Supplementary Information (SI) for Physical Chemistry Chemical Physics.

First-principles Study on Promoting the Performance of Graphene as Anode Materials for Alkali Metal Ion Batteries by Covalent Cross-linking of Rigid Molecules

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Table S1. The calculated charge difference (e^-) between the graphene and intermediate molecule of GPG structure based on Bader charge population. The atoms of the intermediate molecule are marked in red. The formula of the calculation is same with the charge density difference.

Atom	Gain(+)/loss(-)	Atom	Gain(+)/loss(-)	Atom	Gain(+)/loss(-)	Atom	Gain(+)/loss(-)
C1	-0.004307	C28	0.000684	C55	-0.001673	C82	0.003545
C2	0.00582	C29	0.010789	C56	-0.003316	C83	-0.001151
C3	0.009334	C30	-0.009931	C57	0.003669	C84	0.001793
C4	0.02332	C31	0.008753	C58	0.005349	C85	0.004289
C5	0.00415	C32	-0.003052	C59	-0.002413	C86	0.005845
C6	7.3E-05	C33	0.000176	C60	-0.007558	C87	-0.000165
C7	0.001154	C34	-0.001321	C61	0.000326	C88	0.004284
C8	0.001339	C35	5.8E-05	C62	-0.003727	C89	0.00772
C9	0.000811	C36	0.003036	C63	-0.010952	C90	-0.008646
C10	0.002753	C37	-0.00792	C64	0.008573	C91	-0.000561
C11	-0.006214	C38	0.001288	C65	-0.000844	C92	0.000143
C12	-0.002413	C39	0.001457	C66	-0.008102	C93	0.004773
C13	0.000113	C40	0.003963	C67	-0.004929	C94	-0.004562
C14	-0.001826	C41	0.007774	C68	0.013883	C95	-0.001188
C15	0.008392	C42	0.016602	C69	0.018639	C96	0.003094
C16	0.003582	C43	0.003094	C70	-0.130686	C97	0.001709
C17	-0.001453	C44	0.001884	C71	0.000882	C98	0.001129
C18	0.000119	C45	0.0053	C72	0.002802	**C99**	**-0.053586**
C19	0.00173	C46	-0.009973	C73	0.00504	**C100**	**-0.059479**
C20	0.000271	C47	-0.004029	C74	0.018181	**C101**	**-0.0698**
C21	-0.008254	C48	0.019913	C75	0.00066	**C102**	**-0.033718**
C22	0.000607	C49	-0.099589	C76	-0.001216	**C103**	**-0.093812**
C23	0.00168	C50	0.017614	C77	0.006083	**C104**	**-0.032065**
C24	-0.001693	C51	0.003553	C78	-0.000988	**H1**	**0.087974**
C25	-0.033336	C52	0.000266	C79	0.00298	**H2**	**0.135871**
C26	-0.002385	C53	0.010555	C80	-0.022987	**H3**	**0.099153**
C27	0.01603	C54	0.014641	C81	0.000799	**H4**	**0.093951**

Table S2. The calculated charge difference (e^-) between the graphene and intermediate molecule of GPPG structure based on Bader charge population. The atoms of the intermediate molecule are marked in red.

Atom	Gain(+)/loss(-)	Atom	Gain(+)/loss(-)	Atom	Gain(+)/loss(-)	Atom	Gain(+)/loss(-)
C1	-0.011638	C30	-0.011732	C59	-0.001715	C88	0.006583
C2	-0.006086	C31	0.040958	C60	0.006071	C89	0.009893
C3	0.001991	C32	-0.002556	C61	-0.003352	C90	0.010689
C4	0.000535	C33	-0.014238	C62	0.010661	C91	-0.002318
C5	0.005131	C34	-0.001809	C63	0.004008	C92	0.003598
C6	-0.005061	C35	-0.008583	C64	0.008183	C93	0.001545
C7	0.020533	C36	0.000829	C65	-0.00143	C94	-0.021495
C8	0.002648	C37	-0.038972	C66	0.023262	C95	-0.000761
C9	0.011341	C38	-0.001556	C67	-0.083759	C96	-0.000399
C10	0.002758	C39	0.034755	C68	0.005798	C97	-0.000596
C11	-0.005764	C40	0.002368	C69	-0.008704	C98	0.000547
C12	-0.003513	C41	0.012242	C70	-0.023054	**C99**	**-0.080503**
C13	-0.000962	C42	-0.015525	C71	0.000981	**C100**	**-0.066112**
C14	0.00109	C43	-0.001262	C72	0.004548	**C101**	**0.003111**
C15	-0.006984	C44	0.001895	C73	0.01444	**C102**	**-0.0228**
C16	0.004818	C45	-0.029337	C74	0.026284	**C103**	**-0.000135**
C17	0.001273	C46	-0.075775	C75	1E-05	**C104**	**-0.003625**
C18	-0.000596	C47	0.012895	C76	0.001067	**C105**	**-0.08961**
C19	-0.001634	C48	-0.006628	C77	0.003593	**C106**	**-0.01003**
C20	0.002788	C49	-0.060791	C78	0.004098	**O1**	**0.14952**
C21	0.005946	C50	-0.000672	C79	0.002569	**O2**	**0.123742**
C22	-0.002964	C51	0.031745	C80	0.000325	**N1**	**-0.018635**
C23	-0.002884	C52	0.002438	C81	-0.002704	**N2**	**-0.032744**
C24	0.002069	C53	-0.026748	C82	-0.000455	**H1**	**0.043015**
C25	-0.043492	C54	0.000213	C83	0.005514	**H2**	**0.052893**
C26	-0.0373	C55	0.001468	C84	-0.000454	**H3**	**0.008368**
C27	0.029548	C56	-0.011694	C85	0.004173	**H4**	**0.099649**
C28	0.003708	C57	0.021354	C86	-0.004431	**H5**	**0.003699**
C29	-0.004062	C58	0.002525	C87	-0.01622	**H6**	**0.032556**

