

Supplementary Information for

Versatile Fe_2GeS_4 for Li/Na– Fe_2GeS_4 battery cathodes and Li/Na–ion battery anodes

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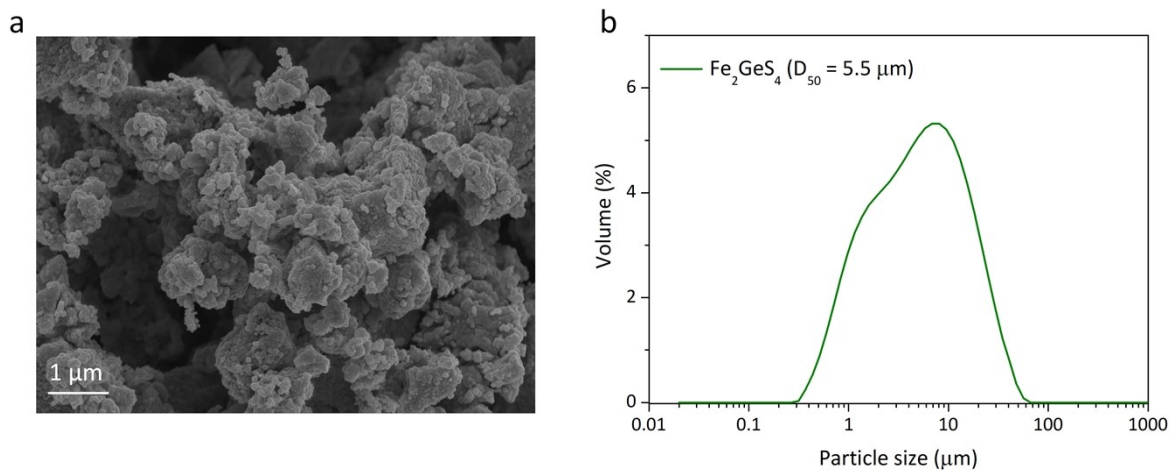


Fig. S1 (a) SEM image and (b) PSA result of the Fe_2GeS_4 .

