# Supplementary Information

## Mitigation of resistance increase in positive electrodes of lithium-ion batteries during floating-cycling modes by operating voltage control

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Table S1. Overview of durability test conditions and parameters.

| Mode | Step | Parameters | Termination |
|---|---|---|---|
| Floating-cycling | 1) CCCV Charge | $I$ = 1.675 A (0.5 C), $V$ = Variant | $t$ > 18 h |
| | 2) CC Discharge | $I$ = –3.35 A (1 C) | $V$ = Variant |
| | 3) Discharge Rest | $V$= open circuit voltage | $t$ > 5 h |

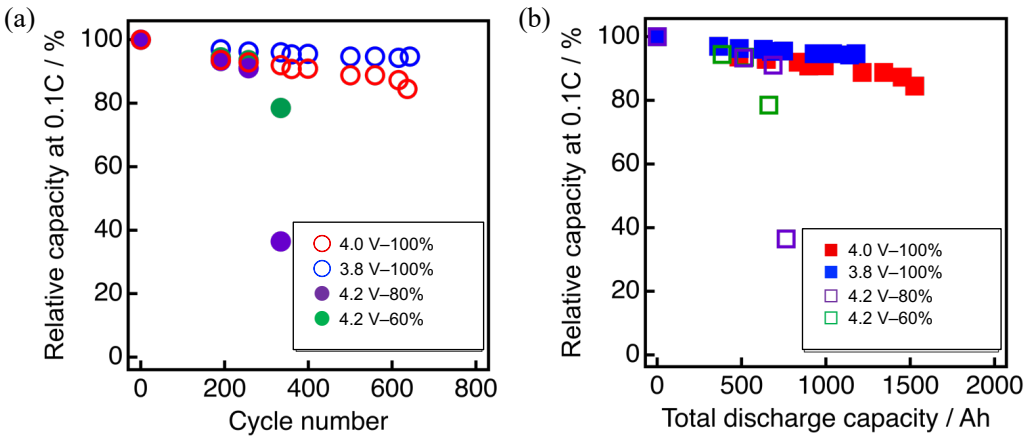



Fig. S1 Relative capacity of the horizontally-set cells at 0.1 C discharging after the durability test versus (a) tested days and (b) total discharge capacity (run at 1 C).

