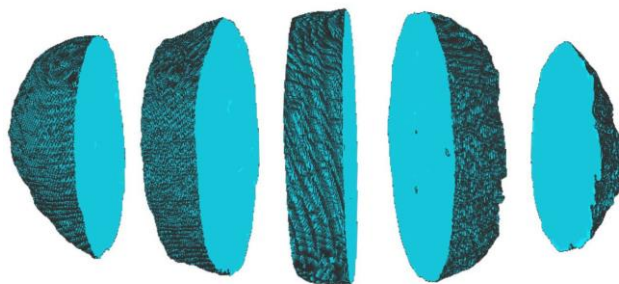
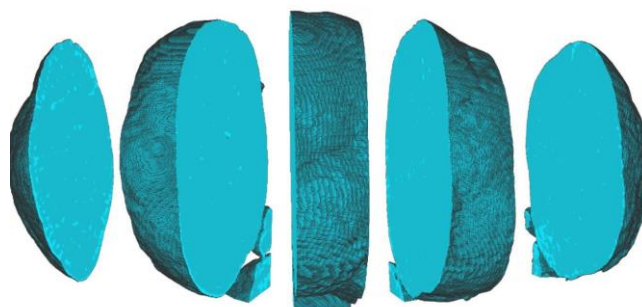

Microstrain screening towards defect-less layered transition metal oxide cathodes

In the format provided by the
authors and unedited

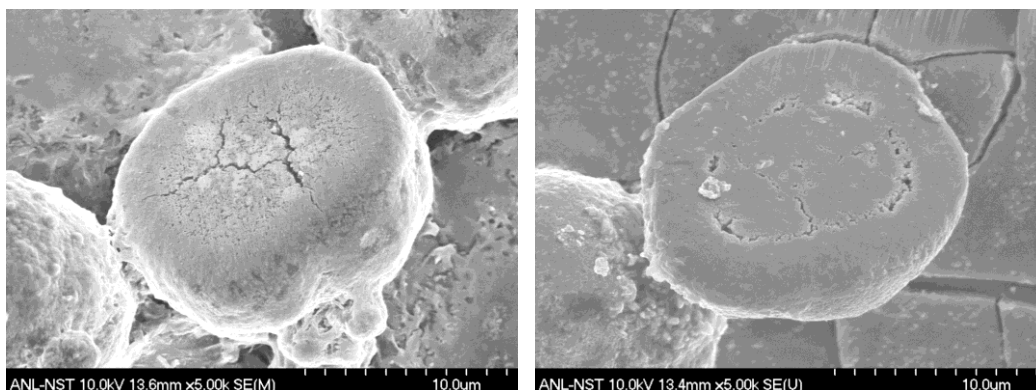
Conventional:



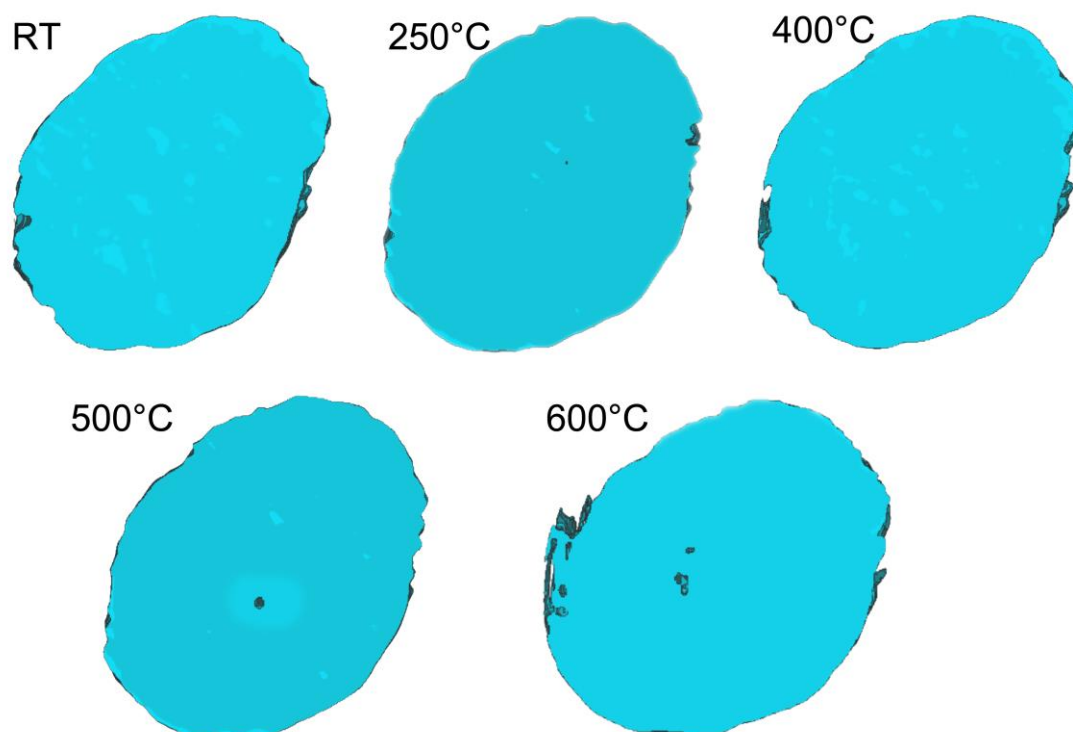
Core-shell:



