Supporting Information

Storage Mechanism of K in Hydrogen Substituted Graphdiyne as Superior Anode

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Figure S1

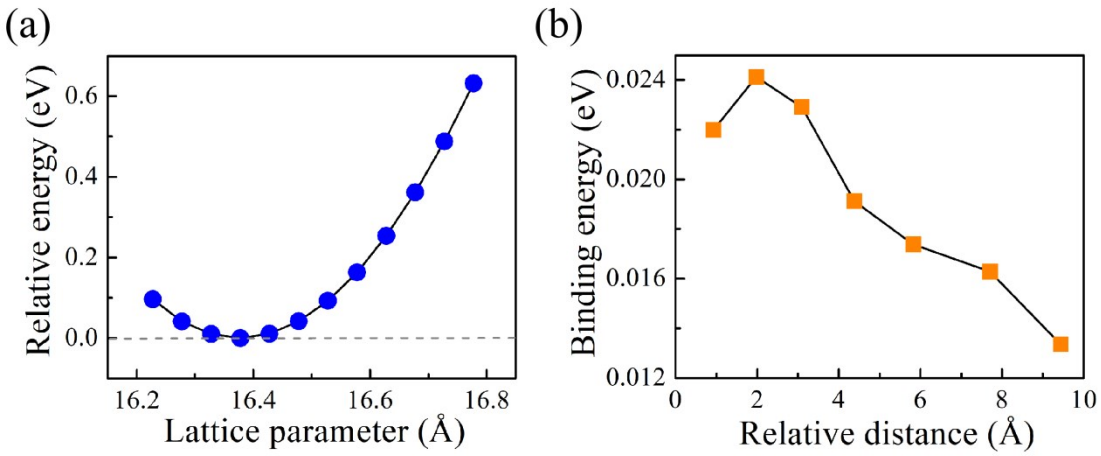

Fig. S1. (a) The relative energy of single-layer HsGDY varies with different lattice constants, and (b) the interlaminar binding energy per atom as a function of the relative distance between two layers in AB stacking HsGDY

Figure S2

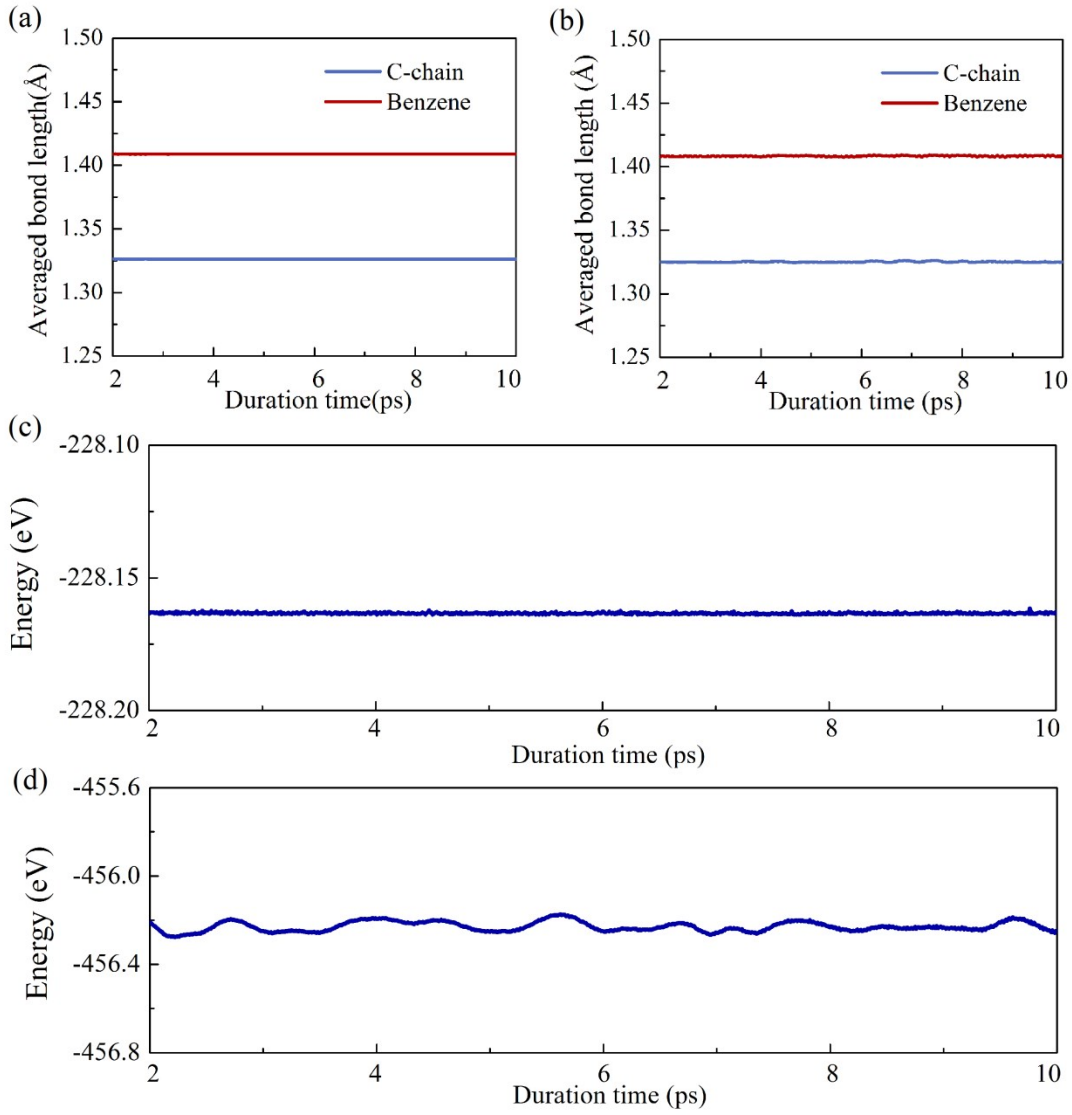

Fig. S2. The averaged carbon bond lengths in C-chains and benzene rings as functions of MD duration time of 8 ps for (a) single-layer and (b) bulk HsGDY, and energies of (c) single-layer and (d) bulk HsGDY as functions of MD duration time at 400k.

