Supporting Information

Effect of Nickel Oxidation State on the Structural and Electrochemical Characteristics of Lithium-rich Layered Oxide Cathodes

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Rietveld refinement was carried out on the ten samples synthesized for this study: Figures S1-S10 included below show the observed XRD patterns and the calculated Reitveld refinement patterns for each sample. The Rp values for each refinement are also included. Table S1 then lists the refined parameters for each sample as well, including the lattice parameters, atomic locations, atomic occupancies, and atomic U values. Mn, Ni, and Co ions were not differentiated in the refinement due to their similar atomic numbers. They are labeled as TM (transition metal) in Table S1. The labels in the parentheses refer to a specific layer from the layered structure. For example, Li (M) refers to the Li atoms in the transition-metal layer. The Li/Ni site mixing value listed in Table 1 in the main article can be found in Table S1, where it is depicted as the TM (Li) occupancy value.

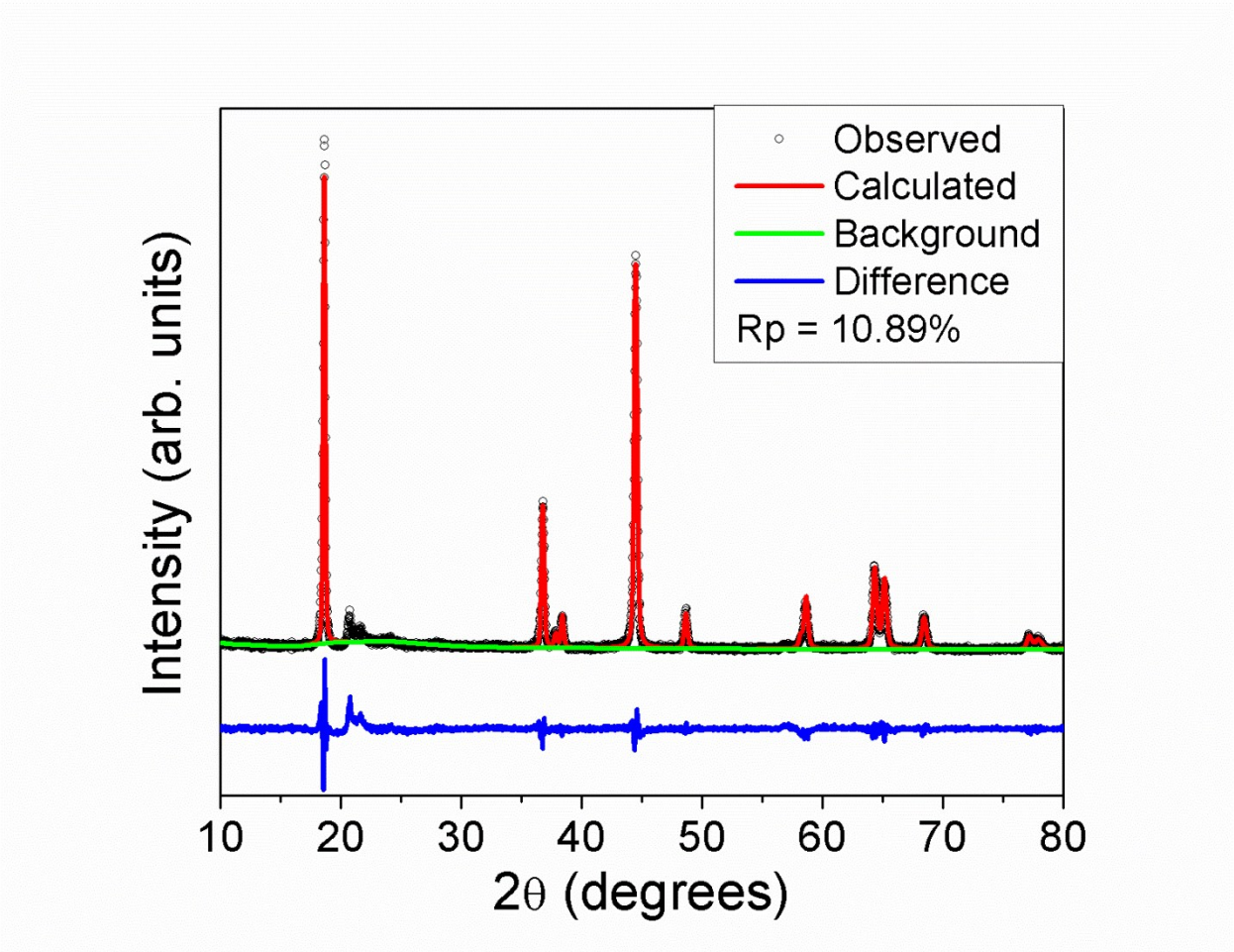

Figure S1: Observed XRD pattern and the calculated Rietveld refinement XRD pattern of the undoped series with $x = 0.00$.

