Supporting Information

A comprehensive investigation of Sr segregation effects on the high-temperature oxygen evolution reaction rate

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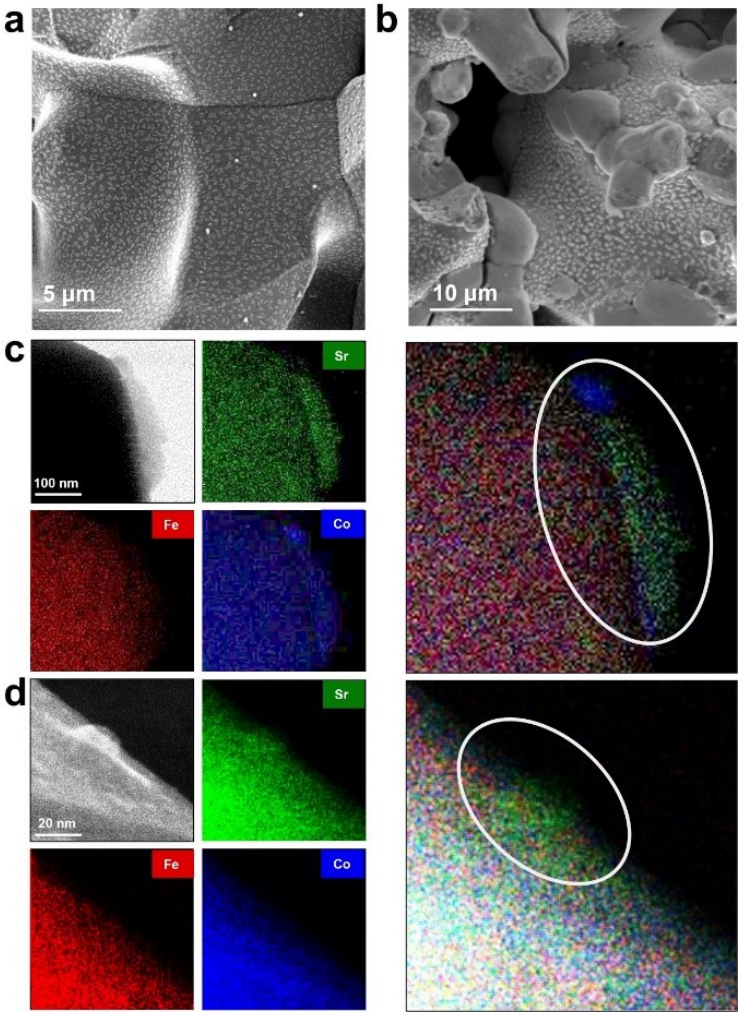
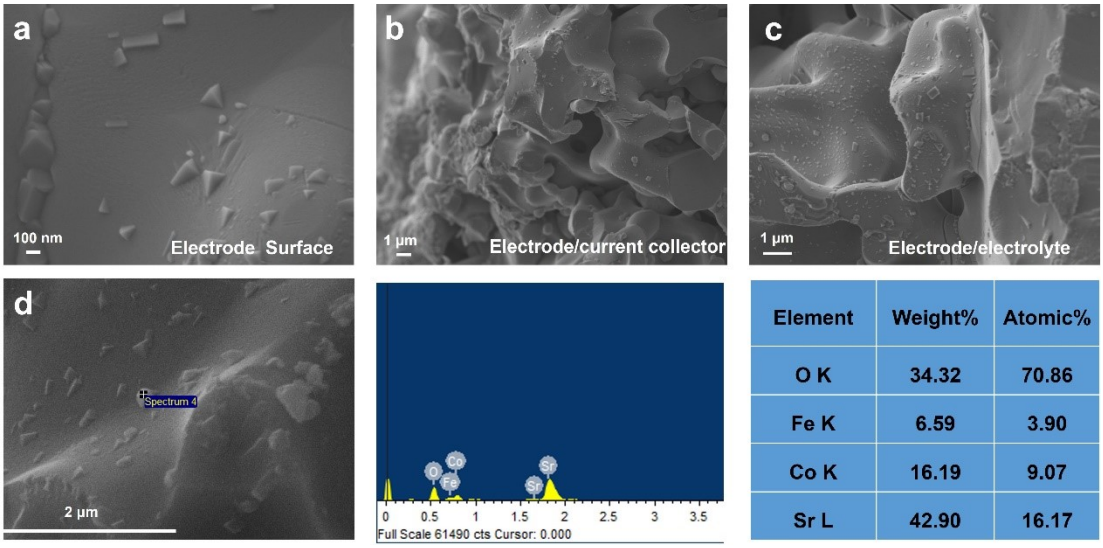


Fig. S1 | (a) SEM images, and **(c)** EDS elemental maps of SCF-SDC anode after O_2 treatment at 800 °C for 3 h, and **(b)** SEM images, and **(d)** EDS elemental maps of SCF-SDC anode after stability test for 487 h.



