Supporting Information

High-valence Ni^{3+} construction and stability by electrochemical de-lithiation boosting oxygen evolution

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Supplementary Figures

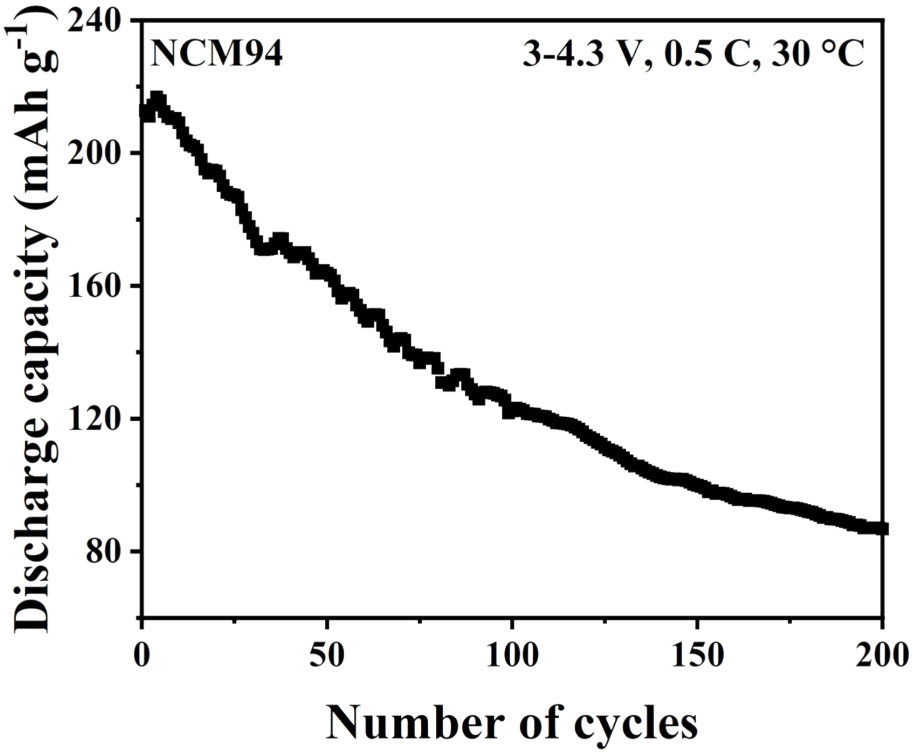

Fig. S1 Charging and discharging cycle curve of NCM94.

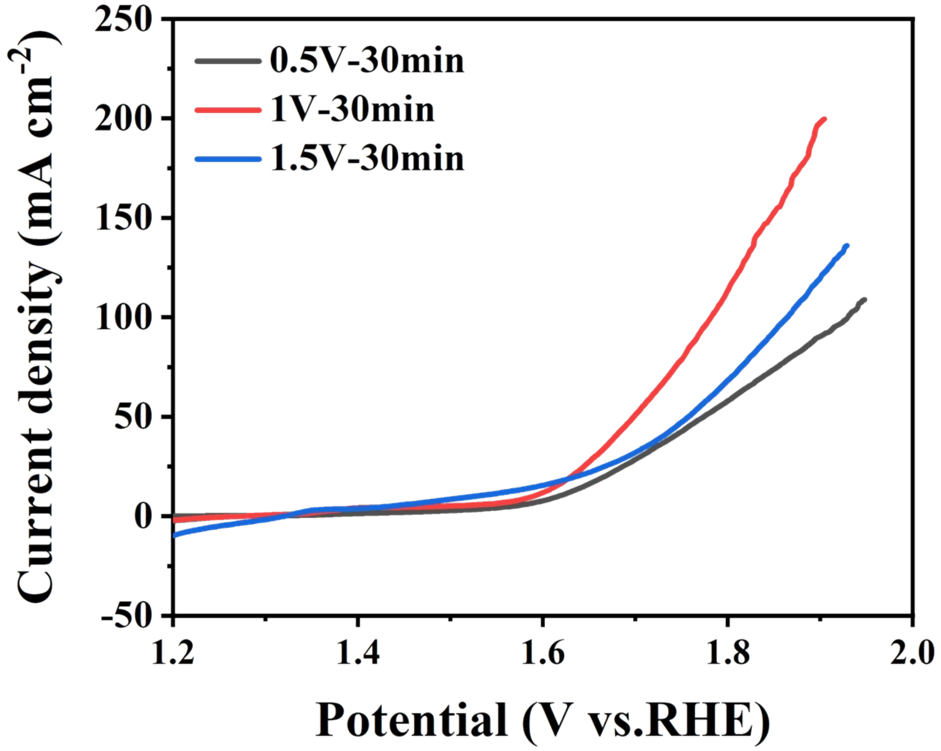

Fig. S2 LSV curves of de-NCM94 after different electrochemical de-lithiation voltages.

