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Investigation of factors affecting the performance of a single-layer nanocomposite fuel cell

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1. Introduction

Fuel cell (FC) is a promising energy conversion technology that converts the chemical energy of the fuel directly to electricity with high efficiency. FCs include a large variety of technologies, some already commercialized and some still in research stage. FCs can be operated with pure H₂, hydrocarbons and other fuels, e.g. methanol. Choosing a suitable FC allows efficient and clean electricity production in both stationary and mobile applications.

Traditional FCs consist of three layers: anode, electrolyte and cathode. Solid oxide fuel cells (SOFCs) [1] and ceramic nanocomposite fuel cells (CNFCs) [2,3] are utilizing this structure. Fuel gas (e.g. H₂) is supplied to the anode and oxidant (e.g. air) to the cathode. The electrolyte transports ions between the electrodes. This three-layer structure requires well-matched thermal expansion coefficients (TECs) for the cell materials to avoid cracking, limiting the range of potential materials. In addition, the manufacturing process of the cell can be complex.

Recently, a single-layer structure has been proposed, noted as single-layer fuel cell (SLFC) or electrolyte-free fuel cell (EFFC) [4-6]. Typical SLFC consists of a composite material that is a mixture of electronic conductor or semiconductor and ionic conductor. This single-layer supports both electrode reactions on the opposite edges of the cell and allows ion diffusion through the cell. This approach eliminates the need of matching TECs and allows simplifying the manufacturing procedure.

The working mechanism of SLFC is presented in Figure 1. Fuel gas (here H₂) is supplied from one side and oxidant (here air, containing O₂) from the other. The electrode reactions occur at the surfaces of the cell, possibly catalyzed by the current collector. Electrons are released on the fuel side and driven through the external circuit to the oxidant side. Charge transfer through the cell is done via
ion conduction. Figure 1 presents two parallel charge transfer mechanisms, based on O\(^2-\) ion and proton conduction respectively. Water vapor and unreacted gases are removed from the cell.

In literature, a large variety of different material candidates for SLFCs is reported. As electronic / semiconductor, typical materials include wide band-gap oxides based on lithium and nickel [7-19], such as lithium nickel zinc oxide (LNZ) [8,17]. The most typical ionic conductors are samarium-doped cerium oxide (SDC) [8,13-15,17,20-23] and gadolinium-doped cerium oxide (GDC) [7,10,19]. Impressive power densities achieved with materials described above include 600 and 760 mW/cm\(^2\) at 550 °C with LNZ-SDC [17] and lithium nickel iron oxide – SDC [8] respectively. Lithium nickel cobalt aluminum oxide (NCAL) has been used as both electronic conductor of the composite material [24-27] and as electrode or external current collector [19,28], typically as painted on a Ni-foam.

Perovskite structured materials, such as lanthanum strontium cobalt ferrite (LSF) [28-31], barium strontium cobalt ferrite [20] and \(\text{Pr}_{0.4}\text{Sr}_{0.6}\text{CoO}_2\text{Fe}_{0.7}\text{Nb}_{0.3}\text{O}_3\) [27] are another alternative for Li- and Ni-based oxides. The ionic conductivity of the SLFCs can be improved by using co-doped cerium oxides [24,26,28,29,31] and mixtures of a doped cerium oxide and alkali carbonates [9,11,12,16,25,27,32]. Careful material selection has led to improved performance: 1187 mW/cm\(^2\) with NCAL – (triple-doped cerium oxide) [24], 1072 mW/cm\(^2\) with NCAL – (SDC – sodium carbonate) [25] and 814 – 1080 mW/cm\(^2\) with LSCF – (Ca, Sm co-doped cerium oxide) [28,29,31] have been reported at 550 °C.

Although SLFCs have shown great potential, several challenges remain, including that scientific knowledge on its principles and performance-affecting factors is yet low. Thus, even basic knowledge about materials suitable for SLFCs and the factors affecting the cell performance and stability is lacking. This work deepens the understanding of the factors affecting the performance of LNZ-based SLFCs. LNZ was chosen as studied material due to existing literature [16-18], our previous experiences [19] and the simplicity of its synthesis procedure [16-19]. The factors analyzed include changing the cell material (applying alkali carbonates and changing the dopant of cerium oxide), modifying the microstructure (changing porosity by adding a pore former) and applying external, catalytically active current collector. The aim is to map systematically how the cell performance responds to the changes discussed above and explain the responds by both microstructural and macroscopic characterization. Understanding the reasons behind the cell performance is crucial to allow the development of high-performing stable SLFCs in the future.

