

Figure S1. EDS mapping of O (a); Al (b); Co (c); Ni (d) of the  $\text{Ni}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}(\text{OH})_2$  commercial precursor.

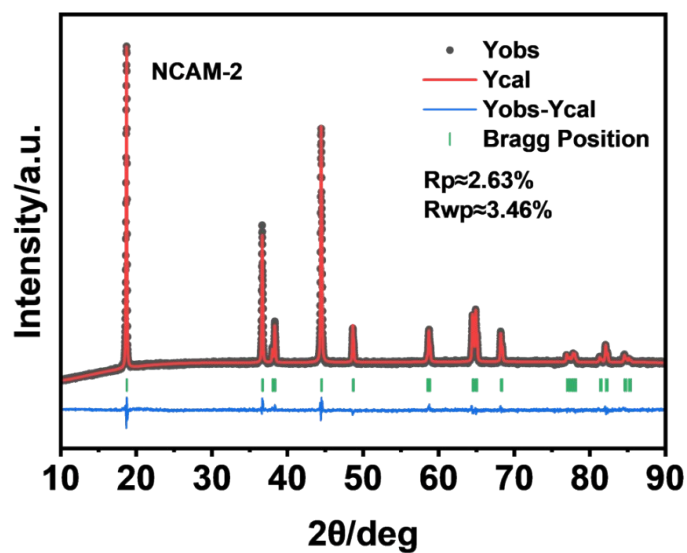


Figure S2. Refined XRD patterns of NCAM-2

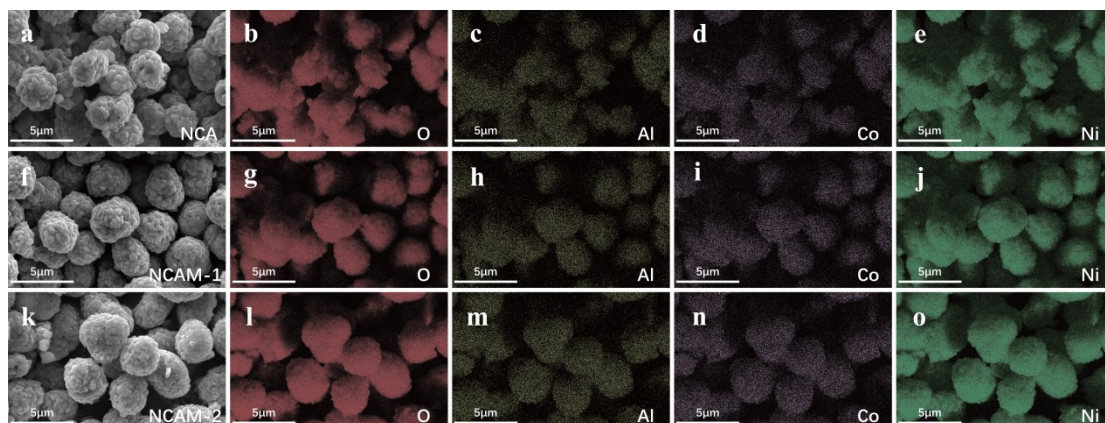


Figure S3. SEM images and EDS mapping of O, Al, Co, Ni of NCA (a-e), NCAM-1 (f-j) and NCAM-2 (k-o).

