

## Supporting information

### **Ferroelectricity-enhanced potassium-ion storage in van der Waals layered CuInP<sub>2</sub>S<sub>6</sub>**

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**Table S1.** Crystallographic parameters of CuInP<sub>2</sub>S<sub>6</sub> refined by the Rietveld method.

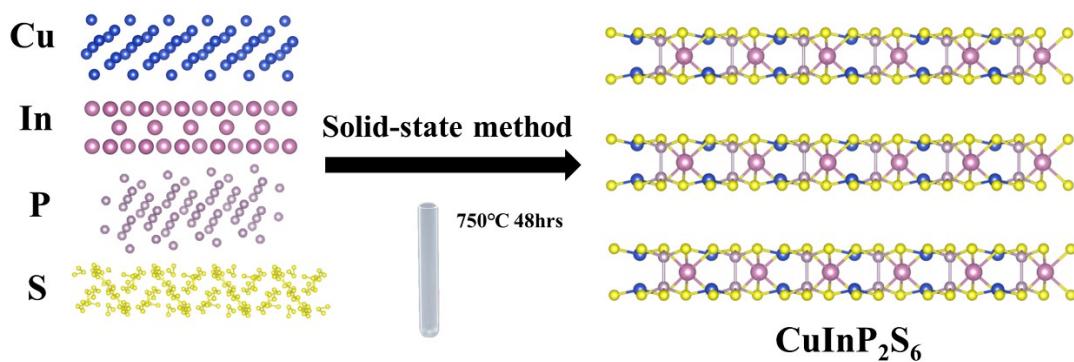
Site	Atom	x	y	z	Occ
Cu_1	Cu	0.0957	0.3355	0.3869	0.875
Cu_2	Cu	-0.069	0.335	0.149	0.1
In_1	In	0	0.00192	0.25	1
P_1	P	0.5686	0.169	0.3491	1
P_2	P	0.4505	0.1674	0.1788	1
S_1	S	0.2808	0.1512	0.395	1
S_2	S	0.2332	0.1645	0.893	1
S_3	S	0.7845	0.0177	0.395	1
S_4	S	0.74	0.1727	0.1336	1
S_5	S	0.7555	0.1747	0.6404	1
S_6	S	0.2722	-0.0057	0.6379	1
a=6.1057		b=10.5569		c=13.5973	
R <sub>wp</sub> =5.19			R <sub>p</sub> =4.29		

**Table S2.** ICP concentrations of the as-synthesized CuInP<sub>2</sub>S<sub>6</sub> (CIPS)

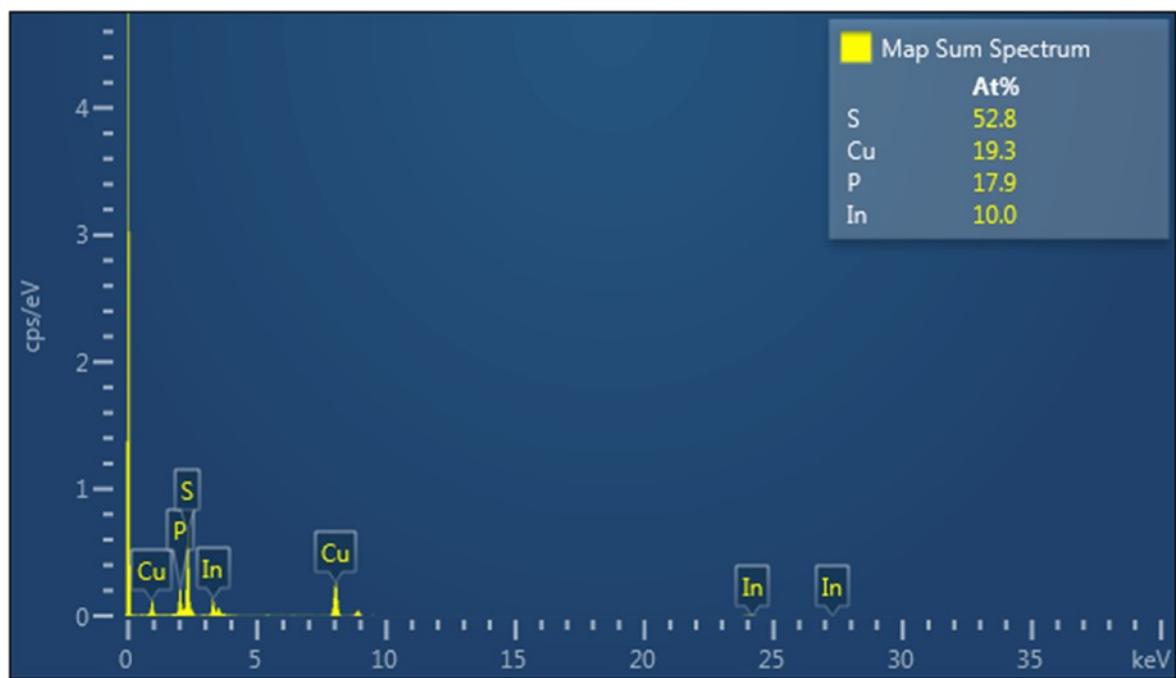
Sample	Cu (at%)	In (at%)	P (at%)	S (at%)
CuInP <sub>2</sub> S <sub>6</sub>	11.69	11.43	20.94	55.94

