In situ imaging of Li-ion transport in Fe₃O₄ battery electrodes

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Li-ion batteries are used for energy storage worldwide in many different applications including cars, portable devices (such as cell phones and laptops), and the electric grid. In order to improve on battery capacity and energy density, it is necessary to understand the local chemical processes that take place inside the battery [1]. Nanoparticles have received considerable attention for use in battery electrodes, as their high surface area to volume ratio improves electrochemical properties. However, the nanoparticles are known to form micrometer-sized aggregates and the transport of Li ions inside these aggregates is not understood. We use X-ray imaging and spectroscopy to elucidate this process.

We study a cathode consisting of aggregates of magnetite (Fe₃O₄) nanoparticles to investigate Li transport and (de)intercalation during battery (dis)charge. We use bulk X-ray absorption spectroscopy (XAS) and transmission X-ray microscopy (TXM) combined with X-ray absorption near edge structure (XANES) at beamlines 4-1 and 6-2c at SSRL, respectively, to investigate magnetite nanoparticle aggregates in the cathode as the battery is cycled. We will also perform *ex situ* experiments to complement the *in situ* results. Together, this will enable us to investigate the validity of candidate models that have been suggested for Li transport [2]: In one case, *intra* particle Li transport is predicted to be fast and nanoparticles should thus be found either in a pure (fully lithiated or delithiated) state. In the other case, faster *inter* particle transport results in some particles being imaged in a mixed oxidation state. *In situ* high-resolution spectromicroscopy is required to determine which model is right, and to measure correct rate constants. Answering these questions could help design batteries with higher energy density and faster charging rates.

- 1. Palacín, M. R. Chem. Soc. Rev. 38, 2565-2575 (2009).
- 2. Knehr, K. W. et al. J. Electrochem. Soc. 162, A2817-A2826 (2015).