Experimental report

02/12/2021

Proposal: CRG-2831 **Council:** 10/2020

Title: Insight into the evolution of the Solid Electrolyte Interphase on Li-metal formed with a Highly concentrated Electrolyte

Research area: Materials

This proposal is a new proposal

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Samples: 4MLiTFSI in DME
2M LiTFSI in DME
LP30 (1MLiPF6 in EC/DMC)

Instrument	Requested days	Allocated days	From	To
SUPERADAM	4	3	06/06/2021	09/06/2021

Abstract:

Metallic Lithium (Li) is the anode material with the highest theoretical specific capacity (3860 mAh/g) for future Li-batteries. Unfortunately, its practical application is severely hindered by uncontrolled growth of Li dendrites and low Coulombic efficiency which mainly results from the unstable solid electrolyte interphase (SEI) formed on the Li-metal anode. Recently, we have reported that highly concentrated electrolytes can enable ultrahigh Coulombic efficiency and cycling stability of Li-metal anodes. With this proposal, we aim to perform in-situ neutron reflectometry (NR) using a custom-built electrochemical cell to directly track the evolution of electrode/electrolyte interface in order to reveal the mechanism behind the extraordinary performance of the high concentrated electrolytes. With neutron reflectometry we can directly probe the evolution of the interfacial structure which will provide new insight on the stability of SEI on Li-metal electrodes and open new routes to rationally design electrolytes for high-energy-density battery systems.

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Local contact: Alexei Vorobiev
Instrument: SuperADAM

In this experiment we performed neutron reflectometry measurements in a custom made electrochemical cell, see figure 1. The neutron beam entered the cell through the side of a Si-block to probe the interphase between the electrolyte and a Cu-electrode. When a potential that is negative enough is applied across this cell, the electrolyte will decompose to form a surface film on the electrode, called a solid electrolyte interphase (SEI). The goal of the experiment was to investigate how this SEI-layer changed when electrolytes with different salt concentrations were used in the cell.

The electrodes were prepared using magnetron sputtering. A 5 nm Ti binding layer was first deposited on the Si-blocks and subsequently a 50 nm Cu electrode layer was deposited. Initial measurements on the sputtered Si-blocks in air showed Cu-fringes over a large q-range, showing a low roughness and high uniformity of the Cu electrode, see figure 2. Furthermore, the reflectivity curves of both substrates used in the experiment were overlapping, indicating that the layer thicknesses and roughnesses were very similar.

During the measurements, the cells in figure 1 were filled with electrolytes containing a mixture of the salt LiFSI (Lithium bis(fluorosulfonyl)imide) and the solvent DME (1,2-Dimethoxyethane) in different ratios. To optimize the contrast for the SEI-layer, a deuterated solvent was used. This gave a scattering length density (SLD) of the electrolyte which was closer to Cu than for a non-deuterated electrolyte which improves the contrast for the SEI layer formed when a potential is applied. Measurements for electrolytes with 1M and 4M salt concentration were carried out and in both cases, a clear change of the reflectivity curves was seen after a potential of +50 mV vs Li/Li^+ had been applied to the Cu-electrode, see figure 3. This shows that the formation of an SEI-layer can be detected in our experiment.

To extract information on the physical characteristics of the SEI the reflectivity curves will be analysed with different models and the results will be correlated to electrochemical measurements as well as post mortem x-ray photoelectron spectroscopy data.