**Table S3.** Comparison of diffusion coefficients between CIPS@G and FePS<sub>3</sub>@G at different stages.

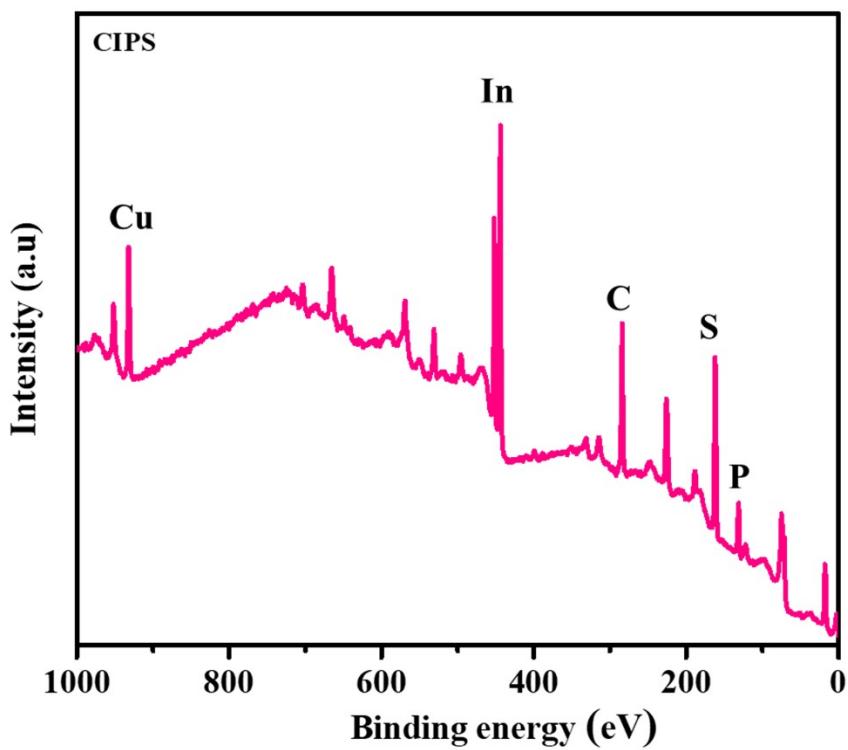
Stage / D <sub>K<sup>+</sup></sub>	Stage I	Stage II	Stage III
CIPS@G	$1.19 \times 10^{-10}$	$3.33 \times 10^{-11}$	$1.12 \times 10^{-11}$
FePS <sub>3</sub> @G	$1.61 \times 10^{-11}$	$1.98 \times 10^{-11}$	$1.98 \times 10^{-11}$



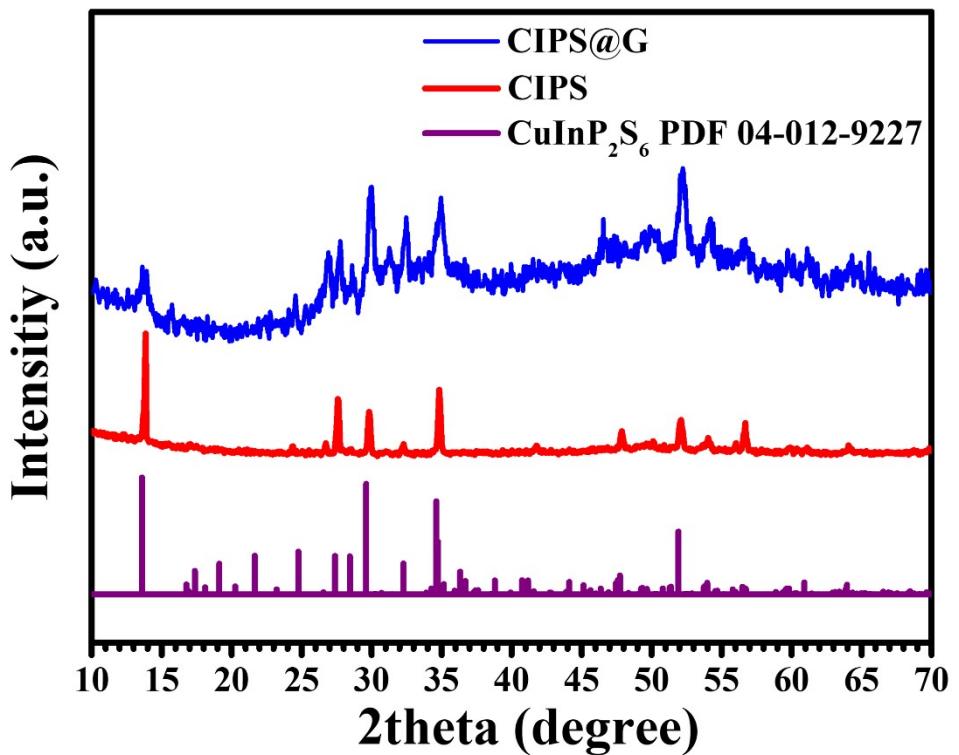
**Figure S1.** Schematic of the synthesis process of CIPS.