Figure S3

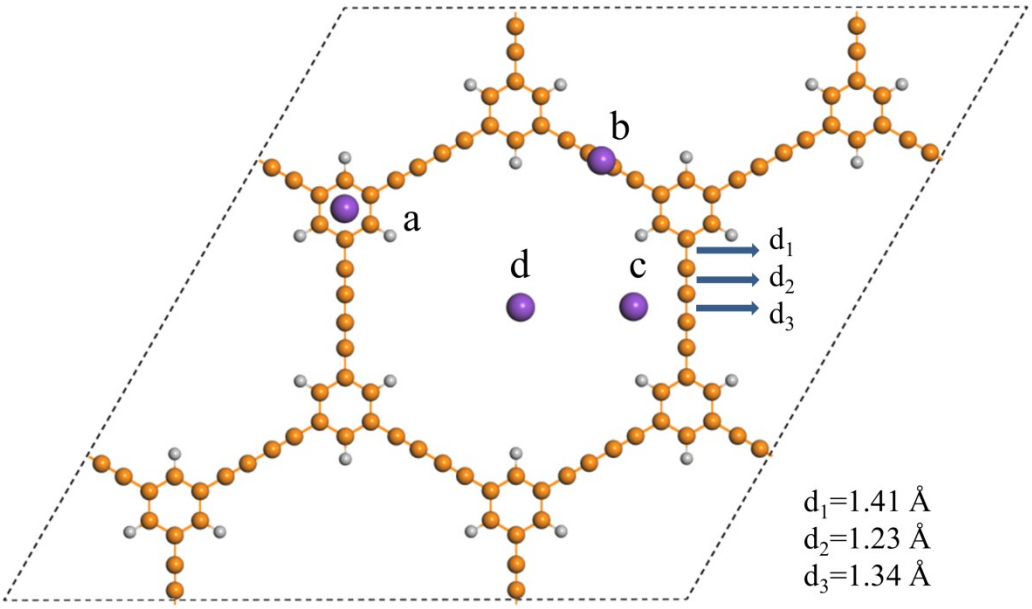

Fig. S3. The top view of the structure of single-layer HsGDY and four potential adsorption sites of K-adsorbed single-layer HsGDY (d2 corresponds to an acetylene bond)

Table S1. The *z*-axis distance of K relative to the carbon layer, and corresponding adsoprtion energy in single-layer HsGDY for different adsorption sites (in Fig. S3)

Site	a	b	c	d
Distance (Å)	2.63	2.48	0	0
Adsorption energy (eV)	1.04	0.99	0.77	0.12

Figure S4

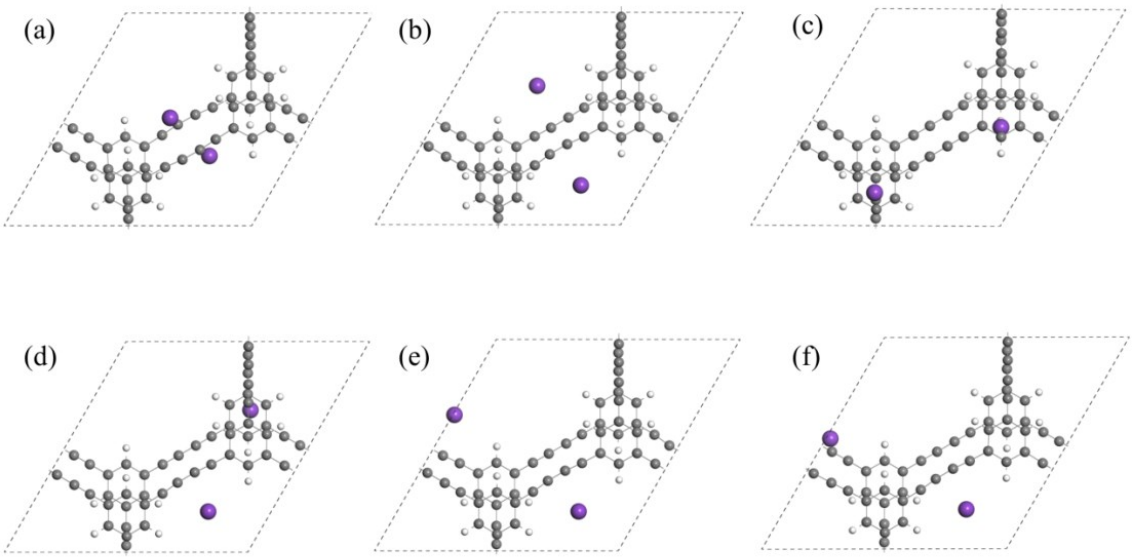

Fig. S4. The top view of various potential low-energy atomic structure models (a-f) with the K/C ratio of 0.042 in bulk HsGDY. The corresponding binding energies are (a)1.64eV, (b)0.80eV, (c)1.08 eV, (d) 0.70 eV, (e)0.86 eV, and (f) 1.20eV, respectively.