2. Experimental methods

All chemicals were purchased from Sigma-Aldrich and used as received unless otherwise stated. Powders were mixed by grinding for 10 min.

LNZ powder was prepared by mixing zinc nitrate hexahydrate, nickel(II) carbonate basic hydrate and lithium carbonate so that elemental ratio Ni:Zn:Li was 8:9:3. After mixing powder was calcined at 800 °C for 180 min and grinded for 10 min. Cerium oxides, samarium/gadolinium doped (15 mol-% for Sm, 10 mol-% for Gd, SDC and GDC respectively) were calcined at 600 °C for 60 min and grinded for 10 min before use. Eutectic sodium-lithium carbonate (NLC) was prepared by mixing sodium and lithium carbonate (57-43 weight-%, melting point 497 °C [2]). Composite SDC-NLC and GDC-NLC powders were prepared by mixing SDC or GDC with NLC (3-1 by weight), calcining at 600 °C for 60 min and grinding for 10 min. LNZ and the ionic conductor (SDC, GDC, SDC-NLC or GDC-NLC) were mixed at 2:3 weight ratio that has been found to be optimal in the literature [11,13] unless otherwise stated, calcined at 700 °C for 120 min and grinded for 10 min. For the powder patches containing a pore former, 5 weight-% of ethyl cellulose (EC) was added after calcining, followed by grinding for 10 min. The cells prepared from EC-containing powders are noted with the tag “-EC”.

Experiments included changing the cell material (applying alkali carbonates and changing the dopant of cerium oxide), modifying the microstructure (changing porosity by adding a pore former) and applying external, catalytically active current collector.
Powders were pressed to pellets in a cylindrical die (diameter 13 mm). Pressing was done at 250 MPa and room temperature for 2 min. After pressing pellets were sintered at 690 °C for 120 min, using < 2 °C/min heating and cooling rates. The summary of the cells presented in this study is given in Table 1.

Lithium nickel cobalt aluminum oxide (NCAL) precursor was prepared by mixing ethanol (Altaia) and glycerol (1:1 by volume) and stirring for 15 min. NCAL was added so that the mass load was 0.4 g/ml, followed by stirring for 60 min. Precursor was then painted from both sides on Ni-foams that were pressed at 100 MPa for 10 s beforehand. NCAL-coated Ni-foams were sintered at 500 °C for 30 min using < 2 °C/min heating and cooling rates.

Electrochemical characterization was done with experimental testing setup purchased from Fiaxell. Gas flow rates were 50 ml/min H₂ + 125 ml/min air (stoichiometric ratio) or 100 ml/min both H₂ and air (equal gas flow pressures on both sides). The operational temperature was monitored by using a thermocouple that was placed next to the cell. Active area was 0.64 cm² and it was limited by the hole cut to the sealant (alumina-silica felt, purchased from Fiaxell). Either Ni-foams or NCAL-coated Ni-foams (noted with tag “-NCAL”) were used as current collectors.

Zahnner Zennium electrochemical workstation was used to perform current-voltage (IV) and electrochemical impedance spectroscopy (EIS) measurements. IV-curves were measured in range 1.2 V → -1.0 V → 1.2 V with a slew rate of 100 mV/s. EIS-spectra were measured in range 100 kHz → 100 mHz → 100 kHz at open circuit condition with 20 mV amplitude. Identical measurements were done at 500 °C, 550 °C and 600 °C under both gas flow rates.

X-ray diffraction (XRD) characterization was done with Rikagu SmartLab X-ray diffractometer, equipped with a rotating anode X-ray source (Cu, 9 kW) and HyPix-3000 detector. Spectra were measured from 2θ = 20° to 2θ = 80° with a scan rate of 6°/min. For high temperature XRD (HT-XRD) measurements in-house built experimental setup was used to heat the sample to the desired temperature (heating rate 50°C/min, 5 min stabilization after reaching the target temperature) after mounting it to the XRD-device. The measurement temperature sequence was room temperature (RT) → 450 °C (below melting point of NLC) → 550 °C (above melting point of NLC) → 450 °C → RT. The mean particle sizes were calculated by using Powder X-ray Diffraction Measurement Analysis Software PDXL, based on the Scherrer equation [33,34].

