Supporting Information

Solid-gas synthesis of stable V_3S_4 nanoflakes: electrochemical characterization as Li-ion battery anode

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Materials:

All the chemicals used in this work were of AR grade. Vanadium oxide (V_2O_5) poweder was purchased from Sigma-Aldrich with 99.95% purity. Ammonium sulfide solution of 20 wt% and Sulfur powder with 98% purity were purchased from Loba chemicals. The electrolyte solution was bought from the Sigma-Aldrich and the Whatman glass microfiber (GF/D) was used as the separator.

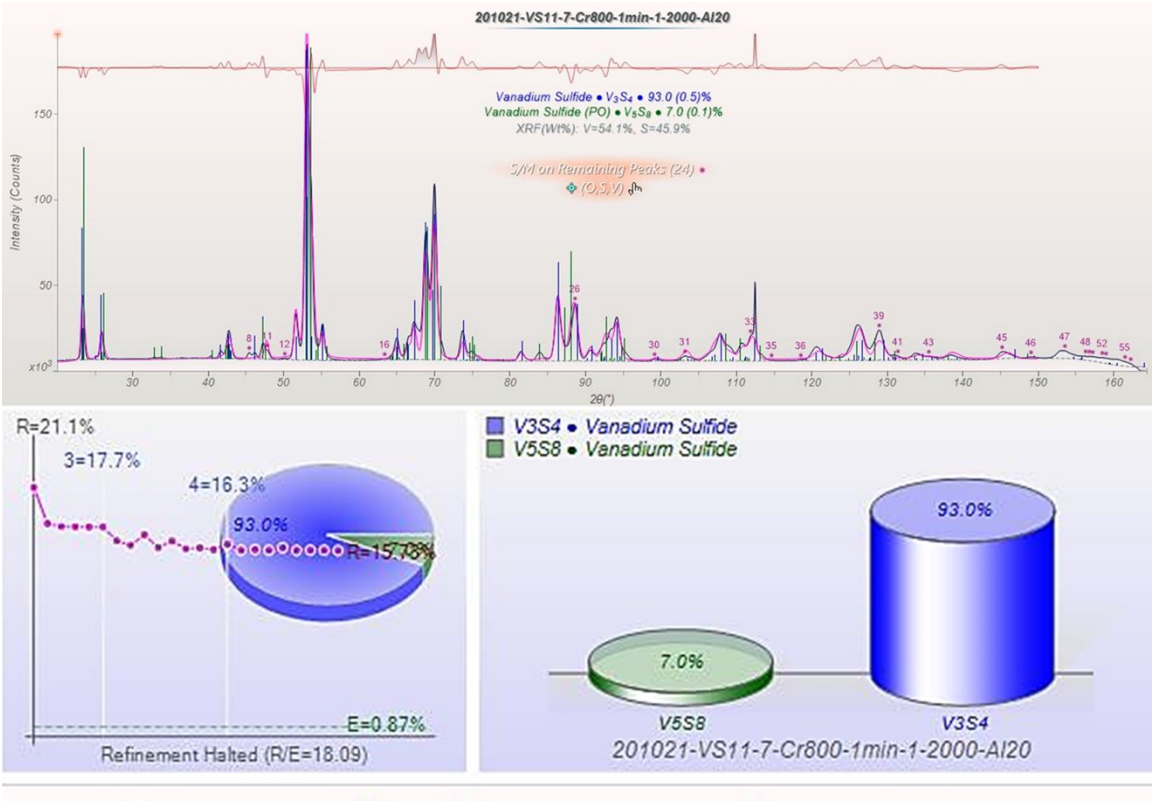

Figure S1: Rietveld refinement of vanadium sulfide (from Fig.1a in the manuscript).

Rietveld analysis carried out using MDI JADE PRO, revealed the phase fraction of V_3S_4 and V_5S_8 are around 93 wt.%, and wt.7% respectively. There were no vanadium oxide phase peaks found to be present from Rietveld refinement.

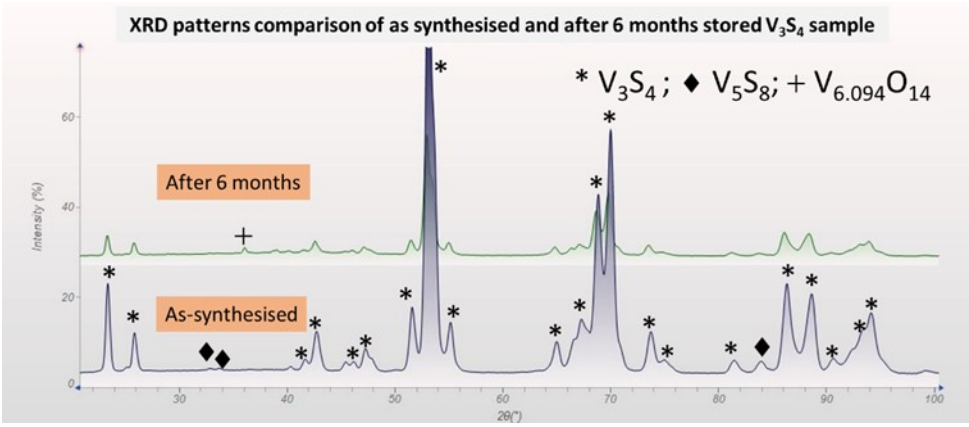

Figure S2: Comparative XRD plot of the as-prepared V_3S_4 (as-prepared and after six months exposure in air).