Figure S5

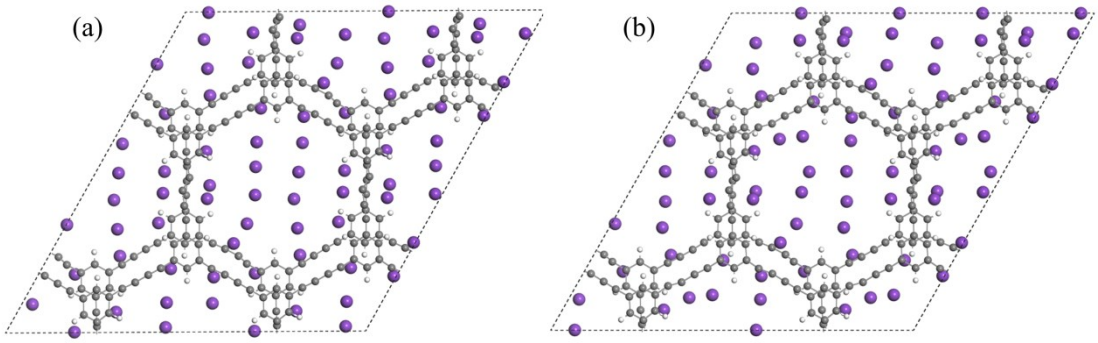

Fig. S5. The top view of two low-energy atomic structure models (a, b) with the K/C ratio of 0.4 in bulk HsGDY. The corresponding binding energies are (a) 1.40eV and (b) 1.42eV, respectively.

Figure S6

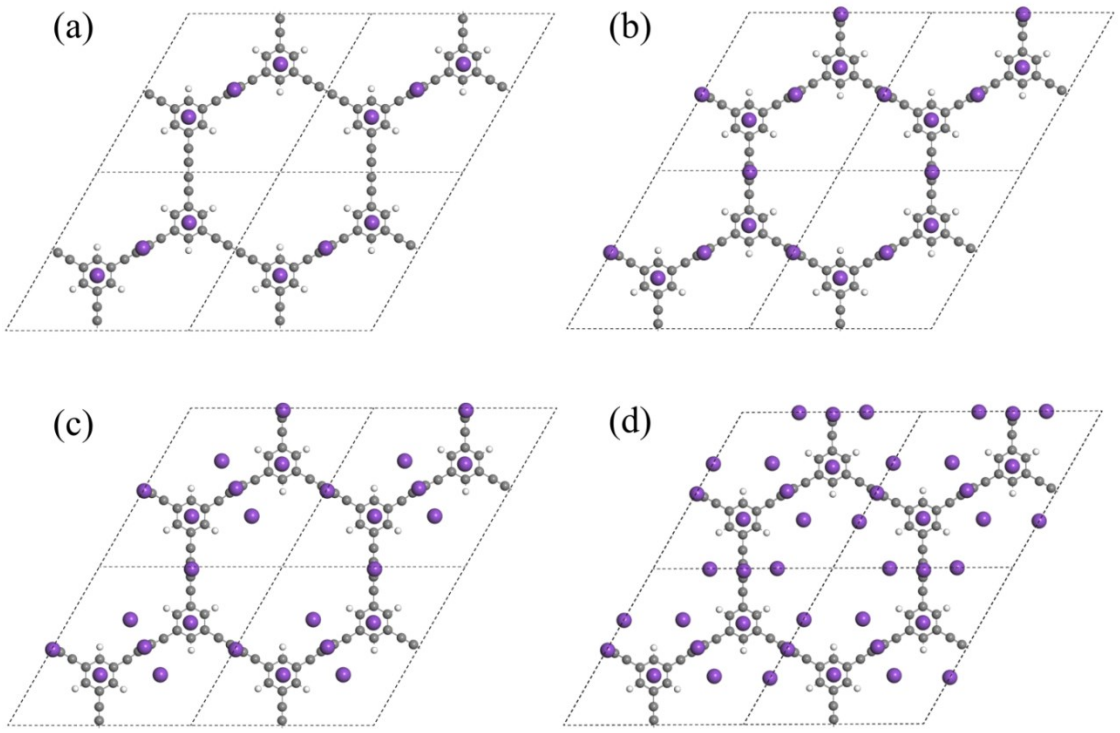

Fig. S6. The top view of stable structures of K-adsorbed single-layer HsGDY with the ratio of K/C of (a) 0.13, (b) 0.21, (c) 0.29 and (d) 0.46

