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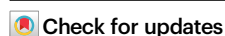
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Revealing multiscale competing processes in the solid-state synthesis of single-crystalline layered oxide positive electrodes

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Solid-state synthesis involves a web of coupled chemical reactions and physical changes that unfold across multiple scales. Efforts to fine-tune its parameters have historically followed heuristic, trial-driven workflows that demand significant time and resources. In this study, we aimed to open this black box by employing multiscale in situ synchrotron imaging and diffraction. Using $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ battery positive electrode material as a model system and Ba-based sintering aids, we reveal dopant segregation, intergranular mass transport, and porosity evolution as key drivers of single-crystalline particle formation. Notably, we uncovered a dynamic competition between particle-level grain coalescence and atomic-scale cation disordering, both of which are thermally activated yet have opposing impacts on battery performance. These findings highlight the coupled, multiscale nature of structure development and offer a mechanistic basis for optimizing the solid-state synthesis process. This framework provides a path toward more controlled, efficient, and scalable production of high-performance battery positive electrode materials.

Solid-state synthesis is a highly complex process governed by intertwined chemical and physical transformations. Optimizing the synthesis conditions has traditionally relied on empirical trial-and-error approaches, which are both inefficient and costly. A high-profile example is in battery industry, where the solid-state synthesis of positive electrode materials is critical to a range of performance matrices^{1,2}, yet its optimization relies on labor-intensive trials. Specifically, $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ (NMC, where $x+y+z=1$) layered oxides are among the most promising candidates for high-energy-density LIB positive electrode, offering a critical pathway to meet the rigorous performance requirements of high demanding applications, such as electric vehicles^{3,4}. To meet the increasing need for improved physical and electrochemical performance, significant efforts have been

devoted to refining the synthesis and manufacturing processes^{5,6}. At a high level, the NMC synthesis protocol may appear straightforward and well-established: co-precipitated precursors are mixed with a lithium source and sintered at high temperatures to produce the final positive electrode material^{7,8}. In practice, however, this process is highly intricate, requiring fine control over numerous variables such as precursor composition^{9,10}, heating profiles¹¹, and sintering additives¹². These parameters are typically optimized through iterative trial-and-error approaches, with the electrochemical performance of the final product serving as the primary feedback metric.

As it is illustrated in Fig. 1, the optimization of the synthesis protocol spans two interlinked domains: process optimization, which focuses on adjusting synthesis conditions (e.g., temperature¹³,

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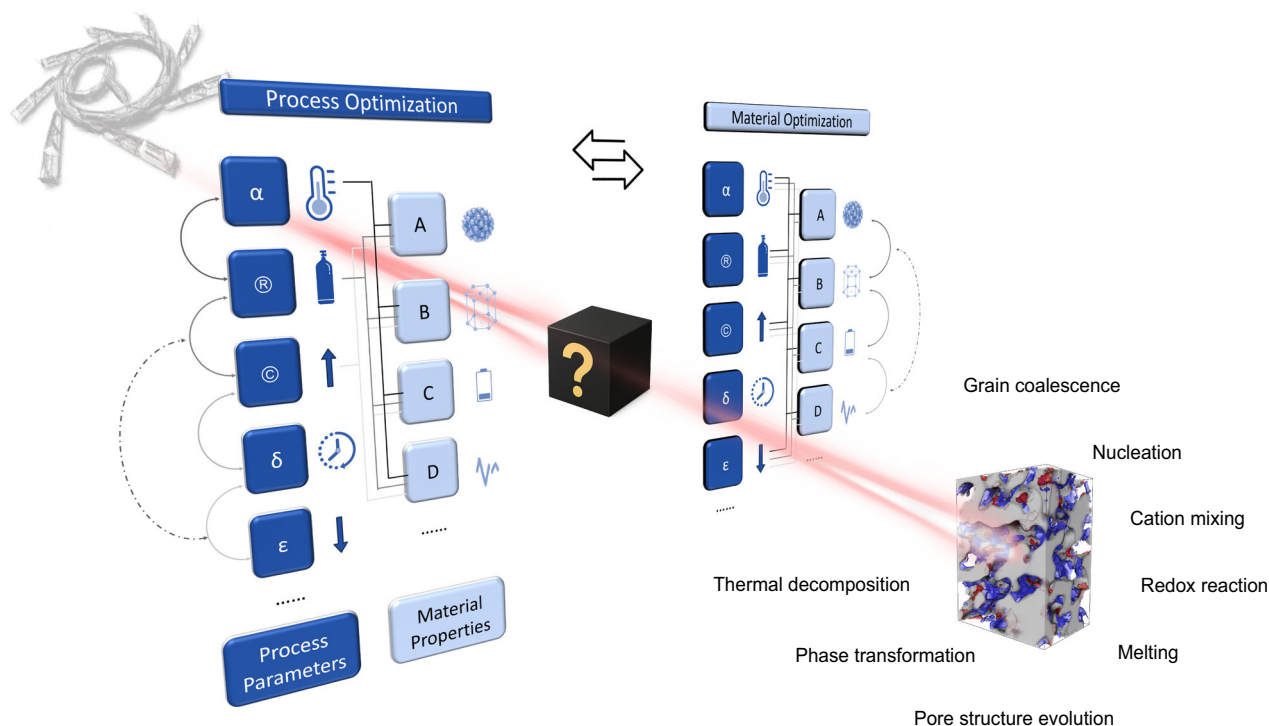


Fig. 1 | Integrating synchrotron characterization into NMC synthesis. Conventional synthesis often relies on trial-and-errors, with limited understanding of the underlying structure evolution. The left panel illustrates a process optimization workflow, where synthetic process parameters (α – ϵ) are varied to generate products with varying material properties (A–D). The right panel shows a material

optimization pathway focused on tailoring specific material properties through fine tuning the synthesis conditions. Synchrotron-based in situ characterization provides multimodal feedback, offering real-time, data-driven insight to inform both optimization pathways.

atmosphere¹⁴, heating and cooling rates¹⁵, and dwell time¹⁶); and material optimization, which tailors microstructure and composition for targeted performance metrics^{17,18}. Currently, these two aspects are often executed sequentially and iteratively, relying heavily on ex situ, offline feedback loops. As a result, the process remains a “black box”, in which the dynamic structural and morphological transformations are not well-understood. This challenge is further amplified by the industry’s shift toward next-generation positive electrode materials, such as single-crystalline NMC particles, which have demonstrated reliable mechanical and electrochemical stability. Their intact grain structures help prevent cracking and degradation during cycling, offering improved structural resilience and extended lifetime^{19–23}.

The synthesis of single-crystalline NMC introduces new layers of complexity. Various sophisticated synthesis techniques such as temperature swing¹⁹, multi-step sintering^{24,25}, the use of sintering aids^{26–28}, and molten salt methods^{9,29} have been developed to tailor the particle morphology. However, these methods often face challenges related to scalability, reproducibility, and process complexity. During sintering, NMC particles undergo rapid and spatially heterogeneous transformations driven by the interplay of lithium diffusion, phase transitions, and thermal decomposition^{30–32}. These dynamic changes significantly influence the final material properties, yet the mechanisms underlying grain nucleation, growth, and coalescence remain poorly understood.