Zeiss Sigma VP Scanning electron microscope (SEM) and secondary electron (SE) detector were used to obtain SEM images from used fuel cell pellets. Jeol JEM-2800 high-resolution transmission electron microscope (HR-TEM) equipped with energy dispersive X-ray spectroscopy (EDX) for elemental analysis was used to characterize the properties of nanopowders.

3. Results and discussion
3.1. Cell microstructure

The XRD-spectra of LNZ-SDC and LNZ-SDC-NLC nanopowders at different temperatures in range 2θ = 20° - 80° are shown in Figure 2a. The observed peaks were corresponding to SDC, LiNiO and LiZnO, indicating the high purity of the characterized powders. Additional peaks related to alkali carbonates were not observed in LNZ-SDC-NLC powder, indicating that carbonates are in amorphous form and thus not visible in XRD-analysis. Particle size analysis revealed significant differences between the different components. At room temperature, the average particle sizes were 36, 45 and 70 nm for SDC, LiNiO and LiZnO respectively. When temperature was increased, also particle sizes grew: at 550 °C the respective values were 41, 47 and 86 nm. The peaks were identified using ICDD files No. 04-015-2396 (SDC), 04-006-8078 (LiNiO) and 04-013-7260 (LiZnO).
Figure 2b shows zoom-in of Figure 2a in range 2° = 32° - 38°. In this scale, the peak shift towards lower angles can be clearly seen for all components, although the shift is much smaller for LiZnO peaks. This phenomenon can be explained by the thermal expansion of the lattices [35-37]: calculated from the lowest-angle peaks of the LNZ-SDC powder, the lattice expansion coefficients from RT to 550 °C were 1.0044, 1.0056 and 1.0027 for SDC, LiNiO and LiZnO respectively. Smaller expansion coefficient explains smaller peak shifts for LiZnO.

SEM was applied to study the surface of used single-layer fuel cell pellets. The LNZ-SDC pellet shows a dense crystalline single-phase structure of LNZ-SDC. In the LNZ-SDC-NLC pellet a dual phase structure can be observed: besides LNZ-SDC crystals there exists an amorphous carbonate phase. Pellets prepared from EC-containing powders show also some holes on the surface, resulting from the evaporation of EC during the sintering. However, the amount of such holes is relatively low due to low EC-content of the respective powders. In all images rod-like structures, possibly hydroxides [2] were observed. SEM images are shown in Supplementary information, Figure S1.

TEM images of LNZ-SDC (Figure 3a) and LNZ-SDC-NLC (Figure 3b) powders show nanoparticles of various sizes. Based on XRD-analysis, the smaller particles are most likely SDC, medium-sized LiNiO, and the large ones LiZnO. Due to electron transparency requirement for TEM analysis, only individual particles and few overlapping particles can be seen: the centers of the particle clusters are too thick for electron transparency and thus are shown as black blurry. A high-resolution TEM image showing crystalline structure of few individual particles from LNZ-SDC powder is shown in Figure 3c. Based on the size, these particles are likely to be SDC.

Elemental analysis of LNZ-SDC-NLC particle cluster based on EDX is shown in Figure 3d-j. Figure 3d shows the bright field image of the studied cluster, whereas Figures 3e-j are elemental maps. Based on these maps, different areas of the particle can be identified as SDC (Ce and Sm maps) and LNZ (Ni and Zn maps). The maps show that Ce and Sm appear only together, whereas Ni and Zn can appear also separately. This is consistent with XRD data that identified three phases: SDC, LiNiO and LiZnO. Based on the Na-map, NLC is spread all over the cluster. Also O is observed in every part of the cluster, as expected. Li was too light element to be detected with a reasonable measurement time.

XRD, SEM and TEM analysis for LNZ-GDC material have been done and reported in our previous work [19] and is not included in this paper.

