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Supplementary Information

On the role of surfaces and interfaces on electrochemical performance and long-term stability of nanostructured LSC thin film electrodes

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Fig. S1. *X*-ray photoelectron spectra of La 3d, Sr 3d, Co 2p, O 1s core-level spectra of the a) as-grown and b) annealed films prepared at 500 °C, 300 °C, and R.T. Main and satellite peak positions are indicated in each plot.



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Fig. S2. Typical plan-view SEM images of LSC films deposited at (a) 500 °C, (b) 300 °C, and (c) room temperature (R.T.), utilized for symmetrical cells after testing at 700 °C for 300 h in air. (d)-(f): corresponding cross-sectional SEM images.

Fig. S2 shows the typical SEM images of the cells after long-term testing. To obtain the plan-view SEM images, the current-collector LSC paste was carefully detached in order to observe the surface of the LSC nanostructure underneath. The cross-sections were obtained by fracturing the cell. Comparing these images with those of the bare LSC thin films annealed in air (Fig. 2 in the main text), we can observe that the LSC grains have undergone further sintering, leading to the increased grain sizes as well as increased pore sizes. For the heated samples, the microstructures have further densified and led to the appearance of well-crystallized grains on the surface. These results indicate that the porosity remained in the room-temperature sample, thus suppressing the sintering effect, whereas this is promoted in the heated samples. This suggests that the presence of initial porosity in the film is beneficial in preventing excessive sintering and densification. Furthermore, the porosity should be sufficiently distributed across the thickness of the film in order to prevent the columnar grains from sintering fully. On the other hand, we note that the LSCF layers for all samples exhibit similar microstructures and no apparent secondary phases can be observed at the interfaces with either LSC or GDC.