Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Design and Understanding of Core/Branch Structured VS_2 Nanosheets @ CNT as High-Performance Anode Material for Lithium-Ion Batteries

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1. Estimation of the weight ratio of VS_2 in the composite

According to our SEM images in Figure 1(a-b), each single-walled CNT is surrounded by many thin VS_2 thin flakes, thus the weight ratio of CNT in the composite is quite small and can be neglected. To provide a reference for the weight ratio of VS_2 in the VS_2@CNT composite, we directly grown VS_2 nanosheet on the high purified single walled CNTs (HP-SWCNT, XFNANO Materials Tech Co., Ltd) with the same method. The corresponding thermogravimetric (TGA) curves for pure HP-SWCNT and VS_2@HP-SWCNT are shown in Figure S1. For pure HP-SWCNT, the weight loss between 500-600 is attributed to the decomposition of HP-SWCNT and the residual weight percent is $W_{HP\text{-}SWCNT}$=9.4%. For the VS_2@HP-SWCNT composite, a continuous weight loss observed below

160°C is due to desorption of water molecules on and within the surface layers. The weight loss caused by the oxidation of VS_2 occurred in the temperature range of 160-500°C. The weight loss between 500-800 is attributed to the decomposition of HP-SWCNTs and the oxidation of V^{4+} to relatively greater and stable V^{5+}.[1] The residual weight percent in VS_2@HP-SWCNT is $W_{VS2@HP\text{-}SWCNT}$=75.8%. Therefore, the weight ratio of VS_2 (X_{VS2}) and HP-SWCNT (1- X_{VS2}) can be calculated based on the following equation: [2, 3]

$$\frac{M_{V_2O_5}}{2M_{VS_2}} \times X_{VS_2} + W_{HP-\text{SWCNT}}(1 - X_{VS2}) = W_{VS_2@HP-SWCNT},$$

$M_{V_2O_5}$ and M_{VS_2} are the molecular weight of V_2O_5 and VS_2. The weight ratio of VS_2 in the composite is estimated to be 95.2%.

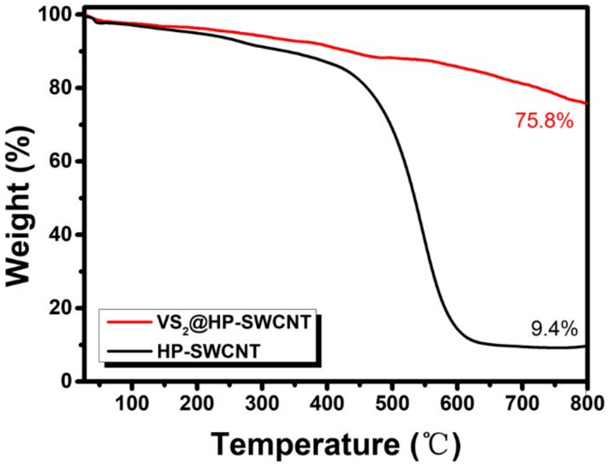

Figure S1. TGA data of (b) pure HP-SWCNT and (c) VS_2@HP-SWCNT.

2. CV curves of the CNT film

To distinguish the contributions of CNT film and VS_2 NS to the acquired CV profiles in Figure 2, a LIB with grown CNT film electrode is also fabricated. Figure S2 (a) shows the comparison of the CV curves of CNT film electrode and VS_2 NS@CNT electrode at scan rate of 0.1 mV/s. Two peaks at 1V and 0.8V with lower intensity are observed during the initial cathodic sweep of the CNT film electrode, while no obvious peaks are observed during the following anodic sweep. Figure S1 (b) shows the CV curves of the CNT film electrode in the first three cycles. It should be noted that no obvious reduction/oxidation peaks are observed in the 2nd and 3rd cycles. Since the CNT films in these two electrode materials are synthesized with the same, the two pairs reduction/oxidation peaks located at 0.7 V/1.7 V and 2.0 V/2.3 V in the VS_2 NS@CNT electrode are resulting from VS_2 NS (Figure 2(a)).

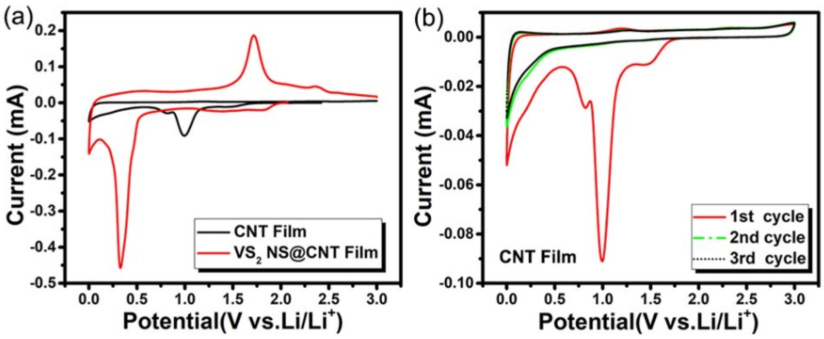

Figure S2. (a) Comparison of the CV profiles of CNT film electrode and VS_2 NS@CNT electrode. (b) First three cycles of the CV profiles of the CNT film electrode.

