Supporting Information

A high entropy O3-$Na_{1.0}Li_{0.1}Ni_{0.3}Fe_{0.1}Mn_{0.25}Ti_{0.25}O_2$ cathode with reversible phase transitions and superior electrochemical performances for sodium-ion batteries

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Equation S1: The ideal configurational entropy (S_{conf}) of NFTMO material was calculated using the formula-

$$S_{conf} = -R\sum_i x_i ln x_i$$

Where R is the gas constant and x_i is the mole fraction of the ith element. The chemical composition of NFMTO is $Na_{1.0}Li_{0.1}Ni_{0.3}Fe_{0.1}Mn_{0.25}Ti_{0.25}O_2$. therefore, the mole fraction of Li, Ni, Fe, Mn and Ti is 0.1, 0.3, 0.1, 0.25 and 0.25 respectively.

Putting the values in Equation 2 we get,

$$S_{conf} = -R(0.1ln0.1 + 0.3ln0.3 + 0.1ln0.1 + 0.25ln0.25 + 0.25ln0.25) = 1.515R$$

Equation S2: The Na^+-ion diffusion coefficient was calculated by applying the galvanostatic intermittent titration technique (GITT) using the following formula[1] -

$$D_{Na^+} = \frac{4}{\pi\tau}\left(\frac{m_B V_m}{M_B S}\right)^2\left(\frac{\Delta Es}{\Delta E\tau}\right)^2 \qquad \left(where,\ \tau \ll \frac{L^2}{D_{Na^+}}\right)$$

D_{Na^+} is the diffusion coefficient of Na^+-ion, τ is the constant current pulse time (s), m_B is the weight of active material (g), V_m is the molar volume ($cm^3\ mol^{-1}$) of the material, M_B is the molecular weight of the cathode material ($g\ mol^{-1}$), S is the contact area of the electrolyte/electrode surface (cm^2), ΔE_s is the steady-state voltage change, ΔE_τ is the transient voltage change during the constant current pulse and L is the diffusion distance of Na^+ from lattice to liquid electrolyte.

