Supplementary

Design of interfaces in efficient Ln$_2$NiO$_{4+\delta}$ (Ln = La, Pr) cathode for SOFCs application

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FULLPROF refinement of the XRD patterns of the Ln$_2$NiO$_{4+\delta}$ (Ln = La, Pr) films prepared by ESD on a screen printed CGO layer on a CGO pellet.

**Fig. S1** FULLPROF refinement of the XRD patterns of La$_2$NiO$_{4+\delta}$ film deposited by ESD on a screen printed CGO layer on a CGO pellet (Fit parameters: $\chi^2 = 12.2$, Bragg R-factor = 1.272 and RF-factor = 0.637 for La$_2$NiO$_{4+\delta}$; Bragg R-factor = 0.925 and RF-factor = 0.442 for CGO).

**Fig. S2** FULLPROF refinement of the XRD patterns of Pr$_2$NiO$_{4+\delta}$ film deposited by ESD on a screen printed CGO layer on a CGO pellet (Fit parameters: $\chi^2 = 26.2$, Bragg R-factor = 1.13 and RF-factor = 0.601 for Pr$_2$NiO$_{4+\delta}$; Bragg R-factor = 0.556 and RF-factor = 0.319 for CGO).
Oxygen overstoichiometry by TGA:

TGA curves of La$_2$NiO$_{4+\delta}$ and Pr$_2$NiO$_{4+\delta}$ are shown in Fig. S3. Two weight changes are observed: the first one occurs around 350 °C, corresponding to the reduction of Ni$^{3+}$ into Ni$^{2+}$ (the oxygen overstoichiometry being reduced down to $\delta = 0$). The second weight loss characterizes the complete reduction of LnNO into La$_2$O$_3$, Pr$_2$O$_3$ and Ni. The $\delta$ value has been calculated from both values of the weight changes using mole conservation principle.

![Fig. S3 TGA curves of La$_2$NiO$_{4+\delta}$ and Pr$_2$NiO$_{4+\delta}$.]
Micrographs of sample 5:

**Fig. S4** shows the SEM micrographs of sample 5, the LaNO cross section and the CGO-LaNO composite base-layer of the LaNO triple layer design on CGO substrate using sequentially SP and ESD.

![SEM micrographs](image)

**Fig. S4** SEM micrographs of sample 5: (a) cross section, (b) CGO-LaNO sub-layer on CGO electrolyte.
The experimental and fitted impedance spectra at 600 °C from the inset of Fig. 8b, is shown in Fig. S5 along with capacitances and exponents of CPE for all 6 architectures in Table S1.

**Fig. S5** Nyquist plots recorded at 600 °C in air at OCV for sample 6. The numbers indicate the logarithm of the measuring frequency.

**Table S1** Capacitances (F cm⁻²) and exponents of CPE for all six architectures (estimated error is < 5%).

<table>
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<th>Sample</th>
<th>CPE₁</th>
<th>CPE₂</th>
<th>CPE₃</th>
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<tr>
<td></td>
<td>n</td>
<td>Q</td>
<td>C₁</td>
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<td>Sample 1</td>
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<td>Sample 5</td>
<td>0.9055</td>
<td>0.0088</td>
<td>3.3x10⁻³</td>
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<tr>
<td>Sample 6</td>
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