## Supporting information

# Lithium Reaction Mechanism and High Rate Capability of $VS_4$-Graphene Nanocomposite for Lithium-ion Batteries Anode Materials

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+ **equally contributed**

### Functionalization of CNT

In the initial step, CNT (Sigma-Aldrich) was treated with a mixture of concentrated sulfuric acid and nitric acid (3:1, 95% and 60%) followed by ultrasonication at 50° C. Further, the product was diluted with water and kept for overnight. The obtained product was filtered and vacuum dried to get the functionalized CNT.

### Synthesis of $VS_4$-10 wt% CNT composite

$VS_4$-10 wt% CNT composite was prepared by following the same procedure used for the synthesis of $VS_4$-rGO. At first, functionalized CNT solution (30 mg/mL) was prepared for the hydrothermal synthesis. $Na_3VO_4$ (0.552 g, 0.003 mol) and $C_2H_5NS$ (1.125 g, 0.015 mol) were dissolved in 115 mL DI water. Then, 5 mL CNT solution was added. The mixture was stirred for 1 h at room temperature, and transferred to a 150 mL Teflon-lined stainless steel autoclave, sealed tightly and kept at 160 $^oC$ for 24 h. The carbon content of the as-prepared $VS_4$-CNT composite was 10 wt% according to elemental analysis.

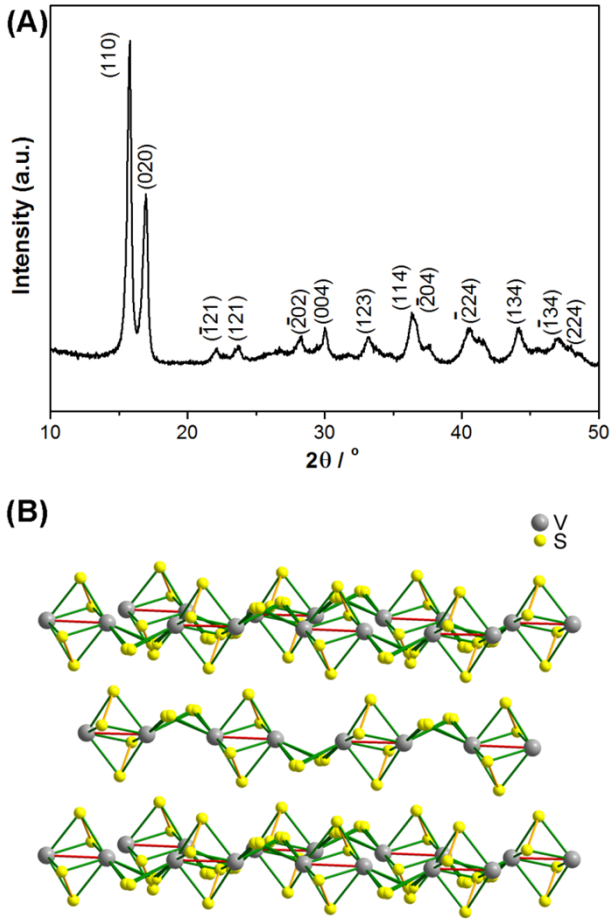


**Fig. S1** (A) Powder XRD pattern of the as-prepared $VS_4$-rGO composite. (B) Structure of linear-chained $VS_4$ with alternating bonding and nonbonding contacts between the octa-coordinated vanadium centers.

