High performing SOFC via multilayer tape casting?

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High performing SOFC via multilayer tape casting?

Aim – Decreased SOFC manufacturing cost while keeping high fuel cell performance:

Substitute processing steps that have low material yield (e.g. spraying) and those not suitable for industrial scale production.

Decrease number of sintering steps and handling efforts.

→ Multilayer tape casting (MTC) and co-sintering of support layer, anode and electrolyte could be a solution!

Questions:

i. Can the entire anode half cell be produced via multilayer tape casting?
ii. Can we co-cast anode and electrolyte layers at thicknesses of just 10-15µm?
iii. Can we obtain optimal microstructures for all half cell components in one single sintering step providing high performing anodes?

Findings

DTU Energy Conversion produce SOC anode half-cells by MTC and co-sintering in a pre-pilot plant. Investigations of microstructure and performance of cells (Ni/3YSZ-Ni/8YSZ-8YSZ-LSM/YSZ) with MTC anodes reveal:

1) A "window" for the co-sintering from 1255°C to 1335°C
2) Uniform microstructures of desired thicknesses, porosities, particle sizes and percolation
3) High initial performance
4) Correlations of microstructure and performance with sintering temperature — increased porosity and decreased Ni particle size with lower sintering temperature ⇒ decreased gas diffusion and charge transfer reaction resistance in the Ni/YSZ anode half cell.

Microstructure

The MTC anode half-cell

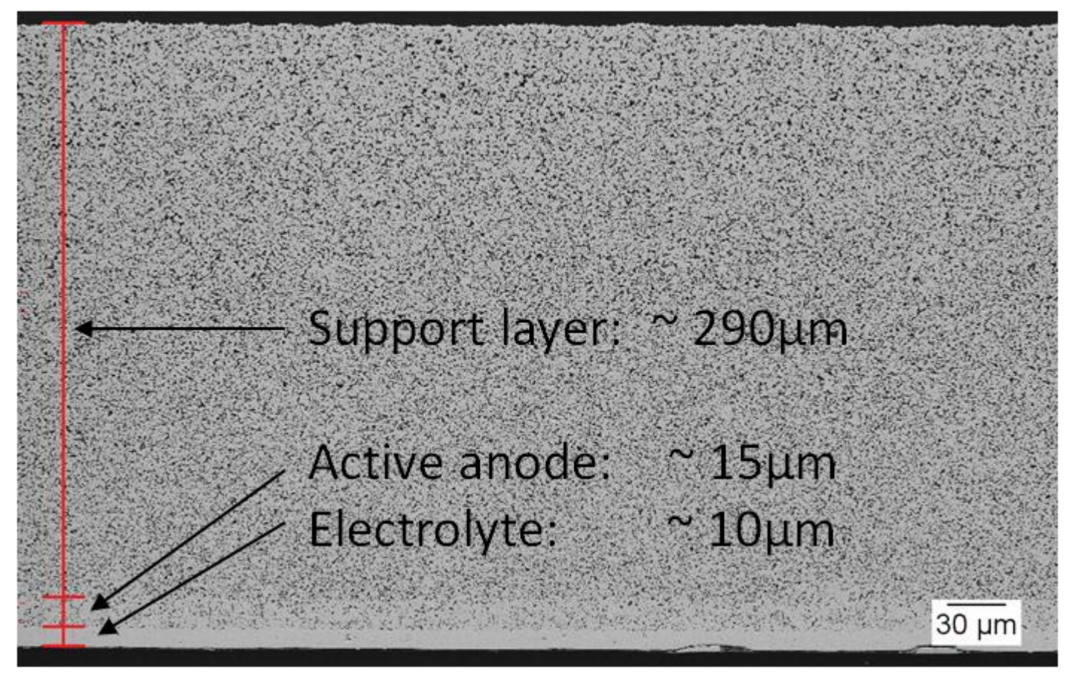

Figure 1: High voltage SE-detector SEM image of reduced, non-tested MTC half-cell sintered at 1315°C.

