

Supporting information:

A multiporous carbon family with superior stability, tunable electronic structures and amazing hydrogen storage capability

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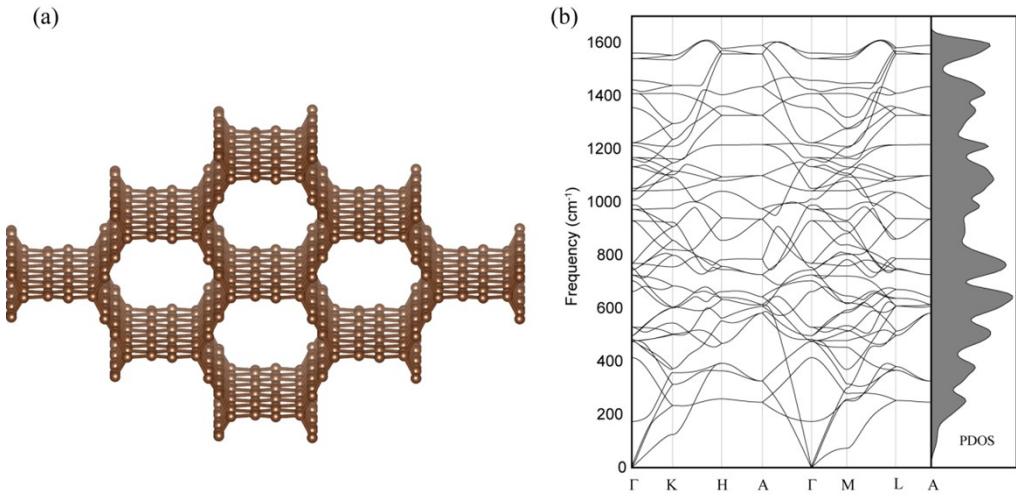


Figure S1. (a) 4-diaphene crystal structure. (b) Phonon dispersion and phonon density of states (PDOS). No imaginary phonon frequency along all high symmetry direction indicates its intrinsic dynamic stability.

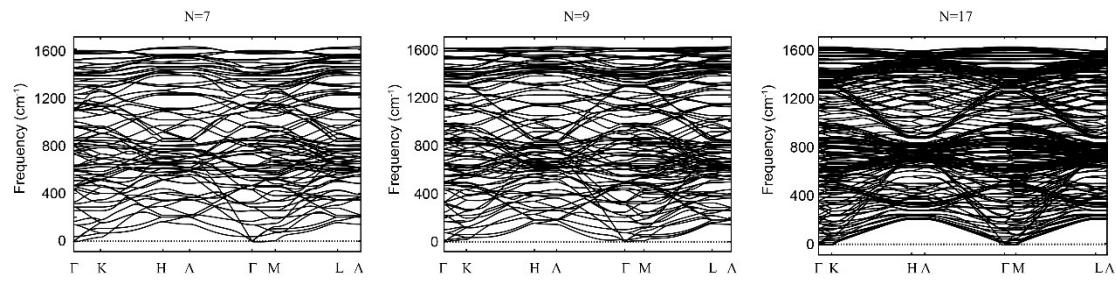


Figure S2. Phonon dispersion calculated using a $2 \times 2 \times 2$ supercell for 7-diaphenes and 9-diaphenes but a $1 \times 1 \times 1$ supercell for 17-diaphenes.

