

## Department of Materials Science and Engineering,

Master's Thesis Defense

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## Exfoliation of Bilayered Vanadium Oxide and Oxide/MXene Heterostructure Assembly for Energy Storage

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Bilayered vanadium oxide  $(\delta - V_2O_5 \cdot nH_2O$  or BVO) synthesized via a sol-gel process is an attractive candidate to be used as a cathode material for Li-ion batteries (LIBs) due to its expanded interlayer spacing, and high theoretical capacity. However, poor electrical conductivity and rapid capacity fading present challenges for achieving high-rate performance and good cycle life in LIBs. A viable solution to mitigating these drawbacks is to fabricate two-dimensional (2D) heterostructures comprised of the alternating BVO and titanium carbide  $(Ti_3C_2T_x)$  MXene layers. First, we demonstrate a simple liquid phase exfoliation of bulk  $\delta$ -Li<sub>x</sub>V<sub>2</sub>O<sub>5</sub>·*n*H<sub>2</sub>O (LVO) powder to obtain a stable colloidal suspension of LVO nanoflakes. Due to the hydrated nature of LVO, we also highlight the importance of controlling interlayer water content with vacuum drying for achieving better cycling stability. The 200°C vacuum dried LVO nanoflake cathode delivered an initial ion storage capacity of 212 mAh g<sup>-1</sup> which was 32% higher than the sample dried at 105°C under vacuum and retained a higher capacity retention during cycling at constant and increasing current densities. Subsequently, electrostatic assembly of LVO and  $Ti_3C_2T_x$  nanoflakes was facilitated by the introduction of cationic species into the mixed suspensions. Through electrochemical testing, the 9:1 weight ratio of LVO: $Ti_3C_2T_x$  cathodes delivered the highest ion storage capacity of 167 mAh  $g^{-1}$  and enabled greater tolerance to high current densities with a capacity retention of ~90% after cycling through increasing current densities. Moreover, 4:1 weight ratio LVO:Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> delivers a capacity of 248.4 mAh g<sup>-1</sup> from a potential window of 1-4V, demonstrating that higher capacities can be achieved at lower potentials where Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> exhibits redox activity. These results demonstrate an environmentally friendly and safe approach to obtaining 2D LVO nanoflakes and offers pathways to constructing novel vanadium oxide based 2D heterostructures for improving electrochemical performance in energy storage devices.