

Fig. S1. Observed, calculated, and difference NPD patterns for the $x=0.25$ sample. Vertical lines show positions of reflections for CoO, NiO and Pr_{1.75}Sr_{0.25}Ni_{0.75}Co_{0.25}O_{4.11(1)} upwards.

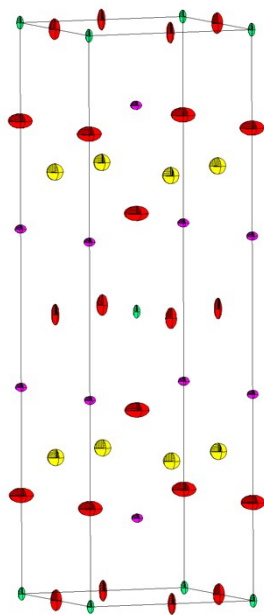


Fig. S2. Atomic displacement ellipsoids in the crystal structure of the $x=0.25$ phase. The oxygen atoms are O1 and O2 (red) as well as O3 (yellow). The cations are Co/Ni (green) and Pr/Sr (violet).

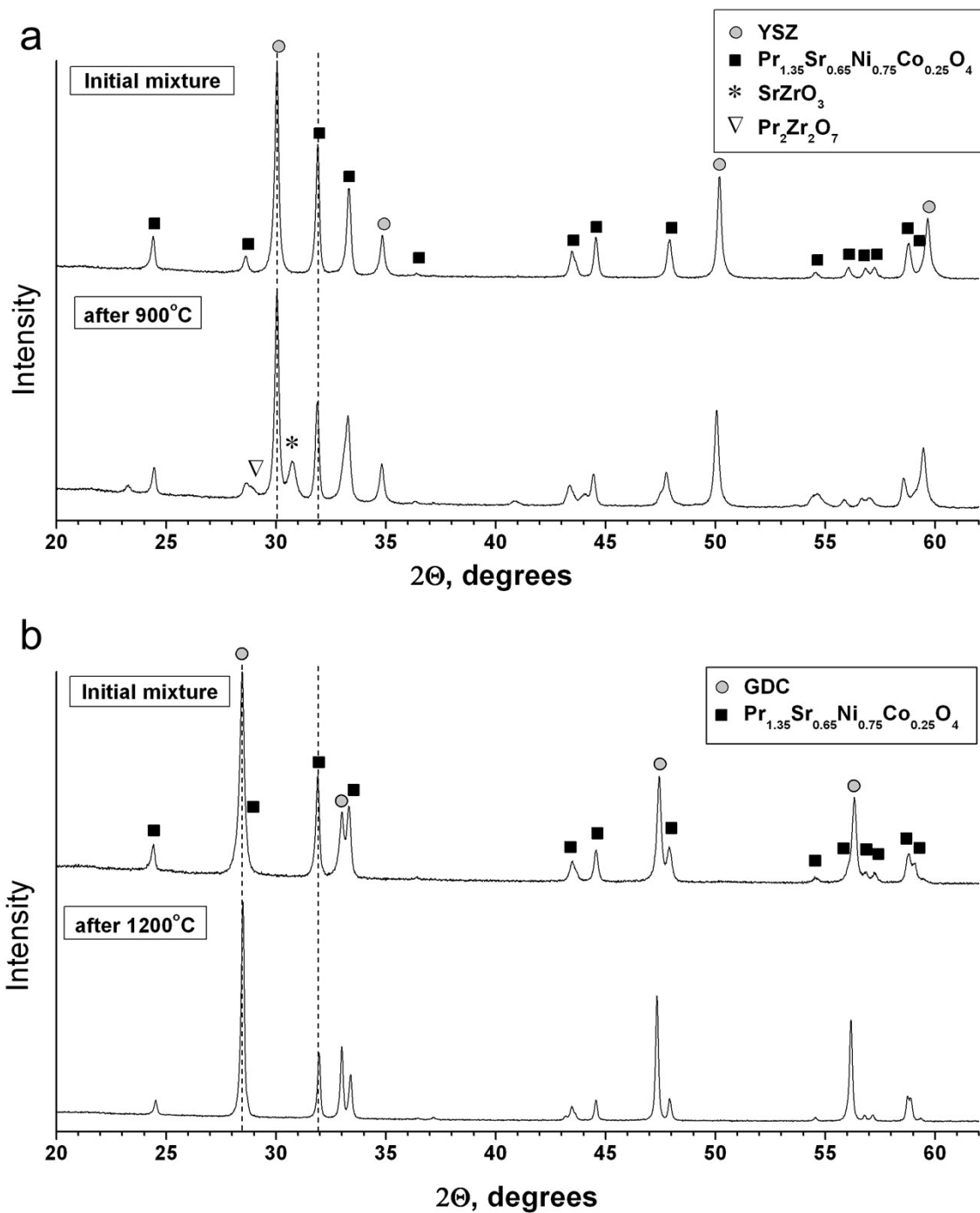


Fig. S3. XRPD patterns of $\text{Pr}_{1.35}\text{Sr}_{0.65}\text{Ni}_{0.75}\text{Co}_{0.25}\text{O}_4/\text{YSZ}$ (a) and $\text{Pr}_{1.35}\text{Sr}_{0.65}\text{Ni}_{0.75}\text{Co}_{0.25}\text{O}_4/\text{GDC}$ (b) mixtures initial (upper patterns) and after annealing for at 900°C (YSZ) and 1200°C (GDC) (lower patterns).

Table S1

Rietveld refinement of the NPD data collected using the POLARIS diffractometer for $\text{Pr}_{2-x}\text{Sr}_x\text{Ni}_{1-x}\text{Co}_x\text{O}_{4+\delta}$ (S.G. $I4/mmm$), $x=0.25$ (first line)^a and 0.75 (second line)^b.

