*Supporting Information*

**Perovskite-type $Nd_{0.75}Sr_{0.25}Co_{0.8}Fe_{0.2}O_{3-\delta}$ cathode for advanced solid oxide fuel cells**

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## Figures

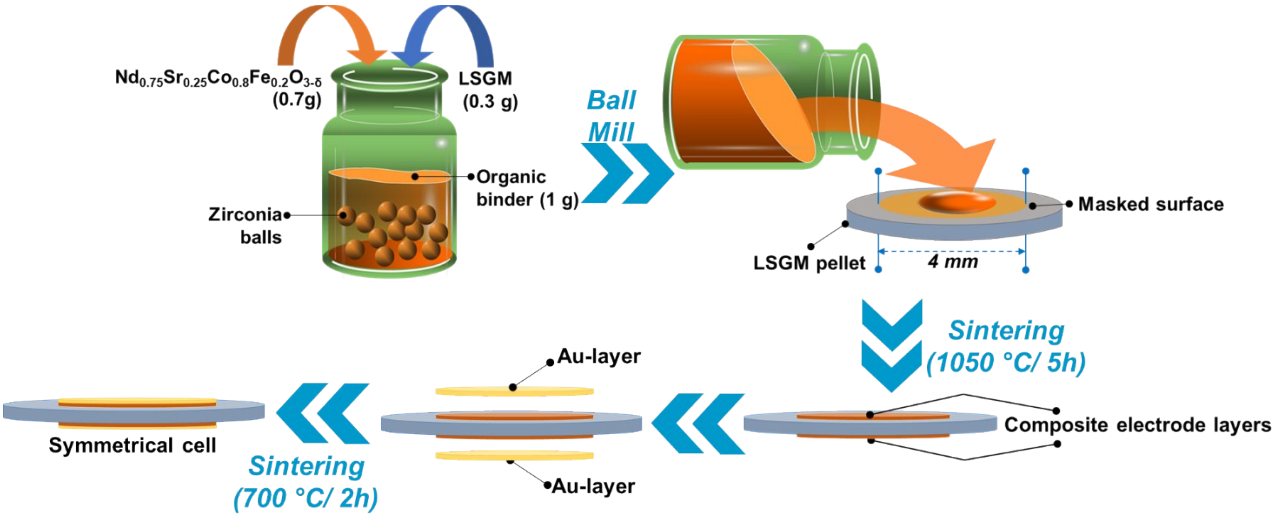


**Figure S1.** Schematic diagram illustrating the half-cell preparation using Nd-based perovskties.

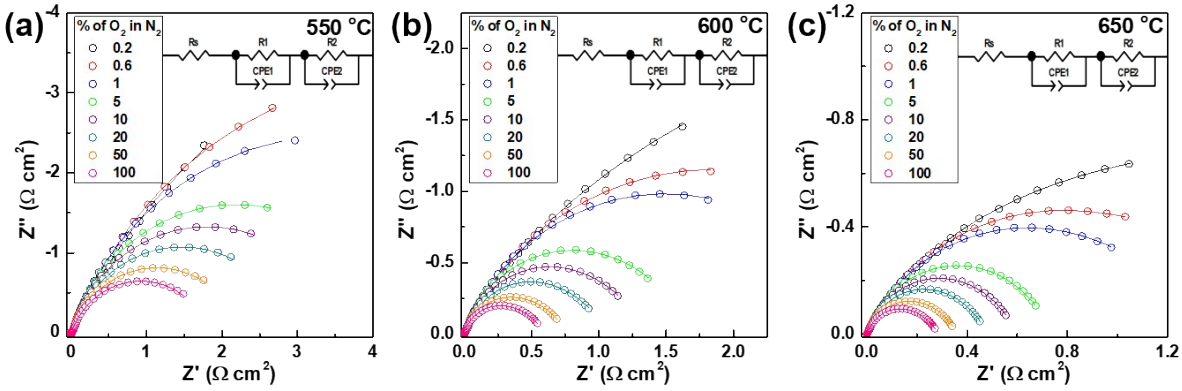


**Figure S2.** Typical Nyquist plot of a symmetrical cell with composite electrode (NSCF+LSGM) layer (electrolyte: LSGM) under OCP at (a) 550 °C, (b) 600 °C and (c) 650 °C with various $pO_2$ concentrations ranging from 0.2-100 % (bal. gas = $N_2$). Data is fitted with two parallel RC circuits (inset) and shown as solid line passing through the original data points (open circles). (Frequency range = 1MHz – 0.1 Hz).

