

Development of High-Energy Cathodes via in-situ Solvothermal Synthesis

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There is tremendous interest in developing low-temperature solvothermal methods for synthesizing high-energy battery electrodes at reduced cost. One of the key hurdles in using solution-based reactions is the difficulty in understanding the reaction pathway and thereby optimizing the reaction for obtaining desired phases and material properties. The ability to probe synthesis reactions in *real time* would reveal which intermediate phase form and when, and provide a better understanding of how temperature, pressure, time and the initial concentrations affect the reaction pathways. Ultimately, a better understanding of the correlation between synthesis conditions, crystallization processes, defect structure and properties would lead to rational design of advanced electrode materials. To this end, we developed specialized in situ reactors for investigating solvothermal synthesis reactions in *real-time* using x-ray diffraction (XRD).

Figure 1 shows the phase transformations that occurred during the hydrothermal synthesis of $\text{Cu}_x\text{V}_2\text{O}_5$ ($x < 1$). At the onset of the reaction the primary peaks are associated with the V_2O_5 phase. As the temperature increased to 180°C the V_2O_5 diffraction peaks disappeared and weak peaks from the $\text{Cu}_x\text{V}_2\text{O}_5$ phase emerged indicating a dissolution-recrystallization reaction. The $\text{Cu}_x\text{V}_2\text{O}_5$ phase grew steadily with time until the reaction was stopped after approximately 17 hours. By refinement, we found that the final product consisted of a Cu deficient phase of $\text{Cu}_{0.36}\text{V}_2\text{O}_5$, which is isostructural with V_2O_5 . There was no evidence of an intermediate phase in this reaction, but intermediate phases were observed with different precursors and under different reaction conditions.

In this presentation, we will show our recent results on solvothermal synthesis of a series of high capacity $\text{Cu}_x\text{V}_2\text{O}_5$ cathodes, and discuss the dependence of the reaction pathway, and morphology, structure of the final product on reaction conditions (precursors, reducing agent, temperature, and time). The lithium reaction mechanisms and the fundamental limits of the electrochemical performance of the $\text{Cu}_x\text{V}_2\text{O}_5$ cathodes (Figure 2) will be also discussed based on the electrochemical measurements and structural characterizations of the as-synthesized and lithiated materials.

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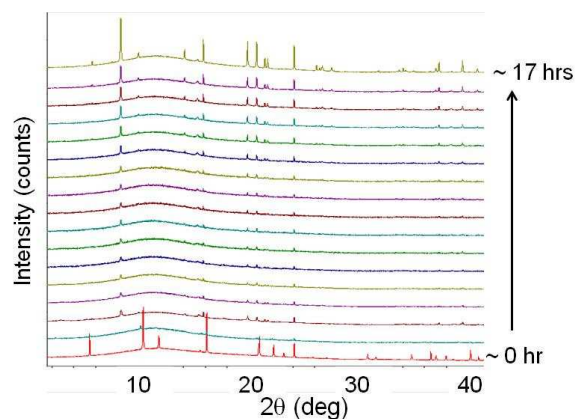


Figure 1. Time-resolved X-ray diffraction patterns showing the transformation from the precursor V_2O_5 to $\text{Cu}_x\text{V}_2\text{O}_5$ via a dissolution-recrystallization reaction process.

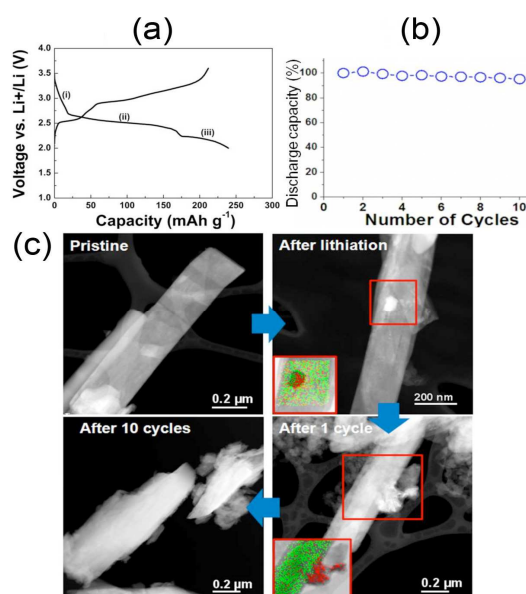


Figure 2. (a) Charge-discharge profiles for the first cycle, (b) cycalibity and (c) Scanning transmission electron microscopy (TEM)-energy-dispersive x-ray spectroscopy (EDS) images of the pristine, lithiated and cycled electrodes showing the extruded Cu identified by EDS mapping (red).