

## Electronic Supporting Information

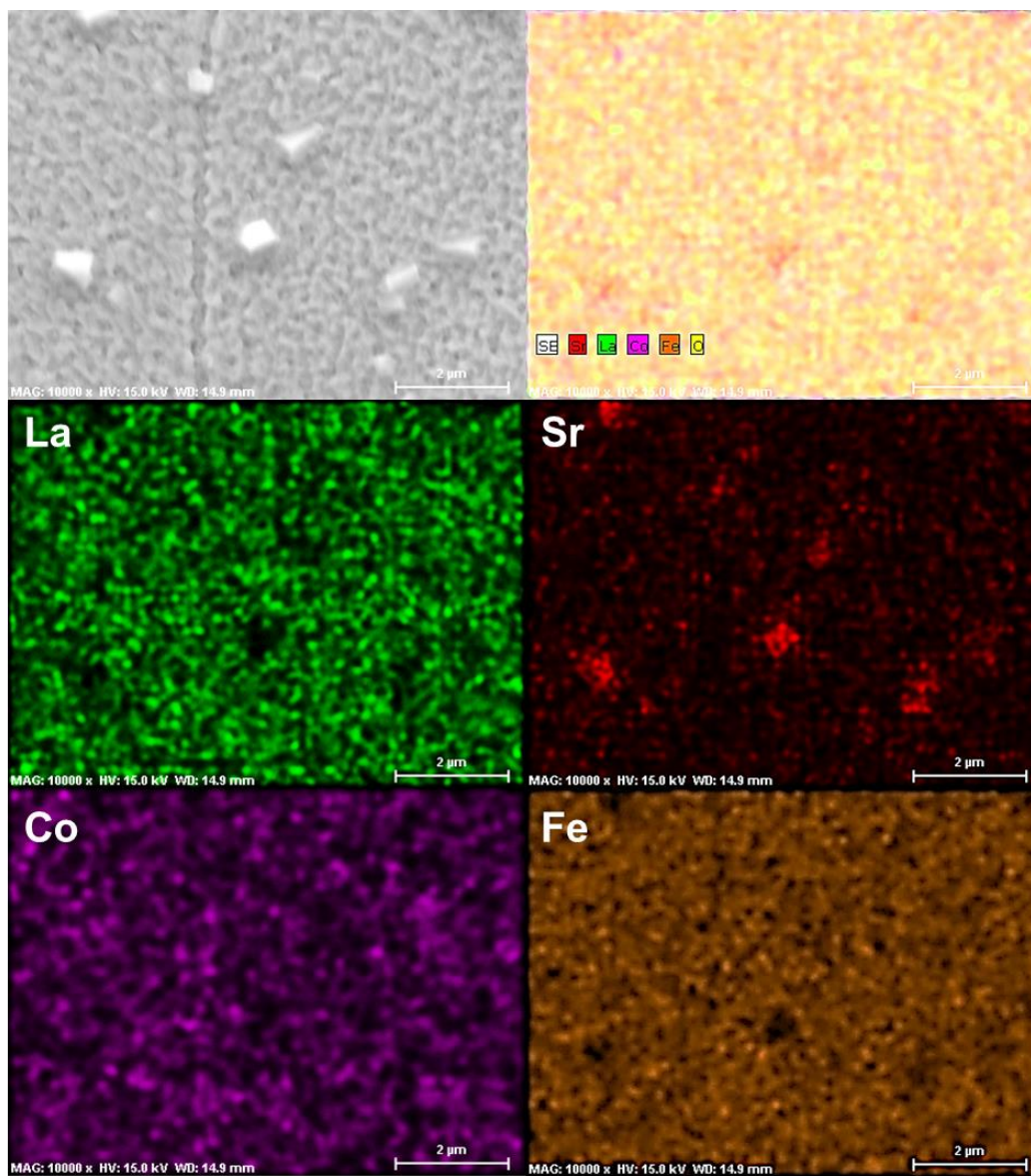
### Effects of Surface Chemical Potentials on Cation Segregation