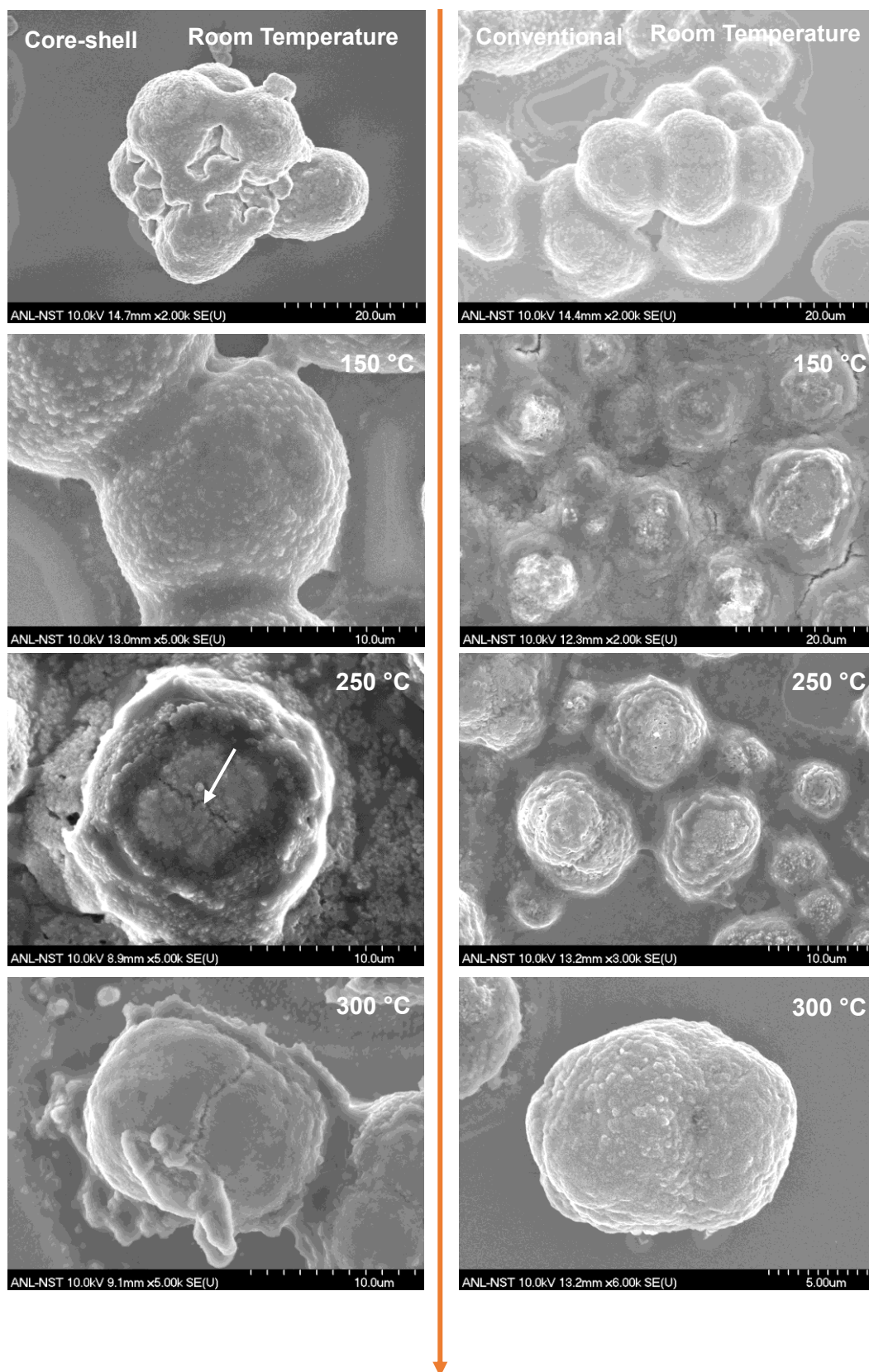
Supplementary Fig. 1: 3D view of conventional and core-shell $\text{Ni}_{0.80}\text{Mn}_{0.10}\text{Co}_{0.10}(\text{OH})_2$ precursors.