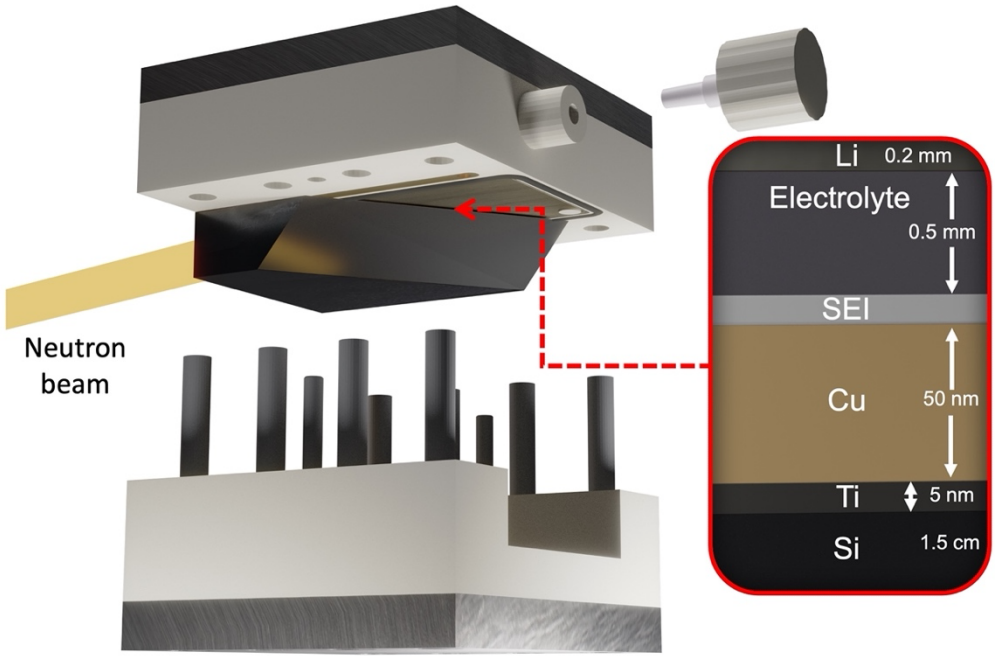

Figure 1. Illustration of the electrochemical cell used for the reflectivity measurements. The neutron beam enters through the back of a 8x5x1.5cm Si-block which was sputtered with Ti and Cu layers. A potential was applied with respect to a Li-counter electrode to form the SEI-layer by electrolyte decomposition.