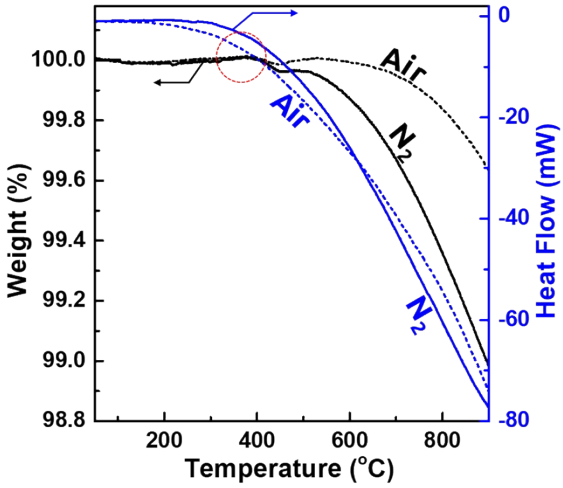


**Figure S3.** TGA (black) and DSC (blue) curves of $Nd_{0.75}Sr_{0.25}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (NSCF) in $N_2$ (solid) and air (dotted), respectively. (Heating rate = 5 °C/min)

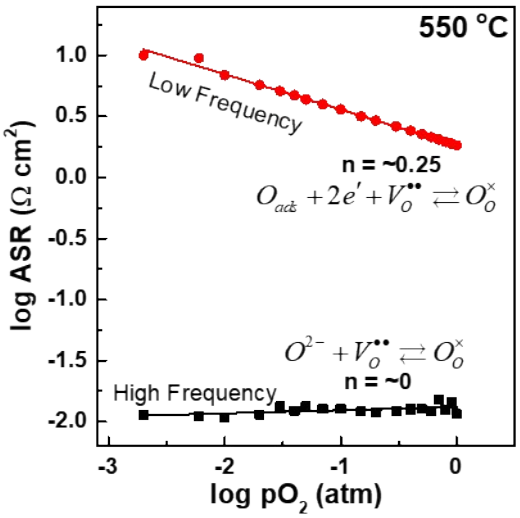


**Figure S4.** Variation of ASRs as a function of $pO_2$ at 550 °C for a symmetrical cell with composite electrode (NSCF+LSGM) layer (electrolyte: LSGM) under OCP. "*n*" indicates the slope obtained from $ASR = ASR_o (pO_2)^{-n}$ and can be translated into the typical reactions occurring at high (■) and low (●) frequency arcs. (Total gas flow = 100 sccm).