Element	Weight%	Atomic%
O K	34.32	70.86
Fe K	6.59	3.90
Co K	16.19	9.07
Sr L	42.90	16.17

Fig. S2 | SEM images SCF anode after treatment for 1 h at pure O_2 atmosphere. **(a)** SCF anode surface, (b) electrode/current collector interface, (c) electrode/electrolyte interface, (d) EDX of Sr aggregation at the boundaries.

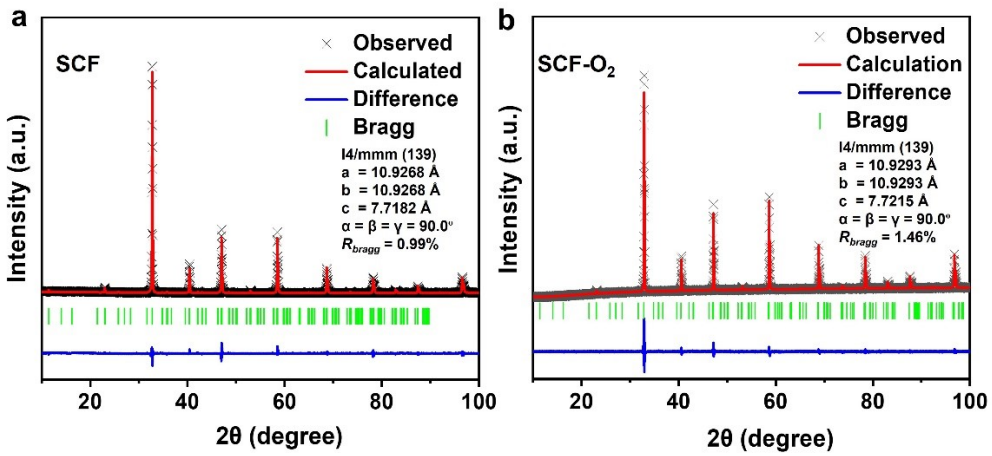


Fig. S3 | Rietveld refinement results of the XRD patterns for **(a)** SCF and **(b)** SCF-O_2.

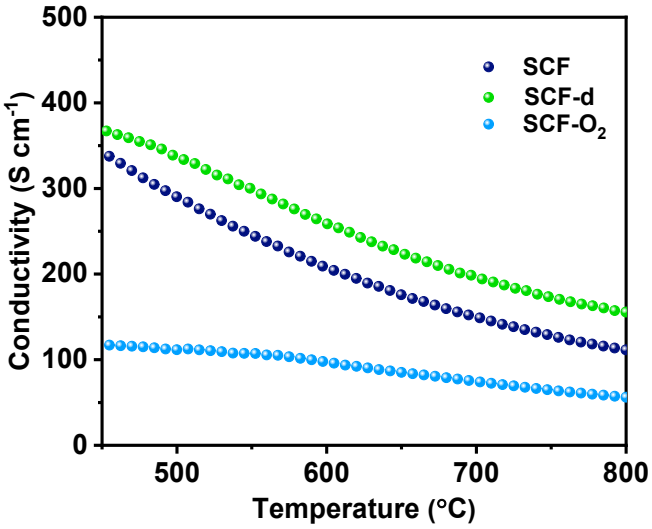


Fig. S4 | Electrical conductivity curves of SCF, SCF-d and SCF-O_2 at different temperatures in air.

