Supplementary material

A nodal flexible-surface three-dimensional carbon network with potential applications in lithium-ion battery anode materials

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Climbing-image nudged elastic band (CI-NEB) method

The Climbing Image Nudged Elastic Band (CI-NEB) approach is a computational technique used in materials science, particularly in the study of Li-ion batteries. It's a variant of the Nudged Elastic Band (NEB) method, which is employed to find saddle points and transition paths between different states of a system. The NEB method works by approximating the transition path as a series of intermediate images between the initial and final states. Each of these images is a snapshot of the system's configuration along the path. The method then iteratively adjusts the positions of these images to minimize the energy along the path while keeping the images spaced evenly and the endpoints fixed. The energy of each image is minimized with respect to the path, but the energy of the images does not necessarily correspond to the actual energy barriers along the path. This can lead to inaccuracies, particularly when there are high energy barriers.

The Climbing Image approach addresses this issue by allowing one of the images to "climb" uphill along the energy profile. This climbing image seeks out the highest energy point along the path, which corresponds to the energy barrier for the transition. By focusing computational effort on this critical point, the method provides a more accurate determination of the transition energy barrier.

In the realm of Li-ion batteries, the CI-NEB method finds particular utility in investigating processes like Li-ion diffusion through electrode materials. This capability enables researchers to unravel intricate mechanisms governing battery performance, ultimately fostering the development of enhanced battery materials.

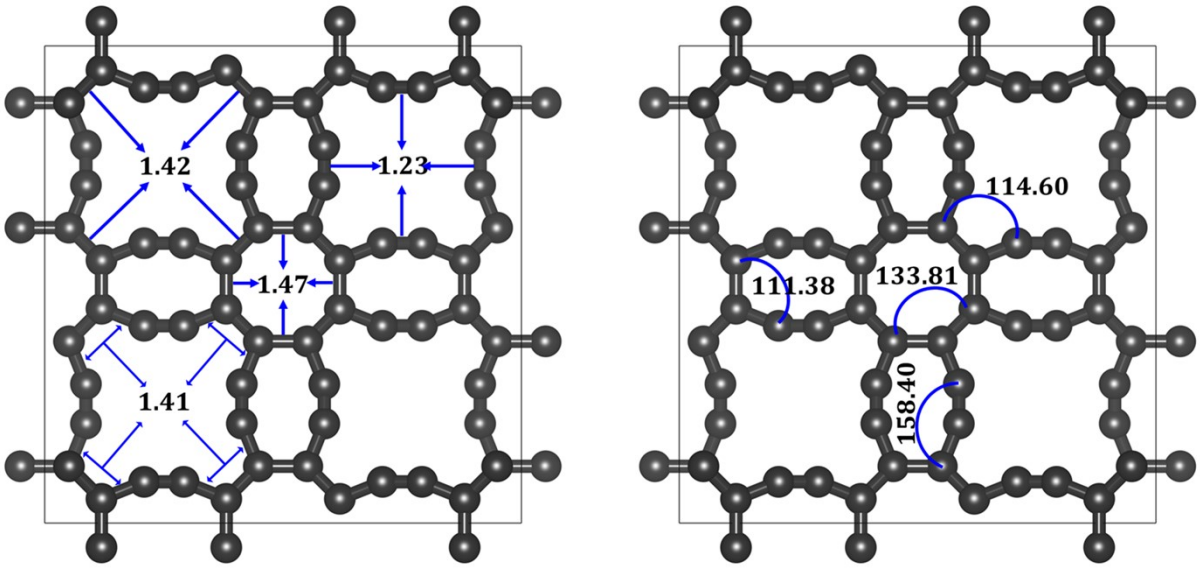


Figure S1. Equilibrium atomic bond lengths (Å) and bond angles (°) of 3D COT network.

A very tiny imaginary frequency (~ 3i cm^{-1}) around the Γ point of phonon band spectrum of 3D COT structure. We have computed phonon band spectrum of 3D COT structure using the 1 × 1 × 1 cell due to the presence of large number (64) of C atoms in the unit cell. The inclusion of 2 × 2 × 2 supercell for the computation of phonon band spectrum of 3D COT structure may eliminate these minute negative frequencies around the Γ point. Such calculations are computationally very demanding and require extensive computational efforts due to the presence of 512 carbon atoms with in the cell. However, previous studies show that the negative frequencies which are less than 10 cm^{-1} around can be ignored and the structure can be consider as dynamically stable.

