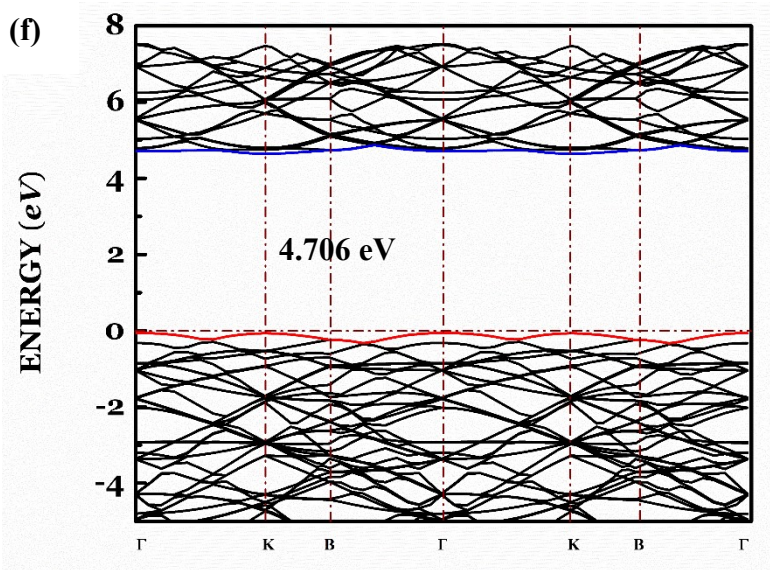





(c)
ENERGY (eV)
0.970 eV
8
6
4
2
0
-2
-4
-6
-8
Γ
K
B
Γ
K
B
Γ


(d)
ENERGY (eV)
1.699 eV
8
6
4
2
0
-2
-4
-6
-8
Γ
K
B
Γ
K
B
Γ


(e)
ENERGY (eV)
2.679 eV
8
6
4
2
0
-2
-4
-6
-8
Γ
K
B
Γ
K
B
Γ



Conduction Band Minimum (CBM)

Valence Band Maximum (VBM)