Supplementary Information

Free standing dual phase cathode tapes – scalable fabrication and microstructure optimization of garnet-based ceramic cathodes

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Supp. Figure 1 top: particle size distribution of the powders used or tape-casting of mixed cathodes via laser-diffraction; bottom left: SEM of the as-received LCO powder; bottom right: SEM of the milled LCO powder

As the LCO particles break in a flake-like manner, the particle size distribution measurement via laser diffraction cannot give an accurate representation of the particle size. SEM confirms the suitability of the milled powder for tape-casting.



Supp. Figure 2 SEM of the PEO-cathode interface

No infiltration of the polymer into the mixed cathode can be detected, as the porosity close to the surface shows clean pore surfaces.



Supp. Figure 3 SEM of thin mixed cathode without gradient after cycling at C/120 for 20 cycles

No micro-cracks between LLZO and LCO particles can be detected after cycling of the full cell.



Supp. Figure 4 Impedance spectra of the PEO:LiTFSI polymer electrolyte at room temperature (blue) and 80°C (orange)

The conductivity of the PEO:LiTFSI at room temperature does not allow for successful cycling at competitive charge and discharge rates. Nevertheless, an increase in temperature beyond the glass transition temperature dramatically increases the conductivity. At the desired operating temperature of 80°C suitable conductivities in the 10⁻⁴ Scm⁻¹ range are reached. The polymer electrolyte can therefore be incorporated into the model cell design without further modifications.





Supp. Figure 5 top: SEM picture of a mixed cathode, bottom left: EDX spectrum of a dark particle, bottom right: EDXX spectrum of a light particle

SEM and EDX analysis were performed on the pristine mixed cathodes. EDX qualitatively confirms the dark particle in the SEM images as LCO and the light particles as LLZO. Quantitative analysis is not possible. As the size of the stimulation bulb in these mixed cathodes can not be determined reliably, and the grain size of around 2-3 μ m is quite small, an interference of neighbouring grains next to or under the ones analysed during the measurement can not be excluded.