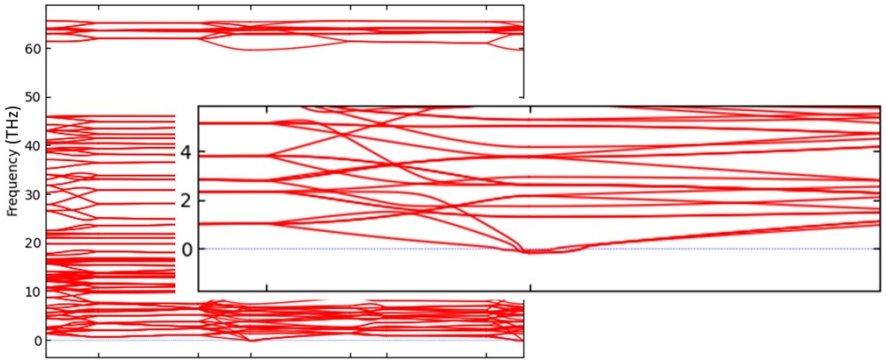


Figure S2. Tiny imaginary frequencies (~ 3i cm^{-1}) around the Γ-point.

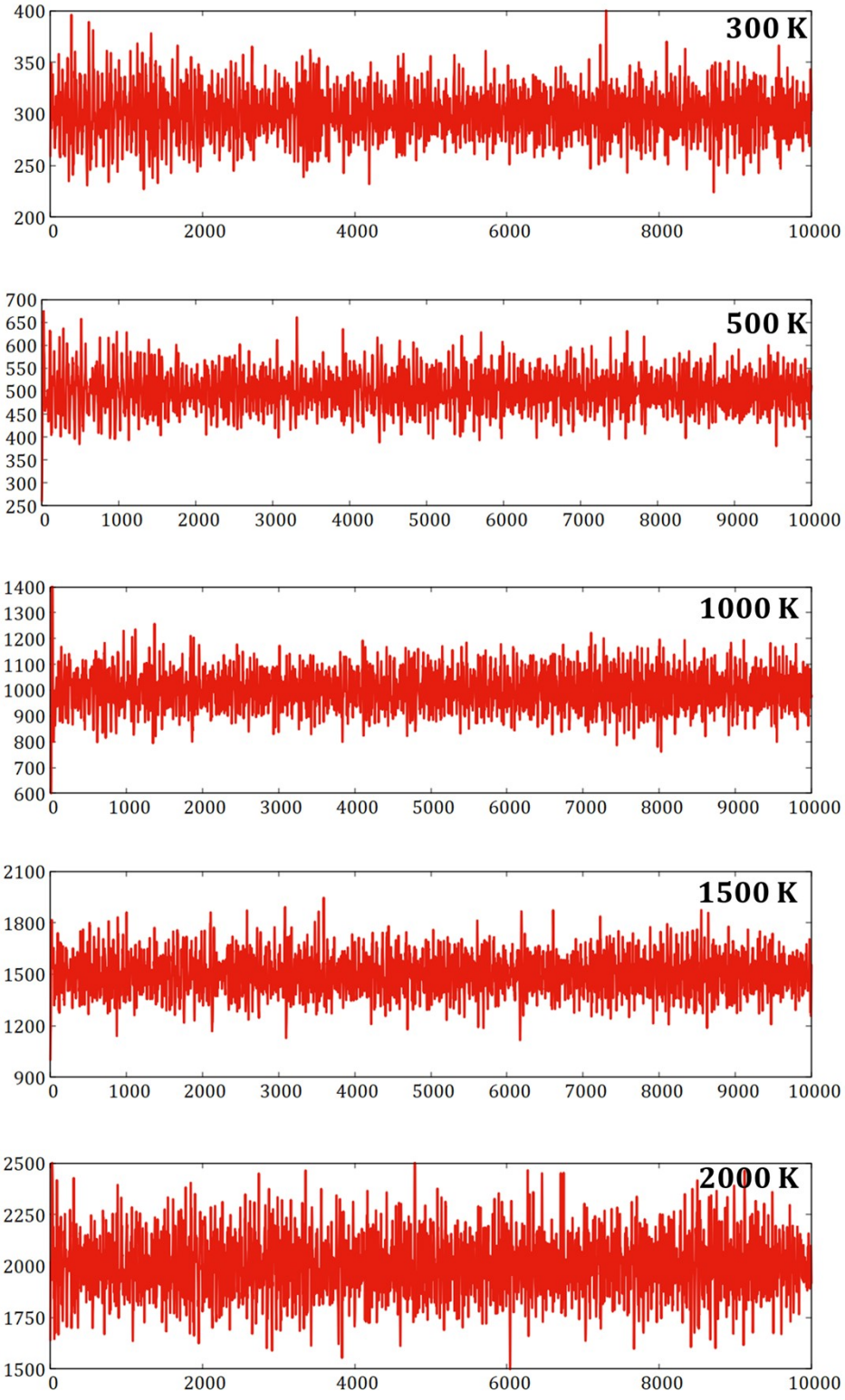


Figure S3. The fluctuation in temperature