

ELECTRONIC SUPPLEMENTAL INFORMATION

Chemical and Electrochemical Lithiation of Van Der Waals Oxytelluride V_2Te_2O

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Neutron refinements

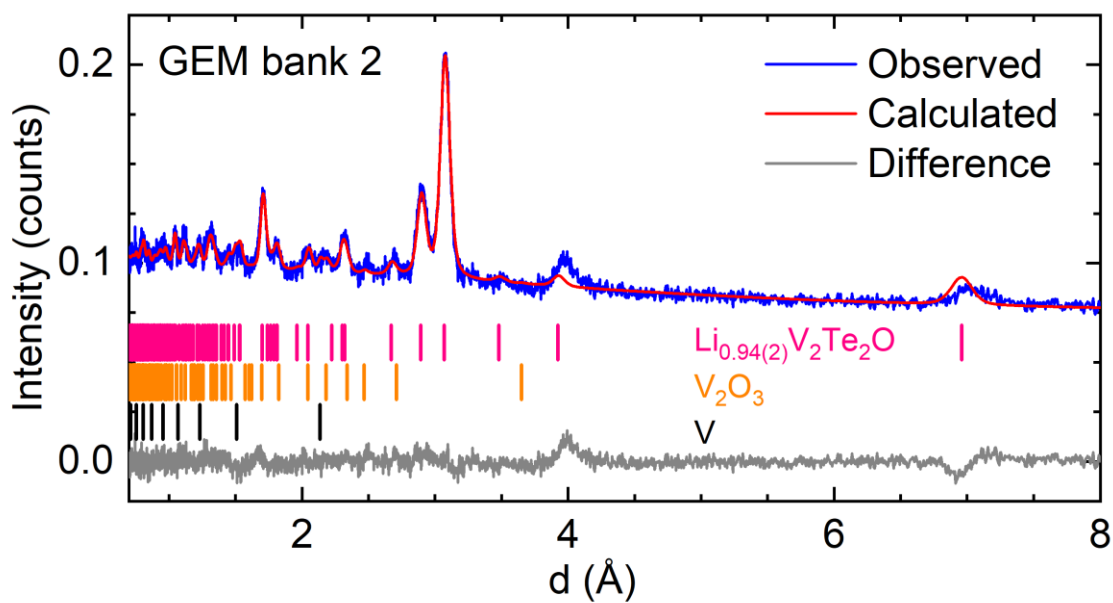


Figure S1: Rietveld refinement of $\text{Li}_{0.94}\text{V}_2\text{Te}_2\text{O}$ against GEM bank 2 data. Blue – experimental data; red – calculated intensities; grey – difference pattern; pink tick marks – Bragg reflection positions for $\text{Li}_{0.94}\text{V}_2\text{Te}_2\text{O}$ (pink), V_2O_3 (orange) and V (black).