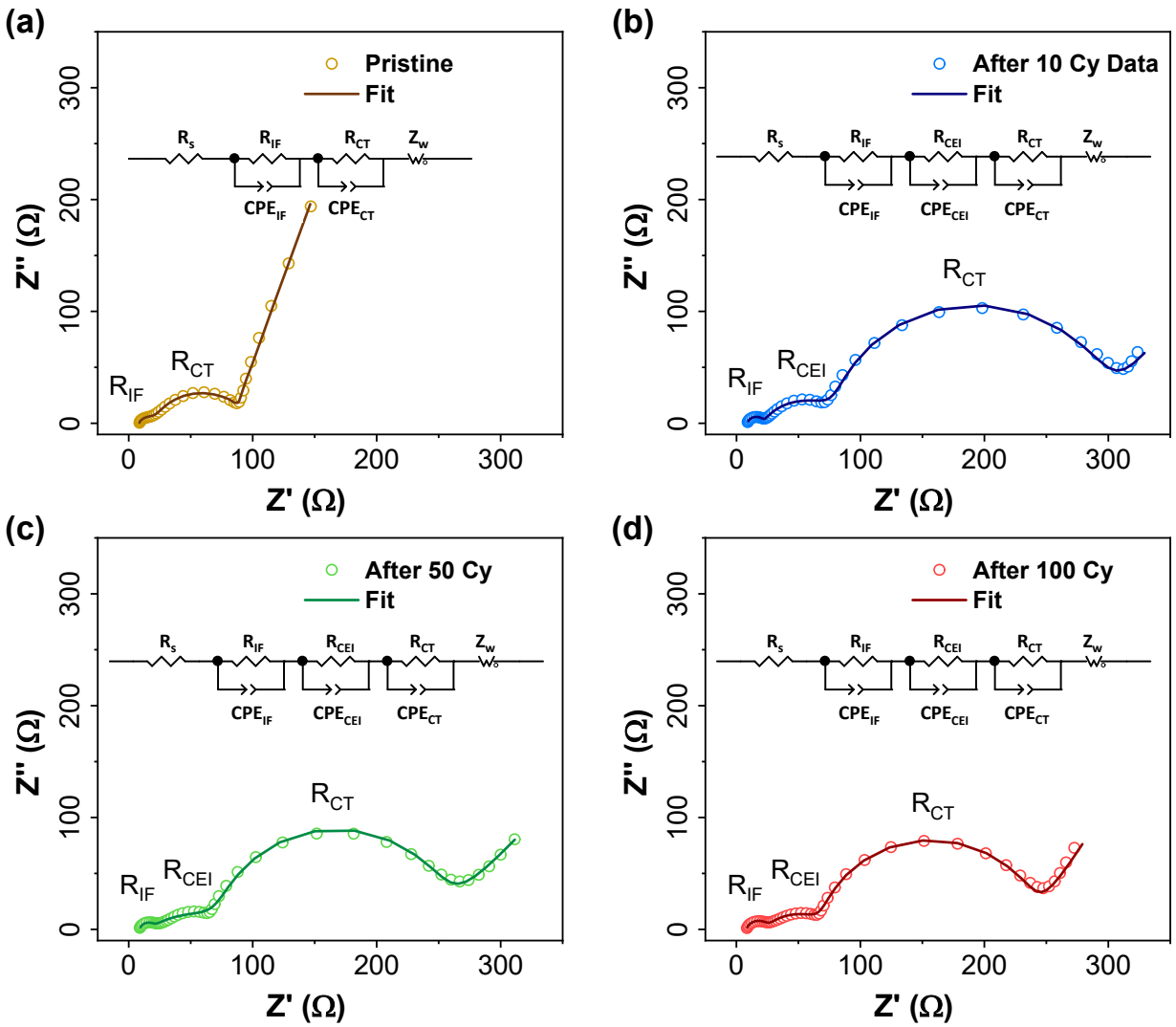

Figure S1: Nyquist plots with equivalent circuit models for fitting the electrochemical impedance spectroscopy data of NFTMO cathode collected at (a) pristine, (b) after 10 cycles, (c) after 50 cycles, (d) after 100 cycles.

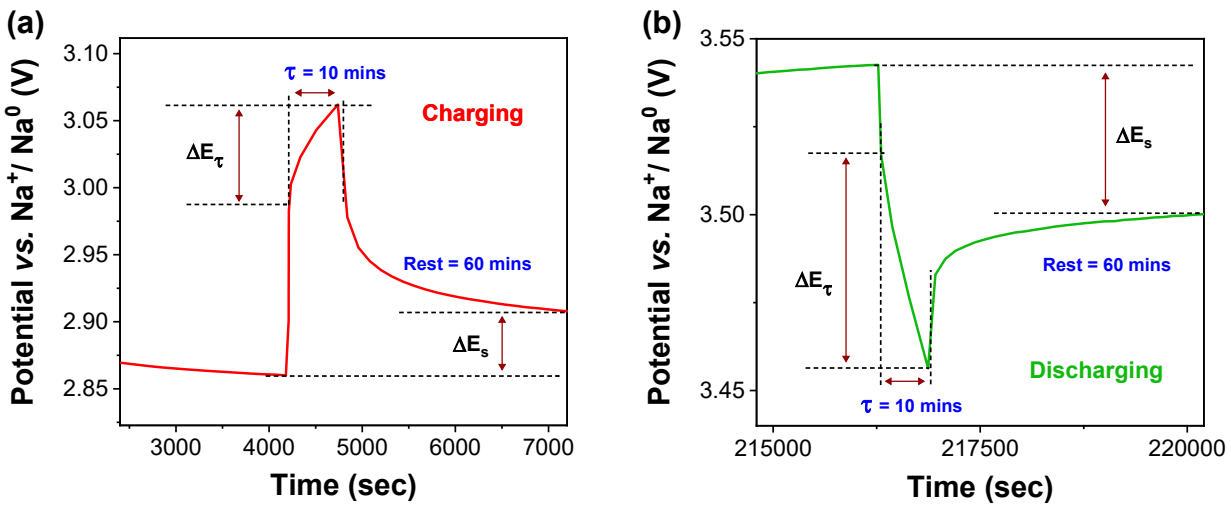

Figure S2: The schematic diagram of a single current pulse for GITT of (a) charging and (b) discharging process and the corresponding parameters.

