Supporting Information

Mitigating Jahn-Teller Distortion and Phase Transition in P2-$Na_{0.67}Ni_{0.33}Mn_{0.67}O_2$ Cathode through Large Sr^{2+} ion Substitution for Improved Performance

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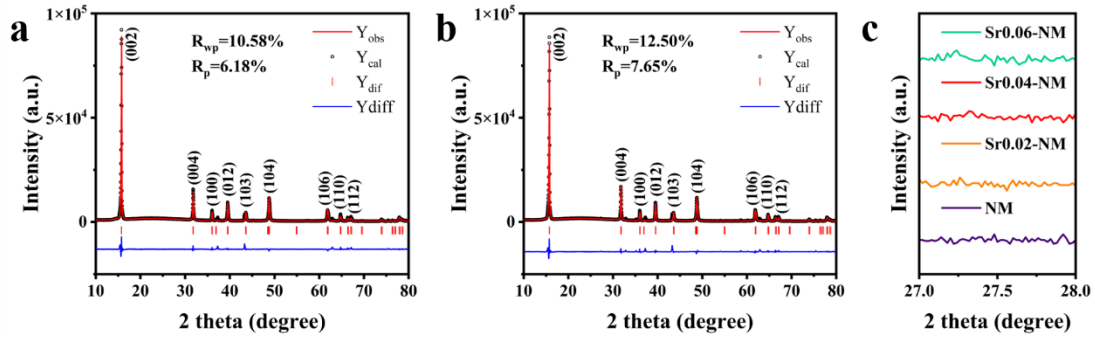

Figure S1. Rietveld refinement of the XRD patterns for (a) Sr0.02-NM and (b) Sr0.06-NM. (c) XRD pattern for the samples at the range of 27~28 degrees.

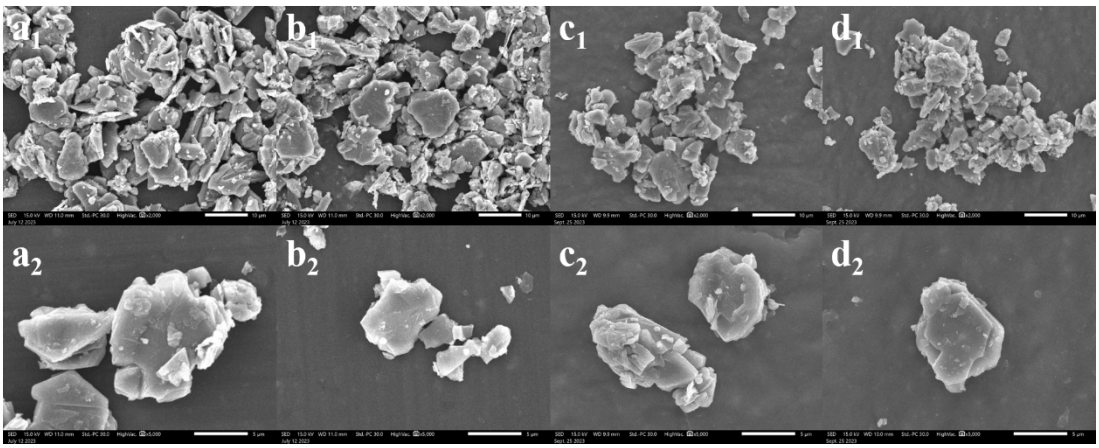

Figure S2. SEM images of (a) NM, (b) Sr0.02-NM, (c) Sr0.04-NM, and (d) Sr0.06-NM.

