Supporting Information

Exploring the Sodium Ion Storage Mechanism of Gallium Sulfide (Ga_2S_3): A Combined Experimental and Theoretical Approach

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[1] *Pei Wang and Miao Liu do the same contribution to this article*

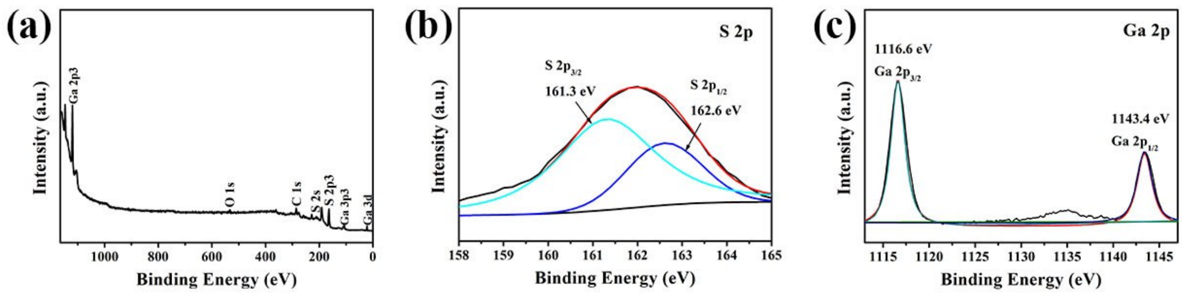


Figure. S1. The full-scan spectra in (a) show the presence of the Ga 2p and S 2p peaks; The Ga 2p and S 2p core levels were scanned respectively. As presented in (b) the fitting of S 2p spectra; (c) the fitting of Ga 2p spectra.