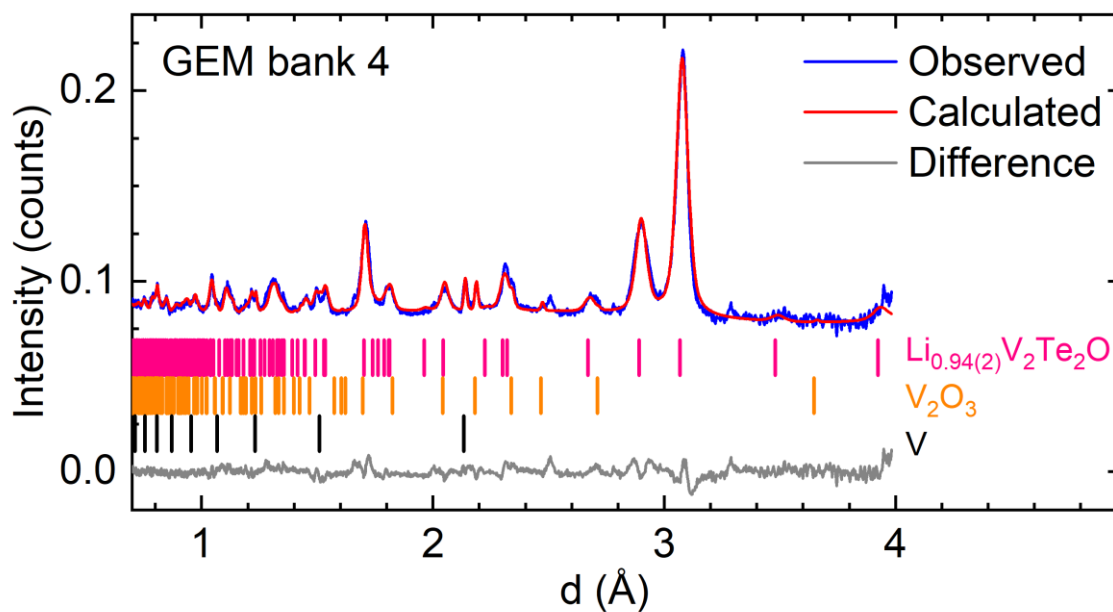


Figure S2: Rietveld refinement of $\text{Li}_{0.94}\text{V}_2\text{Te}_2\text{O}$ against GEM bank 4 data. Blue – experimental data; red – calculated intensities; grey – difference pattern; pink tick marks – Bragg reflection positions for $\text{Li}_{0.94}\text{V}_2\text{Te}_2\text{O}$ (pink), V_2O_3 (orange) and V (black).

Impurity phase observed by PXRD

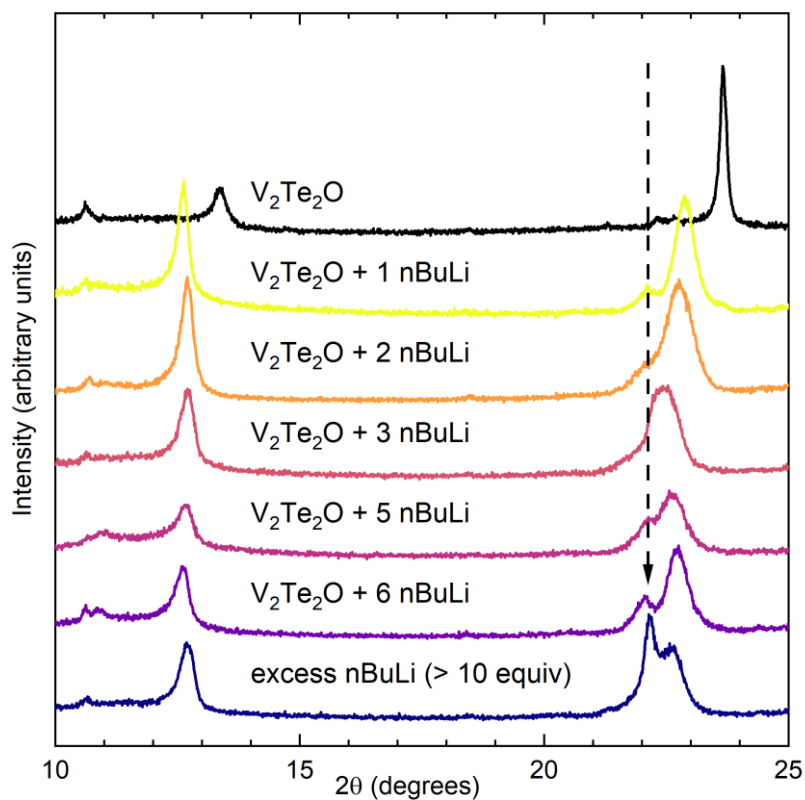


Figure S3: Laboratory PXRD patterns for intercalated samples, showing the increasing amounts of an unindexed secondary phase (peak at $d \sim 4 \text{ \AA}$) with increasing amounts of nBuLi.