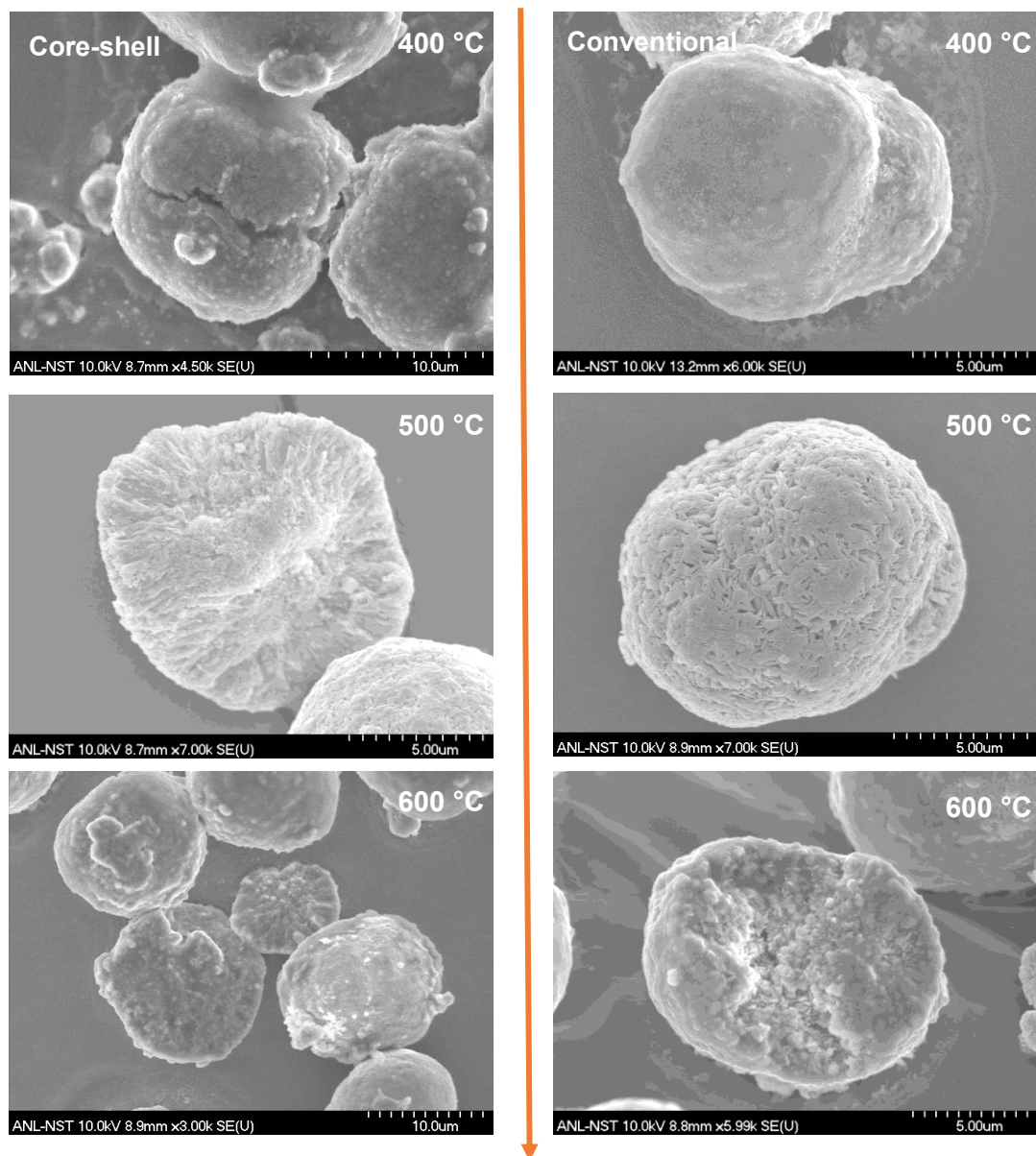
Supplementary Fig. 2: Cross-sectional SEM image of core-shell $\text{Ni}_{0.80}\text{Mn}_{0.10}\text{Co}_{0.10}(\text{OH})_2/\text{NaOH}$ mixture that heated to 250 °C, showing emerging cracks at both core (left) and boundary (right).