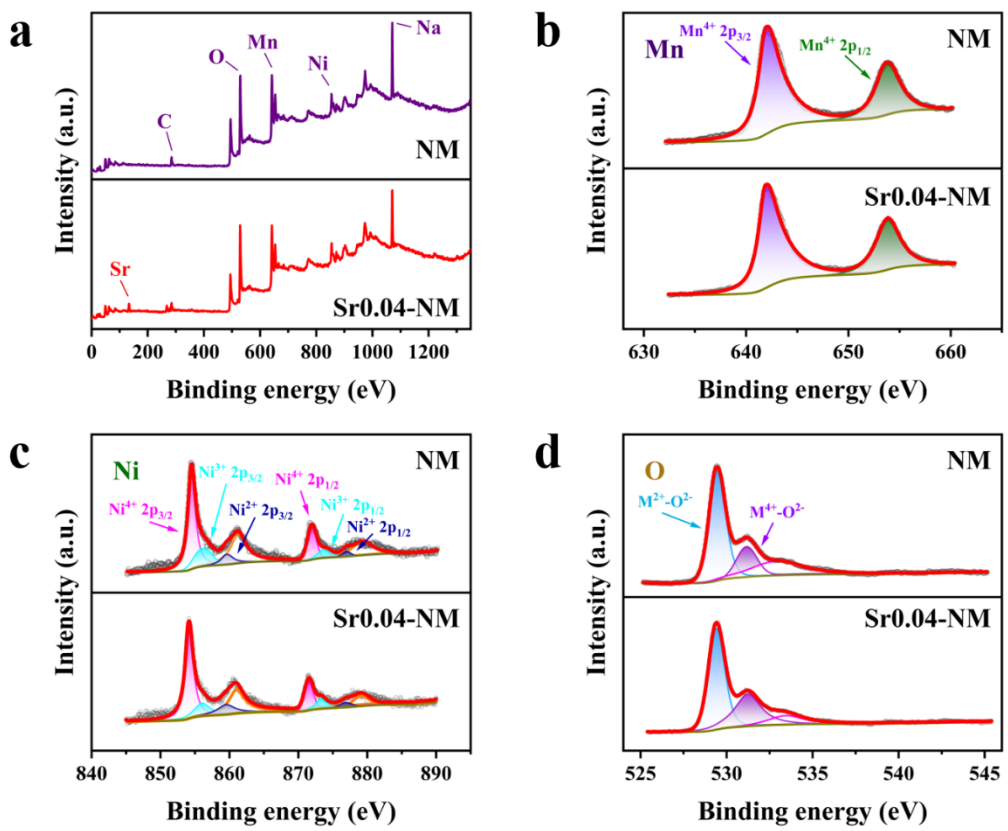

Figure S3. XPS results of powder samples. (a) survey spectrum. Fitting results of (b) manganese, (c) nickel, and (d) oxygen.