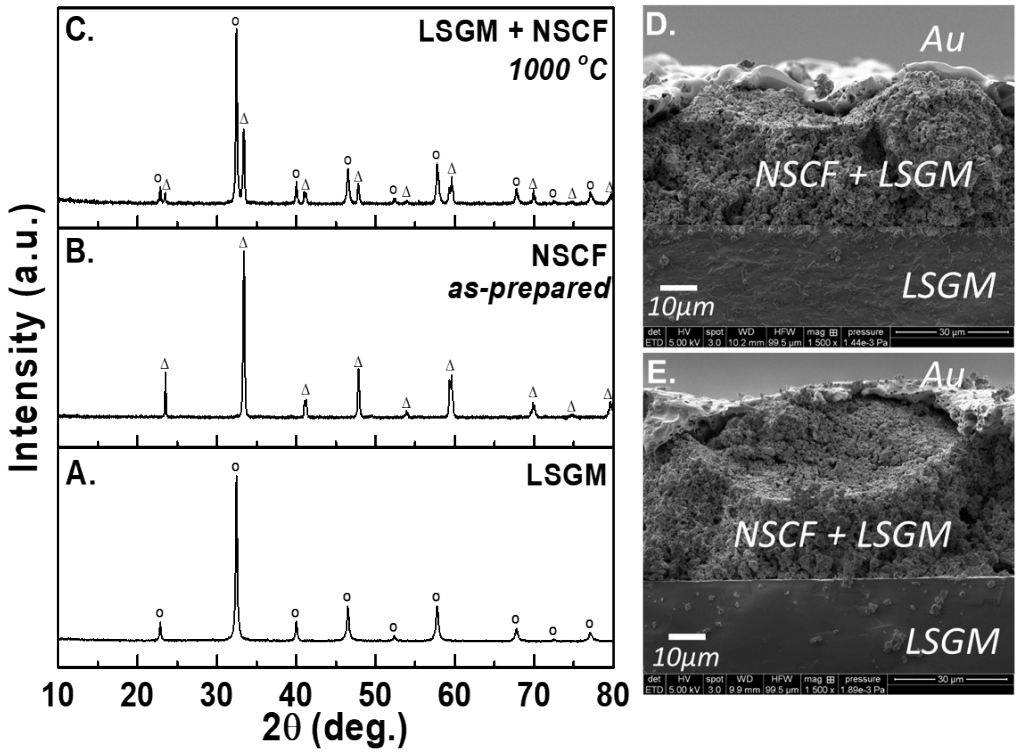


**Figure S5.** PXRD patterns of starting material for composite layer (A) LSGM, (B) as-prepared NSCF and (C) the mixture LSGM+NSCF sintered at 1000 °C for 12 h in air followed by 12 h in $CO_2$. Typical cross-section SEM images of a symmetrical cell composed with dense LSGM electrolyte, NSCF+LSGM composite electrode layer and Au current collector (from bottom to top) (D) before and (E) after the half-cell measurements + 24 h pure $CO_2$ treatment at 800 °C.

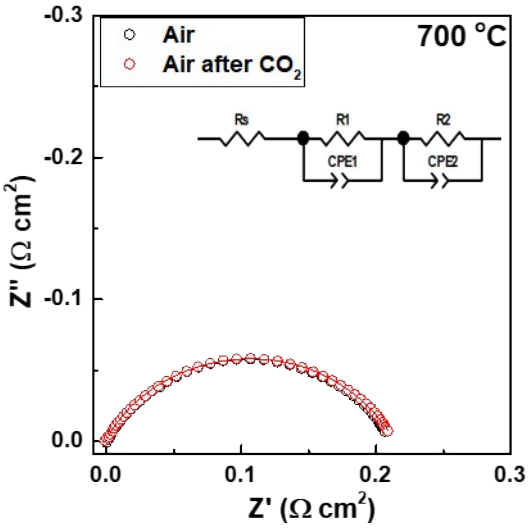


**Figure S6.** Typical Nyquist plot of a symmetrical cell with composite electrode (NSCF+LSGM) layer (electrolyte: LSGM) under OCP at 700 °C in air before (black open circles) and after (red open circles) $CO_2$ exposure for 24 h. Data is fitted with two parallel RC circuits (inset) and shown as solid line passing through the original data points (open circles). Ultrapure $N_2$ was used as balance gas to adjust the $pO_2$. (Frequency range = 1MHz – 0.1 Hz).