Advanced in situ characterization could potentially bridge this gap^{33–35}. Park and colleagues used heated transmission electron microscopy and in situ X-ray diffraction (XRD) to reveal that NMC synthesis is governed by the interplay between intrinsic thermal decomposition of the precursor and lithium incorporation at particle interfaces³⁶. This interplay results in spatially heterogeneous intermediates that affect the final electrochemical properties of the material. However, in situ investigations beyond the atomic scale, especially those using multimodal characterization tools to track morphology

evolution, remain scarce. Directly observing the dynamic evolution of morphology and heterogeneity during sintering is crucial for uncovering the mechanisms behind single-crystalline NMC formation and for establishing scalable protocols that produce robust, defect-resistant structures.

In this study, we aimed to open the black box of traditional solid-state synthesis. Using advanced synchrotron imaging, we directly visualize the sintering dynamics and crystal growth processes that lead to single-crystalline NMC. Using $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ (NMC532) as a model system, we employed a sintering approach based on co-precipitated precursors, Li_2CO_3 , and a Ba-based sintering aid³⁷. The synthesis process was monitored using high-resolution XRD and in situ micro- and nano- tomography, coupled with ex situ nano-resolution X-ray fluorescence (XRF) imaging. These state-of-the-art characterization tools enabled real-time tracking of morphological and structural evolution across multiple length scales. We performed a holistic investigation on individual particles, particle clusters, and powder mounds to reveal how local structural features evolve throughout the entire process. The results show that particle fusion, pore evolution, and grain rearrangement are dynamic and spatially heterogeneous, with recrystallization and densification strongly influenced by particle geometry, packing density, and local thermal environments. Collectively, these factors can lead to persistent structural inconsistencies in the final product, posing potential challenges for long-term electrochemical stability. Our findings provide mechanistic insights into solid-state synthesis and establish a scalable strategy for producing structurally coherent and high-performance single-crystalline positive electrode materials for LIBs.

Results

Compared with polycrystalline counterparts, the synthesis of single-crystalline layered oxides presents additional challenges due to the

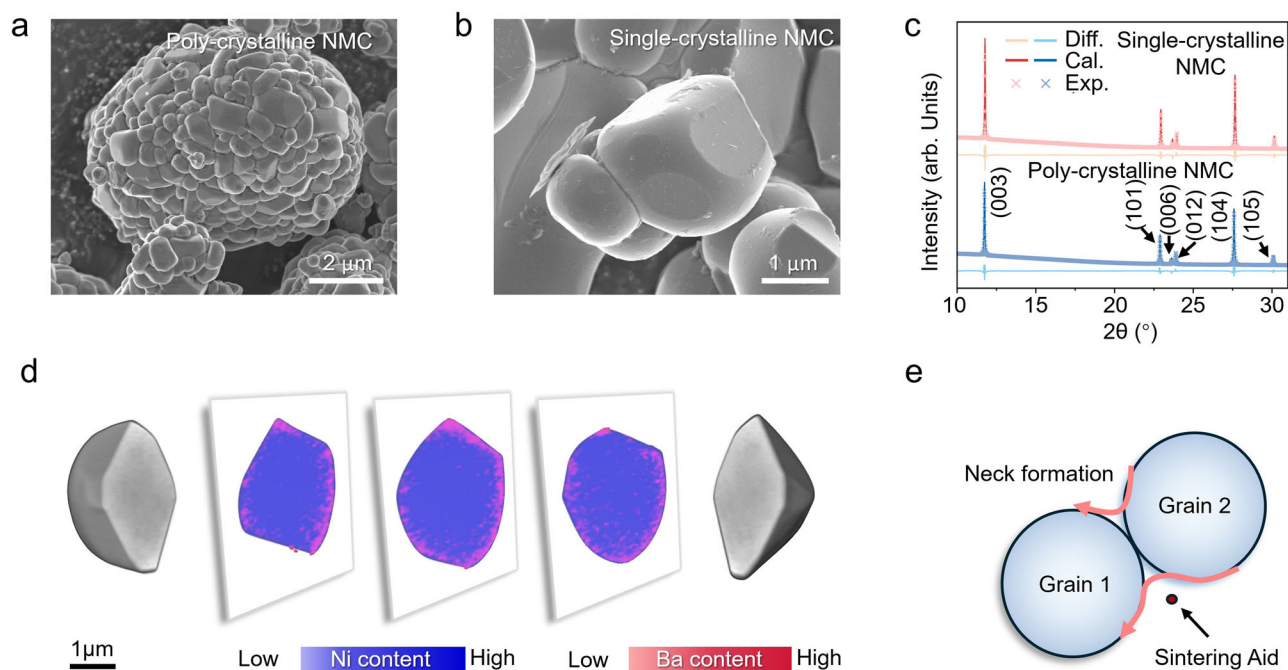


Fig. 2 | Physical and structural characteristics of polycrystalline and single-crystalline NMC particles. **a,b**, Scanning electron microscopy (SEM) images of polycrystalline (**a**) and single-crystalline (**b**) NMC particles. The polycrystalline sample exhibits aggregated primary grains with abundant grain boundaries, while the Ba-assisted single-crystalline counterpart shows smooth surfaces and continuous crystal facets. **c** Synchrotron X-ray diffraction patterns of both samples, indexed to the R-3m layered structure, showing similar crystallographic phase

despite morphological differences, measured at an incident photon energy of 12.7 keV ($\lambda = 0.9763 \text{ \AA}$). **d** Nano-resolution X-ray fluorescence tomography mapping of a single-crystalline particle, indicating a homogeneous distribution of Ni and preferential enrichment of Ba near the particle surface. **e** Schematic illustration of Ba-facilitated intergranular mass transport, where Ba accumulates at grain boundaries and promotes grain fusion by enhancing atomic diffusion during sintering.

inherently polycrystalline nature of co-precipitated precursors. Conventional solid-state sintering tends to preserve this morphology, necessitating specialized thermal protocols to facilitate the transformation into single crystals. Several strategies have been developed to achieve this goal, including molten salt synthesis^{29,38}, high-temperature treatments, temperature-swing protocols¹⁹, and the use of sintering aids^{27,28}. Among them, sintering-aid-assisted approaches are particularly attractive due to their simplicity, low additive content, and cost-effectiveness, often without requiring washing processing steps to remove the extra molten salt. Certain sintering additives can not only promote the transition to single-crystal morphology but also act as dopants to enhance electrochemical performance³⁹.