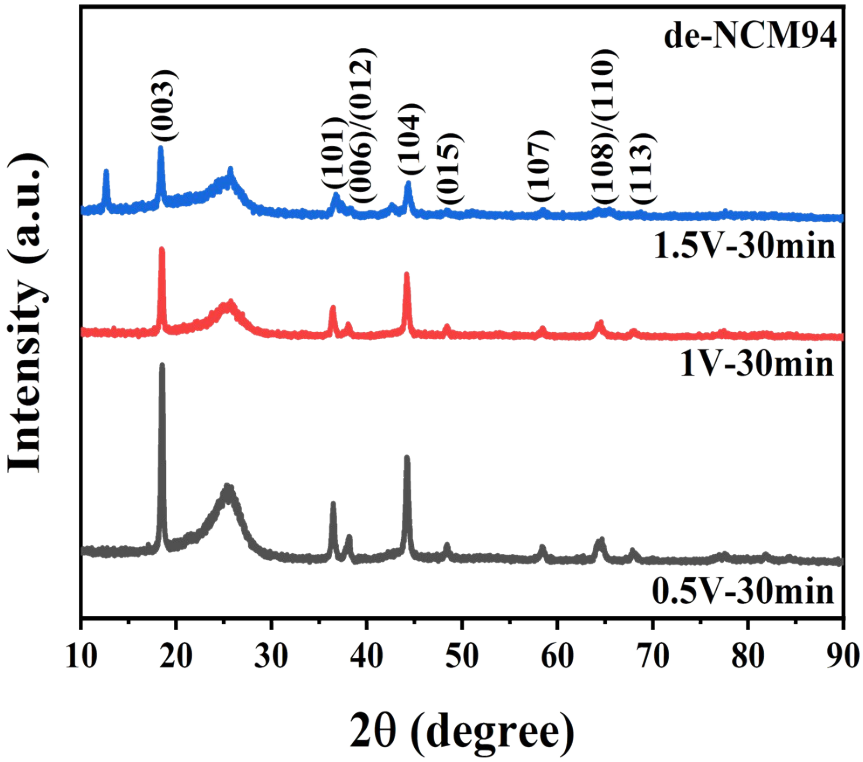

Fig. S3 XRD patterns of de-NCM94 after different electrochemical de-lithiation voltages.

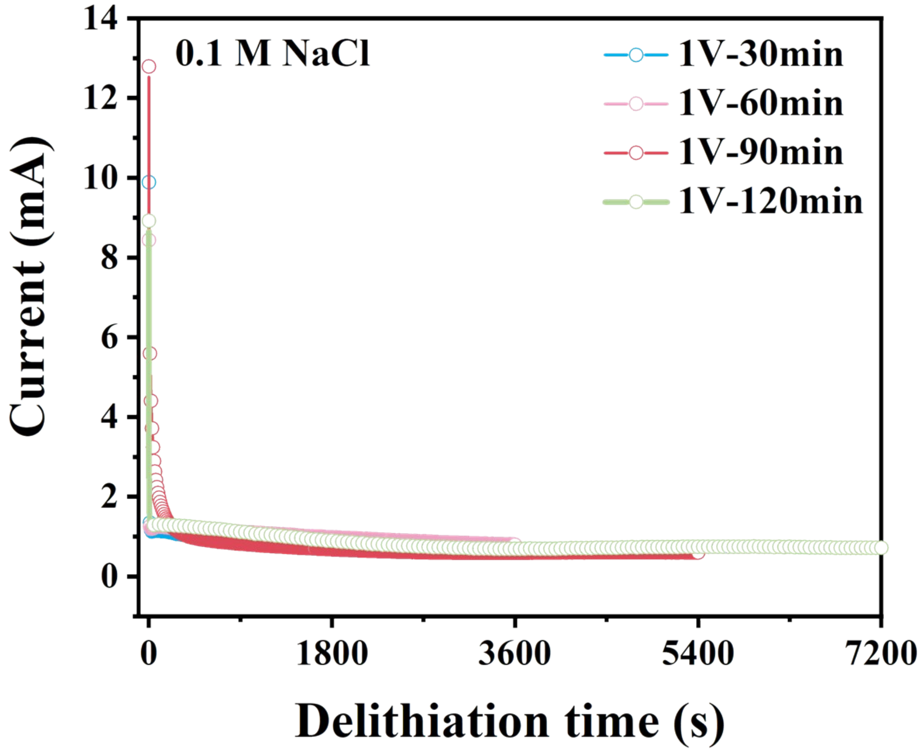

Fig. S4 i-t curves of de-NCM94 de-lithiated at 1 V for different times (30 min, 60 min, 90 min and 120 min).