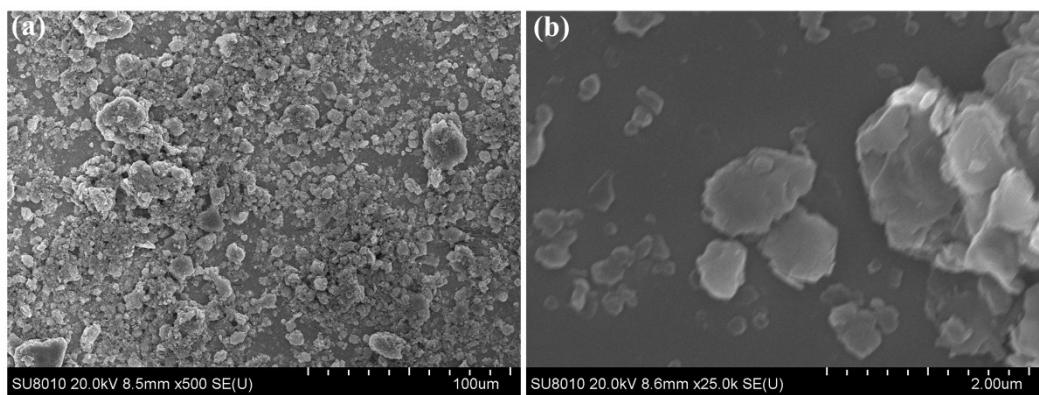
**Figure S2.** EDS mapping result of CIPS.



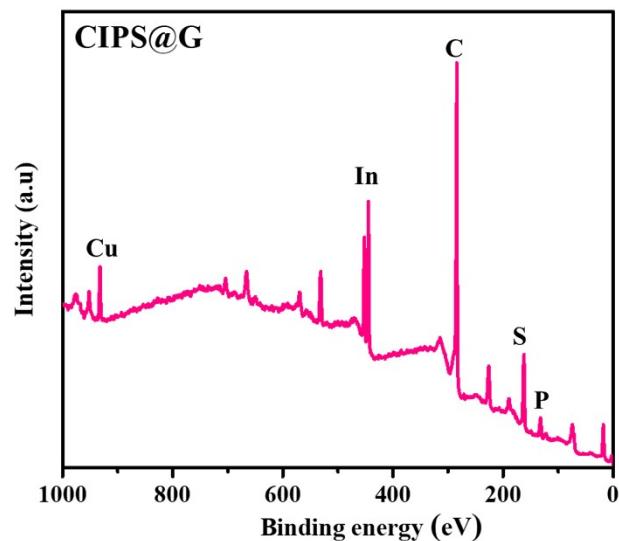
**Figure S3.** The full survey XPS spectrum of CIPS.



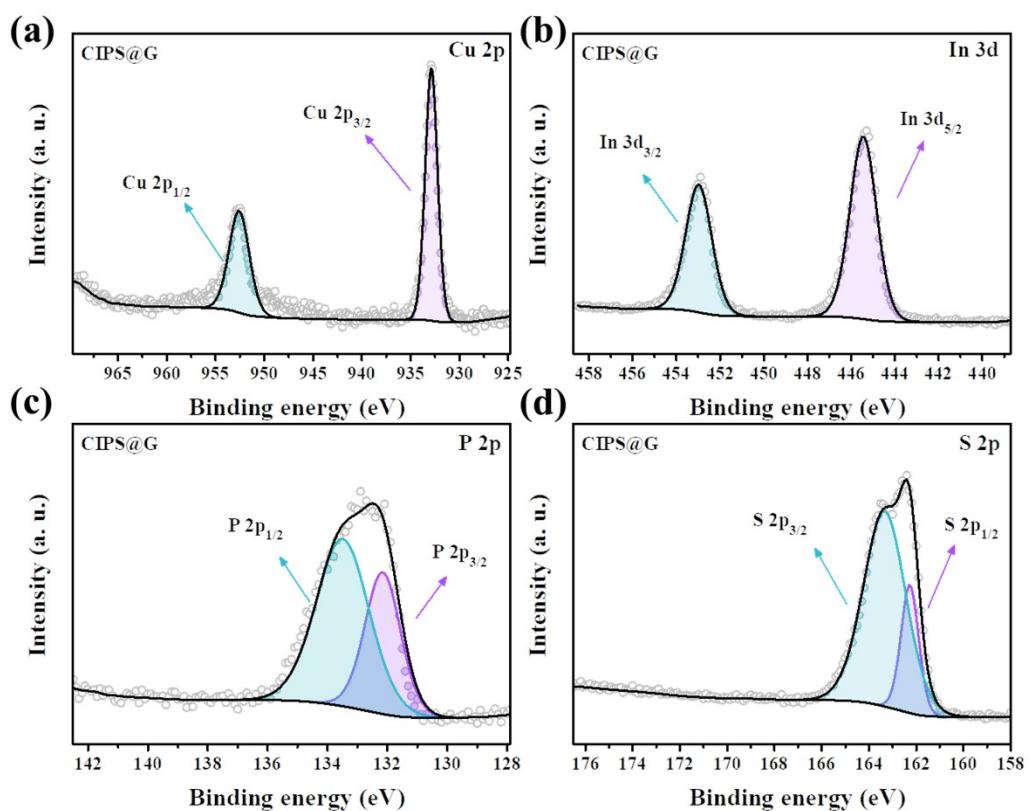
**Figure S4.** XRD pattern of CIPS@G.