Atom	x	y	z	Occupancy	Atomic displacement parameter ($\times 100 \text{ \AA}^2$) ^c			
					$U_{\text{iso}}/U_{\text{eq}}$	U_{11}	U_{22}	U_{33}
Pr/Sr	0	0	0.36142(5)	0.875/0.125	0.54	0.63(1)	0.63(1)	0.63(1)
	0	0	0.36059(5)	0.625/0.375	0.32	0.37(2)	0.37(2)	0.22(2)
Ni/Co	0	0	0.0	0.75/0.25	0.44	0.227(9)	0.227(9)	0.227(9)
	0	0	0	0.25/0.75	0.21	0.02(2)	0.02(2)	0.02(2)
O1	0	0.5	0.0	1.0	1.02	0.67(2)	0.22(2)	2.18(3)
	0	0.5	0.0	1.0	0.57	0.67(2)	0.15(2)	0.88(3)
O2	0	0	0.17212(6)	1.0	2.30	2.92(2)	2.92(2)	1.04(3)
	0	0	0.16669(5)	1.0	1.07	1.12(2)	1.12(2)	0.99(3)
O3	0	0.5	0.25	0.045(2)	1.4(3)			

^a $a=3.82010(5) \text{ \AA}$; $c=12.3900(2) \text{ \AA}$; $\chi^2=3.91$; $R_p=0.0284$; $R_{\text{wp}}=0.0179$.

^b $a=3.79447(3) \text{ \AA}$; $c=12.3255(1) \text{ \AA}$; $\chi^2=7.08$; $R_p=0.0254$; $R_{\text{wp}}=0.0417$.

^c $U_{12}=U_{13}=U_{23}=0.0$ for all atoms.

Table S2

Selected interatomic distances (Å) in the x=0.25 and x=0.75 phases.

Distances		0.25	0.75	
Ni/Co	- O1	1.91005(2)	1.89724(1)	x4
	O2	2.1325(8)	2.0545(6)	x2
Pr/Sr	- O1	2.5684(4)	2.5597(4)	x4
	O2	2.346 (1)	2.3900(8)	x1
	O2	2.7330 (2)	2.7041(1)	x4
	O3	2.3567(4)	-	