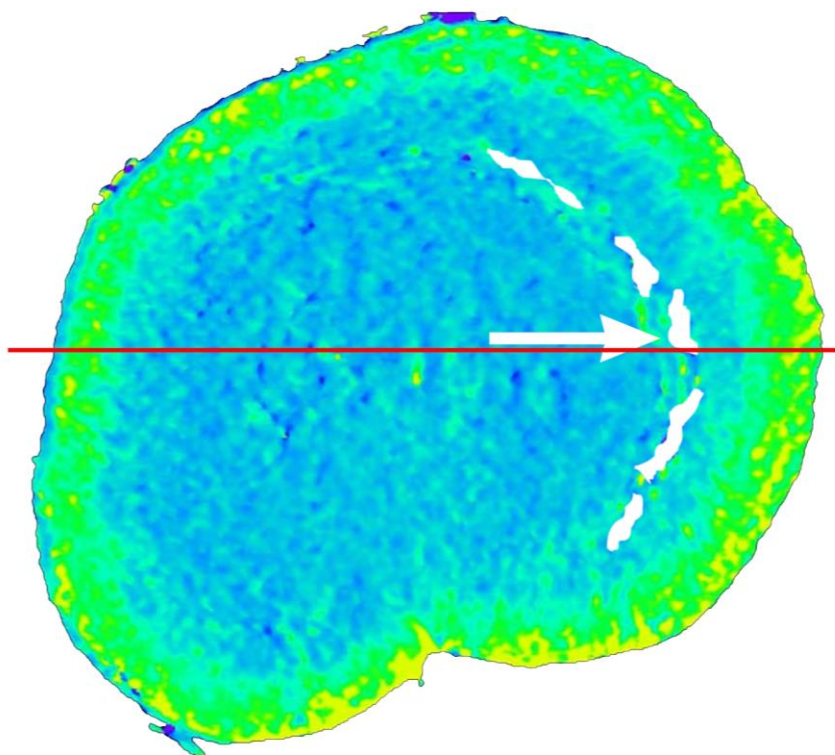
Supplementary Fig. 3: The *in-situ* heating 3D TXM-XANES results of conventional $\text{Ni}_{0.80}\text{Mn}_{0.10}\text{Co}_{0.10}(\text{OH})_2$ particle that was calcinated with NaOH at different temperature, showing absence of cracks or fragmentation



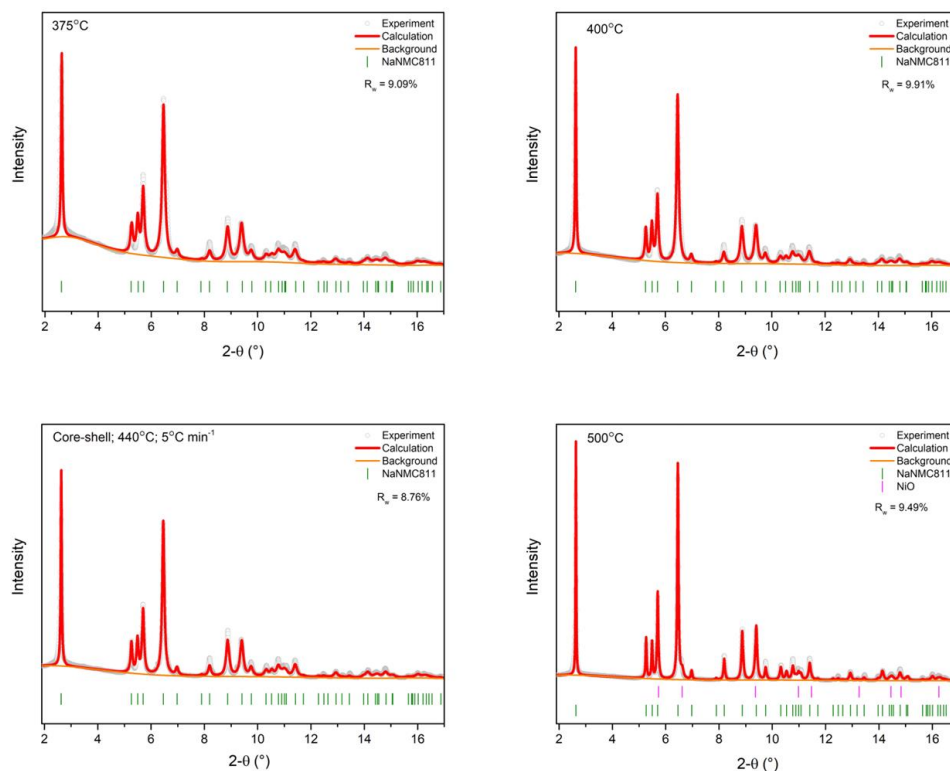
Supplementary Fig. 4: SEM images of core-shell and conventional $\text{Ni}_{0.80}\text{Mn}_{0.10}\text{Co}_{0.10}(\text{OH})_2\text{-NaOH}$ mixtures with a Na-TM molar ratio of 1.0 that were heated from room temperature to 300 °C. Cracks in the core-shell particle appeared at 250 °C and intensified with increasing temperature.