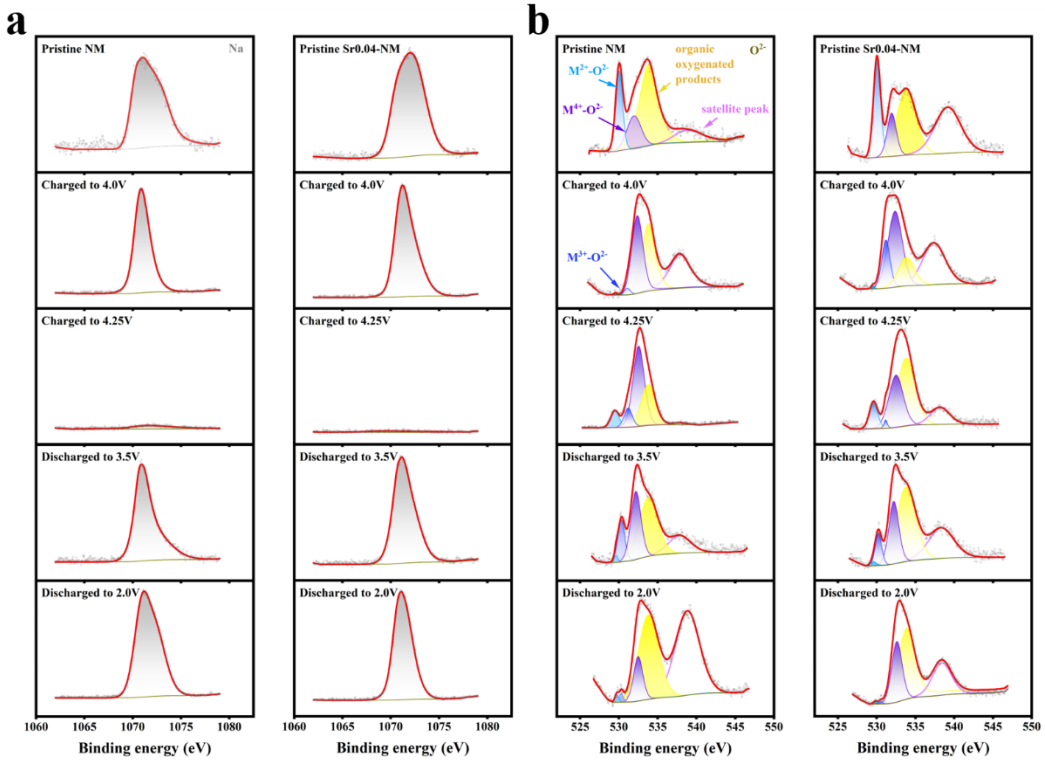

Figure S4. Ex-situ XPS patterns for (a) sodium and (b) oxygen elements at five electrochemical states.

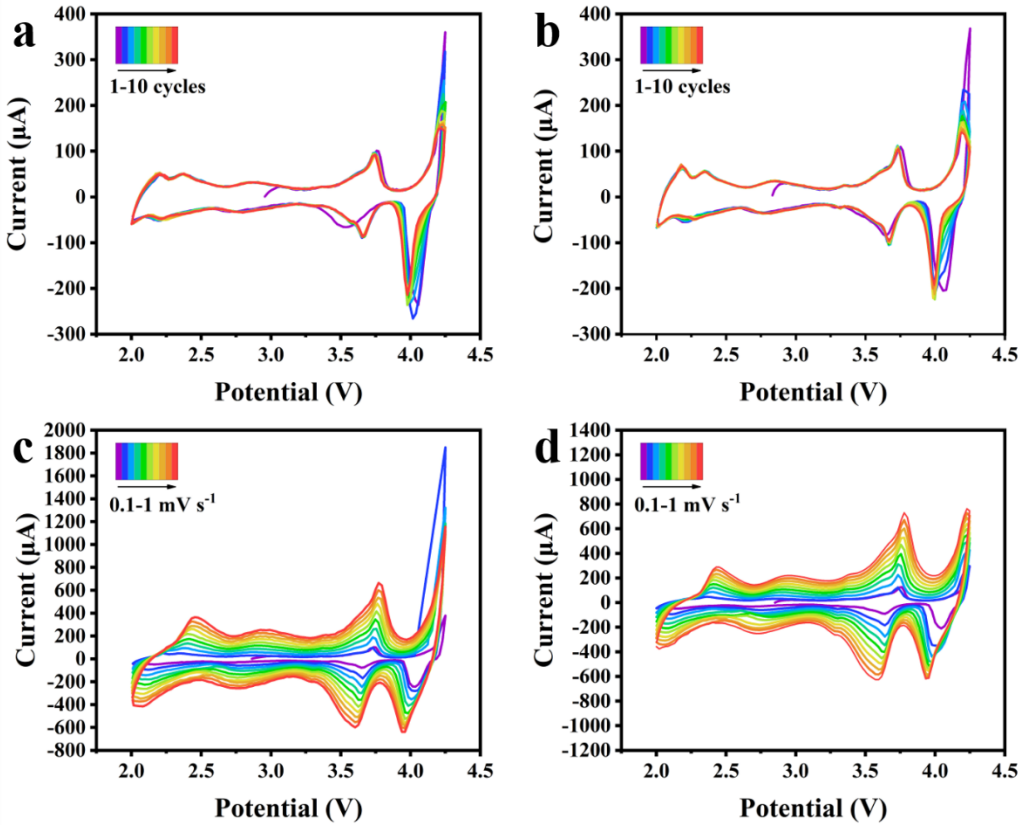

Figure S5. Multi-cycle cyclic voltammetry curves of (a) Sr0.02-NM, (b) Sr0.06-NM, and multi-scan-rate cyclic voltammetry curves of (c) Sr0.02-NM, (d) Sr0.06-NM.