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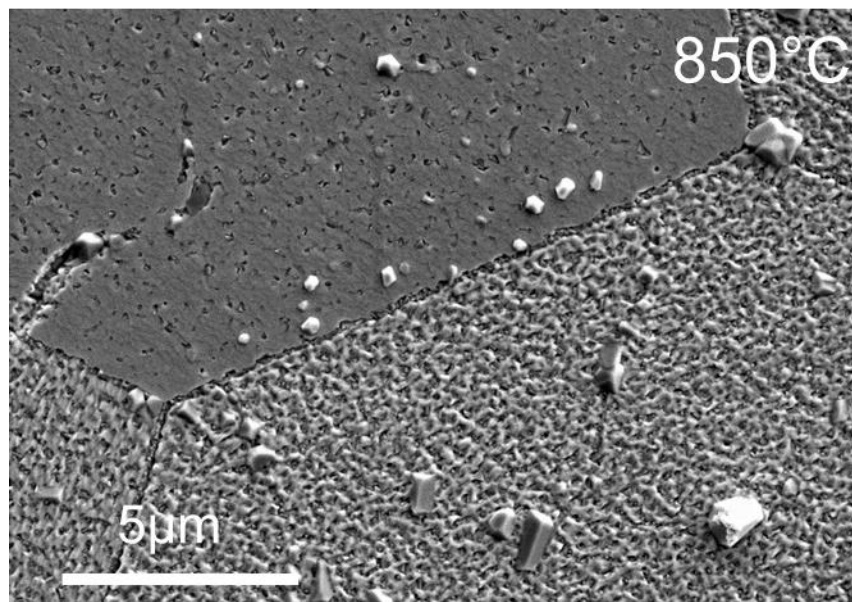
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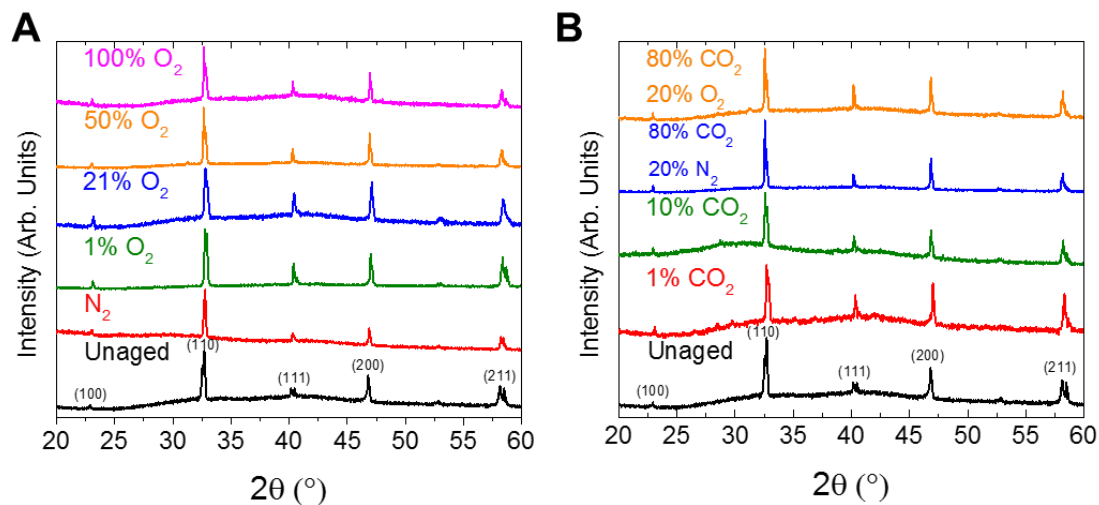
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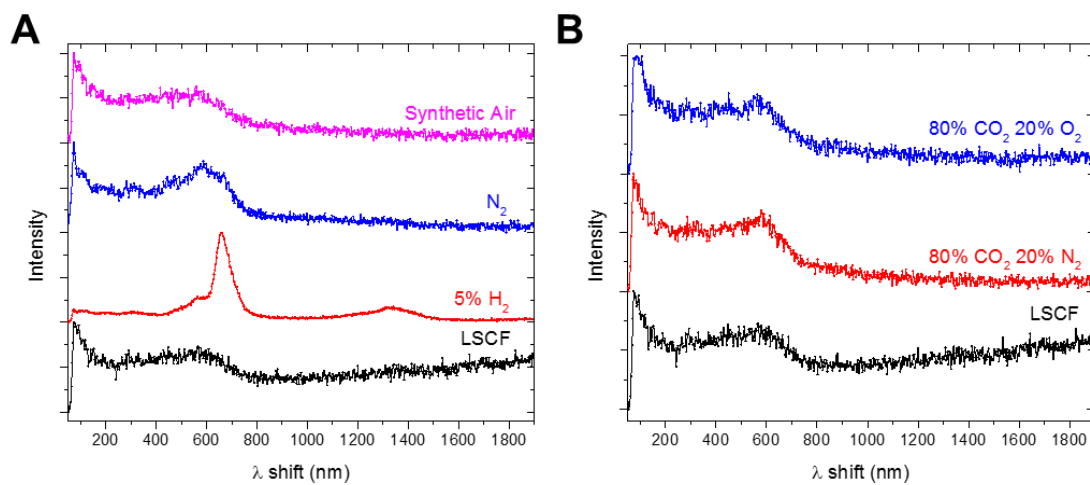
**Fig. S1** EDS on LSCF after aging for 25 hours at 850°C in synthetic air, showing that the main composition of segregated particles is strontium.