Figure S7

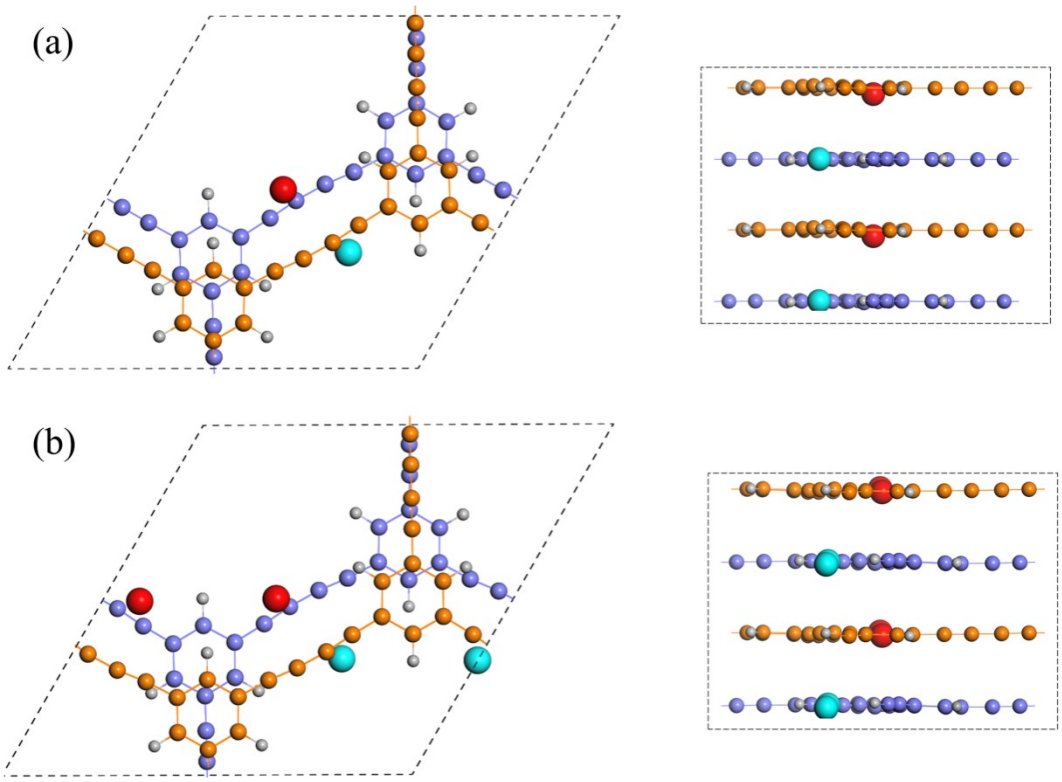

Fig. S7. The top and side view of stable structures of K-adsorbed bulk HsGDY with the ratio of K/C of (a) 0.042 and (b) 0.083. The red and cyan spheres represent K atoms in the orange and purple carbon layers, respectively.

Figure S8

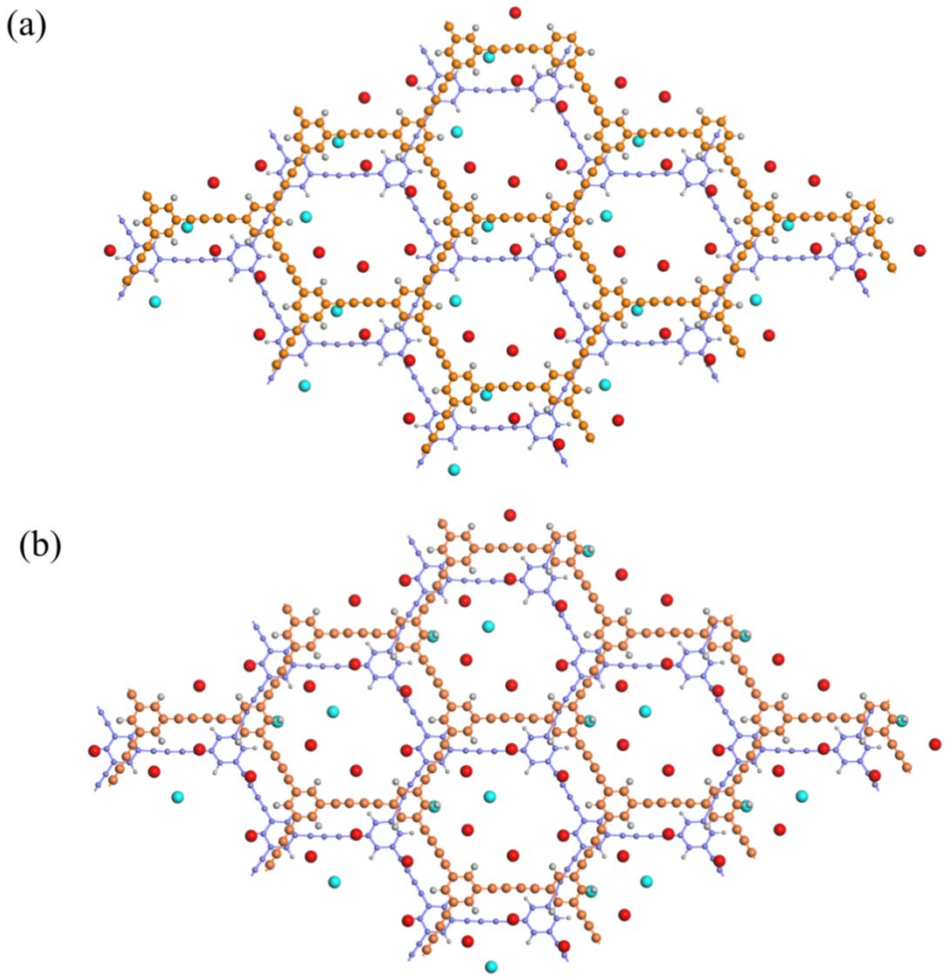

Fig. S8. The side view of stable structures of K-adsorbed bulk HsGDY with the ratio of K/C of (a) 0.15 and (b) 0.17.