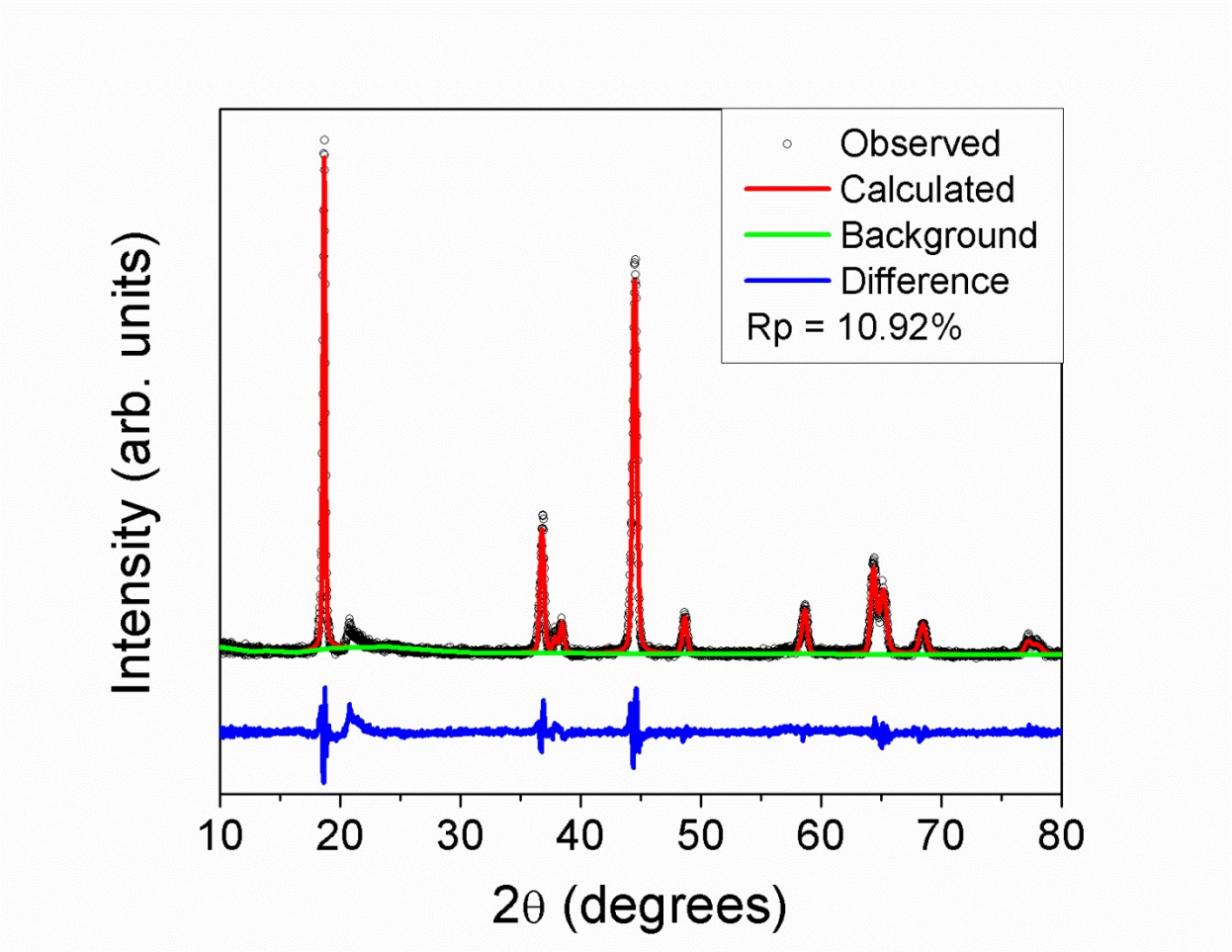

Figure S2: Observed XRD pattern and the calculated Rietveld refinement XRD pattern of the undoped series with $x = 0.02$.

