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Thermally-Driven Mesopore Formation and Oxygen Release in Delithiated NCA Cathode Particles

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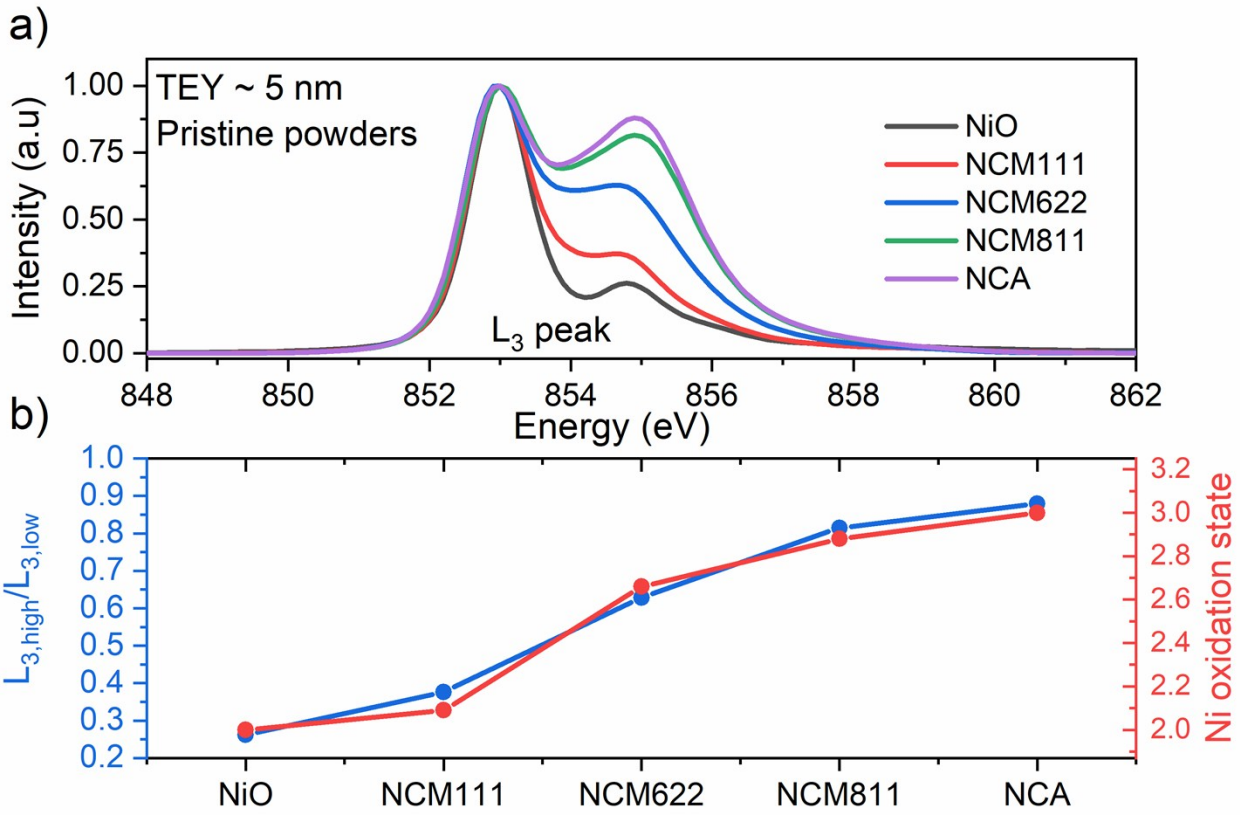

Figure S1: (a) Superimposed soft XAS $L_{3,high}$ and $L_{3,low}$ peaks for various cathode active materials of different Ni content. Ni content correlates with $L_{3,high}$ peak intensity. (b) Correlation of $L_{3,high}/L_{3,low}$ peak ratio with Ni oxidation state.

