

Supplementary Information

An insight into the sodium-ion and lithium-ion storage properties of CuS/graphitic carbon nitride nanocomposite

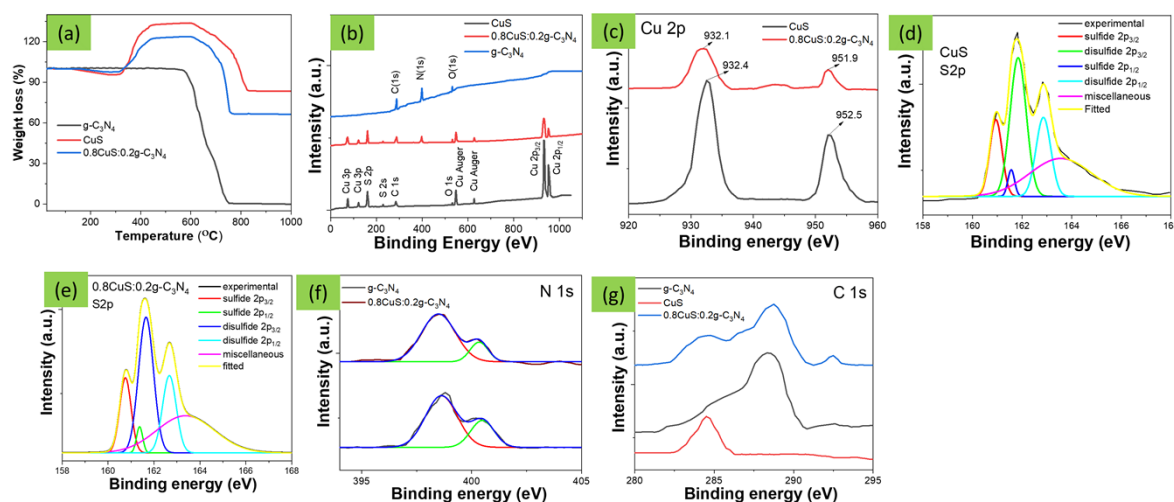


Figure S1: a) TG, b) XPS survey spectra of g-C₃N₄, CuS, 0.8CuS:0.2g-C₃N₄; c) high resolution Cu2p spectrum of CuS and 0.8CuS:0.2g-C₃N₄; Fitted high resolution d) S2p XPS of CuS; e) S2p XPS of 0.8CuS:0.2g-C₃N₄; f) N1s XPS of CuS and 0.8CuS:0.2g-C₃N₄; g) high resolution C1s of g-C₃N₄, CuS, 0.8CuS:0.2g-C₃N₄.

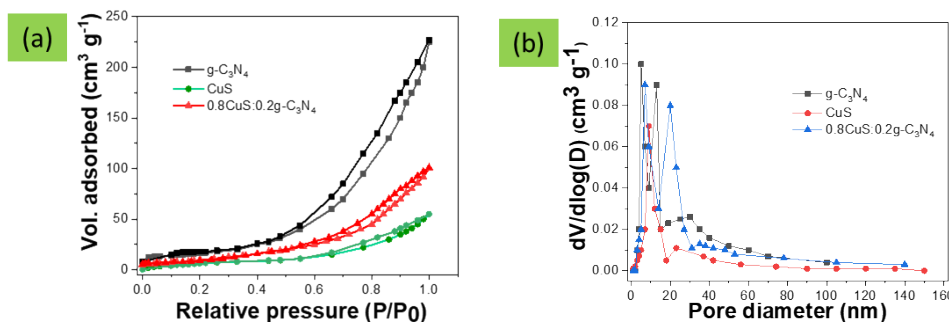


Figure S2: a) BET isotherm and b) BJH pore size distribution plot of g-C₃N₄, CuS, 0.8CuS:0.2g-C₃N₄

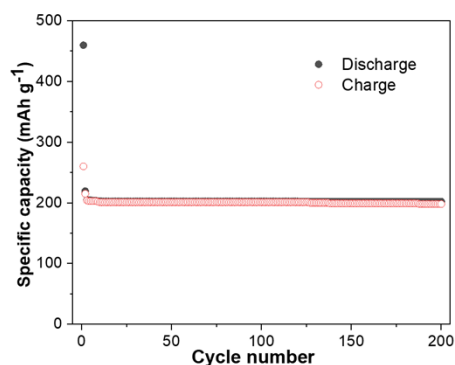


Figure S3: Specific capacity of g-C₃N₄ anode at 0.1 A g⁻¹ current rate in LIB mode

Table S1: Comparison of CuS anode (in this work) with that reported in literature (for LIBs).