Low voltage SEM: The percolating Ni network

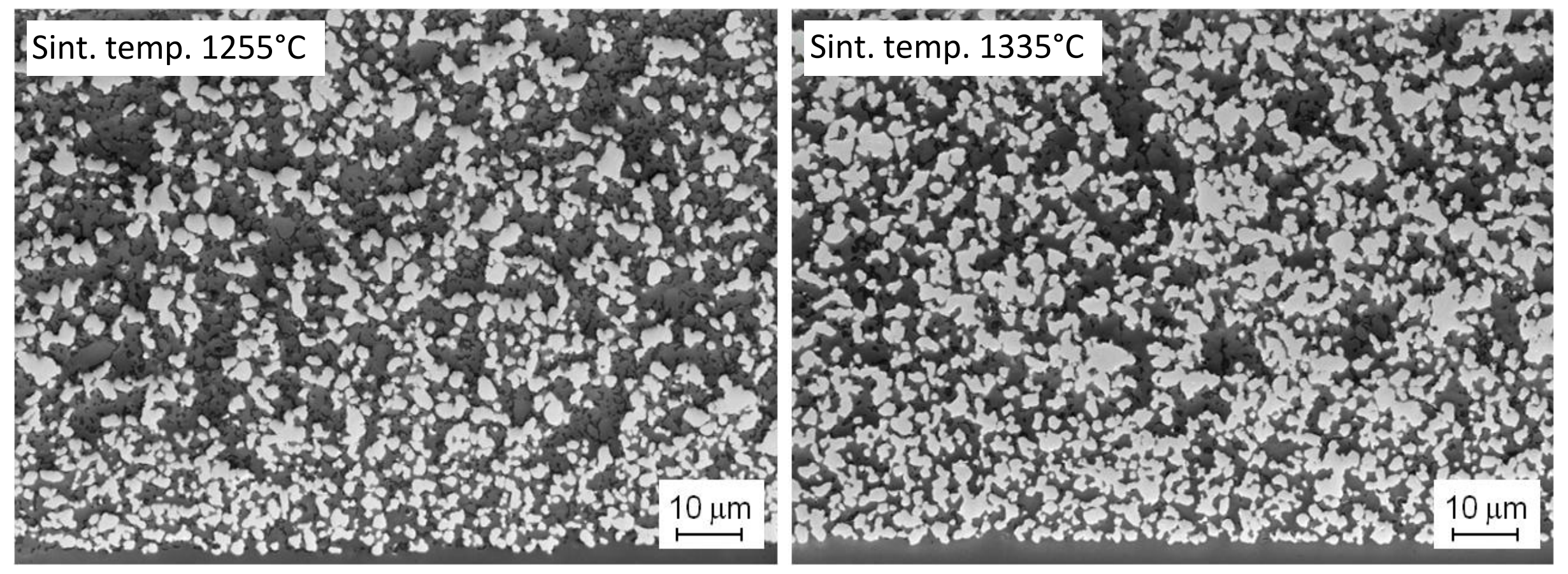

Figure 2: Low voltage (0.9kV) in-lens SEM images of reduced, non-tested MTC half cells. Bright particles constitute the percolating Ni-network.