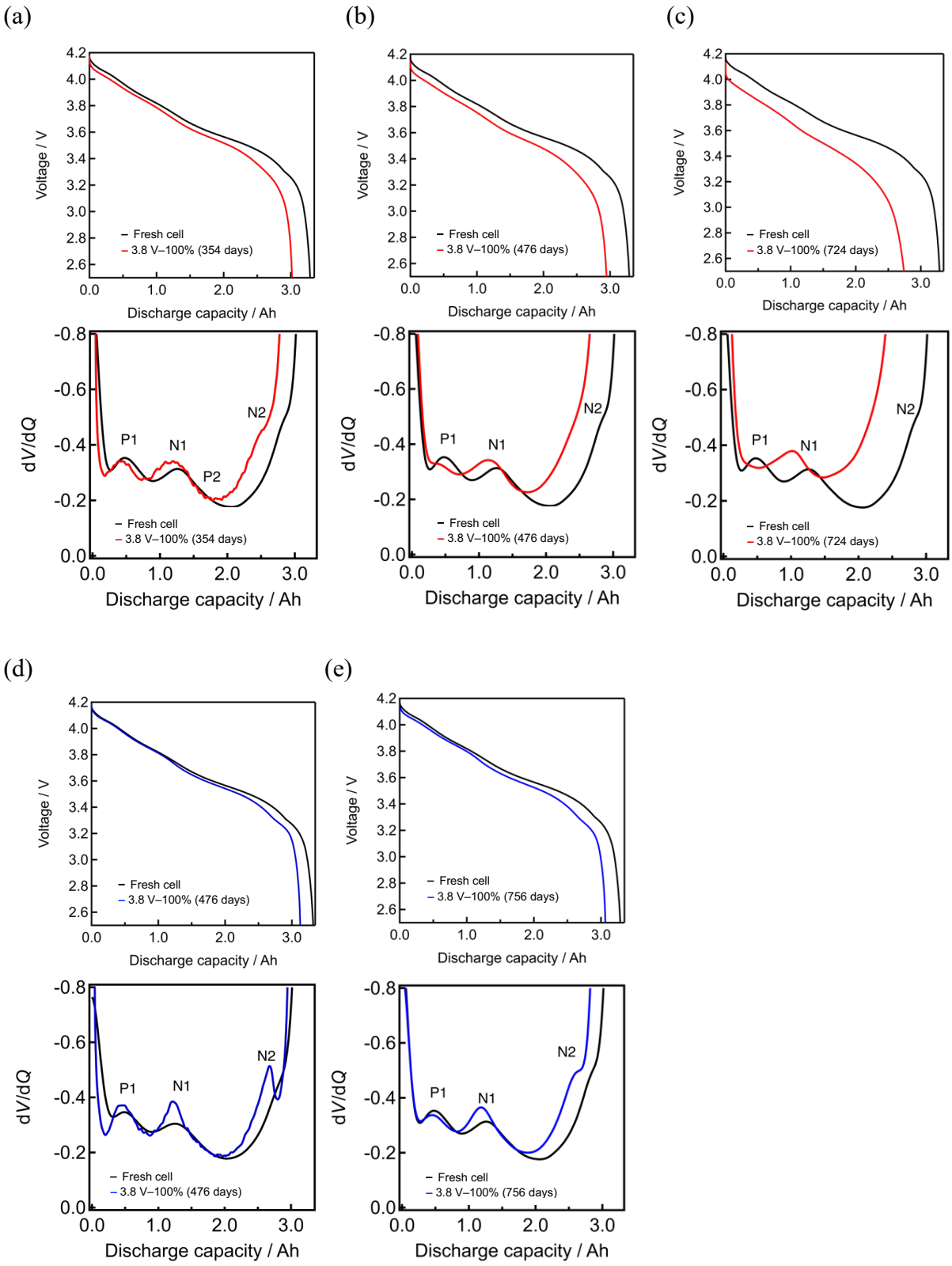



Fig. S2 0.1 C discharge profiles and the corresponding d$V$/d$Q$ profiles of 4.0 V–100% cell after (a) 354, (b) 476 and (c) 724 days and 3.8 V–100% cell after (d) 476 and (e) 756 days.

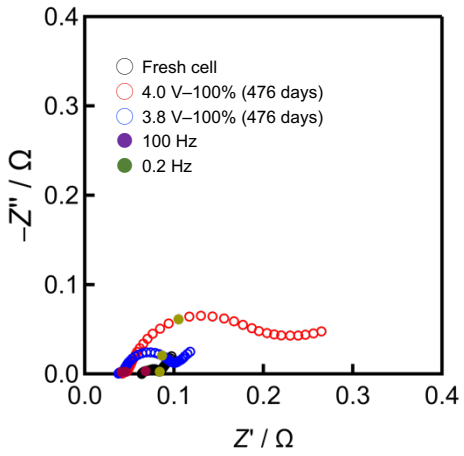



Fig. S3 Nyquist plots of 4.0 V–100% (after 476 days) and 3.8 V–100% (after 476 days) cells at fully charged states.

