

Correlation of electrode expansion and cyclability in graphite-based negative electrodes

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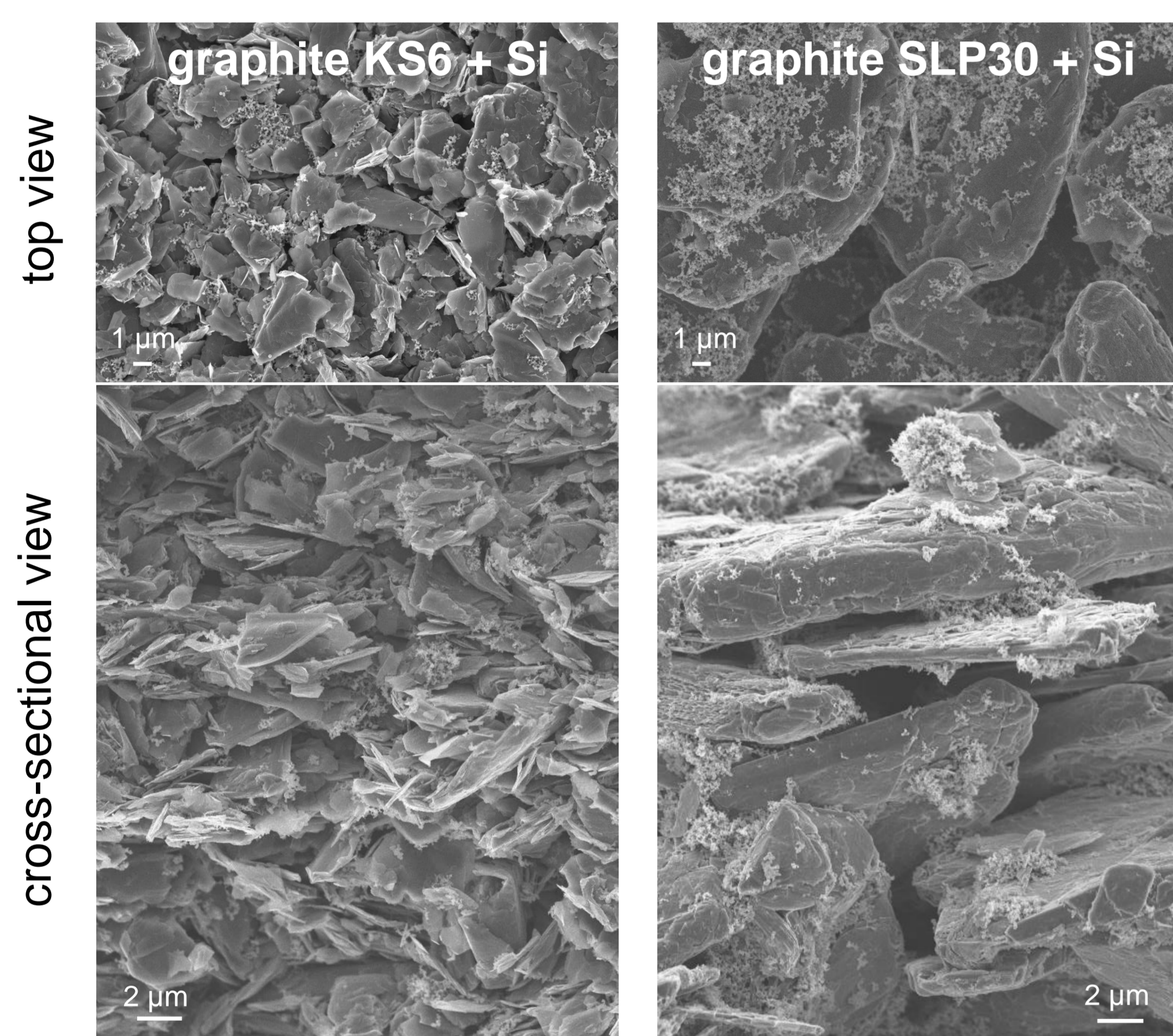
Motivation / Targets

- Develop carbon based anode material with specific charge > 450 mAh/g by addition of small amounts of silicon
- Investigate the influence of graphite / binder combination on cycling
- Understand the relationship between expansion and electrode failure