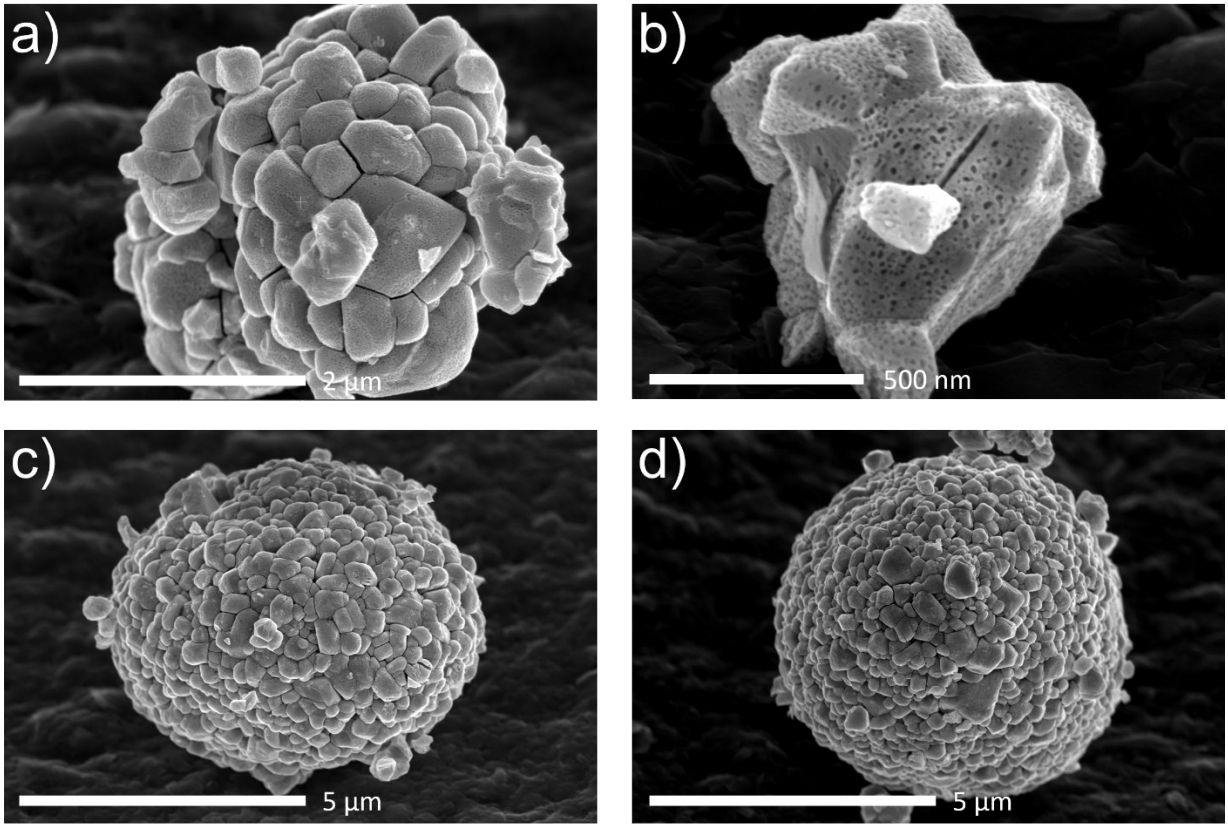

Figure S2: (a-d) Heat-treated $Li_{0.3}NCA$ particles of various sizes showing mesopores on the surface. Independent of particle size, mesopores are homogeneously distributed on the surface of the particles.