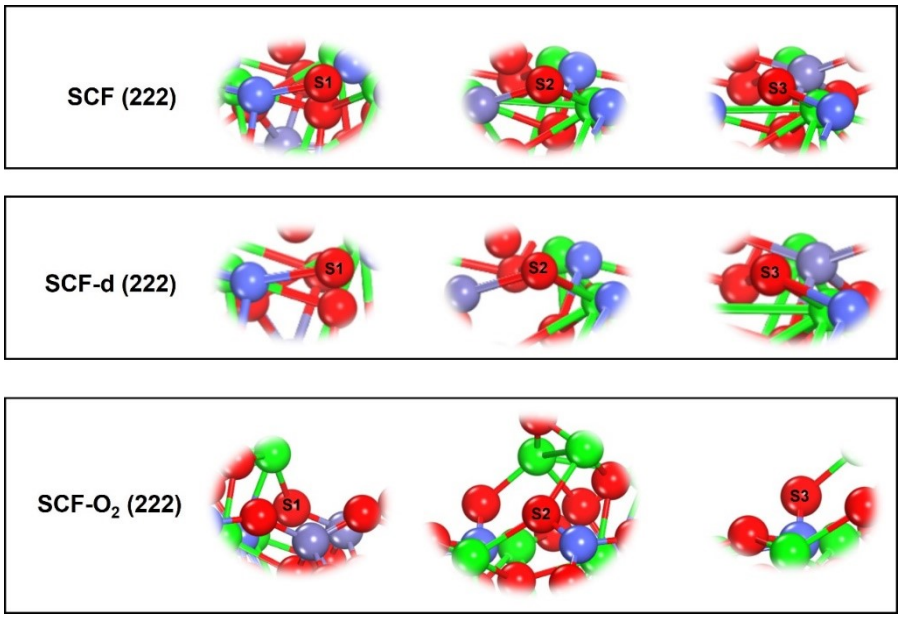


Fig. S5 | The different surface or interface oxygen vacancy sites.

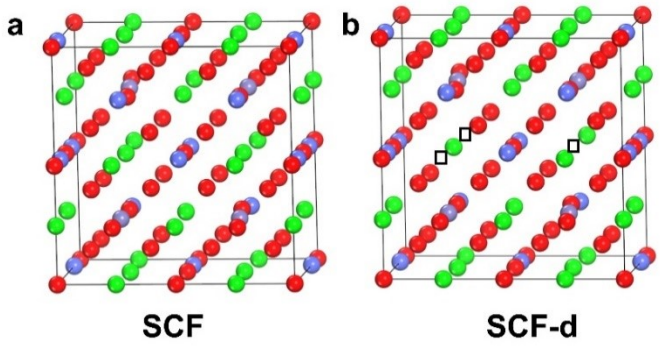


Fig. S6 | Models used for the DFT calculation. **(a)** SCF and **(b)** SCF-d for the calculation of E_{vfb}. Color code: O (red ball), Sr (green ball), Co (dark blue ball), Fe (light blue ball), respectively.

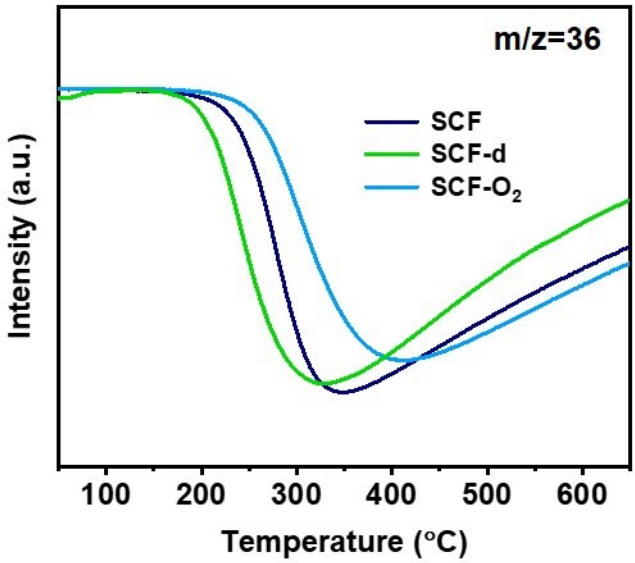


Fig. S7 | $^{18}O_2$ isotope exchange profiles of different samples.