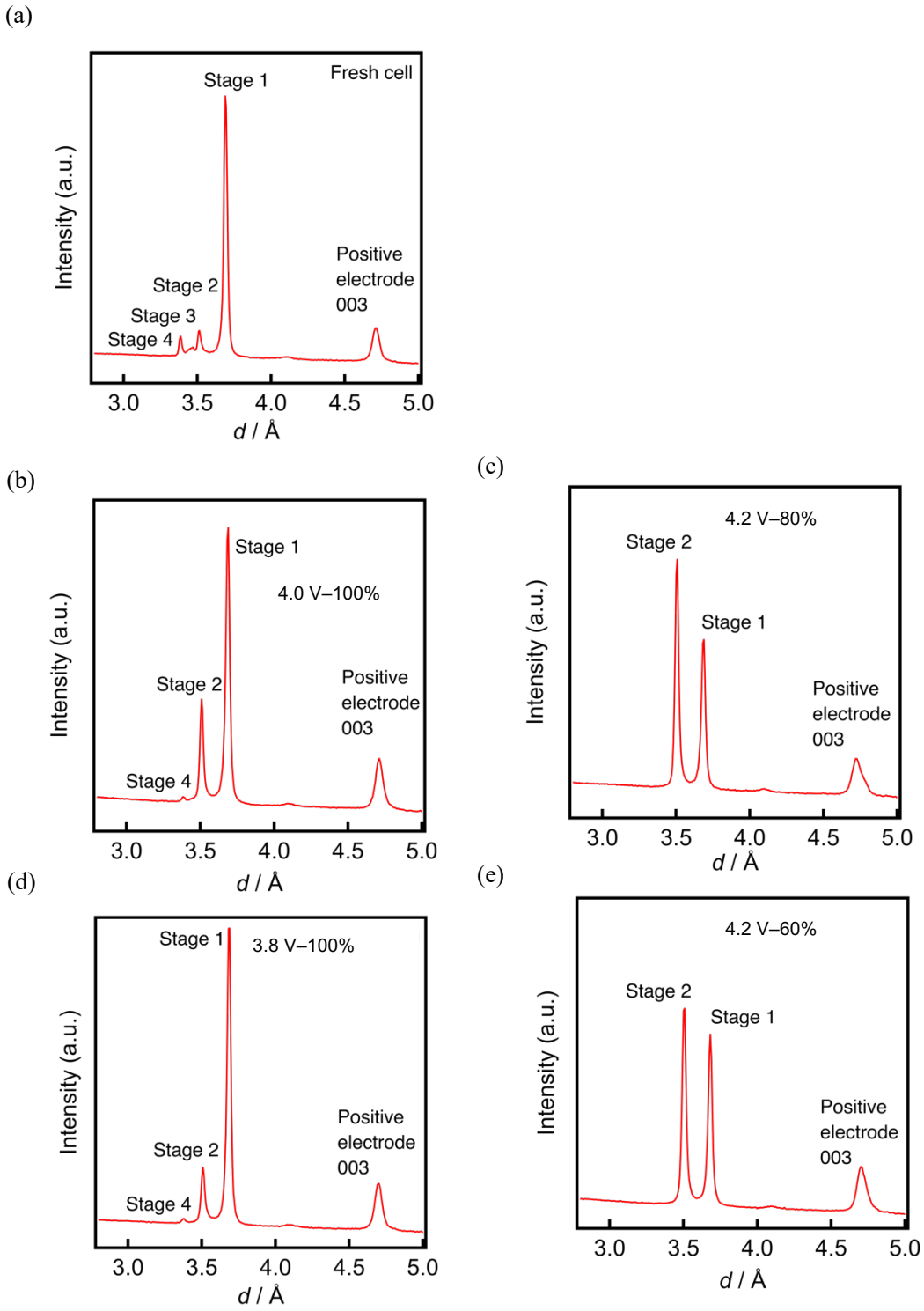



Fig. S4 In-situ neutron diffraction profiles at the end of charging of (a) fresh cell, (b) 4.0 V–100% after 476 days, (c) 4.2 V–80% after 323 days, (d) 3.8 V–100% after 476 days, and (e) 4.2 V–60% after 428 days.