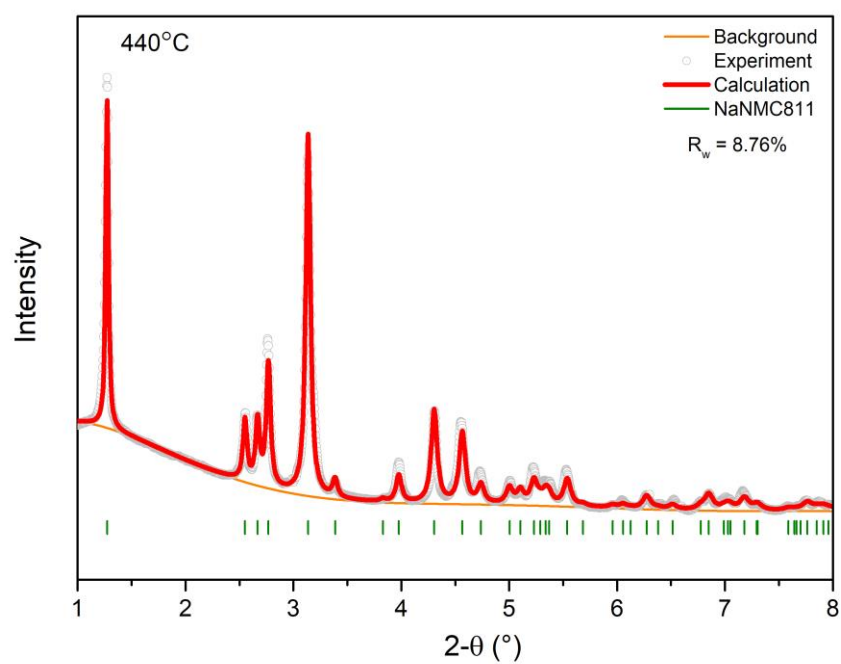
Supplementary Fig. 5: SEM images of core-shell and conventional $\text{Ni}_{0.80}\text{Mn}_{0.10}\text{Co}_{0.10}(\text{OH})_2\text{-NaOH}$ mixtures with a Na-TM molar ratio of 1.0 that were heated from room temperature to 600 °C (Continuation of Supplementary Fig. 4). The conventional particles showed no significant cracks or fragmentation below 500 °C, with only minor fragmentation observed above 600 °C.



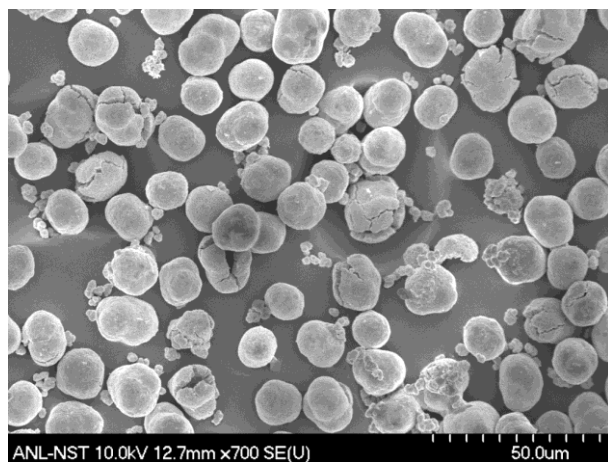
Supplementary Fig. 6: Illustration of collection pathway for white-line-energy intensity profile on the core-shell particle at 250 °C, corresponding to Fig. 3d.