High voltage SEM: Impact of sint. temperature

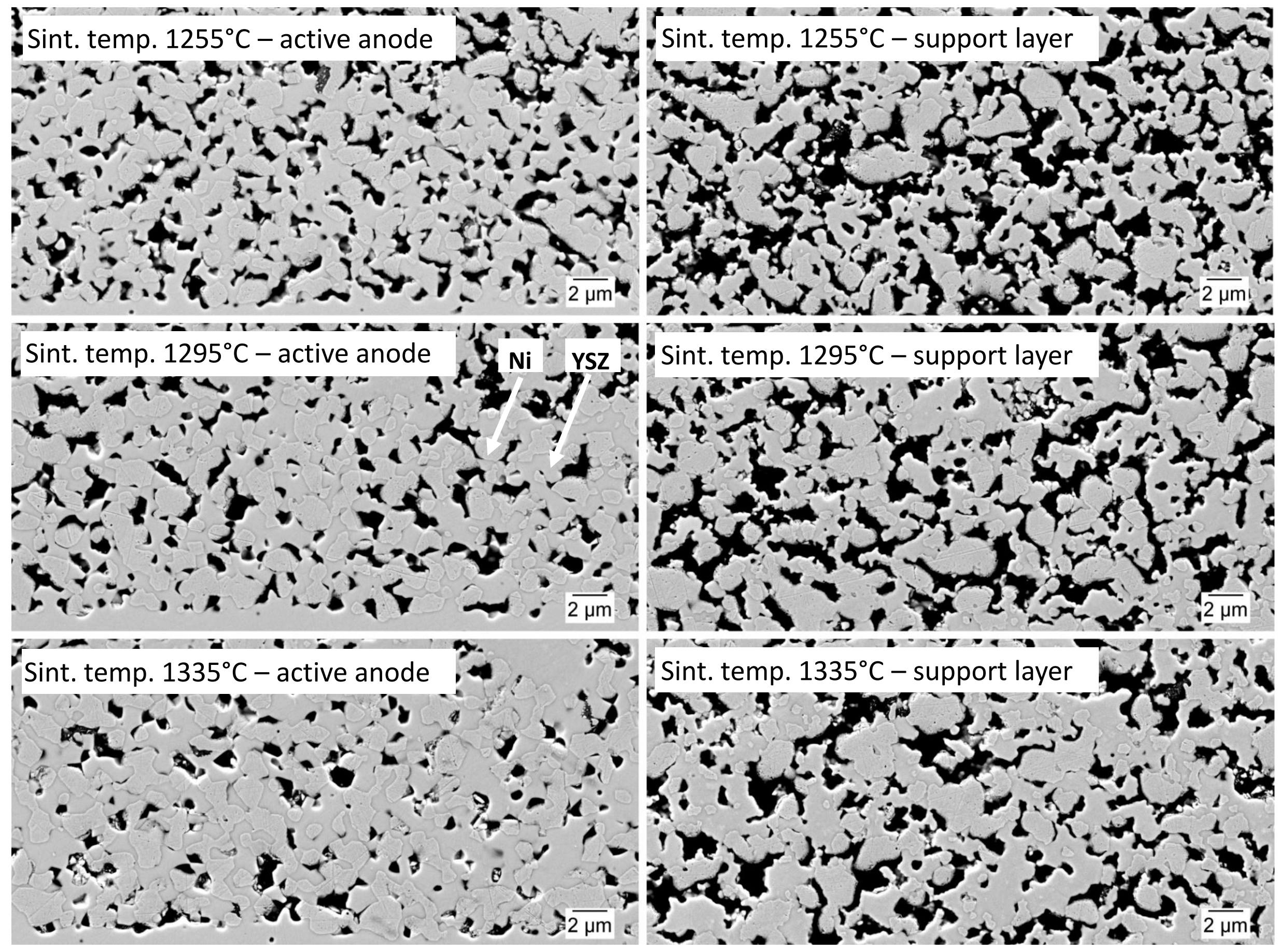

Figure 3: High voltage (8 kV) SE-detector SEM images of reduced, non-tested MTC half-cells. Pores appear black. Ni and YSZ phases are designated in the image of the 1295-cell.