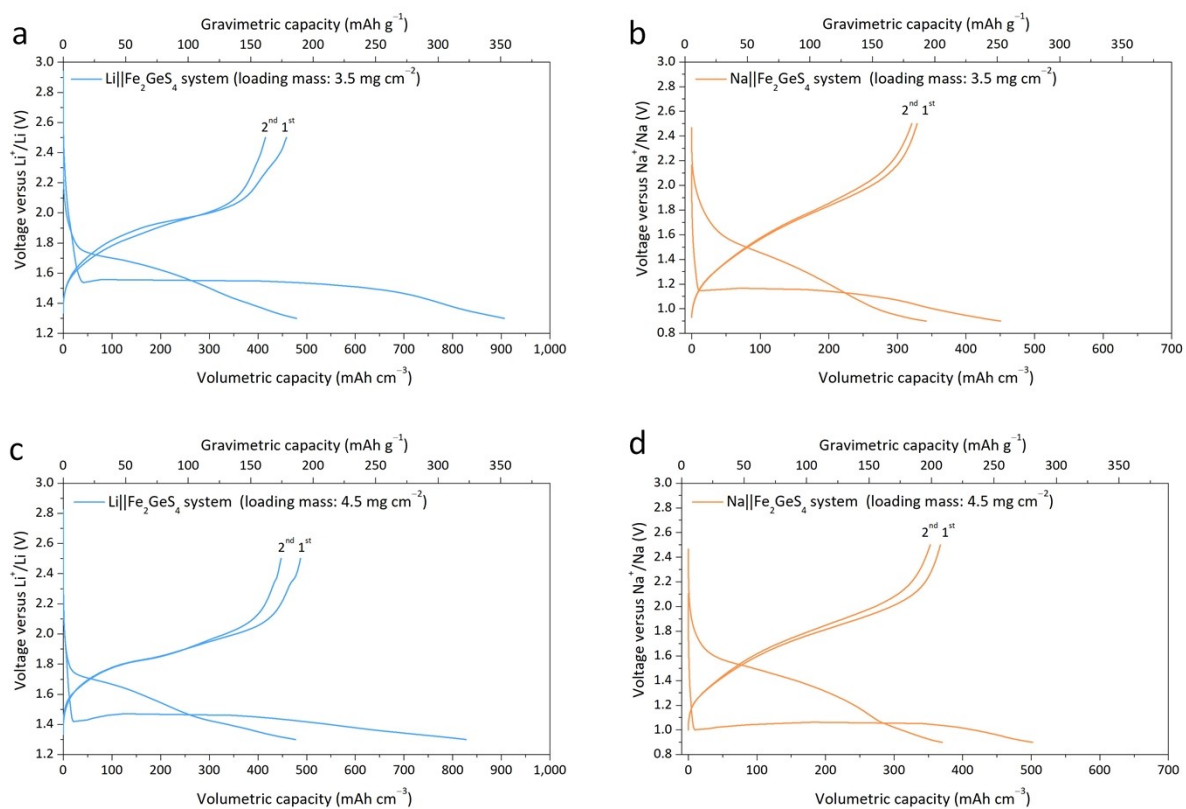


Fig. S2 Electrochemical behavior of Fe_2GeS_4 cathodes at various loading levels of the electrode. Voltage profiles of (a) $\text{Li}||\text{Fe}_2\text{GeS}_4$ and (b) $\text{Na}||\text{Fe}_2\text{GeS}_4$ systems at the loading of $3.5\ \text{mg cm}^{-2}$. Voltage profiles of (c) $\text{Li}||\text{Fe}_2\text{GeS}_4$ and (d) $\text{Na}||\text{Fe}_2\text{GeS}_4$ systems at the loading of $4.5\ \text{mg cm}^{-2}$.

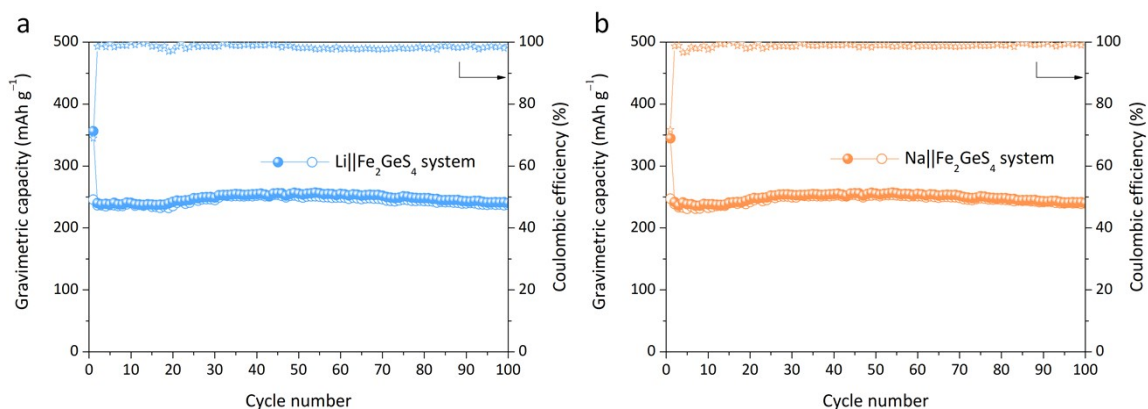


Fig. S3 Cycling performance of (a) Li||Fe₂GeS₄ system (cycling rate: 100 mA g⁻¹) and (b) Na||Fe₂GeS₄ system (cycling rate: 50 mA g⁻¹).