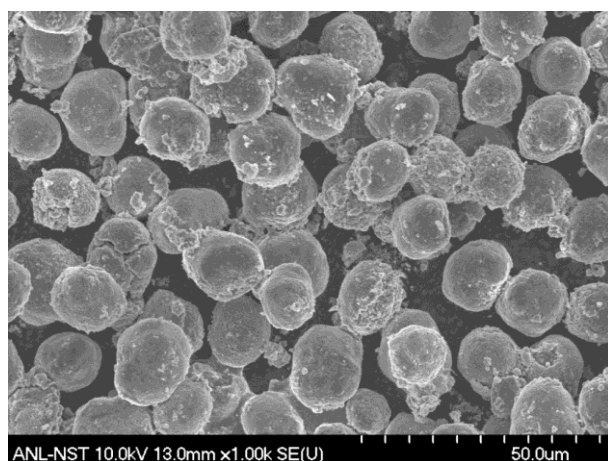
Supplementary Fig. 7: Powder SXRD pattern and corresponding Rietveld refinement results of core-shell NaNMC811 samples that were calcinated at various temperature for 20 h with a heating ramp rate of 5°C min⁻¹. Wavelength is 0.24108Å.



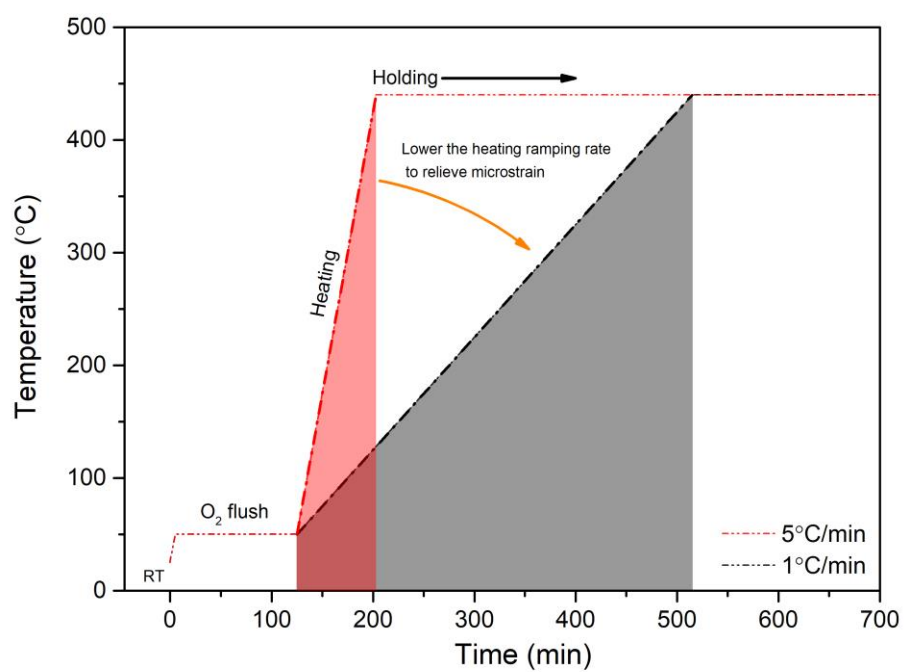
Supplementary Fig. 8: Powder SXRD pattern and corresponding Rietveld refinement results of conventional NaVMC811 sample that was calcinated at 440 °C for 20 h with a heating ramp rate of 5 °C min⁻¹. Wavelength is 0.1173 Å.