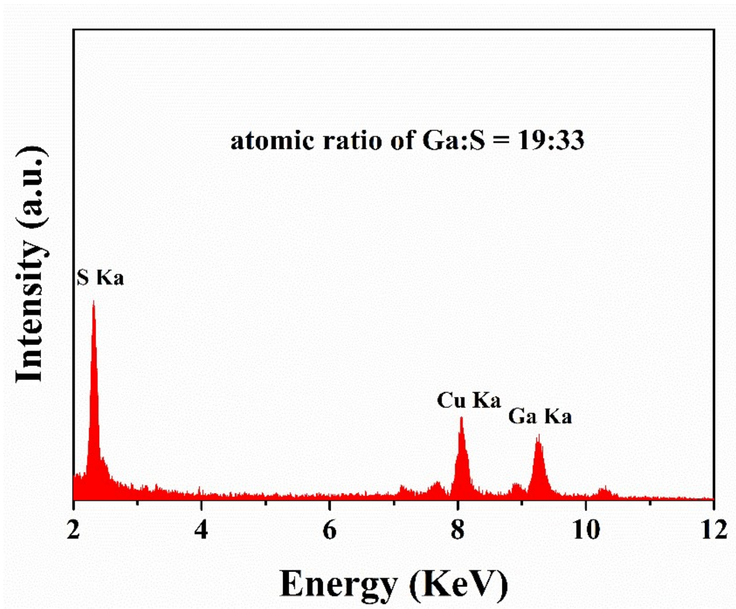


Figure. S2. EDS spectra of as-synthesized Ga_2S_3

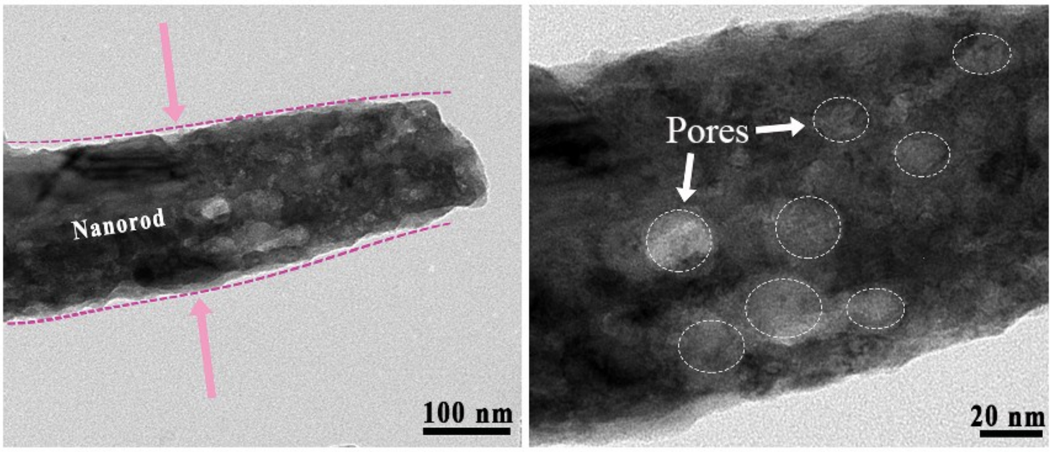


Figure. S3. Typical TEM images of Ga_2S_3 rods to show porous structure inside.

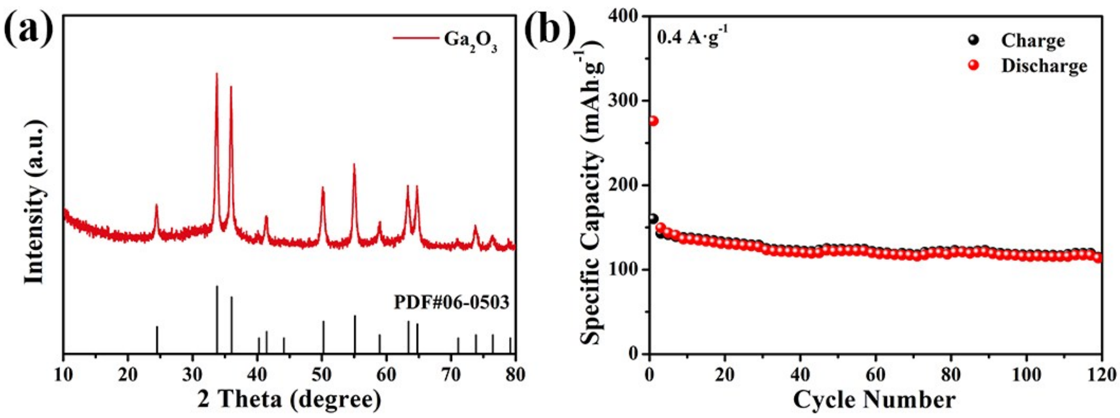


Figure. S4. (a) XRD patterns of Ga_2O_3; (b) cycling performance of Ga_2O_3 at the current density of 0.4 A g^{-1}.

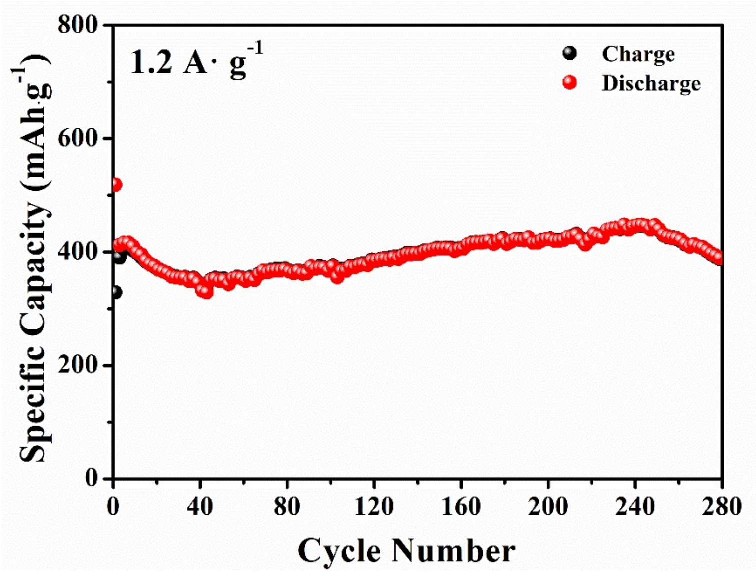


Figure. S5. Long cycling performance of Ga_2S_3 at the current density of 1.2 A g^{-1}.

Table. S1. Comparison of the characteristic parameters of different sulfide-based electrodes for SIBs applications.

