Supplementary Materials

Oxygen transport pathways in Ruddlesden-Popper structured oxides revealed via in-situ neutron diffraction

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Figure S1. Fourier observed nuclear density diagram at 1070 K with a pO_2 of .1 atm for LaSrCo_{0.5}Fe_{0.5}O_{4- δ} (RPn1) at an isosurface level of 0.15, 1.3% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.



Figure S2. Fourier observed nuclear density diagram at 1070 K with a pO₂ of .1 atm for La_{0.3}Sr_{2.7}CoFeO_{7- δ} (RPn2) at an isosurface level of 0.2, 2.1% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.



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Figure S3. Fourier observed nuclear density diagram at 1070 K with a pO₂ of .1 atm LaSr₃Co_{1.5}Fe_{1.5}O_{10- δ} (RPn3) at an isosurface level of 0.25, 2.4% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.



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Figure S4. Fourier observed nuclear density diagram at 1070 K with a pO_2 of .1 atm for LaSrCo_{0.5}Fe_{0.5}O_{4- δ} (RPn1) at an isosurface level of 0.10, 0.9% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.



Figure S5. Fourier observed nuclear density diagram at 1070 K with a pO_2 of .1 atm for $La_{0.3}Sr_{2.7}CoFeO_{7-\delta}$ (RPn2) at an isosurface level of 0.12, 1.3% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.



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Figure S6. Fourier observed nuclear density diagram at 1070 K with a pO₂ of .1 atm LaSr₃Co_{1.5}Fe_{1.5}O_{10- δ} (RPn3) at an isosurface level of 0.25, 1.4% of maximum. The progression of color from blue to green to red denotes an increase in the observed nuclear density, showing the probable positions for the atoms.



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Table S1: Tetragonal space group I4/mmm, Rietveld fit parameters for RPn1 structure at 765K, 865K,and 967 K with sites:

LaSrCo _{0.5} Fe _{0.5} O _{3.942} (RPn1) I4/mmm 765 K								
a,b (Å)	3.859 (1)		χ^2	4.922				
c (Å)	12.738(1)		R _P %	0.1182				
V (ų)	189.6	56(1)	wR _P %	0.0461				
$ ho_{c}$ (g/cm ³)	6.092	2						
	Atomic disp	lacement param	eters (Ų)*100		Fractional			
	U_{11}/U_{iso}	U ₂₂	U ₃₃	Z	Occupancy			
La/Sr1	1.40(2)			0.359(1)				
Co/Fe1	1.17(3)							
01	3.09(4)	3.09(4)	1.46(6)	0.166(1)	1.0			
02	0.64(5)	1.99(6)	2.79(8)		1.0			
LaSrCo _{0.5} Fe _{0.5} O _{3.942} (RPn1) I4/mmm 865 k			К					
a,b (Å)	3.863	3 (1)	χ^2	4.674				
c (Å)	12.78	39(1)	R _P %	0.1139)			
V (ų)	190.84(1)		wR _P %	0.0441				
$ ho_c$ (g/cm ³)	6.055	5						
	Atomic displacement parame		eters (Ų)*100		Fractional			
	U_{11}/U_{iso}	U ₂₂	U ₃₃	Z	Occupancy			
La/Sr1	1.60(2)			0.359(1)				
Co/Fe1	1.34(3)							
01		a (a)	4 (2)(7)	0 4 $(7/4)$	1.0			
01	3.49(5)	3.49(5)	1.63(7)	0.167(1)	1.0			
01	3.49(5) 0.86(5)	3.49(5) 2.32(7)	1.63(7) 2.87(9)	0.167(1)	1.0 1.0			

RPn1: La/Sr1: 4e (0,0,Z), Co/Fe1: 2a (0,0,0), O1: 4e (0,0,Z), O2: 4c (0.5,0,0)