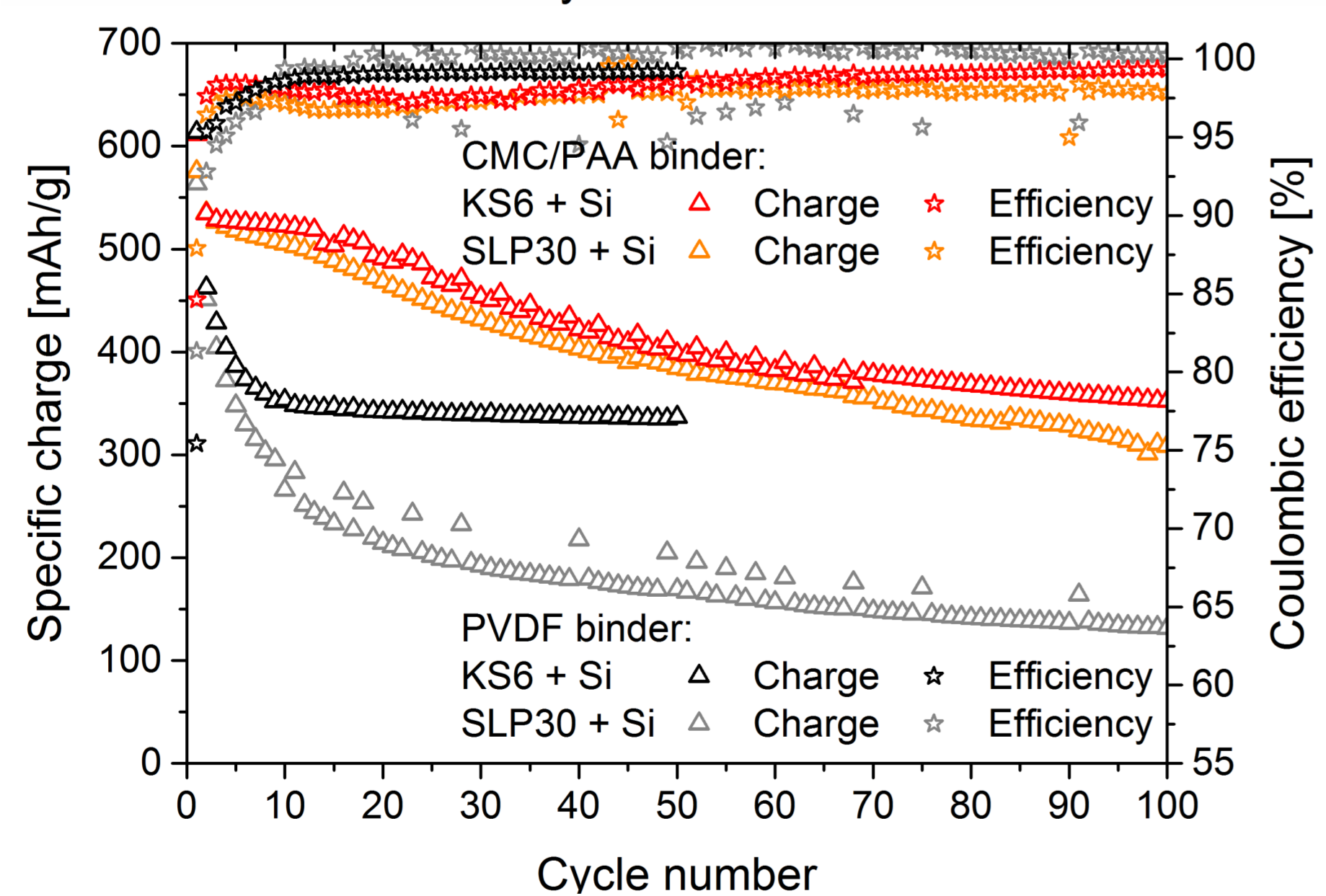