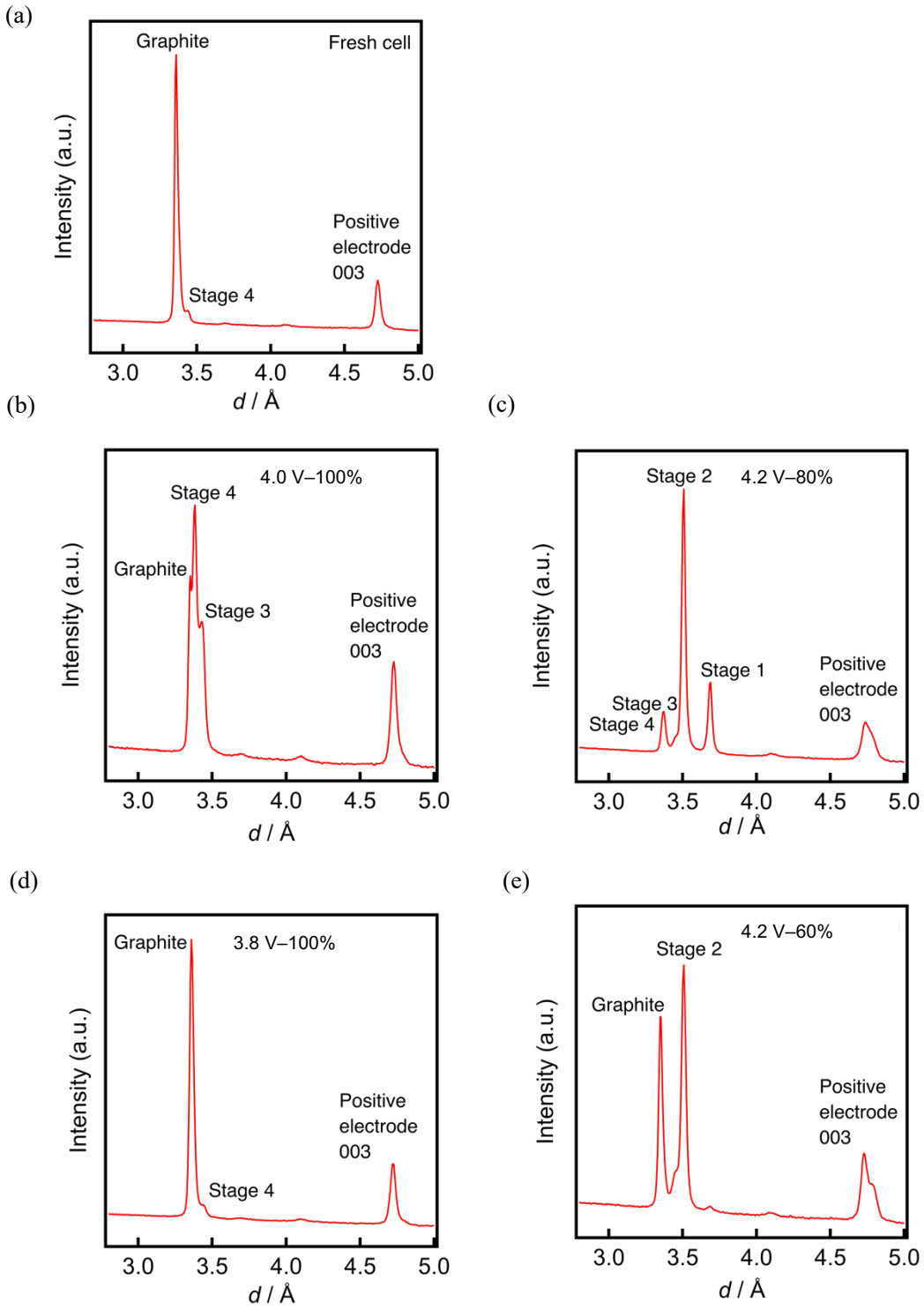



Fig. S5 In-situ neutron diffraction profiles at the end of discharging of (a) fresh cell, (b) 4.0 V–100% after 476 days, (c) 4.2 V–80% after 323 days, (d) 3.8 V–100% after 476 days, and (e) 4.2 V–60% after 428 days.