### 3.2. Electrochemical performance

Replacing the doped ceria with a composite material consisting of doped ceria and alkali carbonates has been observed to improve dramatically the ionic conductivity [2,3,9,11,12,16,25,27,32]. This phenomenon was clearly observed in this study as well. Figure 4 shows IV and EIS data of eight SLFCs with different material compositions. Adding NLC to GDC or SDC improved the cell performance by a factor of 3.2 – 5.7 (Figure 4a), depending on the cell composition: low-end factors are from the cells without NCAL (red → blue lines in Figure 4) and high-end factors from the cells with NCAL (green → cyan). It should be noted that the performance of the cells without NLC was low compared to the literature [19], explaining partly the high performance-increasing factors. The reason for this change can be explained with a dramatic decrease in ohmic resistances of the cells (Figure 4b&c): without NLC the ohmic resistance varied from 2.4 to 2.9 Ω·cm², but when NLC was added the respective values decreased between 0.43 and 0.53 Ω·cm². In addition, the shape of the EIS curve changed significantly (Figures 4b-c). These factors indicate that in the cells without NLC, the ionic conductivity of the cell is the performance-limiting bottleneck. When NLC is added, limited ionic conductivity is still a significant but not a dominating loss factor. It should be noted that the EIS-measurement was
done from 100 kHz to 100 mHz and again back to 100 kHz, so the range of the spectrum was measured twice. The two curves are not completely overlapping, indicating that there may be some minor changes in the cell even during a short (5-6 min) EIS-measurement.

NCAL is a catalytically active material [19] and it is expected to increase the cell performance by boosting the electrode reactions. In the cells without NLC, this phenomenon was not observed: the performance actually decreased by approximately 10 % when NCAL was applied. Figure 4c shows that both the ohmic resistance and the polarization resistance, represented by the diameter of the semicircle, increased when NCAL was applied. However, in NLC-containing cells applying NCAL improved the performance by a factor of 1.3 – 1.4. Figure 4b shows that this is due to a minor improvement in ohmic resistance and a more significant improvement in electrode performance: the low-frequency arc (on the right end) of the EIS-spectrum was much shorter for the cells containing NCAL, indicating a decrease in polarization resistance. With both NLC and NCAL power densities of 240 and 230 mW/cm² (LNZ-GDC-NCL-NCAL and LNZ-SDC-NLC-NCAL respectively) were achieved.

The fact that NCAL had exactly opposite effects to the cell performance in cells with and without NLC was interesting. One possibility to why NCAL increased resistance and decreased performance with non-NLC cells is the high ohmic resistance of the respective cells: since ionic conduction through the cell is clearly a bottleneck, the benefit of NCAL cannot be utilized. Another possibility is that there is some interfacial loss factor between LNZ-(SDC/GDC) and NCAL.

Each cell was characterized at three different temperatures (500, 550 and 600 °C) under two different gas flow rate combinations (125 ml/min air and 50 ml/min H₂ and 100 ml/min both air and H₂), resulting as six different environments in total. In Figure 4, the best data (i.e. the IV-measurement with the highest power density and the EIS at the same environment) is shown. There was a clear trend that the cells with NLC achieved the highest performance at 550 °C and the cells without NLC at 600 °C. The decrease in the performance of NLC-containing cells from 550 °C to 600 °C was not expected, but since it occurred with all four NLC-containing cells presented in Figure 4 it cannot be a mere co-incidence. This may indicate about structural changes happening in the cell during operation. The 100 ml/min of air and H₂ resulted as higher open circuit voltage and cell performance for almost all cells. This indicates that the equal gas flow pressure on both sides results as a better performance than stoichiometric gas flow rates. Only one cell presented in Figure 4 achieved higher power density under 50 ml/min H₂ and 125 ml/min air, but this may be due to some structural variation due to manufacturing procedure.

When comparing the cells with SDC and cells with GDC, no significant difference is observed. The performance differences between cells where only change was replacing SDC by GDC are low and there is no systematic behavior in favor of SDC or GDC.

