Investigation of Mixed Ion and Electron Transport in Single NCM Particles Using Impedance Spectroscopy

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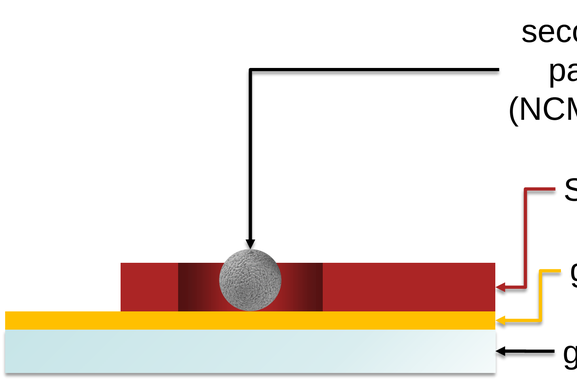
Motivation

- strong interest in improving the lithium ion battery performance driven by industrial and societal needs
- fundamental processes during cycling of batteries faded from the spotlight
- majority of studies focus on composite electrodes, thus properties of electrode material superposed by additives‘ properties
- investigate transport properties of single NCM particles for understanding the fundamental processes as basis for improving corresponding devices

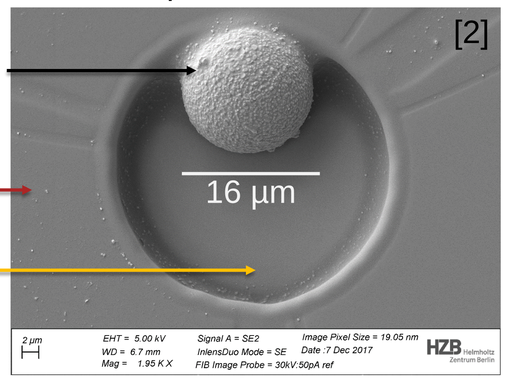
Contacting Single Secondary Particles

- particles arranged in cylindrical traps on a lithographically patterned substrate

sketched cross section

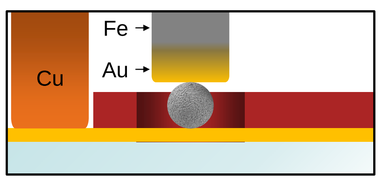


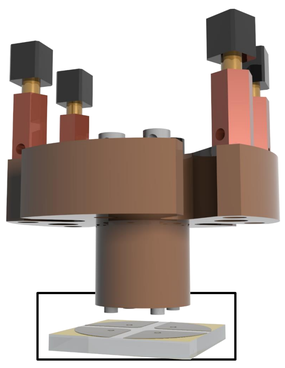
top view in SEM



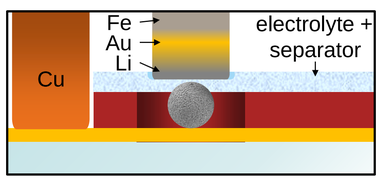
- electrochemical cell designed to contact single particles in different configurations