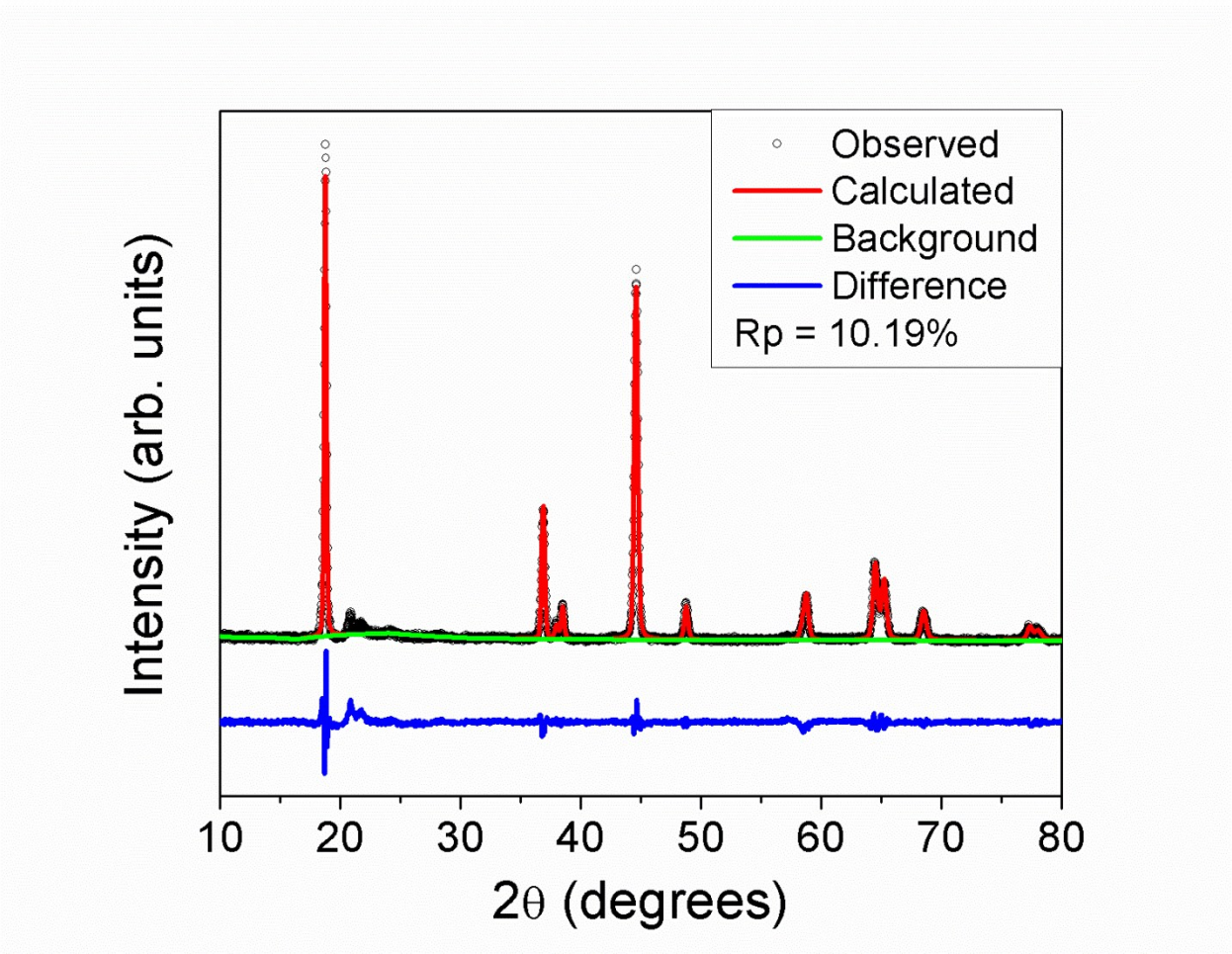

Figure S3: Observed XRD pattern and the calculated Rietveld refinement XRD pattern of the undoped series with $x = 0.05$.

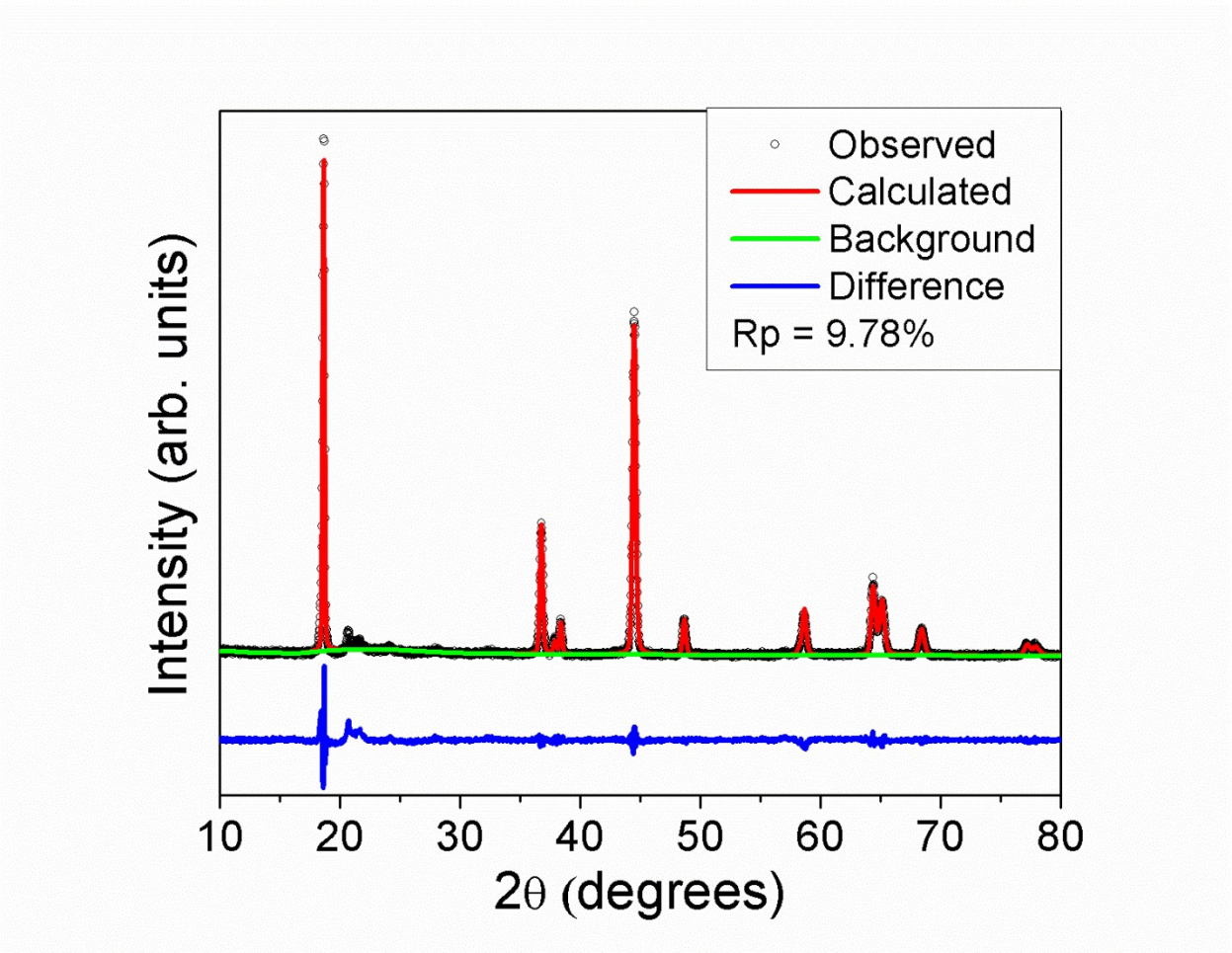

Figure S4: Observed XRD pattern and the calculated Rietveld refinement XRD pattern of the undoped series with $x = 0.10$.