Figure S9

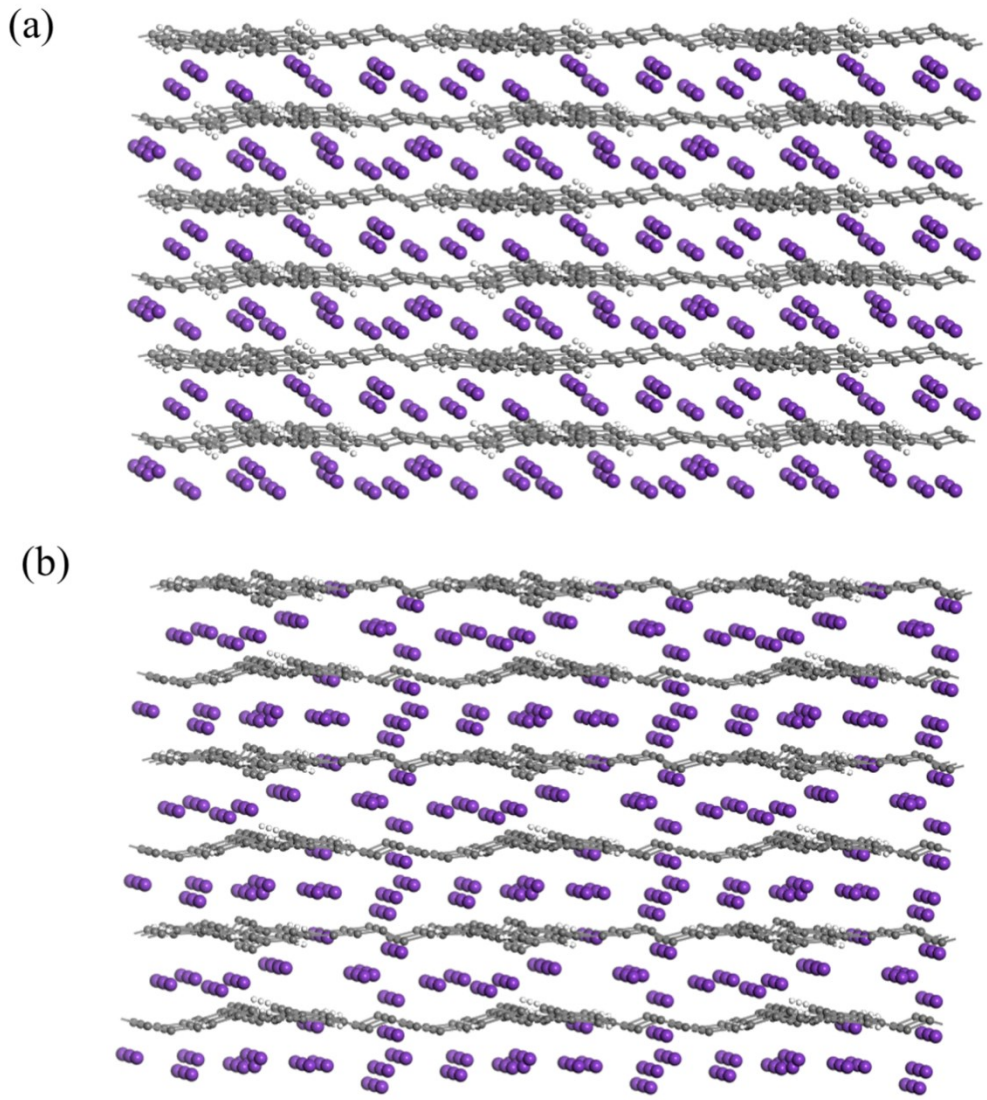

Fig. S9. The side view of stable structures of K-adsorbed bulk HsGDY with the ratio of K/C of (a) 0.4 and (b) 0.48

Figure S10

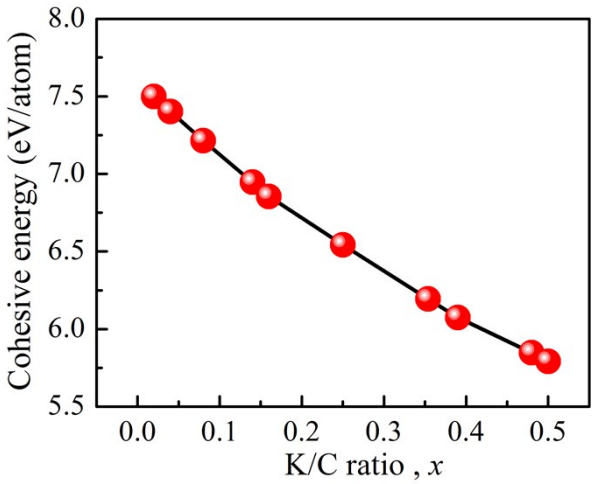

Fig. S10. The cohesive energy bulk HsGDY as a function of the concentration of potassium.

Figure S11

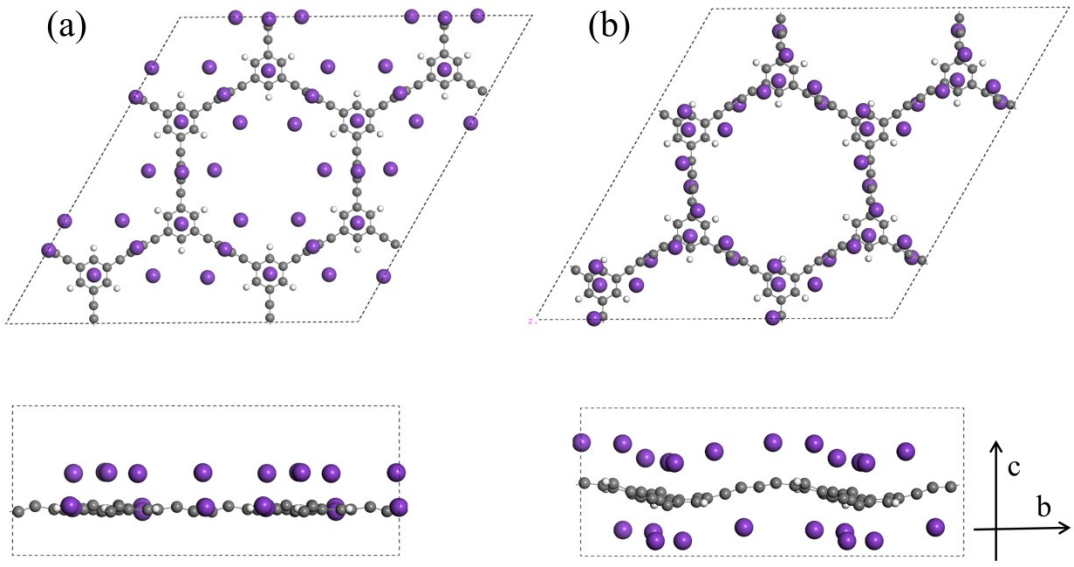

Fig. S11. Top and side views of (a) initiating structure and (b) the structure after 10ps relaxation at 600K with the ratio of K/C of 0.46 in single-layer HsGDY