at 300, 500, 1000, 1500 and 2000 K during AIMD simulations.

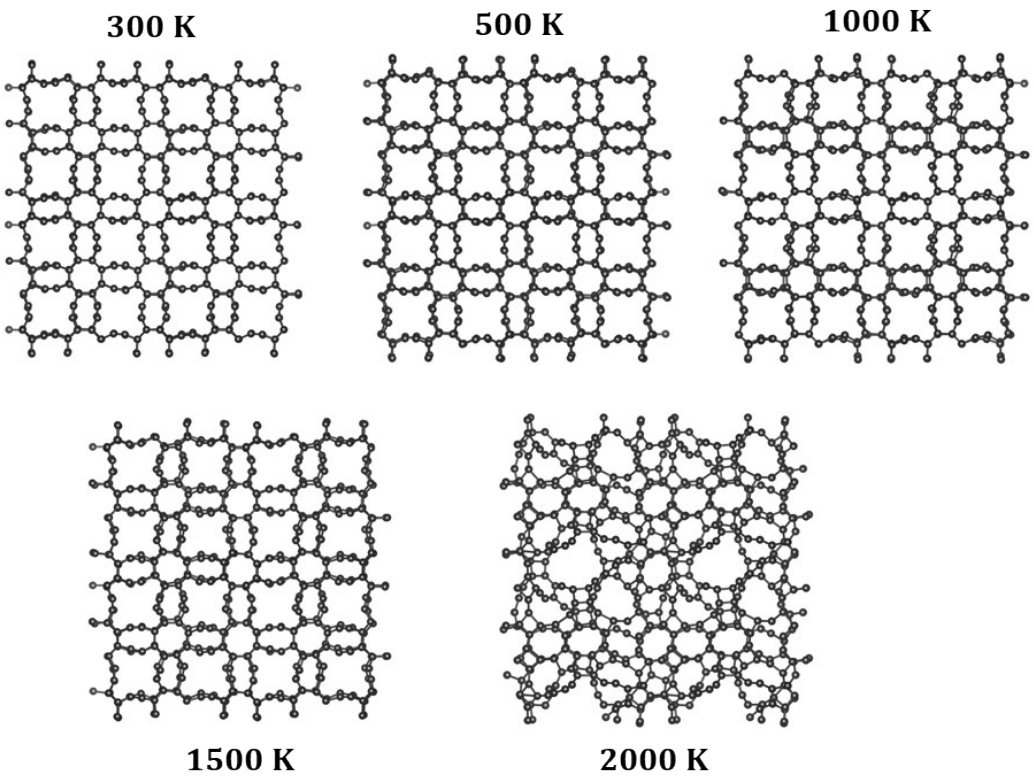


Figure S4. Snapshots of the 3D COTN at the end of the AIMD simulations at different temperatures.

