Supplementary Information

Tailored nano-columnar La_2NiO_4 cathodes for improved electrode performance

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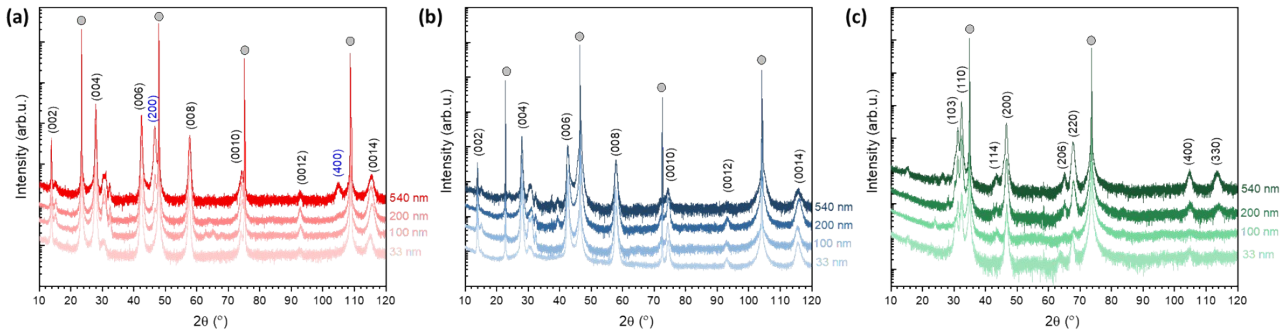
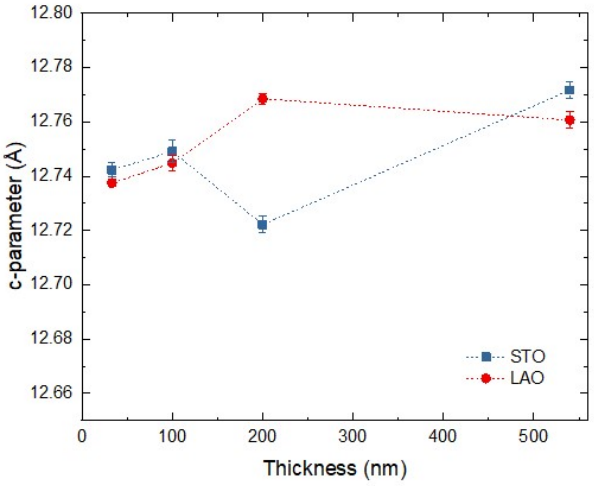
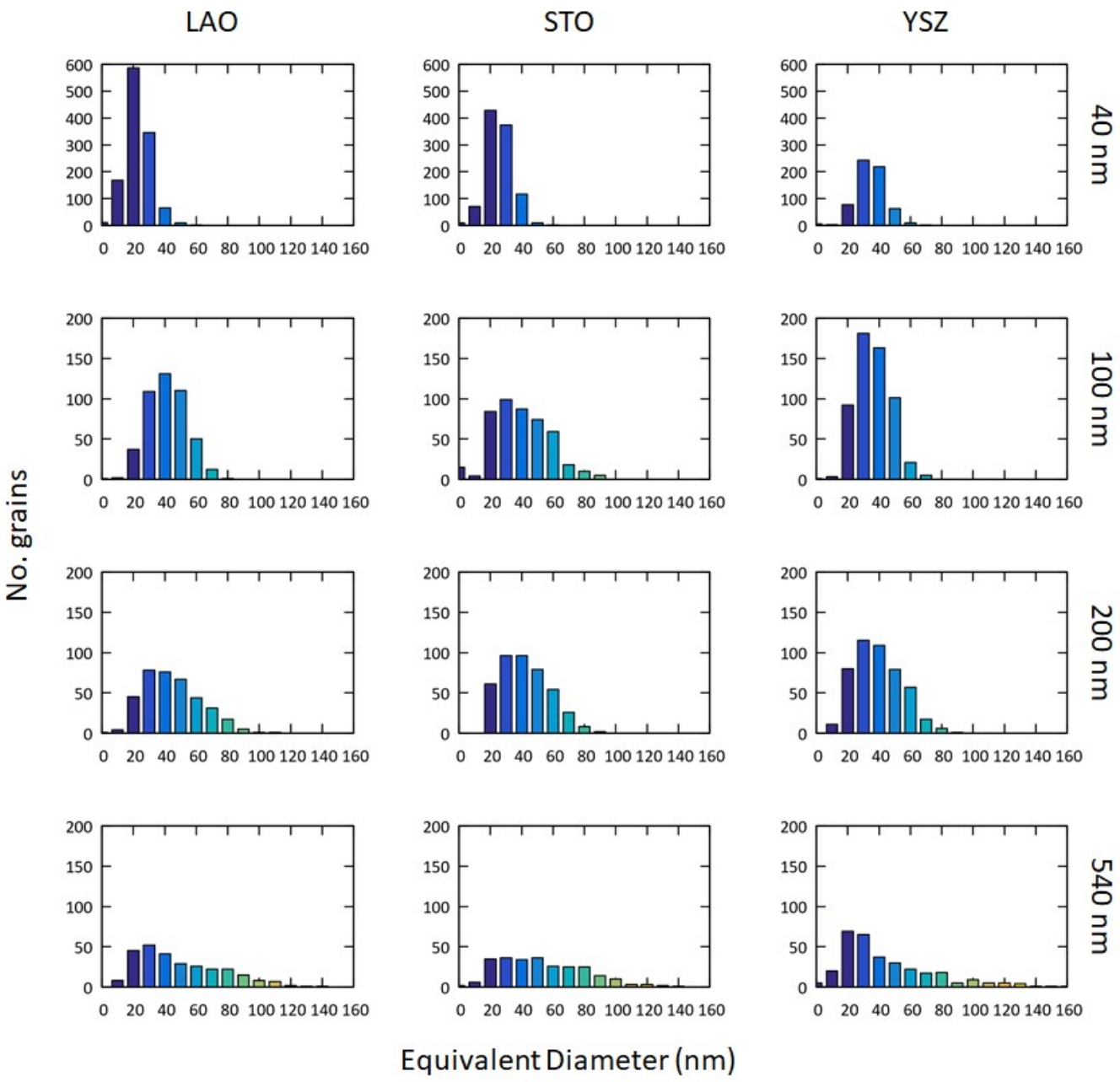