Microstructural analysis: Phase fractions and mean intercept lengths

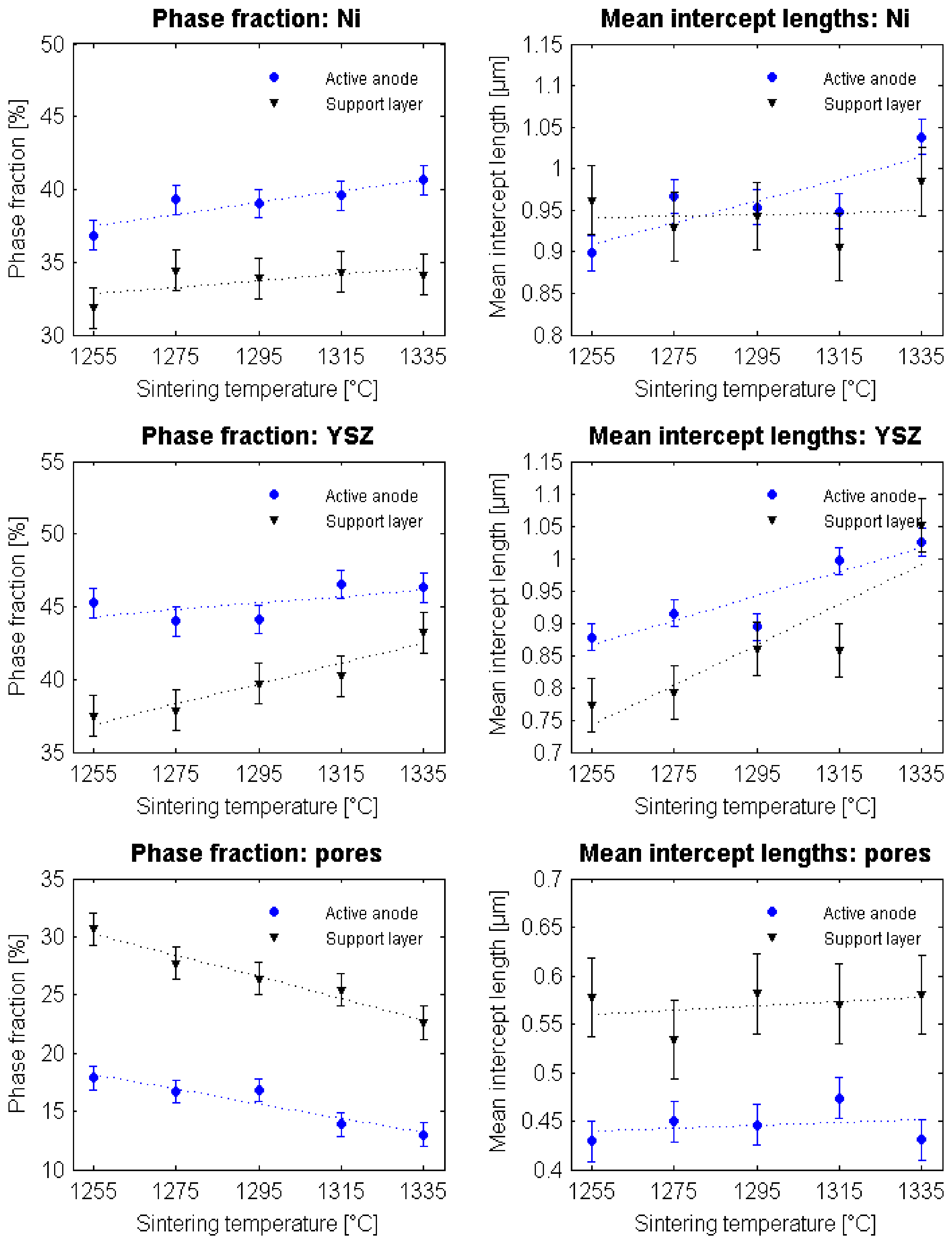

Figure 4: Microstructural analysis of reduced, non-tested half-cells sintered at 1255°C, 1275°C, 1295°C, 1315°C and 1335°C. Uncertainties for the measured intercept lengths are app. ±0.04µm and ±1.4% for phase fractions. Measurements were performed on high voltage SE-detector images, at 11.3 kX (Figure 3), using in-house developed Matlab-based software ManSegv0.31. A minimum of 1000 line intercepts were measured for each phase in anode and support of all 5 cell samples.