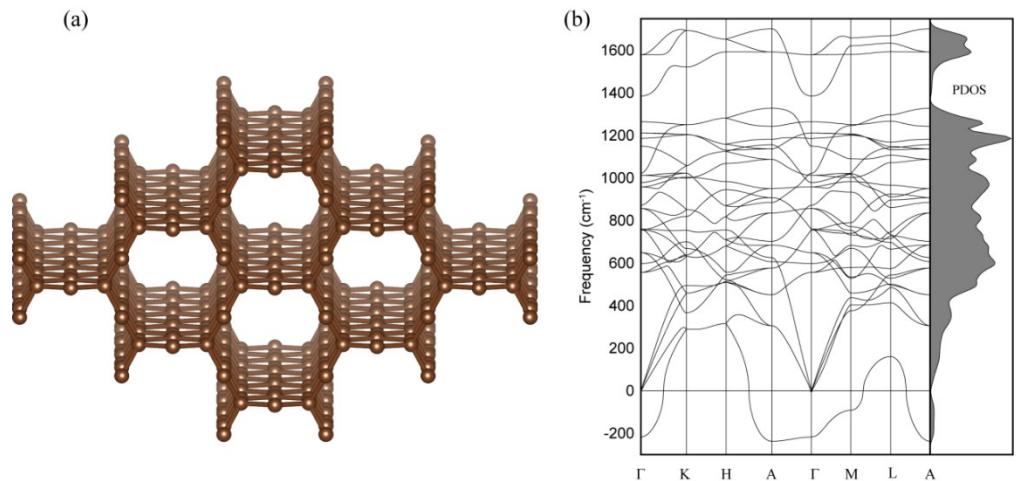


Figure S3. (a) 3-diaphene crystal structure. (b) Phonon dispersion and phonon density of states (PDOS). Broad range of wave vectors with imaginary phonon frequency along Γ -K, Γ -A, Γ -M and A-L, indicates its dynamical instability.

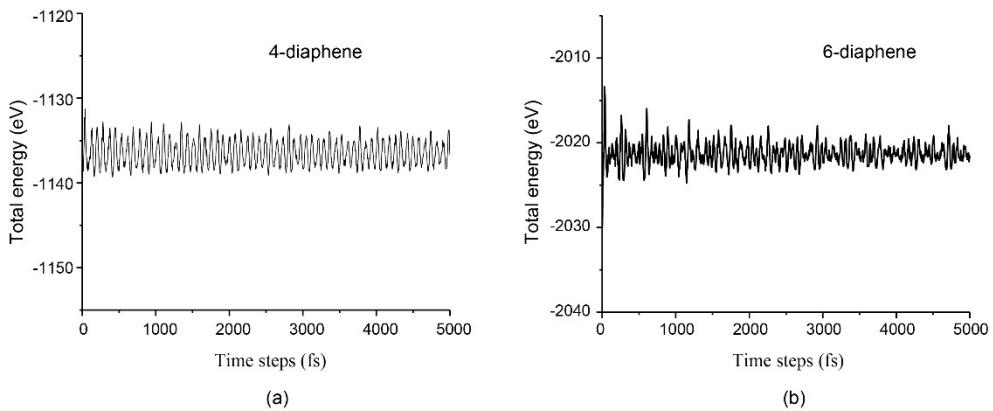


Figure S4. NVT molecular dynamics simulations for 4-diaphene (a) and 6-diaphene (b) at 300K.

Table S1 and Table S2 are stiffness and compliance coefficients, respectively. Angular anisotropy Young's modulus (E_θ) and shear modulus (G_θ) can be calculated by the following equations given by Voigt.^[1]

$$E_\theta = 1/[S_{11} \cdot \sin^4\theta + S_{33} \cdot \cos^4\theta + (2S_{13} + S_{44}) \cdot \cos^2\theta \cdot \sin^2\theta] \quad (S1)$$

$$G_\theta = 1/[S_{44} + (S_{11} - S_{12} - 0.5S_{44}) \cdot \sin^2\theta + 2(S_{11} + S_{33} - 2S_{13} - S_{44}) \cdot \cos^2\theta \cdot \sin^2\theta] \quad (S2)$$

In Eqs. (S1) and (S2), the original $(1-\cos^2\theta)$ terms have been replaced by $\sin^2\theta$. Note that when θ equals 0° and 90° , Eq. (S1) shows that Young's modulus E_θ is $1/S_{33}$ and $1/S_{11}$, respectively. Similarly, the shear modulus G_θ is $1/S_{44}$ at 0° and $2/(S_{44} + 2S_{11} - 2S_{12})$ at 90° , respectively.

