

Optimization of in-situ 3D electron diffraction in an electrochemical liquid cell for crystal structure determination of energy materials

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Over the last decades, the increased environmental pollution and vast fossil consumption generated a need for renewable energy sources. As these renewable energy sources are not always available, this also needs next-generation energy storage devices, such as lithium-ion batteries and solid oxide fuel cells. Despite the great interest in these systems, there are still gaps in the knowledge about the crystal structure evolution and phase transitions of these energy materials during the electrochemical reactions due to the submicron size of the active particles, which impede single crystal diffraction with X-rays or neutrons. Filling these gaps is crucial for understanding why a particular material functions better or worse than other closely related materials.

3D electron diffraction can be applied to submicron sized single crystals and is a powerful tool for determining the crystal structure and studying the structural changes during the electrochemical reaction [1]. However, *ex situ* experiments are not sufficient to solve all the questions and leave room for misinterpretation of artefacts due to, for instance, air and vacuum exposure and relaxation between cycling and structure determination and inherent differences between different crystals. Therefore, we aim to apply *in situ* 3D electron diffraction in a liquid filled electrochemical cell to study the crystal structure evolution upon electrochemical cycling in the transmission electron microscope.

Whereas our group was able to obtain *in situ* 3DED data of charged particles after a single cycle [2], *in situ* observation of ongoing reactions with electron diffraction has not been realized yet. One challenge is the strong scattering of the electrons by the thick liquid layer, which significantly decreases the signal-to-noise ratio [3, 4]. For obtaining data after a single cycle, this thick layer of liquid can be partially evaporated using an intense electron beam [3]. However, this procedure leaves contamination behind and prevents further cycling.

Our study aims to perform *in situ* 3D electron diffraction at different stages of the electrochemical process within the same experiment and therefore, without the need for evaporating part of the liquid. Our preliminary experiments on gold nanoparticles established this possibility. However, gold is an ideal system because of its high atomic number and the possibility to introduce the particles into the electrochemical cell by flushing. Studying complex and lower atomic number compounds of which the particles cannot be flushed through the cell will require optimization of the experimental conditions. Controlling all the parameters during the experiments, such as particle deposition, liquid thickness, bulging of the windows, beam irradiation and flow rate, is challenging. In this presentation, I will discuss the hurdles, the solutions and the results I have obtained so far.

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