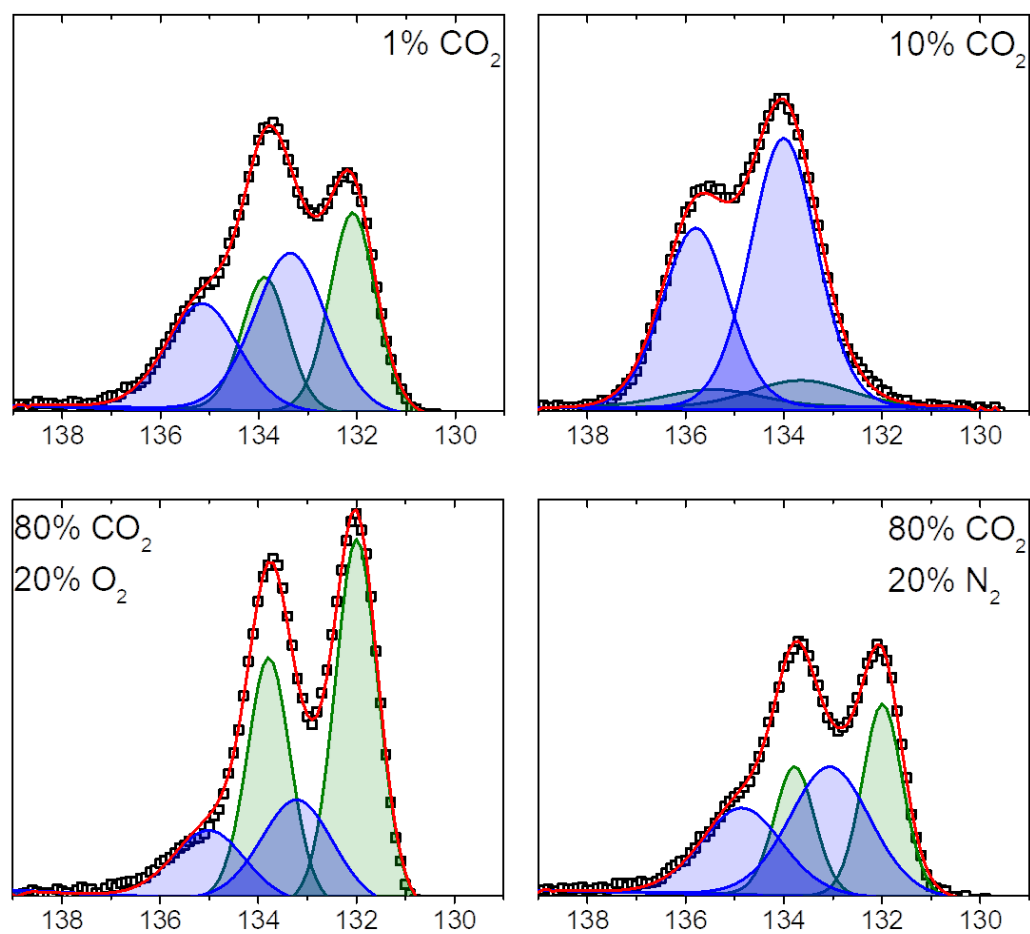
**Fig. S2** SEM of LSCF aged for 25 hours at 850°C in synthetic air, showing the high level of surface degradation