Figure S12

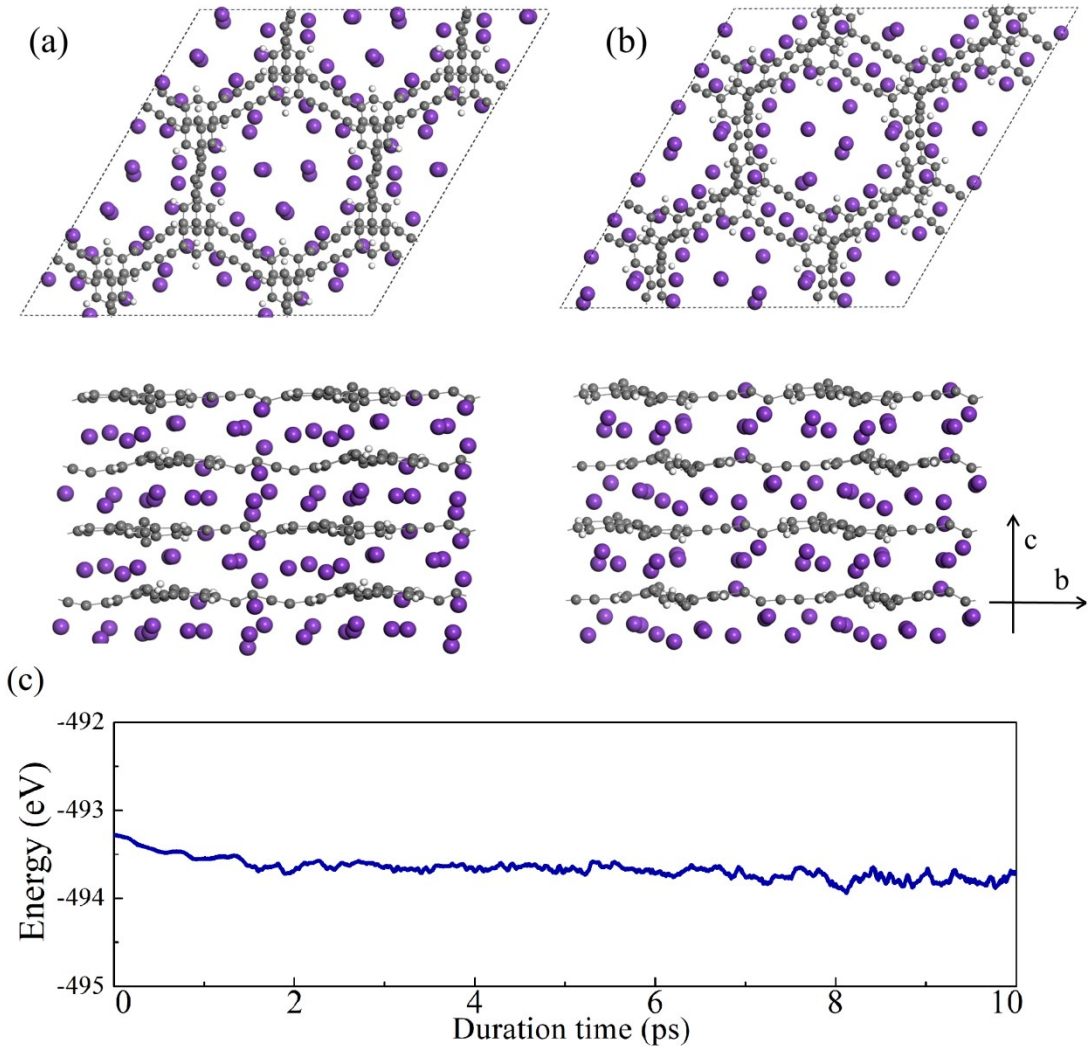

Fig. S12. Top and side views of (a) initiating structure and (b) the structure of after 10ps relaxation at 600K with the ratio of K/C of 0.48 in bulk HsGDY, (c) energy of bulk HsGDY as a function of molecular dynamical duration time at 600 K with the K/C ratio of 0.48