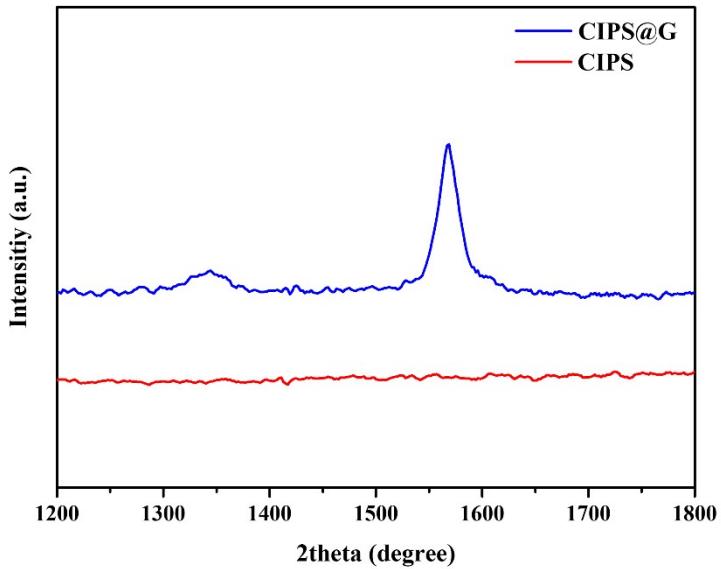
**Figure S5.** SEM image of CIPS@G.



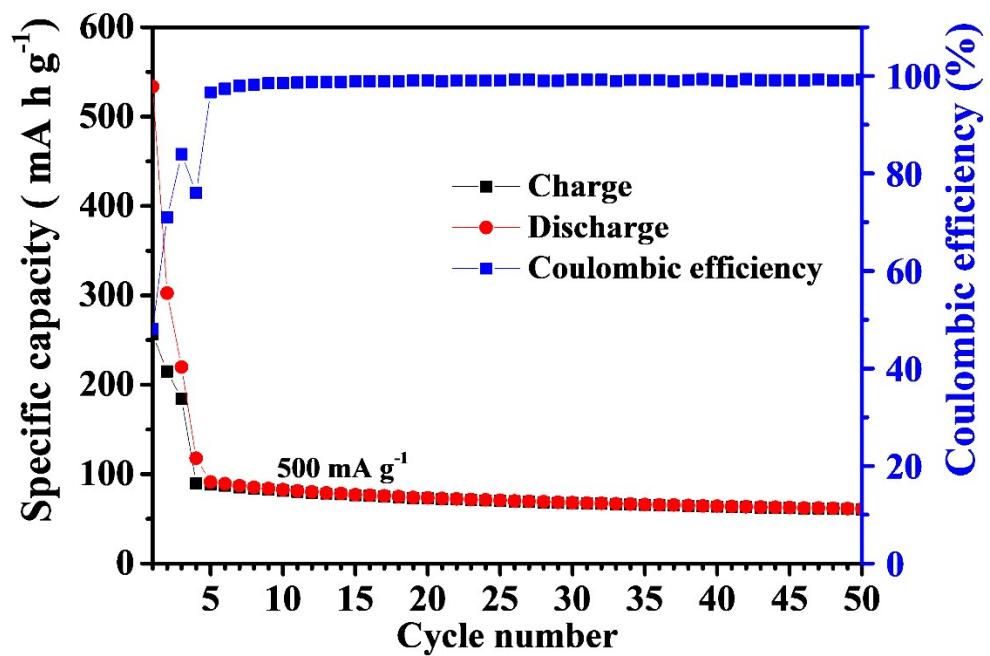
**Figure S6.** The full survey XPS spectrum of CIPS@G.



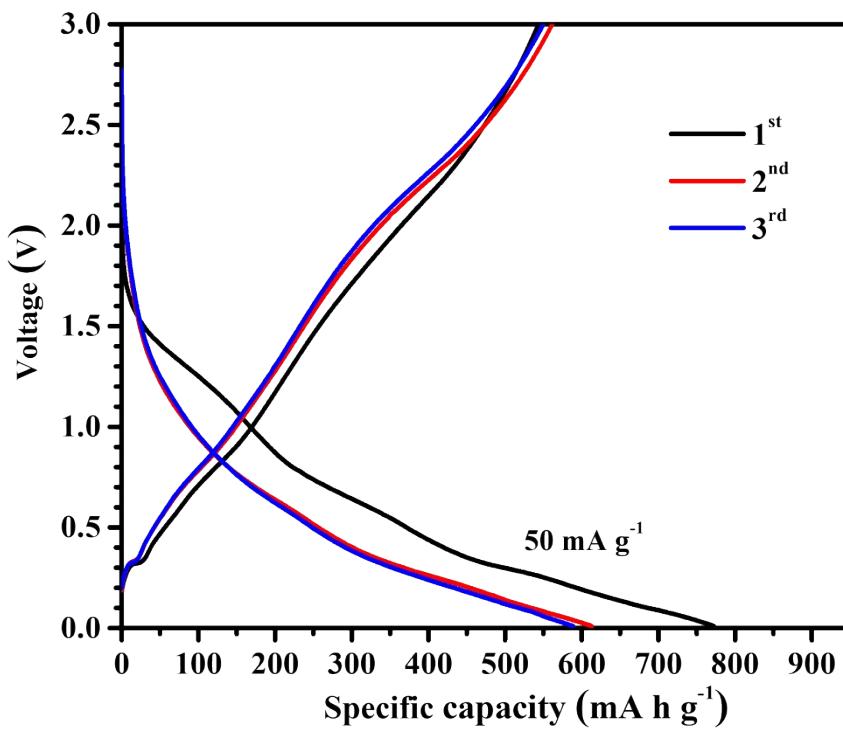
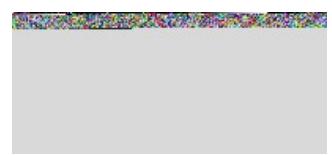
**Figure S7.** High-resolution (a) Cu 2p, (b) In 3d, (c) P 2p, and (d) S 2p XPS spectra of CIPS@G nanoflakes.