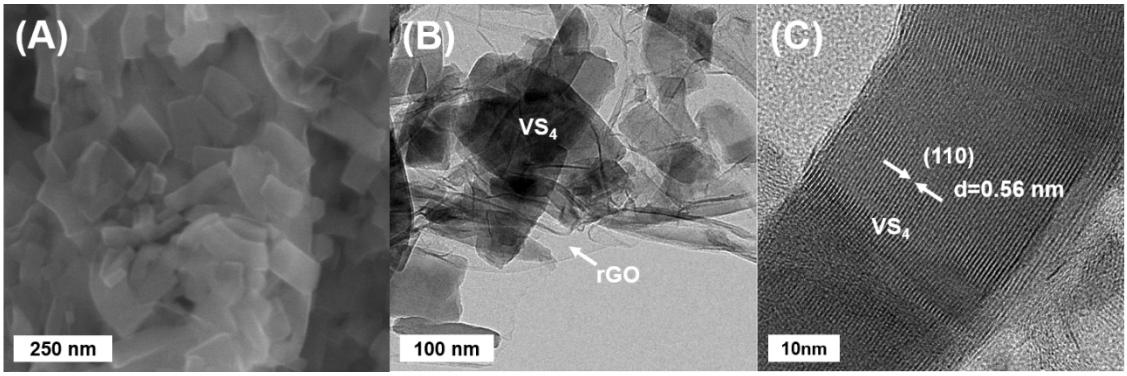


**Fig. S2** (A) SEM (B) TEM, and (C) HR-TEM of $VS_4$ in the as-prepared $VS_4$-rGO composite.

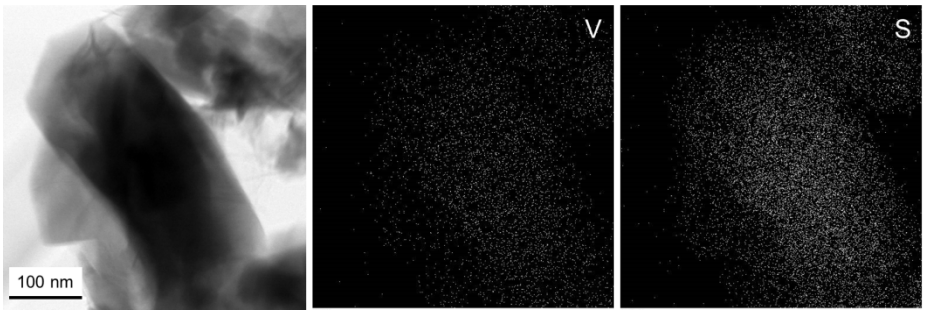


**Fig. S3** EDS mapping images of the $VS_4$-rGO composite.

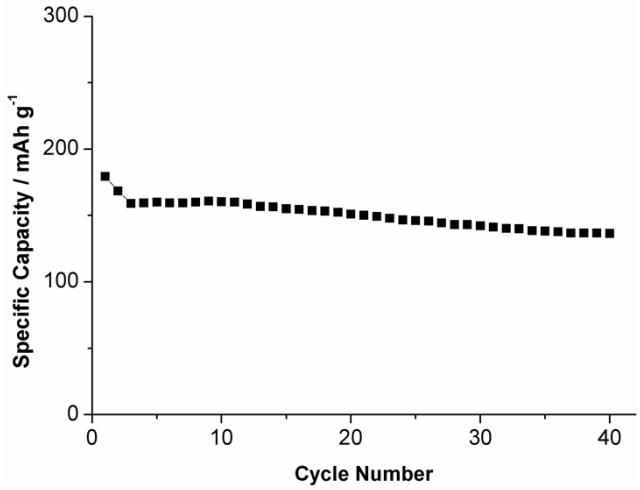


**Fig. S4** Capacity retention of rGO at a rate of 4 C (1 C=1000 mA $g^{-1}$).

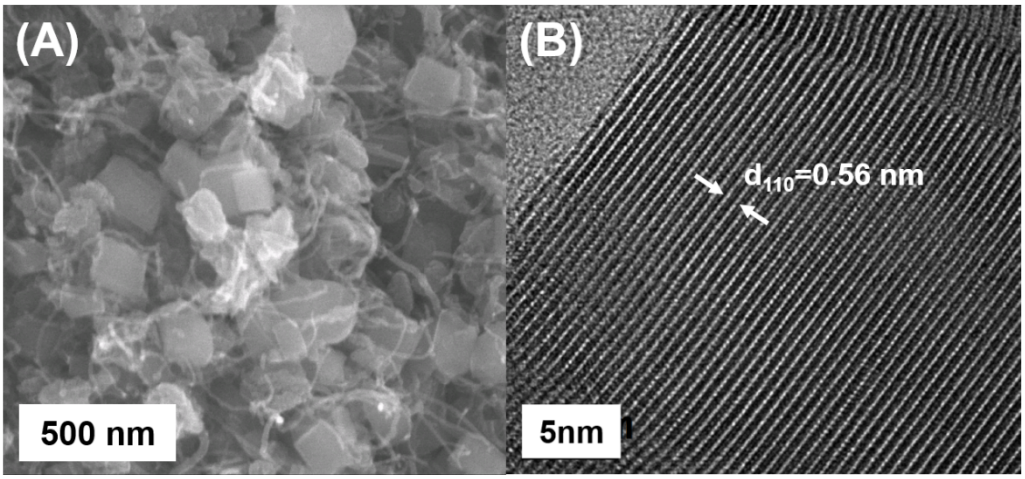


**Fig. S5** (A) SEM and (B) TEM images of the as-prepared $VS_4$-10 wt% CNT composites

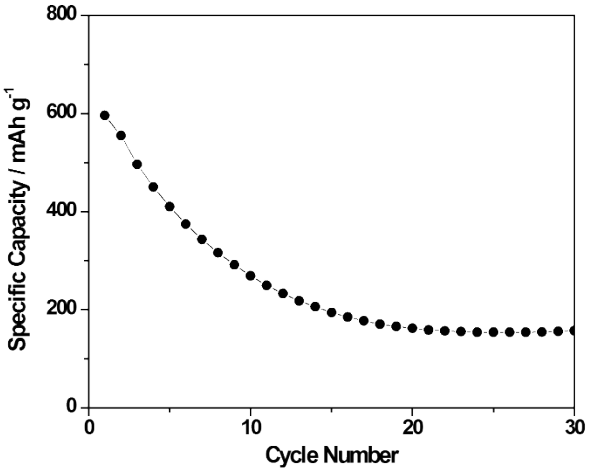


**Fig. S6** Cycle performance of VS4-10 wt% CNT composites at 4C and 23 °C