

Reversible Intercalation of Mg^{2+} in V_2O_5 at Elevated Temperatures Leads to Enhanced Electrochemical Performances

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Barriers to lithium ion battery development, such as the limited energy density of Li-ion batteries and the scarcity of Li, have led to great interest in exploring multi-valent ionic batteries containing earth abundant intercalation elements, such as magnesium or calcium [1]. Magnesium-ion batteries have the potential of becoming the next stable, rechargeable, and high-energy-density batteries that can rise above the limitations of current Li-ion batteries. Reversible intercalation of Mg^{2+} ions is a crucial part in constructing a stable battery, which, when combined with high capacity, charts a promising pathway towards high performance and future designs of Mg-ion batteries. High levels of reversible Mg^{2+} intercalation have been reported in $\alpha\text{-V}_2\text{O}_5$ by cycling assembled cells at an elevated temperature of 110°C , the benefit of which is retained at room temperature [2].

Experiments were carried out with assembled cells consisted of pressed V_2O_5 powders as the cathode, ionic liquid $\text{MgTFSI}_2\text{-PY14TFSI}$ as the electrolyte, and activated carbon cloths or Mg foil as the anode. Electrochemical experiments were conducted within a potential range that the ionic liquid is stable, and all cells were fabricated in a glove box filled with Ar gas. After cycling at room temperature and 110°C , the samples were characterized structurally with transmission electron microscopy (TEM, JEM 3010, JEOL) and scanning transmission electron microscopy (STEM, JEM-ARM200CF, JEOL) equipped with a Cs corrector and using a cold field-emission source, among other techniques including composition analysis using a Scanning transmission X-ray microscopy (STXM) combined with X-ray absorption spectroscopy (XAS). Current research is ongoing to explore in-situ characterization techniques, such as TEM or STEM combined with capabilities of simultaneous heating and electrochemical cycling. If realized, this can open up more possibilities to deepen the understanding of structural and chemical developments during the intercalation process.

The ex-situ experiments demonstrate that $\alpha\text{-V}_2\text{O}_5$ can intercalate at least one mole of Mg^{2+} reversibly at 110°C , as opposed to <0.6 mole at 25°C . The capacity of the $\alpha\text{-V}_2\text{O}_5$ is increased 20-fold, from 16 mAh g^{-1} at 25°C to 295 mAh g^{-1} at 110°C , matching the performance of Li-ion batteries, as shown in Figure 1. This increase in electrochemical performance is accompanied by structural changes, as observed by STEM in Figure 2. The morphology of the $\alpha\text{-V}_2\text{O}_5$ powders drastically change from platelets to delaminated layers. Particles with delaminated morphologies show the change in Mg content, which echoes with the change in capacity, calculated to be 1 mol Mg^{2+} per mole $\alpha\text{-V}_2\text{O}_5$. These findings pave the road toward reversible Mg-ion batteries with high degrees of Mg^{2+} intercalation and high potential, and current efforts in developing in-situ characterization functionalities will lead to imperative capabilities [3].

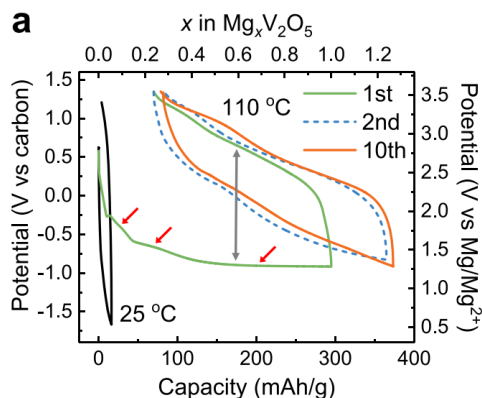


Figure 1: The voltage profiles from the electrochemical cycling of α - V_2O_5 in an ionic liquid Mg^{2+} electrolyte at 25 and 110 °C, showing the increased capacitance with the grey arrow, and reversibility between the 2nd and 10th cycles [2].

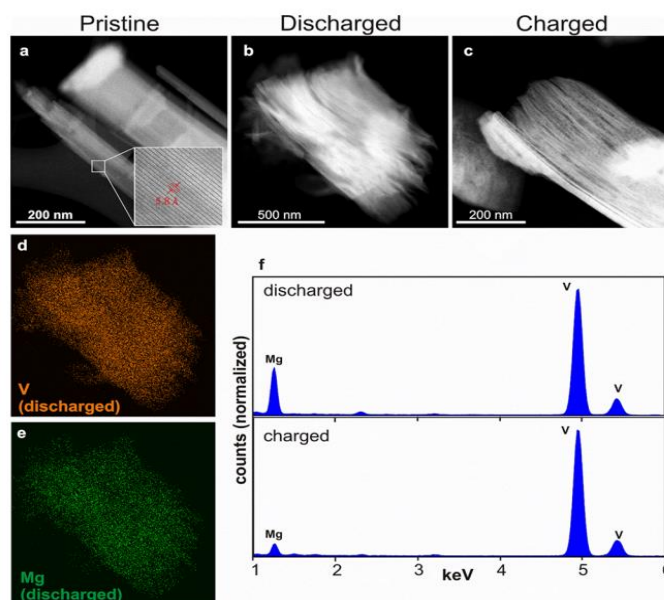


Figure 2: Morphological and compositional changes upon electrochemical cycling. High angle annular dark field images of pristine (a), discharged (b), and recharged samples (c). Elemental mapping of discharged particles for V (d) and Mg (e). (f) EDS spectra of discharged (top) and recharged (bottom) samples. [2]

References:

- [1] J Muldoon et al., *Chem. Rev.* **114** (2014), p. 11683. DOI: 10.1021/cr500049y
 [2] HD Yoo et al., *ACS Energy Lett.* **4**(7) (2019), p. 1528. DOI: 10.1021/acsenerylett.9b00788
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