In this study, we employed Ba as a sintering aid, based on prior reports indicating its dual function in promoting grain growth and improving cycling stability through lattice incorporation³⁷. Under identical sintering conditions, the Ba-containing precursor yielded single-crystalline NMC532 after calcination (Fig. 2a), whereas the undoped precursor retained a polycrystalline morphology (Fig. 2b). Synchrotron X-ray powder diffraction results (Fig. 2c) confirm that trace Ba incorporation does not significantly alter the crystal structure. Both the polycrystalline and single-crystalline materials exhibit a well-defined layered structure with an R-3m space group, and their lattice parameters remain unchanged (Table S1). Electrochemical testing of 0.2 Ah pouch cells paired with graphite negative electrode revealed distinct performance differences (Fig. S1). After 300 cycles, the single-crystalline NMC retained 97% of its initial capacity, in contrast to only 89% for the undoped, polycrystalline material. Coin cell tests further supported this observation, as shown in Figs. S2, S3. Notably, the single-crystalline sample exhibited more stable cycling stability at both 4.3 V and 4.5 V cutoff voltages. The observed enhancement is attributed to the improved chemical and mechanical robustness conferred by the single-crystalline particle structure, which is believed to

mitigate intergranular cracking and structural degradation. To further investigate the role of Ba additive in microstructural evolution during synthesis, we employed nano-resolution x-ray fluorescence (XRF) tomography using a nano-focused hard x-ray probe to reconstruct three-dimensional elemental maps with a spatial resolution of ~30 nm. Interestingly, the sintering aid was found to be preferentially enriched along particle boundaries, with evidence suggesting the formation of a local solid solution with transition metals near these interfacial regions (Fig. 2d). Given that the precursors consist of polycrystalline aggregates, intergranular contact during high-temperature sintering likely initiates mass transport along grain boundaries, ultimately leading to grain coalescence and boundary fusion. These results align with prior hypotheses that sintering additives may segregate to interfacial regions and promote grain fusion by facilitating local diffusion. Trace amounts of the additive that accumulate in neck regions between adjacent grains, reducing the energy barrier for interfacial mass transport and enhancing atomic mobility across primary grains (Fig. 2e). Such additive-assisted mass transport likely accelerates Ostwald ripening and contributes to the formation of single-crystalline particles. However, the dynamic evolution remains poorly understood, and further time-resolved studies are needed to guide the process optimization^{40–42}.

To investigate the chemical dynamics during sintering, thermogravimetric analysis (TGA) was performed on two systems, precursor and Li_2CO_3 mixtures with and without the sintering aid, to uncover the thermal profile during synthesis. As shown in Fig. 3a (additional details in Fig. S4), both samples exhibited similar thermal events. A pronounced weight loss near 335 °C (Fig. 3b) corresponds to the decomposition of $\text{TM}(\text{OH})_2$ and the structural transition from layered to rock salt-type phases^{30,33,43}. Interestingly, the introduction of a sintering aid catalyzed this decomposition at slightly lower temperatures, as evidenced by a leftward shift in the TGA curve. A similar trend was

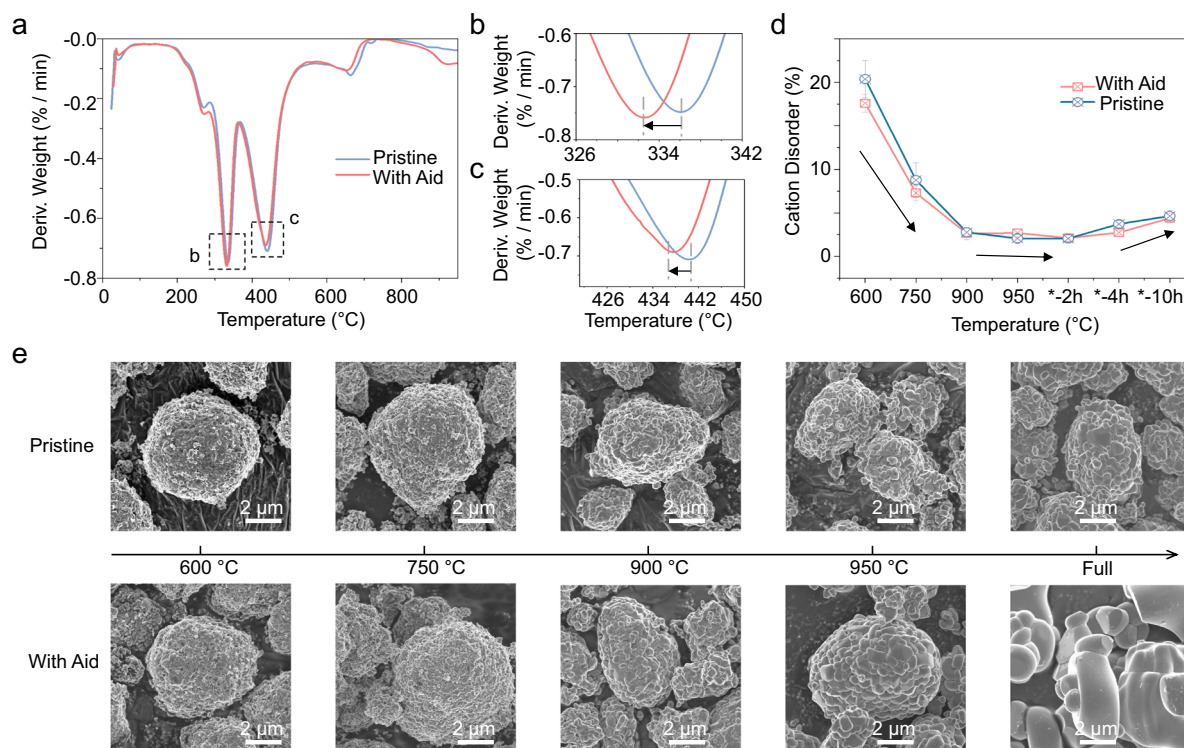


Fig. 3 | Comparison of polycrystalline and single-crystalline NMC evolution during sintering. **a–c** DTG curves show that the Ba sintering aid lowers decomposition temperatures, promoting earlier reaction onset. **d** Cation disordering decreases during temperature ramping and re-emerges with prolonged high-temperature dwelling; the Ba sintering aid has little effect on this trend. Error bars

denote the uncertainty associated with the structural parameter fitting process. **e** SEM images reveal that the undoped sample retains polycrystalline features, while the sample with Ba sintering-aid eventually transforms into single-crystalline particles.

observed at ~440 °C (Fig. 3c), where the decarbonation of Li_2CO_3 and lithium incorporation occurred³⁶. These results suggest that the sintering aid facilitates both transitions at reduced temperatures, potentially by modifying local chemical environments or enhancing interfacial ion transport between precursor particles. Such catalytic behavior could lower the energy barrier for phase transitions, enabling earlier onset of decomposition and lithium incorporation during sintering. While the major lithium-related reactions, such as hydroxide decomposition and carbonate decarbonation, are mostly completed below 600 °C, the exposure to higher temperatures primarily drive the reorganization of the lattice structure and particle morphology.

This stage includes atomic-scale processes like cation disordering, which plays a crucial role in determining the structural integrity and electrochemical reversibility of layered oxides⁴⁴. To investigate these changes, we carried out synchrotron XRD measurements (Figs. S5–S6), which allow direct tracking of crystallographic evolution. The results show that at 600 °C, both samples completed the lithiation process and formed well-defined layered structures. As illustrated in Fig. 3d, the level of cation disordering continues to decrease between 600 and 900 °C. However, additional heating and dwelling at 950 °C led to a gradual increase in cation disordering. These observations point to three key insights: (1) as the temperature increases toward the sintering target of 950 °C, the material transitions from a disordered to an ordered layered phase; (2) prolonged exposure to high temperatures can reverse this ordering transition; and (3) the addition of a sintering aid has minimal impact on cation ordering during heating.

