

Supplementary Materials

Synthesis and Properties of the Gallium-Containing Ruddlesden-Popper Oxides with High-Entropy B-Site Arrangement

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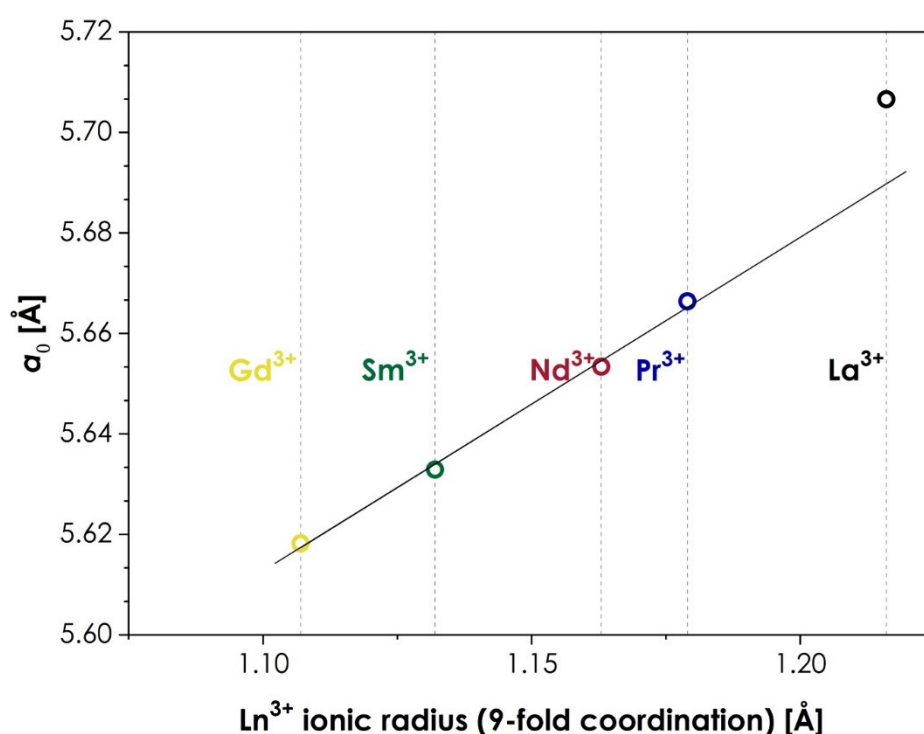


Figure S1. The dependence between the normalized, quasi-cubic lattice parameter a_0 and the ionic radii of the Ln^{3+} ions for the $\text{LnSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_4$ ($\text{Ln} = \text{La, Pr, Nd, Sm, or Gd}$) series. The linear regression line based on the Gd-, Sm-, Nd-, and Pr-based compositions is extrapolated on the value of ionic radius corresponding to La^{3+} .