Air exposure PXRD

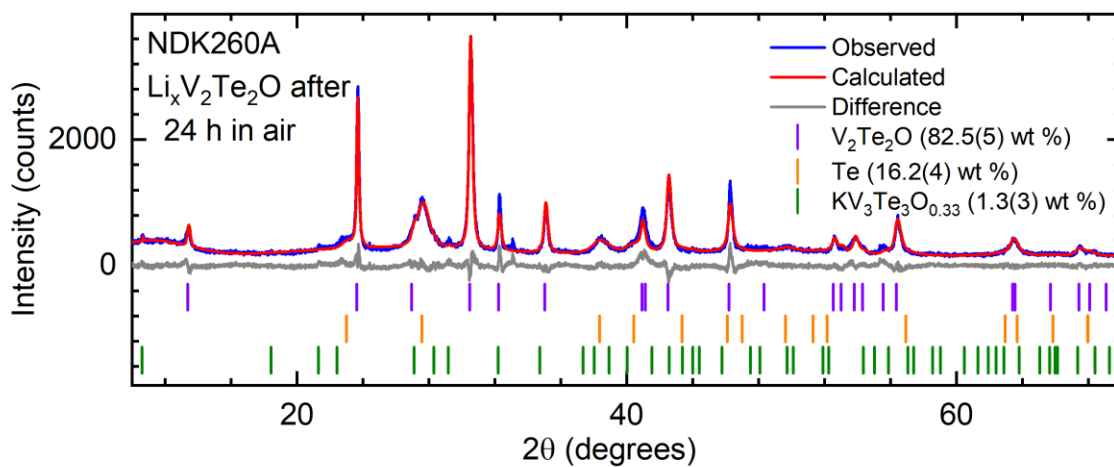


Figure S4: Rietveld refinement of laboratory PXRD data for $\text{Li}_{0.94}\text{V}_2\text{Te}_2\text{O}$, 24 h after exposure to moist air. The sample had fully decomposed into $\text{V}_2\text{Te}_2\text{O}$ + Te.

Water reaction PXRD

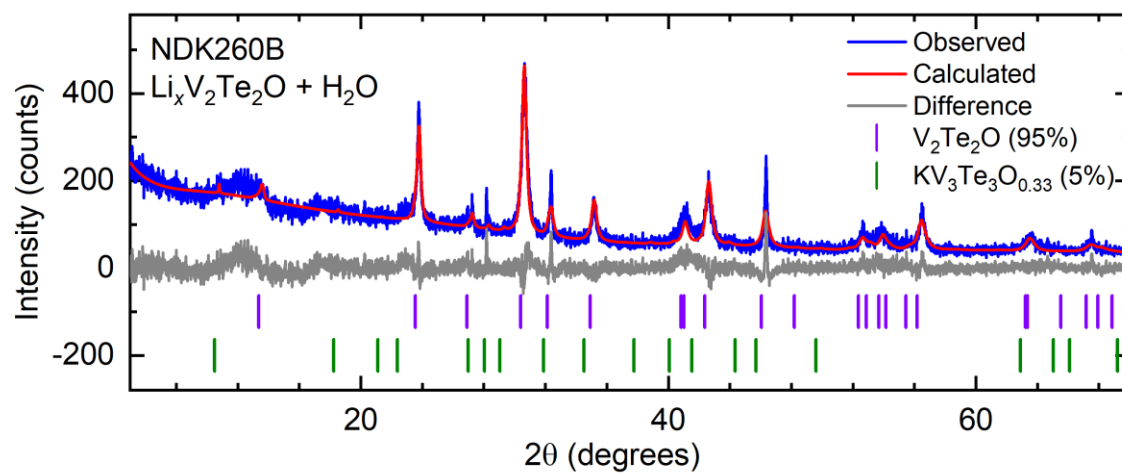


Figure S5: Rietveld refinement of laboratory PXRD data for the product of the reaction $\text{Li}_{0.94}\text{V}_2\text{Te}_2\text{O} + \text{H}_2\text{O}$ (under inert atmosphere).