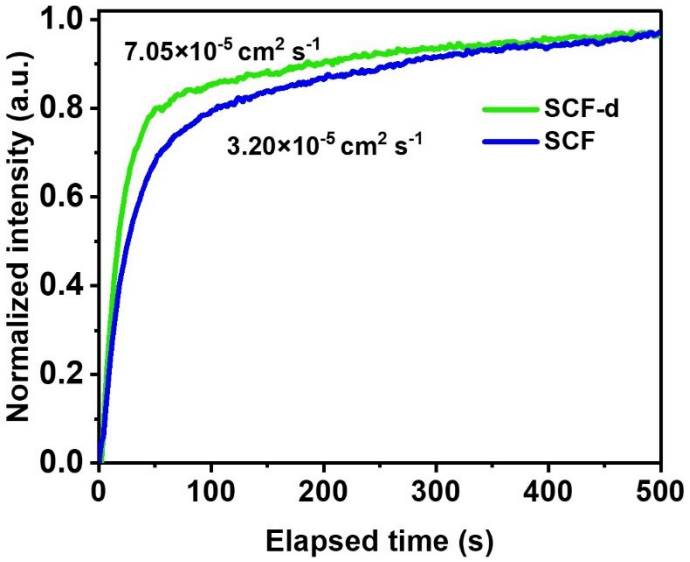


Fig. S8 | ECR curves of SCF and SCF-d in air at 800 °C.

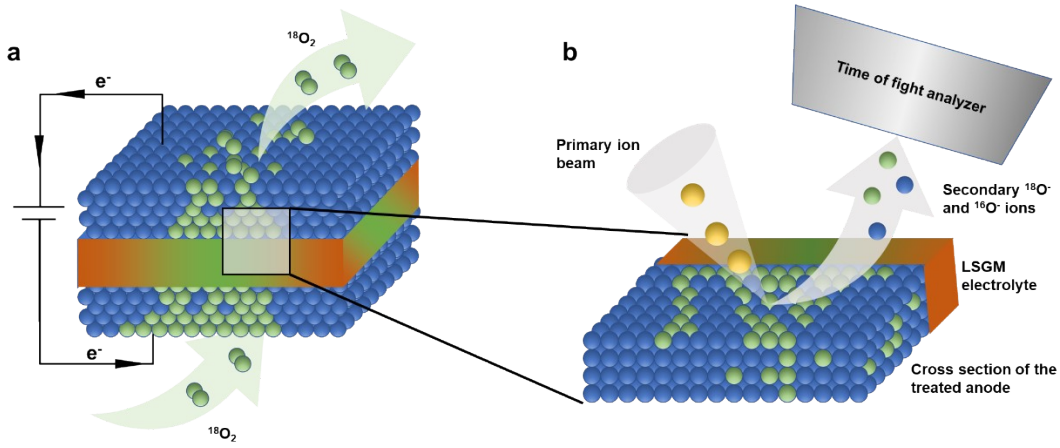


Fig. S9 | Schematic diagram of *quasi in-situ* TOF-SIMS measurement. **(a)** The pre-treatment of SOEC with different anode by $^{18}O_2$ gas, and the grey box is the test area for elemental mapping of TOF-SIMS. **(b)** Schematic diagram of TOF-SIMS measurement. Color code: ^{18}O (green balls), ^{16}O (blue balls), and Bi (yellow balls).

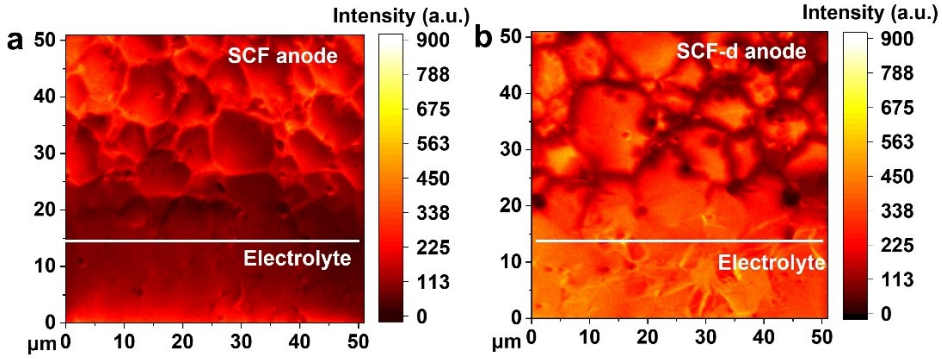


Fig. S10 | Two-dimensional spectra of TOF-SIMS for different samples. Total O^- signal intensity distribution of **(a)** SCF and **(b)** SCF-d.