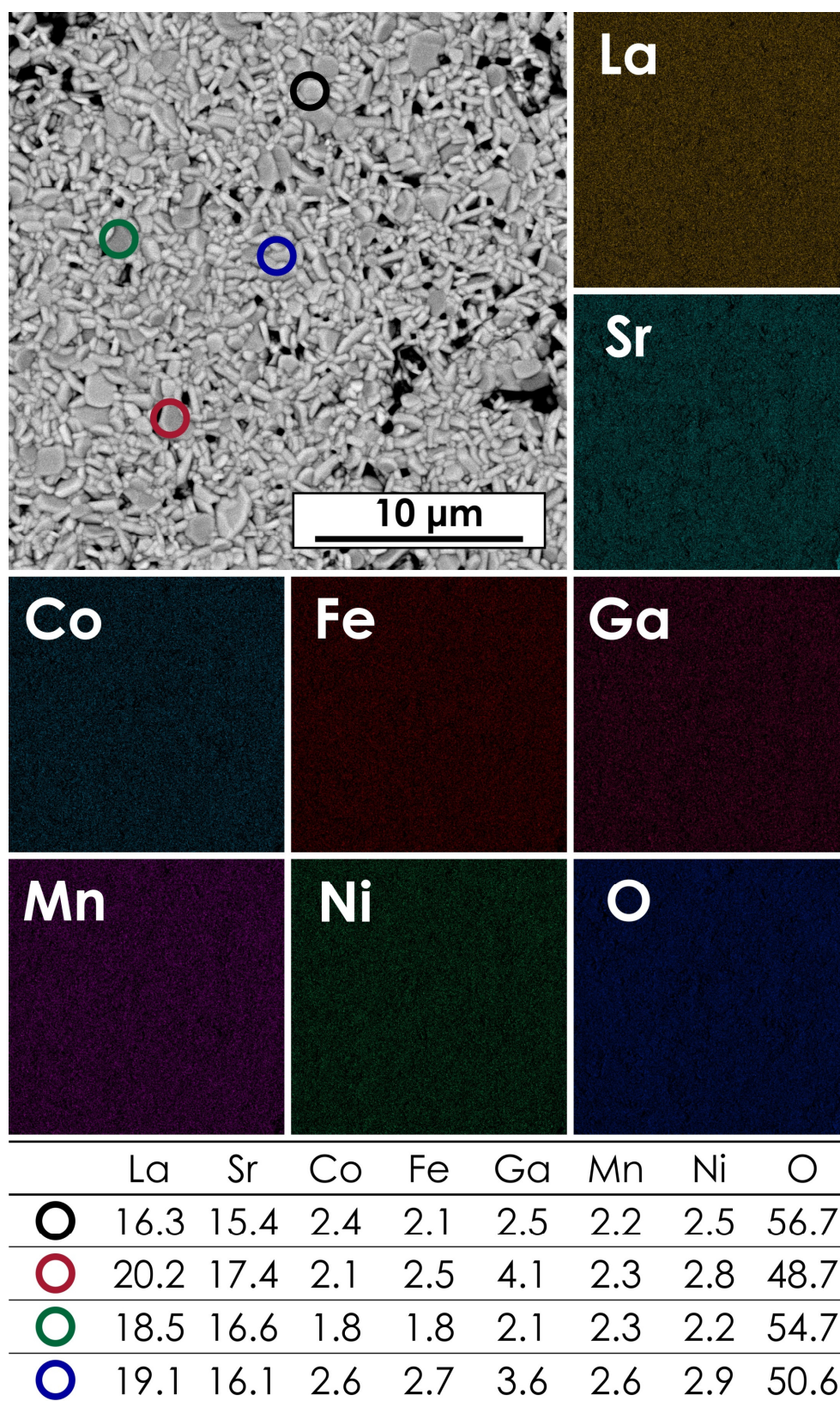


Figure S2. The results of EDS mapping and point analysis for the $\text{LaSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_4$ pellet sintered at 1200 °C for 20 h.