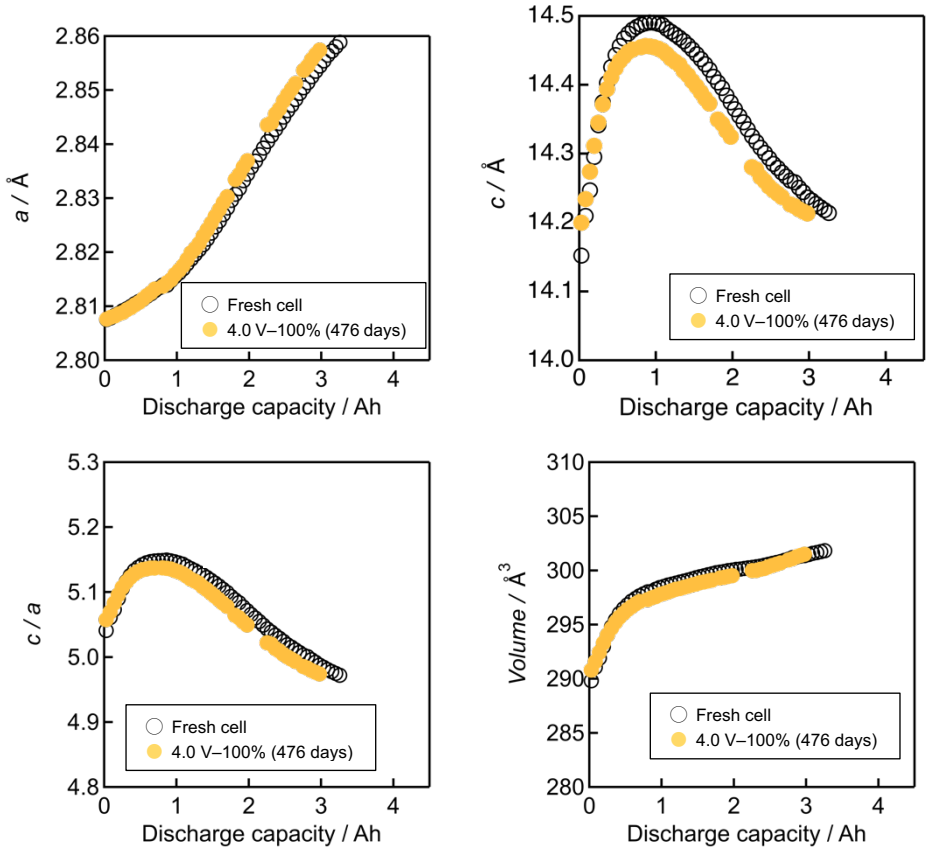



Fig. S6. Evolutions of lattice constants *a* and *c*, ratio *c*/*a* and unit cell volume of the layered positive material aged by 4.0 V–100% mode.