Ref.	Synthesis method	Cycling performance		
		Initial discharge/charge (mAh g ⁻¹)	Reversible capacity (mAh g ⁻¹) / current density (A g ⁻¹)	No. of cycles
1	Hydrothermal	525/311	50 / 0.05	10
2	Co-precipitation	708/570	400 / 0.25	100
3	Sol-gel	775/561	390 / 0.11	250
4	Liquid phase	1380/850	259 / 0.1	50
5	Microwave	505/450	379 / 0.2	100
6	Hydrothermal	690/500	203 / 0.1	100
This work	Co-precipitation	930/693	304 / 0.1	200

1. Tao, H.-C.; Yang, X.-L.; Zhang, L.-L.; Ni, S.-B. One-pot facile synthesis of CuS/graphene composite as anode materials for lithium ion batteries. *J. Phys. Chem. Solids* **2014**, *75*, 1205–1209.
2. Feng, C.; Zhang, L.; Yang, M.; Song, X.; Zhao, H.; Jia, Z.; Sun, K.; Liu, G. One-pot synthesis of copper sulfide nanowires/reduced graphene oxide nanocomposites with excellent lithium-storage properties as anode materials for lithium-ion batteries. *ACS Appl. Mater. Interfaces* **2015**, *7*, 15726–15734.
3. Zhou, M.; Peng, N.; Liu, Z.; Xi, Y.; He, H.; Xi, Y.; Liu, Z.; Okada, S. Synthesis of sub-10 nm copper sulphide rods as high-performance anode for long-cycle life Li-ion batteries. *J. Power Sources* **2016**, *306*, 408-412.
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5. Li, H.; Wang, Y.; Huang, J.; Zhang, Y.; Zhao, J. Microwave-assisted synthesis of CuS/graphene composite for enhanced lithium storage properties. *Electrochim. Acta* **2017**, *225*, 443–451

6. Zhang, J.; Zhao, Y.; Zhang, Y.; Li, J.; Babaa, M.-R.; Liu, N.; Bakenov, Z. Synthesis of microflower-like vacancy defective copper sulfide/reduced graphene oxide composites for highly efficient lithium-ion batteries. *Nanotechnology* **2020**, *31*, 095405 (6pp)

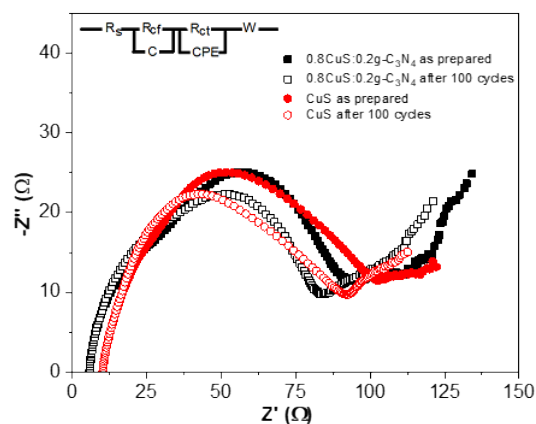


Figure S4: Nyquist plot of as prepared and after 100 cycle run of CuS and 0.8CuS:0.2g-C₃N₄ electrode.

Table S2: Electrochemical performance of CuS based composite LIB anodes reported in literature.

