Li/Ag2VO2PO4 batteries: the roles of composite electrode constituents on electrochemistry.

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The primary battery cathode material silver vanadium phosphorous oxide, Ag<sub>2</sub>VO<sub>2</sub>PO<sub>4</sub>, was utilized as a model system to systematically study the impact of the constituents of a composite electrode, including polymeric and conductive additives, on electrochem. Notably, although highly resistive, this bimetallic cathode can be discharged as a pure electroactive material in the absence of a conductive additive as it generates an in situ conductive matrix via a reduction displacement reaction resulting in the formation of silver metal nanoparticles. Three different electrode compositions were investigated: Ag<sub>2</sub>VO<sub>2</sub>PO<sub>4</sub> only, Ag<sub>2</sub>VO<sub>2</sub>PO<sub>4</sub> with binder, and Ag<sub>2</sub>VO<sub>2</sub>PO<sub>4</sub> with binder and carbon. Constant current discharge, pulse testing and impedance spectroscopy measurements were used to characterize the electrochemical properties of the electrodes as a function of depth of discharge. In situ EDXRD was used to spatially resolve the discharge progression within the cathode by following the formation of Ag<sup>0</sup>. Ex situ XRD and EXAFS modeling were used to quantify the amount of Ag<sup>0</sup> formed. Results indicate that the metal center reduced ( $V^{5+}$  or Ag<sup>+</sup>) was highly dependent on composite composition (presence of PTFE, carbon), depth of discharge (Ag<sup>0</sup> nanoparticle formation), and spatial location within the cathode. The addition of a binder was found to increase cell polarization, and the percolation network provided by the carbon in the presence of PTFE was further increased with reduction and formation of Ag<sup>0</sup>. This study provides insight into the factors controlling the electrochemistry of resistive active materials in composite electrodes.