Supplementary information to Operando Neutron Depth Profiling to Determine the Spatial Distribution of Li in Li-ion Batteries

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In the paper only analysed NDP spectra are shown. In this supplement a typical spectrum is described in more detail to explain what features are obtained. In the paper a ^{6}Li metal foil on top of copper is used as one of the component for calibrating lithium metal experiments. This straightforward experiment can be described by a step function going from the ^{6}Li concentration in de metal to zero ^{6}Li concentration in the copper window, shown in the inset in figure S1. First step is to account for neutron beam intensity losses, through capture reactions neutron density decreases with depth causing the detection efficiency to reduce for lithium ions located deeper within the sample. This effect is indicated in the inset of figure S1 where the measurable concentration decreases with increasing depth compared to the blue initial concentration step function. Note that in most cases this effect is only minor but for samples with a high ^{6}Li concentration this becomes relevant. The next steps involve convolution with the geometrical error and the sample surface roughness, here they are both approximated by a Gaussian function, the geometrical error is obtained from a reference sample SRM2137 (NIST) and the surface roughness is fitted around 3 micron. Conversion from depth to energy is the inverse of the operations described in the paper, now the purple dotted line is obtained. This clearly resembles the concentration measured, there is however some intensity visible in the window. This is another effect arising from the high ^{6}Li concentration and subsequent counting rates, since the charged particles arrive at such an intensity that two particles are occasionally detected as a single particle having the sum of the kinetic energies. In the spectrum this results into the small lithium concentration stretching into the current collector/window making it prone to misinterpretation. The probability of having two hits within a short time is equal to the single hit probability squared, hence shape of this anomaly is found by convolution of the spectrum with itself and the intensity is related to the detector dead time squared. Summation of the yellow and purple lines leads to the blue line which closely resembles the measured data using only the sample surface roughness as a free parameter.

Figure S1. Deconvolution of neutron depth profiling measurement. The green line reflects the measured spectrum obtained in 5 minutes, 1 count per monitor second is equivalent to 1.018×10^{15} ^{10}B atoms/cm^{2}. Primary counts reflect the detector output if all particles are correctly separated and secondary counts are due to two particles measured as one, the simulated spectrum is the summation of the two, accurately reflecting the measured spectrum. The inset reflects the starting point in blue and the measurable concentration in red, lowered due to lower neutron intensity in deeper parts of the sample.