Engineering Activated Carbon via Chemical Vapor Deposition for High-Performance Core-Shell Carbon Anodes in Sodium-Ion Batteries

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Abstract: The growing need for sustainable energy storage has revived interest in sodium-ion batteries (SIBs). While hard carbons are commonly used in SIB anodes, the link between their structure and Na-storage mechanism is only partially understood. Activated carbons (ACs), with their abundant micropores and high surface area, show promise as a low-cost anode material. However, their performance is limited by structural disorder and excessive solid-electrolyte interphase (SEI) formation, which causes active-species loss and hinders high specific capacities. As recently shown, these limitations can be overcome by sealing AC particles with a semi-permeable coating via chemical vapor deposition (CVD) to form core–shell carbon structures, delivering reversible capacities of ≈ 400 mAh g^{-1}.[1] This study explores the interplay between the porosity characteristics of ACs, the CVD conditions, and the resulting performance as SIB anodes.

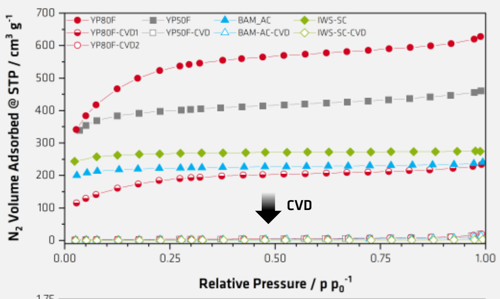
Specific Surface Area & Porosity

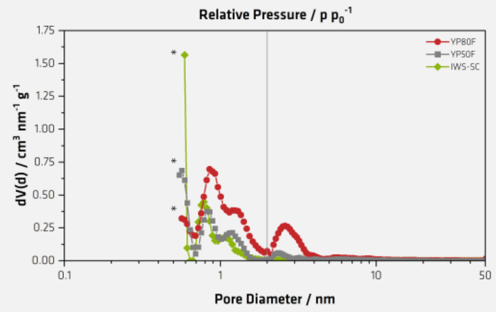
- Four different AC materials with varying properties and morphology were treated via a solvent-based CVD process[1]
- Aim: Low N_2-surface area (BET-method, 77 K) of < 20 $m^2\ g^{-1}$ by closing open porosity
- For YP80F: CVD-Process had to be conducted twice, possibly, due to a higher SSA and TPV

Name	SSA / $m^2\ g^{-1}$	TPV / $cm^3\ g^{-1}$	SSA after CVD / $m^2\ g^{-1}$
YP80F	1696	0.96	12.4 (591*)
YP50F	1220	0.71	13.5
IWS-SC	682	0.43	1.38
BAM-AC	670	0.37	8.84

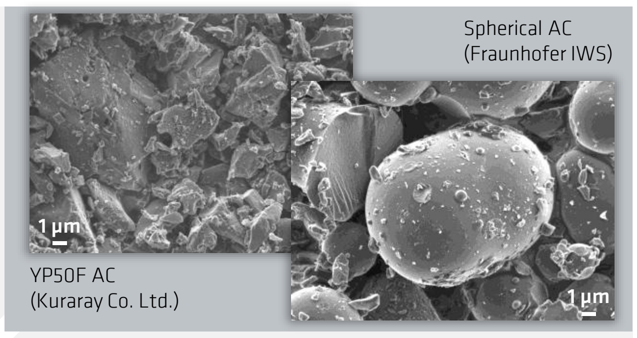
SSA: Specific Surface Area, TPV: Total Pore Volume