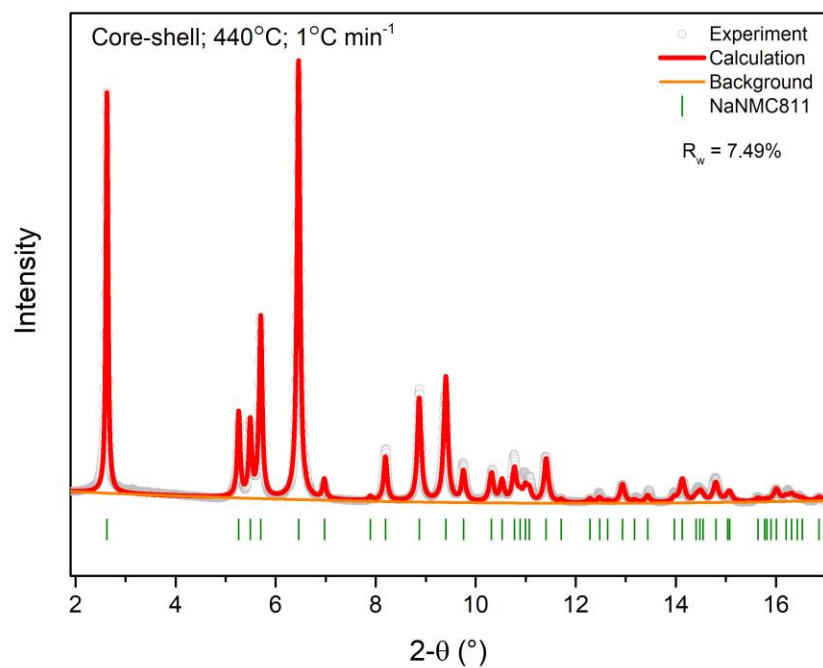
Supplementary Fig. 9: SEM image of core-shell NaNMC811 particles that were calcinated at 440 °C for 20 h, showing particle cracking.



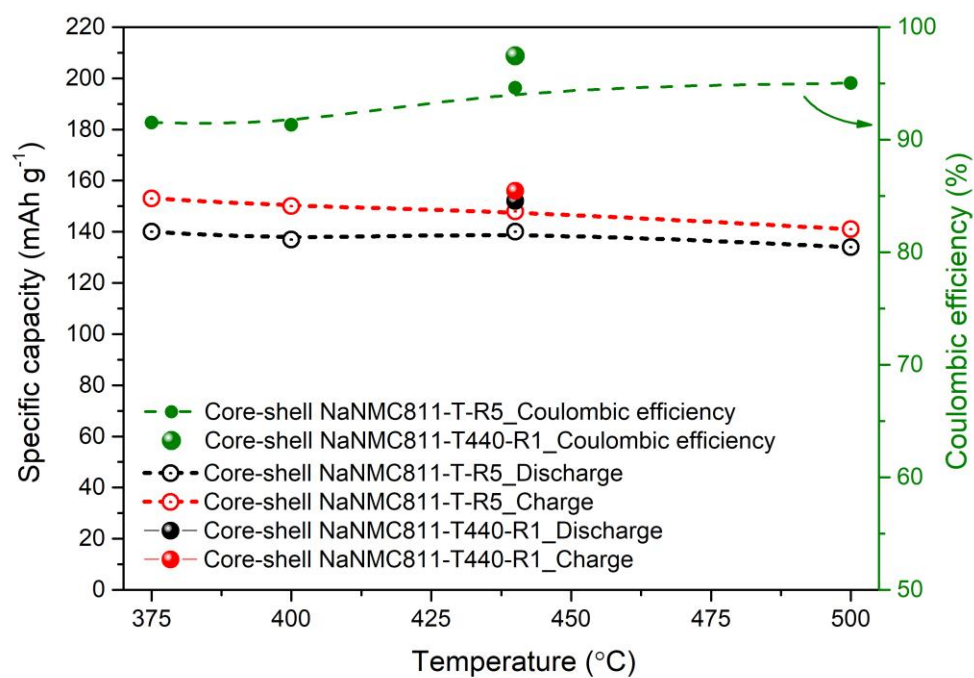
Supplementary Fig. 10: SEM image of core-shell NaNMC811 particles that were calcinated at 600 °C for 20 h, showing fragmentation and small-particle adherence.