Table S1. Stiffness coefficients (GPa)

Direction	xx	yy	zz	xy	yz	zx
xx	186.72	174.73	42.94	0.00	0.00	0.00
yy	174.73	186.72	42.94	0.00	0.00	0.00
zz	42.94	42.94	621.91	0.00	0.00	0.00
xy	0.00	0.00	0.00	150.07	0.00	0.00
yz	0.00	0.00	0.00	0.00	150.07	0.00
zx	0.00	0.00	0.00	0.00	0.00	5.99

Table S2. Compliance coefficients (0.01/GPa)

Direction	xx	yy	zz	xy	yz	zx
xx	4.31	-4.03	-0.02	0.00	0.00	0.00

yy	-4.03	4.31	-0.02	0.00	0.00	0.00
zz	-0.02	-0.02	0.16	0.00	0.00	0.00
xy	0.00	0.00	0.00	0.67	0.00	0.00
yz	0.00	0.00	0.00	0.00	0.67	0.00
zx	0.00	0.00	0.00	0.00	0.00	16.68

The mechanical stability is evaluated by Born-Huang criteria.^[2] For hexagonal structures, the stiffness coefficients in Table 1 satisfy $C_{11}>0$, $C_{33}>0$, $(C_{11}-C_{12})>0$, $C_{44}>0$ and $(C_{11}+C_{12})C_{33}-2C_{13}^2>0$. The Voigt bulk modulus B_V , shear modulus G_V and Young's modulus E_V are calculated by equations S3, S4 and S5 and they are 168.5 GPa, 110.2 GPa and 271.4 GPa, respectively.

$$B_V = (2C_{11} + C_{33} + 4C_{13} + 2C_{12})/9 \quad (\text{S3})$$

$$G_V = (3.5C_{11} + C_{33} - 2C_{13} - 2.5C_{12} + 6C_{44})/15 \quad (\text{S4})$$