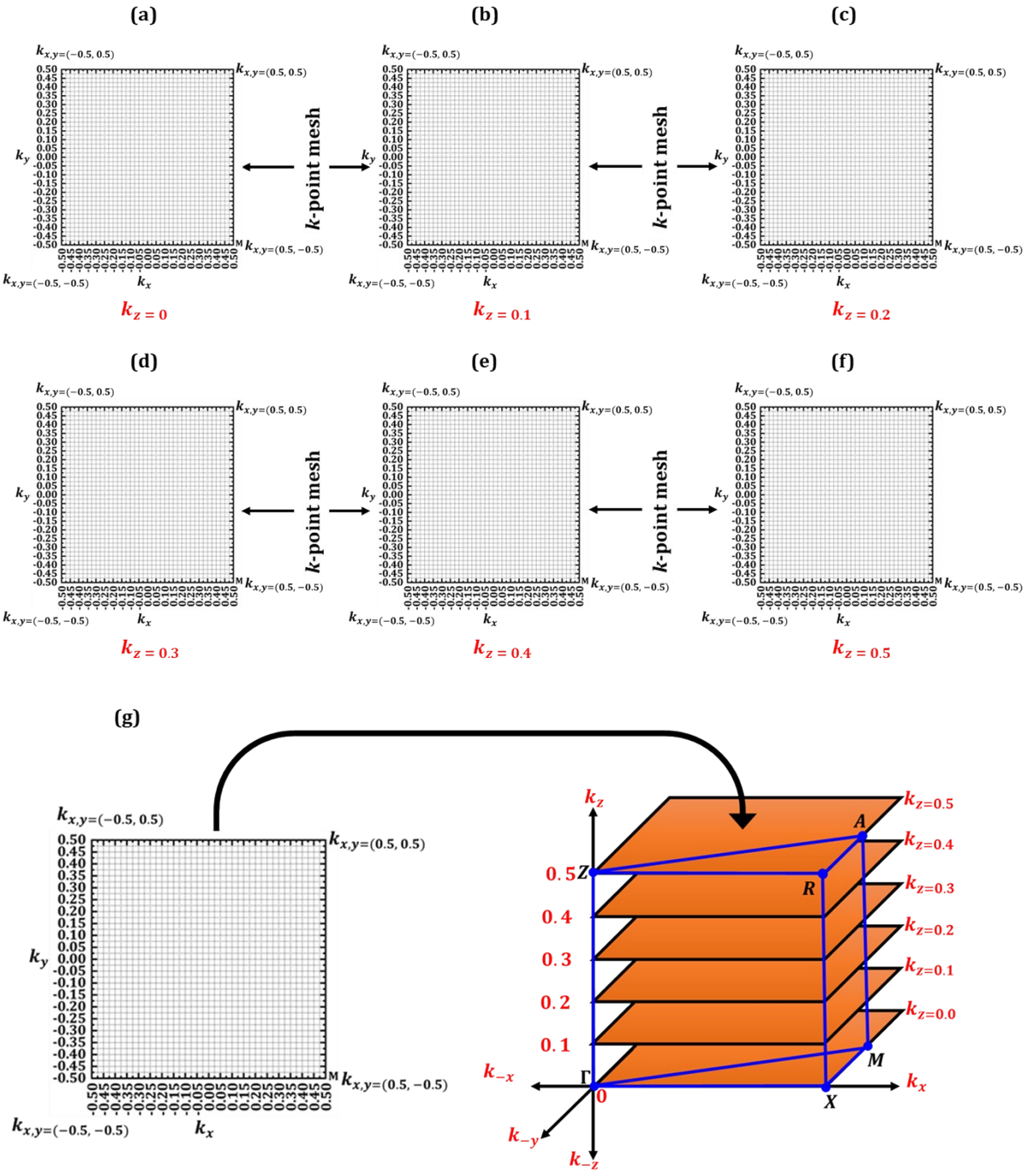


Figure S5. (a) The k_{xy}-plane is spanned with a finely gridded k-points in the entire tetragonal Brillouin zone by employing $k_z = 0.0$. The k_z value is changed to (b) 0.1, (c) 0.2, (d) 0.3, (e) 0.4 and (f) 0.5 to span the Brillouin zone with the k-points along the Z-axis. This procedure spans the entire tetragonal Brillouin zone with finely gridded k-points as in (g). In each case the Dirac nodal points were identified.