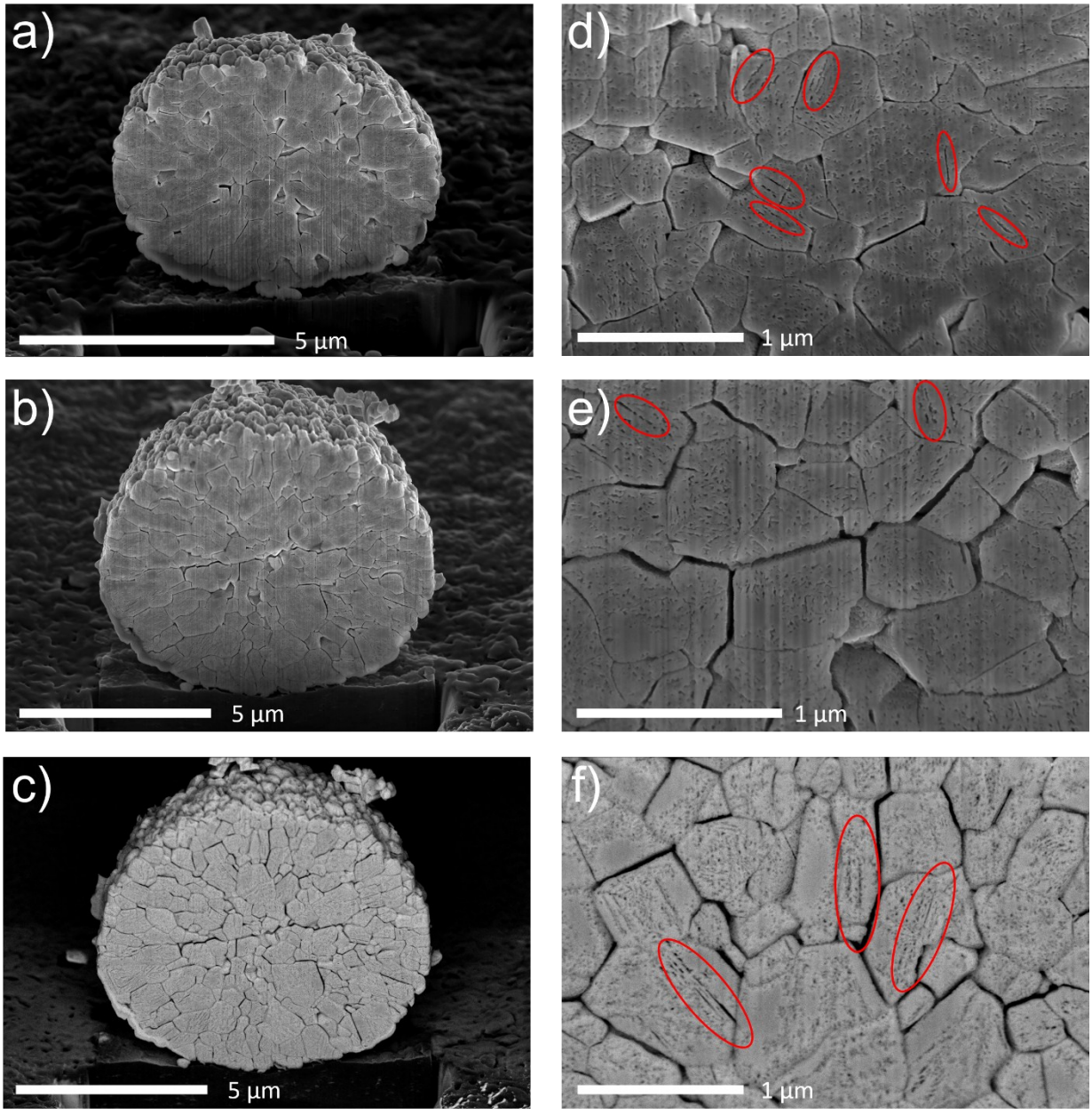

Figure S3: (a-c) Cross-sectional SEM images for four different particles after focused ion beam milling. Cross-sections of particles also indicate a homogenous distribution of mesopores. (d-f) High magnification of cross-sectional images corresponding to the cross-sectional images (a-c). High magnifications show how mesopores appear along intragranular cracks (red circles) and throughout the entire particle.