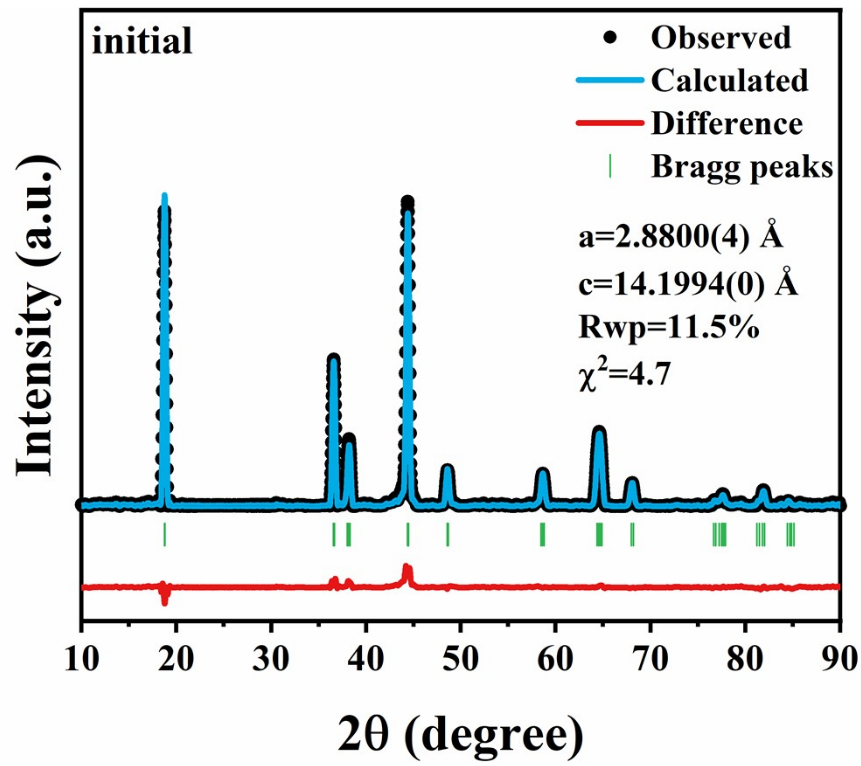

Fig. S5 Rietveld refinement results of NCM94-initial.

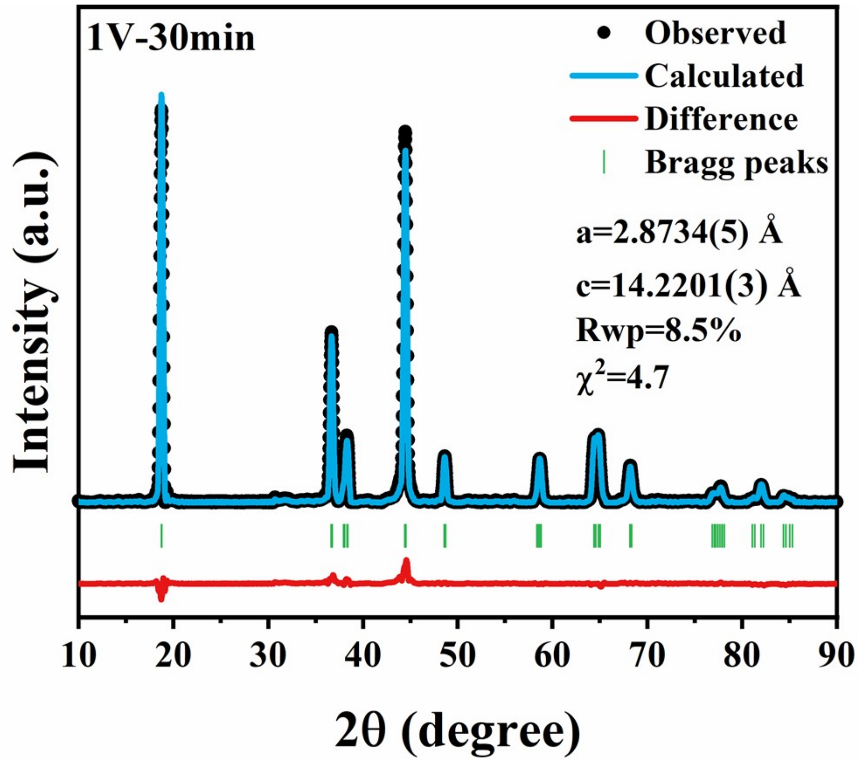

Fig. S6 Rietveld refinement results of NCM94-1V-30min.