The amount of suitable reaction sites for the electrode reactions is depending strongly on the porosity of the cell. The porosity can be increased by adding a pore former (here EC) to the powder before pressing it into pellet. Increasing porosity should improve the electrode reaction rates if the amount of available reaction sites is the bottleneck for the cell performance. However, since the entire cell consist of a single layer, increasing porosity also increases the ohmic resistance due to twisting of ionic conduction pathways and makes the gas mixing through the cell more likely. Implementing EC was observed to decrease the cell performance by 15 – 24 % in all studied cases (LNZ-SDC, LNZ-SDC-NCAL, LNZ-SDC-NLC, LNZ-SDC-NLC-NCAL). This is due to increased ohmic resistance of the cells. For the best-performing material (LNZ-SDC-NLC-NCAL) the power density dropped from 230 to 190 mW/cm² and ohmic resistance increased from 0.44 to 0.59 Ω·cm². There is no evidence of improved electrode performance due to EC, indicating that the electrode reaction
rates are not increased significantly by using external pore former. There was a concern with using EC that it may cause gas cross-over through the cell. If a cell suffers from gas cross-over, it’s open circuit voltage would be unstable. Based on the electrochemical data, there is no reason to suspect that direct gas mixing through the cell has occurred since the open circuit voltages of the cells did not fluctuate during the measurement sequence. This indicates that either there are no continuous pores through the cell or the gases are consumed by the fuel cell reactions before they diffuse into each other, which is consistent with the computational model developed by Liu et al. [9]. However, direct observation of gas atmospheres on the opposite edges of the cell was not performed. IV- and EIS-data are presented in Supplementary information, Figure S2. Also the performance of the cells with EC showed the same temperature trend as the cells shown in Figure 4: the peak performance was achieved at 550 °C with NLC-containing cells and at 600 °C for non-NLC cells.

In most SLFC studies, the semiconductor and ionic conductor are mixed with a 40-60 weight ratio, that has been found to be optimal [11,13]. Increasing the amount of ionic conductor should lead to straighter ion conduction pathways through the cell, at the cost of decreased amount of suitable reaction sites on the edges. However, when a catalytically active current collector is used, the optimal ratio may differ from the literature. Three cells, consisting of LNZ and SDC-NLC with 40-60, 36-64 and 32-68 weight ratios were fabricated and characterized with NCAL current collector to study this issue. The electrochemical data is presented in Figure 5.

At 500 °C, the 40-60 cell has a superior performance compared to others (Figure 5a). The most significant difference in the EIS-data (Figures 5b-d) is the length of the low-frequency arcs of 36-64 and 32-68 cells, suggesting that the electrode reactions rates are limiting the performance of these cells. When the temperature is increased to 550 °C, 40-60 cell still has highest power density, but differences are much smaller than at 500 °C, suggesting that with increased temperature the electrode reaction kinetics improvements are playing important role. At 600 °C, the 36-64 cell reached power density of 300 mW/cm² and also 32-68 cell (280 mW/cm²) surpassed 40-60 cell (260 mW/cm²). At this temperature, the electrode reaction rates seem to be sufficient even with lower amount of LNZ, so the benefit of increased ionic conductivity can be utilized. The EIS-plots (Figures 5b-d) shows that at 600 °C the low-frequency arcs of the 36-64 and 32-68 cells are compatible to the 40-60 cell. It should be noted that the 40-60 cell reached highest power density at 550 °C similarly than NLC-containing cells in other patches (Figure 4, Figure S2), but both 36-64 and 32-68 cells improved performance from 550 °C to 600 °C (like all the cells that did not contain NLC). The exact reason behind this phenomenon is not known, but it can be concluded that the carbonate content and the mass ratio between the ionic conductor and the semiconductor obviously affects to the optimal operational temperature of a SLFC.

When comparing the performance of the cells prepared by similar manufacturing procedure but by different researchers or by same researcher but at different times, the key question is that are these results actually comparable. When the electrochemical data of two LNZ-SDC cells and two LNZ-SDC-NLC-NCAL cells that are prepared via similar manufacturing procedure but at different times was analyzed, a moderate performance difference was observed. The IV- and EIS-plots are shown in Supplementary information, Figure S3. The variation between two cells that are supposed to be identical can be greater than the variation due to a material or structural difference (e.g. when comparing SDC and GDC, Figure 4).

4. Conclusions

A large number of SLFCs were manufactured and systematically characterized. Compared to the reference cells (LNZ-GDC and LNZ-SDC) the most significant performance-increasing factor was
adding NLC to doped ceria (3:1 weight ratio of SDC/GDC and NLC). This lead to a huge improvement in ionic conductivity and overall cell performance. The cell performance was boosted even further when catalytically active NCAL current collector was applied to the cells containing NLC.

Increasing porosity by using EC as external pore former led to decreased cell performance due to a higher ohmic resistance, most likely caused by twisting of ion conduction channels through the cell. Increasing porosity was not observed to boost electrode reactions. However, since the amount of pore former was relatively low (5 weight-%), improvement in electrode reaction rates may occur when higher amount of pore former is used.