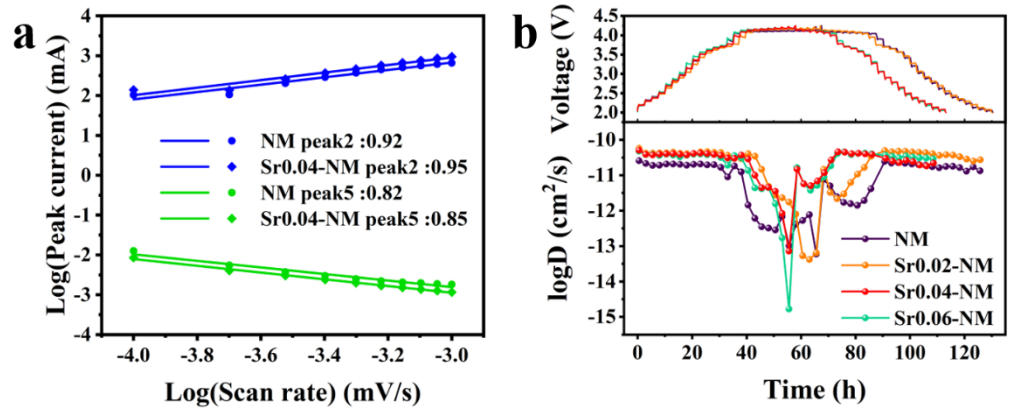

Figure S6. The peak currents of the (a) peaks 2 and 5 of NM and Sr0.04-NM, (b) GITT test (time *vs.* log D).

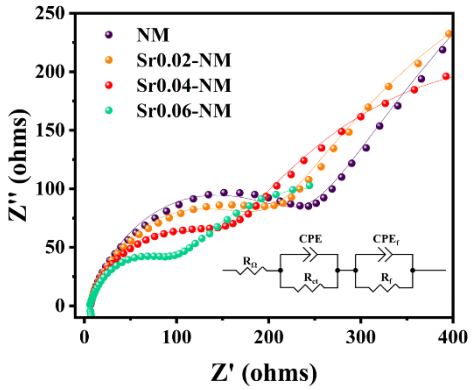

Figure S7. The Nyquist plots of the electrochemical impedance spectroscopies test with an equivalent circuit embedded.

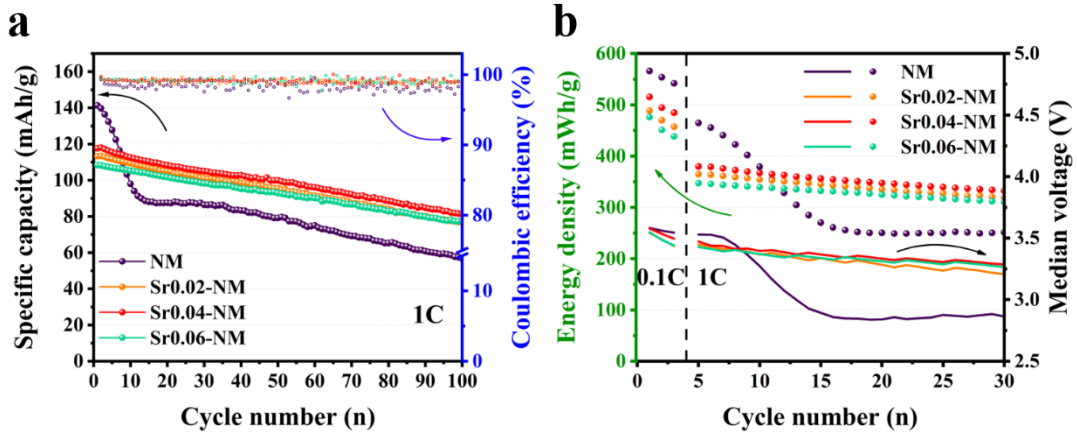

Figure S8. Electrochemistry performance. (a) Cycle performance of samples at 1 C between 2.0 V and 4.25 V. (b) Energy density and median voltage of the electrodes.

