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By a combination of the bulk XRD study with the local TXM and STEM analysis at different length scales, we were able to confirm the formation of the nanocomposites in NM9505 with 0.95Li, consisting of the main layered phase and minor RS-structured phase intergrown within the same ccp oxygen framework, with the epitaxial orientation relationship (illustrated in Fig. 2d).

An important cell parameter in the layered phase is the Li slab thickness, serving as an indicator of the structural ordering of the layered phase. Interestingly, Li-deficient samples displayed a larger Li slab (Table S2) because there is less Li/Ni mixing in those layered domains in the composite-structured NM9505. In addition, the domain size of the layered phase increased with the rise of Li content, as displayed in Fig. 3d, highlighting the critical role of lithiation in driving crystal growth.

As reported in the literature, the primary particle size is impacted by various factors during calcination, including chemical composition, temperature, and oxygen pressure35,46,47,48. We demonstrate here the strong dependence of primary particle size on Li content in the precursors (and so the Li stoichiometry in the synthesized CAMs), which is corroborated by multiscale modeling, providing insights into the role of Li stoichiometry in governing phase progression and crystallization.

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