Supporting Information

Storage of Na in Layered Graphdiyne as High Capacity Anode Materials for Sodium Ion Batteries

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Fig. S1

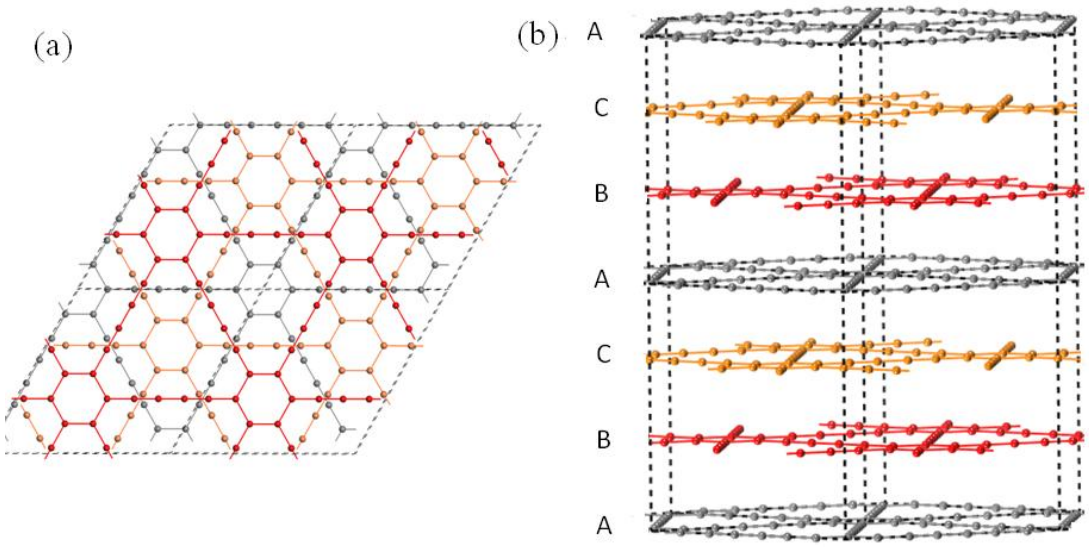


Fig. S1. Top and side views of the atomic structure of bulk GDY with ABC stacking. Grey, red and yellow balls represent carbon atoms in different layers.

Fig. S2

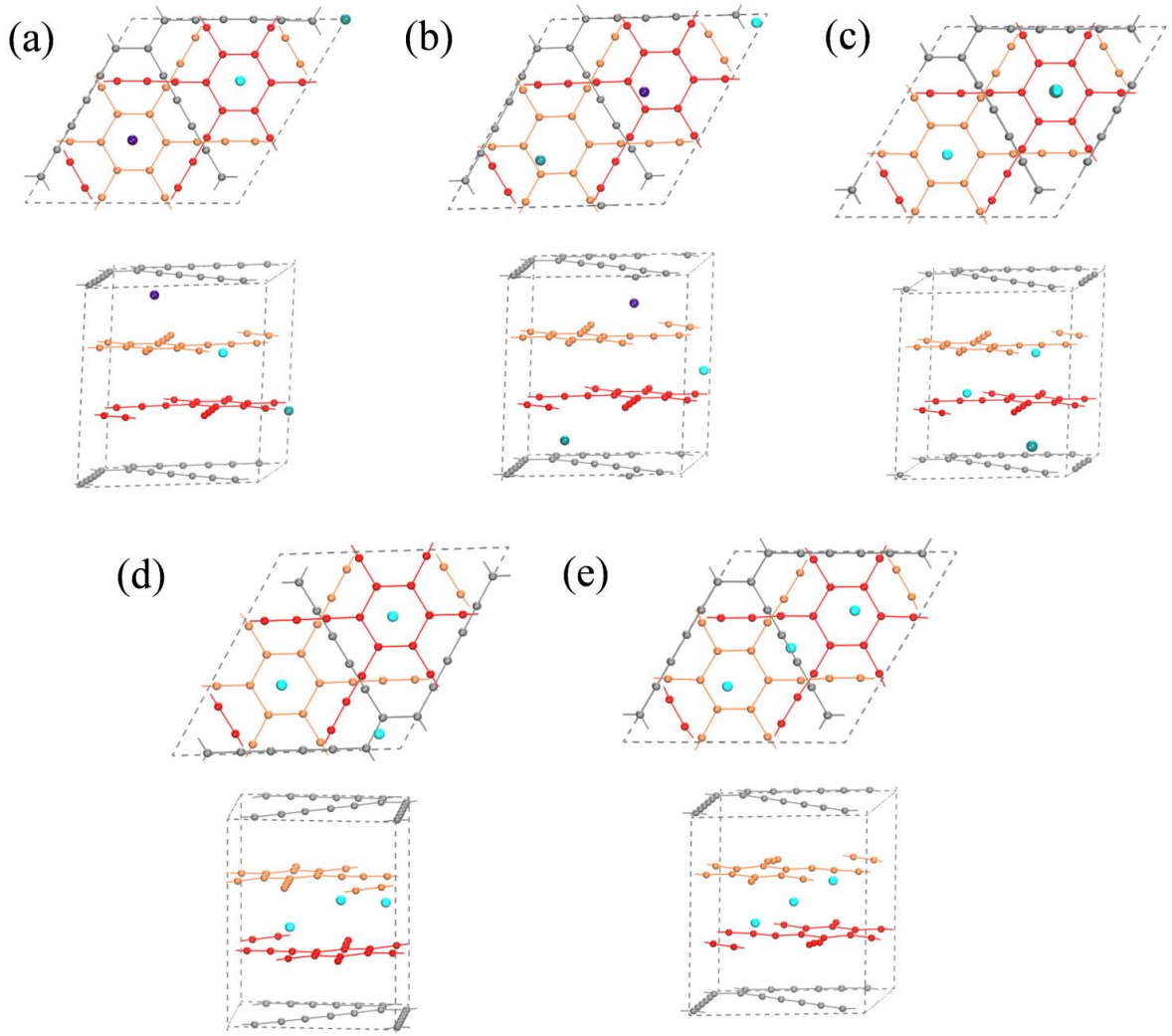


Fig. S2. The top and side view of various atomic structures of $Na_{0.056}C$ and the corresponding binding energies including (a) 2.93eV, (b) 2.82 eV, (c) 2.91 eV, (d) 2.84 eV, and (e) 2.50 eV, respectively.

