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The Effect of Mechanical Cycling on the Electronic Conductivity of Composite Electrodes

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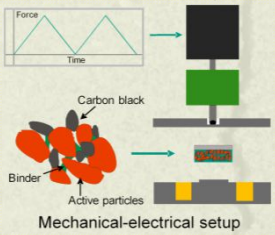
Motivation

During battery operation, active materials change their volume. This can cause mechanical damage in the form of cracked active particles [1]. Even when the active material is not damaged, binder joints can move and carbon black (CB) can redistribute [2]. Here we explore how this affects the electrode performance. In order to isolate these mechanical effects, we exclude electrochemical changes by pure mechanical cycling. This serves as an experimental tool to determine changes of the conductive network and can be used as a test of the mechanical reliability.

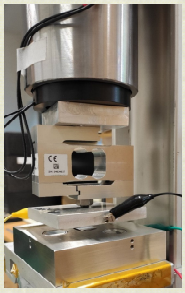
[1] Chen, D. et al. Electrochemical Acta 2018, 259, 939-948. DOI: 10.1016/j.electacta.2017.10.179

Experimental Setup

- Dry electrode sheet placed between two metal plates
- Force is chosen to cause similar stresses than during battery operation (as estimated by *in situ* substrate curvature experiments [2])
- Simultaneous measurement of resistivity
- Variation of T



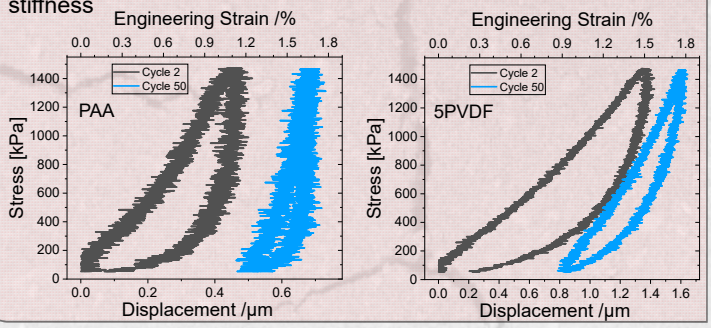
Mechanical-electrical setup



[2] Janzen, M. et al. Energy Technology 2021, 9(6), 2000867. DOI: 10.1002/ente.202000867

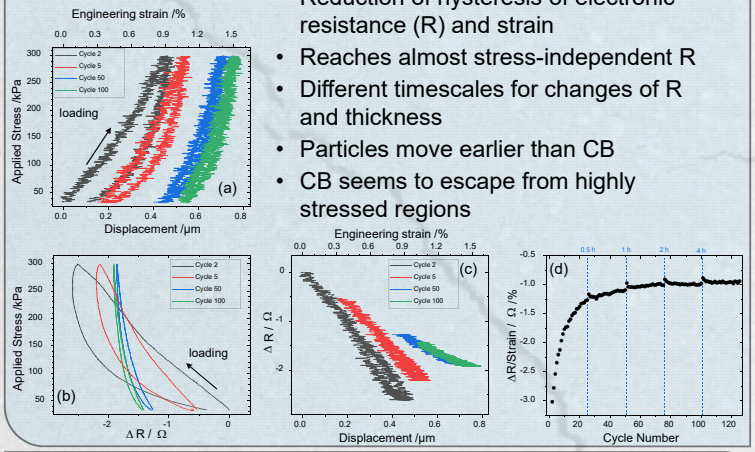
Results 1. Comparison of Binder Materials

- Binder interconnects particles and binder with higher modulus stiffens the composite electrode
- Also increased amount of binder (from 5% to 10% PVDF) leads to a stiffer electrode
- Choice of binder material and amount allows tuning of electrode stiffness



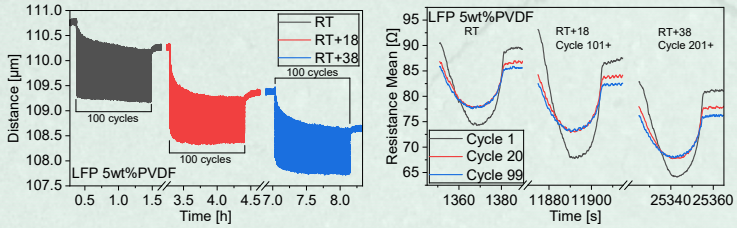
Results 2. Electrode Rearrangement by Mechanical Cycling

- Reduction of hysteresis of electronic resistance (R) and strain
- Reaches almost stress-independent R
- Different timescales for changes of R and thickness
- Particles move earlier than CB
- CB seems to escape from highly stressed regions



Results 3. Thermal Activation

- During cycling, R and thickness evolve asymptotically
- Increasing T initiates new rearrangement and leads to smaller values
- Possibly caused by softening/plasticity of polymeric binder
- Athermal processes also cause changes in the conductive network [3] but are not in agreement with our findings.



[3] Becker, V. et al. Energy Technology 2021, 9(6), 2000886. DOI: 10.1002/ente.202000886

Conclusion

- Mechanical cycling leads to a rearrangement of the electrode. This was observed in the mechanical strain data, its hysteresis and in the electronic conductivity.
- During cycling, mechanical behavior and electronic resistivity depend on temperature. It can be expected that complex mechanical changes occur when a battery is operated at different temperatures.

Next steps

- Influence of solvents on mechanical and electronic properties
- Fatigue testing of electrodes by fast mechanical cycling

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