LaSrCo _{0.5} Fe _{0.5} O _{3.942} (RPn1) I4/mmm 967 K								
a,b (Å)	3.868 (1)	χ^2	4.522				
c (Å)	12.840	(1)	<i>R_P</i> %	0.1055				
V (ų)	192.08	(1)	wR _P %	0.0423				
$ ho_c$ (g/cm ³)	5.999							
	Atomic displacement parame				Fractional			
	U_{11}/U_{iso}	U ₂₂	U ₃₃	Z	Occupancy			
La/Sr1	1.90(2)			0.359(1)				
Co/Fe1	1.53(3)							
01	3.52(7)	3.52(7)	1.8(1)	0.167(1)	0.971(8)			
02	1.0(1)	2.7(1)	3.3(1)		1.0			

Table S2: Tetragonal space group I4/mmm, Rietveld fit parameters for RPn2 structure at 765K, 865K,and 967 K with sites:

RPn2: La/Sr1: 4e (0,0,Z), La/Sr2: 2b (0,0,0.5), Co/Fe1: 4e (0,0,Z), O1: 4e (0,0,Z), O2: 8g (0,0.5,Z), O3: 2a (0,0,0)

La _{0.3} Sr _{2.7} CoreO ₆	_{5.378} (KPNZ)	14/mmm /05	ĸ				
a,b (Å)	3.88	8(1)	χ^2	5.333			
c (Å)	20.306(1)		R _P %	0.1677			
<i>V</i> (ų)	306.	97(1)	wR _P %	0.0678	3		
$ ho_{c}$ (g/cm ³)	5.38	2					
	Atomic dis	placement param	eters (Ų)*100		Fractional		
	U ₁₁ /U _{iso}	U ₂₂	U ₃₃	Z	Occupancy		
La/Sr1	2.22(4)			0.317(1)			
La/Sr2	1.93(6)						
Co/Fe1	1.33(4)			0.100(1)			
01	2.92(6)			0.195(1)	1.0		
02	2.43(9)	1.40(8)	3.6(1)	0.092(1)	0.956(6)		
03	3.1(2)				0.70(1)		
La _{0.3} Sr _{2.7} CoFeO _{6.378} (RPn2) I4/mmm 865 K							
La _{0.3} Sr _{2.7} CoFeO ₆	_{5.378} (RPn2)	I4/mmm 865	К				
La _{0.3} Sr _{2.7} CoFeO ₆ a,b (Å)	5.378 (RPn2) 3.90	I4/mmm 865 0(1)	$\frac{\mathbf{K}}{\chi^2}$	4.918			
La _{0.3} Sr _{2.7} CoFeO ₆ a,b (Å) c (Å)	5.378 (RPn2) 3.90 20.3	14/mmm 865 0(1) 56(1)		4.918 0.1532	 !		
La _{0.3} Sr _{2.7} CoFeO ₆ a,b (Å) c (Å) V (Å ³)	5.378 (RPn2) 3.90 20.3 309.	I4/mmm 865 0(1) 56(1) 59(1) 59(1)	Κ <i>χ</i> ² <i>R_P %</i> <i>wR_P %</i>	4.918 0.1532 0.0637			
La _{0.3} Sr _{2.7} CoFeO ₆ a,b (Å) c (Å) V (Å ³) ρ _c (g/cm ³)	5.378 (RPn2) 3.90 20.3 309. 5.31	I4/mmm 865 0(1) 56(1) 59(1) 5		4.918 0.1532 0.0637	<u>.</u>		
La _{0.3} Sr _{2.7} CoFeO ₆ a,b (Å) c (Å) V (Å ³) ρ _c (g/cm ³)	5.378 (RPn2) 3.90 20.3 309. 5.31 Atomic dis	I4/mmm 865 0(1) 56(1) 59(1) 5 placement param	χ^2 χ^2 $R_P \%$ w $R_P \%$ veters (Å ²)*100	4.918 0.1532 0.0637	Fractional		