Electrochemical performance

Performance analysis: EIS, DRT, iV

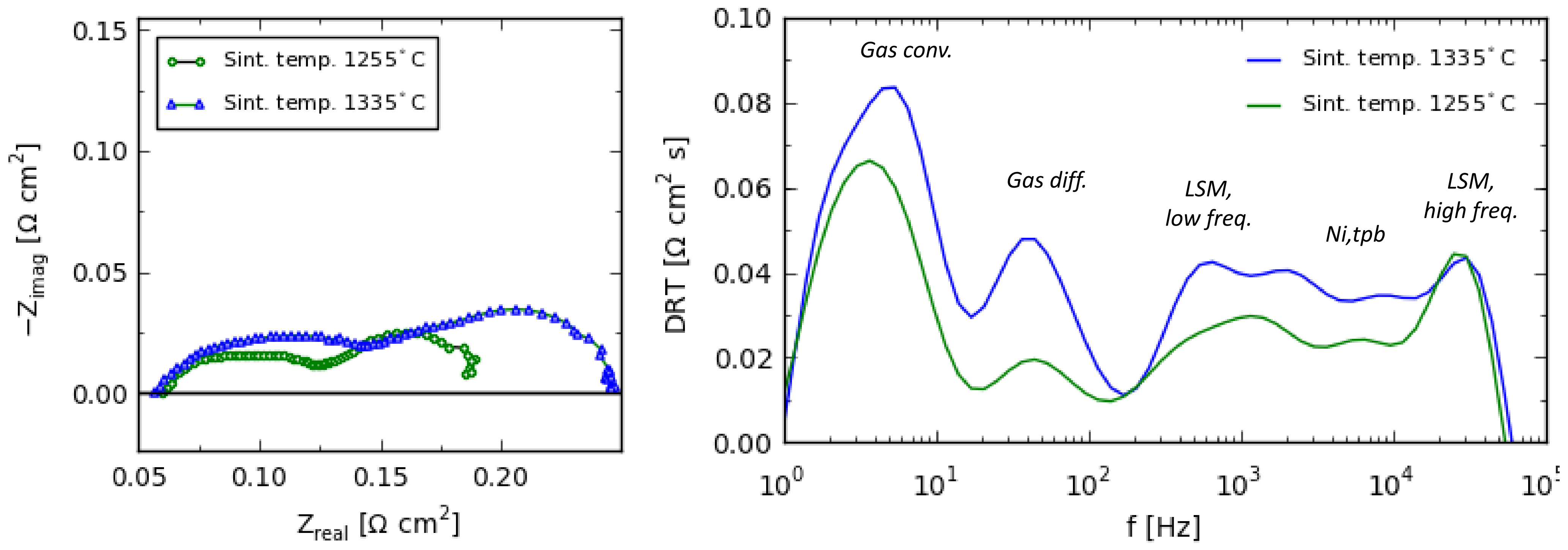

Figure 5: Nyquist (left) and DRT plot (right) of impedance spectra (IS) at 850°C, OCV, air to the cathode, 21-23% H_2O in H_2 to the anode. The Nyquist plot is corrected for inductance.