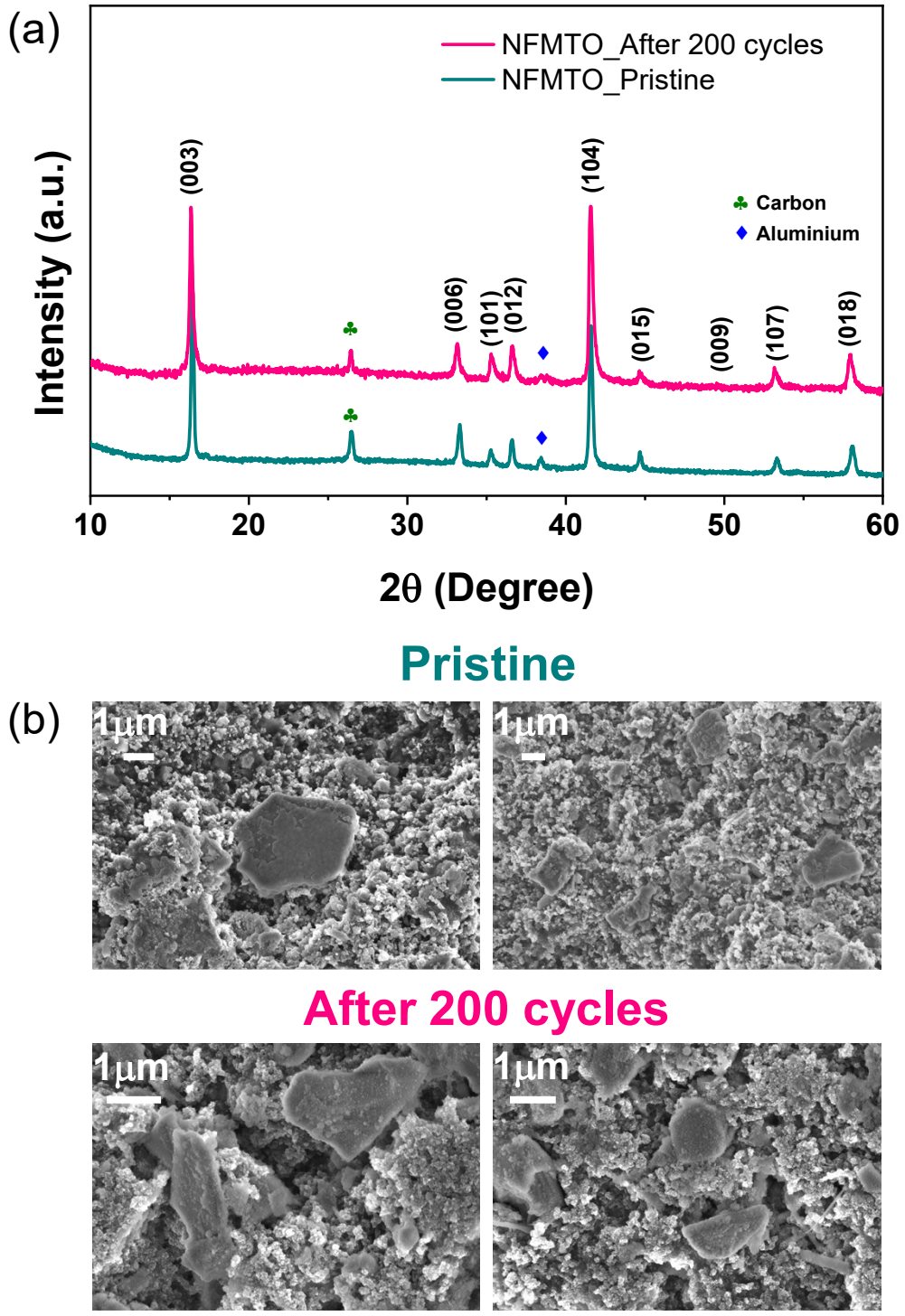

Figure S3: (a) PXRD and (b) SEM images of NFMTO electrode in the pristine state and after 200 cycles in the potential window of 4.0-2.0 V *vs.* Na^+/ Na^0 at 0.1 C rate**.**

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Table S1: Structural parameters of $Na_{1.0}Li_{0.1}Ni_{0.3}Fe_{0.1}Mn_{0.25}Ti_{0.25}O_2$ cathode

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Title $Na_{1.0}Li_{0.1}Ni_{0.3}Fe_{0.1}Mn_{0.25}Ti_{0.25}O_2$
Lattice type R
Space group name R -3 m
Space group number 166

Lattice parameters

a = 2.979(4) Å α = 90.00°
b = 2.979(4) Å β = 90.00°
c = 16.020(5) Å γ = 120.00°
V = 123.165 $Å^3$

Structure parameters

Atom	x	y	z	Occ.	U	Site	Sym
1 O	0.000	0.000	0.236	1.000	0.009	6c	3m
2 Fe	0.000	0.000	0.500	0.100	0.000	3b	-3m
3 Mn	0.000	0.000	0.500	0.250	0.000	3b	-3m
4 Ni	0.000	0.000	0.500	0.300	0.000	3b	-3m
5 Ti	0.000	0.000	0.500	0.250	0.000	3b	-3m
6 Li	0.000	0.000	0.500	0.100	0.000	3b	-3m
7 Na	0.000	0.000	0.000	1.000	0.028	3a	-3m

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Table S2: Electrochemical performance comparison of reported multi-element based (binary to high entropy) layered oxide cathode materials.