Fig. S3

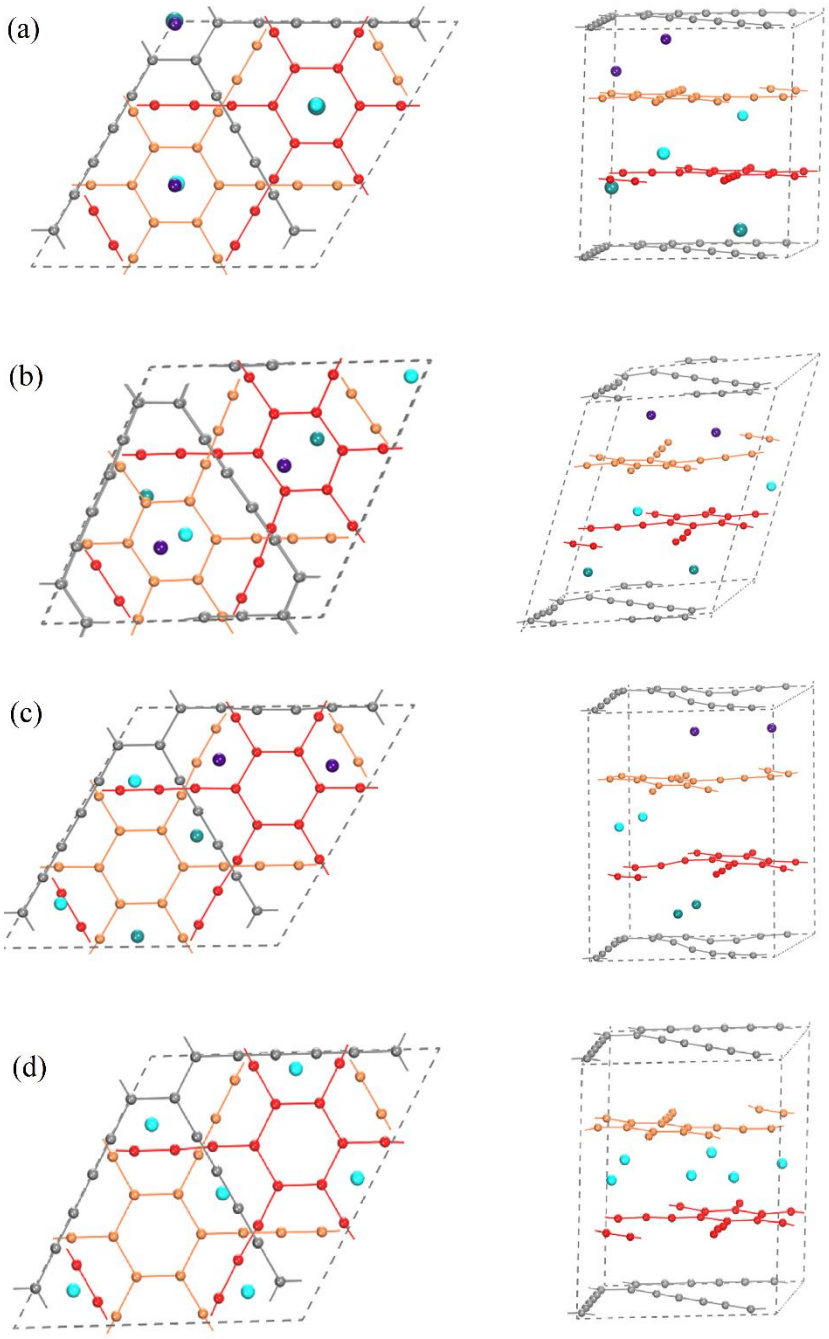


Fig. S3. The top and side view of various atomic structures of $Na_{0.11}C$ and the corresponding binding energies including (a) 2.75eV, (b) 2.62 eV, (c) 2.32 eV and (d) 2.12 eV, respectively.