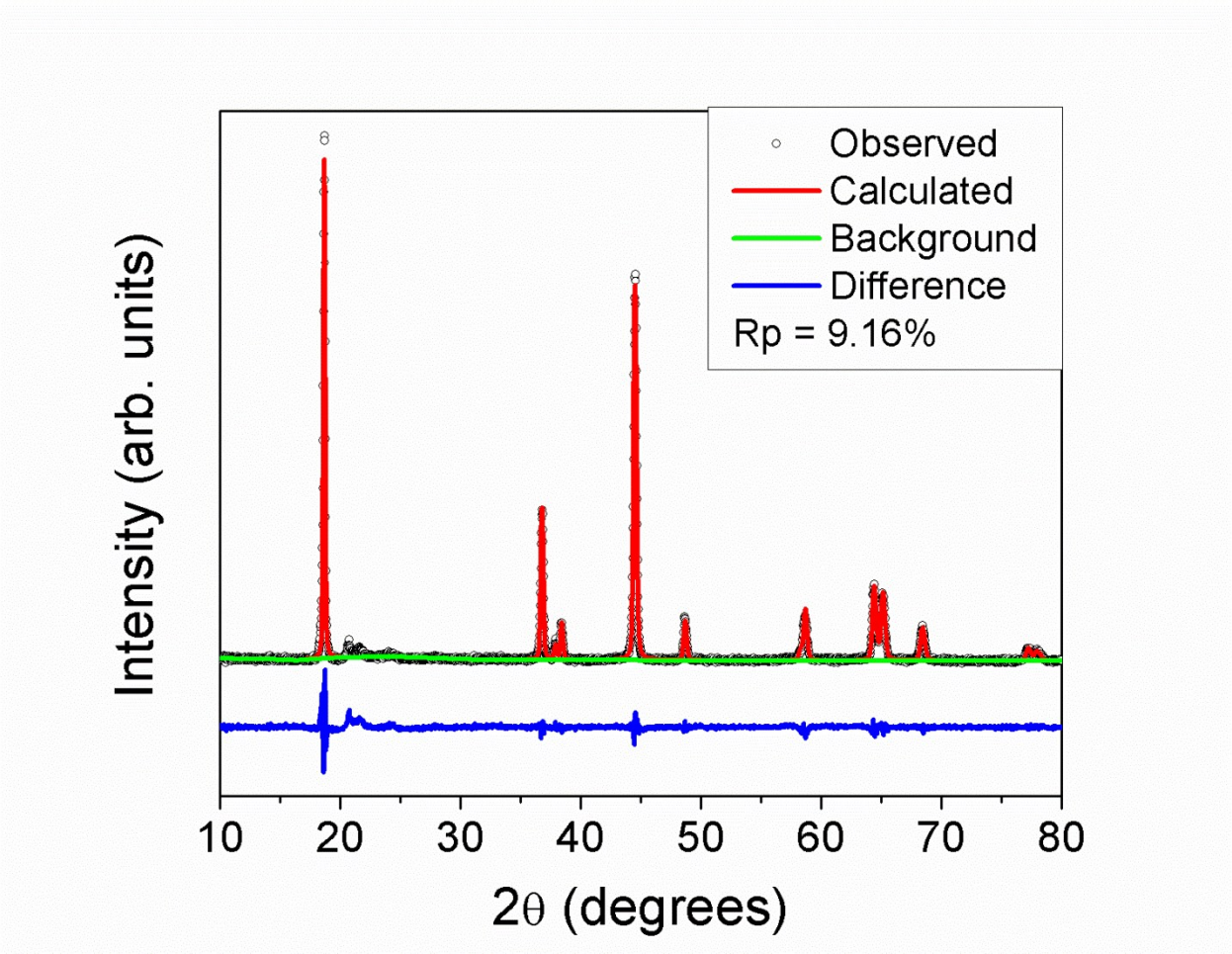

Figure S5: Observed XRD pattern and the calculated Rietveld refinement XRD pattern of the undoped series with $x = 0.15$.