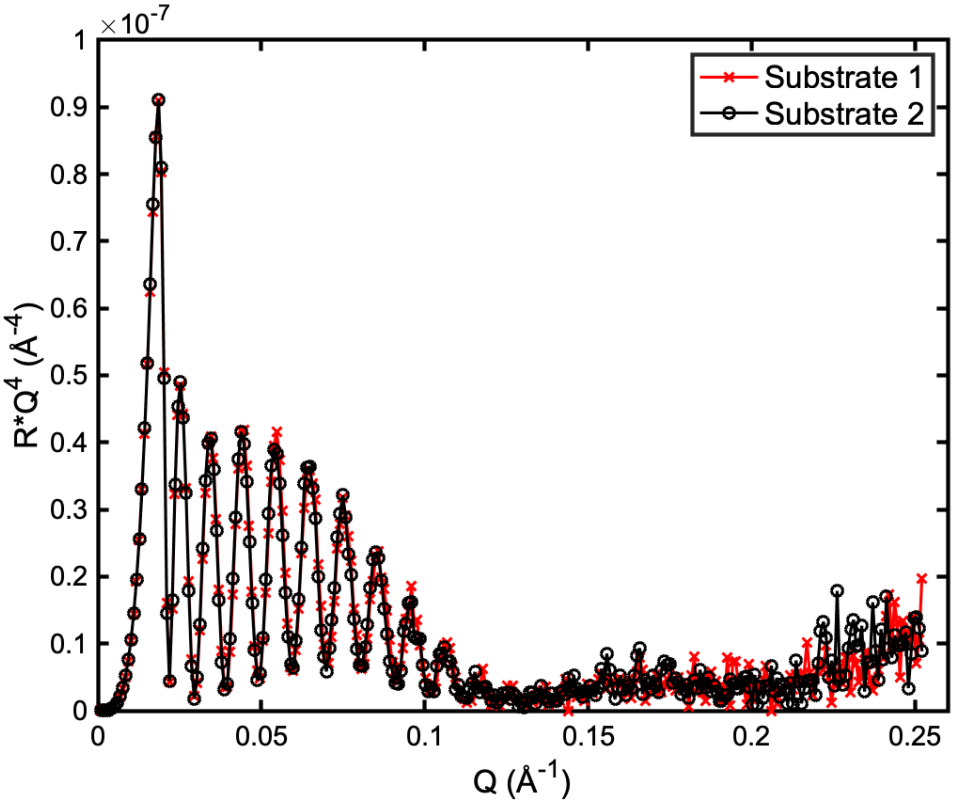

Figure 2. Reflectivity profiles of the sputtered substrates, measured in air.

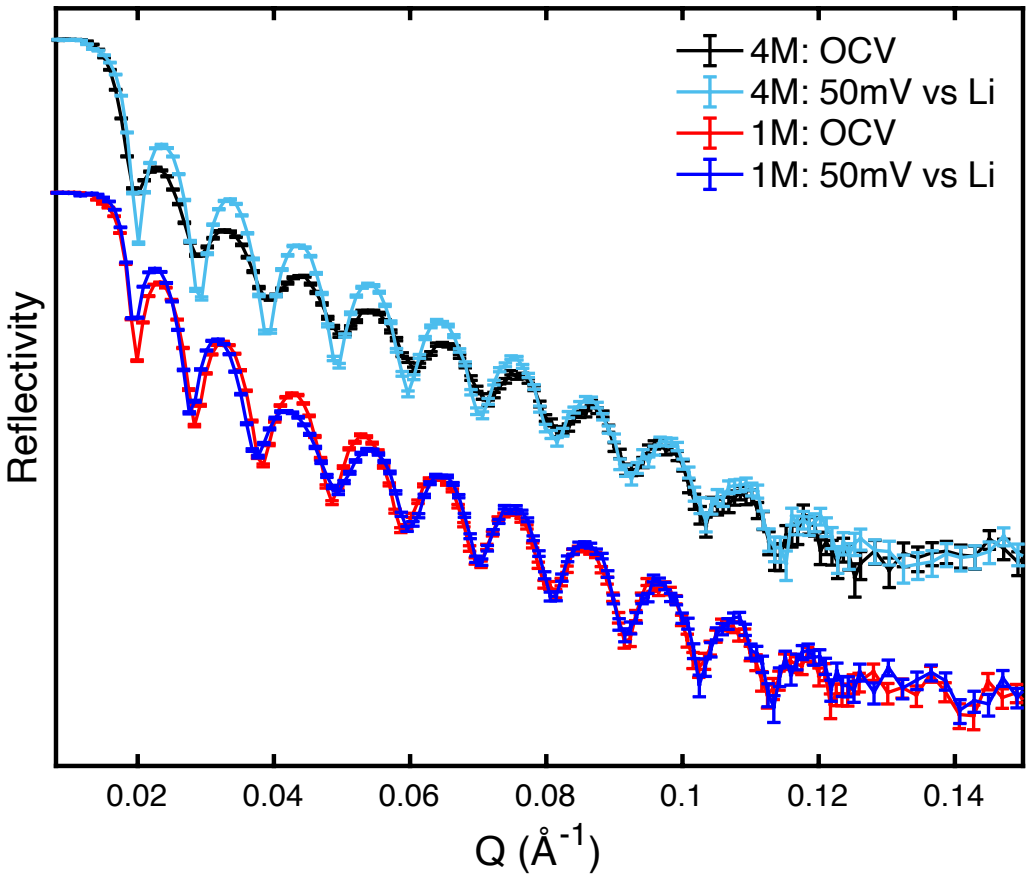

Figure 3. Reflectivity profiles from the electrochemical cell with the two different electrolytes (1M and 4M salt concentration) and with two different potentials applied. OCV denotes open circuit voltage (no potential applied).