Fig. S4

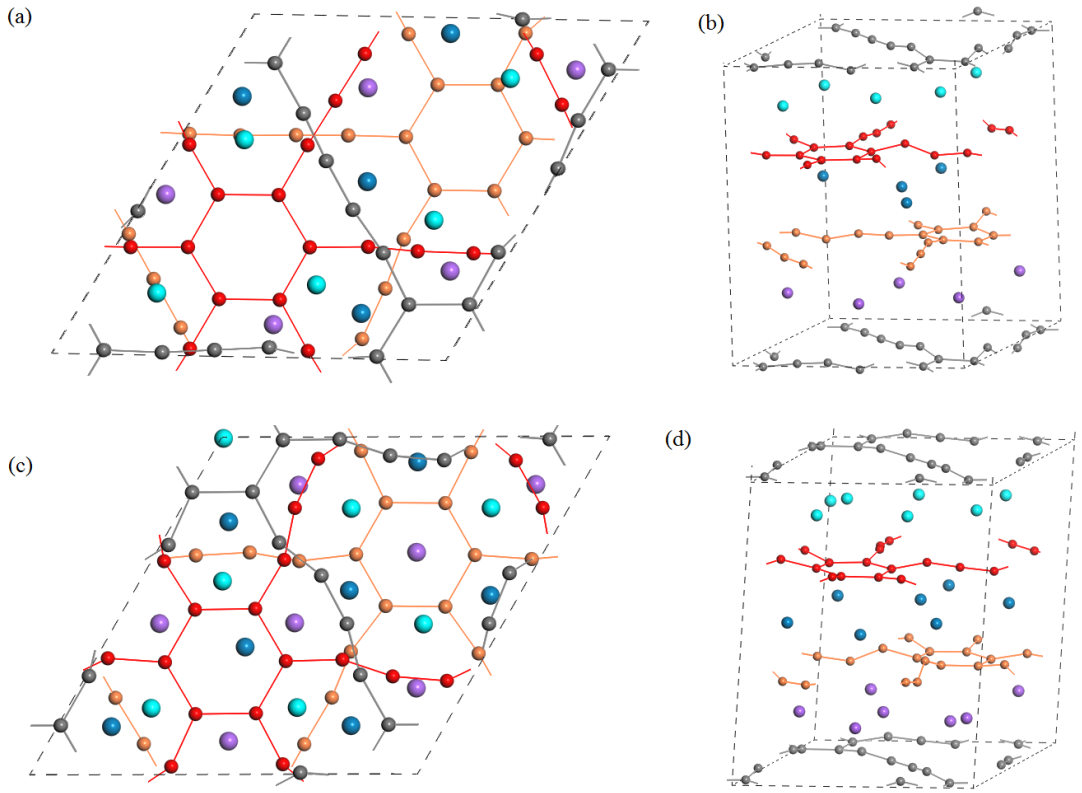
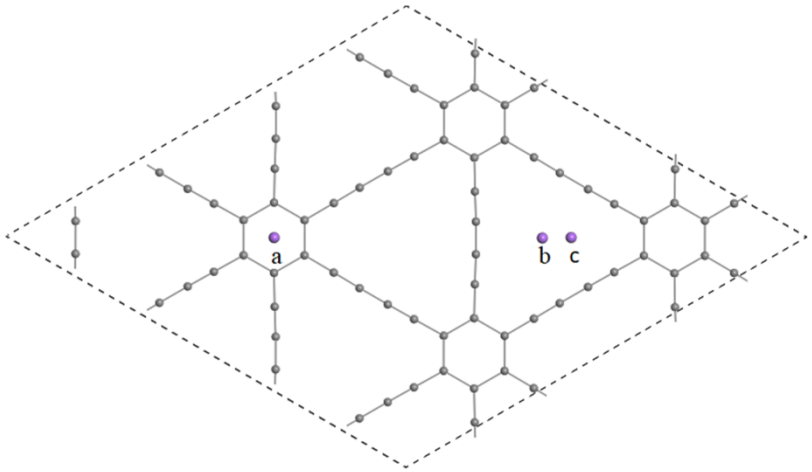


Fig. S4. The top and side views of atomic structures of low-energy stable Na_xC, including the models of (a, b) $Na_{0.26}C$ and (c, d) $Na_{0.39}C$.

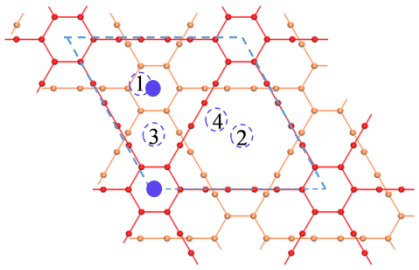
Fig. S5

(a)

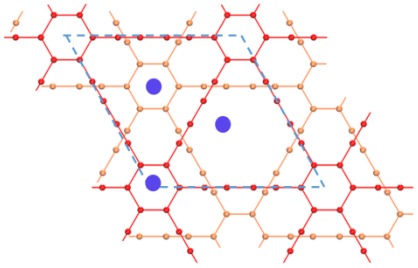


(b)

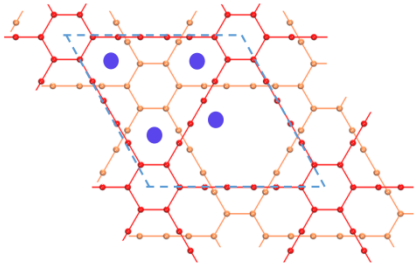
Step	1	2	3	4	5	6	7
Na number in one interlayer	2	3	4	6	7	8	……
Na number in three interlayers bulk GDY	6	9	12	18	21	24	……



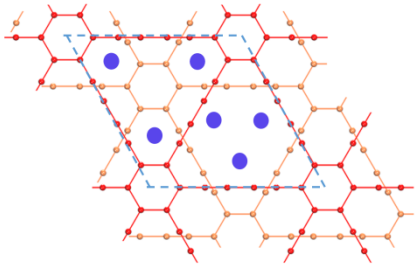
Step 1



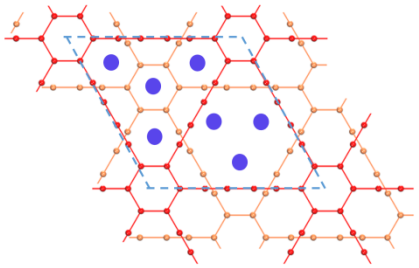
Step 2



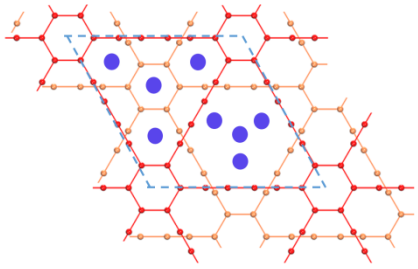
Step 3



Step 4



Step 5



Step 6

Fig. S5 (a) Top view of the atomic structure of single-layer GDY and the possible Na adsorption sites on single-layer GDY, and (b) schematic diagram of Na intercalation processes in bulk GDY. In (b), the schematic structure of each step is shown for Na intercalation in one interlayer of bulk GDY. In our simulation, the bulk GDY with ABC-stacking have three interlayers in one unite cell. In the table, the number of Na intercalated in each step is shown.