$$E_V = \frac{9B_V G_V}{3B_V + G_V} \quad (\text{S5})$$

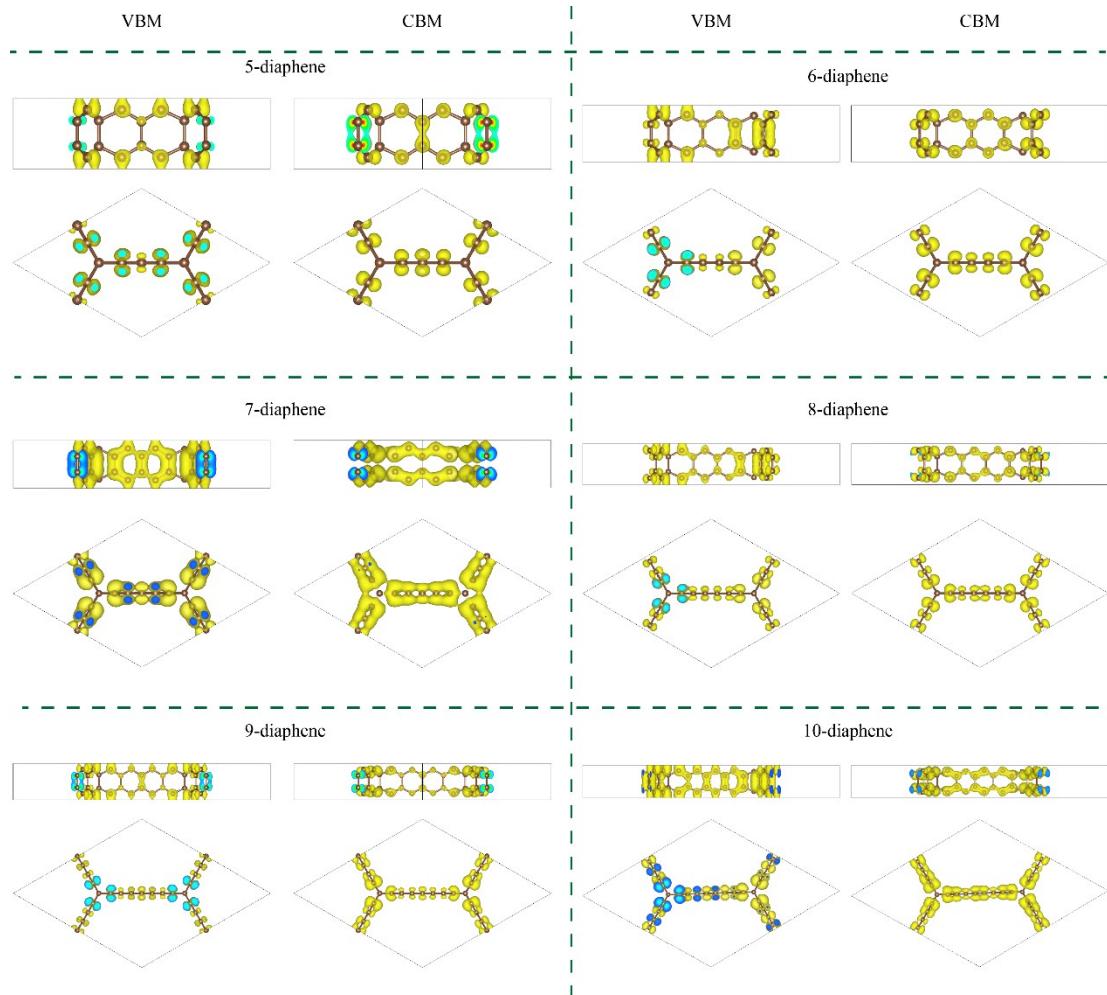


Figure S5. More localized valence band maxima (VBM) and conduction band minimum (CBM) charge densities for the semiconductive N -diaphenes and more delocalized VBM and CBM charge densities for the metallic N -diaphenes.

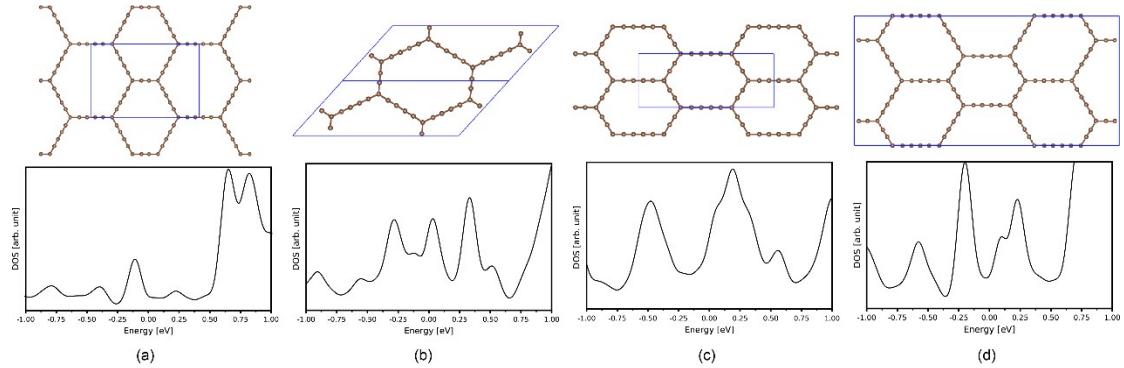


Figure S6. Top views of a few crystal structures ($N_1^m N_2^n$ -diaphene) built from armchair graphene nanoribbons (AGNRs) and their density of states (DOS). The N_1 and N_2 are feature size parameters. The m and n denotes the number of AGNRs with N_1 and N_2 parameters in the unit cell, respectively. The names of (a)-(d) structures are $4^1 6^5$ -diaphene, $4^3 6^3$ -diaphene, $4^4 6^2$ -diaphene and $4^4 6^{16}$ -diaphene, respectively.