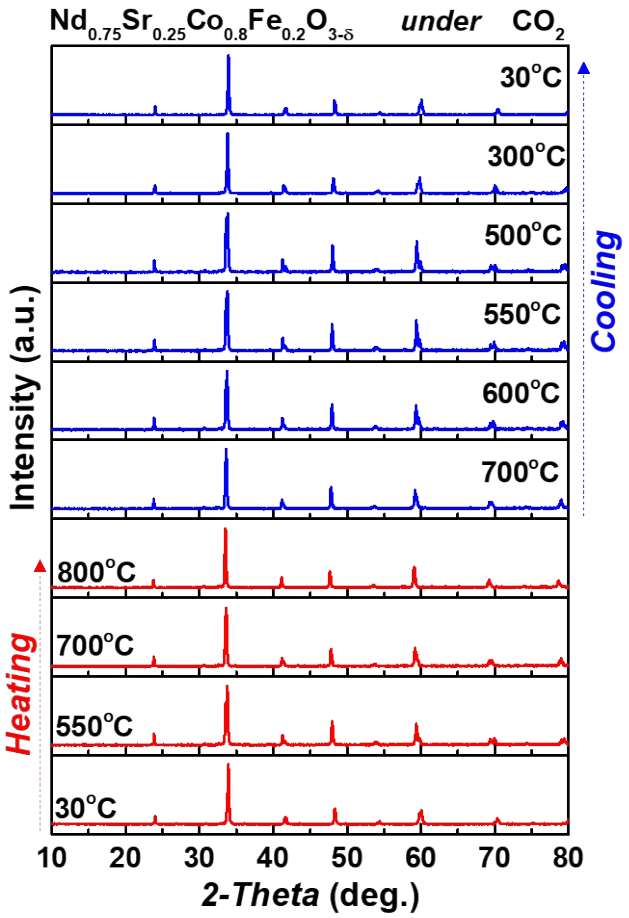


**Figure S7.** In-situ high temperature PXRD patterns of as-prepared $Nd_{0.75}Sr_{0.25}Co_{0.8}Fe_{0.2}O_{3-\delta}$ under pure $CO_2$ obtained from heating (30 – 800 °C; red patterns) and cooling (800 – 30 °C; blue patterns) cycles. (Total gas flow = 20 sccm).

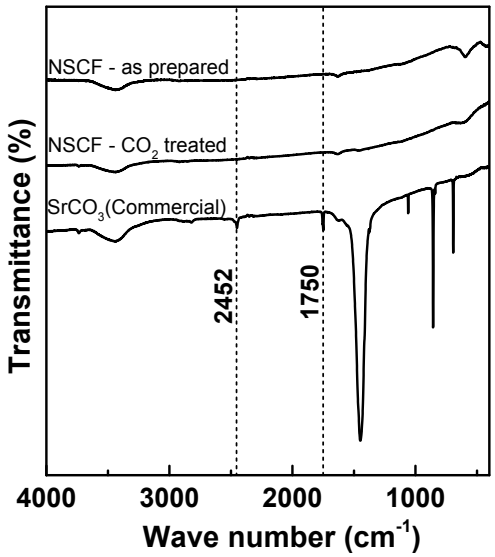


**Figure S8.** FT-IR spectra obtained on $Nd_{0.75}Sr_{0.25}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (NSCF) before and after $CO_2$-treatemnt at 800 °C for 24h.

**Tables**

**Table S1.**: The powder X-ray Rietveld refinement results for $Nd_{0.75}Sr_{0.25}Co_{0.8}Fe_{0.2}O_{3-\delta}$. ($R_p$ = 7.22; $R_{wp}$ = 9.12; $\chi^2$ = 1.23; $a$ = 5.3816(3) Å, $b$ = 7.6218(1) Å, $c$ = 5.4191(7) Å; space group = *Pnma*)

| Atom | Wyckoff-site | *x/a* | *y/b* | *z/c* | Occupancy | $U_{iso}$ (Å$^2$) |
|---|---|---|---|---|---|---|
| Nd | 4*c* | -0.0192(2) | 1/4 | -0.0002(3) | 0.75 | 0.0195(3) |
| Sr | 4*c* | -0.0192(2) | 1/4 | -0.0002(3) | 0.25 | 0.0195(3) |
| Co | 4*b* | 0 | 0 | 1/2 | 0.80 | 0.0222(9) |
| Fe | 4*b* | 0 | 0 | 1/2 | 0.20 | 0.0081(8) |
| O1 | 4*c* | 1/2 | 1/4 | 0.0726 | 1.00 | 0.1016(3) |
| O2 | 8*d* | 0.2459(4) | 0.0239(6) | 0.7947(2) | 0.99 | 0.0257(6) |