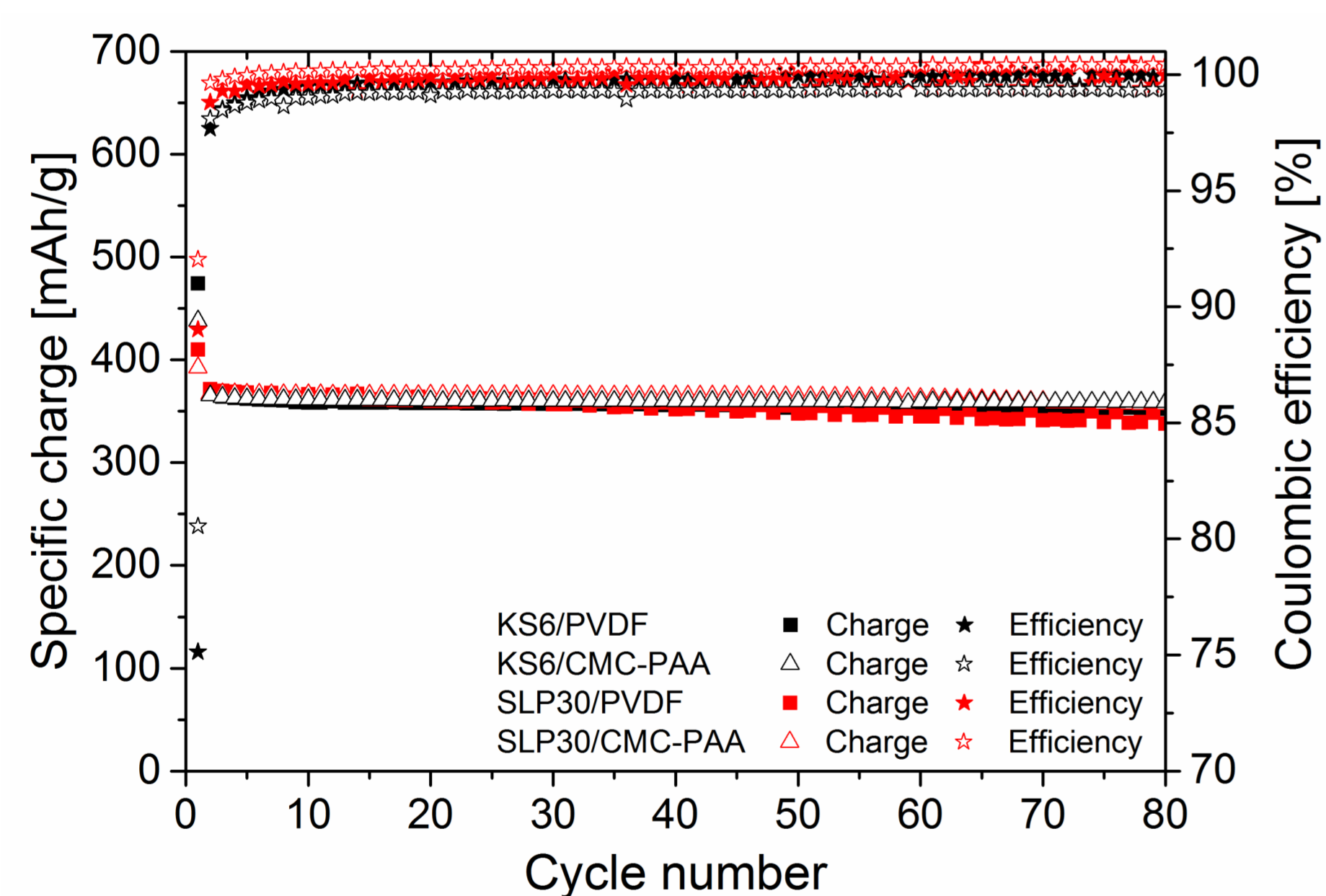
Experimental