Figure S13

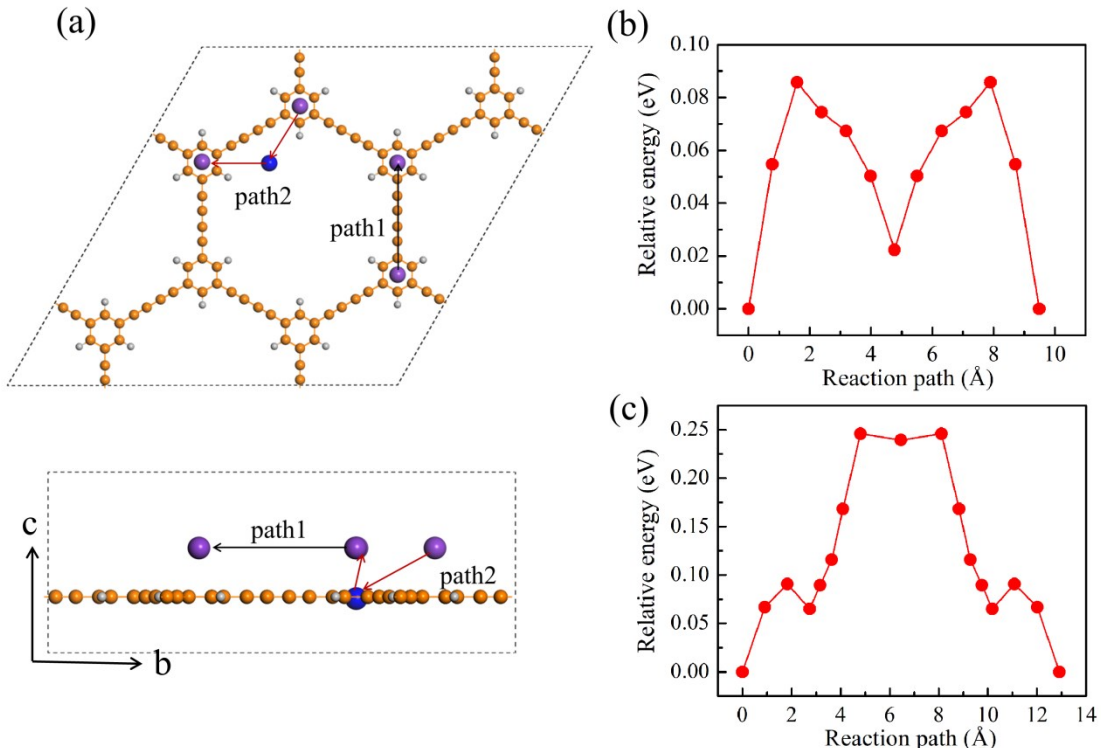

Fig. S13. (a) Schematic diagrams of diffusion in single-layer HsGDY, corresponding potential energy barrier of (b) path-1 and (c) path-2. In (a), the purple balls and blue ball correspond to the site- *a* and *c* (in Fig. S2), respectively

Figure S14

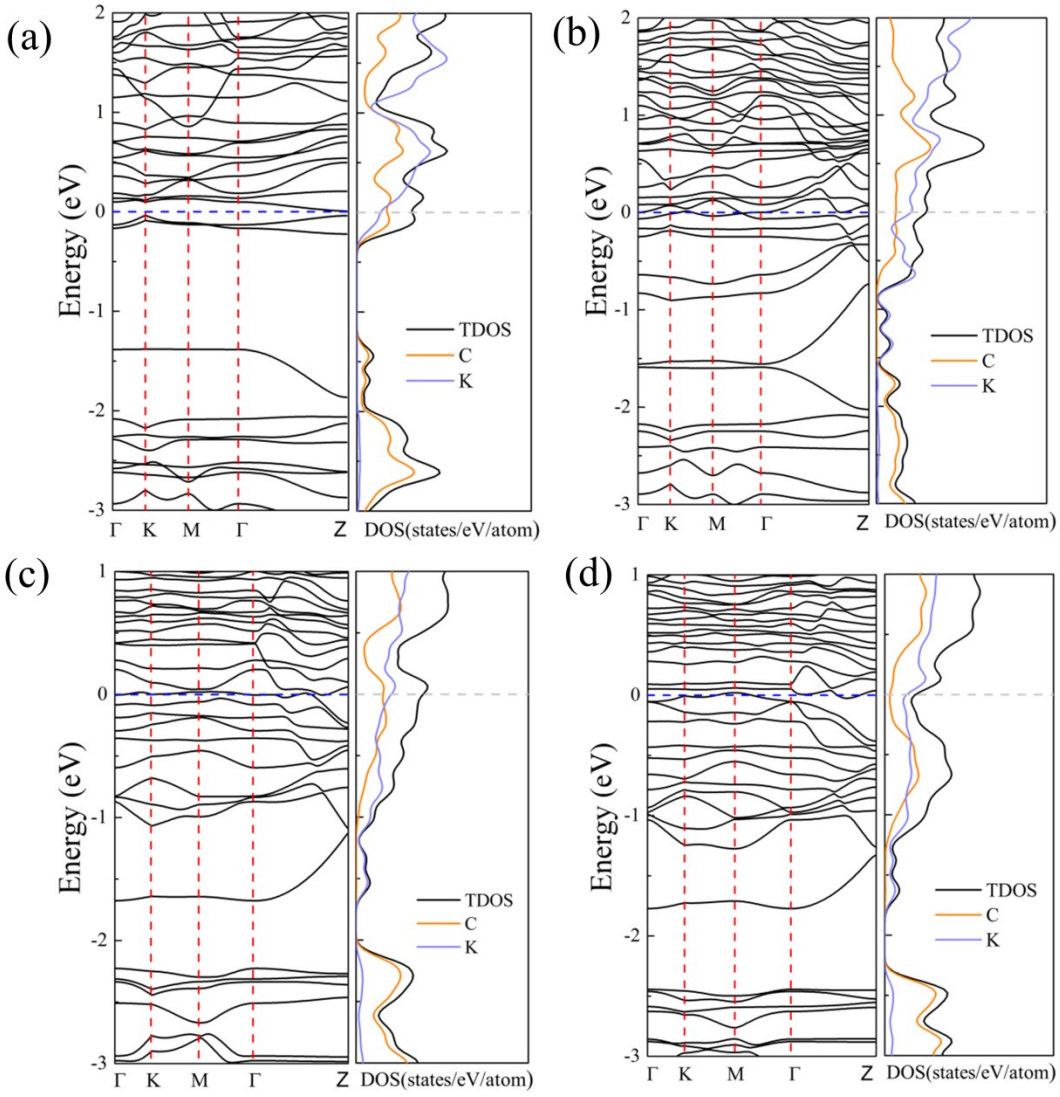

Fig. S14. Band structure and partial density of states of bulk HsGDY with the ratio of K/C of (a) 0.083, (b) 0.25, (c) 0.4 and (d) 0.5