3. Morphology of VS_2 after the 1st cycling process

As shown in Figure S3, the morphology of VS_2 flakes after the 1st cycling process were broken into several pieces by cracks and more active sites are exposed for the following cycling processes.

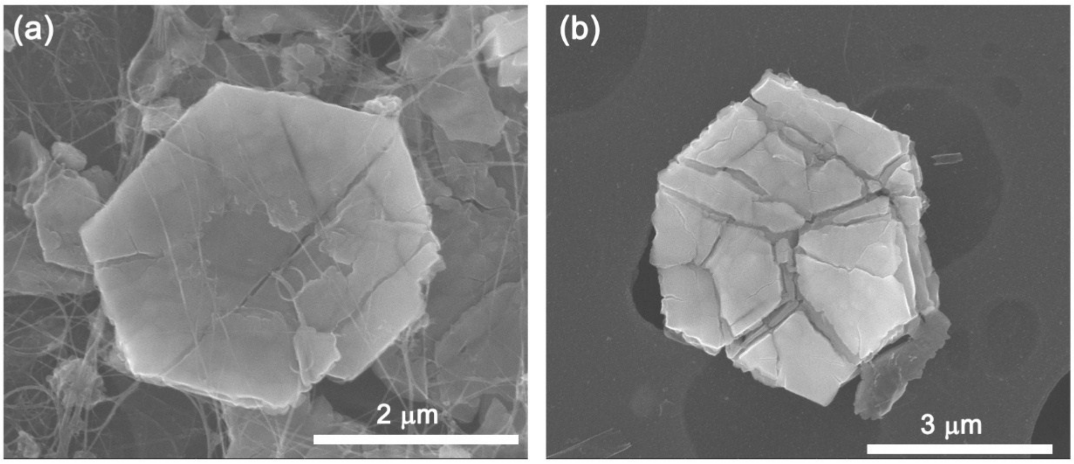

Figure S3. SEM images showing the morphology of VS2 flakes after the 1st cycling process.

4. Morphology Evolution of VS_2 NS@CNT

The morphology evolution of VS_2 NS is *in-situ* investigated by the SEM. Figure S4(a) shows the experimental setup, where the VS_2@CNT, LiO_2 and Li metal act as anode, electrolyte and cathode, respectively. The lithiation process is initiated by applying a -2V voltage on the VS_2@CNT and the current is limited to 100nA. As indicated by the red arrows, volume expansions are clearly observed in the interior region of the VS_2 NS (Figure S4 (b) and (c)).

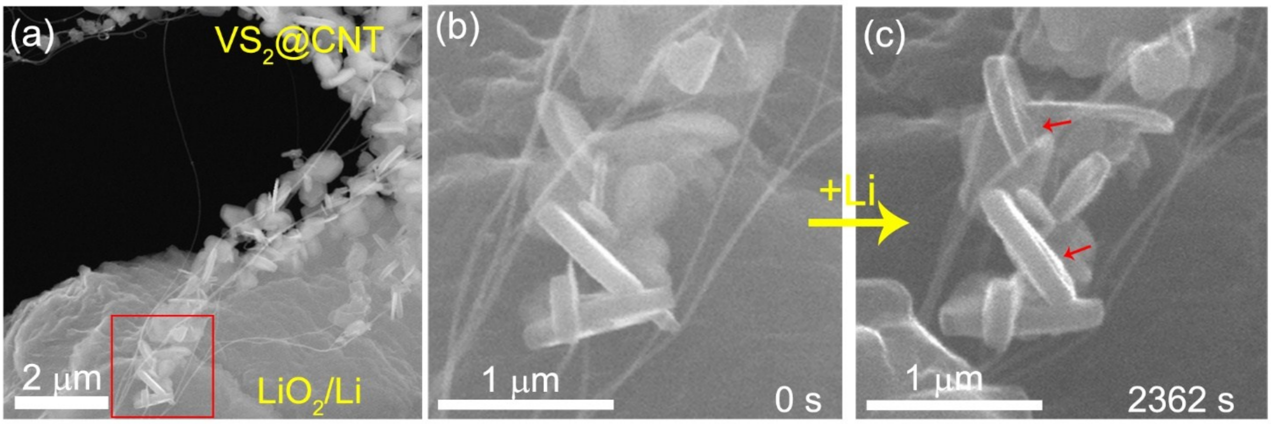

Figure S4. (a) Experimental setup for the in-situ SEM investigation on the discharging/charging processes. SEM images of the red framed region in (a) after (b) 0s and (c) 2362s lithiation process.