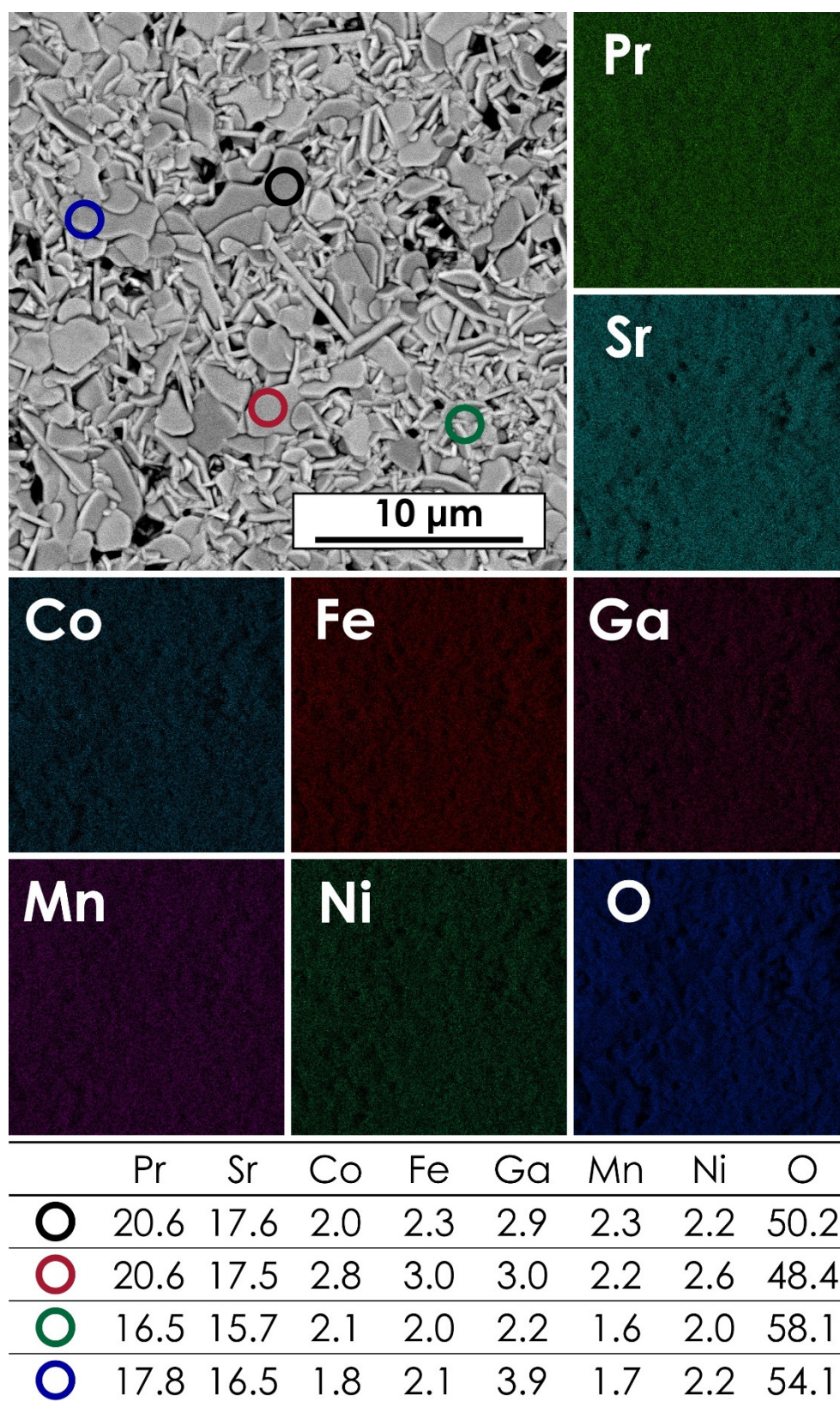


Figure S3. The results of EDS mapping and point analysis for the $\text{PrSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_4$ pellet sintered at 1200 °C for 20 h.