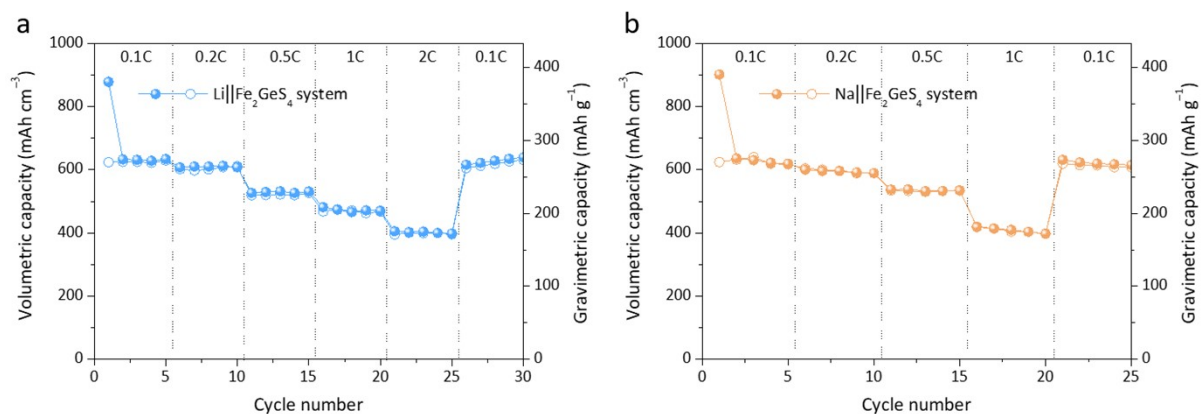


Fig. S4 Rate capability results of Fe₂GeS₄ cathode for Li||Fe₂GeS₄ and Na||Fe₂GeS₄ systems. High-rate cycling performance of (a) Li||Fe₂GeS₄ at various rates (1C: 250 mA g⁻¹) and (b) Na||Fe₂GeS₄ at various rates (1C: 250 mA g⁻¹).

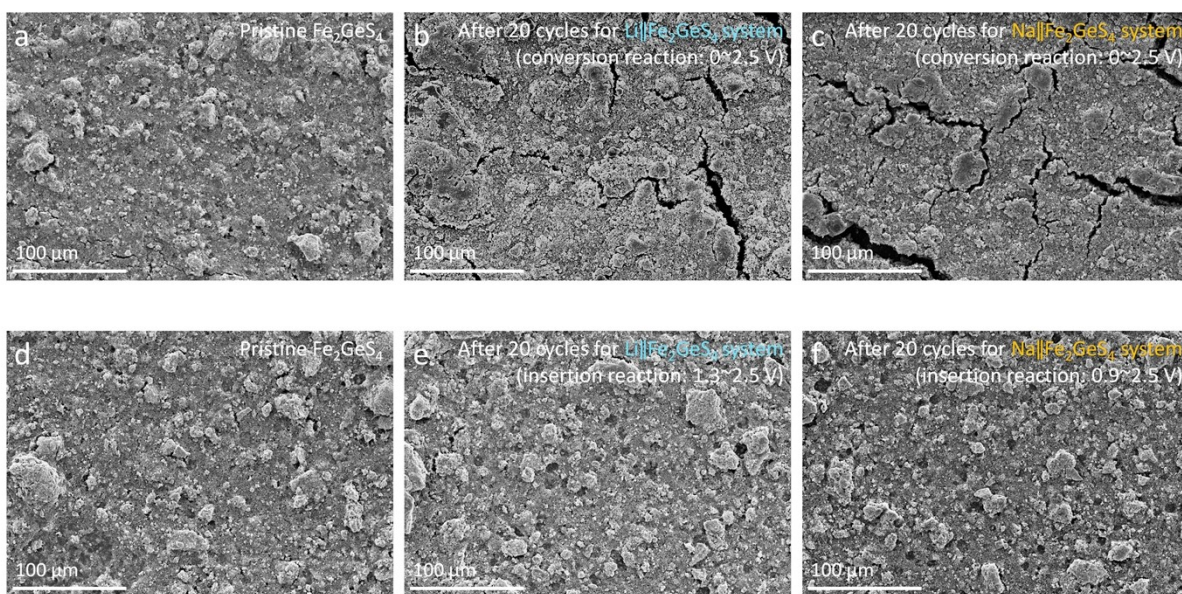


Fig. S5 Morphological stability of Fe_2GeS_4 cathode in $\text{Li}||\text{Fe}_2\text{GeS}_4$ and $\text{Na}||\text{Fe}_2\text{GeS}_4$ systems. *Ex situ* SEM images of Fe_2GeS_4 electrode (a) before cycle and (b, c) after 20 cycles in the conversion reaction voltage range (0–2.5 V) for (b) $\text{Li}||\text{Fe}_2\text{GeS}_4$ and (c) $\text{Na}||\text{Fe}_2\text{GeS}_4$. *Ex situ* SEM images of (d) before cycle and (e, f) after 20 cycles in the insertion reaction voltage range for (e) $\text{Li}||\text{Fe}_2\text{GeS}_4$ (1.3–2.5 V) and (f) $\text{Na}||\text{Fe}_2\text{GeS}_4$ (0.9–2.5 V).

