High-rate Operando X-ray Studies of Lithium-ion Battery Materials

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The maximum power output and minimum charging time of a lithium-ion battery – key parameters for its use in, for example, transportation applications – depend on mixed ionic–electronic diffusion. While the discharge/charge rate and capacity can be tuned by varying the composite electrode structure, ionic transport within the active particles represents a fundamental limitation. Thus, to achieve high rates, particles are frequently reduced to nanosize dimensions despite this being disadvantageous in terms of volumetric packing density as well as cost, stability, and sustainability considerations. As an alternative to nanoscaling, we show that complex niobium tungsten oxides with topologically frustrated polyhedral arrangements and dense μm-scale particle morphologies can rapidly and reversibly intercalate large quantities of lithium. Multielectron redox, buffered volume expansion, and extremely fast lithium transport approaching that of a liquid lead to extremely high volumetric capacities and rate performance as very recently reported in both crystallographic shear structure and bronze-like niobium tungsten oxides[1]. The active materials Nb16W5O55 and Nb18W16O93 offer new strategies toward designing electrodes with advantages in energy density, scalability, electrode architecture/complexity and cost as alternatives to the state-of-the-art high-rate anode material Li4Ti5O12.

Characterisation of the complex material evolution will be presented, focusing on insights from operando X-ray diffraction (figure) performed during high-rate battery operation with cycles lasting only a few minutes. Appropriate time resolution to capture the numerous stages of phase evolution was achieved with a synchrotron X-ray source and an area detector, allowing individual patterns to be recorded in seconds. Materials and mechanisms that enable lithiation of μm particles in minutes have implications for high power applications, fast charging devices, all-solid-state batteries, and general approaches to electrode design and material discovery.