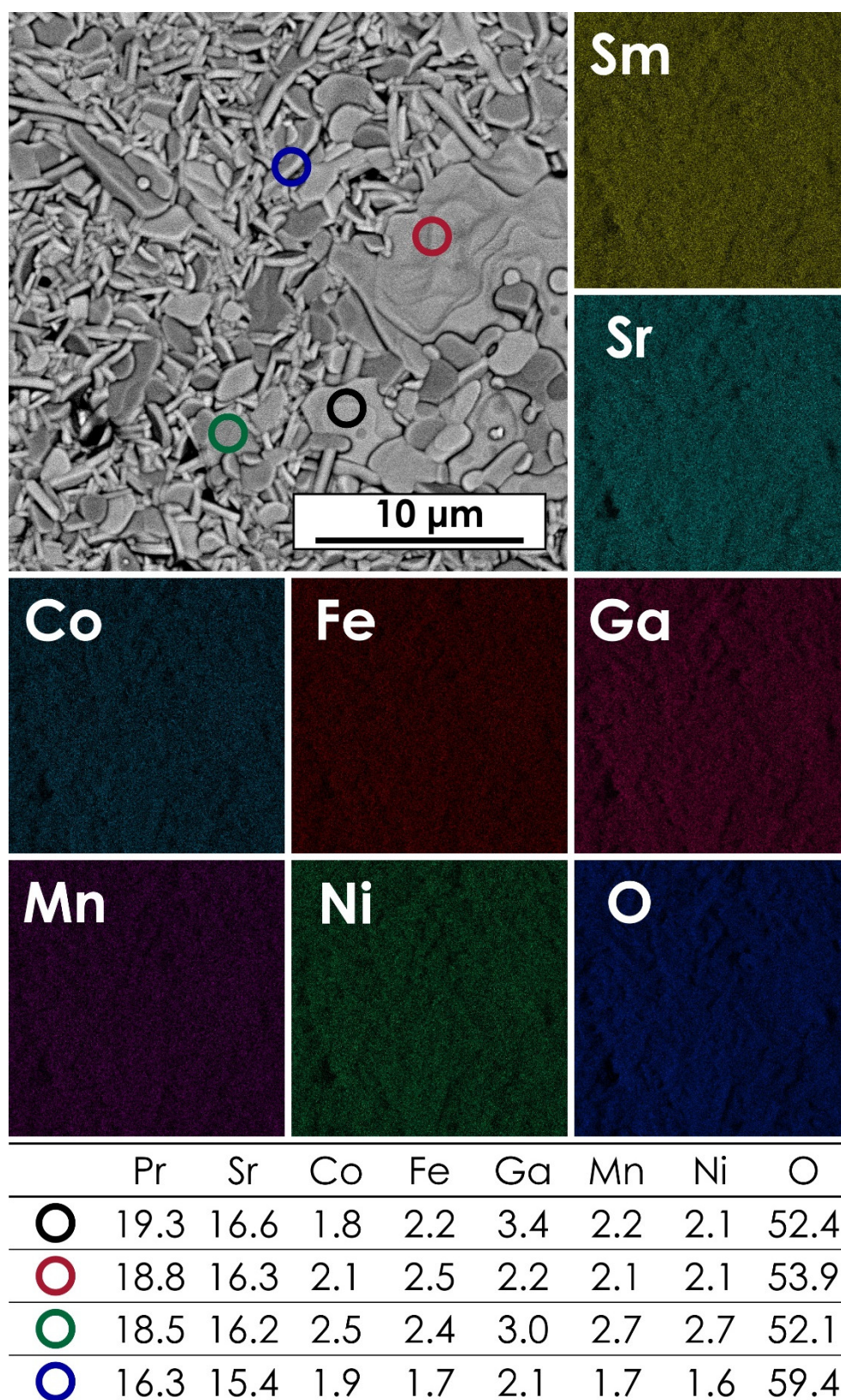


Figure S4. The results of EDS mapping and point analysis for the $\text{SmSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_4$ pellet sintered at 1200 °C for 20 h.