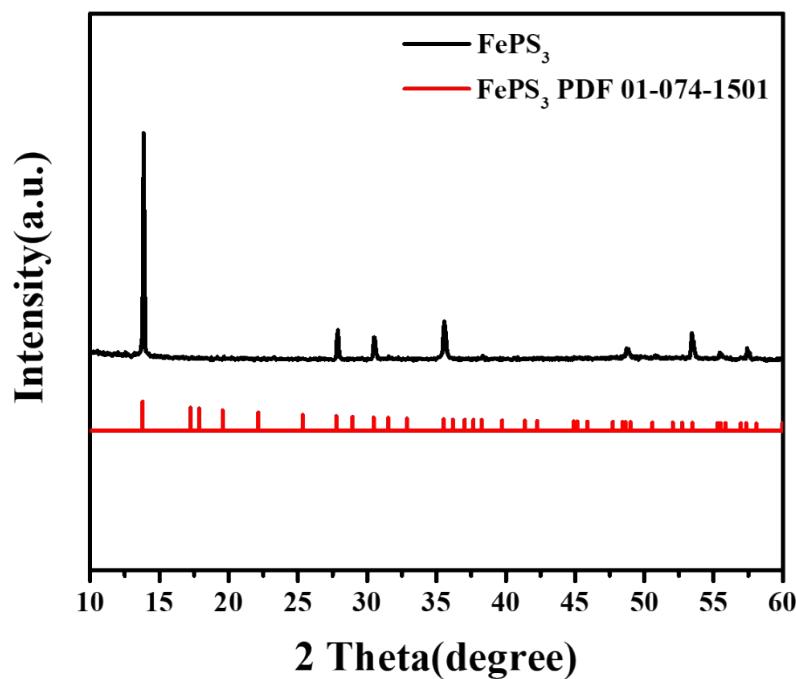
**Figure S8.** Raman spectra of CIPS and CIPS@G.



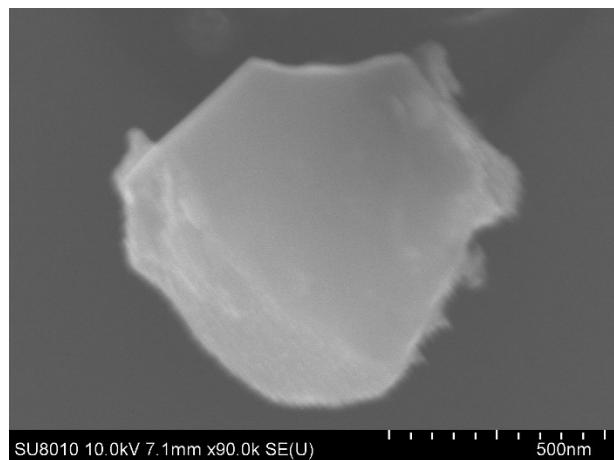
**Figure S9.** Electrochemical performance of pure CIPS.



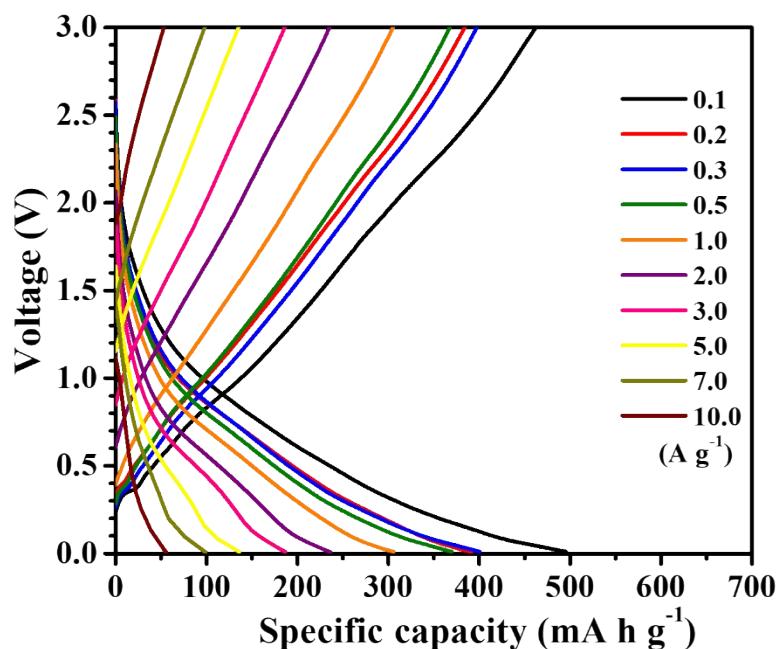
**Figure S10.** GCD curve of CIPS@G at a current density of  $50 \text{ mA g}^{-1}$ .