Table S1. The binding energies (E_b) between Na and single-layer GDY and corresponding distance between Na and carbon layer (in Fig. S3) for the adsorption of Na on single-layer DGY at different sites with the concentration of 0.056.

Site	a	b	c
Binding energy (eV)	1.59	2.53	2.08
Distance (Å)	2.15	0.00	1.38

Fig. S6

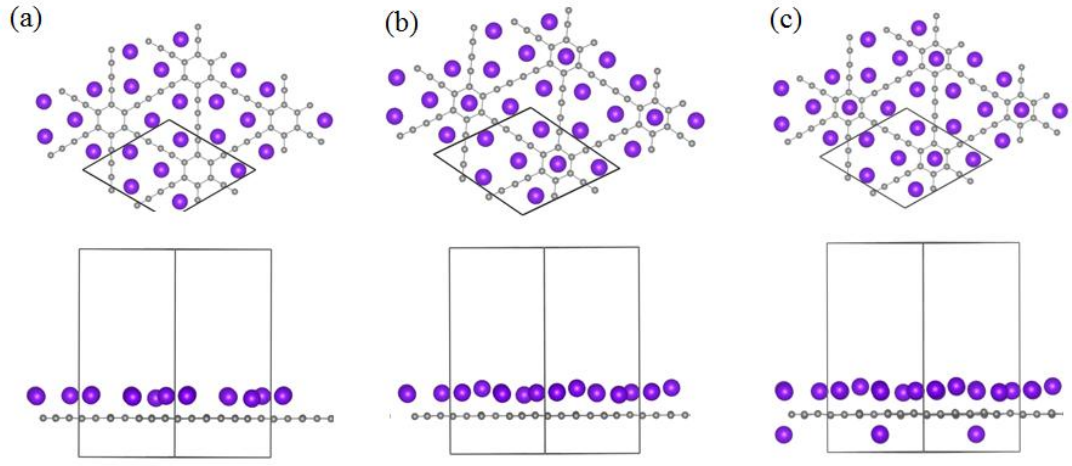


Fig. S6. The top and side views of atomic structures of Na-adsorbed single-layer GDY with the concentration of *x*= (a)0.333, (b)0.39, and (c) 0.444 (in a cell of 18 carbon atoms with 6, 7, and 8 Na atoms, respectively).

Fig. S7

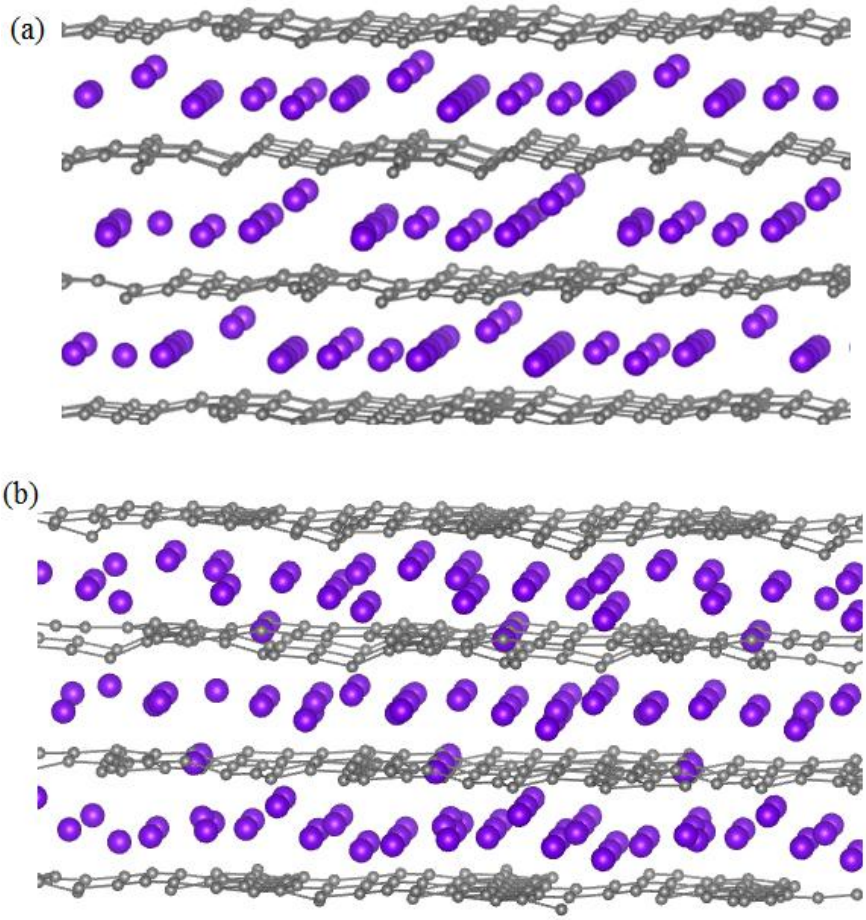


Fig. S7 The side views of atomic structures of Na-inserted GDY with the concentration of *x*= (a) 0.39, and (b) 0.444 (in a cell of 54 carbon atoms with 21 and 24 Na atoms, respectively)