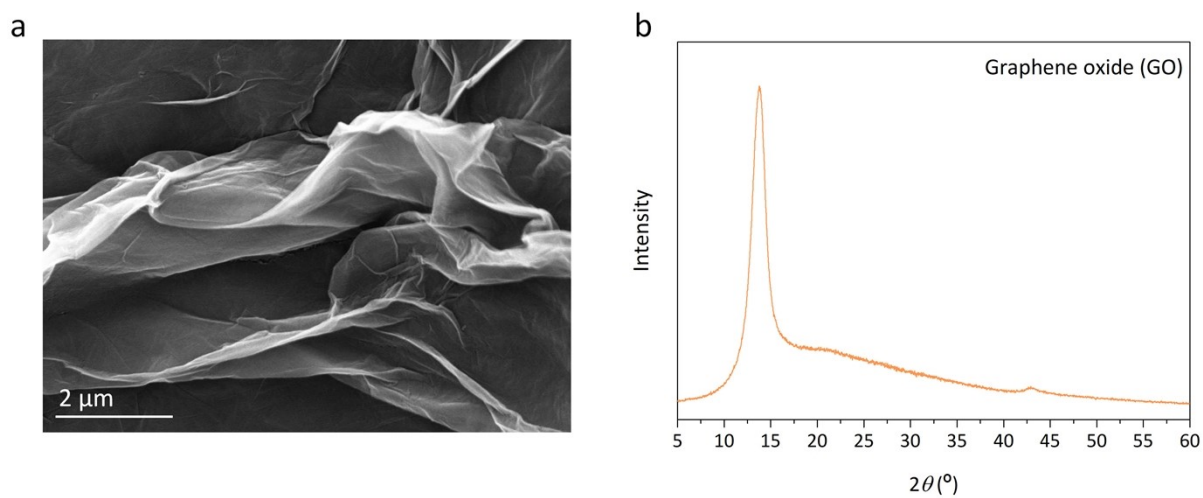


Fig. S6 (a) SEM image and (b) XRD pattern of graphene oxide (GO).

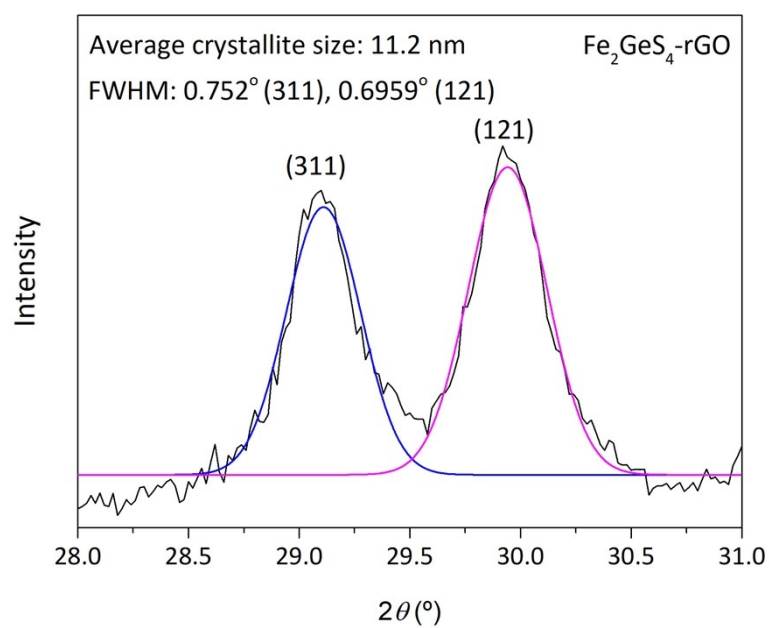


Fig. S7 Average crystallite size of $\text{Fe}_2\text{GeS}_4\text{-rGO}$ calculated using (311) and (121) planes of the XRD result.