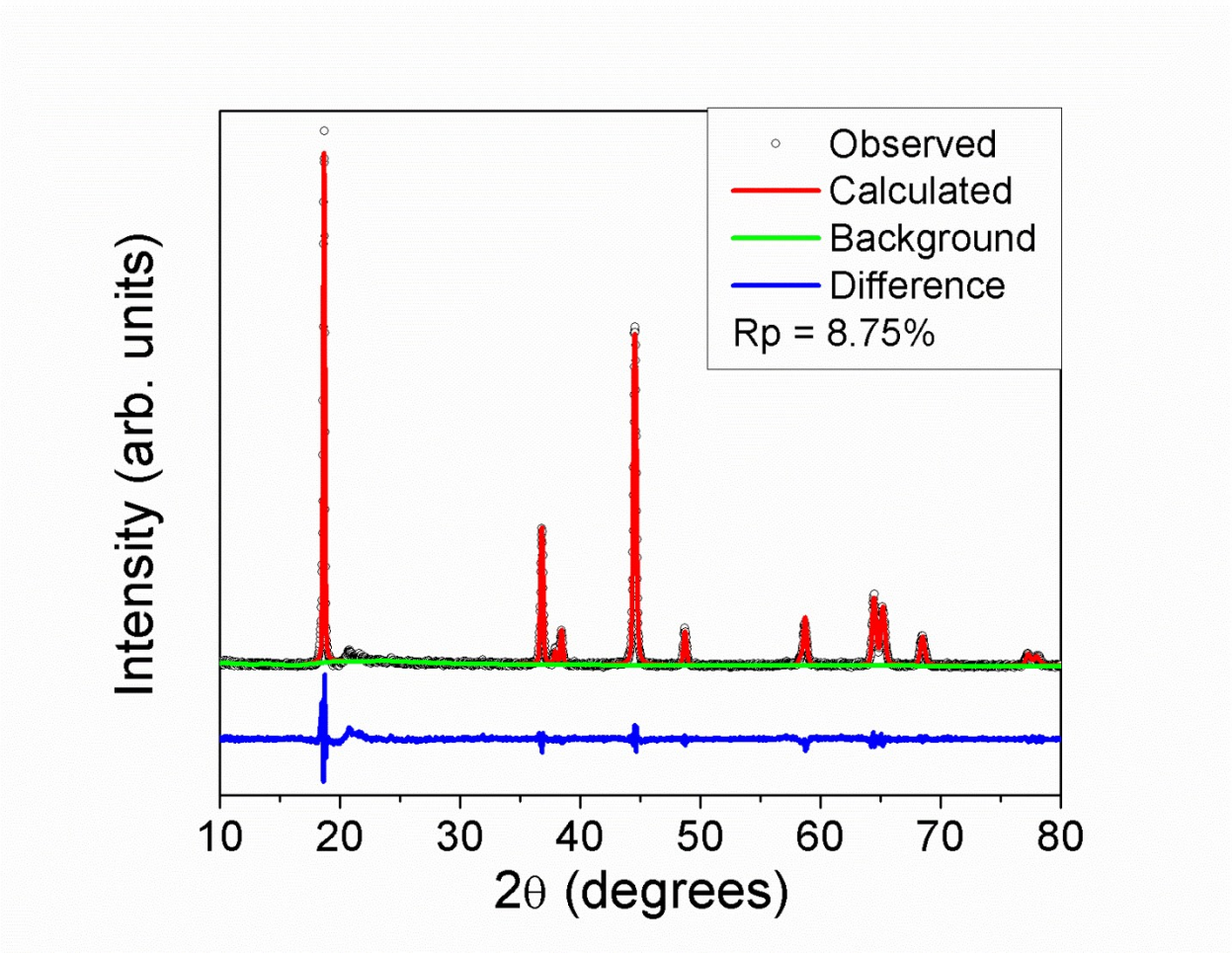

Figure S6: Observed XRD pattern and the calculated Rietveld refinement XRD pattern of the undoped series with $x = 0.20$.

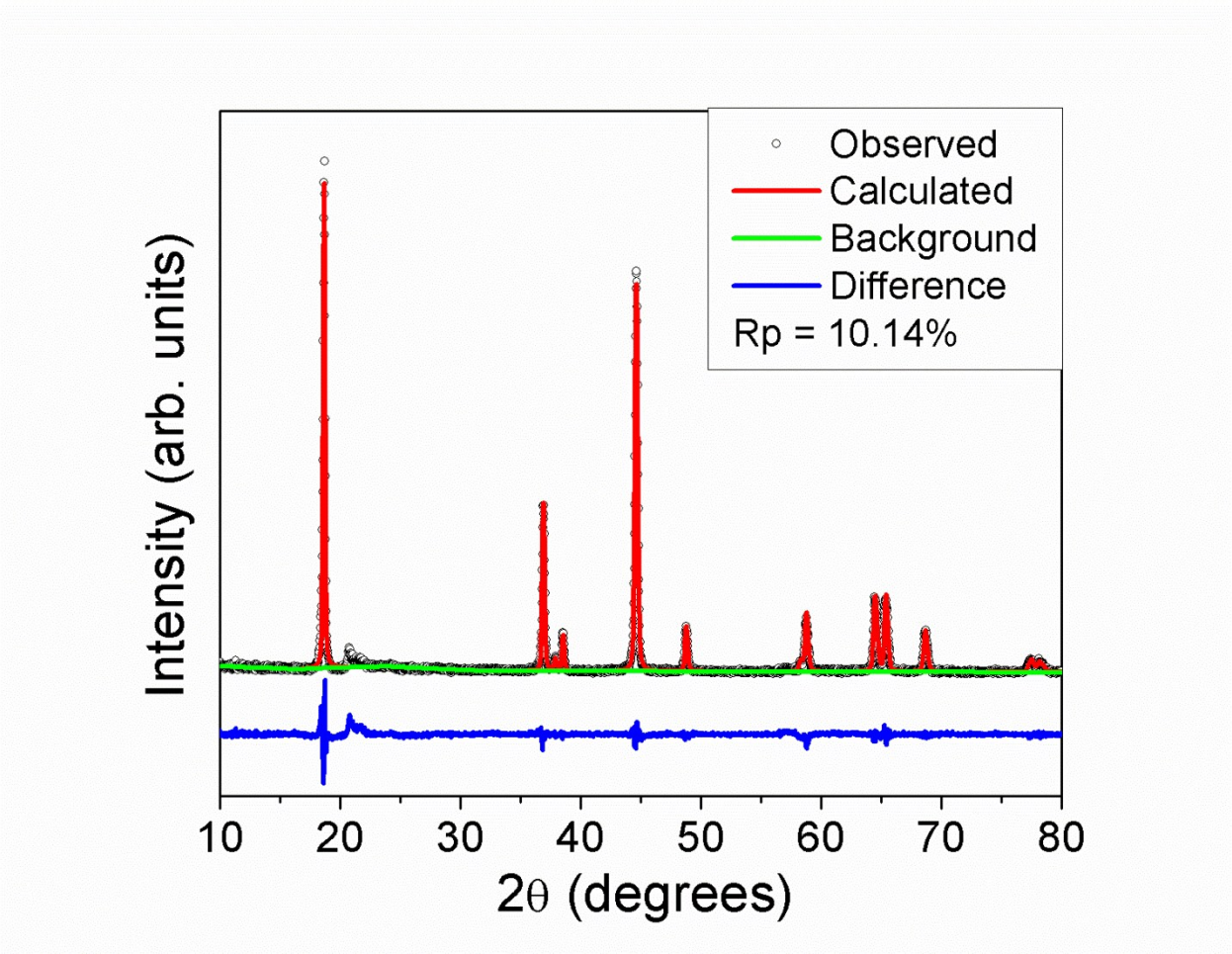

Figure S7: Observed XRD pattern and the calculated Rietveld refinement XRD pattern of the Co series with x = 0.00.