electronic charge transport

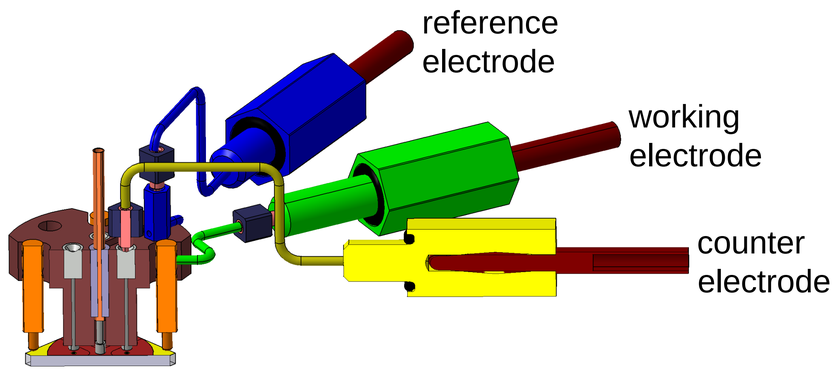




ionic charge transport

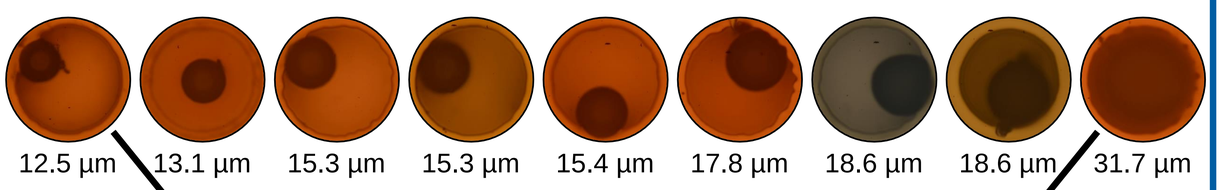


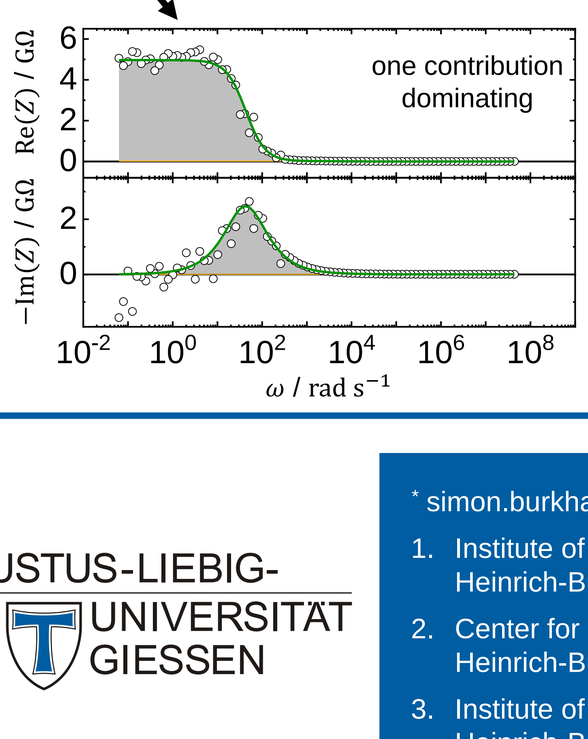
- three electrode electrochemical cell for experiments on single particles

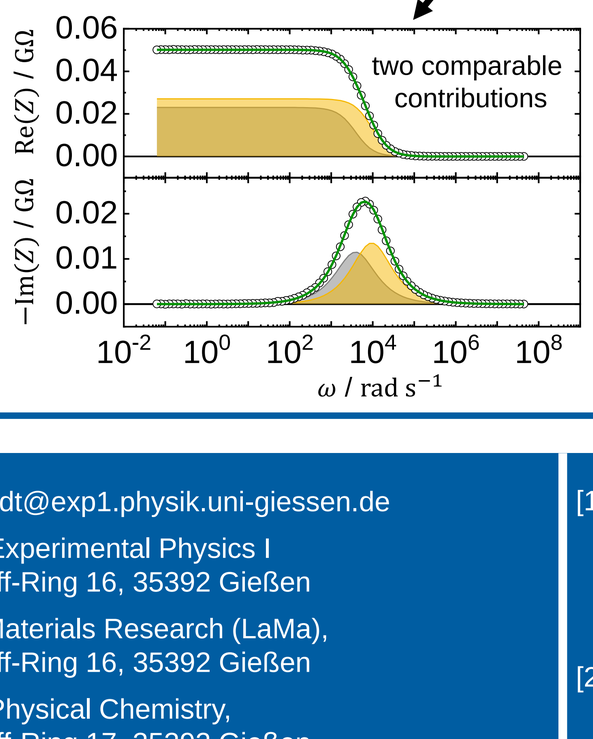


Impedance of Single Secondary Particles

- dimension of secondary particles influences impedance signal

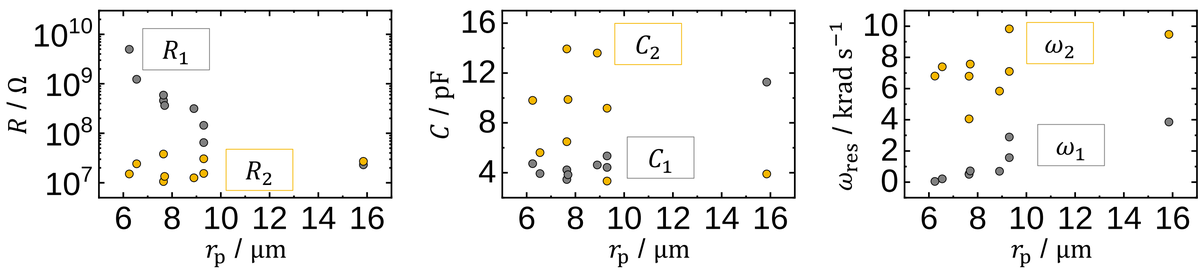






Two Different Electron Transport Processes

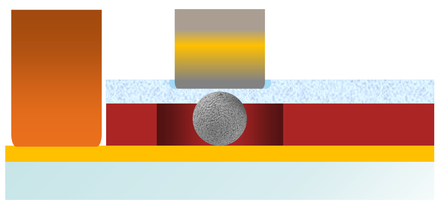
- simulation of impedance with equivalent circuit consisting of two RC elements in series