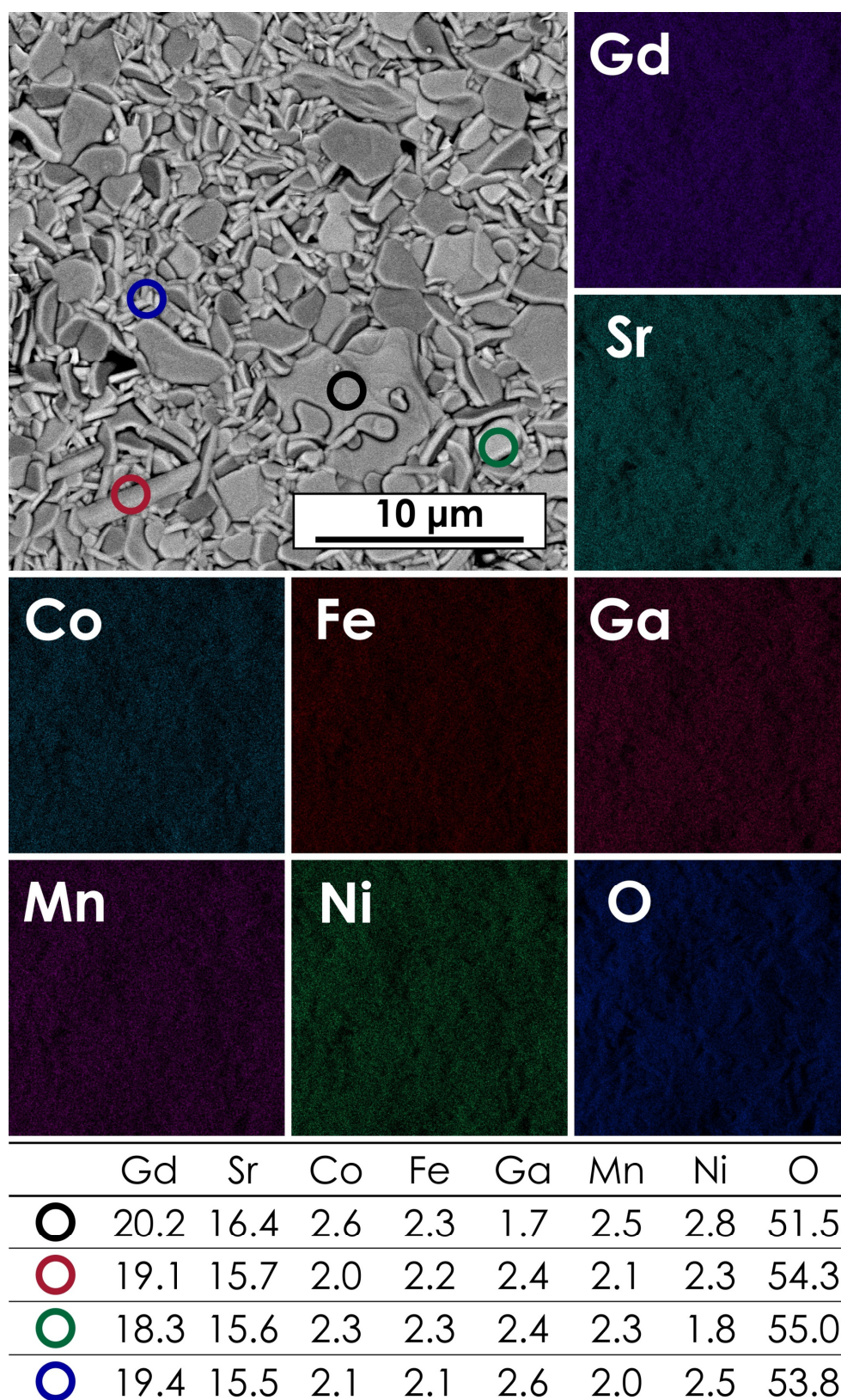


Figure S5. The results of EDS mapping and point analysis for the $\text{GdSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_4$ pellet sintered at 1200 °C for 20 h.