Anode matierls	Cycle number	Current	Cycling capacity	Ref
Bimetallic sulfide hollow nanocubes	150	500 mA g^{-1}	87 mAh g^{-1}	1
FeS microspheres	45	200 mA g^{-1}	500 mAh g^{-1}	2
$Co_{1-x}S$/C nanoparticles	130	1000 mA g^{-1}	320 mAh g^{-1}	3
Co_9S_8 quantum dots	500	300 mA g^{-1}	628 mAh g^{-1}	4
CoS_x quantum dots	200	1000 mA g^{-1}	414 mAh g^{-1}	5
CoS_2 nanoparticles	1000	1000 mA g^{-1}	315 mAh g^{-1}	6
CoS_2/rGO composite	100	100 mA g^{-1}	400 mAh g^{-1}	7
CoS_2/carbon composite	60	500 mA g^{-1}	330 mAh g^{-1}	8
M-CoS@C	100	200 mA g^{-1}	532 mAh g^{-1}	9
CoS/rGO	30	100 mA g^{-1}	426 mAh g^{-1}	10
CuS nanodisks	100	100 mA g^{-1}	350 mAh g^{-1}	11
FeS@Fe_3C@graphitic carbon	100	200 mA g^{-1}	575 mAh g^{-1}	12

MoS_2/Co_9S_8/C nanoboxes	100	500 mA g^{-1}	546 mAh g^{-1}	13
SnS@C nanotubes	100	200 mA g^{-1}	426 mAh g^{-1}	14
MoS_2 nanoflakes	100	100 mA g^{-1}	474 mAh g^{-1}	15
NiS/carbon composites	100	200 mA g^{-1}	483 mAh g^{-1}	16
Sn@Sb_2S_3/rGO	60	200 mA g^{-1}	600 mAh g^{-1}	17
Sb_2S_3/C	100	200 mA g^{-1}	545 mAh g^{-1}	18
SnS	50	100 mA g^{-1}	410 mAh g^{-1}	19
SnS_2@rGO	100	50 mA g^{-1}	509 mAh g^{-1}	20
SnS_2/S/rGO	80	100 mA g^{-1}	450 mAh g^{-1}	21
SnS/CNS	100	1000 mA g^{-1}	474 mAh g^{-1}	22
VS_2 microflowers	150	200 mA g^{-1}	330 mAh g^{-1}	23
VS_4@GS nanocomposites	200	500 mA g^{-1}	250 mAh g^{-1}	24
ZnS@C	100	100 mA g^{-1}	768 mAh g^{-1}	25
ZnS quantum dots	100	100 mA g^{-1}	759 mAh g^{-1}	26
$(Co_{1/3}Fe_{2/3})Se_2$	60	300 mA g^{-1}	594 mAh g^{-1}	27

$NiSe_2/C$ porous nanofiber	100	200 mA g^{-1}	468 mAh g^{-1}	28
$NiSe_2$/rGO	1000	1000 mA g^{-1}	346 mAh g^{-1}	29
Nickel-doped Co_9S_8 hollow nanoparticles	200	2000 mA g^{-1}	355 mAh g^{-1}	30
Co_9Se_8/rGO nanocomposites	100	50 mA g^{-1}	406 mAh g^{-1}	31
$Bi_{0.25}Sb_{1.75}Te_3$	100	200 mA g^{-1}	406 mAh g^{-1}	32
$Fe_{1-x}S@Na_{2.9}PS_{3.95}Se_{0.05}$	100	100 mA g^{-1}	494.3 mAh g^{-1}	33
$CoSe_2/(NiCo)Se_2$ box-in-box hollow nanocubes	80	200 mA g^{-1}	497 mAh g^{-1}	34
$CoSe_x$–rGO	50	300 mA g^{-1}	420 mAh g^{-1}	35
$CoSe_x$	50	300 mA g^{-1}	215 mAh g^{-1}	35
Fe_7Se_8@NC nanoparticles	1200	100 mA g^{-1}	339 mAh g^{-1}	36
$FeS_{2-x}Se_x$	6000	2000 mA g^{-1}	220 mAh g^{-1}	37
$FeTe_2$-rGO	80	200 mA g^{-1}	293 mAh g^{-1}	38
Mo_6T_8 (T = S, Se)	50	20 mA g^{-1}	60.3 mAh g^{-1}	39
$MoSe_2/C$	100	500/1000 mA g^{-1}	423/395 mAh g^{-1}	40
$MoSe_2$ grown on N,P-co-doped carbon nanosheets	1000	500 mA g^{-1}	378 mAh g^{-1}	41