Ref.	Sample	Cycling performance				Rate performance
		Initial discharge/charge (mAh g ⁻¹)	ICE (%)	Reversible capacity (mAh g ⁻¹) / current density (A g ⁻¹)	No. of cycles	Reversible capacity (mAh g ⁻¹) / current density (A g ⁻¹)
1	CuS/graphene	827/484	58.5	296/0.05	25	---/--
2	CuS nanowires/rGO	908/630	69.4	620/0.28	100	300/2.8
3	CuS/PANI/graphene	1655/1266	76.5	1265/0.1	250	374/5
4	CuS/rGO	1236/658	53.2	399/0.1	50	608/1.6
5	CuS/Cu _{1.8} S	1130/707	62.5	450/0.26	1000	195/0.8
6	CuS/graphene	627/525	83.7	348/2	1000	370/4
7	FeS ₂ /CuS	1394/999	71.7	843.3/1.0	600	530.4/10
8	CuS/rGO	882/670	75.9	585/0.11	100	150/1.6
This work	CuS/g-C ₃ N ₄	981/855	87.2	552 /1.0	1000	478.4/2

1. Tao, H.-C.; Yang, X.-L.; Zhang, L.-L.; Ni, S.-B. One-pot facile synthesis of CuS/graphene composite as anode materials for lithium ion batteries. *J. Phys. Chem. Solids* **2014**, *75*, 1205–1209.

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- Zhang, J.; Zhao, Y.; Zhang, Y.; Li, J.; Babaa, M.-R.; Liu, N.; Bakenov, Z. Synthesis of microflower-like vacancy defective copper sulfide/reduced graphene oxide composites for highly efficient lithium ion batteries. *Nanotechnology* **2020**, *31*, 095405 (6pp)

Table S3: Comparison of CuS anode (in this work) with that reported in literature (for SIBs).

Ref.	Synthesis method	Cycling performance			
		Initial discharge/charge (mAh g ⁻¹)	ICE (%)	Reversible capacity (mAh g ⁻¹) / current density (A g ⁻¹)	No. of cycles
1	Microwave	440/400	90.9	311.8 / 0.1	50
2	Solvothermal	700/550	78.6	300 / 0.2	100
3	Hydrothermal	---/400	---	150 / 1.0	100
4	Dealloying	484/444	91.7	132.6 / 5.0	5000
5	Thermal	640/555	86.7	517 / 5.0	2000
6	Hydrothermal ion exchange	456.9/418.6	91.6	418.6 / 0.1	100
This	Co-	738/635	86.0	212/1.0	800

work	precipitation				
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1. Li, J.; Yan, D.; Lu, T.; Qin, W.; Yao, Y.; Pan, L. Significantly improved sodium-ion storage performance of CuS nanosheets anchored into reduced graphene oxide with ether-based electrolyte. *ACS Appl. Mater. Interfaces* **2017**, *9*, 2309–2316.
2. Kim, N. R.; Choi, J.; Yoon, H. J.; Lee, M. E.; Son, S. U.; Jin, H.-J.; Yun, Y. S. Conversion reaction of copper sulfide based nanohybrids for sodium-ion batteries. *ACS Sustainable Chem. Eng.* **2017**, *5*, 9802-9808.
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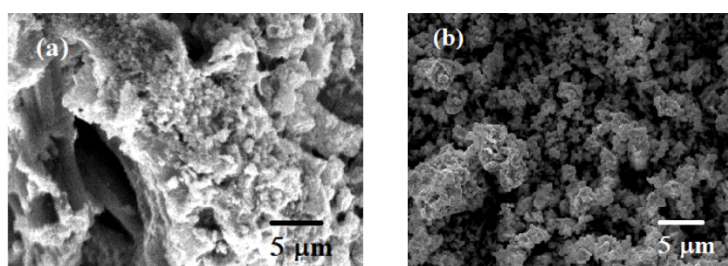


Figure S5: SEM of a) CuS electrode; b) 0.8CuS:0.2g-C₃N₄ composite electrode after 800 cycle run in SIB mode.

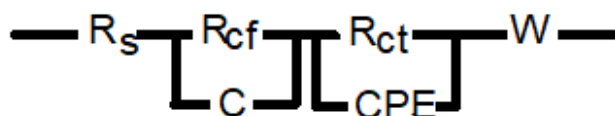


Figure S6: Equivalent circuit diagram for Nyquist plot for as prepared and after run of 100 cycles of CuS and 0.8CuS:0.2g-C₃N₄ composite electrode in SIB mode.