La _{0.3} Sr _{2.7} CoFeO ₆ a,b (Å) c (Å) V (Å ³) ρ _c (g/cm ³)	5.378 (RPn2) 3.90 20.3 309. 5.31 Atomic dis U ₁₁ /U _{iso}	I4/mmm 865 0(1) 56(1) 59(1) 5 placement param U ₂₂	K χ^2 $R_P \%$ $wR_P \%$ weters (Å ²)*100 U ₃₃	4.918 0.1532 0.0637 Z	Fractional Occupancy		
La _{0.3} Sr _{2.7} CoFeO _e a,b (Å) c (Å) V (Å ³) ρ _c (g/cm ³) La/Sr1	5.378 (RPn2) 3.90 20.3 309. 5.31 Atomic dis U ₁₁ /U _{iso} 2.60(5)	14/mmm 865 0(1) 56(1) 59(1) 5 placement param U ₂₂	χ^2 χ^2 $R_P \%$ w $R_P \%$ veters (Å ²)*100U ₃₃	4.918 0.1532 0.0637 Z 0.317(1)	Fractional Occupancy		
La _{0.3} Sr _{2.7} CoFeO _e a,b (Å) c (Å) V (Å ³) ρ _c (g/cm ³) La/Sr1 La/Sr2	5.378 (RPn2) 3.90 20.3 309. 5.31 Atomic dis U ₁₁ /U _{iso} 2.60(5) 2.25(7)	I4/mmm 865 0(1) 56(1) 59(1) 5 placement param U22	K χ^2 $R_P \%$ $wR_P \%$ heters $(Å^2)*100$ U_{33}	4.918 0.1532 0.0637 Z 0.317(1)	Fractional Occupancy		
La _{0.3} Sr _{2.7} CoFeO _e a,b (Å) c (Å) V (Å ³) ρ _c (g/cm ³) La/Sr1 La/Sr2 Co/Fe1	5.378 (RPn2) 3.90 20.3 309. 5.31 Atomic dis U ₁₁ /U _{iso} 2.60(5) 2.25(7) 1.61(4)	14/mmm 865 0(1) 56(1) 59(1) 5 placement param U ₂₂	χ^2 $R_P \%$ w $R_P \%$ veters (Ų)*100U_{33}	4.918 0.1532 0.0637 Z 0.317(1) 0.100(1)	Fractional Occupancy		
La _{0.3} Sr _{2.7} CoFeO _e a,b (Å) c (Å) V (Å ³) ρ _c (g/cm ³) La/Sr1 La/Sr2 Co/Fe1 O1	5.378 (RPn2) 3.90 20.3 309. 5.31 Atomic disp U ₁₁ /U _{iso} 2.60(5) 2.25(7) 1.61(4) 3.32(6)	14/mmm 865 0(1) 56(1) 59(1) 5 placement param U ₂₂	χ^2 $R_P \%$ w $R_P \%$ veters (Ų)*100U_{33}	4.918 0.1532 0.0637 Z 0.317(1) 0.100(1) 0.195(2)	Fractional Occupancy 1.0		
La _{0.3} Sr _{2.7} CoFeO _e a,b (Å) c (Å) V (Å ³) ρ _c (g/cm ³) La/Sr1 La/Sr2 Co/Fe1 O1 O2	5.378 (RPn2) 3.90 20.3 309. 5.31 Atomic dis U ₁₁ /U _{iso} 2.60(5) 2.25(7) 1.61(4) 3.32(6) 2.9(1)	14/mmm 865 0(1) 56(1) 59(1) 5 placement param U22 1.53(9) 1.53(9)	K χ^2 $R_P \%$ w $R_P \%$ eters (Å ²)*100 U ₃₃ 3.9(1)	4.918 0.1532 0.0637 Z 0.317(1) 0.100(1) 0.195(2) 0.091(1)	Fractional Occupancy 1.0 0.941(6)		