**Table S2.** ASRs under various $pO_2$ (~$10^{-2}$ – 1 atm) in the temperature range 550-700 °C.

| $pO_2$ (atm) | $R_p$ (Ω. cm$^2$) | | | |
|---|---|---|---|---|
| | 550 °C | 600 °C | 650 °C | 700 °C |
| 0.02 | 10.04 | 5.82 | 3.12 | 2.14 |
| 0.2 | 2.95 | 1.02 | 0.48 | 0.21 |
| 1 | 1.85 | 0.58 | 0.28 | 0.12 |

**Table S3a.** 2-Theta, *d*-spacing and corresponding (hkl) of $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ (LSGM) and LSGM in composite with NSCF as illustrated in Figs. S5A and S5C, respectively.

| $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ (LSGM) | | | LSGM in composite* | |
|---|---|---|---|---|
| $2\vartheta$ (deg.) | *d-spacing* (Å) | (hkl) | $2\vartheta$ (deg.) | *d-spacing* (Å) |
| 22.847 | 3.8892 | (001) | 22.859 | 3.8872 |
| 32.549 | 2.7561 | (011) | 32.462 | 2.7538 |
| 40.001 | 2.2521 | (111) | 40.019 | 2.2511 |
| 46.499 | 1.9514 | (002) | 46.521 | 1.9505 |
| 52.359 | 1.7459 | (012) | 52.426 | 1.7439 |
| 57.778 | 1.5944 | (112) | 57.767 | 1.5947 |
| 67.759 | 1.3818 | (022) | 67.772 | 1.3816 |
| 72.456 | 1.3033 | (122) | 72.537 | 1.3025 |
| 77.04 | 1.2368 | (013) | 77.074 | 1.2364 |

**Table S3b.** 2-Theta, *d*-spacing and corresponding (hkl) of as-prepared $Nd_{0.75}Sr_{0.25}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (NSCF) and NSCF in composite with LSGM as illustrated in Figs. S5B and S5C, respectively.

| $Nd_{0.75}Sr_{0.25}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (NSCF) | | | NSCF in composite* | |
|---|---|---|---|---|
| $2\vartheta$ (deg.) | *d-spacing* (Å) | (hkl) | $2\vartheta$ (deg.) | *d-spacing* (Å)) |
| 23.477 | 3.7861 | (101) | 23.476 | 3.7863 |
| 33.358 | 2.6838 | (121) | 33.377 | 2.6823 |
| 41.048 | 2.1986 | (022) | 41.019 | 2.1985 |
| 41.222 | 2.1882 | (220) | 41.246 | 2.1869 |
| 47.78 | 1.902 | (202) | 47.806 | 1.9011 |
| 47.891 | 1.8979 | (040) | 47.931 | 1.8964 |
| 53.877 | 1.7003 | (222) | 53.93 | 1.6987 |
| 59.32 | 1.5566 | (123) | 59.399 | 1.5547 |
| 59.561 | 1.5509 | (240) | 59.621 | 1.5494 |
| 69.839 | 1.3456 | (242) | 69.898 | 1.3447 |
| 70.039 | 1.3423 | (400) | 70.101 | 1.3413 |
| 74.713 | 1.2695 | (143) | 74.899 | 1.2668 |
| 79.579 | 1.2036 | (323) | 79.694 | 1.2022 |

**composite made of 1:1 weight ratio of LSGM+NSCF.*