- 4.75 wt% Si, 90.25 wt% intercalating carbon, 1 wt% SuperC65, 4 wt% binder
- Baseline: 95 wt% intercalating carbon, 1 wt% SuperC65, 4 wt% binder
- Electrochemistry: 20 mA/g 1st cycle, then 50 mA/g, 5 mV – 1.5 V vs Li⁺/Li
- Electrolyte: 1M LiPF₆ in EC/DMC 1:1 (w:w)

Graphite / Binder influence

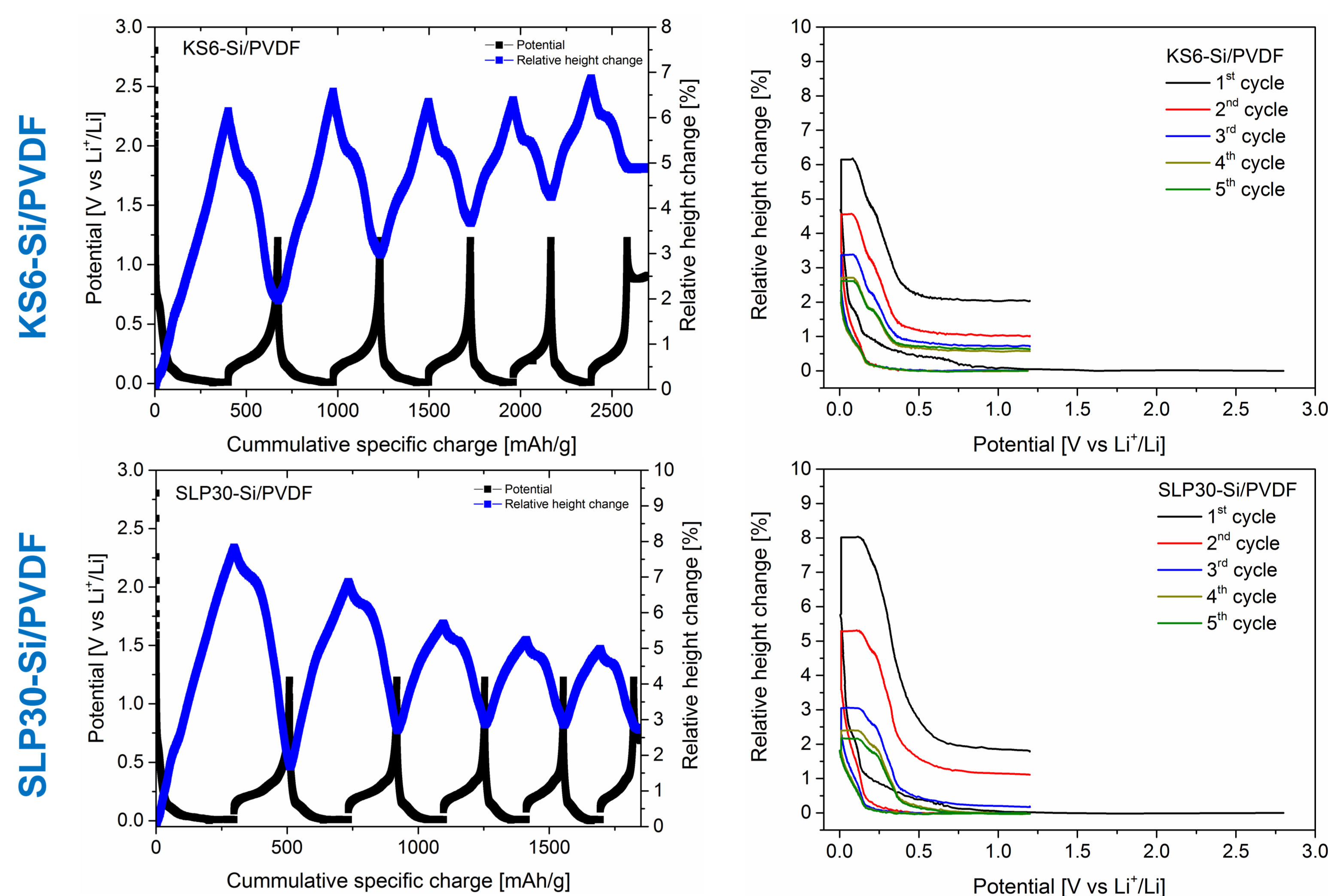


- Larger particles & higher aspect ratio for SLP30 than KS6
- preferred orientation of SLP30 parallel to substrate