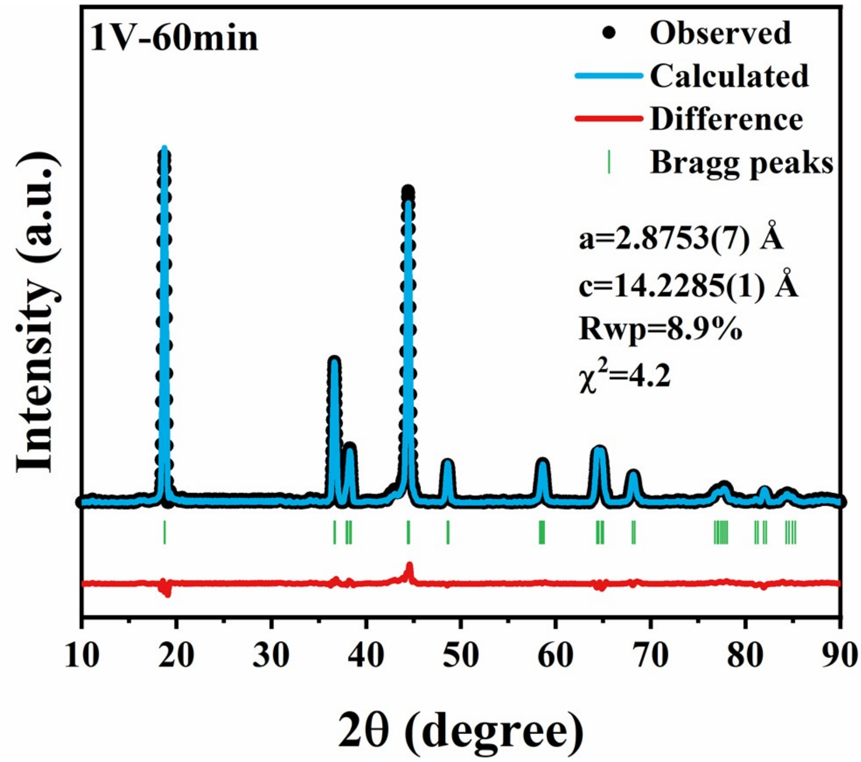

Fig. S7 Rietveld refinement results of NCM94-1V-60min.