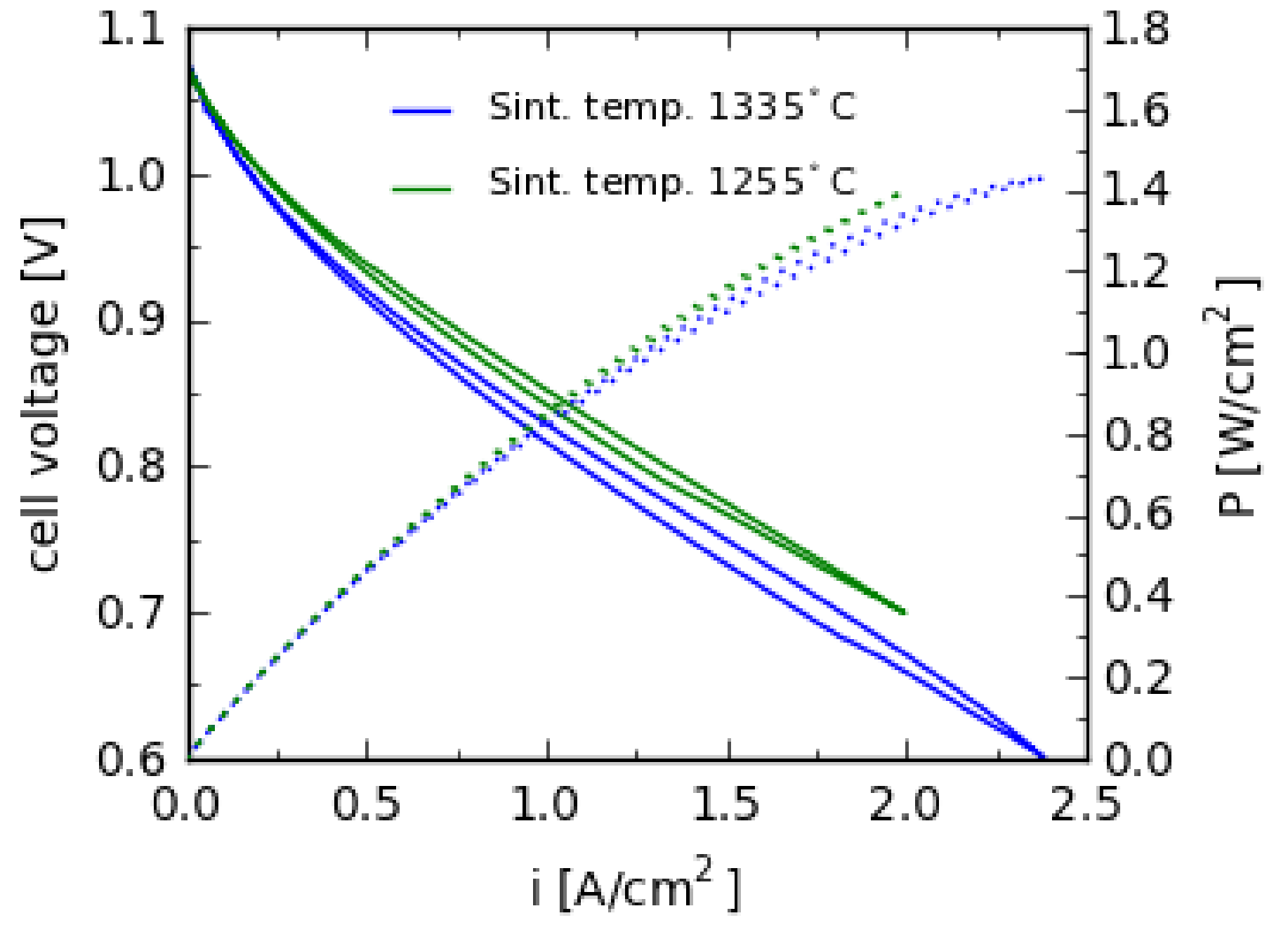

Figure 6: IV curves at 850°C, air to the cathode, 4% H_2O in H_2 to the anode.

EIS analysis: CNLS fitting

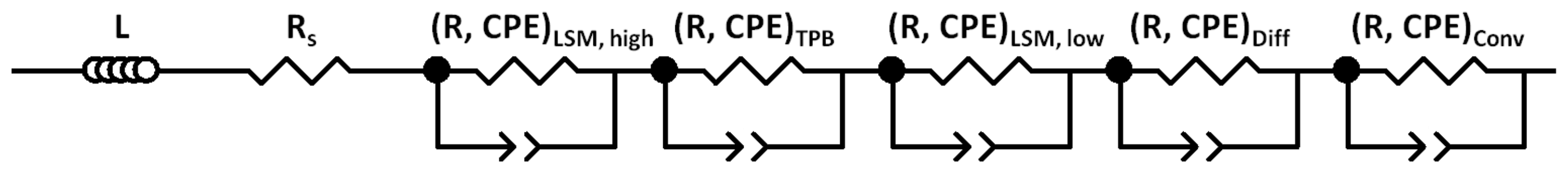

Figure 7: Equivalent circuit model applied in CNLS fitting of IS shown in Figure 5. Inductance L and series resistance R_s are connected in series with 5 RQ elements, representative of the polarisation resistance. Corresponding results are given in Table 1.

Sint. temp.	ASR_{FU} (mΩcm²)	R_s (mΩcm²)	R_{tpb} (mΩcm²)	$f_{s,tpb}$ (Hz)	R_{Diff} (mΩcm²)	$f_{s,Diff}$ (Hz)	R_{Conv} (mΩcm²)	$f_{s,Conv}$ (Hz)
1255°C	128	51	52	8442	15	39	51	4
1335°C	158	42	85	7112	34	39	61	4

Table 1: ASR_{FU} at 700 mV from iV-curves in Figure 6 and resistances and summit frequencies from CNLS-fit of the IS shown in Figure 5. Estimated error for the resistances ~3 mΩcm².

Sintering temperature: 1255°C ⟶ 1335°C

Microstructure	> <	Cell performance
1) Lower porosity (support: from 31% to 23%; active anode: from 18% to 13%)		1) ~2-fold increase in gas diffusion resistance
2) Ni particle coarsening (active anode: from 0.88 µm to 1.03 µm)		2) Increase in charge transfer reaction resistance from 52 mΩcm² to 85 mΩcm² (850 °C)

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