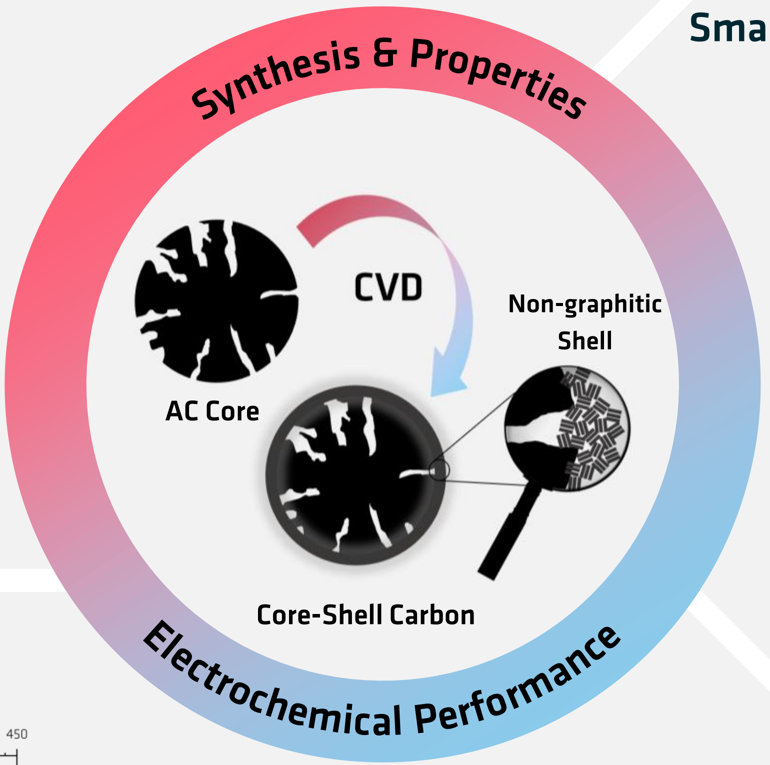
*SSA after 1st CVD treatment





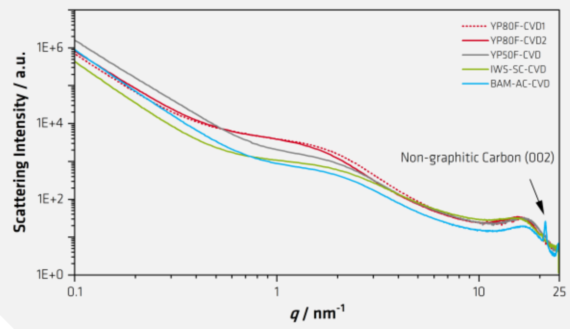
* Artifacts from QSDFT





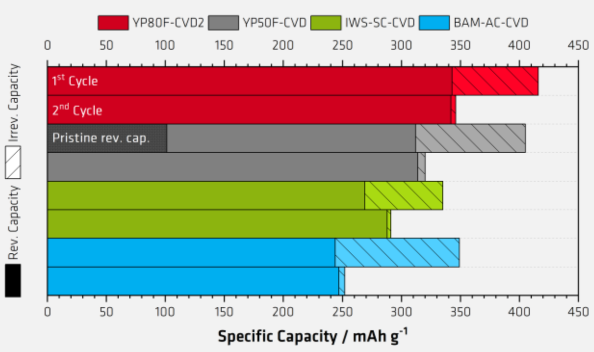
Small-angle X-ray Scattering (SAXS)

- All samples clearly show remaining internal porosity after the CVD-process
- SAXS confirms higher share in micro- and (small) meso-porosity for YP80F
- CVD of BAM-AC yields non-graphitic multi-layered carbon, indicating a thick shell



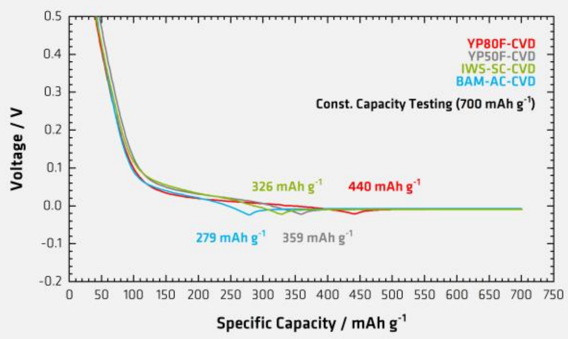
Sodium-Ion Battery Anodes

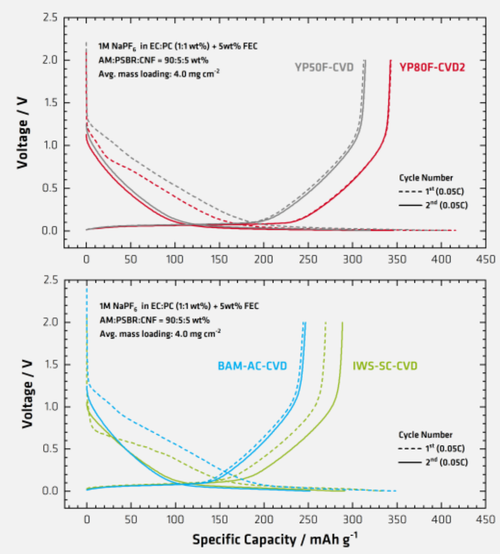
Half-Cell Performance vs. Na-Metal



- Pristine YP50F with 700 mAh g^{-1} irrev. capacity
- Higher SSA and TPV of pristine materials leads to increased reversible capacity after CVD
- Significant increase of rev. capacity for IWS-SC-CVD in the 2nd cycle, possibly, due to active species trapping in the initial cycle

- YP80F-CVD with highest micro- and mesoporosity share showed best performance with max. capacity of 440 mAh g^{-1}
- BAM-AC-CVD revealed the highest irrev. losses and lowest spec. capacity, possibly, due to formation of thick shell





Conclusion: Commercial ACs with varying SSA and porosity were treated with a CVD-process to create a core-shell structure comprising a porous AC core and a non-graphitic carbon shell. By sealing the open porosity of the core, the formation of excessive SEI is effectively mitigated, while enabling efficient reversible sodium storage within the core's porosity. SAXS proved that porosity was maintained, however, a thick shell was apparently formed for one of the samples. Generally, a higher initial SSA and TPV led to higher reversible capacity after CVD-treatment. Hence, the porosity features of the AC core seems to allow a rough prediction of the Na-capacity of the materials. The factors leading to the formation of the thick carbon shell still need to be understood. Future studies could explore the scalability of the process and the incorporation of the CVD-treated materials into solid-state battery applications.[2]

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