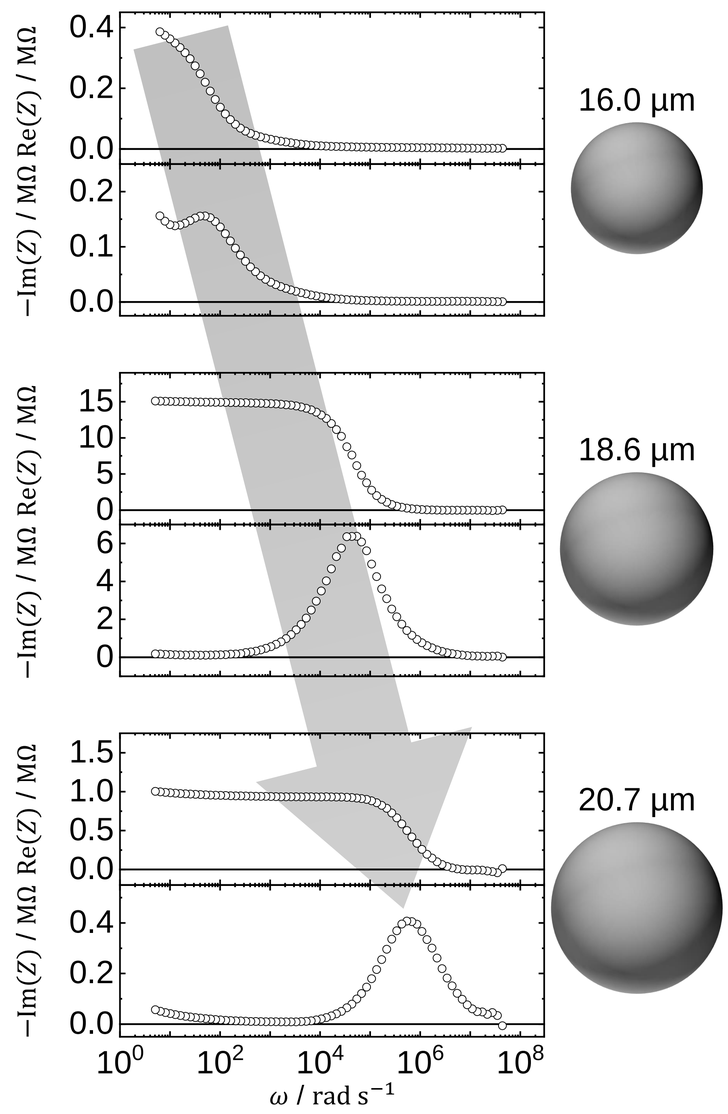
- assignment of transport processes to size-dependent resistance R_1 and size-independent resistance R_2

Size Dependence in Single Particle Batteries

- half cell of a lithium-ion battery realized
- cathode consisting of single secondary particle



- diameter of secondary particle has been varied
- resonance frequency increases with increasing particle size
- ionic charge transport in secondary particles:
  - charge transfer from electrolyte to particle
  - diffusion in grains
  - diffusion along grain boundaries



Summary

- electrochemical cell designed to measure impedance of single secondary particles
- investigation of electronic and ionic charge transport in single particles
- influence of particles’ diameter on both transport processes revealed
- increase of resonance frequency observed with increasing size of particles in electronic as well as ionic charge transport
- further experiments for characterization of single particles necessary

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[1] $Li(Ni_{0.33}Co_{0.33}Mn_{0.33})O_2$ (NCM-111) secondary particles have been prepared by Dr. Joachim Binder and co-workers at Karlsruhe Institute of Technology (KIT), Institute for Applied Materials, Department for Ceramic Materials and Technologies (joachim.binder@kit.edu)

[2] SEM investigation performed by Markus Osenberg, Department of Materials Science and Technology, Technische Universität Berlin (markus.osenberg@tu-berlin.de)