

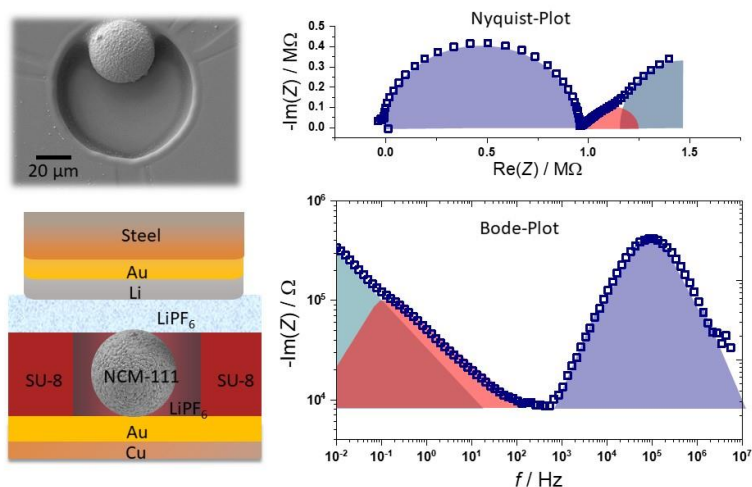
ELECTROCHEMICAL PROPERTIES OF MICRO-BATTERIES WITH SINGLE NCM-111 SECONDARY PARTICLES AS CATHODE

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Although lithium ion batteries (LIB) are already used in numerous applications, e.g. as power source in portable devices, the optimization of the battery performance, such as life-time, cyclability and energy density, is of large interest for using LIBs in e.g. electrical vehicles or temporary storage systems for renewable energy sources. Typically, the performance of the cathode active material is investigated using composite electrodes. Advanced composite electrodes consist of a complex architecture with comparably large secondary particles (10 μm - 30 μm) of the active material built up from nanometer sized primary particles. Furthermore, they also contain additives influencing the electrochemical properties of the composite electrode. To avoid such influences and to further optimize the performance of the cathode's active material a detailed understanding of the impact of the cathode architecture on the ionic and electronic transport processes is necessary. Here, we present the investigation of the ionic and electronic transport properties of single secondary particles of the active cathode material $\text{Li}(\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3})\text{O}_2$ (NCM-111) using electrochemical impedance spectroscopy. For this purpose, single NCM-111 particles have been arranged in traps on a glass substrate deposited with copper or gold as electrode using the meniscus force deposition method [1,2]. The traps on top of the metal film were prepared by patterning of a SU-8 resist films using photolithography. Using a specially designed electrochemical cell, the single secondary particles were contacted using a stainless steel cylinder. To determine the electronic conductivity of the single secondary particle the top electrode was coated with gold as ion blocking metal electrode. For measuring the micro-battery performance, the cell was assembled using a separator impregnated with liquid electrolyte (1M LiPF₆ in 50:50 PC:DMC) and a cylinder with lithium foil as anode. Impedance measurements were performed with an amplitude of 50 mV in the frequency range between 10 mHz and 7 MHz..



The impedance measurements using gold as blocking electrodes for the ionic transport reveal two transport processes, which are attributed to a contact resistance and the electronic transport in the NCM-111 particle. By a systematic analysis of the results it was possible to determine the partial electronic conductivity of single secondary particles of mixed-ionic electronic conducting materials, which is in good agreement with values obtained from measurements on pressed pellets. Furthermore, by studying the impedance response of different cell arrangements, it was possible to identify the different transport processes visible in the impedance spectra of the micro-battery.

Figure 1 –Nyquist- and Bode-plot of the impedance spectrum of a microbattery revealing several transport processes.

[1] Y. Cui, M. T. Bjrk, A. Liddle, C. Snnichsen, B. Bousset, and A. P. Alivisatos, "Integration of colloidal nanocrystals into lithographically patterned devices", *Nano. Lett.*, 4, 1093 (2004).

[2] A. Fabian, M.T. Elm, D.M. Hofmann, and P.J. Klar, "Hierarchical structures of magnetic nanoparticles for controlling magnetic interactions on three different length scales", *Appl. Phys. Lett.*, 121, 224303 (2017).