La _{0.3} Sr _{2.7} CoFeO _{6.378} (RPn2) I4/mmm 967 K							
a,b (Å)	3.912(1)	χ^2	4.529			
c (Å)	20.409(1)	R _P %	0.1459			
<i>V</i> (ų)	312.28(1)	wR _P %	0.0587			
$ ho_c$ (g/cm ³)	5.265						
Atomic displacement parame			neters (Ų)*100		Fractional		
	U_{11}/U_{iso}	U ₂₂	U ₃₃	Z	Occupancy		
La/Sr1	2.87(6)			0.317(1)			
La/Sr2	2.52(7)						
Co/Fe1	1.76(5)			0.101(1)			
01	3.65(7)			0.196(2)	1.0		
02	3.4(1)	1.8(1)	4.4(2)	0.091(1)	0.942(6)		
03	4.7(3)				0.61(2)		

Table S3: Tetragonal space group I4/mmm, Rietveld fit parameters RPn3 structure at 765K, 865K, and967 K with sites:

RPn3: La/Sr1: 4e(0,0,Z), La/Sr2: 4e (0,0,Z), Co/Fe1: 4e (0,0,Z), Co/Fe2: 2a (0,0,0), O1: 4e (0,0,Z), O2: 8g (0,0.5,Z), O3: 4e (0,0,Z), O4: 4c (0,0.5,0)

LaSr ₃ Co _{1.5} Fe _{1.5} O _{9.582} (RPn3) I4/mmm 765 K							
a,b (Å)	3.879((1)	χ^2	3.064			
c (Å)	28.235(1)		R _P %	0.1407			
V (ų)	424.90(1)		wR _P %	0.0546			
$ ho_c$ (g/cm ³)	5.737						
	Atomic displ	acement param	eters (Ų)*100		Fractional		
	U_{11}/U_{iso}	U ₂₂	U ₃₃	Z	Occupancy		
La/Sr1	1.9(1)			0.2013(1)			
La/Sr2	2.0(1)			0.0696(2)			
Co/Fe1	1.1(1)			0.1398(1)			
Co/Fe2	1.5(1)						
01	2.7(2)			0.2106(2)	1.0		
02	2.0(2)			0.1378(1)	1.0		
03	3.5(3)			0.0675(3)	1.0		
O4	5.3(4)	1.4(2)	7.3(5)		1.0		
LaSr ₃ Co _{1 5} Fe _{1 5} O _{9 582} (RPn3) 14/mmm 865 K							

a,b (Å)	3.889(1)		χ^2	2.834	
c (Å)	28.366(1	28.366(1)		0.1260	
<i>V</i> (ų)	429.09(2	429.09(2)		0.0505	
$ ho_{c}$ (g/cm ³)	5.65.3				
	Atomic displace	ement param	eters (Ų)*100		Fractional
	U_{11}/U_{iso}	U ₂₂	U ₃₃	Z	Occupancy
La/Sr1	2.1(1)			0.2013(1)	
La/Sr2	2.4(1)			0.0704(2)	
Co/Fe1	1.4(1)			0.1404(1)	
Co/Fe2	1.9(1)				
01	3.4(1)			0.2113(2)	1.0
02	2.2(1)			0.1380(2)	0.98(1)
03	4.1(3)			0.0664(3)	0.99(2)
O4	7.9(7)	1.6(3)	8.8(7)		0.94(2)

LaSr ₃ Co _{1.5} Fe _{1.5} O _{9.582} (RPn3) I4/mmm 967 K							
a,b (Å)	3.900(1)		χ^2	2.786			
c (Å)	28.523(1)	R _P %	0.1577			
V (Å ³)	433.78(2)	wR _P %	0.0587			
$ ho_c$ (g/cm ³)	5.568						
	Atomic displace	ement param	neters (Å ²)*100		Fractional		
	U_{11}/U_{iso}	U ₂₂	U ₃₃	Z	Occupancy		
La/Sr1	2.5(1)			0.2013(1)			
La/Sr2	2.8(1)			0.0710(2)			
Co/Fe1	1.5(1)			0.1409(1)			
Co/Fe2	2.7(1)						
01	3.8(2)			0.2114(2)	1.0		
02	2.5(1)			0.1380(2)	0.98(1)		
O3	4.7(3)			0.0654(3)	0.95(2)		
04	12(1)	2.2(4)	11.4(9)		0.89(4)		