Table S3. The adsorption energy (eV) of Li/Na/K ions at each sites of the GPG and GPPG. H*n* represents the adsorption on the hollow site numbered *n*, Bn_1-n_2 represents the adsorption on the C-C bond common to rings numbered n_1 and n_2, and Tn_1-n_2-n_3 presents the adsorption on the top of C atom common to rings numbered n_1, n_2 and n_3.

initial site	GPG						GPPG					
	Li		Na		K		Li		Na		K	
	Site	E_a	Site	E_a	Site	E_a	Site	E_a	Site	E_a	Site	E_a
H1	H1	-0.95	H1	-0.95	T1-2-3	-1.61	H1	-1.45	H1	-1.31	H1	-1.87
H2	H2	-0.31	H2	-0.68	H2	-1.61	H2	-0.39	H2	-0.35	B2-3	-1.89
H3	H1	-0.94	H3	-0.52	B2-3	-1.58	H2	-1.04	H1	-1.34	B2-3	-1.89
H4	H4	-0.2	H5	-0.61	T5-13-14	-1.39	H4	-0.63	H1	-1.25	B1-9	-1.86
H5	H5	-0.4	H5	-0.6	T5-13-14	-1.4	H7	-1.67	H7	-1.4	H7	-1.93
H6	H6	-0.45	B5-6	-0.61	H6	-1.39	H7	-1.62	H7	-1.35	H7	-1.91
H7	H7	-0.32	H6	-0.6	H6	-1.38	H7	-1.67	H7	-1.31	H7	-1.95
H8	H1	-0.92	H1	-0.92	B1-2	-1.59	H8	-1.55	H8	-1.28	H8	-1.8
H9	H9	-0.27	H1	-0.94	B1-2	-1.6	H1	-1.54	T1-8-9	-1.18	B1-9	-1.85
H10	H10	-0.29	H1	-0.95	B1-2	-1.6	H10	-0.45	B1-9	-1.31	B1-9	-1.87
H11	H11	-0.15	H11	-0.25	H11	-1.12	H11	-0.3	H11	-0.34	H11	-1.15
H12	H12	-0.16	B1-2	-0.88	H2	-1.59	H12	-0.29	H12	-0.36	H2	-1.63
H13	H13	-0.16	H5	-0.61	T5-13-14	-1.4	H13	-0.29	H13	-0.36	H13	-1.16
H14	H14	-0.15	H14	-0.51	T5-13-14	-1.4	H14	-0.29	H14	-0.36	H7	-1.95
H15	H15	-0.15	H15	-0.23	H15	-1.12	H15	-0.33	H15	-0.35	H15	-1.16
H16	H16	-0.16	H16	-0.24	B2-3	-1.58	H16	-0.39	H7	-1.37	H7	-1.8
H17	H17	-0.15	H17	-0.25	H17	-1.11	H17	-0.32	H17	-0.35	H17	-1.12
H18	H18	-0.1	H18	-0.19	H18	-1.08	H18	-0.26	H18	-0.27	H18	-1.08
H19	H19	-0.12	H19	-0.2	H19	-1.11	H19	-0.29	H8	-1.33	H8	-1.82
H20	H20	-0.12	H20	-0.2	H20	-1.09	H20	-0.41	B7-8	-1.29	B7-8	-1.81
H21	H21	-0.14	H21	-0.22	H21	-1.1	H21	-0.34	H7	-1.39	H7	-1.91
H22	H22	-0.13	H22	-0.21	H22	-1.1	H22	-0.27	H22	-0.23	H22	-1.02
H23	H23	-0.13	H23	-0.21	H23	-1.11	H23	-0.26	H23	-0.29	H7	-1.12