Another important aspect is the morphological evolution, commonly examined using ex situ scanning electron microscopy (SEM). Figure 3e shows SEM images of samples quenched at various stages of the sintering process. During heating from 600 to 950 °C, it appears that both samples retained their polycrystalline nature without any significant morphological evolution. Upon reaching the high-

temperature dwelling stage at 950 °C, the system containing the Ba sintering aid underwent a transition from polycrystalline to single-crystalline morphology, whereas the undoped sample remained unchanged. This is somewhat puzzling, as it contradicts the observation that significant lattice reconstruction occurs within the lower temperature range. Additionally, high-resolution SEM images revealed localized material redistribution near grain boundaries on the particle surfaces, providing further experimental evidence supporting the hypothesis of enhanced mass transport at sintering necks at a lower temperature (Fig. S7).

These ex situ SEM samples were rapidly quenched during sintering to preserve their nonequilibrium states. However, the resulting microstructures may not accurately reflect the true thermodynamic and kinetic pathways present under in situ conditions. Moreover, SEM is inherently limited to capturing surface morphology due to the shallow penetration depth of the electron probe. A more complete understanding of these processes requires real-time, in situ, and three-dimensional observation under operational synthesis conditions. As a result, we conducted in situ nano-tomography of the precursor mixtures under controlled sintering conditions. Building on traditional transmission X-ray Microscopy (TXM), we utilize a micro-furnace on the sample holder as a heat source, allowing for in situ synthesis of single-crystalline NMC materials while using X-ray microscope to monitor the transformation (as shown in Fig. S8). This setup enabled us to observe the 3D morphological evolution at both the particle and particle cluster scales, providing insights that are otherwise inaccessible. As shown in Fig. 4a, at 600 °C, the sample retains the morphology of the precursor (also see Fig. S9), showing particle sizes of 1–2 microns, despite TGA indicating that lithiation has already taken place at this stage. At this temperature, the sample exhibits low crystallinity, evident from the weak and broadened XRD peaks (Fig. S10). As the temperature rises to 750 °C, the sample undergoes a transformation

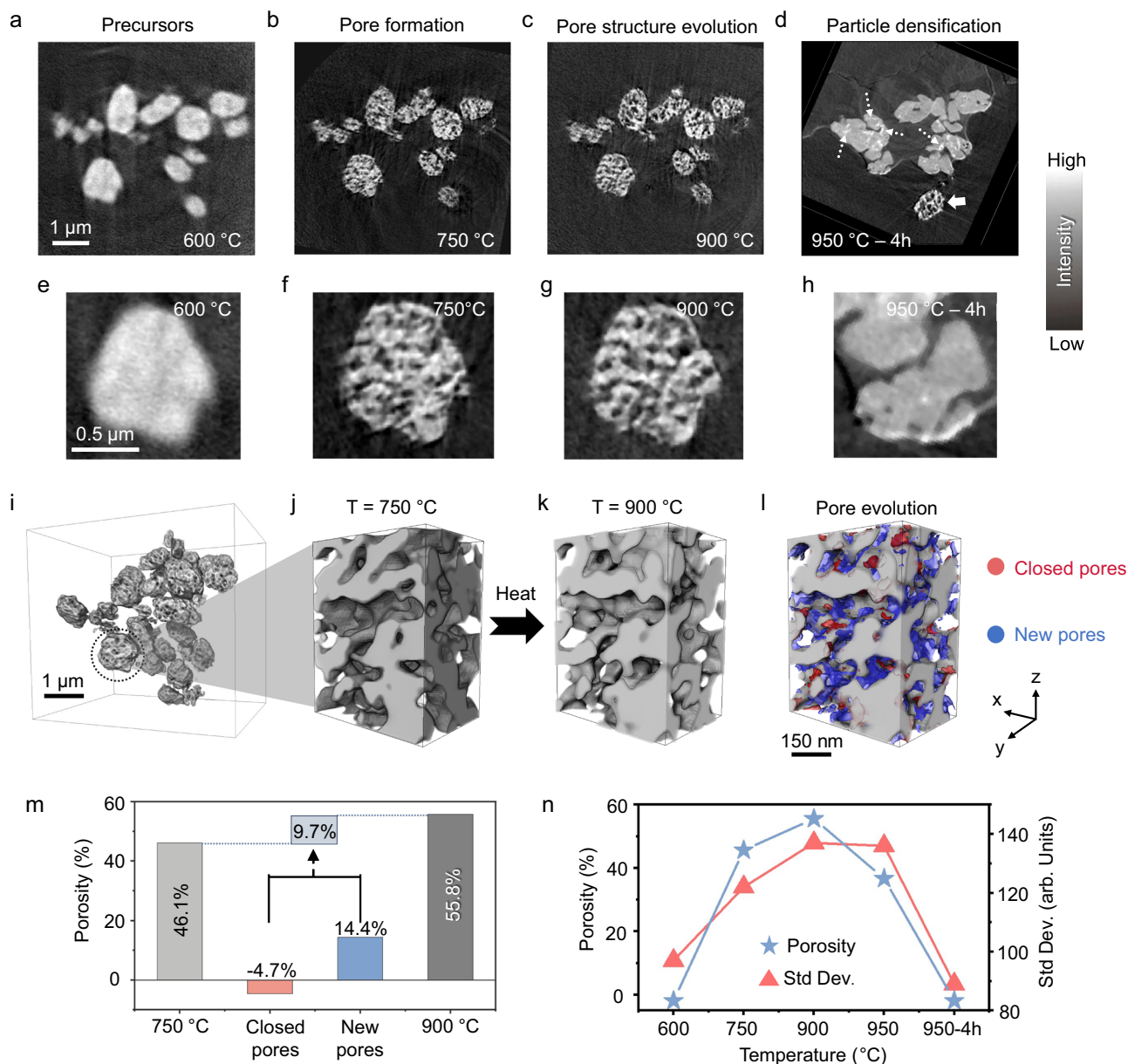


Fig. 4 | Morphological evolution of single-crystalline NMC particle clusters during sintering. Nano-tomographic slices of a single-crystalline NMC particle cluster at 600 °C (**a**), 750 °C (**b**), 900 °C (**c**), and after isothermal hold for 6 h (**d**). Dashed arrows in (**d**) indicate dopant segregation, while the bold arrow marks a particle that did not transform into a single-crystalline structure. **a–d** share a common scale bar. **e–h** Enlarged views of a selected particle from (**a–d**), showing progressive changes in grain boundary fusion, densification, and internal structure at various sintering stages. **e–h** Share a common scale bar. **i** 3D rendering of the

imaged particle cluster at 750 °C. **j, k** Internal structure of a selected particle (highlighted by the dashed circle in (**i**)) at 750 °C (**j**) and 900 °C (**k**). **l** Visualization of pore evolution within the region shown in (**j, k**), with blue regions representing newly developed pores and red regions indicating closed pores. **j–l** share a common scale bar. **m** Quantitative analysis of pore volume changes within the sub-particle volume analyzed in (**j, k**). **n** Evolution of porosity and standard deviation of optical intensity across the material at different sintering stages.