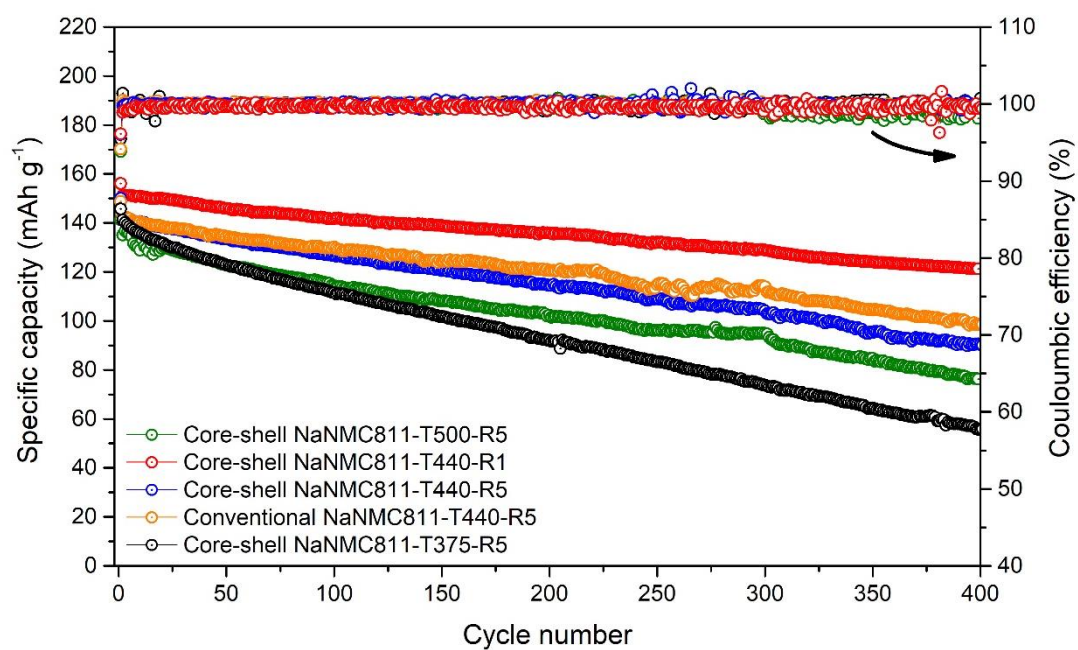
Supplementary Fig. 11: Schematic illustration of the heating ramp rate control.



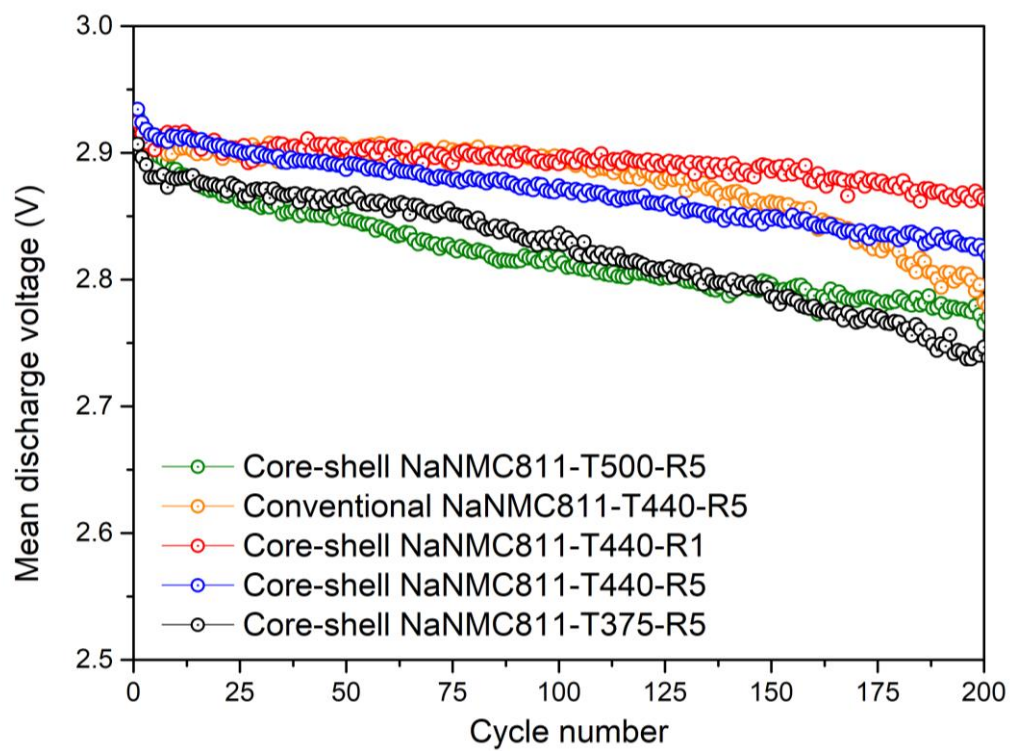
Supplementary Fig. 12: Powder SXRD pattern and corresponding Rietveld refinement results of core-shell NaNMC811 sample that was calcinated at 440 °C for 20 h with a heating ramp rate of 1 °C min⁻¹. Wavelength is 0.24108 Å.



Supplementary Fig. 13: Initial charge/discharge capacities and Coulombic efficiency of core-shell cathodes.



Supplementary Fig. 14: Cycling stability of core-shell NaNCM811 cathodes and conventional NaNCM811-T440-R5 sample at 0.5C.



Supplementary Fig. 15: Mean discharge voltage of NaNMC811 cathodes.