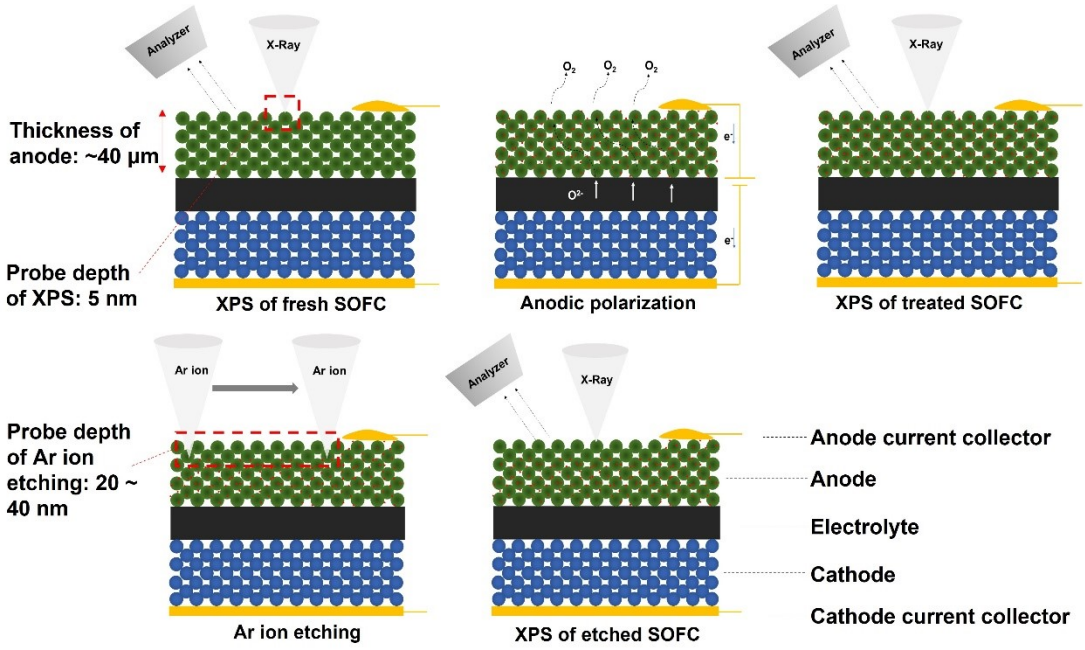


Fig. S11 | Schematic diagram of Ar ion etching and XPS measurements.

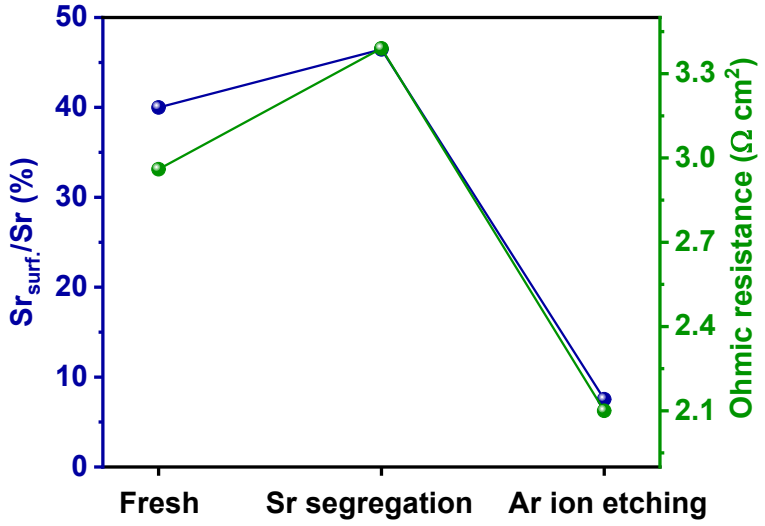


Fig. S12 | Variation of ohmic resistance at OCV and the amount of surface Sr species of SCF electrode under different conditions.

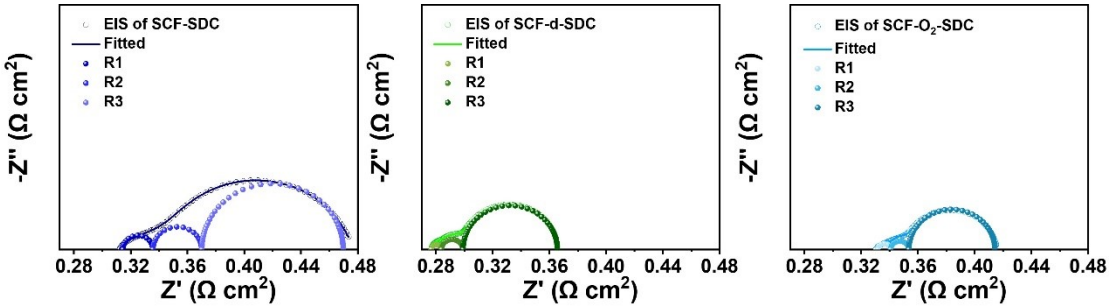


Fig. S13 | The CNLS fitting results of EIS of SOECs with different anodes.