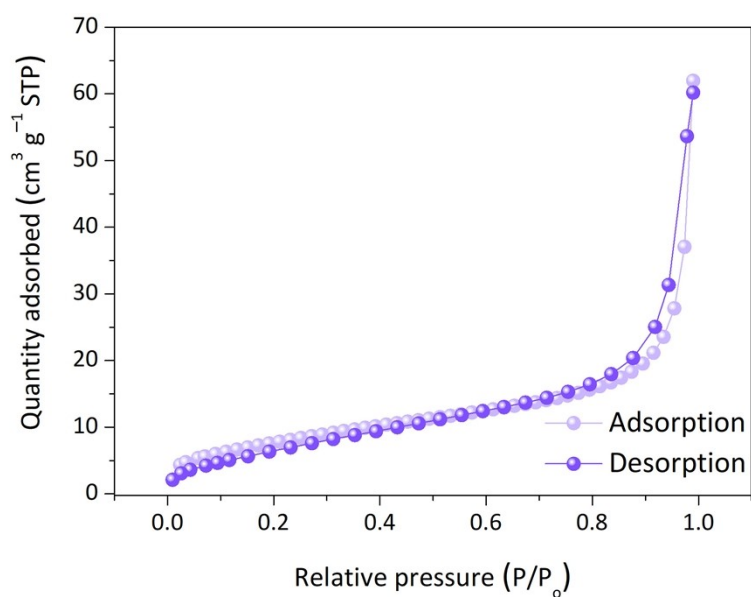


Fig. S8 N_2 adsorption–desorption isotherm plot for Fe_2GeS_4 .

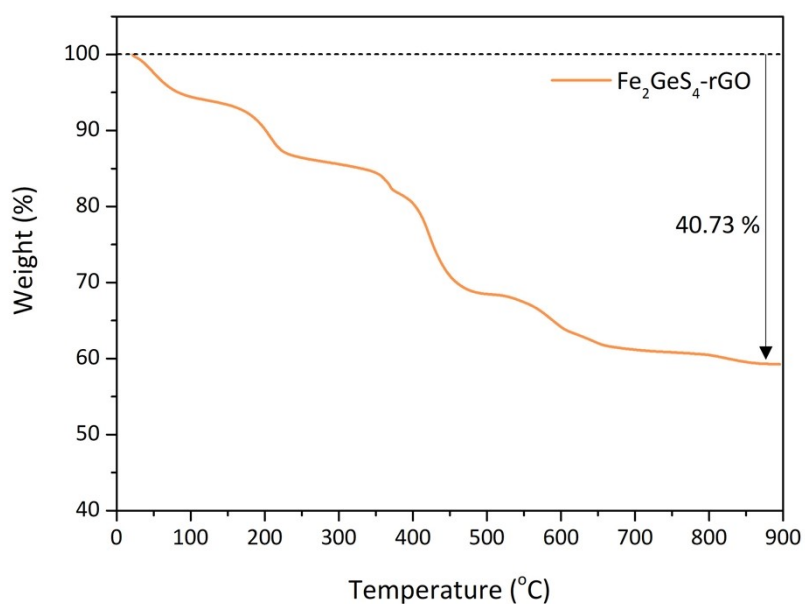


Fig. S9 TGA curve of Fe₂GeS₄-rGO obtained in air atmosphere from room temperature to 900 °C at a rate of 10 °C min⁻¹.

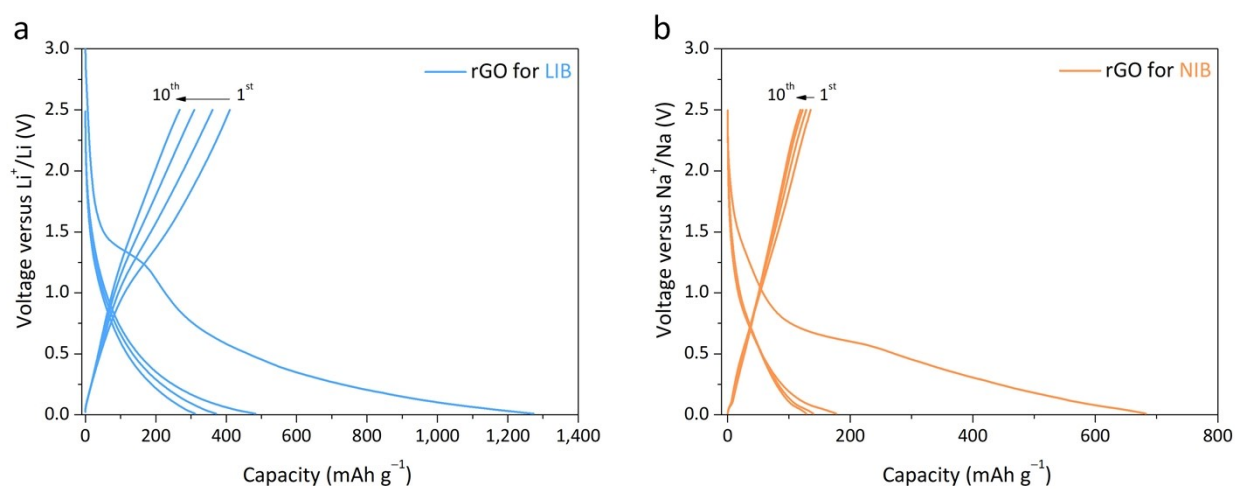


Fig. S10 Electrochemical behavior of rGO. Voltage profiles of rGO for (a) LIB (current density: 100 mA g⁻¹) and (b) NIB (current density: 50 mA g⁻¹).