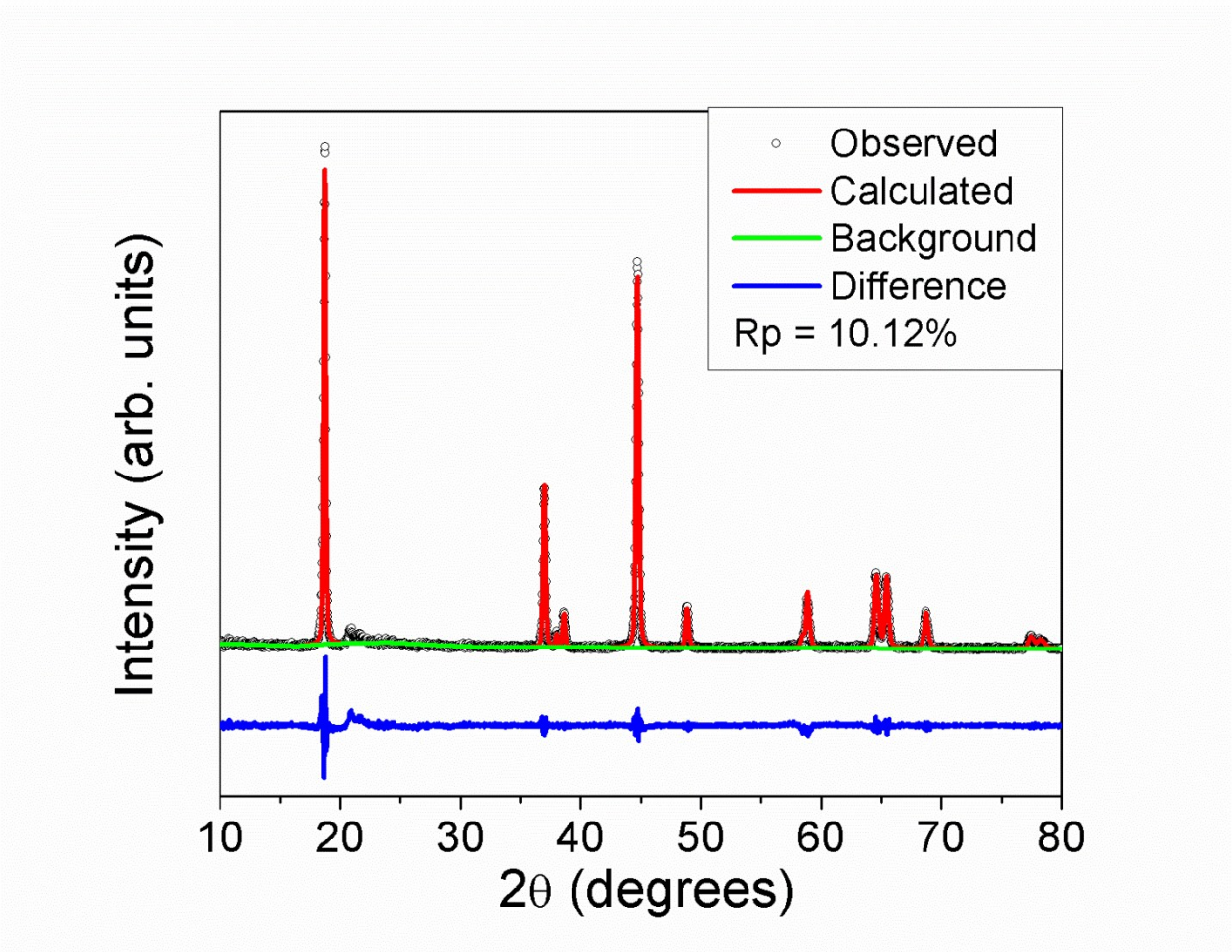

Figure S8: Observed XRD pattern and the calculated Rietveld refinement XRD pattern of the Co series with $x = 0.04$.