Fig. S8

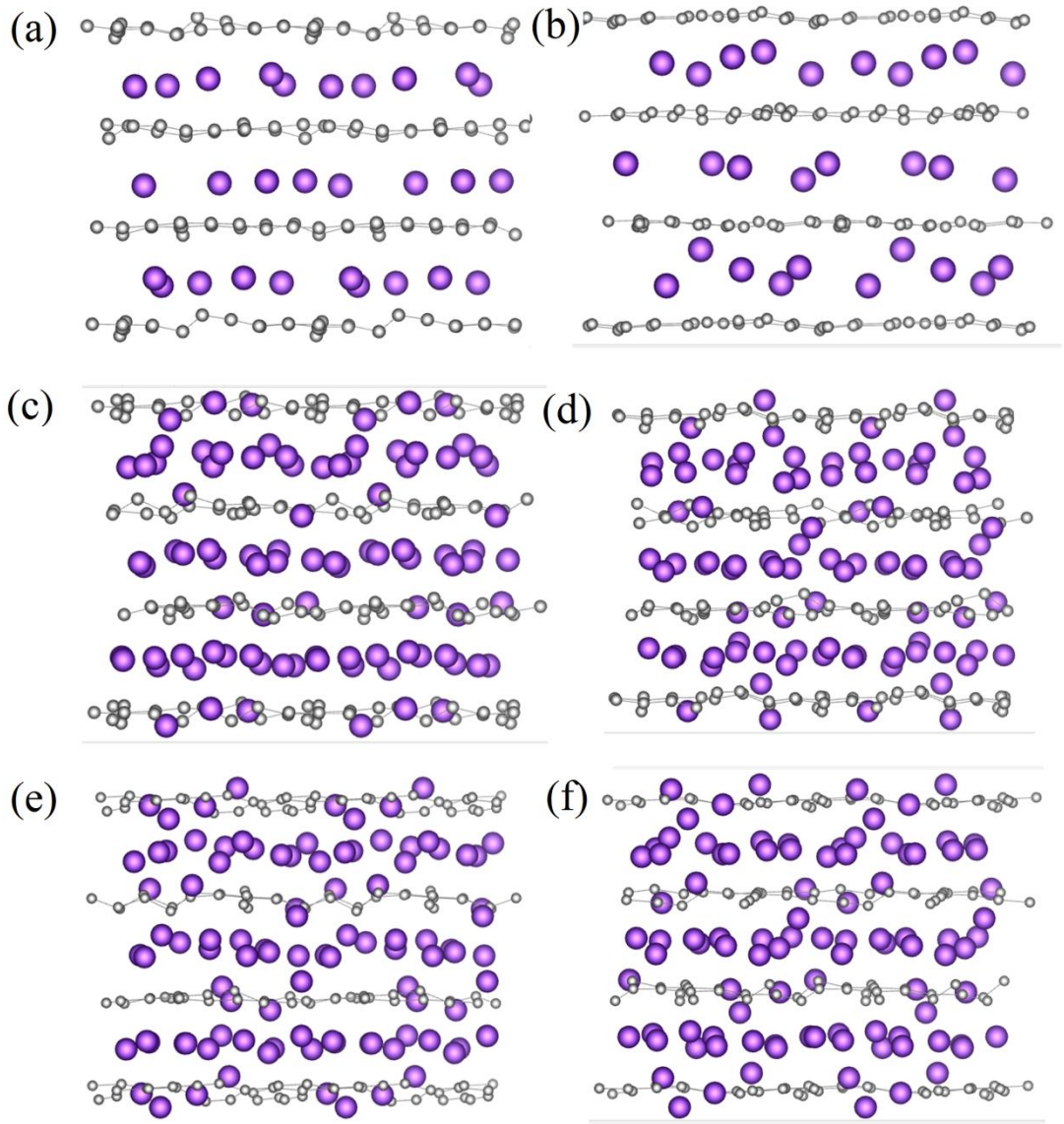


Fig. S8 The side views of atomic structures of GDY with Na concentration of (a, b) 0.26, (c, d) 0.78, and (e, f) 0.83 under the temperature of 0K and 300K.

Fig. S9

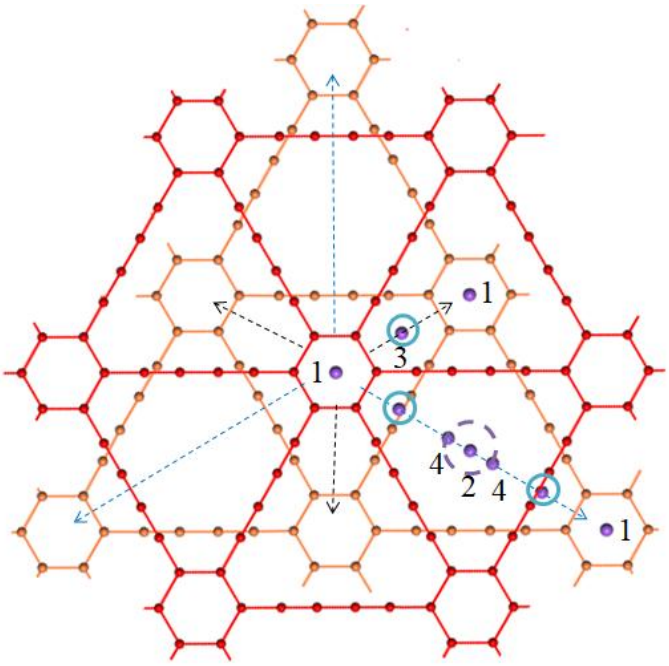


Fig. S9 Schematic diagram of diffusion paths of Na ion in the interlayer, including three equivalent path-1(blue dot line) and three equivalent path-2 (black dot line).