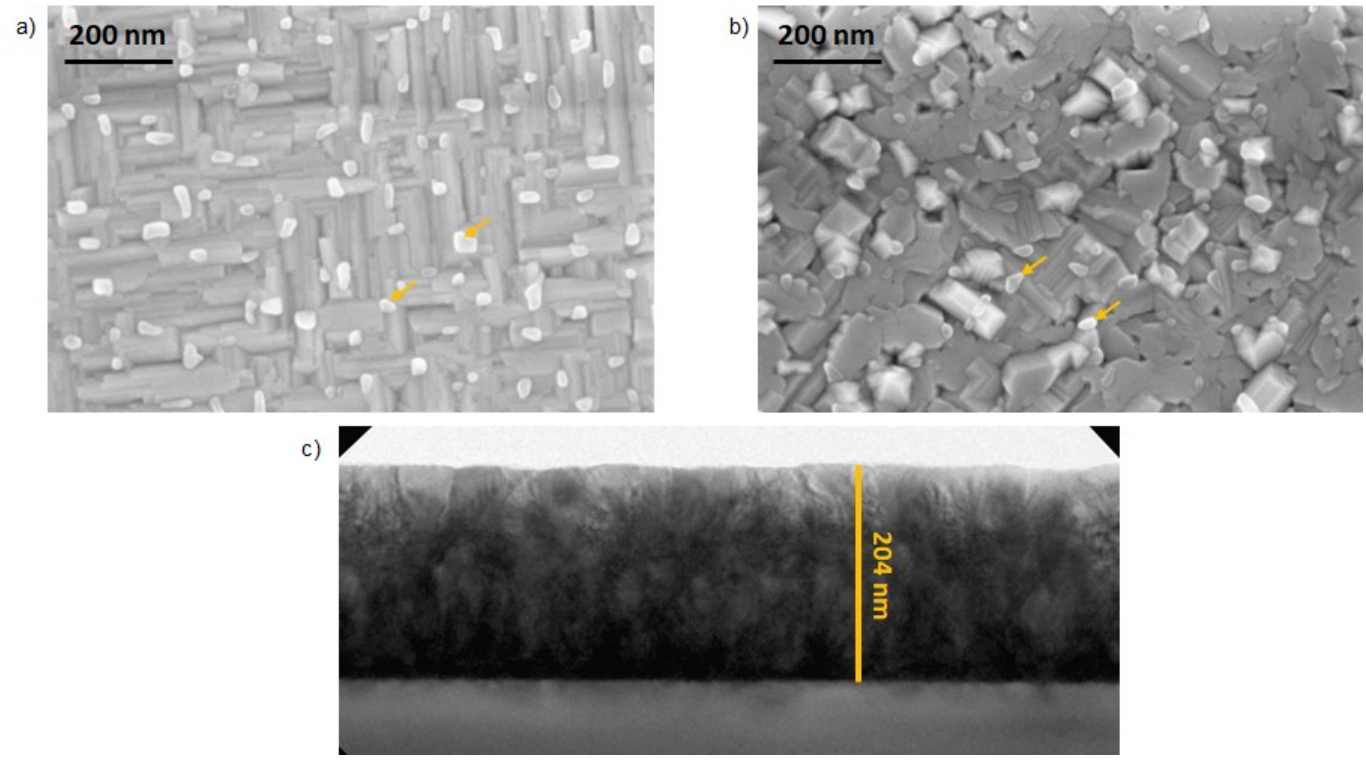
ESI-Figure 1: XRD pattern of L2NO4 thin films with different thickness deposited at 650 °C on a) LAO, b) STO and c) YSZ. Substrate peaks are marked by grey dots and the observed L2NO4 planes are labelled according to ICDD reference 04-015-2147.