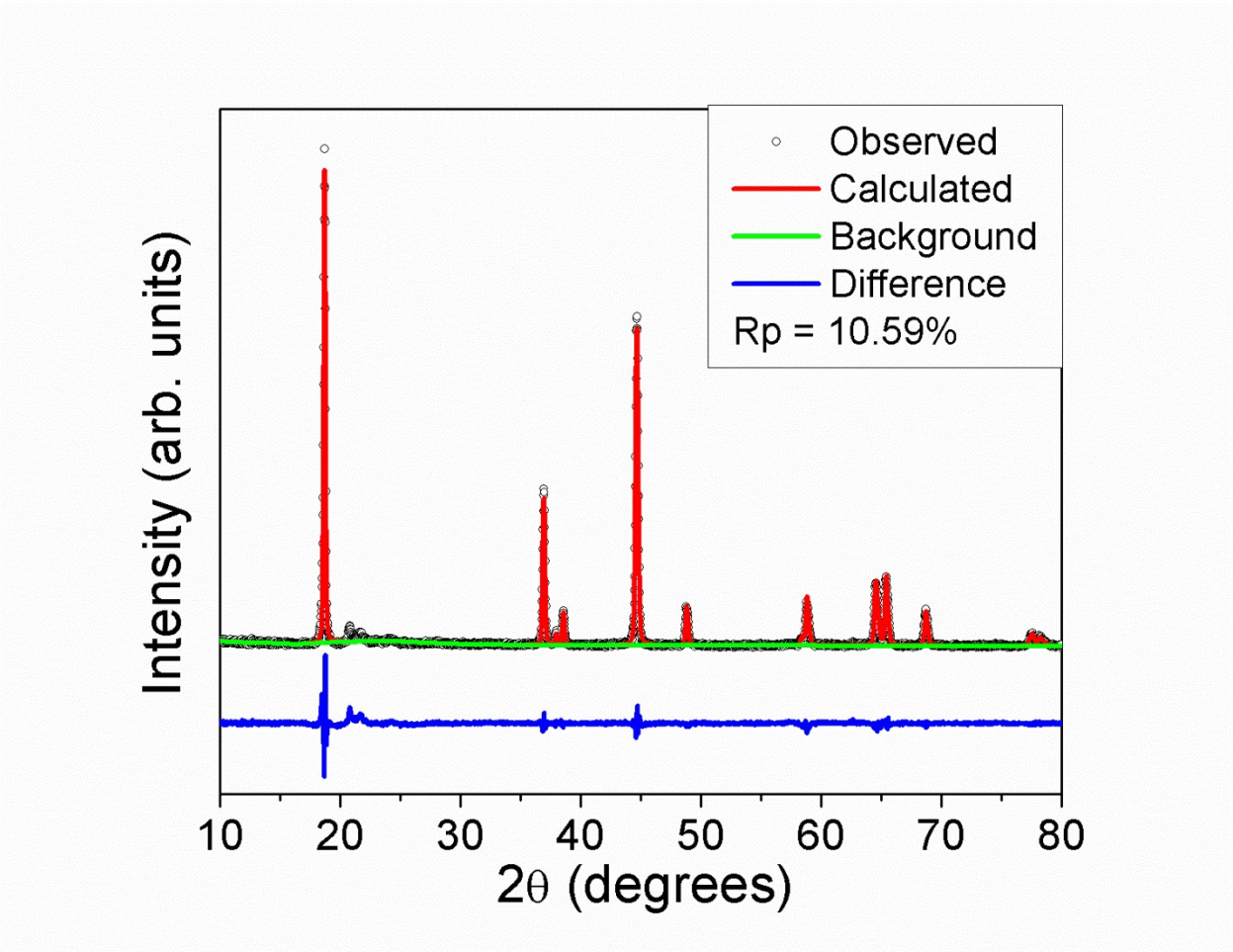

Figure S9: Observed XRD pattern and the calculated Rietveld refinement XRD pattern of the Co series with x = 0.09.

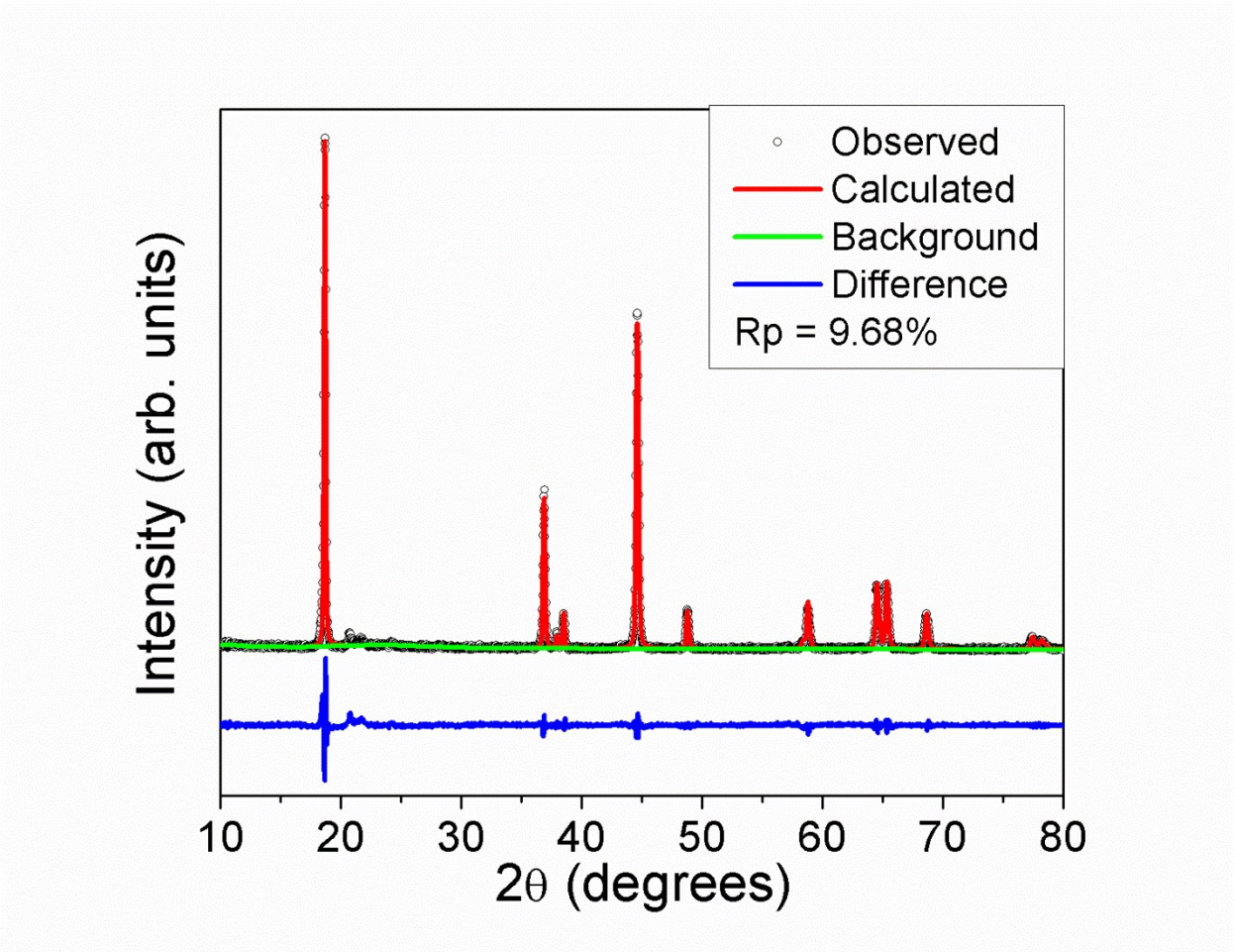

Figure S10: Observed XRD pattern and the calculated Rietveld refinement XRD pattern of the Co series with $x = 0.14$.