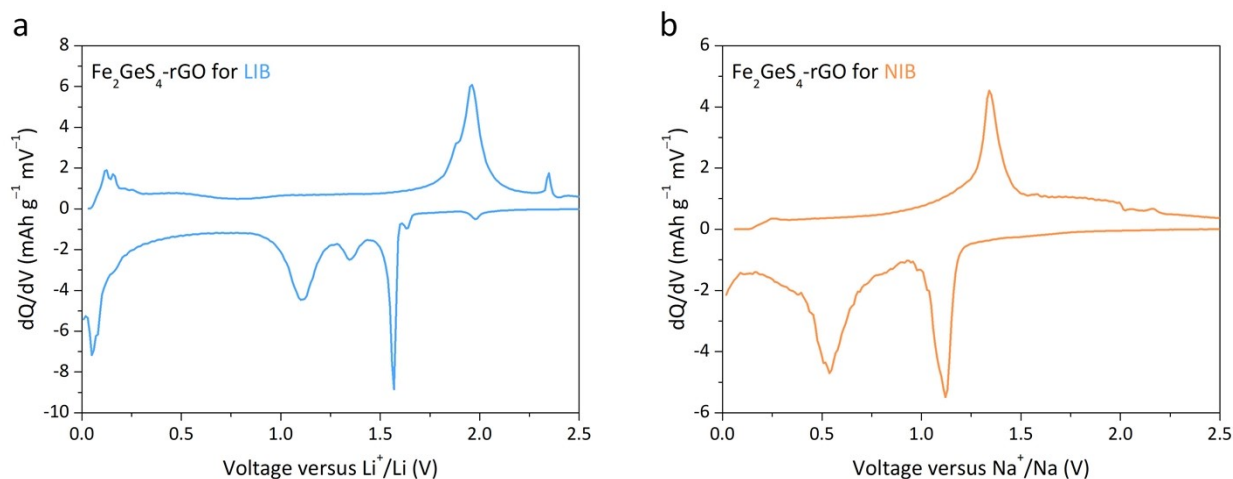


Fig. S11 dQ/dV plots of Fe_2GeS_4 -rGO for (a) LIB and (b) NIB.

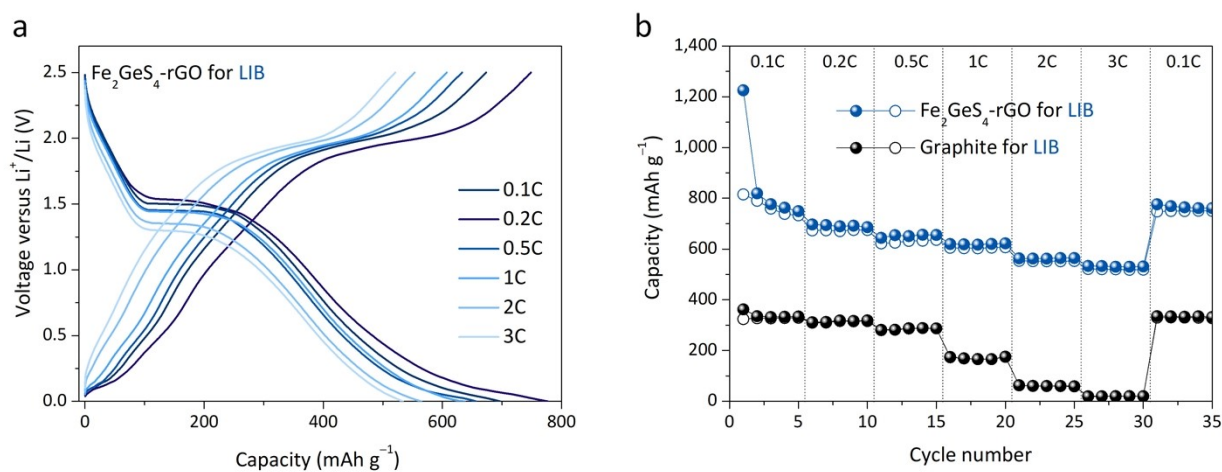


Fig. S12 Li-rate capability results of Fe_2GeS_4 -rGO. (a) Voltage profiles at various rates of Fe_2GeS_4 -rGO. (b) Cycling performance of Fe_2GeS_4 -rGO at various rates (1C: 730 mA g^{-1}) and graphite (1C: 320 mA g^{-1}).