Fig. S10

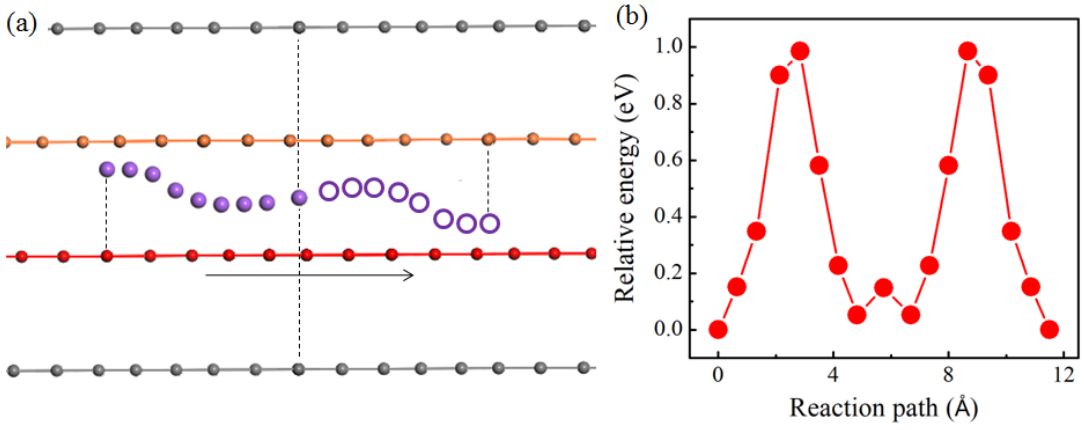


Fig. S10. (a) Schematic diagram of interlayer diffusion path-1 from site-1 to site-2 through site-4 and (b) corresponding energy surface with energy barrier.

Fig. S11

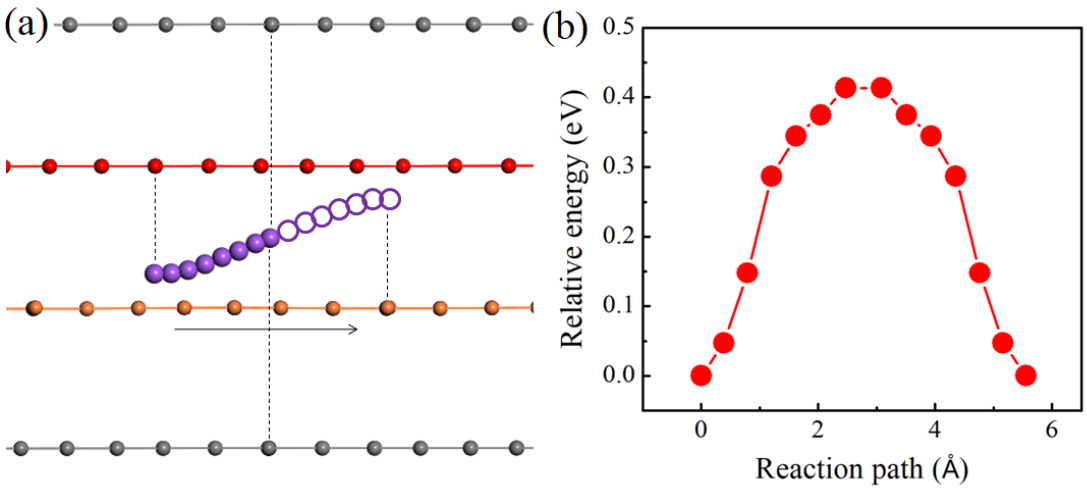


Fig. S11 (a) Schematic diagram of interlayer diffusion path-2 from site-1 to the nearby site-1 through the overlapping region of two triangular pores and (b) corresponding energy surface with energy barrier.

Fig. S12

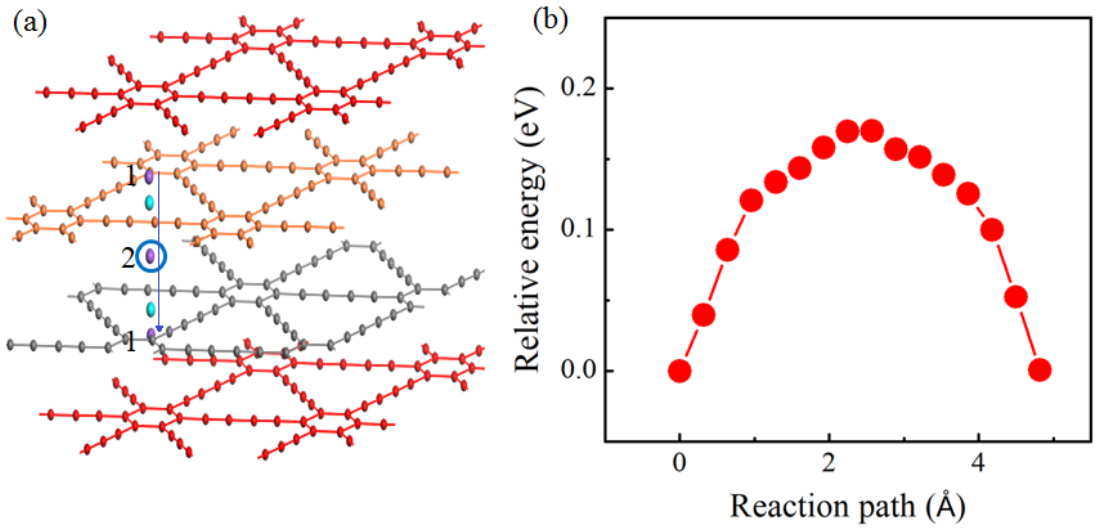


Fig. S12. (a) Schematic diagram of vertical diffusion path from site-1 to the next site-1 through two triangular pores successively and (b) corresponding energy surface with energy barrier.