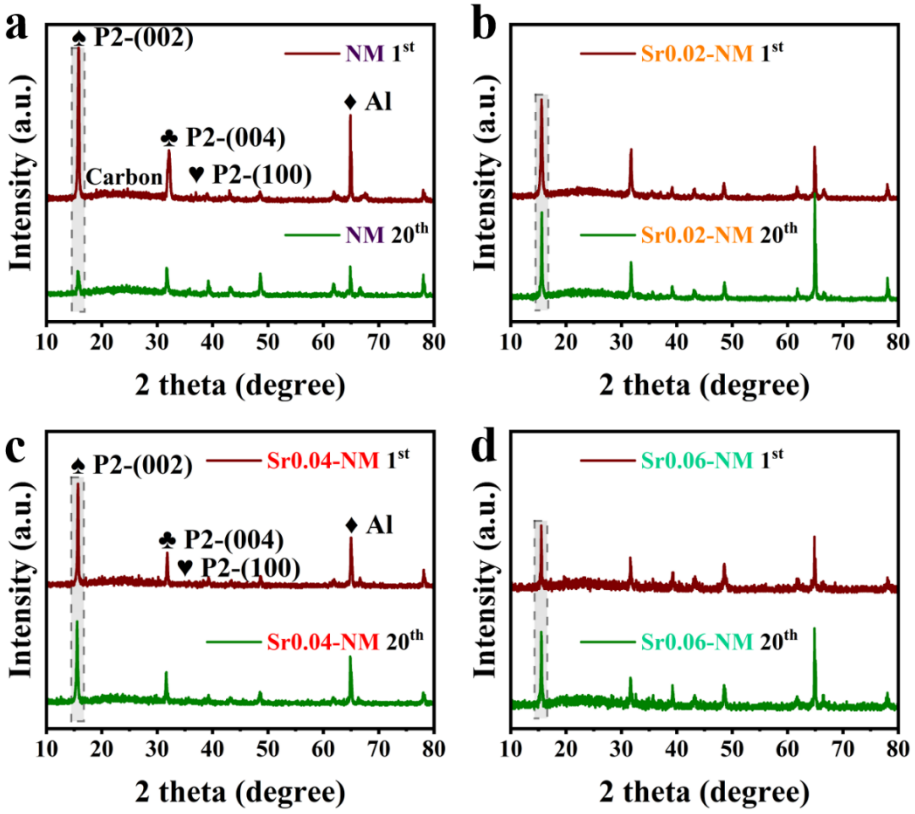

Figure S9. The XRD patterns of the electrodes before and after 20 cycles at 1 C rate for NM and Sr0.04-NM.