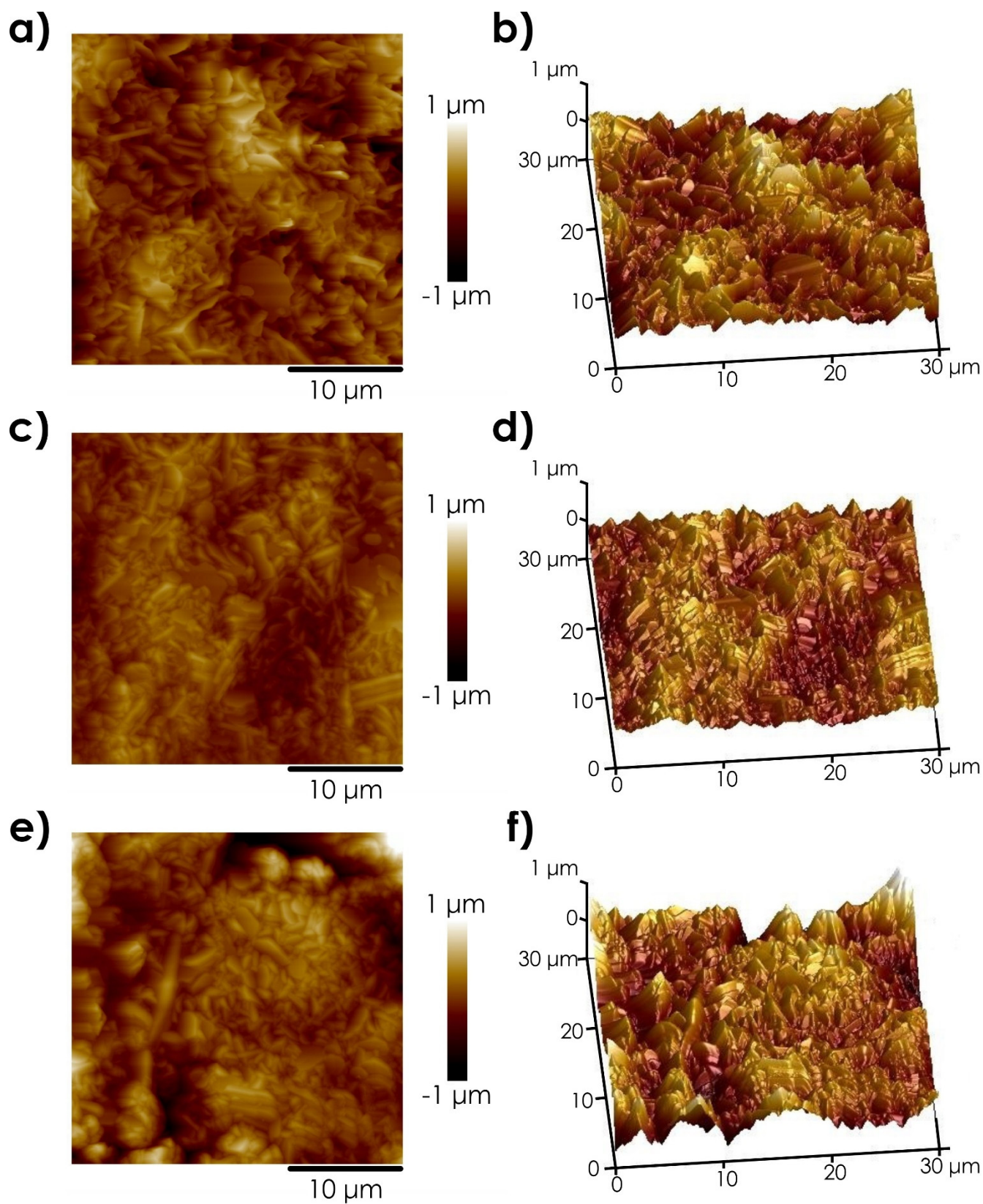


Figure S6. The results of the AFM mapping (tapping mode): (a) 2D micrograph of the PrSr(Co,Fe,Ga,Mn,Ni)O₄ pellet; (b) 3D micrograph of the PrSr(Co,Fe,Ga,Mn,Ni)O₄ pellet; (c) 2D micrograph of the NdSr(Co,Fe,Ga,Mn,Ni)O₄ pellet; (d) 3D micrograph of the NdSr(Co,Fe,Ga,Mn,Ni)O₄ pellet; (e) 2D micrograph of the SmSr(Co,Fe,Ga,Mn,Ni)O₄ pellet; (f) 3D micrograph of the SmSr(Co,Fe,Ga,Mn,Ni)O₄ pellet.