into a porous structure, with signs of recrystallization occurring within the secondary particles. This corresponds to the thermal event observed at 700 °C in the TGA profile and aligns with an increase in XRD peak intensity, signaling improved crystallinity. The transition to single-crystalline particles takes place during the dwell period at the target temperature of 950 °C (Fig. 4d). Throughout this period, particle boundaries fuse, grains merge, and the internal porosity diminishes. After the hold period, density inconsistencies can be observed at the grain boundaries, likely due to compositional segregations, consistent with observations from XRF tomography (Fig. 2d). Interestingly, even after the 6-hours dwell period, some particles still failed to transform into the single-crystalline formation. These failed particles

tend to be more isolated from others, suggesting that mass transport processes vary from particle to particle, leading to heterogeneity at the particle cluster scale. Although there is no significant macroscopic change in the 750–900 °C temperature range, localized cross-sections reveal evolving pore structures within individual particles, indicating progressive internal transformations. Registration and segmentation of 3D nano-tomography data allow for more accurate and quantitative analysis of the pore evolution. In Fig. 4i–l, cross-sectional views and porosity calculations of the same particle reveal that, while the overall external morphology remains similar, notable changes occur in internal pore structures as the temperature increases. Specifically, some larger pores expand, while smaller pores close, likely due to mass

transport and redistribution of primary particles during heating. Overall, the pore volume fraction increases from 46.1% to 56.8% between 750 °C and 900 °C. This change involves the closure of 4.7% of the original pores (red) and the development of new pores at 14.4% volumetric ratio (blue). Despite the absence of single-crystal formation at this stage, mass transport and stress relaxation are already underway, driving pore evolution and affecting local structural heterogeneity. These early transformations play a critical preparatory role, establishing morphological and energetic conditions that are essential for achieving long-range crystallographic coherence and, ultimately, for ensuring the structural integrity and electrochemical stability of the final material. In Fig. 4n, the quantitative analysis of each set of 3D tomogram reveals that, during sintering, particles initially in a uniform, lithiated state undergo recrystallization, leading to increased porosity. At first, the pore volume rises with increasing temperature but subsequently decreases as more pores close, ultimately forming single-crystalline NMC particles. By tracking changes in internal particle density, we observe that this process introduces heterogeneity, which arises due to variations in particle location, morphology, size, and dopant distribution. This density heterogeneity initially intensifies as the temperature rises, but as heating continues, increased mass diffusion and reaction kinetics drive the material toward a stable equilibrium, resulting in a dense single-crystalline structure. This is consistent with prior studies on additive-assisted sintering, such as molten salt synthesis. For example, Chen-Wiegart et al. showed that enhanced mass transport, pore coarsening, and ligament evolution in molten salt environments promote single-crystal formation⁴⁵. In our Ba-assisted system, similar mechanisms apply: Ba segregates to interfacial regions, accelerates intergranular diffusion, and facilitates grain coalescence. These pore transformations are thus not merely morphological artifacts but are essential intermediate steps toward single-crystal conversion.

The particle-to-particle inconsistencies observed within a particle cluster prompted us to investigate the dynamics at a larger scale during the single-crystalline NMC positive electrode material synthesis. To achieve this, we conducted in situ studies on powder mound dynamics using synchrotron-based micro-tomography. In this experimental setup, we positioned several Infrared lamps around the sample as heat sources and placed a thermocouple directly beneath the powder mixture of lithium salt, precursor, and Ba additive to achieve an accurate temperature calibration (see Fig. S11 for experimental setup). The as-loaded sample at room temperature (~25 °C) serves as the pristine state for micro-CT analysis. At this stage, the powder mound exhibits no signs of densification or local reaction, as confirmed by the uniform contrast and narrow density distribution shown in Fig. S12. Starting from this initial state, this configuration enabled real-time 3D monitoring of morphological evolution of the powder mound during sintering. In Fig. 5a, cross-sectional views in the yz direction reveal that, overall, the volume of the powder mound decreases while its density increases as the temperature rise. However, this densification process exhibits pronounced heterogeneity. At 600 °C, a localized collapse and densification first appears on the left side of the mound. This effect becomes more prominent at 800 °C, where scattered densification regions within the powder mound initiate internally and then spread outward. During the high-temperature hold, the powder mound morphology continues to evolve, indicating ongoing particle movement even at this stage.

Overall, the synthesis of single-crystalline NMC leads to significant densification at the powder mound scale, driven by particle rearrangement. This relative particle movement is influenced by forces generated during melting of the lithium salts, recrystallization, morphology evolution, and grain boundary merging within and among individual particles, as observed via nano-tomography. Additionally, gas diffusion forces arising from precursor dehydration and Li₂CO₃ decarbonation in early reaction stages contribute to this densification

effect. Notably, different parts of the powder mound exhibit varied movement directions and rates, even at the same temperature. This process appears to proceed via two distinct patterns. The first is localized, near-surface densification onset, which then extends forward (Fig. 5b). This behavior can be attributed to the proximity of surface particles to the heat source, likely subjecting them to higher local temperatures and allowing them to react sooner than the overall chemical reaction. Moreover, the surface offers more favorable diffusion conditions for gases produced during dehydration and CO₂ release, further accelerating reaction kinetics. The second is the initiation of internal densification at scattered, randomly distributed sites within the bulk, which then propagates outward (as shown in Fig. 5c). This scattered propagation is likely due to the obstruction of heat transfer and gas diffusion as surface densification progresses. Particles near existing pore channels retain more favorable reaction conditions, allowing them to begin reacting. As illustrated in the 800 °C cross-section, these scattered onset locations are more likely to occur near these channels, where the local configurations facilitate the onset of the reaction. The onset location could also correlate with the uniformity of Li and precursor mixing, dopant segregation at the powder mound scale. These two distinct reaction patterns collectively govern the collapse of the powder mound, resulting in the formation of new macroscopic pore channels, as shown in Fig. 5d. These newly formed pore channels can influence gas diffusion kinetics, modulating the downstream reaction pattern developments. The structural details of the heated powder mound under these interacting regulatory effects are shown in Fig. S13. The powder mound's overall density progressively and monotonically increases throughout the sintering process. As illustrated in Fig. 5e, which tracks relative density evolution, the most significant density change does not occur during the lithiation process (below 600 °C) but rather during the 600–900 °C heating phase. This window corresponds to the recrystallization, porosity reconstruction, and particle densification. Notably, at 800 °C, the powder mound's density distribution exhibits a distinct multimodal pattern. And this multimodal distribution gradually transitions into a unimodal form as the sintering process reaches completion. Overall, as shown in Fig. 5f, the powder mound's volume progressively decreases while its density increases throughout the sintering process. The reaction heterogeneity initially intensifies, peaking around 800 °C, before gradually diminishing.

Discussion

In this study, we investigate the synthesis process for single-crystalline NMCs32 aided by trace amount of Ba additive. We applied a multi-scale approach to reveal the dynamic morphology evolution and its underlying mechanisms. This sintering process, as illustrated in Fig. 6a, involves complex, multi-scale phenomena resulted from intertwined physical and chemical transformations that ultimately determine the formation and performance of the final product.