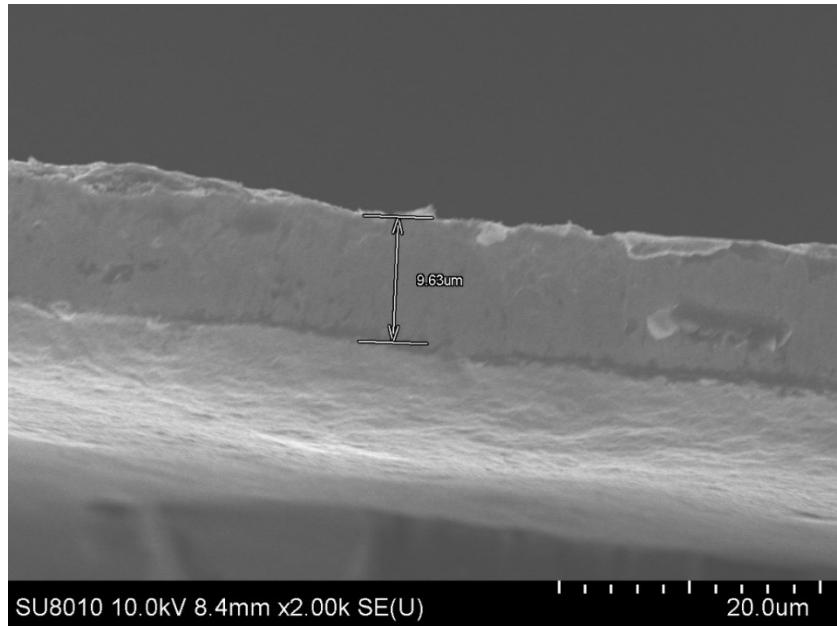
**Figure S11.** XRD pattern of  $\text{FePS}_3$ .



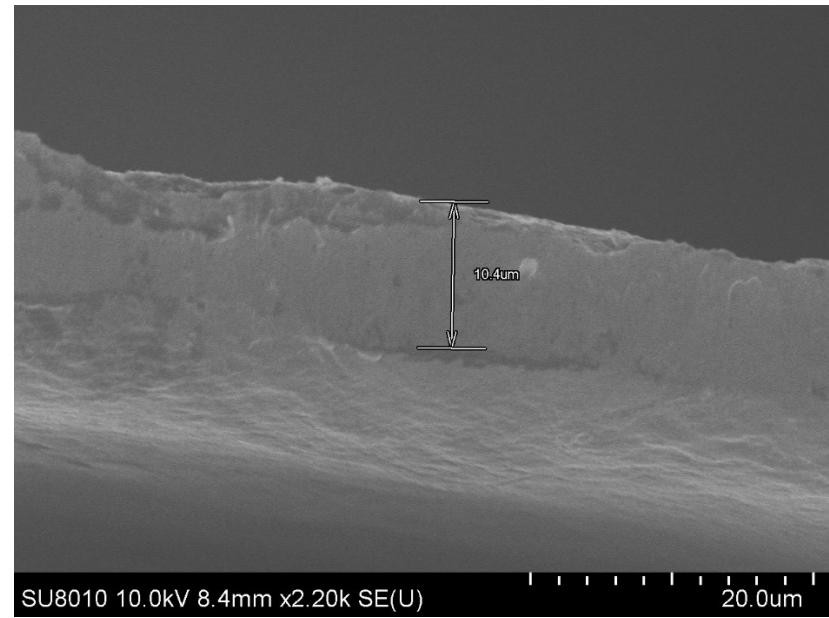
**Figure S12.** SEM image of  $\text{FePS}_3@\text{G}$ .



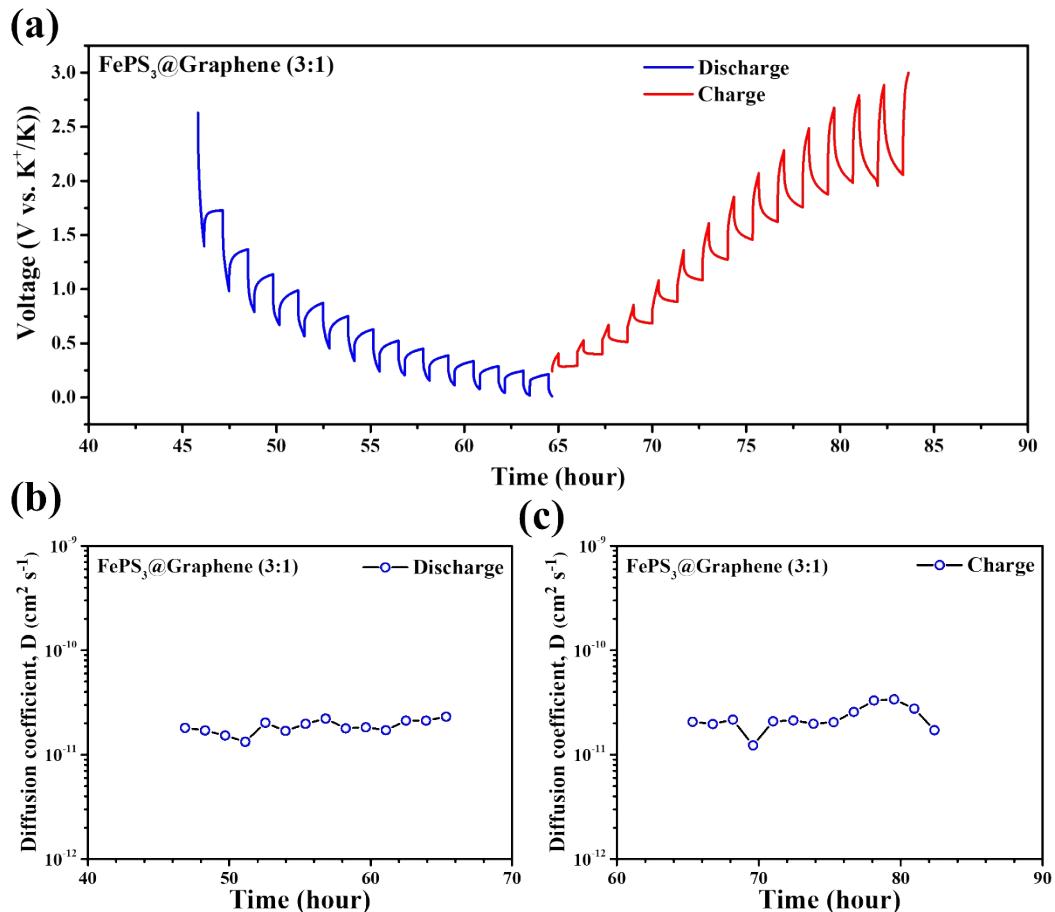
**Figure S13.** Galvanostatic charge/discharge curves of  $\text{CIPS}@\text{G}$  at various current densities.



