Supporting Information

Synthesis of a novel book-like $K_{0.23}V_2O_5$ and their electrochemical

behavior in lithium batteries

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Fig.S1. TGA of precursor of K_{0.23}V₂O₅

The thermal properties of the obtained $K_{0.23}V_2O_5$ precursor were studied using TG analysis. Fig. S1 displays the TG curve for the $K_{0.23}V_2O_5$ precursor. It shows a first weight loss around 400 °C, which is attributed to the evaporation of residual water and the removal of chemically bound water in the sample. The following slight drop in weight above 600 °C corresponds to the decomposition of of $K_{0.23}V_2O_5$ phase. The weight loss of 1.5 % between 100 and 600°C is very little. This behavior implies that the formation of $K_{0.23}V_2O_5$ phase is completed at this stage. So, the samples were annealed at 400 °C, 450 °C and 500 °C in this work.



Fig. S2. Element distribution mapping images of $K_{0.23}V_2O_5$.

Fig. S 2(a) shows the original SEM image of $K_{0.23}V_2O_5$ without any treatment. The EDS elemental mapping results for all three elements in the same area are shown in Fig. S4 (b, c, d), which indicates the homogeneous distribution of the three elements.



Fig.S3. EIS of $K_{0.23}V_2O_5$ annealed at 500°C cathode before cycle and after 50 cycles(a); EIS of $K_{0.23}V_2O_5$ at 2.45 V of discharge and V_2O_5 at 2.25 V of discharge after 3 cycles(b); the corresponding plots of the real part of impedance (Z') as a function of the inverse square root of the angular frequency (ω -1/2) in the Warburg region (c), (d);

Fig.S3 (a) shows the impedance spectra of the $K_{0.23}V_2O_5$ annealed at 500°C cathode before cycle and discharged at the current 50 mA g⁻¹ from 1.5 to 3.8 V after 50 cycles. Before cycle, the impedance spectra showed obvious impedance characteristic for the reason of the poor electrical conductivity of $K_{0.23}V_2O_5$. However, the impedance of the electrode become much more small after 50 cycles on account of lithiation. Fig.S3 (b) presents samaller EIS of $K_{0.23}V_2O_5$ at 2.45 V of discharge compared with V_2O_5 at 2.25 V of discharge after 3 cycles. The corresponding plots of the real part of impedance (Z') as a function of the inverse square root of the angular frequency (ω -^{1/2}) in the Warburg region are presented in Fig. S 3(c, d).



Fig. S4. The electrochemical performance of V₂O₅

Fig. S4. The electrochemical performance of V_2O_5 prepared by a hydrothermal method with a post-sintering process. (a) CVs at a scan rate of 0.1 mV s⁻¹ showing the first, second and third cycles at a voltage window of 2.0~4.0 V, which is different from the CVs of $K_{0.23}V_2O_5$. The cathodic peaks at 2.61, 3.28 and 3.49 V are clearly observed in the second cycle, indicating the multistep Li⁺ ions intercalation process. In addition, there are three main anodic peaks appearing at the potentials of about 2.25, 3.14 and 3.34 V, corresponding to the Li⁺ ion extraction; (b) Long-term cycling performance at the current density of 50 mA g⁻¹ in the voltage range of 2.0~4.0 V versus Li⁺/Li.The electrode only retains a specific discharge capacity of 80 mA h g⁻¹ after 80 cycles with a capacity retention of 32% comparing with 166 mA h g⁻¹ (68%) of $K_{0.23}V_2O_5$.



Fig. S5. The TEM of $K_{0.23}V_2O_5$ electrode after 50 cycles.

Fig. S5 shows the TEM image of $K_{0.23}V_2O_5$ electrode that was stopped at 3.8V where Li ions were absolutely extrated from $K_{0.23}V_2O_5$ electrode at 1C after 50 cycles, which exhibits that the multilayered structures keep stable after 50 cycles. As shown in Fig. S6 (a, b), multilayered structures are still maintained at 1C after 50 cycles. The corresponding electron diffraction(c) and HRTEM image (d) are almost same with of which showed in Figure. 2 (e, f), indicating good structural stability during charging/discharging.



Fig. S6. XPS spectra for the $K_{0.23}V_2O_5$ electrode before and after 50 cycles under 1 C

Fig. S6 (a, b) shows full spectra the $K_{0.23}V_2O_5$ electrode stopped at 3.8V where Li ions was absolutely extracted before and after 50 cycles under 1 C, and the peaks of $V2p_{3/2}$, O-1s, K2p3 and C1s could be found, impling that the V and K ions kept stable; Fig. S6 (c, d) shows the high resolution XPS core level spectra of $V2p_{3/2}$ of $K_{0.23}V_2O_5$ electrode before and after charge– discharge measurement. For a freshly prepared electrode, the peak at 530.5 eV in the O-1s region is attributable to lattice oxygen (O–V) in $K_{0.23}V_2O_5$.¹ The peak for $V2p_{3/2}$ at 517.2 eV can be indexed to the V⁵⁺, and the shoulder peak at lower energy of 516.1 eV confirms the existence of the V⁴⁺.^{2,3} After cycling at 1C for 50 cycles, the $K_{0.23}V_2O_5$ electrode was carefully disassembled and washed by EC/DEC pure solution (EC/DEC =1 : 1 v/v) in an Ar-filled glove box, and then detected by XPS spectra. As can be seen, the V-2p3/2 region does not show any difference after electrochemical measurement. The existence of V⁵⁺ (517.2 eV) and V⁴⁺ (516.1 eV) after charge– discharge measurement demonstrates that the crystallographic structure is maintained unchanged. In addtion, the amount of kalium element is similar from its initial value to its final value, impling good structural stability during charging/discharging.

References

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