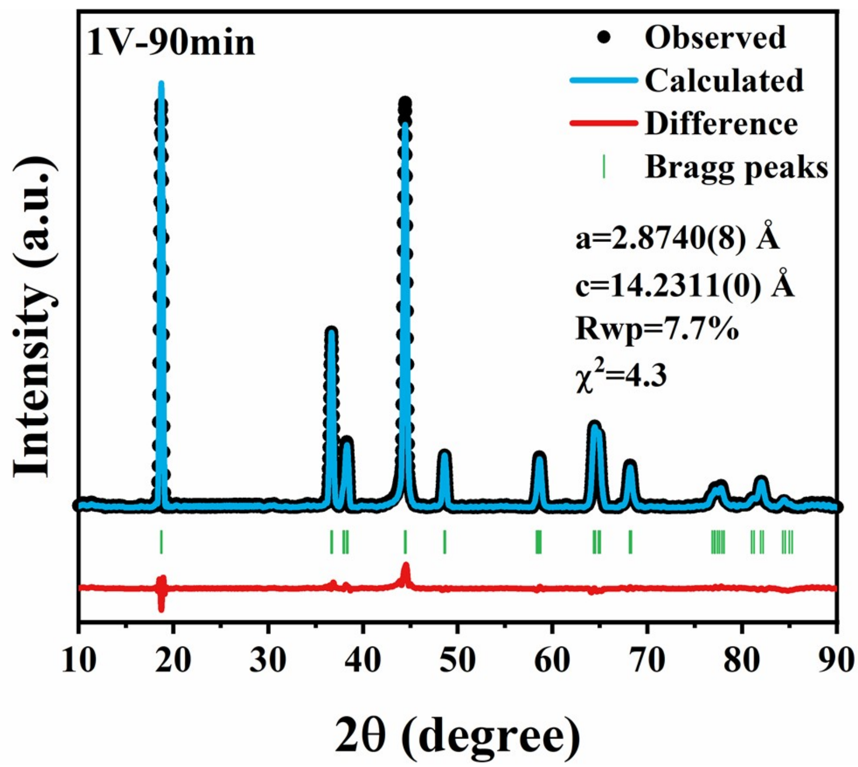

Fig. S8 Rietveld refinement results of NCM94-1V-90min.

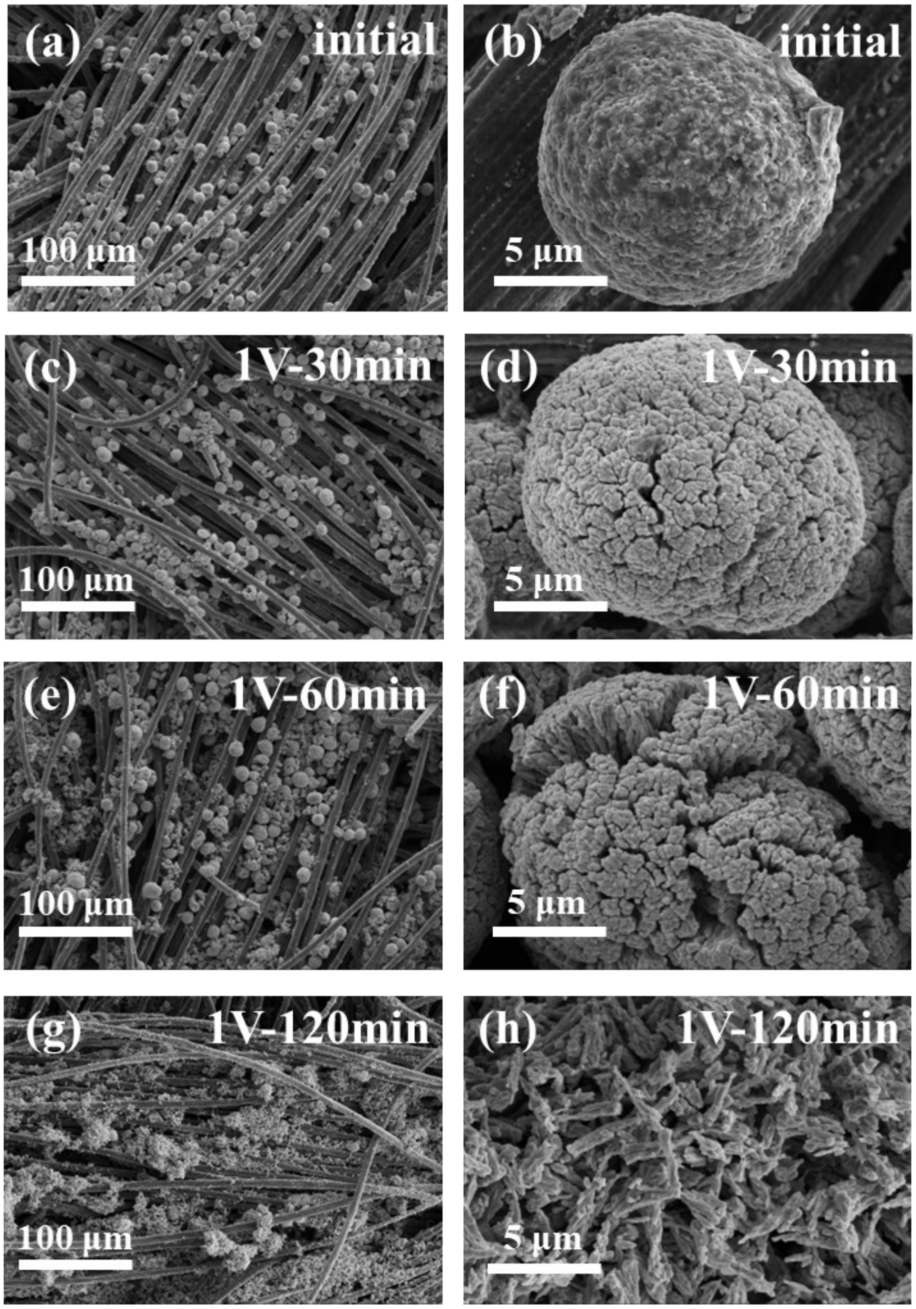

Fig. S9 SEM images of de-NCM94 at different magnifications after varied electrochemical de-lithiation time.