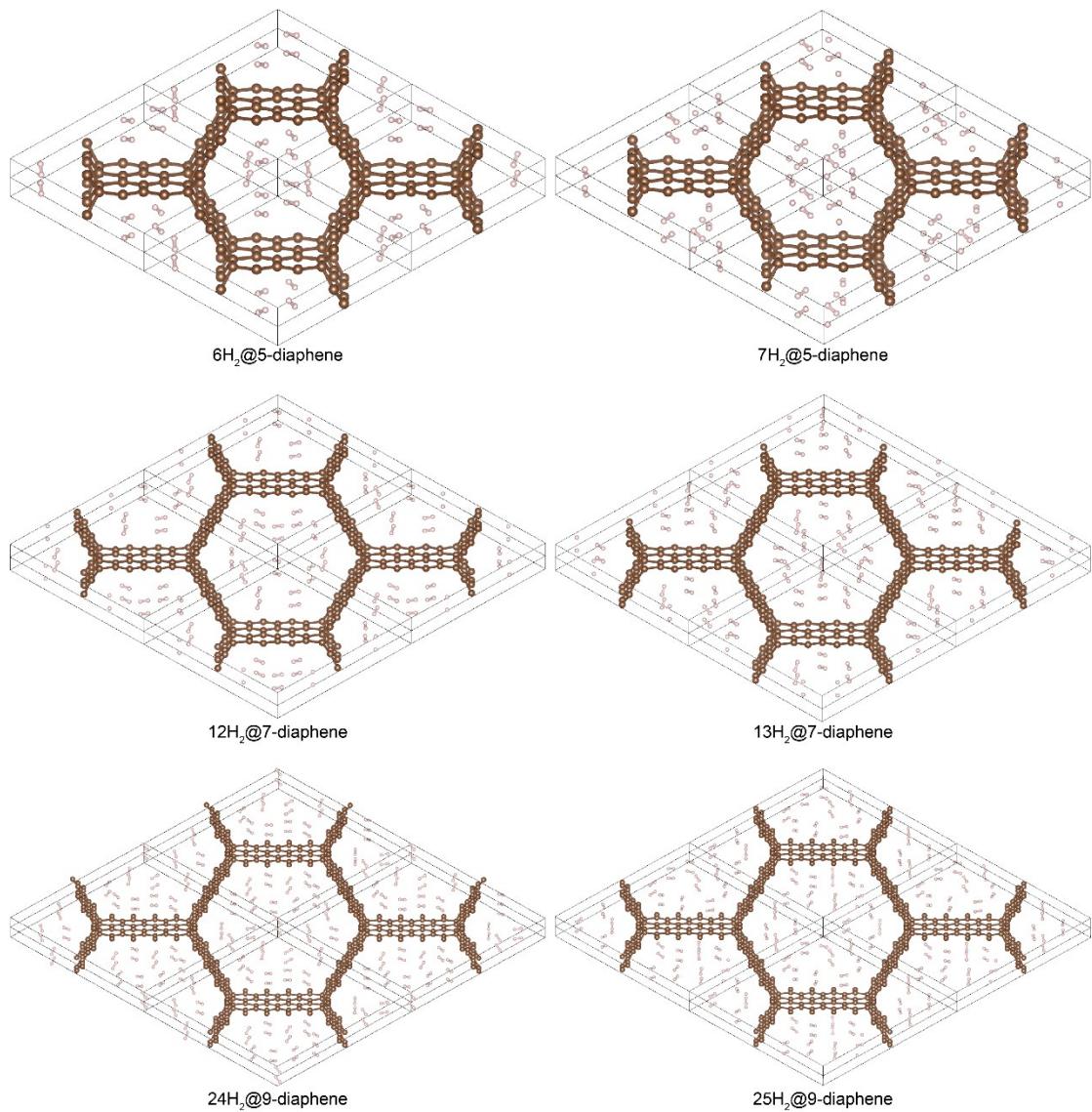


Figure S7. Adsorption geometries of H_2 molecules in N-diaphenes.

References

- [1] W. Voigt, "Lehrbuch Der Kristallphysik.", B. G. Teubner, Leipzig and Berlin, 1910; 2nd ed., 1928.
- [2] M. Born, K. Huang, Dynamical Theory of Crystal Lattices, Clarendon, Oxford, 1954.