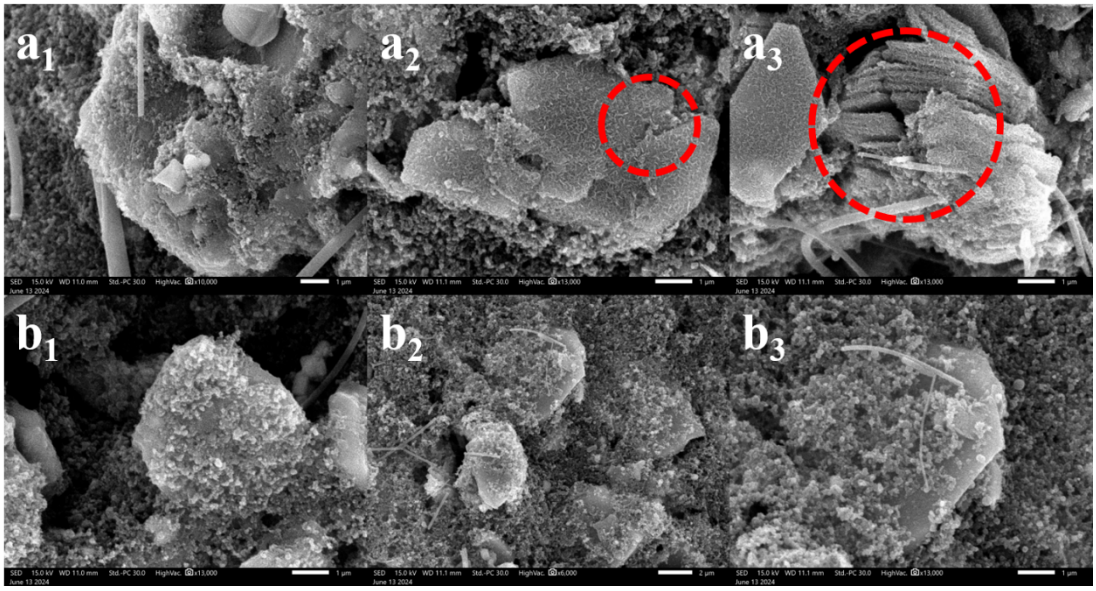

Figure S10. SEM images of the electrodes for (a) NM and (b) Sr0.04-NM. (a_1) and (b_1) are the images of pristine electrodes before testing. (a_2), (a_3), (b_2), and (b_3) belong to the electrodes after 20 cycles at a 5 C current rate.