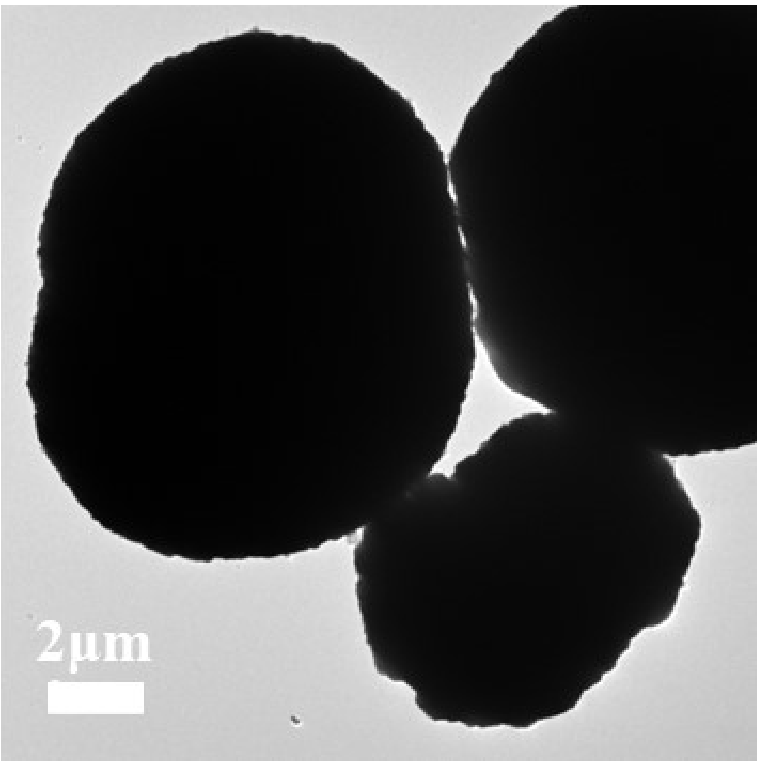

Fig. S10 TEM image of initial NCM94.

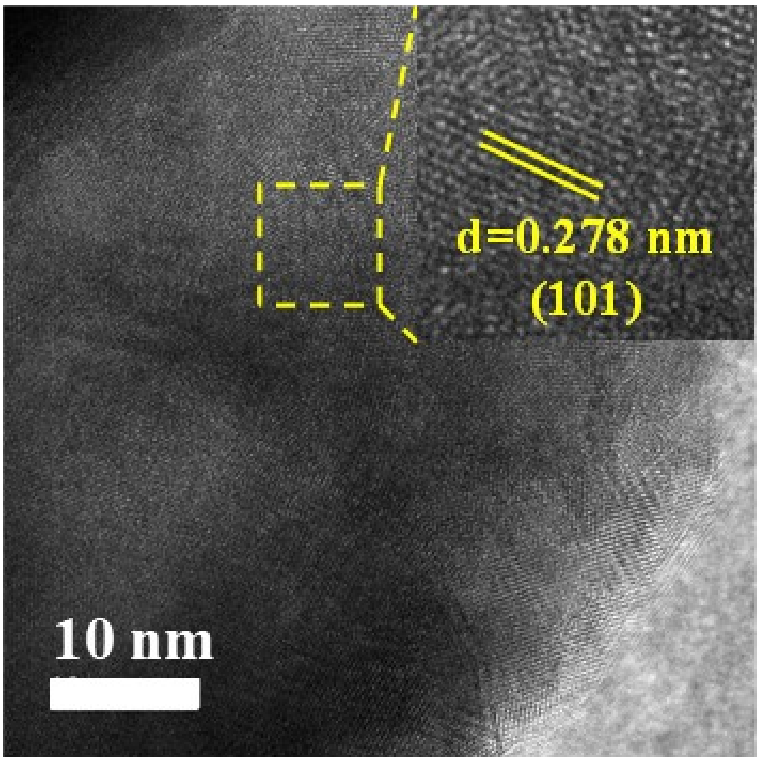

Fig. S11 HRTEM image of initial NCM94.