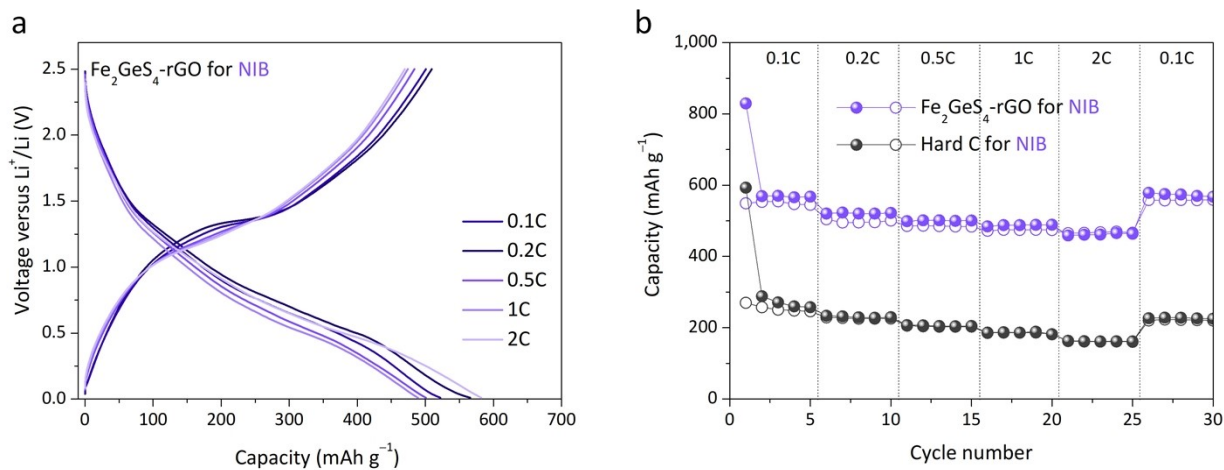


Fig. S13 Na-rate capability results of Fe₂GeS₄-rGO. (a) Voltage profiles at various rates of Fe₂GeS₄-rGO. (b) Cycling performance of Fe₂GeS₄-rGO at various rates (1C: 560 mA g⁻¹) and hard-C (1C: 250 mA g⁻¹).

Table S1. Lattice constants and atomic parameters of Fe₂GeS₄ refined from XRD result

Fe ₂ GeS ₄					
Space group: Pnma, Orthorhombic					
Mw = 312.32 g/mol				R _{bragg} = 7.794%	
a = 12.4708(4) Å, b = 7.2222(3) Å, c = 5.9050(1) Å				R _p = 18.4%	
V = 531.84(3) Å ³				R _{wp} = 15.1%	
Density: 3.903 g/cm ³				χ ² = 9.55	
Atom	Wyckoff site	X	Y	Z	Occ
Fe (1)	4a	0	0	0	0.3433
Fe (2)	4c	0.2266(6)	0.25	0.5053(6)	0.3837
Ge	4c	0.4412(6)	0.25	0.0803(2)	0.3797
S (1)	4c	0.4012(3)	0.25	0.7181(8)	0.3402
S (2)	4c	0.5774(3)	0.25	0.2345(2)	0.3683
S (3)	8d	0.3368(1)	0.0099(5)	0.2497(5)	0.7807

Table S2. Theoretical capacity of Fe₂GeS₄

Battery system	Gravimetric capacity	¹ Volumetric capacity	² Volumetric capacity
	[mAh/g]	[mAh/cm ³]	[mAh/cm ³]
Li-ion storage	1008	3934	2328
Na-ion storage	772	3013	1783

¹: gravimetric capacity × density, ²: gravimetric capacity × tap density

Density: 3.903 g/cm³, Tap density: 2.31 g/cm³