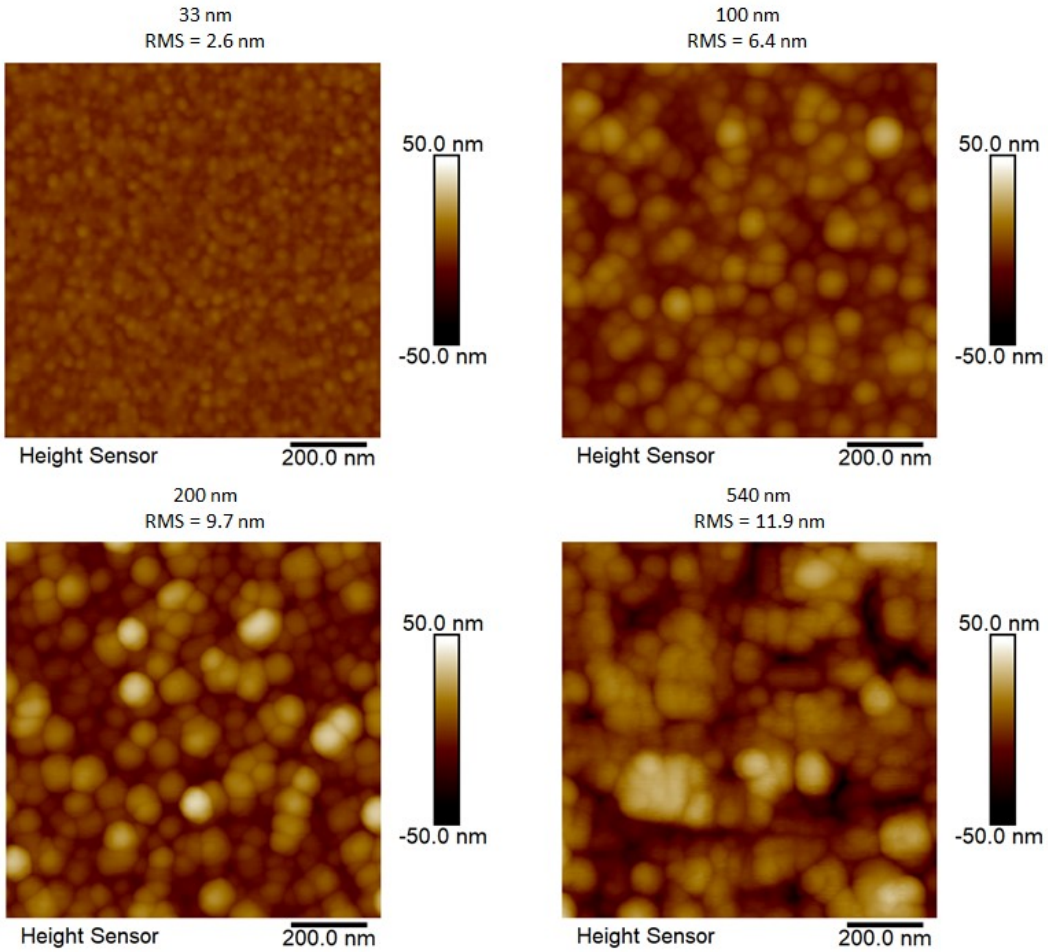
ESI-Figure 2: *c*-parameter as a function of thickness for ”as-deposited“ L2NO4 films on LAO and STO single crystal substrates.