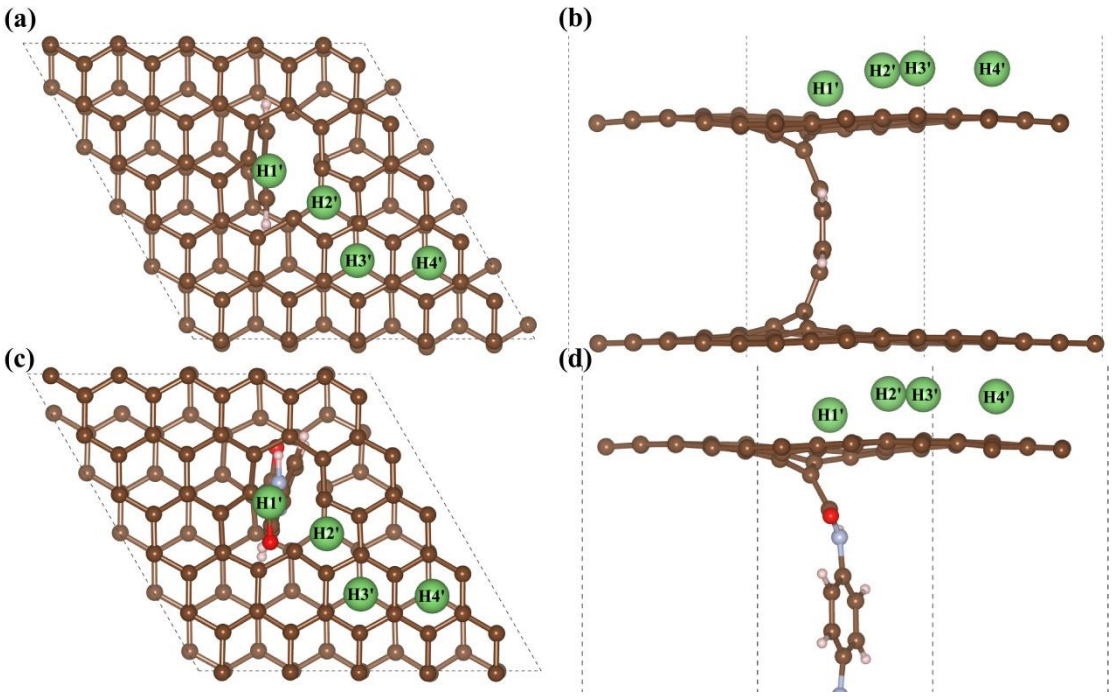

Figure S1. The possible adsorption sites of Li/Na/K ions at the outer side of (a)-(b) GPG and (c)-(d) GPPG.

Table S4. The adsorption energies (eV) of Li/Na/K ions at the outer side of GPG and GPPG.

	GPG			GPPG		
Sites	Li	Na	K	Li	Na	K
H1'	-0.58	-0.57	-1.11	-0.78	-0.78	-1.36
H2'	-0.52	-0.37	-1.03	-0.73	-0.57	-1.21
H3'	-0.16	-0.11	-0.84	-0.36	-0.31	-1.03
H4'	-0.07	-0.03	-0.76	-0.28	-0.24	-0.97