$MoSe_2$	120	100/1000 mA g^{-1}	543/491 mAh g^{-1}	42
$Fe_{0.3}Nb_{0.7}S_{1.6}Se_{0.4}$ Nanosheets	750	1000 mA g^{-1}	250 mAh g^{-1}	43
RGO/Ni_3S_2/Ni	100	/	433 mAh g^{-1}	44
Ni_3Se_2	100	500 mA g^{-1}	339.7 mAh g^{-1}	45
$Ni_{0.85}Se$/C hollow nanowires	100	0.2C	390 mAh g^{-1}	46
Nitrogen-doped CoSe/C	50	500 mA g^{-1}	531.6 mAh g^{-1}	47
N-Doped Graphene@Sb_2Se_3	50	100 mA g^{-1}	548.6 mAh g^{-1}	48
Sb_2Se_3/rGO	500	1000 mA g^{-1}	417 mAh g^{-1}	49
Sb_2Te_3/C	400	1000 mA g^{-1}	360 mAh g^{-1}	50
SnSe NSCs nanosheet clusters	100	200/2000/5000 mA g^{-1}	271/183/70 mAh g^{-1}	51
SnSe nanoplates	50	300 mA g^{-1}	558 mAh g^{-1}	52
SnTe/C	100	50 mA g^{-1}	316 mAh g^{-1}	53
Sb_2Te_3@RGO	80	100 mA g^{-1}	514 mAh g^{-1}	54
Nano Te@C	1000	60 mA g^{-1}	345 mAh g^{-1}	55
Ga_2S_3	**100**	**400 mA g^{-1}**	**476 mAh g^{-1}**	**this work**

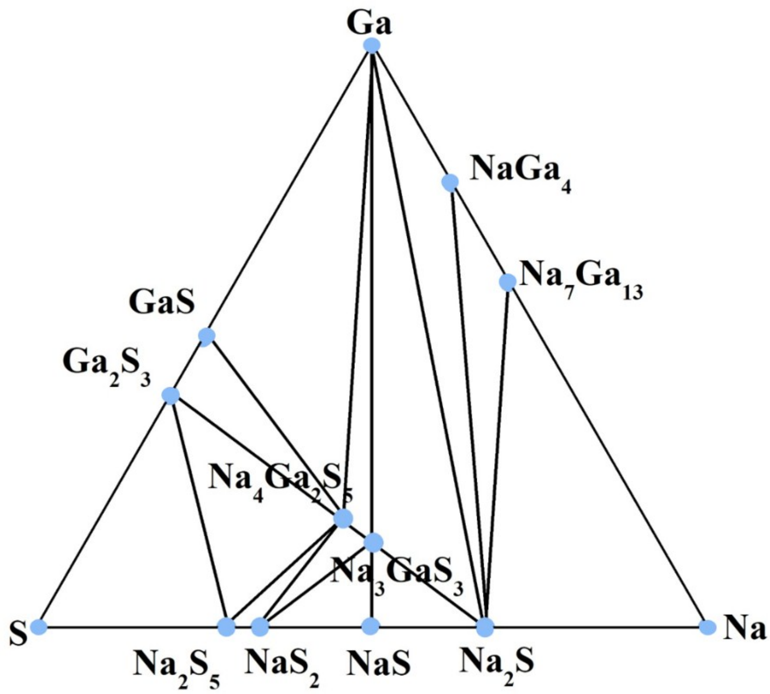


Figure. S6. The phase diagram presents all the thermodynamically stable phases in the Ga-S-Na chemical system (blue dots)