Table S1. The results of the iodometric titration measurements conducted for the $\text{LnSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_{4+\delta}$ (Ln = La, Pr, Nd, Sm, or Gd) series.

Composition	Powder mass[g]	Volume of titrant [ml]	Oxygen content	Oxygen non-stoichiometry	Average oxygen non-stoichiometry	Deviation
$\text{LaSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_{4+\delta}$	0.0508	12.017	4.006	0.006	0.006	0.004
	0.0513	12.017	4.002	0.002		
	0.0513	12.217	4.009	0.009		
$\text{PrSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_{4+\delta}$	0.0507	12.017	4.009	0.009	0.015	0.006
	0.0508	12.217	4.015	0.015		
	0.0509	12.417	4.021	0.021		
$\text{NdSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_{4+\delta}$	0.0507	11.817	4.006	0.006	0.008	0.005
	0.0506	12.017	4.014	0.014		
	0.0509	11.817	4.004	0.004		
$\text{SmSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_{4+\delta}$	0.0510	11.817	4.011	0.011	0.013	0.003
	0.0508	11.817	4.012	0.012		
	0.0502	11.817	4.017	0.017		
$\text{GdSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_{4+\delta}$	0.0501	11.617	4.019	0.019	0.013	0.006
	0.0511	11.617	4.011	0.011		
	0.0514	11.617	4.008	0.008		

Table S2. The results of the Rietveld analysis conducted for diffractograms from HT-XRD measurement of $\text{SmSr}(\text{Co,Fe,Ga,Mn,Ni})\text{O}_{4+\delta}$ (cooling run).

Temperature [°C]	Space group	$a = b$ [Å]	c [Å]	V_0 [Å ³]	a_0 [Å]	GoF	Rwp
1000	$I4/mmm$	3.8473	12.6261	186.887	5.7173	2.81	2.85
900	$I4/mmm$	3.8421	12.5973	185.960	5.7079	2.87	2.90
800	$I4/mmm$	3.8367	12.5670	184.991	5.6979	2.88	2.91
700	$I4/mmm$	3.8313	12.5365	184.017	5.6879	2.68	2.82
600	$I4/mmm$	3.8264	12.5066	183.114	5.6786	2.44	2.69
500	$I4/mmm$	3.8212	12.4775	182.195	5.6691	2.47	2.71
400	$I4/mmm$	3.8165	12.4486	181.321	5.6600	2.50	2.72
300	$I4/mmm$	3.8120	12.4212	180.493	5.6514	2.88	2.92
200	$I4/mmm$	3.8076	12.3940	179.683	5.6429	3.91	3.41
100	$I4/mmm$	3.8039	12.3710	179.001	5.6358	2.24	2.60
25	$I4/mmm$	3.8012	12.3539	178.503	5.6305	2.03	2.48

Table S3. The results of the Rietveld analysis conducted for diffractograms from HT-XRD measurement of NdSr(Co,Fe,Ga,Mn,Ni)O_{4+δ} (cooling run).

Temperature [°C]	Space group	$a = b$ [Å]	c [Å]	V_0 [Å ³]	a_0 [Å]	GoF	Rwp
1000	<i>I4/mmm</i>	3.8590	12.7046	189.194	5.7408	4.65	3.51
900	<i>I4/mmm</i>	3.8537	12.6757	188.250	5.7312	4.68	3.53
800	<i>I4/mmm</i>	3.8485	12.6460	187.297	5.7215	4.77	3.56
700	<i>I4/mmm</i>	3.8433	12.6161	186.357	5.7119	4.47	3.44
600	<i>I4/mmm</i>	3.8381	12.5843	185.383	5.7020	4.35	3.41
500	<i>I4/mmm</i>	3.8332	12.5542	184.465	5.6925	4.03	3.28
400	<i>I4/mmm</i>	3.8284	12.5239	183.561	5.6832	3.97	3.25
300	<i>I4/mmm</i>	3.8238	12.4949	182.696	5.6743	3.96	3.27
200	<i>I4/mmm</i>	3.8196	12.4675	181.890	5.6659	3.82	3.20
100	<i>I4/mmm</i>	3.8154	12.4413	181.109	5.6578	4.98	3.68
25	<i>I4/mmm</i>	3.8125	12.4245	180.594	5.6524	4.85	3.67