5. Microstructural analysis of the lithiated/delithiated VS_2 NS

The FFT patterns of the triangular shaped NS and the crystalline matrix in Figure S5(a) are shown respectively in Figure S5(b) and Figure S5(c). According to our analysis, the triangular shaped NS and the crystalline matrix is indexed to Li_2S.

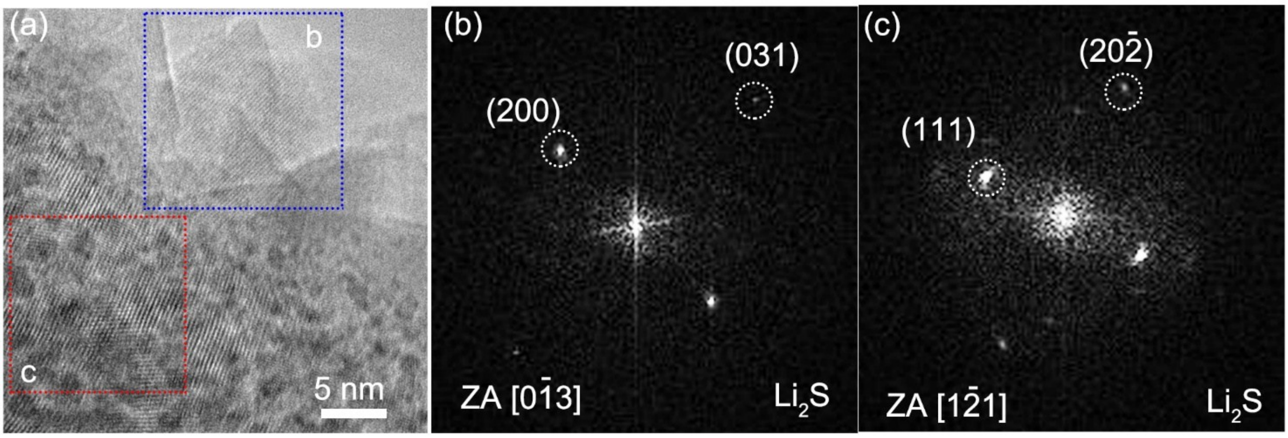

Figure S5. Microstructural analysis of the lithiated VS_2 NS. (a) HRTEM image of the lithiated VS_2. The FFT patterns of the region marked by the blue frame (b) and red frame (c) in (a).

Figure S6 shows the enlarged view of the VS_2 nanosheet (Figure 6) before and after delithiation, from which we can see that the crystalline Li_2S crystal gradually disappear and the particle size of V is decreased during this process.

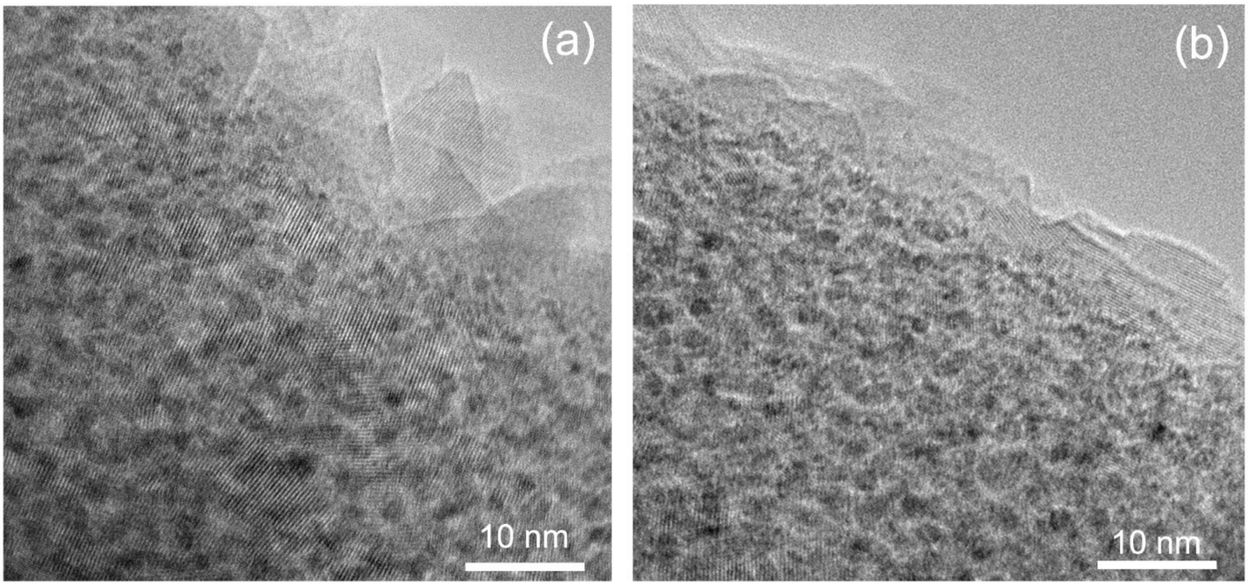

Figure S6. Particle size comparison of V before (a) and after (b) delithiation process.

References:

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