Magnetometry

Magnetic measurements were made using a Quantum Design MPMS-XL or MPMS-3 SQUID magnetometer. A powder sample of mass 20–30 mg (accurately measured in each case) was placed in a gelatine capsule inside a plastic straw as the sample holder. The susceptibility $\chi \approx M/H$ was determined by measuring the magnetic moment (M) on warming from 5–350 K in an applied field (H) of 100 Oe after cooling in the applied field (FC) or in zero field (ZFC). A magnetisation hysteresis loop was measured as a function of applied field in the range ± 5 T at temperatures of 5 and 300 K.

The susceptibility curves for the lithiated samples (Figure S6) increase sharply below $T_c \sim 40$ K, indicating ferromagnetic (FM) interactions. The $M(H)$ data at low temperature show soft ferromagnetism with a very small hysteresis. However, for several reasons we believe that this may originate from the unidentified impurity phase with its main diffraction peak at $d \sim 4$ Å. The maximum magnetisation (at $T \sim 30$ K) is only $5 \times 10^{-4} \mu_B$ per formula unit of $\text{Li}_x\text{V}_2\text{Te}_2\text{O}$ and the susceptibility drops again below 30 K rather than continuing to increase upon cooling, which suggests that the FM fraction of the total sample is small. Furthermore, the onset temperature of the transition is identical across samples made using different amounts of nBuLi, whereas different values of x in $\text{Li}_x\text{V}_2\text{Te}_2\text{O}$ would be expected to produce different electron counts and thus different T_c 's. At 100–350 K the susceptibility slowly decreases in a non-Curie-Weiss fashion and the absolute values of the magnetisation are of the same order as powder samples of the parent material $\text{V}_2\text{Te}_2\text{O}$. The behaviour is consistent with Pauli paramagnetic behaviour across the compositional range, as per the parent $\text{V}_2\text{Te}_2\text{O}$ [1].

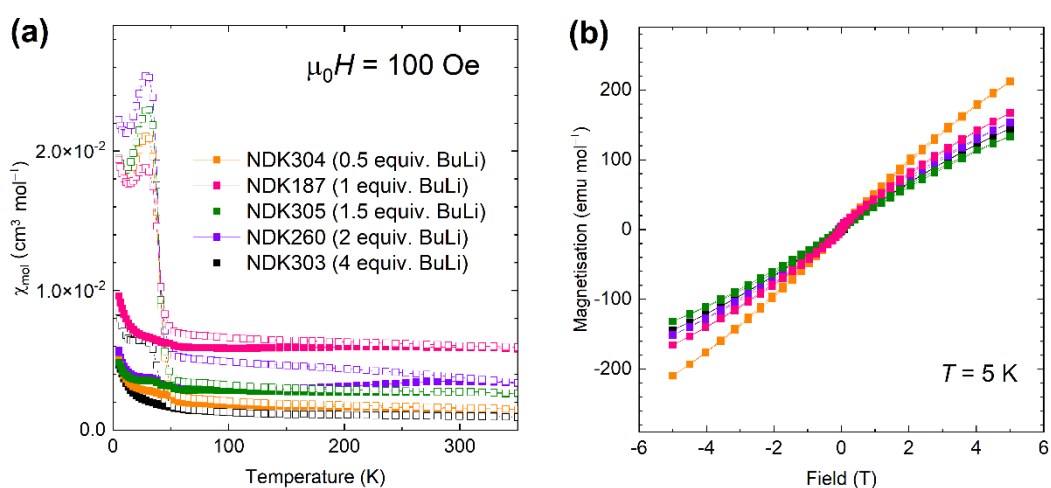


Figure S6: Magnetic data for $\text{Li}_x\text{V}_2\text{Te}_2\text{O}$ samples. (a) Magnetic susceptibility at 100 Oe measured in the ZFC (filled symbols) and FC (open symbols) regimes. (b) Hysteresis loop at 5 K.

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- [1] A. Ablimit *et al.*, “ $\text{V}_2\text{Te}_2\text{O}$: A Two-Dimensional van der Waals Correlated Metal,” *Inorg. Chem.*, vol. 57, no. 23, pp. 14617–14623, 2018.