The effect of the mass ratio between semiconductor (LNZ) and ionic conductor was studied with cells using SDC-NLC as ionic conductor and NCAL current collectors. At 500 °C, the cell with conventional 40-60 mass ratio had superior performance due to limited electrode reaction kinetics of the 36-64 and 32-68 cells. However, when the temperature was increased, the cells with a lower LNZ-content improved significantly and at 600 °C, both the 36-64 and the 32-68 cells surpassed the performance of the 40-60 cell. These results indicate that with catalytically active current collectors, a lower amount of semiconducting material can increase the cell performance at higher temperatures.

Finally, an important observation about the consistency was made. Even cells that were manufactured via identical experimental procedure but in different cell sets showed variation in performance. This makes it rather challenging to compare directly the results from cells that are not from the same set. Especially, when comparing results reported by different research groups it is challenging to ensure that the reported results are actually comparable with each other.

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References


Figure 1. Working principle of a SLFC. The fuel and oxidant gases are supplied from the opposite sides of the cell. Electrons are released on the fuel side and bonded on the oxidant side. Ion conduction allows charge transfer through the cell.

Table 1. Summary of the SLFCs reported in this paper. “Base structure” refers to the structure that is similar for all cells within the patch. “Variables” shows which experimental parameters (e.g. NLC vs. no NLC) were varied within the patch. “Figure(s)” shows the number(s) of the Figure(s) where the electrochemical data of the cells is shown. Hereafter individual cells and powders are referred with syntax “base structure-variables” (e.g. LNZ-SDC-EC).

Figure 2. XRD-spectra of LNZ-SDC (thin lines) and LNZ-SDC-NLC (thick lines) nanopowders with sequence RT (blue) → 450 °C (green) → 550 °C (red) → 450 °C (purple) → RT (cyan) in range 2θ = 20°-80° (a). Zoom-in for range 2θ = 32°-38° (b).
Figure 3. TEM images of LNZ-SDC (a) and LNZ-SDC-NLC (b) nanopowders. A high-resolution image showing crystalline structure of individual particles (c). Bright field image of LNZ-SDC-NLC cluster (d). Elemental maps of the cluster shown in (d) for Ce, Sm, Ni, Zn, Na and O respectively (e-j). Scale bar lengths: 100 nm for (a-b), 10 nm for (c) and 200 nm for (d-j).

Figure 4. IV- and IP-curves of eight SLFCs with different material combinations (a). EIS-data of the respective cells presented as Nyquist plots: 100 kHz → 100 mHz → 100 kHz (b-c). The data shown is at 600 °C for the cells without NLC and at 550 °C for the cells with NLC. The IV-measurement with the highest power density was chosen for each cell. Gas flows: 100 ml/min for both H₂ and air unless otherwise stated.

Figure 5. I-V and I-P curves showing the effect of mass ratio [LNZ-(SDC-NLC)](a). EIS data of the respective cells presented as Nyquist plots: 100 kHz → 100 mHz → 100 kHz (b-d). The IV-measurement with the highest power density was chosen for each cell/temperature combination. Gas flows: 100 ml/min for both H₂ and air unless otherwise stated.

Figure S1. SEM images of used LNZ-SDC (a), LNZ-SDC-EC (b), LNZ-SDC-NLC (c) and LNZ-SDC-NLC-EC (d) pellets. All images were taken from the fuel side after electrochemical characterization. Scale bar is 20 μm.

Figure S2. IV- and PV-curves of eight SLFCs showing the effect of EC to the cell performance (a). EIS data of the respective cells presented as Nyquist plots (b-c). The data shown is at 600 °C for the cells without NLC and at 550 °C for the cells with NLC. The IV measurement with the highest power density was chosen for each cell. Gas flows: 100 ml/min for both H₂ and air unless otherwise stated.

Figure S3. IV- and PV-curves of four SLFCs showing performance difference of cells that should be identical (a). EIS data presented as Nyquist plots (b-c). The data shown is at 600 °C for the cells without NLC and at 550 °C for the cells with NLC. The IV measurement with the highest power density was chosen for each cell. Gas flows: 100 ml/min for both H₂ and air unless otherwise stated.