**Fig. S3** XRD of pristine LSCF surface and aged at 700°C for 25 hours in each gas environment. (a) Different oxygen partial pressures: 100%  $O_2$ , 50%  $O_2$ , 21%  $O_2$ , 1%  $O_2$ , 100%  $N_2$ . (b) Different carbon dioxide oxygen partial pressures: 80%  $CO_2$  / 20%  $O_2$ , 80%  $CO_2$  / 20%  $N_2$ , 10%  $CO_2$  / 20%  $O_2$ , 70%  $N_2$ , and 1%  $CO_2$  / 20%  $O_2$ , 79%  $N_2$ .



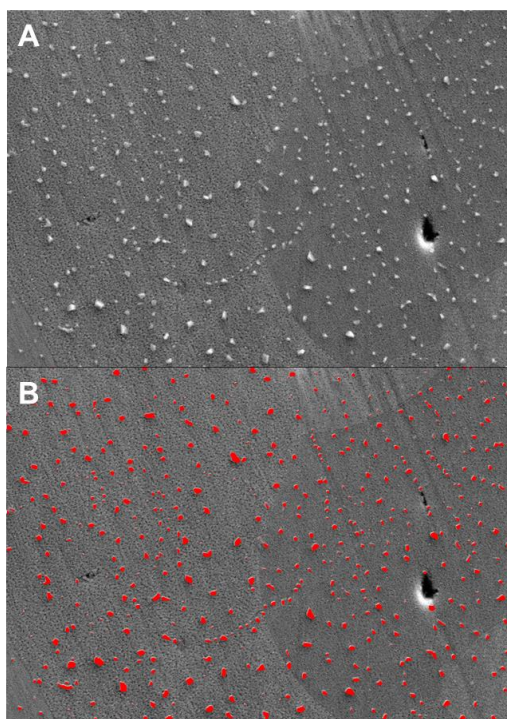
**Fig. S4** Raman spectroscopy of pristine LSCF surface and aged for 25 hours at 700°C in each gas environment (a) without  $CO_2$  (b) with  $CO_2$ .



**Fig. S5** CO<sub>2</sub> Effects on Surface Chemistry of LSCF. XPS of Sr 3d spectra in different CO<sub>2</sub> environments. LSCF samples were aged at 700°C for 25 hours in the listed environment. LSCF lattice Sr peaks are shown in green and surface segregated Sr peaks are shown in blue.

Peak Class	Unaged	1% O <sub>2</sub>	20% O <sub>2</sub>	50% O <sub>2</sub>	100% O <sub>2</sub>	1% CO <sub>2</sub>	10% CO <sub>2</sub>	80% CO <sub>2</sub> 20% O <sub>2</sub>	80% CO <sub>2</sub> 20% N <sub>2</sub>
Sr3d <sub>5/2</sub> - Surface	1.00	1.33	1.86	1.79	1.03	1.69	2.63	1.12	1.54
Sr3d <sub>5/2</sub> - Bulk	1.00	1.96	0.66	0.59	1.43	1.36	0.38	2.34	1.15

**Fig. S6** Figure S6. Table of XPS spectra values of the area under each curve, normalized to Sr3d<sub>5/2</sub> – Bulk peak intensity.



**Fig. S7** Overview of procedure for using Trainable WEKA segmentation in ImageJ to determine particle size distribution (a) original image (b) surface particles identified by Trainable WEKA highlighted in original image.