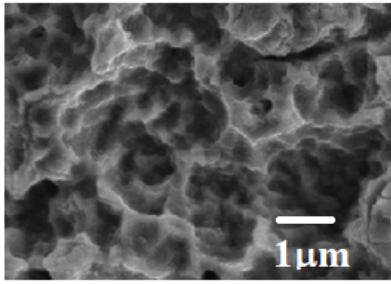


Figure S7: SEM of Cu current collector after 800 cycle run (anode material removed)

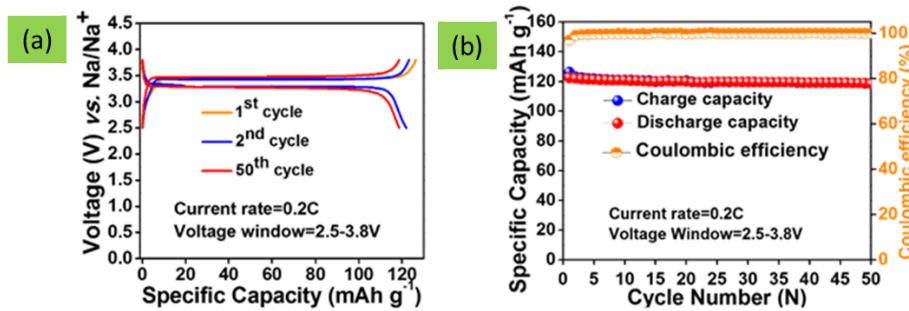


Figure S8: (a) GDC performance of NVP, (b) Cycling performance of NVP at 0.5C; potential range from 2.5 to 3.8 V vs. Na/Na⁺. NVP has been synthesized using the procedure as described in our earlier work [Carbon 143 (2019) 402-412]. The NVP electrodes were prepared by casting a slurry of NVP: Super C-65 (Timcal): Polyvinylidene fluoride (PVDF) in 8:1:1 weight ratio in N-Methyl-2- pyrrolidone (NMP) medium onto Al foil (16 mm). The electrodes were dried in vacuum at 100°C for about 8 h, and the half cell was assembled in an Ar-filled glove box, where both moisture and oxygen levels were kept at less than 1 ppm. 1 M NaClO₄ in a mixture of ethylene carbonate (EC), propylene carbonate (PC) (1:1 v/v) and fluoroethylene carbonate (FEC) (3 wt%) was used as the electrolyte, and a porous borosilicate glass fiber (GF/D, Whatman) was used as a separator.

Table S4: Electrochemical performance of CuS based composite SIB anodes reported in literature.

Ref.	Sample	Cycling performance			Rate performance	
		Initial discharge/charge (mAh g ⁻¹)	ICE (%)	Reversible capacity (mAh g ⁻¹) / current density (A g ⁻¹)	No. of cycles	Reversible capacity (mAh g ⁻¹) / current density (A g ⁻¹)
1	CuS/rGO	780/550 (NaClO ₄ /FEP, 0.005–3.0 V)	70.5	392.9/0.1	50	359.5/1.0
2	CuS /a-SWCNT	710/600	84.5	410/0.1	500	315/3.0
3	CuS/CTAB	747.5/ 684.9	91.6	312.5/10	1000	21.1/40
4	Cu _{1.8} S-C/C	380/350	92.1	372/0.05	110	286/2.8

	core/shell					
5	CuS _x @C	555.4/369.3	66.5	208/2	6300	133.2/10
This work	CuS/g-C ₃ N ₄	755/695	92.1	424 /1.0	800	408/2.0

1. Li, J.; Yan, D.; Lu, T.; Qin, W.; Yao, Y.; Pan, L. Significantly improved sodium-ion storage performance of CuS nanosheets anchored into reduced graphene oxide with ether-based electrolyte. *ACS Appl. Mater. Interfaces* **2017**, *9*, 2309–2316.
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