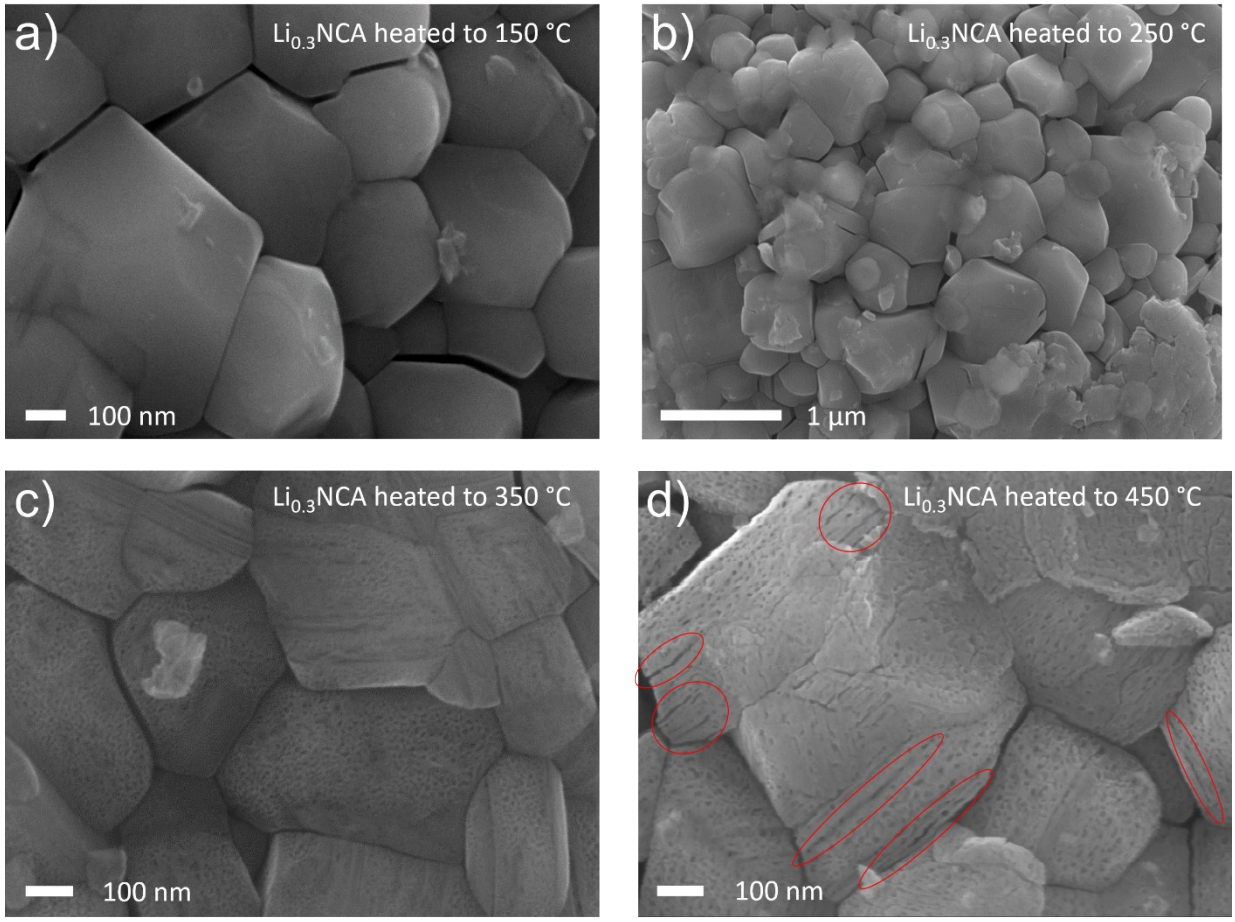

Figure S4: SEM images of the surface of four individual delithiated NCA particle heated to (a) 150 °C, (b) 250 °C, (c) 350 °C, and (d) 450 °C. No mesopores are observed on particles that were heated to 150 or 250 °C. Evolution of mesopores seem to start at temperatures above 300 °C. (d) Mesopores seem to appear along intragranular cracks (red circles).