At temperatures below 600 °C, significant solid-state reactions occur, forming a layered structure with poor crystallinity. As the temperature rises above 600 °C, notable lattice-level transformations take place, including the suppression of cation disordering and the release of lattice strain. This initiates a recrystallization process that coincides with internal morphological changes. During this stage, internal pores form and evolve with increasing temperature. In the presence of Ba-based sintering aid, these pores undergo gradual coarsening, followed by progressive closure during the isothermal hold at 950 °C. This sequence of events ultimately drives the transition to a single-crystalline particle structure.

These transformations are both heterogeneous and asynchronous, influenced by factors such as dopant distribution, interparticle spacing, and precursor morphology, each of which can impact the success of single-crystal formation. At the powder mound scale, non-equilibrium densification proceeds via two distinct reaction modes:

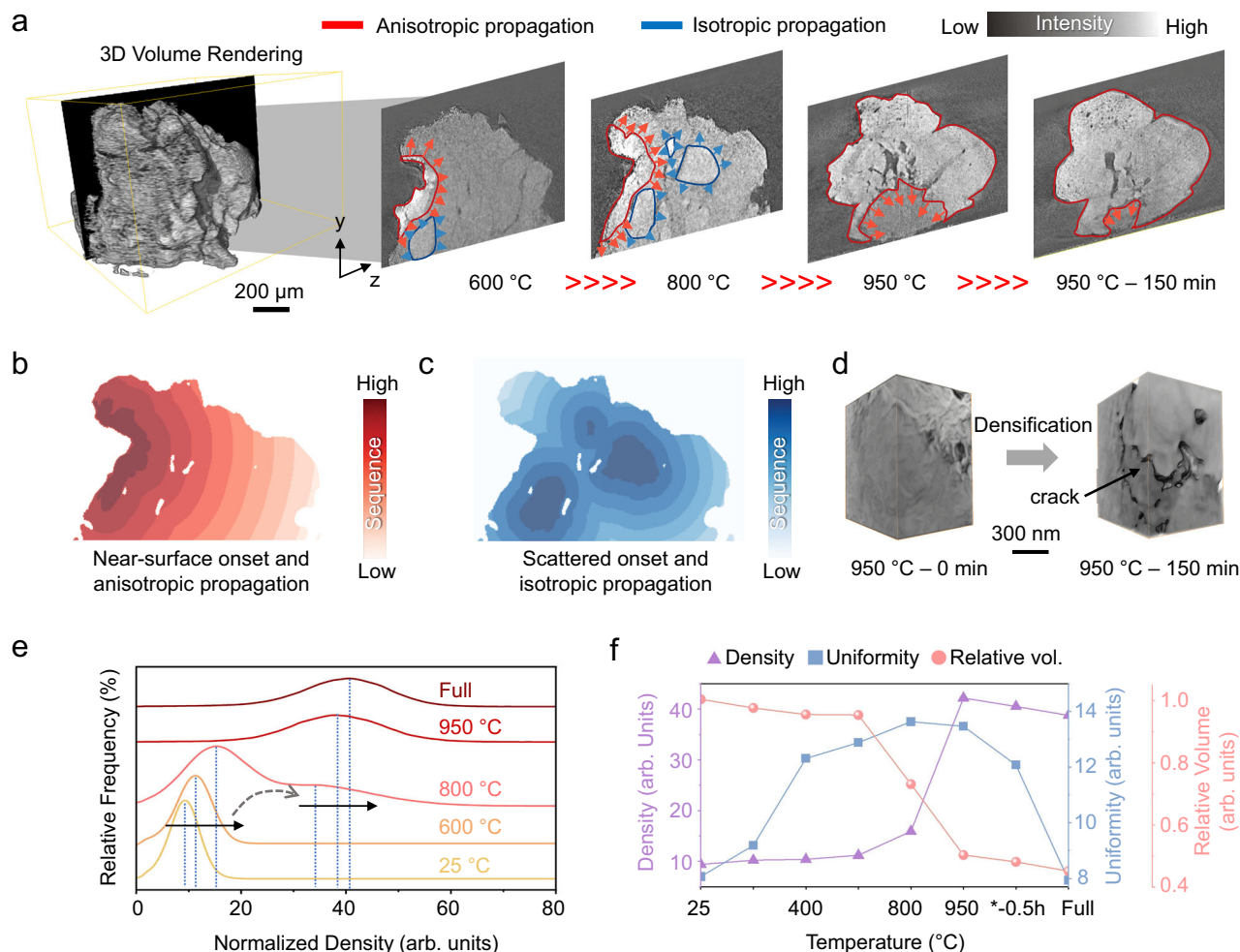


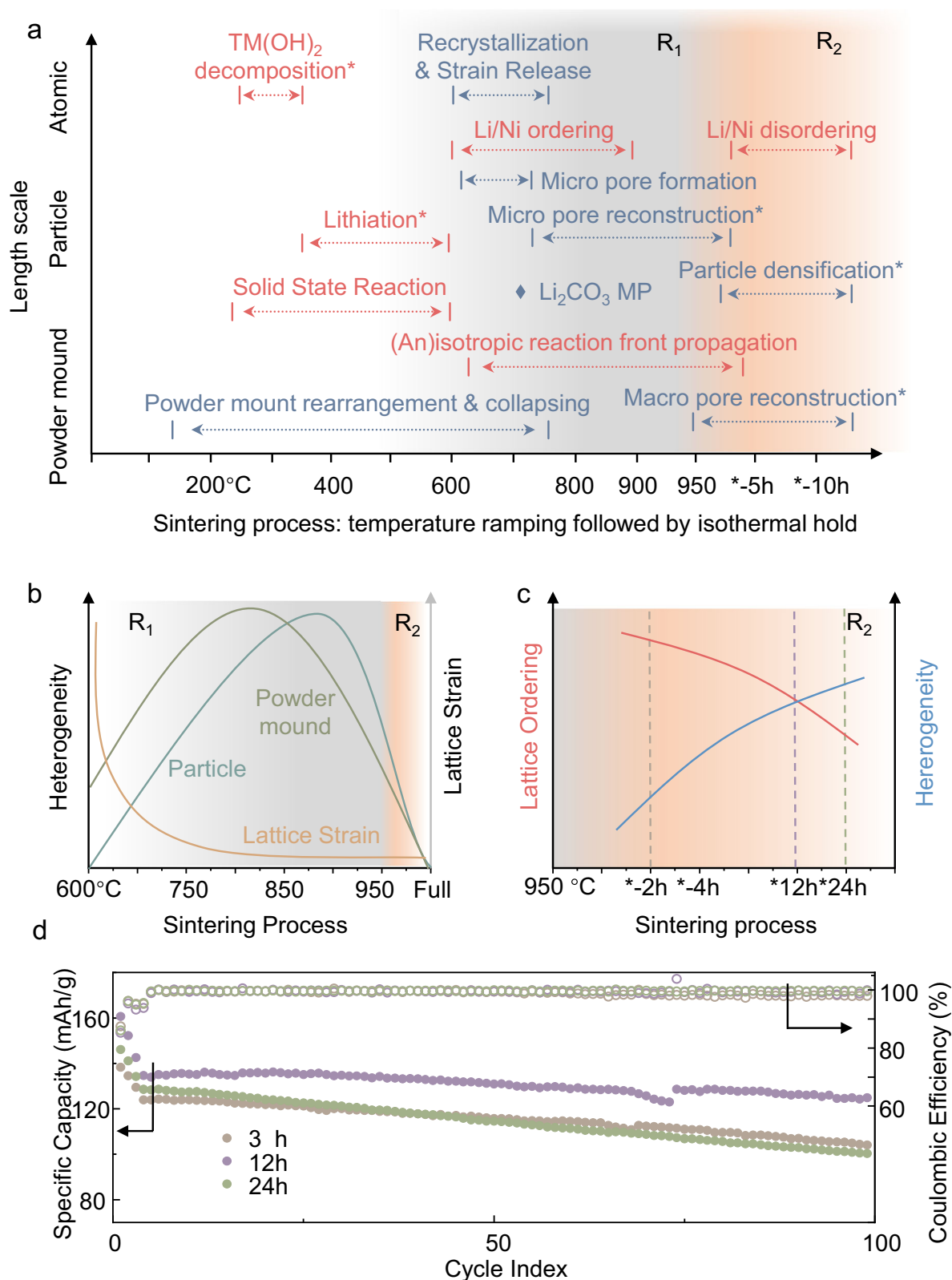
Fig. 5 | Structural evolution of single-crystalline NMC at the powder mound scale during sintering. **a** 3D rendering of the imaged powder mound and an arbitrarily selected yz slice at 600 $^{\circ}\text{C}$, 800 $^{\circ}\text{C}$, 950 $^{\circ}\text{C}$, and after isothermal hold. Schematics of two distinct reaction patterns: **(b)** near-surface onset followed by anisotropic propagation and **(c)** scattered onset followed by isotropic outward propagation. **d** Zoomed-in 3D rendering reveals local densification and macroscopic crack formation. **e** Density histograms at different sintering stages. The