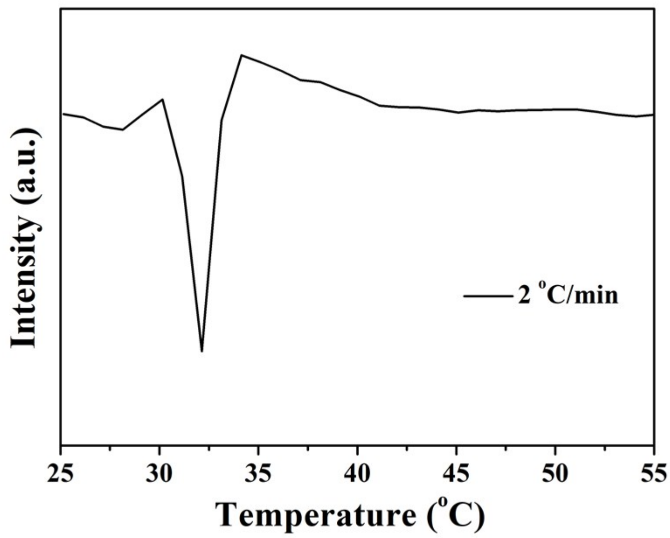


Figure S7. DSC traces (2◦/min, Ar flow) for electrodes discharged at 0.5 V

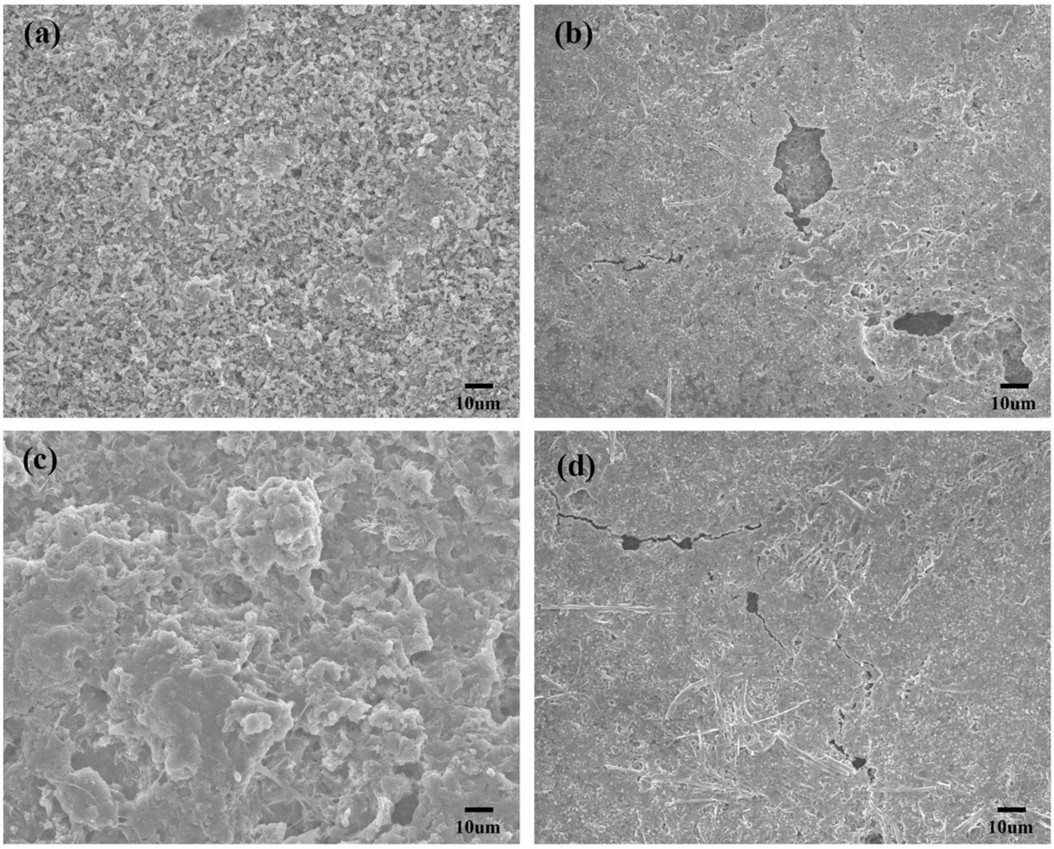
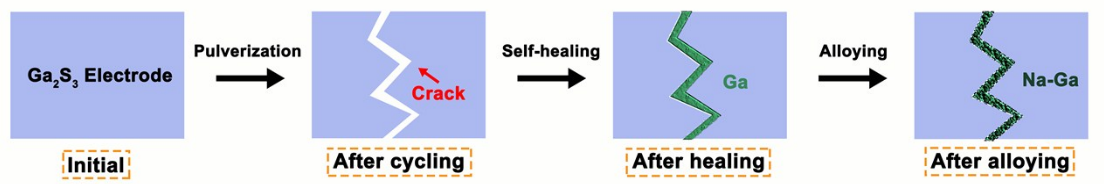


Figure S8. SEM observation of Ga_2S_3 electrode during the discharge: (a) initial state; discharged at (b) 0.7 V (c) 0.4 V and (d) 0.01 V.