Video S1: FIB-SEM milling for a delithiated NCA particle.
https://drive.google.com/open?id=19p0T8S6oIhVJ10SBVdG9Jc6xfV2qeU-d

Video S2: FIB-SEM milling for a delithiated and heat-treated NCA particle.
https://drive.google.com/open?id=1qvTCIFQ6VG5DJDPrHSb1tH01axGldaDe

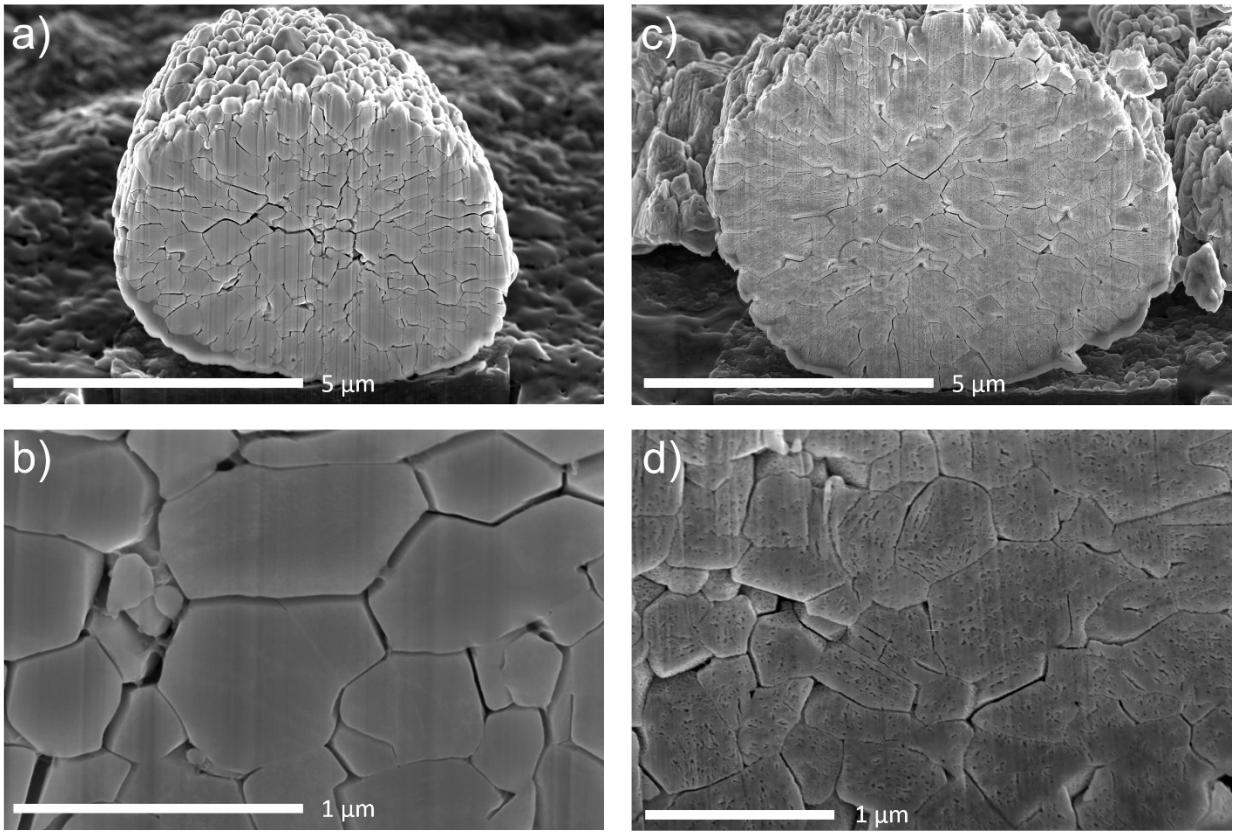

Figure S5: Cross-sectional SEM images for two different particles after focused ion beam milling. (a) Cross-sectional area for a delithiated NCA particle. (b) High magnification of the cross-sectional image shown in (a). (c) Cross-sectional area for a delithiated and heat-treated NCA particle. (d) High magnification of the cross-sectional image shown in (c).