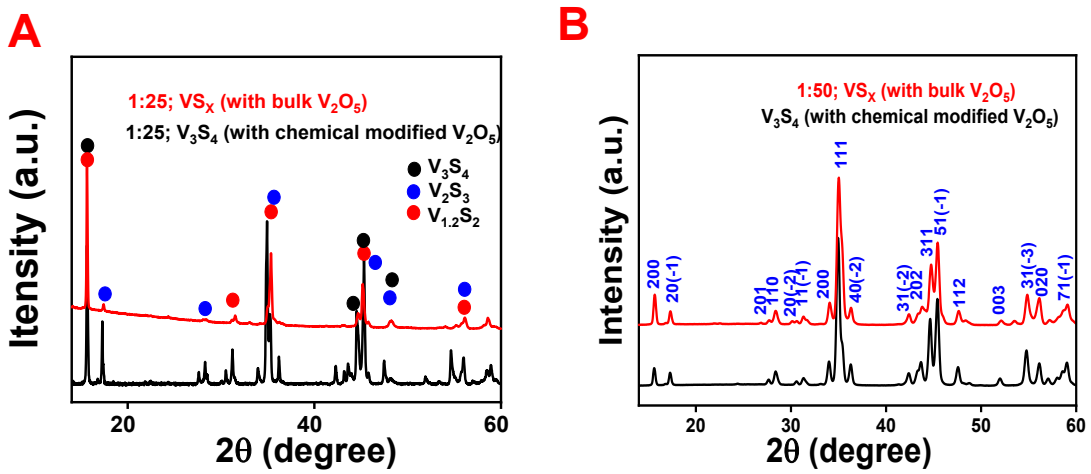

Figure S3: Comparative XRD profile of vanadium sulfide prepared from chemical modified V_2O_5 and ball milled V_2O_5 with different sulfur powder (A) 1:25 and (B) 1:50.

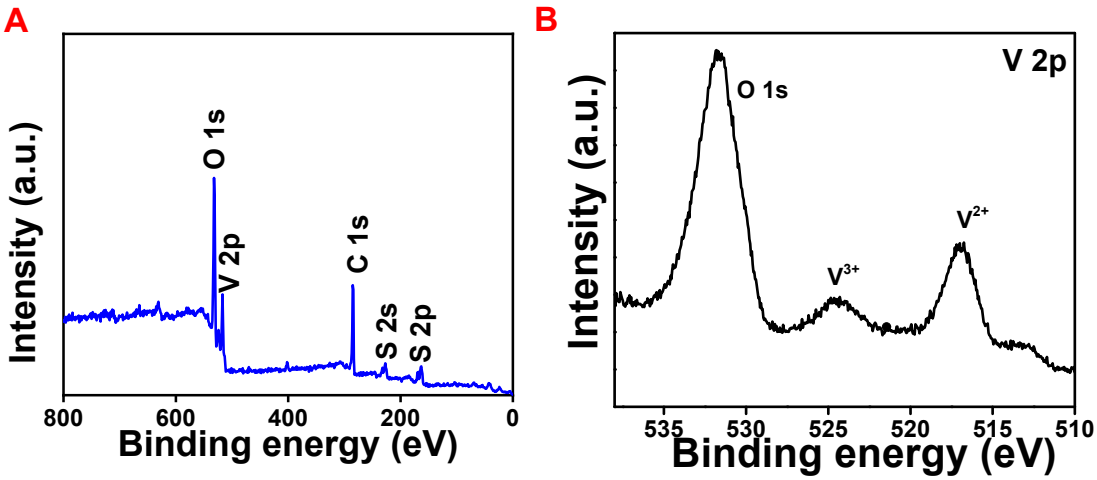

Figure S4: X-ray photoelectron spectroscopy (XPS) of the as-prepared V_3S_4 nanoflakes; (A) wide range spectrum and (B) narrow range V 2p with O 1s spectra.