Figure S15

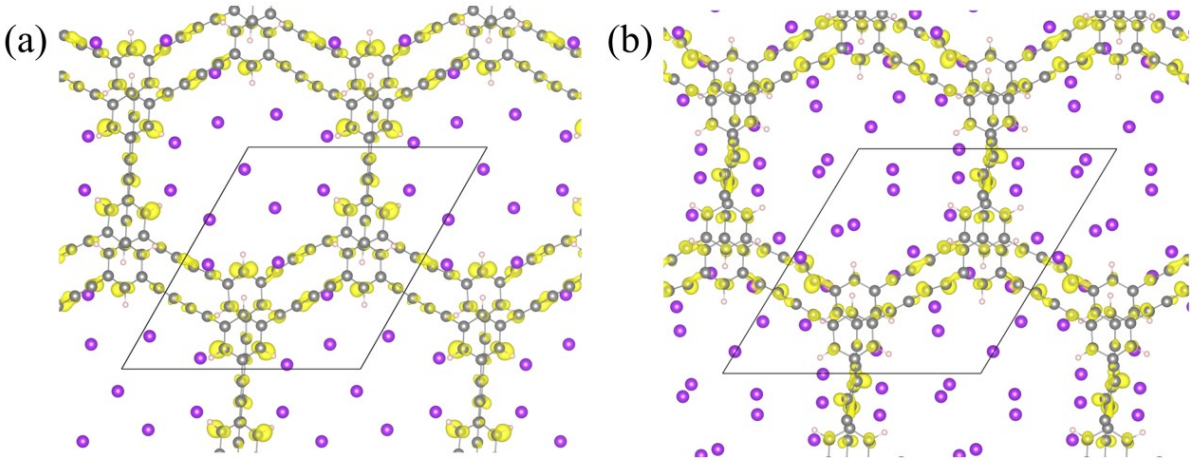

Fig. S15. Distribution of electron charge difference for bulk HsGDY with the ratio of K/C of (a) 0.25 and (b) 0.5

Figure S16

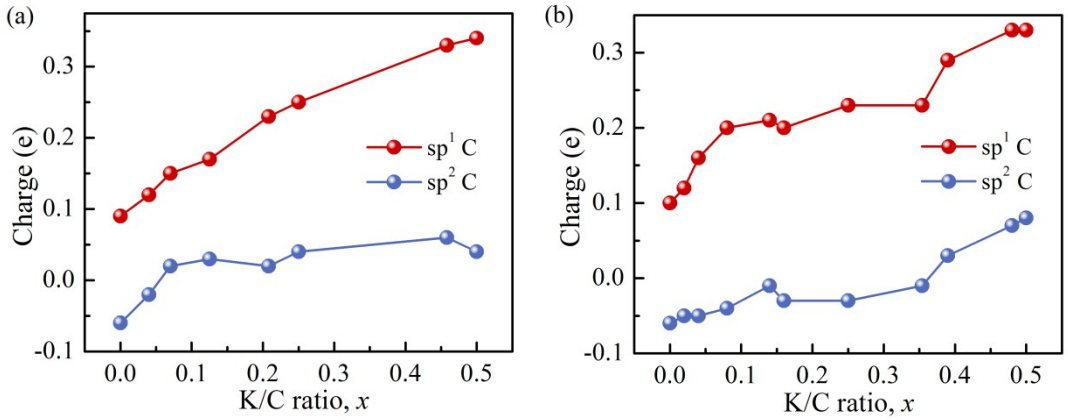

Fig. S16. Bader charge transferred from K to sp^1 and sp^2 carbon in (a) single-layer and (b) bulk HsGDY as functions of K/C ratio.

Table S2 Structural information of bulk HsGDY is calculated from DFT.

Space group of bulk HsGDY : CMCM			
Structural parameters			
a=16.37 Å b=16.37 Å c=6.85 Å		α=90° β=90° γ=60°	
Atoms' coordinates			
Atom	a/x	b/y	c/z
C1	0.774348	0.600272	0.403447
C2	0.440883	0.267253	0.403433
C3	0.874002	0.550437	0.403727
C4	0.960537	0.506863	0.403976
C5	0.62521	0.600328	0.403483
C6	0.292094	0.267266	0.403497
C7	0.575436	0.550493	0.403782
C8	0.532522	0.506898	0.404025
C9	0.724493	0.550821	0.403352
C10	0.391539	0.217184	0.403208
C11	0.441385	0.117498	0.403042
C12	0.484592	0.030949	0.403118
C13	0.62491	0.749807	0.403468
C14	0.292155	0.415838	0.403949
C15	0.575084	0.849393	0.403438
C16	0.531909	0.935892	0.403383
C17	0.575772	0.699681	0.403578
C18	0.241899	0.366649	0.403818
C19	0.142163	0.416418	0.404008
C20	0.055629	0.459404	0.404103
C21	0.724219	0.699685	0.403538
C22	0.391584	0.366581	0.403766
C23	0.441512	0.416355	0.403947
C24	0.485013	0.459367	0.404051
C25	0.846299	0.460846	0.901437
C26	0.512379	0.127665	0.90166
C27	0.946063	0.411104	0.901309
C28	0.032573	0.368222	0.901452
C29	0.696591	0.460938	0.901478
C30	0.364001	0.12774	0.90164
C31	0.646647	0.411178	0.901388
C32	0.603145	0.368149	0.901384

C33	0.796032	0.411697	0.901319
C34	0.46321	0.077626	0.901877
C35	0.512883	0.977997	0.902247
C36	0.55605	0.891476	0.902418
C37	0.696748	0.610078	0.901968
C38	0.363713	0.276674	0.901853
C39	0.646833	0.709865	0.902195
C40	0.603562	0.796419	0.90236
C41	0.647254	0.560224	0.901766
C42	0.313951	0.227147	0.901722
C43	0.214267	0.277048	0.901512
C44	0.127657	0.320722	0.901476
C45	0.796141	0.56016	0.901735
C46	0.462994	0.227057	0.901727
C47	0.512734	0.27698	0.901523
C48	0.555635	0.320608	0.901398
H1	0.517645	0.229155	0.403665
H2	0.76284	0.474038	0.403699
H3	0.253766	0.492625	0.404167
H4	0.499013	0.738261	0.404016
H5	0.25345	0.229141	0.403763
H6	0.762319	0.738344	0.403964
H7	0.589149	0.089228	0.901273
H8	0.834345	0.33491	0.901136
H9	0.325311	0.35347	0.901571
H10	0.570488	0.598374	0.901568
H11	0.32577	0.089228	0.901292
H12	0.834649	0.598418	0.901502