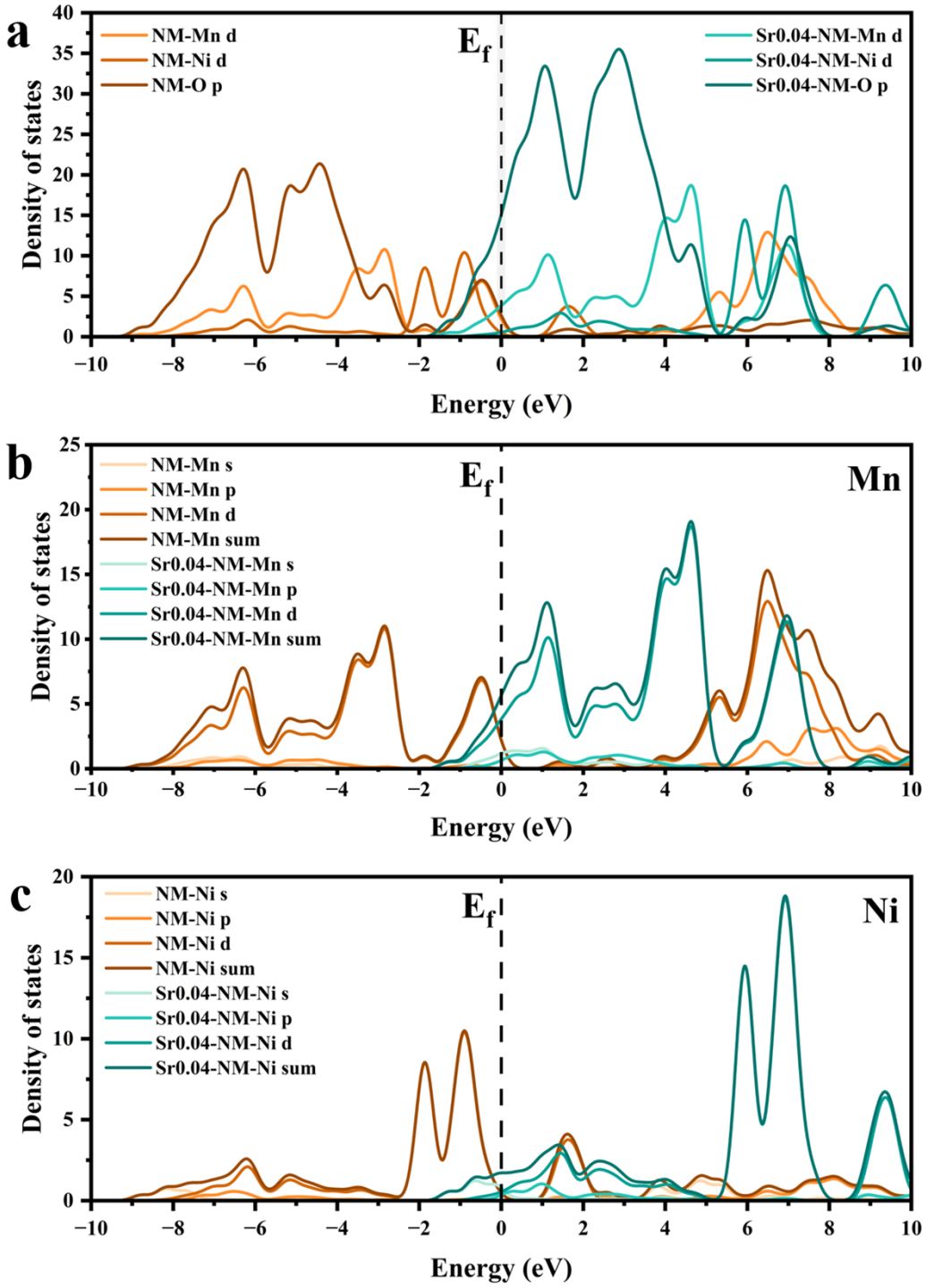

Figure S11. DFT calculations of NM and Sr0.04-NM. (a) The total density of states and the pDOS of (b) manganese and (c) nickel.

Table S1. The crystallographic results of NM from XRD refinement.

Space group = $P6_3/mmc$		**R_p = 6.16%**		**R_{wp} = 10.65%**		
a (Å) = b (Å) = 2.878780		**c (Å) = 11.270700**		**V (Å^3) = 80.902**		
Atom	**Wyckoff position**	**x**	**y**	**z**	**U_{iso}**	**Occ.**
O	4f	0.6667	0.3333	0.0878	0.00923	1.0473
Na_f	2b	0.0000	0.0000	0.2500	0.14409	0.5085
Na_e	2d	0.3333	0.6667	0.7500	0.12125	0.7670
Ni	2a	0.0000	0.0000	0.0000	0.14846	0.3199
Mn	2a	0.0000	0.0000	0.0000	-0.03354	0.7306

Table S2. The crystallographic results of Sr0.02-NM from XRD refinement.

Space group = $P6_3/mmc$		**R_p = 6.18%**		**R_{wp} = 10.58%**		
a (Å) = b (Å) = 2.879209		**c (Å) = 11.263156**		**V (Å^3) = 80.861**		
Atom	**Wyckoff position**	**x**	**y**	**z**	**U_{iso}**	**Occ.**
O	4f	0.6667	0.3333	0.091788	-0.00164	0.8581
Na_f	2b	0.0000	0.0000	0.2500	0.06815	0.0681
Na_e	2d	0.3333	0.6667	0.7500	0.04325	0.3984
Ni	2a	0.0000	0.0000	0.0000	0.01514	0.3363
Mn	2a	0.0000	0.0000	0.0000	0.00938	0.6668
Sr_f	2b	0.0000	0.0000	0.2500	0.06544	0.0067
Sr_e	2d	0.3333	0.6667	0.7500	0.05310	0.0133

Table S3. The crystallographic results of Sr0.04-NM from XRD refinement.

Space group = $P6_3/mmc$		**R_p = 6.76%**			**R_{wp} = 11.30%**	
a (Å) = b (Å) = 2.879814		**c (Å) = 11.255483**			**V (Å^3) = 80.789**	
Atom	**Wyckoff position**	**x**	**y**	**z**	**U_{iso}**	**Occ.**
O	4f	0.6667	0.3333	0.094207	0.02081	0.9573
Na_f	2b	0.0000	0.0000	0.2500	0.09217	0.3814
Na_e	2d	0.3333	0.6667	0.7500	0.13286	0.4986
Ni	2a	0.0000	0.0000	0.0000	0.02229	0.3220
Mn	2a	0.0000	0.0000	0.0000	0.00800	0.6529
Sr_f	2b	0.0000	0.0000	0.2500	0.12355	0.0131
Sr_e	2d	0.3333	0.6667	0.7500	0.02977	0.0267

Table S4. The crystallographic results of Sr0.06-NM from XRD refinement.

Space group = $P6_3/mmc$		**R_p = 7.82%**			**R_{wp} = 12.65%**	
a (Å) = b (Å) = 2.878919		**c (Å) = 11.267184**			**V (Å^3) = 80.852**	
Atom	**Wyckoff position**	**x**	**y**	**z**	**U_{iso}**	**Occ.**
O	4f	0.6667	0.3333	0.093658	0.03380	0.9029
Na_f	2b	0.0000	0.0000	0.2500	0.19654	0.3359
Na_e	2d	0.3333	0.6667	0.7500	0.17187	0.4603
Ni	2a	0.0000	0.0000	0.0000	-0.04947	0.3300
Mn	2a	0.0000	0.0000	0.0000	0.15520	0.6659
Sr_f	2b	0.0000	0.0000	0.2500	0.01867	0.0298
Sr_e	2d	0.3333	0.6667	0.7500	0.08738	0.0314

Table S5. ICP-OES results. The mass and mole fraction of each element of NM and Sr0.04-NM.