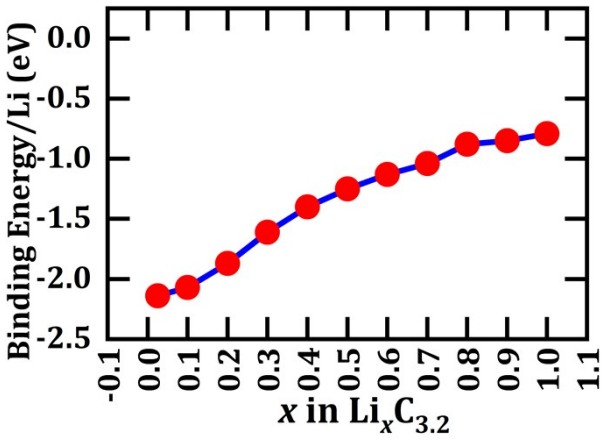
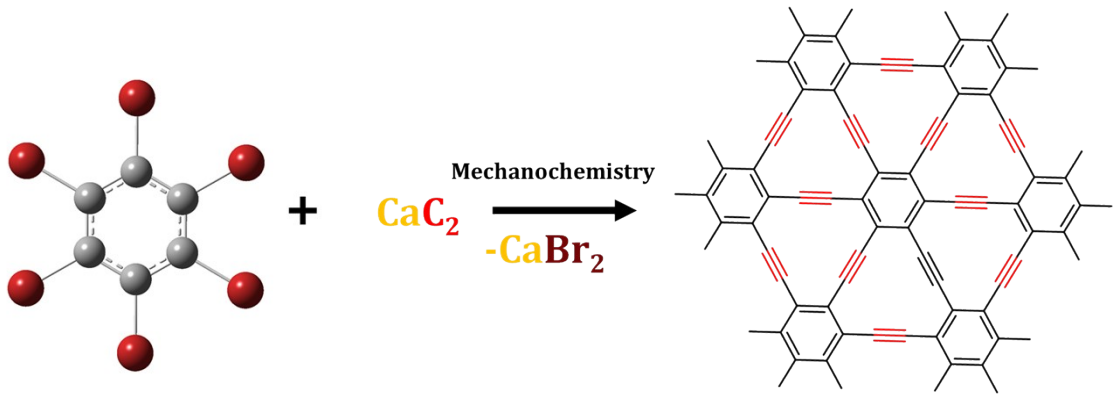


Figure S6. Average binding energy of Li atoms at different Li concentration in 3D COT network.



Synthetic route for experimental synthesis of γ-graphyne[1]

Figure S7. (a) The plausible synthetic procedure for the experimental fabrication of 3D COT network using octabromocyclooctatetraene and calcium carbide similar to that of (b) γ-graphyne which was fabricated using hexabromobenzene and CaC_2 as the precursors by ball-milling method.

Table S1. Equilibrium atomic density (ρ) of different carbon allotropes.

S. No	C allotrope	ρ(g/cm^3)
1	COTN	1.44
2	T-carbon	1.50
3	bct-C_{16}	2.17
4	Diamond	2.86
5	Graphite	2.22

Table S2. Computed fermi velocities of holes and electrons along different crystallographic directions.

Fermions	Fermi velocity (× 10^6 m/s)							
	D1		D2		D3		D4	
	D1⟶ Z	D1⟶ A	D2⟶ M	D2⟶ Γ	D3⟶ Γ	D3⟶ Z	D4⟶ X	D4⟶ Γ
holes	7.01	7.82	8.70	8.70	2.49	3.14	6.08	6.08
electrons	7.82	7.01	8.70	8.70	3.14	2.01	6.08	5.02

References:

(1) Li, Q.; Li, Y.; Chen, Y.; Wu, L.; Yang, C.; Cui, X. Synthesis of γ-Graphyne by Mechanochemistry and Its Electronic Structure. *Carbon N Y* **2018**, *136*, 248–254. https://doi.org/https://doi.org/10.1016/j.carbon.2018.04.081.