Table S1: Rietveld refinement parameters for the undoped and Co series of materials

Undoped x = 0.00			R_p: 10.89%				
Atom	*x/a*	*y/b*	*z/c*	Occupancy	U_{iso}(Å^2)	Lattice Parameters (Å)	
O	0	0	0.2421(2)	1	0.02414(2)	a	2.85987(8)
TM (M)	0	0	0.5	0.7548(2)	0.02091(6)	b	2.85987(8)
Li (M)	0	0	0.5	0.2452(2)	0.02091(6)	c	14.2461(6)
Li (Li)	0	0	0	0.9278(2)	0.07676(4)		
TM (Li)	0	0	0	0.0722(2)	0.07676(4)		

Undoped x = 0.02			R_p: 11.95%				
Atom	*x/a*	*y/b*	*z/c*	Occupancy	U_{iso}(Å^2)	Lattice Parameters (Å)	
O	0	0	0.2433(2)	1	0.02345(2)	a	2.8616(1)
TM (M)	0	0	0.5	0.7369(2)	0.01666(6)	b	2.8616(1)
Li (M)	0	0	0.5	0.2631(2)	0.01666(6)	c	14.2554(7)
Li (Li)	0	0	0	0.9369(2)	0.05827(4)		
TM (Li)	0	0	0	0.0631(2)	0.05827(4)		

Undoped x = 0.05			R_p: 10.19%				
Atom	*x/a*	*y/b*	*z/c*	Occupancy	U_{iso}(Å^2)	Lattice Parameters (Å)	
O	0	0	0.2417(3)	1	0.02551(3)	a	2.8600(1)
TM (M)	0	0	0.5	0.7373(5)	0.01709(9)	b	2.8600(1)
Li (M)	0	0	0.5	0.2627(5)	0.01709(9)	c	14.2195(9)
Li (Li)	0	0	0	0.9373(5)	0.06739(7)		
TM (Li)	0	0	0	0.0627(5)	0.06739(7)		

Undoped x = 0.10			R_p: 9.78 %				
Atom	*x/a*	*y/b*	*z/c*	Occupancy	U_{iso}(Å^2)	Lattice Parameters (Å)	
O	0	0	0.2426(2)	1	0.02822(2)	a	2.8614(1)
TM (M)	0	0	0.5	0.7440(5)	0.02104(9)	b	2.8614(1)
Li (M)	0	0	0.5	0.2560(5)	0.02104(9)	c	14.2386(11)
Li (Li)	0	0	0	0.9440(5)	0.07159(8)		
TM (Li)	0	0	0	0.0560(5)	0.07159(8)		

Undoped x = 0.15			R_p: 9.16%				
Atom	*x/a*	*y/b*	*z/c*	Occupancy	U_{iso}(Å^2)	Lattice Parameters (Å)	
O	0	0	0.2431(2)	1	0.03284(2)	a	2.8615(9)
TM (M)	0	0	0.5	0.7410(5)	0.02049(8)	b	2.8615(9)
Li (M)	0	0	0.5	0.2590(5)	0.02049(8)	c	14.2318(10)

Li (Li)	0	0	0	0.9410(5)	0.07856(7)		
TM (Li)	0	0	0	0.0590(5)	0.07856(7)		

Undoped x = 0.20			R_p: 8.75%				
Atom	*x/a*	*y/b*	*z/c*	Occupancy	U_{iso}(Å^2)	Lattice Parameters (Å)	
O	0	0	0.2422(2)	1	0.03080(2)	a	2.8591(8)
TM (M)	0	0	0.5	0.7552(4)	0.02207(9)	b	2.8591(8)
Li (M)	0	0	0.5	0.2448(4)	0.02207(9)	c	14.2215(9)
Li (Li)	0	0	0	0.9552(4)	0.06603(7)		
TM (Li)	0	0	0	0.0448(4)	0.06603(7)		

Co doped x = 0.00			R_p: 10.14%				
Atom	*x/a*	*y/b*	*z/c*	Occupancy	U_{iso}(Å^2)	Lattice Parameters (Å)	
O	0	0	0.2422(2)	1	0.02282(2)	a	2.8507(9)
TM (M)	0	0	0.5	0.7516(5)	0.01498(9)	b	2.8507(9)
Li (M)	0	0	0.5	0.2484(5)	0.01498(9)	c	14.2250(10)
Li (Li)	0	0	0	0.9516(5)	0.08784(9)		
TM (Li)	0	0	0	0.0484(5)	0.08784(9)		

Co doped x = 0.04			R_p: 10.12%				
Atom	*x/a*	*y/b*	*z/c*	Occupancy	U_{iso}(Å^2)	Lattice Parameters (Å)	
O	0	0	0.2419(2)	1	0.02796(3)	a	2.8511(1)
TM (M)	0	0	0.5	0.7722(5)	0.02168(9)	b	2.8511(1)
Li (M)	0	0	0.5	0.2278(5)	0.02168(9)	c	14.2133(10)
Li (Li)	0	0	0	0.9722(5)	0.05845(8)		
TM (Li)	0	0	0	0.0278(5)	0.05845(8)		

Co doped x = 0.09			R_p: 10.59%				
Atom	*x/a*	*y/b*	*z/c*	Occupancy	U_{iso}(Å^2)	Lattice Parameters (Å)	
O	0	0	0.2418(2)	1	0.02807(3)	a	2.8502(8)
TM (M)	0	0	0.5	0.7902(5)	0.02133(9)	b	2.8502(8)
Li (M)	0	0	0.5	0.2098(5)	0.02133(9)	c	14.2113(9)
Li (Li)	0	0	0	0.9902(5)	0.03967(9)		
TM (Li)	0	0	0	0.0098(5)	0.03967(9)		

Co doped x = 0.14			R_p: 9.68%				
Atom	*x/a*	*y/b*	*z/c*	Occupancy	U_{iso}(Å^2)	Lattice Parameters (Å)	
O	0	0	0.2416(2)	1	0.03167(3)	a	2.8523(7)
TM (M)	0	0	0.5	0.7895(5)	0.02288(8)	b	2.8523(7)

Li (M)	0	0	0.5	0.2105(5)	0.02288(8)	c	14.2136(8)
Li (Li)	0	0	0	0.9895(5)	0.04319(8)		
TM (Li)	0	0	0	0.0105(5)	0.04319(8)		