x-axis represents the rescaled 8-bit grayscale intensity of the reconstructed CT volumes (0–255). Original datasets were acquired as 16-bit volumes (0–65536) and uniformly rescaled to 8-bit prior to analysis to ensure consistency across temperatures. Histograms were calculated using the rescaled 8-bit voxel intensities. **f** Quantitative analysis of the powder mound's relative density, uniformity and volume at different sintering stages.

(1) a near-surface onset followed by anisotropic inward propagation, and (2) a scattered internal onset followed by isotropic outward progression. These densification pathways are governed by reaction kinetics and particle mobility, which in turn are modulated by the homogeneity of Li-precursor mixing, local porosity, and the packing configuration of the mound. Such mesoscale factors play a critical role in determining the uniformity and quality of the final sintered product.

Interestingly, as shown in Fig. 6b, there is a temperature-dependent coupling in heterogeneity and lattice strain. Across scales, the degree of heterogeneity first increases, peaking between 800–900 $^{\circ}\text{C}$, before declining during the isothermal hold at 950 $^{\circ}\text{C}$. The lattice-level strain appears to be decreasing monotonically. This trend may favor a prolonged isothermal hold, however, a closer look at the transformations during the extended isothermal hold (see schematics in Fig. 6c) would suggest otherwise. On the one hand, prolonged isothermal hold at 950 $^{\circ}\text{C}$ indeed promotes the morphological transformation from polycrystalline particles to single-crystalline particles, where grain growth is driven by mass transfer, grain coalescence, and pore closure. On the other hand, over-sintering will lead to Li/Ni cation disordering and mixing, which are detrimental to the

battery rate capability. In our experiments, this phenomenon was validated by holding the sintering process at 950 $^{\circ}\text{C}$ for 3 hours, 12 h, and 24 h. The resulting samples were subjected to electrochemical tests as shown in Fig. 6d (more details could be access in Figs. S14–16). The electrochemical tests reveal that, compared to the samples held for 3 h and 24 h, the 12-h isothermal hold resulted in superior cycling stability and higher electrochemical capacity under both voltage conditions. As mentioned earlier, the isothermal holding process is a multi-scale heterogeneity reduction process. A short holding time fails to eliminate heterogeneity and does not complete the single-crystallization process. Conversely, for the 24-hour sample, while prolonged holding provides sufficient time for heterogeneity reduction and complete single-crystallization, the extended thermal exposure, however, increases Li/Ni cation disordering, which hinders lithium-ion diffusion and ultimately reduces the electrochemical capacity. Crucially, these transformations are interdependent and tightly coupled, defining a narrow processing window for optimal performance. Recognizing and managing this competition offers a mechanistic basis for tailoring sintering protocols, and underscores the broader importance of controlling multi-scale dynamics during synthesis.



While this work focused on the synthesis of NMC, the approach we use to uncover it, i.e. the multiscale, time-resolved, in situ tracking of structural evolution, serves as a generalizable strategy for decoding synthesis–structure–property relationships in a wide range of functional materials. In systems as diverse as Ni-rich layered oxides, Li-rich disordered rock salts, or even solid electrolytes, similar competitions exist between beneficial and detrimental transformations across length

scales. The strength of our methodology lies not in resolving one particular conflict, but in providing a framework to identify, observe, and ultimately manage these hidden interactions. As such, the “competition” between crystallization and disordering in NMC should be viewed not as an endpoint, but as a demonstration of how to open the black box of solid-state synthesis, a toolset that can be extended across chemistries to inform rational design and accelerate synthesis optimization.

Fig. 6 | Temperature-dependent physical and chemical processes during NMC sintering. **a** Schematic summary of key events across different length scales (atomic, particle, and powder mound) during sintering, illustrating the temporal sequence of reactions and morphological transitions. Red indicates chemical transformations, blue represents physical changes, and the asterisks denote the processes in which Ba doping played a significant role. Two distinct regimes are defined: R1, corresponding to the temperature-dependent physical and chemical evolution during the heating ramp, and R2, representing the time-dependent structural and morphological adjustments during the isothermal holding stage. **b** Evolution of heterogeneity and lattice strain across the sintering process. Heterogeneity initially increases due to concurrent multi-scale transformations during R1 and later decreases during R2 as recrystallization and densification proceed.

c Competing trends of lattice ordering (red) and particle densification (blue) during isothermal holding. Prolonged sintering leads to partial cation disordering, while particle morphology continues to densify. Vertical lines indicate samples corresponding to different isothermal holding durations (3 h, 12 h, and 24 h). The 3 h sample exhibits lower cation disordering but higher heterogeneity, whereas the 24 h sample shows higher cation disordering but lower heterogeneity; the 12 h condition represents an optimized balance between minimized heterogeneity and suppressed disordering. **d** Electrochemical cycling performance in a Li||NMC coin cell of samples sintered at 950 °C for 3 h, 12 h, and 24 h, respectively. Optimized isothermal hold improves structural ordering and densification, yielding enhanced capacity retention.

Looking forward, this work reinforces the central role of synthesis in determining the structure–property relationships that underpin battery performance. In many cases, materials discovery is limited not by a lack of promising chemistries, but by an incomplete understanding of how processing governs structure formation across multiple scales. By opening the black box of solid-state synthesis, we can move beyond empirical optimization to a quantitative, feedback-informed science, which may ultimately enable the development of predictive models that link thermal histories, reaction pathways, and microstructural outcomes.