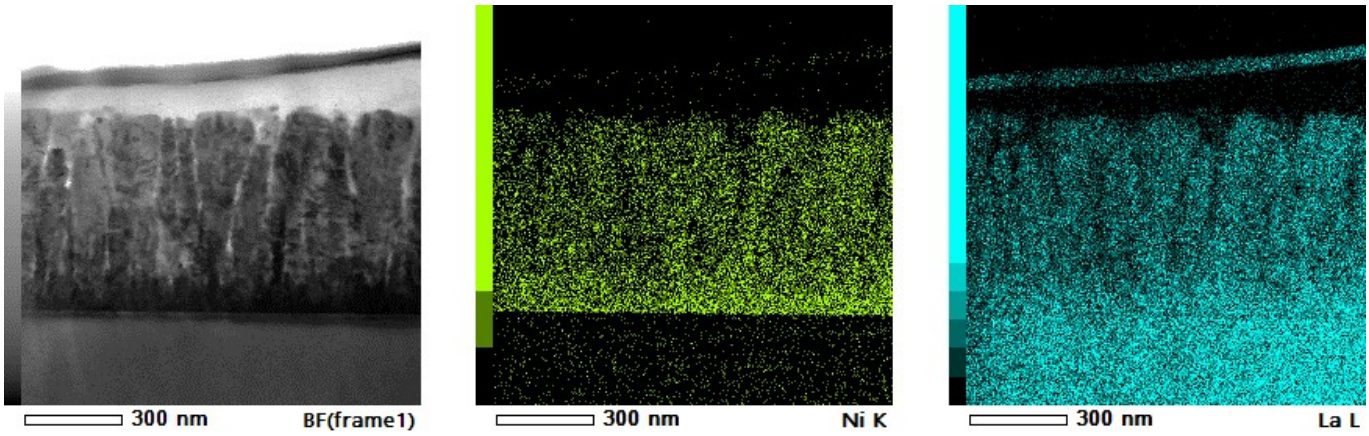
ESI-Figure 3: Grain size distribution, i.e. number of grains for each grain size, for all L2NO4 film thicknesses and substrates, analysed within an area of 1.13×0.76 µm.



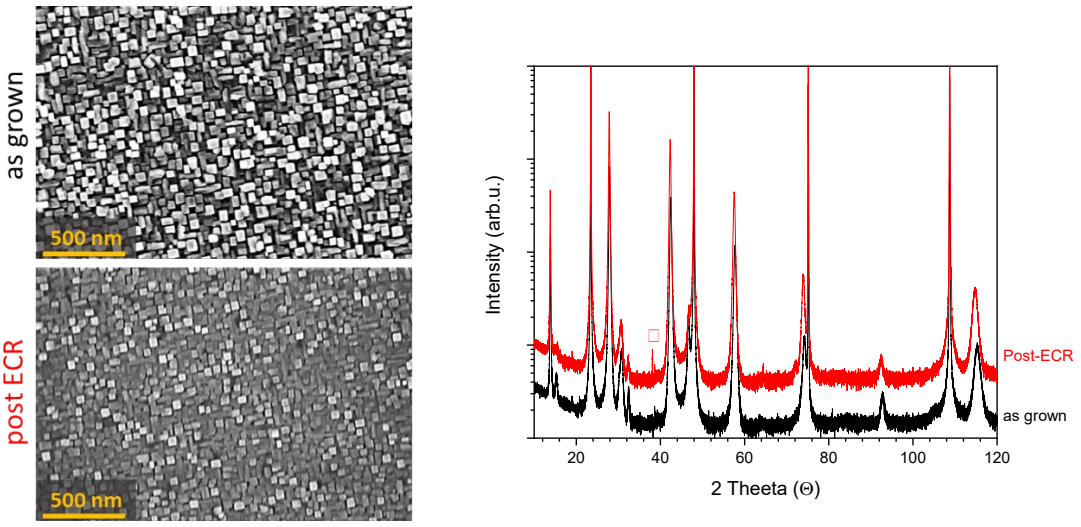
ESI-Figure 4: SEM analysis of 200 nm thick L2NO4 film deposited at 750 °C on a) LAO and b) YSZ substrate. Deposition at elevated temperature results in a dense structure with a closed, flat surface, as confirmed by TEM analysis, shown in c) for the L2NO4/LAO sample. Segregation of excess La on the surface is observed in the form of La_2O_3 particles (yellow arrows). The La/Ni ratio in the precursor solution was optimised for depositions at 650 °C. The solution-to-layer transfer ratio however depends on the deposition temperature. The La segregation can be avoided by reducing the La/Ni ratio in the precursor solution. However, no influence of excess La was found on the exchange activity.