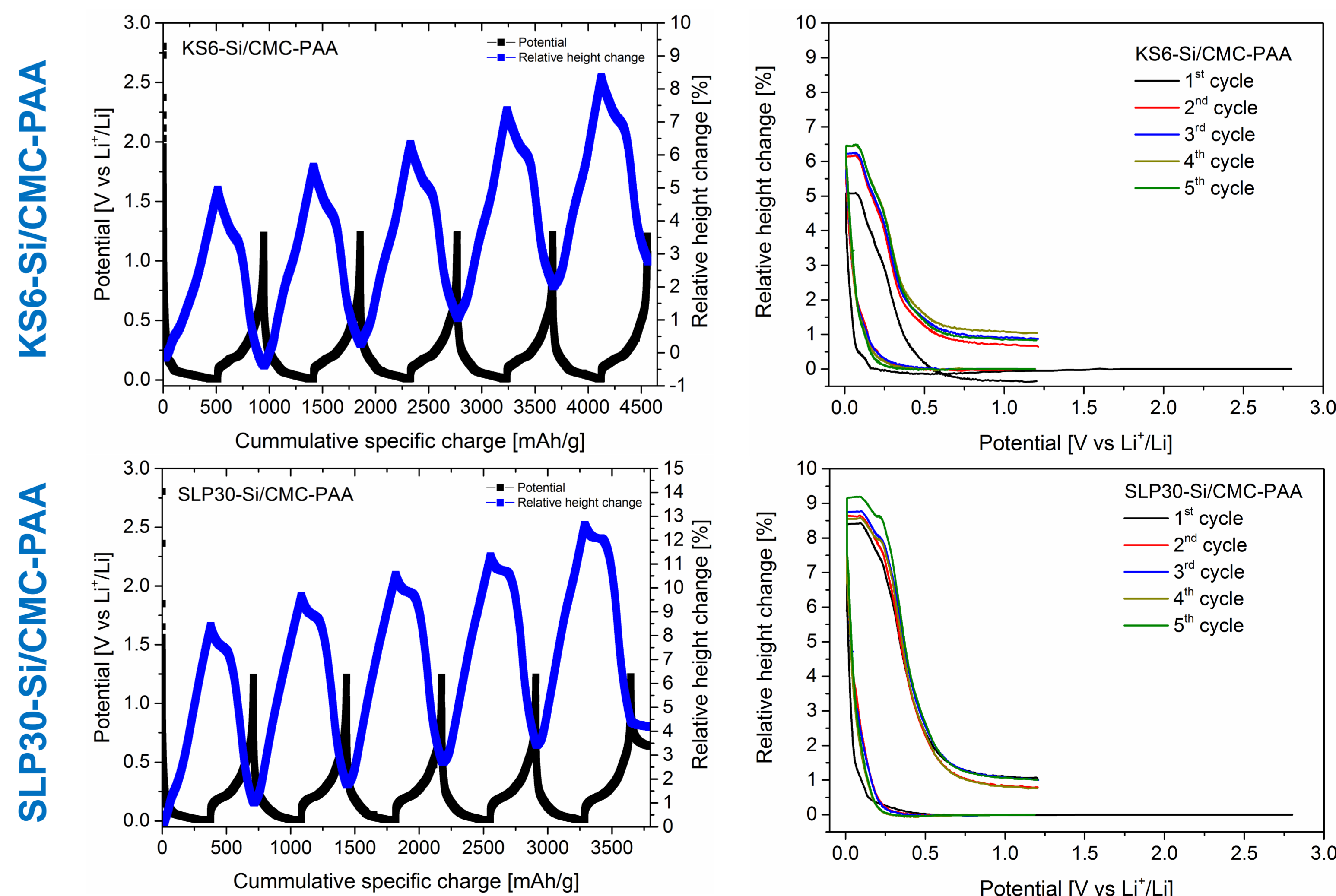


- Graphite only: stable cycling regardless of the binder
- Graphite with silicon: improved cycling stability for both KS6 and SLP30 based electrodes with CMC-PAA crosslinked binder

Electrochemical *in situ* dilatometry



- PVDF is a stiff binder
 - KS6 and SLP30 electrodes expand irreversibly ca. 2% in 1st cycle
 - Capacity fading linked to “loss” of active Si → lower expansion in subseq. cycles
- CMC-PAA is an elastic binder
 - Irreversible expansion of ca. 1% → recovered every cycle
 - Holds expansion of up to 9% with low irreversibility
 - Capacity fade is **not** due to “loss” of active Si → different failure mechanism



Conclusions

- Both KS6 and SLP30 electrodes show stable cycling regardless of the binder
- Different failure mechanism with PVDF
 - KS6-Si keeps the graphite charge, SLP30-Si fails even below graphite's capacity
- Binder has strong influence on accommodation of volume changes
- Failure mechanism is influenced by the binder type:
 - CMC-PAA holds larger expansion avoiding loss of active Si
 - *in situ* dilatometry provides new insights into binder performance