Fig. S13

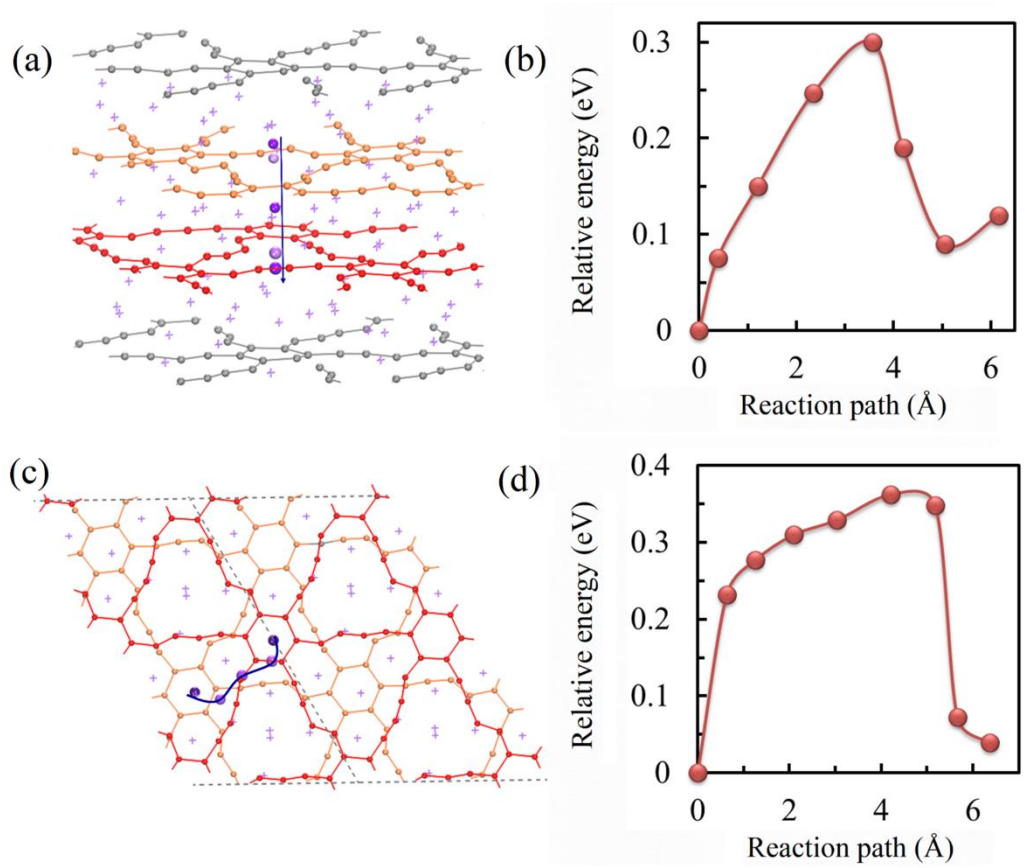


Fig. S13. Schematic diagram of diffusion paths (a, c) and corresponding potential barriers (b, d) of $Na_{0.54}C$. (a) Vertical diffusion path through two triangular pores and (b) corresponding energy surface with energy barrier. (c) Interlayer diffusion through the overlapping region of two triangular pores and (d) corresponding energy surface with energy barrier.

Fig. S14.