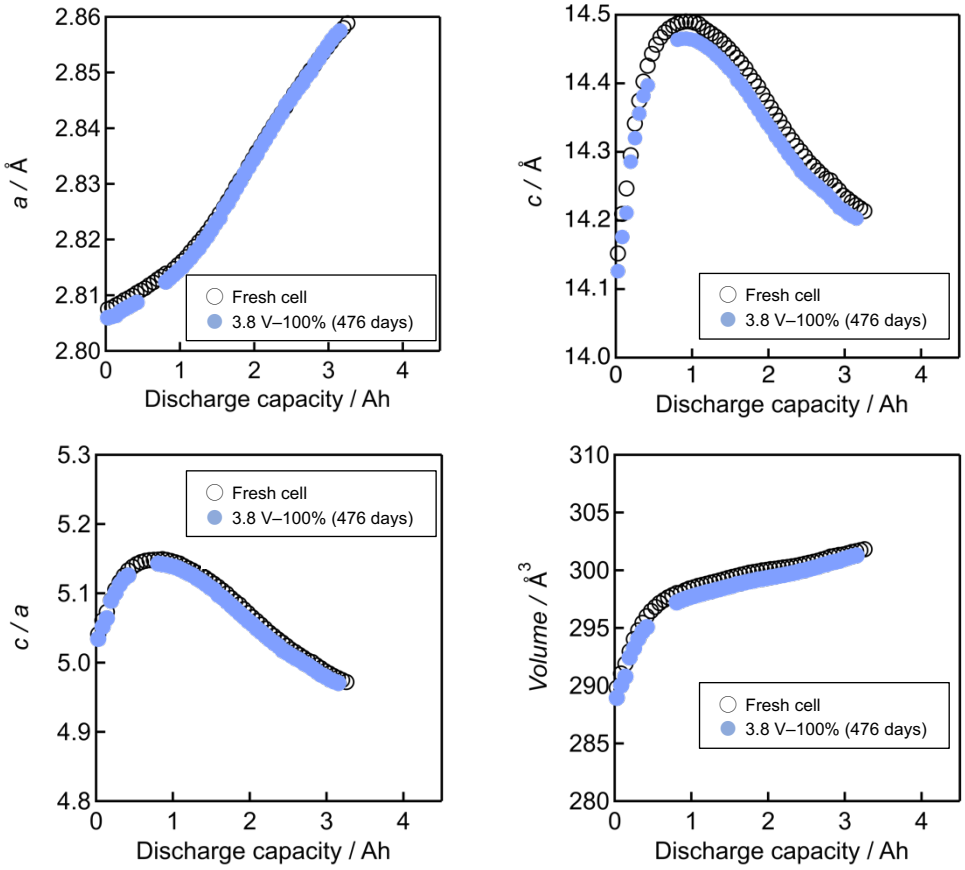