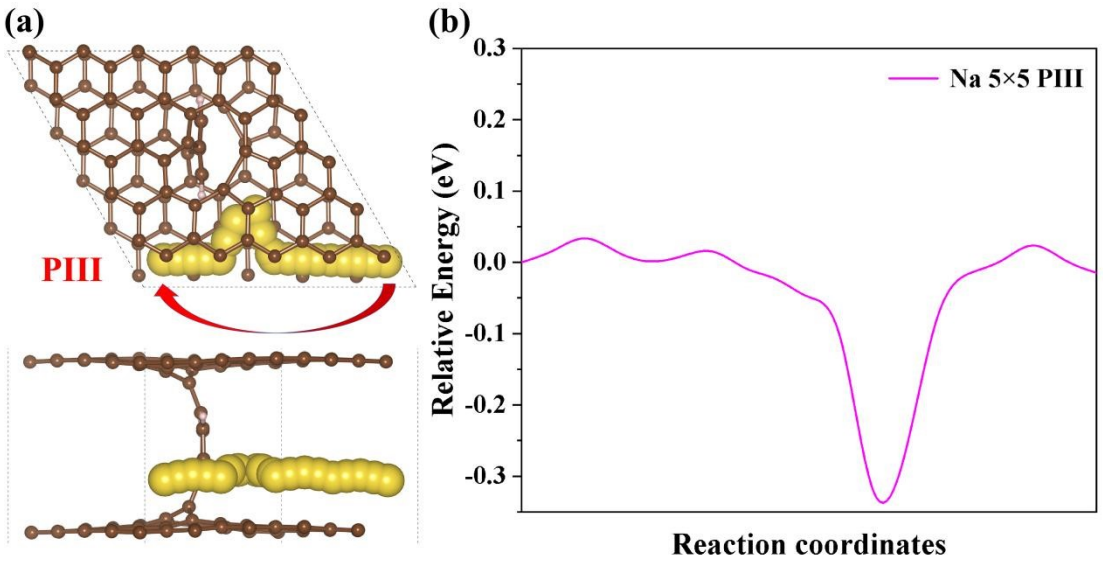

Figure S2. (a) Top and side views of PIII for the diffusion of Na-ions in 5×5 GPG super cell and (b) the corresponding diffusion energy barrier.

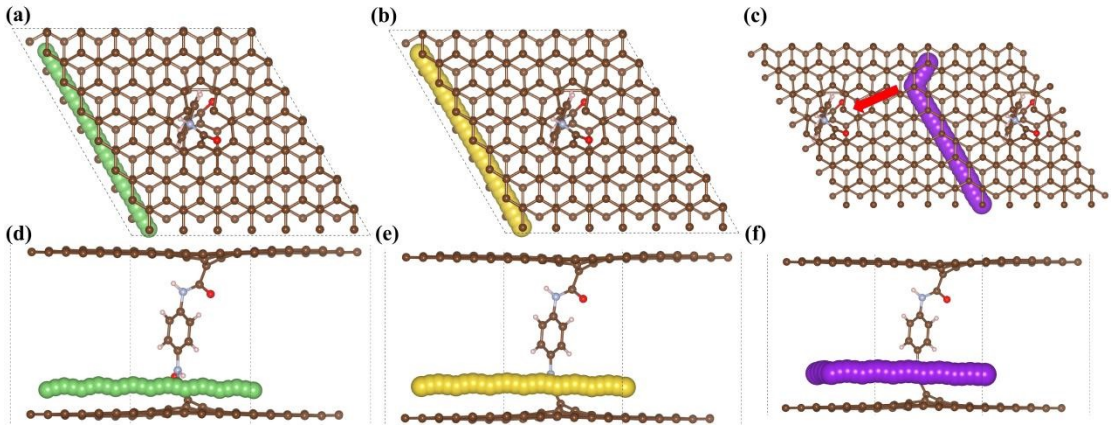

Figure S3. Top and side views of the PIII diffusion paths of Li, Na, K ions in 7×7 GPPG super cell.

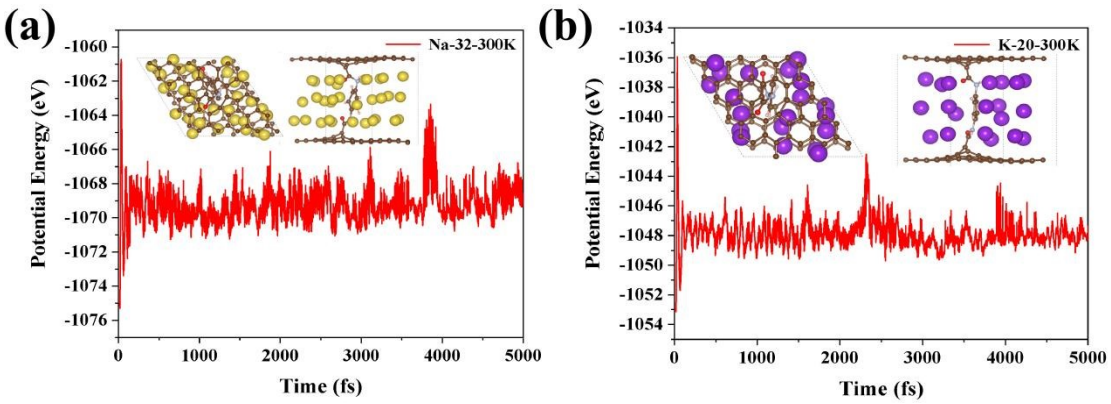

Figure S4. AIMD simulations for structures of GPPG with the maximum adsorption capacity of (a) Na, (b) K ions at 300 K.