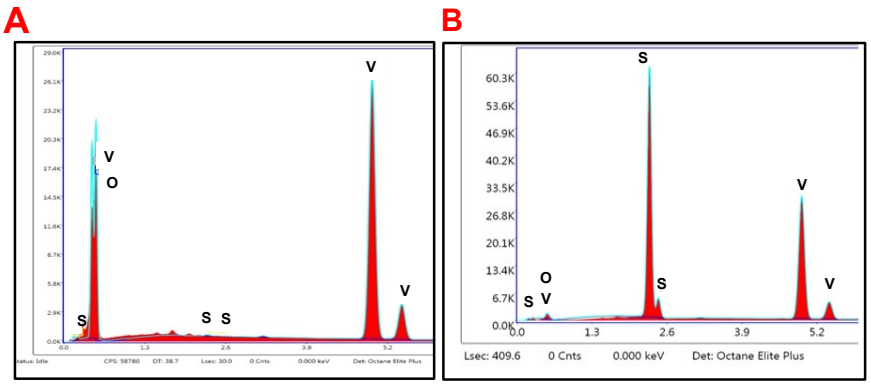

Figure S5: EDS spectra of **(A)** chemically modified product and **(V)** V_3S_4 nanoflakes.

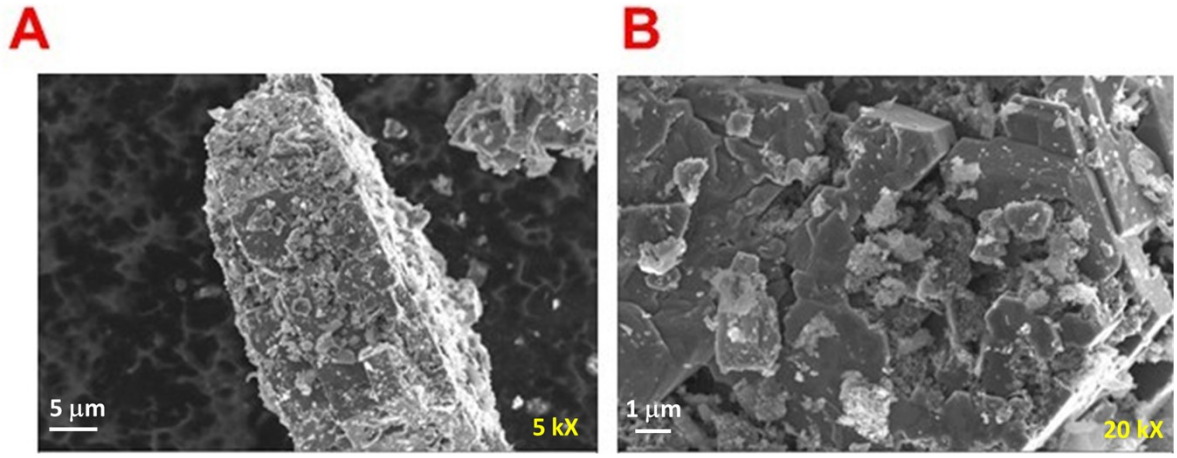

Figure S6: The FESEM image of the vanadium sulfide sample prepared by ball-milled V_2O_5 precursor (without chemical modification).

Table S1: Various synthesis strategies and comparative specific capacity values of the different V_3S_4 and its composite.

Materials	**Synthesis strategy**	**Precursors and synthesis parameters**	**Specific capacity (mA h g^{-1}) at specific current (mA g^{-1}) as LIB anode**	**Ref.**
V_3S_4-3D graphene	Hydrothermal followed by annealing	**Hydrothermal**: ammonium vanadate and ammonia **Sulfidation**: H_2S gas and Argon, 900 °C	1165 at 0.07	S1

V_3S_4@C nanosheets	Hydrothermal followed by sulfidation	Hydrothermal: V_2O_5 powder, 1, 4-benzenedicarboxylic acid and ascorbic acid **Sulfidation**: thioacetamide, nitrogen gas, 500 °C	1065 at 0.1	S2
V_3S_4/carbon	Electrospinning followed by thermal sulfidation	**Electrospinning:** vanadium(V)oxytriisopropoxide and Polyvinyl acetate **Sulfidation:** sulfur, argon and hydrogen gas, 800 °C	790 at 0.01	S3
V_3S_4/N,S-GO	Hydrothermal followed by annealing	Hydrothermal: NH_4VO_3 and $(NH_2)_2CS$, graphene oxides **Sulfidation**: Argon gas, 600 °C	1150 at 0.05	S4
V_3S_4 nanoflakes	Hydrothermal followed by solid-gas reaction	Hydrothermal: V_2O_5 powder, $(NH_4)_2S$ **Sulfidation:** sulfur and hydrogen gas, 500 °C	781 at 0.025	**This work**

References:

S1 Q. Liu, W. Yao, L. Zhan, Y. Wang and Y. A. Zhu, *Electrochim. Acta*, 2018, **261**, 35–41.

S2 Y. Liu, Z. Sun, X. Sun, Y. Lin, K. Tan, J. Sun, L. Liang, L. Hou and C. Yuan, *Angew. Chemie Int. Ed.*, 2020, **59**, 2473–2482.

S3 S. Husmann, Ö. Budak, A. Quade, A. Frank, A. Kruth, C. Scheu, A. Tolosa and V. Presser, *J. Power Sources*, 2020, **450**, 227674.

S4 N. Wu, D. Miao, X. Zhou, L. Zhang, G. Liu, D. Guo and X. Liu, *Nanomaterials,* 2019, **9**, 1638.