Supporting Information: A FIB-SEM based correlative methodology for X-ray nanotomography and Secondary Ion Mass Spectrometry: An application example in lithium batteries research

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**Sample preparation:** Preparation of the sample material including lift-out and transfer to the substrate was carried out in a Xenon (Xe)-Plasma FIB (Thermo Fisher Helios PFIB G4 CXe) equipped with a micromanipulator (Kleindiek MM3A). The advantage of a Xe-Plasma FIB over the widespread gallium (Ga)-FIB lies in the larger beam current, allowing to keep milling times for large volumes in a useful range. For practical reasons of geometry inside the FIB-SEM it was not possible to cut the pillar perpendicular to its vertical axis and fix it onto the carbon substrate. Instead, cutting was performed under an angle of 38° which is the angle of the ion beam against the stage plane of the instrument used in this study. The preparation consists of three steps (see Figure S1):

1. Cut a plane of ~150 x 150 µm² under an angle of 38° into the surface of the carbon substrate (Figure S1 a &b).
2. For stability reasons and to make the pillar stand out of the surface, a pedestal was cut from a thin needle of stainless steel with ~100 µm diameter. The needle was placed upright on a holder without pre-tilt and no stage tilt was used for the cutting so the angle of the cut face on both ends was again 38° (Figure S1 c & d). This piece was transferred to the previously cut angled area of the substrate using the micromanipulator and fixed with ion beam induced platinum deposition (Figure S1 e & f). The process was adapted from the well-established FIB lift-out preparation of TEM samples.
3. A pillar with a diameter of 50 µm was prepared from the sample volume with annular milling using Xe ion beam currents from 2.5 µA down to 60 nA (Figure S1 g & h). The top edge of the pillar was fixed to the micromanipulator with ion beam induced platinum deposition and cut free from the sample under an angle of 38°, matching the angle of the steel pedestal on the carbon substrate (Figure S1 i). After transfer to the substrate, the pillar was attached to the pedestal using FIB-based deposition and cut free from the manipulator tip (Figure S1 j & k) using ion beam. To increase mechanical stability, further deposition was carried out around the circumference of the pillar.



Figure S1: Step 1: a) Carbon substrate. b) Cut a sloped surface, 38 °, of ~150 µm width

Step 2: c) Stainless steel needle as received. d) Preparation of pedestal for transfer to carbon substrate. Cuts under 38° angle relative to the needle axis to match the angle of target area on the carbon substrate. Only a small connection to the needle is left until the micromanipulator is attached for transfer. e) Micromanipulator with pedestal hovering above the carbon substrate target area. Gas injection needle (GIS) for platinum precursor coming from the right side. f) Pedestal positioned and fixed to the carbon substrate.

Step 3: g) Cathode material glued to a pin. h) Pillar cut from the cathode material ready for lift-out. i) Pillar fixed to the micromanipulator and cut free on the bottom. GIS coming from the left side due to image scan rotation. j) Manipulator with pillar hovering above the pedestal.

Step 4: k) SEM image of the final assembly carbon substrate – steel pedestal – cathode material pillar. l) X-ray transmission image of the assembly. Steel pedestal is clearly visible due to atomic number contrast.

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Figure S2: Video of the reconstructed sample volume which has been analyzed by nanoCT. The diameter of the sample pillar is approximately 50 μm.



Figure S3: Correlative SEM, EDS and SIMS images of the same ROI. The EDS elemental maps of Ni (yellow), Co (blue) and C (green) elucidate the identity of the particles. The correlative SIMS images showing the chemical distributions of 7Li+ (green), 27Al+ (blue) and 59Co+ (red) reveal further details due to its capability of detecting low-Z elements such as Li and nm-scaled lateral resolution. (scale bar = 5 μm)



Figure S4: SIMS chemical maps of the raw cathode sample (pre-milling to pillar shape). 7Li+ = green. 27Al+ = blue. 58Ni+ = yellow. 59Co+ = red. The image on the top right represents an SE-SEM secondary electron image of the same ROI and the image on the bottom right is an overlap of the four individual elemental maps. (scale bar = 10 μm)