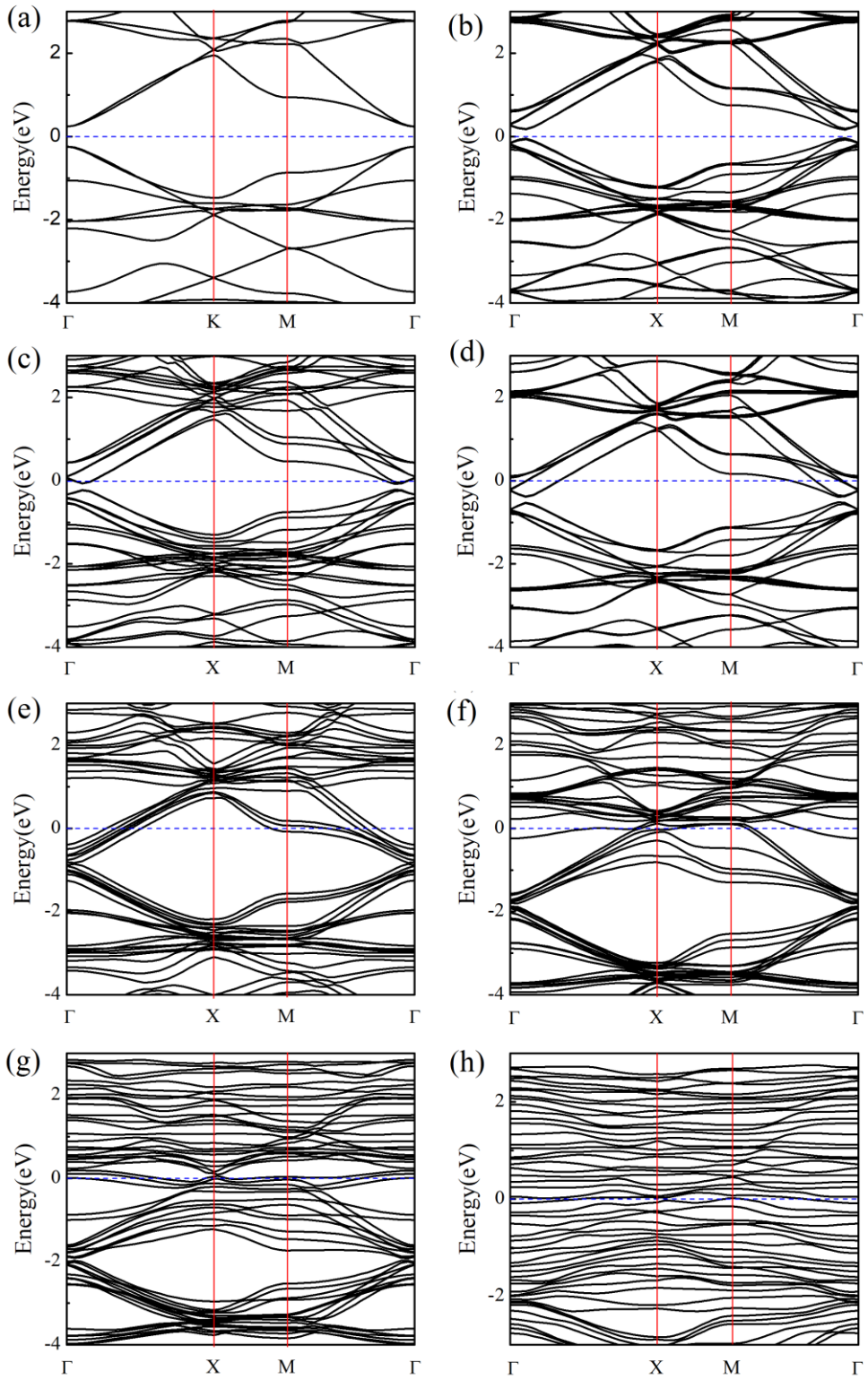


Fig. S14. Band structure of (a) GDY sheet, (b) bulk GDY, and bulk GDY with Na concentration of (c) 0.02, (d) 0.06, (e) 0.11, (f) 0.22, (g) 0.39, (h) 0.61

Fig. S15.

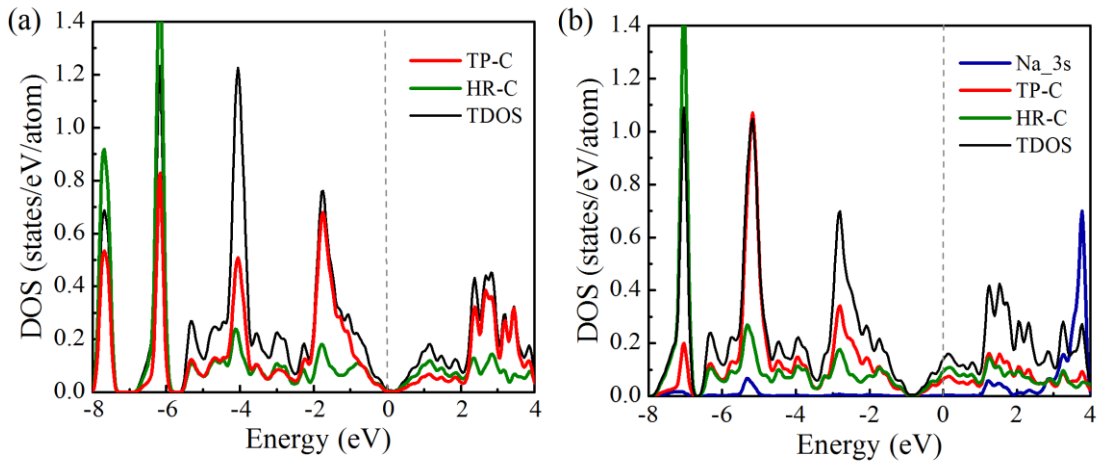


Fig. S15. Total density of states (DOS) and partial density of states of carbon from trigonal pore (TP-C), carbon from hexagonal ring (HR-C), and Na_3s for (a) bulk GDY, and (b) bulk GDY with Na concentration of 0.06.

Fig. S16

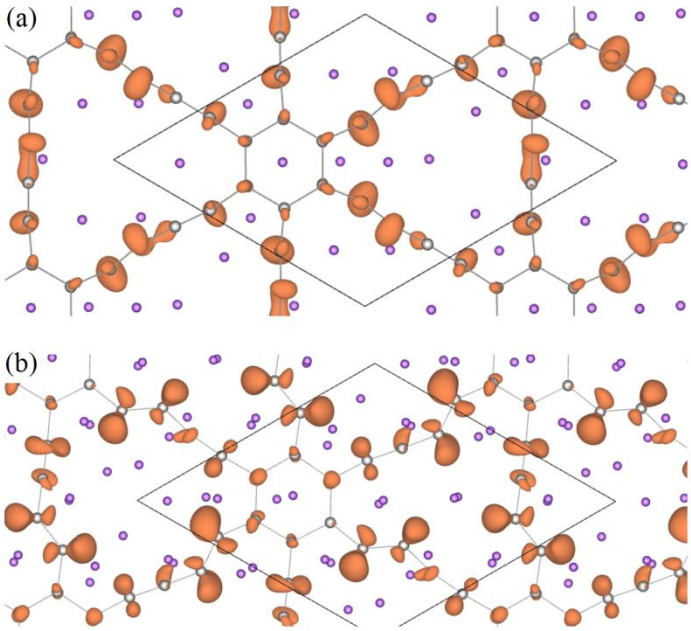


Fig. S16 The distribution of electron charge difference for bulk GDY with Na concentration of 0.39 and 0.83.