Element	Designed mol ratio	wt%	mol%	Normalization
Na	**0.67**	**12.19131**	**0.530288**	**0.575234**
Ni	**0.33**	**14.27771**	**0.243261**	**0.263879**
Mn	**0.67**	**33.93238**	**0.617649**	**0.67**
Na	**0.67**	**12.21079**	**0.531135**	**0.566816**
Sr	**0.04**	**2.700095**	**0.030812**	**0.032882**
Ni	**0.33**	**14.62216**	**0.249129**	**0.265866**
Mn	**0.67**	**34.49133**	**0.627823**	**0.67**

Table S6. The fitting results of the electrochemical impedance spectroscopies.

Electrodes	R_{Ω}	R_{ct}
NM	**6.075**	**221.2**
Sr0.02-NM	**6.095**	**184.3**
Sr0.04-NM	**6.323**	**129.8**
Sr0.06-NM	**5.85**	**87.39**

Table S7. The results of the XRD peak intensity in **Figure S8** are based on the diffraction peak of Al.

Electrodes	Pristine ratio to intensity of Al	Ratio of electrodes after 20 cycles	Retention
NM	1.73	0.75	43%
Sr0.02-NM	1.82	0.83	45%
Sr0.04-NM	2.08	1.71	82%
Sr0.06-NM	1.15	0.94	82%

Table S8. Electrochemical properties of Ni/Mn-based cathodes under different voltage window

Cathodes	Voltage range (V)	Rate capacity		Cyclic stability			Ref.
		Maximum capacity (rate) (mAh g^{-1})	High rate capacity (rate) (mAh g^{-1})	Initial capacity (rate) (mAh g^{-1})	Cycle numbers	Retention (%)	
P2-$Na_{0.59}Sr_{0.04}Ni_{0.33}Mn_{0.67}O_2$	2.0-4.25	154.2 (0.1 C)	68.3 (10 C)	117.6 (1 C)	100	69	This work
P2-$Na_{0.57}Ni_{0.33}Mn_{0.67}O_2$	2.0-4.25	171.1 (0.1 C)	25.8 (10 C)	141.3 (1 C)	100	40.1	This work
O3-$NaNi_{1/3}Fe_{1/3}Mn_{1/3}O_2$	2.0-4.0	~131 (0.1 C)	~82 (10 C)	~119 (1 C)	100	63	11
O3/P2 $Na_{0.88}Ni_{0.45}Mn_{0.55}O_2$	2.0-4.0	127.2 (0.05 C)	~78 (10 C)	106.7 (1 C)	250	71.1	15
P2-$Na_{0.78}Al_{0.05}Ni_{0.33}Mn_{0.60}O_2$	2.0-4.5	123.9 (0.1 C)	41.2 (5 C)	131 (0.1 C)	50	83.9	19
P2-$Na_{0.75}Ca_{0.04}[Li_{0.1}Ni_{0.2}Mn_{0.67}]O_2$	2.0-4.3	133.1 (0.1 C)	68.8 (20 C)	136.9 (0.1C)	50	94.2	27
P2-$Na_{0.67}Ni_{0.15}Fe_{0.2}Mn_{0.65}F_{0.05}O_{1.95}$	1.5-4.3	229 (0.1 C)	100 (10 C)	~229 (0.1 C)	50	87.7	35
P2-$Na_{2/3}[Ni_{0.3}Co_{0.1}Mn_{0.6}]O_2$	2.0-4.3	161.6 (0.1 C)	~90 (10 C)	161.6 (0.1 C)	50	79.2	45
P2-$Na_{2/3}[Ni_{1/3}Mn_{2/3}]O_2$	2.3-4.5	134 (0.01 C)	None	134 (0.01 C)	10	64	53