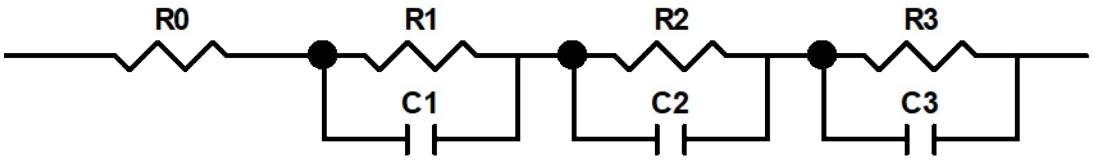


Fig. S14 | Equivalent circuit for the CNLS fitting of EIS curves.

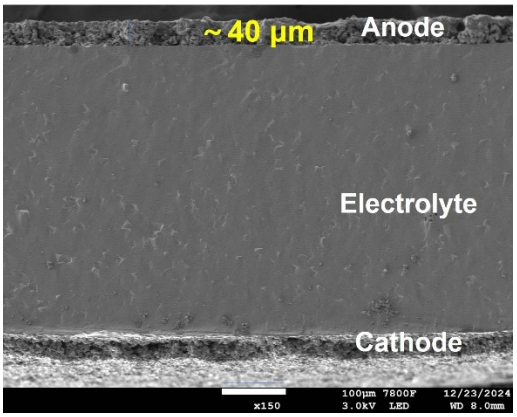


Fig. S15 The SEM image of the SOEC.

Table S1 | Fitting results of Sr 3*d* XPS spectra of SCF and SCF-d.

Sample	**Binding energy (eV)**				**$Sr_{sur.}$/Sr (%)**
	Sr^{3+} (3/2)	Sr^{4+} (3/2)	Sr^{3+} (1/2)	Sr^{4+} (1/2)	
SCF	132.82	134.13	134.60	135.93	39.99
SCF-O_2	131.12	132.76	133.30	134.77	60.60

Table S2 | Positron annihilation lifetime measurement results for SCF and SCF-O_2.

Sample	C_1		C_2		C_3	
	τ_1 (ps)	I_1 (%)	τ_2 (ps)	I_2 (%)	τ_3 (ps)	I_3 (%)
SCF	102.1	21.2	199.5	78.0	1384	0.856
SCF-O_2	112.8	37.9	207.1	61.2	1527	0.91

Table S3 | Rietveld refinement parameters of SCF sample.

Atom	Wyck.	s.o.f.	x	y	z
O1	16m	0.909569	0.119014	0.119014	0.277600
O2	16k	0.894607	0.124127	0.624127	0.250000
Sr1	8j	1.000000	0.242032	0.500000	0.000000
Sr2	8i	1.000000	0.254532	0.000000	0.000000
O3	8h	1.000000	0.237501	0.237501	0.000000
Fe1	8f	0.300000	0.250000	0.250000	0.250000
Fe2	4e	0.300000	0.000000	0.000000	0.250000
Fe3	4d	0.300000	0.000000	0.500000	0.250000
O4	4c	1.000000	0.000000	0.500000	0.000000
O5	2a	1.000000	0.000000	0.000000	0.000000
Co1	8f	0.700000	0.250000	0.250000	0.250000
Co2	4e	0.700000	0.000000	0.000000	0.250000
Co3	4d	0.700000	0.000000	0.500000	0.250000

Table S4 | Rietveld refinement parameters of SCF-O_2 sample.