Schematic R1. Illustrate the self-healing process of Ga-based material upon discharge.

Assemble of K Ion Battery. To prepare working electrodes, active material, graphene and binder (polyvinylidene fluoride, PVdF) were mixed in a weight ratio of 70:20:10; 1-methyl-2-pyrrolidinone solvent (Aldrich, 99%) was added to form slurry. The well-mixed slurry was coated onto a Cu foil as a current collector using a doctor blade method and then dried under vacuum at 80 °C overnight. Coin-type half-cells were assembled inside an argon-filled glove box (<0.1 ppm, H_2O and O_2), using the metallic potassium foil as the counter electrode, the Whatman glass fiber as the separator and 0.5 L mol^{-1} CF_3SO_3K solution in diethylene glycol dimethyl ether as the electrolyte. The charge-discharge cycling was performed within the voltage range of 0.01−3.00 V vs. K/K$^+$ on a battery test instrument (CT2001A, LAND).

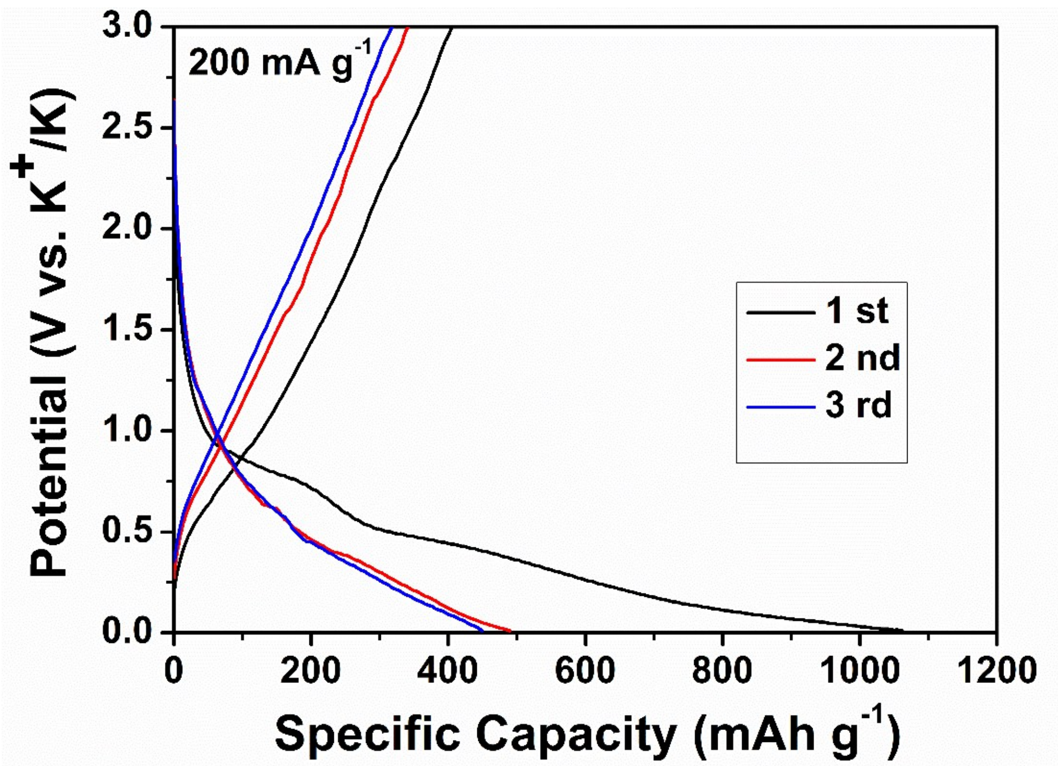


Figure S9. Galvanostatic charge-discharge voltage profiles of the Ga_2S_3 for K ion battery

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