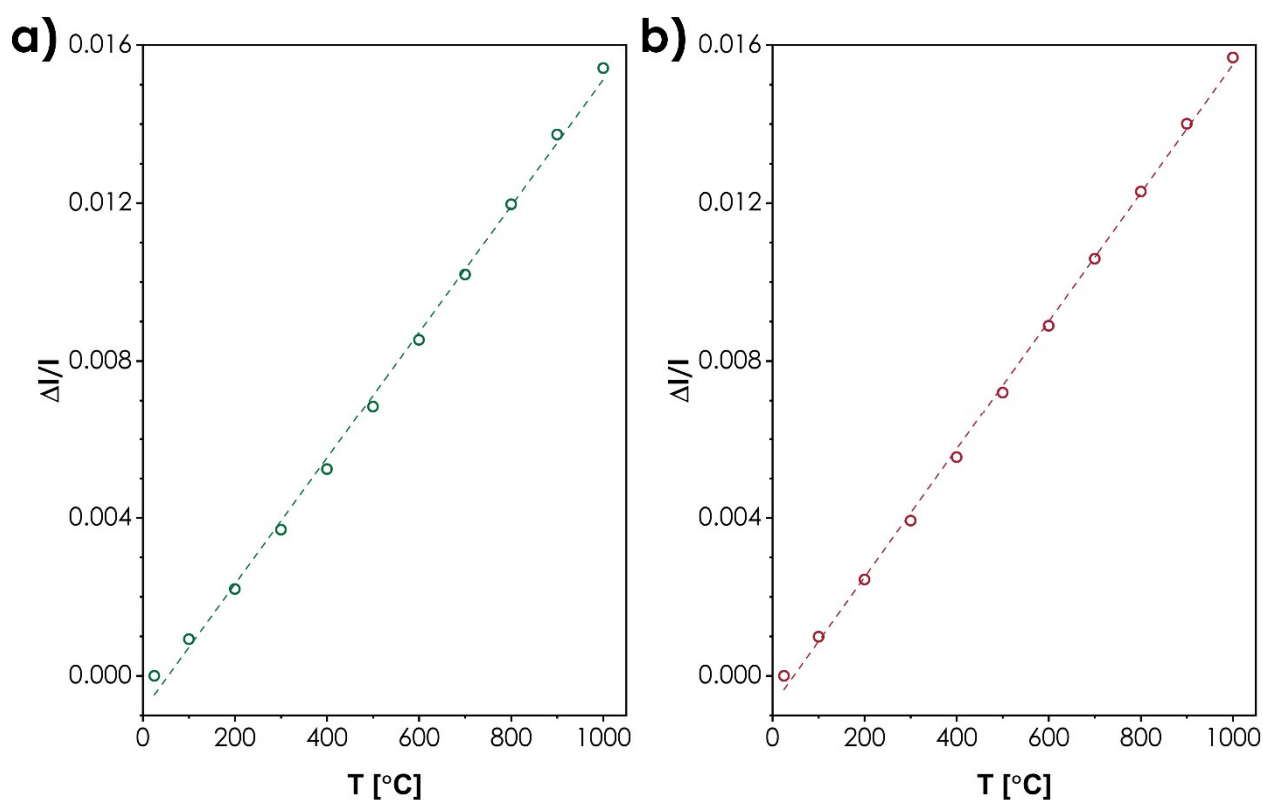


Figure S7. Thermal expansion behavior determined from the unit cell expansion during HT-XRD measurement for: (a) SmSr(Co,Fe,Ga,Mn,Ni)O_{4+δ}; (b) NdSr(Co,Fe,Ga,Mn,Ni)O_{4+δ}.