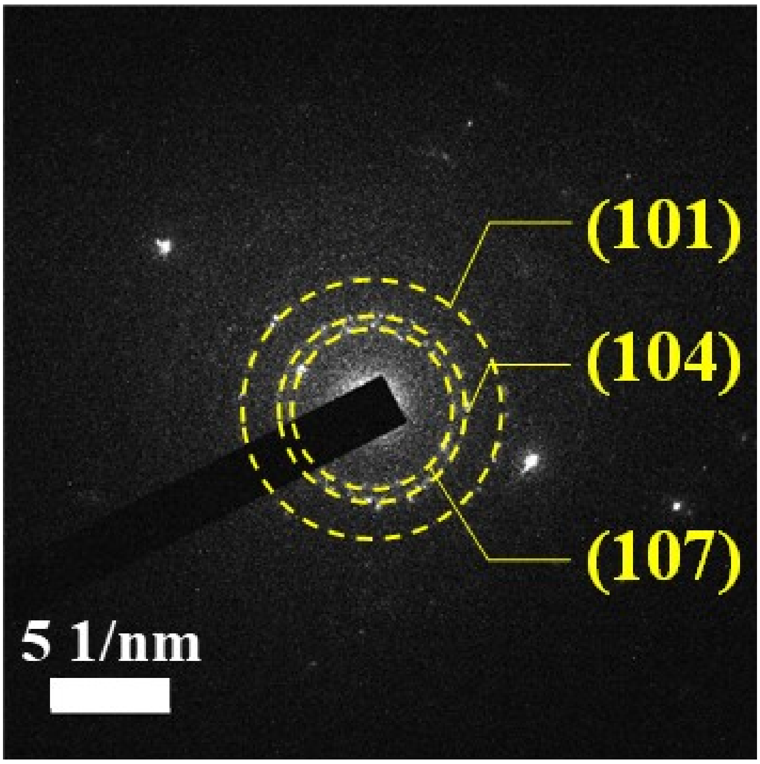

Fig. S12 SAED image of initial NCM94.

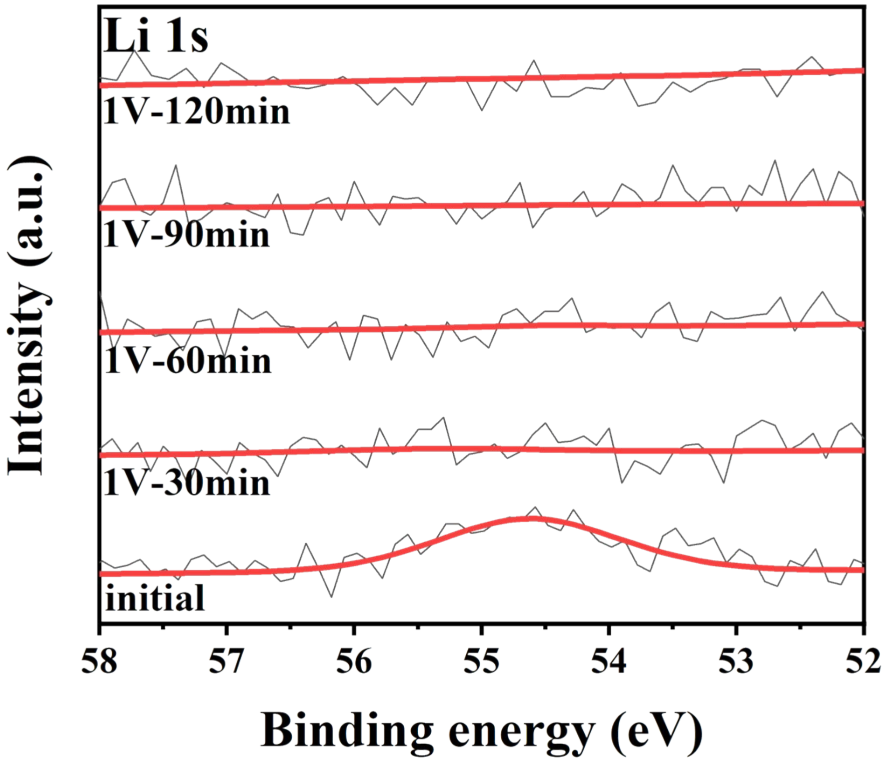

Fig. S13 Li 1s XPS spectra of de-NCM94 after varied electrochemical de-lithiation time.