Methods

Synthesis

The poly-crystalline and single-crystalline samples were prepared using a high-temperature solid-state sintering method. The precursor, with the molecular formula $\text{Ni}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}(\text{OH})_2$ (GEM Co., Ltd.), along with Li_2CO_3 (99.5%, Sichuan Tianqi Lithium Industry Co., Ltd.) and $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$ (98%, Sinopharm Chemical Reagent Co., Ltd.), was mixed at a Li/TM ratio of 1.06 (where TM represents transition metal elements) and a Ba^{2+} concentration of 0 ppm (poly-crystalline) or 5000 ppm (single-crystalline). Prior to heat treatment, the powders were thoroughly mixed using an agate mortar and pestle for 30 min to ensure compositional homogeneity. The mixtures were first preheated in a tube furnace at 600 °C for 5 h to ensure homogeneous reaction between the lithium salts and precursors. Subsequently, high-temperature sintering was conducted at 950 °C for 12 h in ambient air. The heating rate for both steps was maintained at 10 °C·min⁻¹. After the high-temperature dwell, the furnace was allowed to cool naturally to room temperature, corresponding to an average cooling rate of approximately 3–5 °C·min⁻¹ over the 950–200 °C range. Following sintering, the as-obtained products were gently ground and sieved through an 800-mesh screen to remove large agglomerates and ensure uniform particle size distribution. To validate the competing mechanism, additional samples were prepared with isothermal hold durations of 3 h and 24 h for comparison.

Electrochemistry

N-methyl-2-pyrrolidone (NMP, 99.5%, Sigma-Aldrich) was used as the solvent to prepare positive electrode slurry by dispersing the active material (NMC), acetylene black (battery grade, MTI), and polyvinylidene difluoride (PVDF, Solef 5130, Solvay) in a weight ratio of 8:1:1. The solvent-to-solid mass ratio in the slurry was approximately 2.5:1. The slurry was coated onto aluminum foil (thickness: 15 μm, battery grade) by an automatic coater on one side and dried in a vacuum oven at 120 °C for 4 h. The resulting electrode sheet was punched into 12 mm diameter discs, yielding an active material mass loading of approximately 3.5 mg cm⁻² per electrode.

For coin cell assembly, CR2025 half-cells were constructed in an argon-filled glovebox. Stainless steel cases and stainless steel wave springs (MTI Co., Ltd.) were used; the spring constant was not specified by the manufacturer. The electrolyte consisted of a 1 M LiPF₆ (battery

grade, BASF) solution in a mixed solvent of ethyl methyl carbonate (EMC), dimethyl carbonate (DMC), and ethylene carbonate (EC) (1:1:1 by volume). Approximately 60 μL of electrolyte was added to each coin cell. Lithium metal foil (thickness: 50 μm, 99.9%, MTI Co., Ltd.) was used as the counter electrode as received, Celgard 2400 polypropylene separator (thickness: 25 μm) as the separator, and the prepared positive electrode disc as the working electrode. Careful electrode alignment was employed during stacking to ensure uniform electrochemical performance. All current values normalized to the mass of active material are referred to as specific currents. Coin cells were initially activated at a specific current of 0.1 C, 0.2 C, and 0.5 C for three cycles, followed by long-term cycling at 1 C (where 1 C = 150 mAh/g), within a voltage window of 2.8–4.3 V or 2.8V–4.5 V. All electrochemical measurements were conducted at 25 ± 2 °C in a temperature-controlled laboratory environment (no climatic chamber was used). The Coulombic efficiency (CE) was calculated as the ratio of the discharge capacity to the charge capacity in the immediately preceding cycle.

For pouch cell assembly, graphite was used as the negative electrode with a negative-to-positive (N/P) capacity ratio of 1.2. The positive electrode mass loading was approximately 12 mg cm⁻², incorporating 4% PVDF binder and 4% active nano-carbon, in alignment with practical commercial designs. The graphite loading was calculated to be 6.2 mg cm⁻². During cycling, an external stack pressure (~0.3 MPa) was applied to the pouch cells using mechanical clamps. After electrolyte injection, the pouch cells were rested for wetting, followed by a degassing step prior to final heat sealing. Pouch cells were subjected to galvanostatic cycling using a Neware battery test system (CT-4008T-5V20mA-164, Shenzhen, China) at 25 °C. The cycling protocol involved charging and discharging the cells between 2.5 V and 4.2 V.

Characterization

The microstructure of the samples was observed using a field emission scanning electron microscope (ZEISS, Gemini 500). Synchrotron X-ray diffraction measurements were conducted at the 11–3 beamline of the Stanford Synchrotron Radiation Lightsources. An energy beam of 12.7 keV was used for the experiments, with a Rayonix 225 detector. The LaB₆ peak was utilized to calibrate the momentum transfer parameter in the diffraction data for each sample. Thermal gravimetric-differential thermal analysis (TG-DTA) curves were collected using a SDT650 with a scanning rate of 5 °C/min between room temperature and 950 °C under an air atmosphere.

Nano-fluorescence mapping procedure

The nano-fluorescence mapping was carried out at the Hard X-ray Nanoprobe beamline (NSLS-II, BNL), where a 9.6 keV X-ray beam was focused into a 30-nm spot using a Fresnel zone plate. For tomographic reconstruction, a dataset consisting of 51 projections was collected across an angular range from –75° to 75° with a 3° step size. Subsequent visualization and analysis of the reconstructed data were performed with the Avizo software package.

In situ X-ray nano-tomography

The Full-field X-ray Imaging (FXI) beamline (18-ID) at the National Synchrotron Light Source II (BNL) was utilized to perform in situ 3D nano-tomography via Transmission X-ray Microscopy. Imaging was conducted with a 32 μm field of view and achieved a spatial resolution of around 40 nm. For each tomographic scan, the sample was rotated through 180° at a constant rate of 4°/s, with an individual exposure time of 0.05 s for each collected projection. During the test, an automated micro-muffle furnace was used to heat the sample, simulating the sintering process. The data reconstruction and spectral fitting were performed using our in-house developed PyXAS software⁴⁶.

In situ X-ray micro-tomography

The experiments were performed at the Advanced Light Source on Beamline 8.3.2. This beamline utilizes a 4.37 T superbend magnet source and a multilayer monochromator to deliver a 25 keV monochromatic X-ray beam. Detection was achieved using a 50 mm LuAG:Ce scintillator and a 10x Olympus objective, coupled with a PCO. For each scan, 2625 images were captured over 180°, with an exposure time of 250 ms.

The precursor and lithium carbonate and Ba salt mixture powder mound was placed on a holder above a thermocouple. The surrounding area was heated using IR lamps, creating a sintering environment. Temperature control during the heating process was achieved by adjusting the voltage applied to the IR lamps, with data collected at various temperatures and heating times.

Data availability

The data supporting the findings of this study are available within the article and its Supplementary Information files. Source data are provided with this paper.

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Competing interests

The authors declare no competing interests.

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