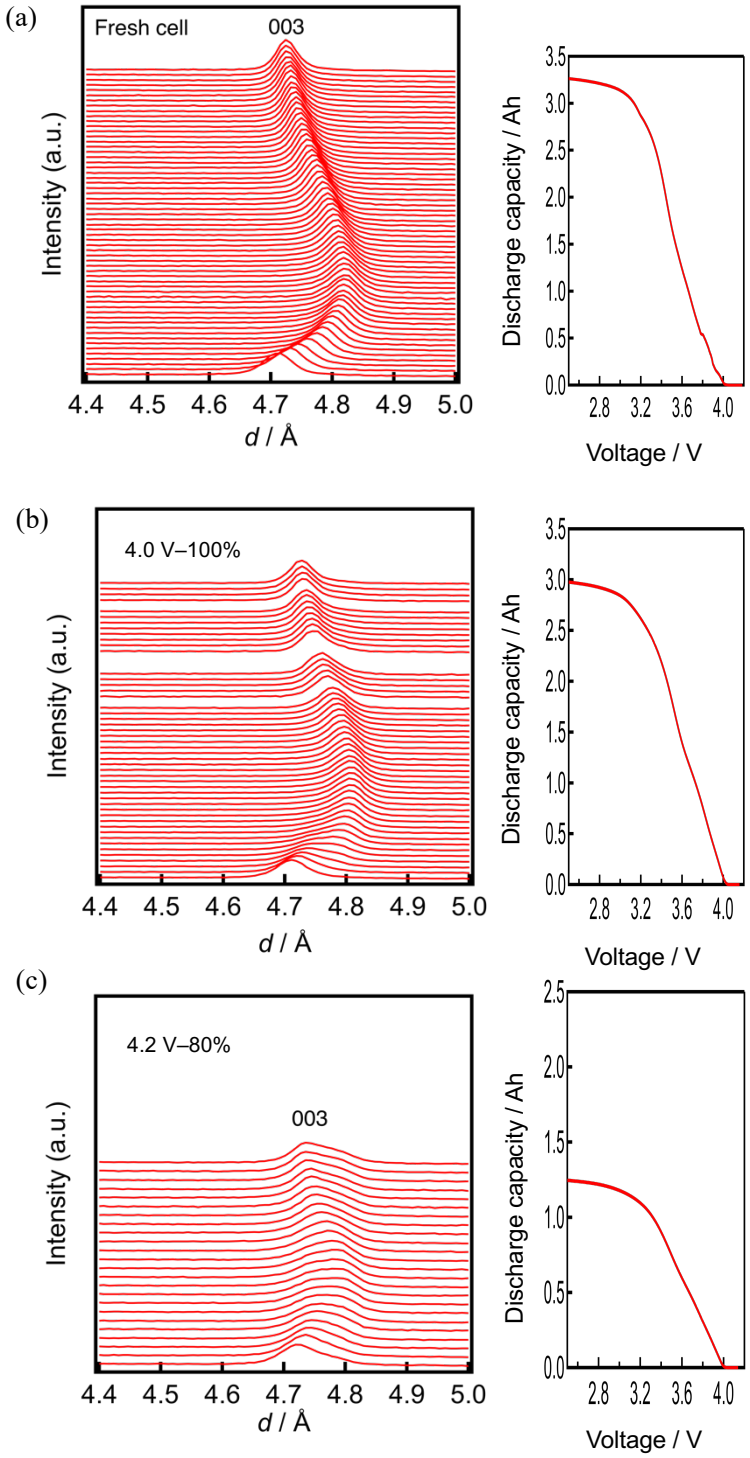