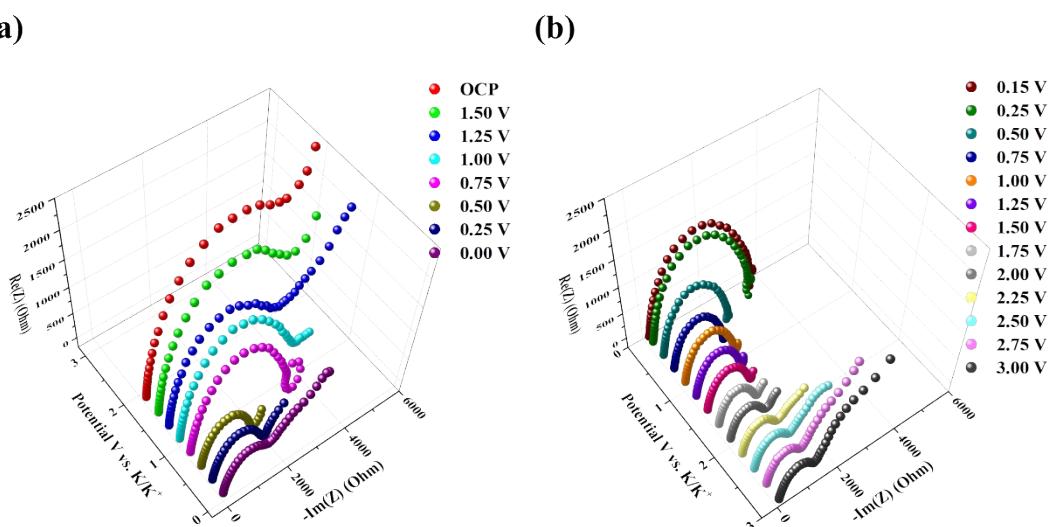
**Figure S14.** Cross-sectional SEM images and the thickness of CIPS@G electrodes.



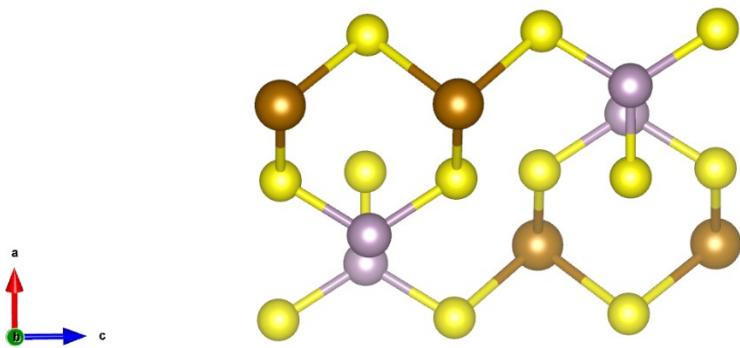
**Figure S15.** Cross-sectional SEM images and the thickness of FePS<sub>3</sub>@G electrodes.



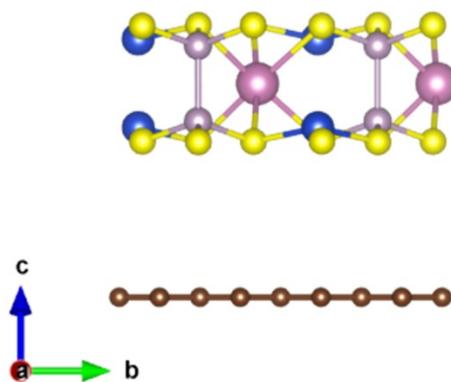
**Figure S16.** (a) GITT curves of FePS<sub>3</sub>@G with 20 min current pulse of 50 mA g<sup>-1</sup> followed by 1 hour relaxation and corresponding diffusion coefficients of (b) discharge and (c) charge process.



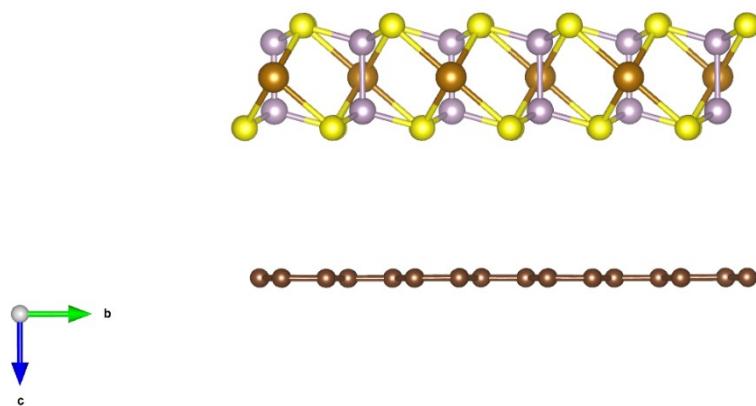
**Figure S17.** In-situ EIS test performed in selected potentials. (a) Discharging process.(b) Charging process.