Atom	Wyck.	s.o.f.	x	y	z
O1	2a	0.494631	0.000000	0.000000	0.277600
O2	16m	0.609941	0.119000	0.119000	0.000000
Sr1	8j	1.000000	0.242300	0.500000	0.000000
Sr2	8i	1.000000	0.251000	0.000000	0.000000
O3	8h	1.000000	0.234000	0.234000	0.000000
Fe1	4e	0.300000	0.000000	0.000000	0.250000
Fe2	8f	0.300000	0.250000	0.250000	0.250000
Fe3	4d	0.300000	0.000000	0.500000	0.250000
O4	16k	1.000000	0.123700	0.623700	0.250000
O5	4c	1.000000	0.000000	0.500000	0.000000
Co1	4e	0.700000	0.000000	0.000000	0.250000
Co2	4e	0.700000	0.000000	0.000000	0.250000
Co3	8f	0.700000	0.250000	0.250000	0.250000

Table S5 | Fitting results of Fe 2*p* XPS spectra of SCF and SCF-d.

Sample	**Binding energy (eV)**					**Fe^{4+}/Fe (%)**
	Fe^{3+} (3/2)	Fe^{4+} (3/2)	Fe^{3+} (1/2)	Fe^{4+} (1/2)	Satellite	
SCF	709.45	711.39	722.79	724.22	714.67	46.17
SCF-d	709.47	711.23	722.78	724.36	714.88	52.50

Table S6 | Fitting results of Co 2*p* XPS spectra of SCF and SCF-d.

Sample	**Binding energy (eV)**					**Co^{4+}/Co (%)**
	Co^{3+} (3/2)	Co^{4+} (3/2)	Co^{3+} (1/2)	Co^{4+} (1/2)	Satellite	
SCF	781.01	779.71	796.44	794.94	784.71	43.65
SCF-d	781.18	779.78	796.68	795.12	784.73	47.66

Table S7 | ^{57}Fe-Mössbauer parameters for SCF and SCF-d.

Sample	Sub-spectra	Fe species	IS (mm s^{-1})	QS (mm s^{-1})	Content (%)
SCF	D1	Fe^{3+} (CN=5)	0.23(1)	0.28(3)	37.58
	D2	Fe^{4+} (CN=6)	-0.07(2)	0.50(3)	33.06
	D3	Fe^{3+} (CN=6)	0.29(3)	0.56(4)	18.70
	S1	Fe^{4+} (CN=6)	-0.17(1)	-	10.66
SCF-d	D1	Fe^{3+} (CN=5)	0.27(1)	0.28(3)	27.29
	D2	Fe^{4+} (CN=6)	-0.02(1)	0.51(5)	37.88
	D3	Fe^{3+} (CN=6)	0.33(3)	0.56(4)	20.95
	S1	Fe^{4+} (CN=6)	-0.12(2)	-	13.88

The reference used for the IS test is α-Fe

Table S8 | E_{vfs} of different sites in SCF, SCF-d and SCF-O_2.

Sample	Surface oxygen vacancy formation energy (eV)		
	Site 1	**Site 2**	**Site 3**
SCF	0.90	2.19	0.40
SCF-d	0.72	2.00	0.23
SCF-O_2	2.08	3.21	0.48

Table S9 | E_{vfb} of different sites in SCF and SCF-d.

Sample	Bulk oxygen vacancy formation energy (eV)			
	Site 1	**Site 2**	**Site 3**	**Site 4**
SCF	1.10	1.00	0.78	0.73
SCF-d	-0.12	-0.12	-0.66	-0.33

Table S10 | *In-situ* XPS analysis of O 1*s* spectra for SCF and SCF-d anodes under different conditions.

Sample	Binding energy (eV)				$O_{ads.}/O$ (%)
	H_2O	OH-	O^-/O_2^{2-}	$O_{lat.}$	
SCF-SDC RT, OCV	532.50	531.10	529.76	528.41	61.69
SCF-SDC 1000 K, OCV	532.50	531.02	529.63	528.40	26.01
SCF-SDC 1000 K, 1 mA	532.49	531.00	529.62	528.41	32.06
SCF-d-SDC RT, OCV	532.51	531.10	529.74	528.46	63.50
SCF-d-SDC 1000 K, OCV	532.49	530.94	529.60	528.39	25.29
SCF-d-SDC 1000 K, 1 mA	532.57	531.00	529.59	528.45	35.42

Table S11 | Resistance of each electrode process for SOECs with different anodes.

Sample	R1 (Ω cm^2)	R2 (Ω cm^2)	R3 (Ω cm^2)
SCF-SDC	0.0213	0.0344	0.0997
SCF-d-SDC	0.0072	0.0152	0.0665
SCF-O_2-SDC	0.0091	0.0128	0.0607