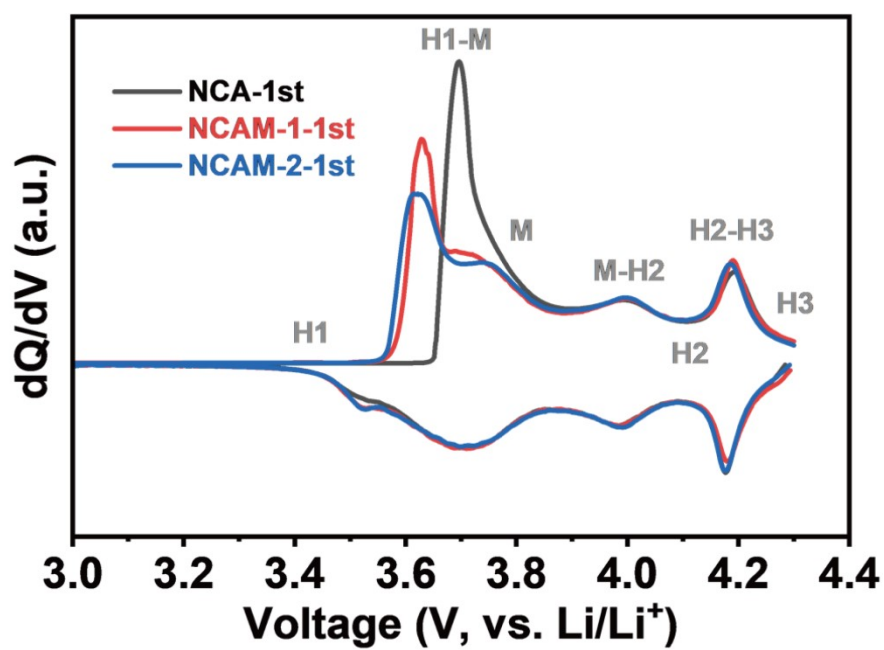


Figure S4. CV curves of NCA, NCAM-1 and NCAM-2 cathode materials, and their phase transitions.

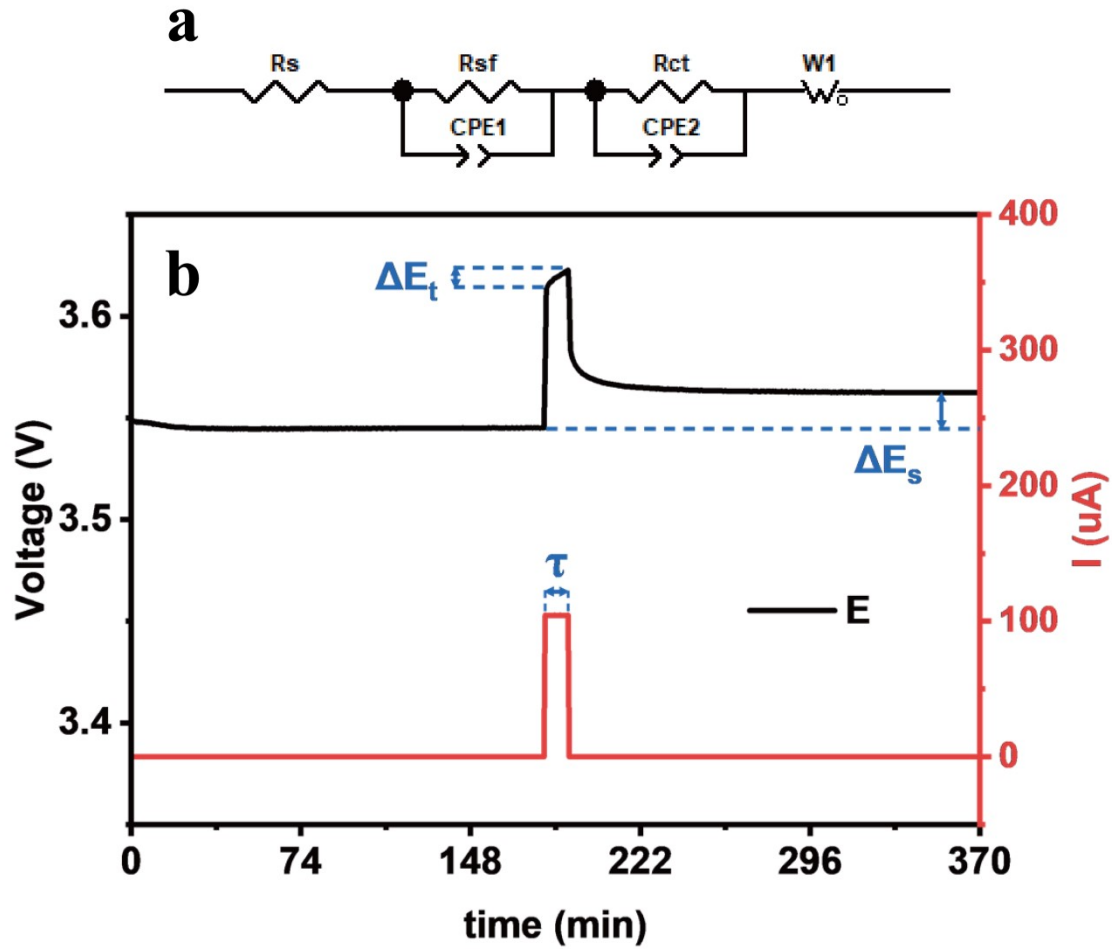


Figure S5. EIS equivalent fitting circuit (a); Single pulse of the GITT test (b).