Fig. S7. Evolutions of lattice constants *a* and *c*, ratio *c*/*a* and unit cell volume of the layered positive material aged by 3.8 V–100% mode.


(a)
Fresh cell
003
Intensity (a.u.)
4.4 4.5 4.6 4.7 4.8 4.9 5.0
d / Å
Discharge capacity / Ah
3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
2.8 3.2 3.6 4.0
Voltage / V
(b)
4.0 V–100%
Intensity (a.u.)
4.4 4.5 4.6 4.7 4.8 4.9 5.0
d / Å
Discharge capacity / Ah
3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
2.8 3.2 3.6 4.0
Voltage / V
(c)
4.2 V–80%
003
Intensity (a.u.)
4.4 4.5 4.6 4.7 4.8 4.9 5.0
d / Å
Discharge capacity / Ah
2.5 2.0 1.5 1.0 0.5 0.0
2.8 3.2 3.6 4.0
Voltage / V

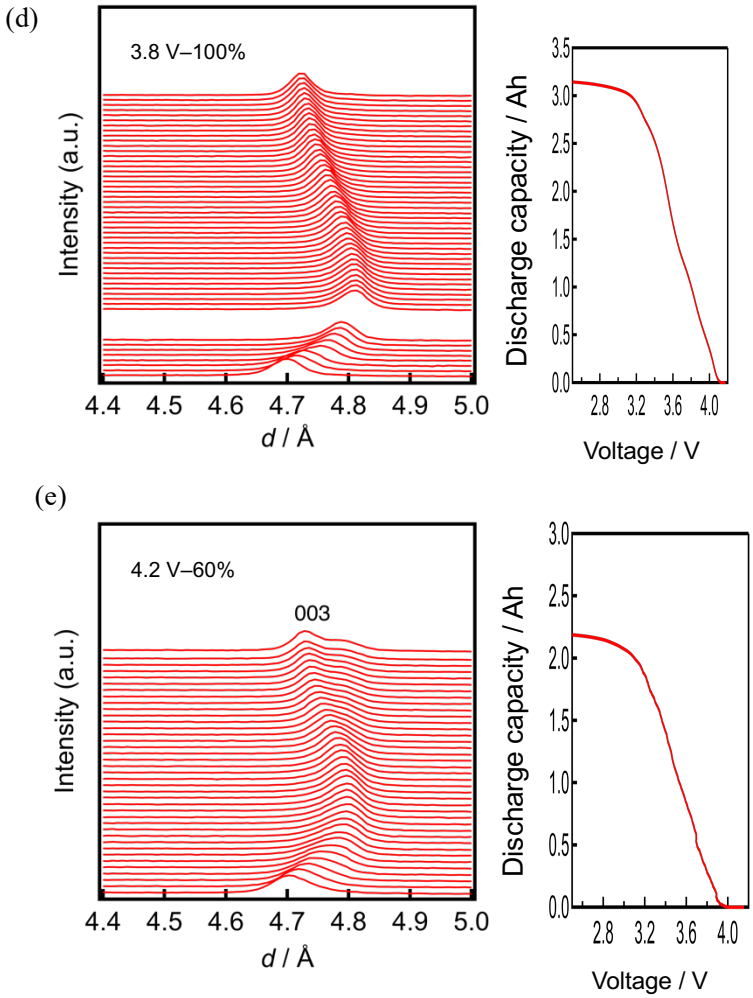



Fig. S8 Operando neutron diffraction and voltage profiles of layered positive 003 reflection during 0.1 C discharging (a) fresh cell, (b) 4.0 V–100% after 476 days, (c) 4.2 V–80% after 323 days, (d) 3.8 V–100% after 476 days, and (e) 4.2 V–60% after 428 days.

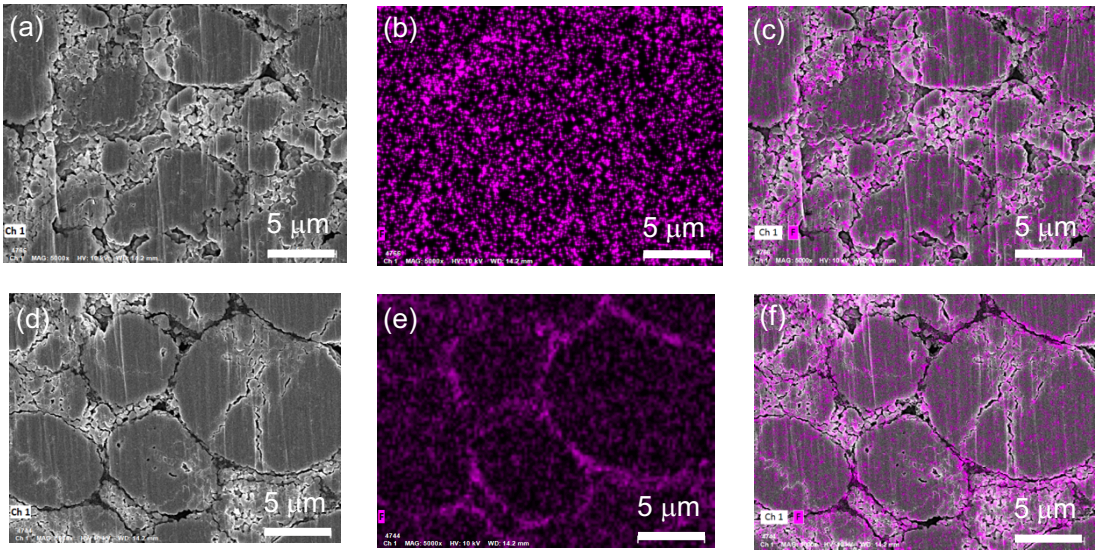



Fig. S9. (a), (d) SEM only, (b), (e) EDX only and (c), (f) the overlapped images of positive electrode particles derived from (a), (b), (c) 4.2 V–60% and (d), (e), (f) 3.8 V–100% aged cells.