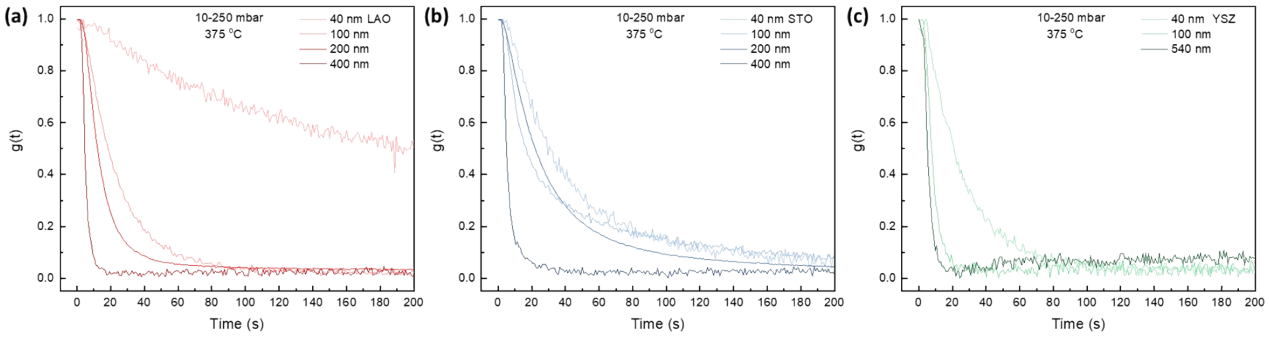
ESI-Figure 5: Atomic force microscopy of L2NO4/LAO samples, revealing increasing roughness (RMS) from 2.6 to 11.9 nm with increasing film thickness from 33 to 540 nm.



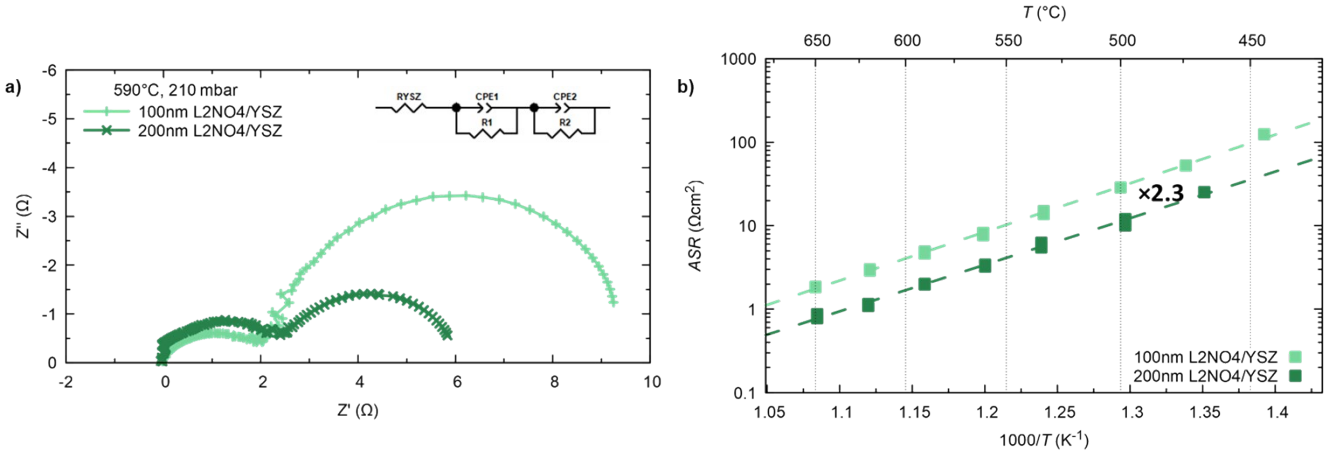
ESI-Figure 6: STEM EDX analysis of 540 nm thick L2NO4/LAO sample.



ESI-Figure 7: Analysis of microstructural stability of L2NO4 by SEM and XRD after functional characterisation at temperatures up to 600 °C with a total annealing duration of 24h. The very narrow line at around 38 ° in the post ECR spectra (red diamond) comes from the Ag electrodes.



ESI-Figure 8: Normalised conductivity transients of L2NO4 thin films at 375°C after a change of pO_2 from 10-250 mbar. Films of different thickness deposited on a) LAO, b) STO and c) YSZ.



ESI-Figure 9: a) Electrochemical impedance spectroscopy of nano-columnar L2NO4/YSZ measured in dry air in the frequency range from 1 MHz to 1 Hz at 590 °C. The equivalent circuit, used to fit the EI spectra, is shown in the inset, with a resistive contribution for YSZ in series to a high and low frequency Randles cell for the counter and the L2NO4 working electrode, respectively. b) L2NO4 contribution over the reciprocal temperature for 100 and 200 nm thick L2NO4.