The simplified formula of GITT are as follows:

$$D = \frac{4}{\pi\tau} \left( \frac{mV_M}{MS} \right)^2 \left( \frac{\Delta E_s}{\Delta E_t} \right)^2 \quad \left( \tau \ll \frac{L^2}{D} \right)$$

Figure S4b shows a single pulse of the GITT test, which  $\tau$  represents the duration of the current pulse (600 s),  $m$ ,  $V_M$  and  $M$  denotes the mass, the molar volume ( $202.3 \times 10^{-7} \text{ m}^3 \cdot \text{mol}^{-1}$ ) and the molar mass ( $97.61 \text{ g} \cdot \text{mol}^{-1}$ ) of the active material, respectively.  $S$  represents the electrode surface area ( $1.131 \times 10^{-4} \text{ m}^2$ ),  $\Delta E_s$  indicates the open-circuit potential difference before and after the current pulse, while  $\Delta E_t$  denotes the voltage variation during the current pulse.

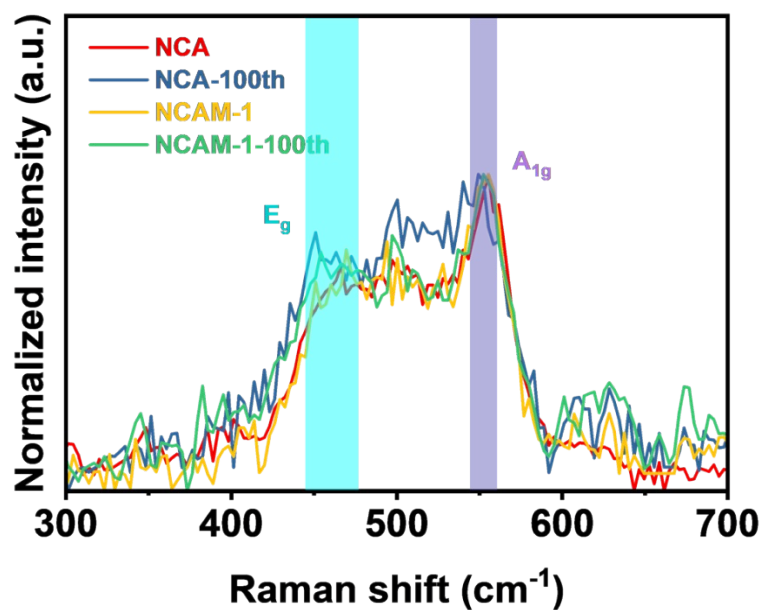


Figure S6. Raman spectra of NCA and NCAM-1 before and after 100 cycles of charge-discharge.

Table S1. Parameters of NCA, NCAM-1 and NCAM-2

Sample	a=b(Å)	c(Å)	c/a	$I_{(003)}/I_{(104)}$	Li <sup>+</sup> /Ni <sup>2+</sup> (%)
NCA	2.8685	14.1782	4.9427	1.20	4.35
NCAM-1	2.8691	14.1816	4.9429	1.60	2.23
NCAM-2	2.8693	14.1854	4.9439	1.36	3.64

Table S2. The  $R_{sf}$  and  $R_{ct}$  of NCA and NCAM-1

	Cycle number	$R_{sf}(\Omega)$	$R_{ct}(\Omega)$
NCA	1st	15.66	93.38
	100th	50.17	707.8
NCAM-1	1st	7.96	44.22
	100th	8.73	158.5

Table S3. The mean  $D_{Li+}$  of NCA and NCAM-1

	Mean $D_{Li+}$ of charge ( $cm^2 s^{-1}$ )	Mean $D_{Li+}$ of discharge( $cm^2 s^{-1}$ )
NCA	2.08062E-10	2.74127E-10
NCAM-1	3.10156E-10	3.51085E-10