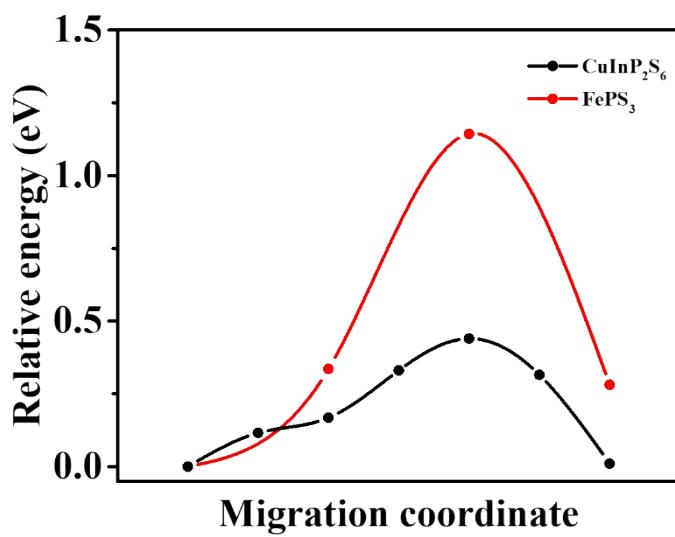
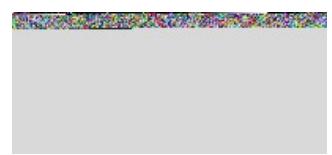
**Figure S18.** Crystal structure of  $\text{FePS}_3$ .



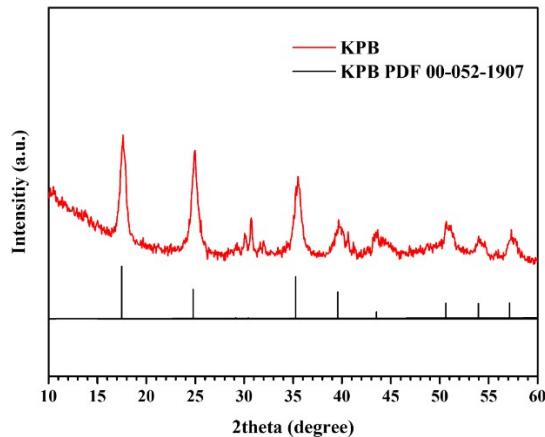
**Figure S19.** Crystal structure of CIPS@G.



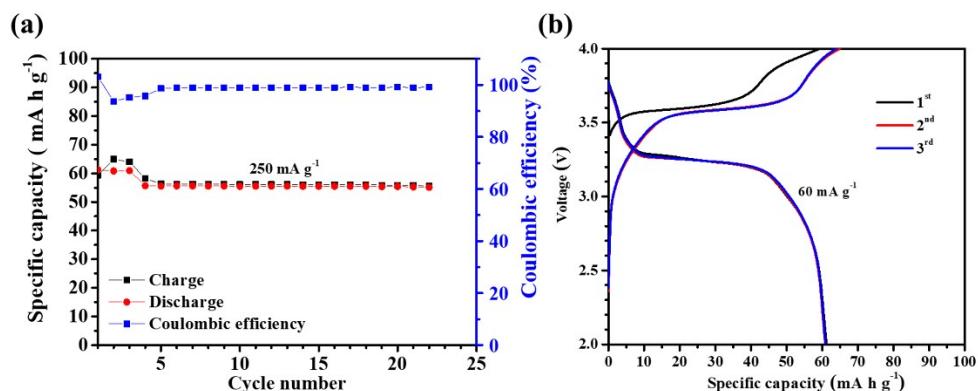
**Figure S20.** Crystal structure of  $\text{FePS}_3@G$ .



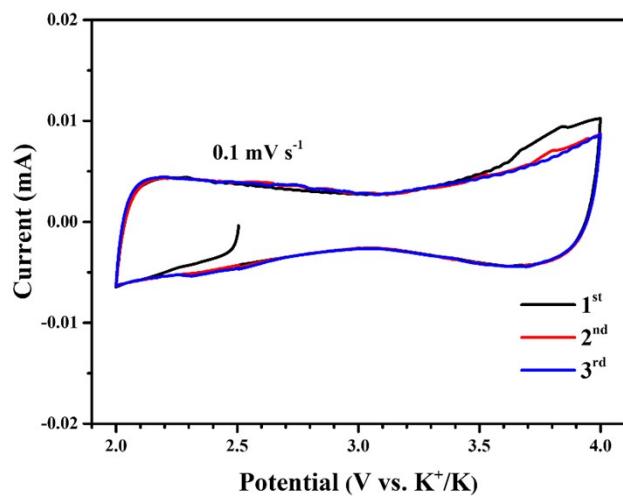
**Figure S21.** Calculated energy barriers of K between CIPS layers and graphene, and between  $\text{FePS}_3$  and graphene.



**Figure S22.** XRD pattern of PB.



**Figure S23.** (a) Cycling performances of PB at  $250 \text{ mA g}^{-1}$  and (b) corresponding galvanostatic charge-discharge curves.



**Figure S24.** Cyclic voltammetry curves of AC electrodes at  $0.1 \text{ mV s}^{-1}$ .