No.	**Material**	**Potential Window (V *vs* Na^+/Na^0)**	**Applied Rate (C)**	**Q_{Dis} ($mAh \cdot g^{-1}$)**	**V_{avg} (V)**	**Energy Density ($Whkg^{-1}$)**	**Capacity Retention**	**Ref.**
[1]	$NaNi_{0.5}Mn_{0.5}O_2$	3.8-2.2	0.02 C	125	~2.9	362.5	75% (0.1 C, 50 cycles)	2
[2]	$NaNi_{0.33}Mn_{0.33}Fe_{0.33}O_2$	4.0-2.0	0.1 C	136	3.15	428.4	85.88% (0.1C, 50 cycles)	3
[3]	$NaNi_{0.5}Mn_{0.2}Ti_{0.3}O_2$	4.0-2.0	0.05 C	135	3.2	432	85% (1 C, 200 cycles)	4
[4]	$Na[Ni_{0.4}Mn_{0.4}Fe_{0.2}]_{0.95}Ti_{0.05}O_2$	4.0-2.0	0.1 C	160.7	~3.1	498.2	83.8% (1C, 200 cycles)	5

[5]	$NaNi_{0.4}Fe_{0.2}Mn_{0.2}Ti_{0.2}O_2$	4.2-2.0	0.1 C	145	3.2	464	84% (0.1 C, 200 cycles)	6
[6]	$NaNi_{0.45}Mn_{0.2}Ti_{0.3}Zr_{0.05}O_2$	4.0-2.0	0.05 C	141.4	3.0	424	70% (0.05 C, 200 cycles)	7
[7]	$NaNi_{0.45}Mn_{0.4}Ti_{0.1}Cu_{0.05}O_2$	4.0-2.0	0.1 C	124	3.1	384	70.2% (1 C, 500 cycles)	8
[8]	$NaNi_{0.25}Co_{0.25}Mn_{0.25}Ti_{0.25}O_2$	3.9-2.0	0.1 C	116	3.15	365.4	75% (5 C, 400 cycles)	9
[9]	$Na_{0.85}Li_{0.1}Ni_{0.175}Fe_{0.2}Mn_{0.525}O_2$	4.2-2.0	0.1 C	130	~3.4	429	95% (1 C, 100 cycles)	10
[10]	$NaNi_{0.3}Cu_{0.1}Fe_{0.2}Mn_{0.2}Ti_{0.2}O_2$	3.9-2.0	0.1 C	130	~3.15	409.5	71% (0.5 C, 500 cycles	11
[11]	$NaNi_{0.25}Co_{0.25}Fe_{0.25}Mn_{0.125}Ti_{0.125}O_2$	4.1-2.0	0.1 C	128	~3.1	396.8	96% (1 C, 100 cycles)	12
[12]	$NaNi_{0.12}Cu_{0.12}Mg_{0.12}Fe_{0.15}Co_{0.15}Mn_{0.1}Ti_{0.1}Sn_{0.1}Sb_{0.04}O_2$	3.9-2.0	0.1 C	110	~3.1	341	83% (0.5 C, 500 cycles)	13
[13]	$NaNi_{0.25}Mg_{0.05}Cu_{0.1}Fe_{0.2}Mn_{0.2}Ti_{0.1}Sn_{0.1}O_2$	4.0-2.0	0.1 C	130.8	3.21	421	75% (1 C, 500 cycles	14
[14]	$Na_{0.83}Li_{0.1}Ni_{0.25}Co_{0.2}Mn_{0.15}Ti_{0.15}Sn_{0.15}O_{2-\delta}$	4.2-2.0	0.1 C	123.3	~3.2	394.6	87.2% (2 C,200 cycles)	15
[15]	$NaLi_{0.1}Ni_{0.3}Fe_{0.1}Mn_{0.25}Ti_{0.25}O_2$	4.0-2.0	0.1 C	123	3.2	393.6	88% (1 C, 200 cycles) 77% (2 C, 400 cycles)	Our Work

Table S3: Fitted parameters from EIS of NFMTO cathode collected at (a) pristine, (b) after 10 cycles, (c) after 50 cycles, (d) after 100 cycles.

State	R_s (Ω)	R_{IF} (Ω)	R_{CEI} (Ω)	R_{CT} (Ω)
Pristine	8.094	13.25	-	72.01
After 10 Cycles	8.79	12.76	59.9	210
After 50 Cycles	8.44	15.39	39.06	163.8
After 100 Cycles	8.16	15.76	47.34	161.1

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