Supporting information

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Section I. Calculation Process

S1 Calculation process for sodium intercalation coefficients

The low frequency line in the Nyquist plot is associated with the sodium ion diffusion in the electrode materials. Thus the sodium ion diffusion coefficient could be calculated from the low frequency plots according to the following equation [S1-S3]:

$$\mathbf{D} = R^2 T^2 / 2A^2 n^4 F^4 C^2 \sigma^2 \tag{1}$$

Where *R* is the gas constant, *T* is the absolute temperature, *A* is the surface area of the cathode, *n* is the number of electrons per molecule during oxidation, *F* is the Faraday constant, *C* is the concentration of lithium ion, σ is the Warburg factor which is relative with *Z*' [S1-S3].

$$Z' = R_e + R_{ct} + \sigma \omega^{-1/2} \tag{2}$$

Where ω is the frequency. Based on the relationship between Z' and the reciprocal square root of frequency in the low frequency region, the sodium diffusion coefficients can be calculated.

References:

[S1] H. Liu, Q. Cao, L. J. Fu, C. Li, Y. P. Wu, H. Q. Wu, *Electrochem. Commun.* 2006, 8, 1553-1557.

[S2] Q. Cao, H. P. Zhang, G. J. Wang, Q. Xia, Y. P. Wu, H. Q. Wu, *Electrochem. Commun.* 2007, 9, 1228-1232.

[S3] A. J. Bard, L. R. Faulkner, Electrochemical Methods, second edition, Wiley, 2001, p.231.

Section II. Supporting Tables

Table S1 Atomic parameters of the "top" precursor Na₂Fe(SO₄)₂·4H₂O refined from the XRD data. Space group: $P2_{1/c}$, R_{wp} =5.31%, R_{p} =3.59%. a=5.5638(7) Å, b=8.2718(0) Å, c=11.1895(2) Å, β =100.198(6)°

Atom	Wyck. site	x	у	Z
Na	4 <i>e</i>	0.6212(3)	0.5661(6)	0.8618(1)
Fe	2a	0.5	0.5	0.5
S	4 <i>e</i>	0.8765(2)	0.7878(7)	0.6331(8)
O1	4 <i>e</i>	0.8132(8)	0.6351(6)	0.5687(8)
02	4 <i>e</i>	0.7078(0)	0.9181(7)	0.5769(6)
O3	4 <i>e</i>	1.1354(0)	0.8361(1)	0.6286(0)
O4	4 <i>e</i>	0.8489(2)	0.7758(6)	0.7616(7)
05	4 <i>e</i>	0.3702(2)	0.5446(1)	0.6616(8)
O6	4 <i>e</i>	0.3177(0)	0.7174(9)	0.4170(3)
H1	4 <i>e</i>	0.1857(7)	0.6830(0)	0.6737(7)
H2	4 <i>e</i>	0.3345(0)	0.4862(6)	0.6904(9)
H3	4 <i>e</i>	0.1016(3)	0.6670(3)	0.3374(2)
H4	4e	0.2085(4)	0.7929(1)	0.4368(9)

Table S2 Atomic parameters of the "down" product off-stoichiometry alluaudite Na_{2+2x}Fe_{2-x}(SO₄)₃ refined from the XRD data. Space group: C2/c, a=12.8330(0) Å, b=12.9733(7) Å, c=6.5460(5) Å, $\beta=115.802(8)^{\circ}$

Atom	Wyck. site	x	у	z
Na1	4 <i>e</i>	0	0.7701(8)	0.25
Na2	4b	0	0	0
Na3	4 <i>e</i>	0	0.5073(8)	0.25
Fe1	8 <i>f</i>	0.7367(2)	0.1248(0)	0.1702(4)
S 1	4e	0	0.1984(0)	0.25
011	8 <i>f</i>	0.0667(6)	0.9735(9)	0.8389(2)
012	8 <i>f</i>	0.3948(0)	0.2255(8)	0.5303(1)
S2	8 <i>f</i>	0.7795(7)	0.6012(1)	0.7991(3)
O21	8 <i>f</i>	0.7304(0)	0.7389(9)	0.7267(0)
O22	8 <i>f</i>	0.3665(4)	0.9796(4)	0.4609(0)
O23	8 <i>f</i>	0.3247(0)	0.6162(4)	0.5996(5)
O24	8 <i>f</i>	0.3011(4)	0.1400(5)	0.0442(0)

Table S3 BET area, pore volume and carbon content of the muscle-inspired $Na_{2+2x}Fe_{2-x}(SO_4)_3/SWNT$ sample and the reference sample.

Materials	BET area $/m^2g^{-1}$	Pore volume/m ³ g ⁻¹	Carbon content/%
Muscle-inspired	64.1	0.16	4.03
Reference	6.2	0.03	0

Section III. Supporting Figures

Figure S1 SEM images of the reference sample.