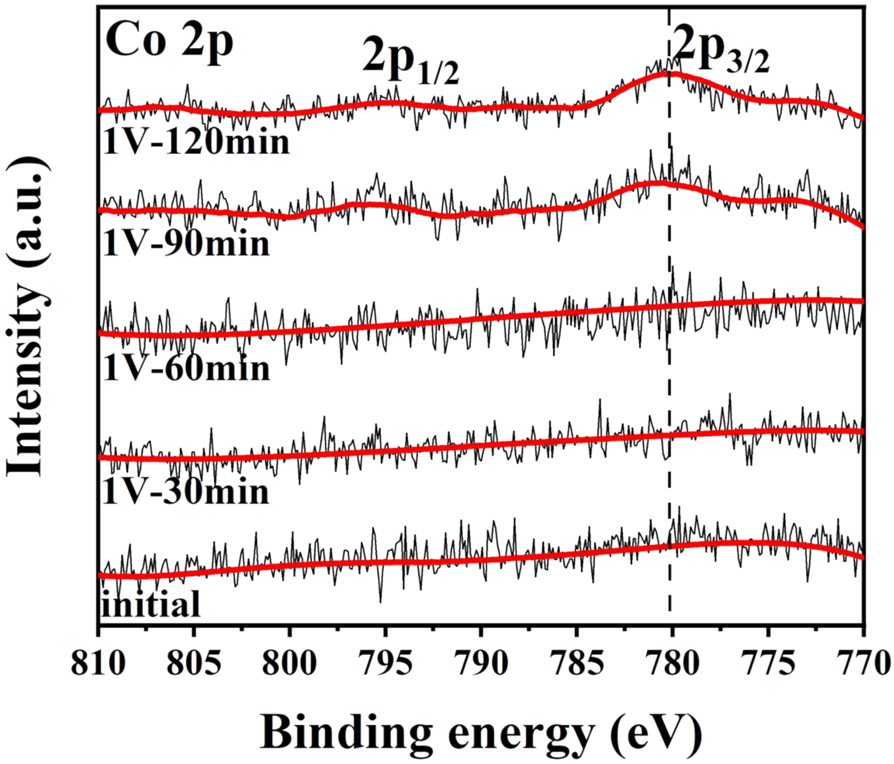

Fig. S14 Co 2p XPS spectra of de-NCM94 after varied electrochemical de-lithiation time.

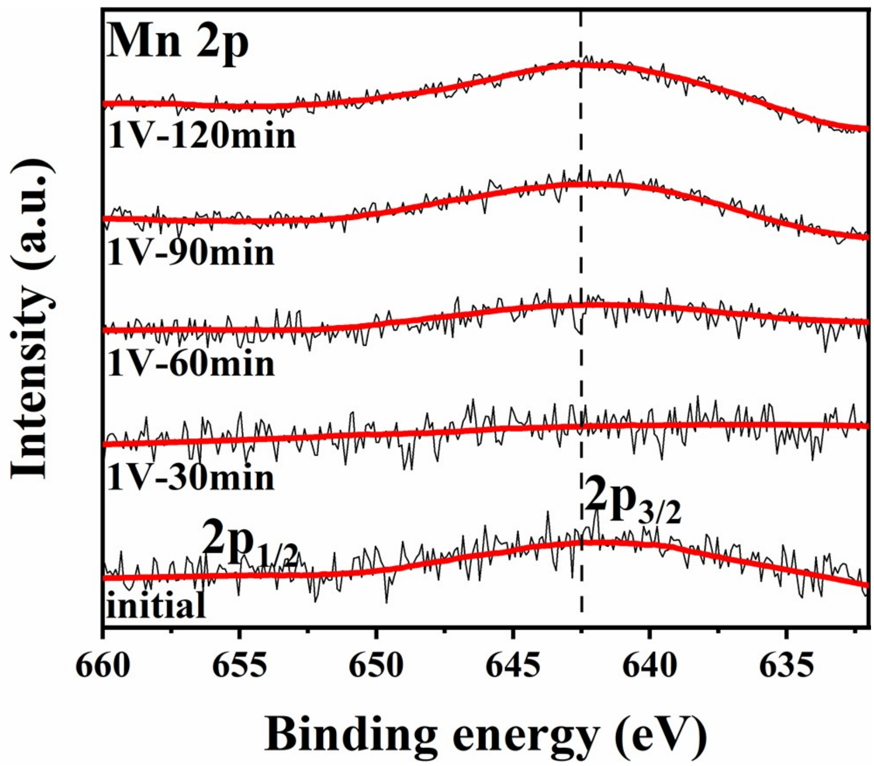

Fig. S15 Mn 2p XPS spectra of de-NCM94 after varied electrochemical de-lithiation time.

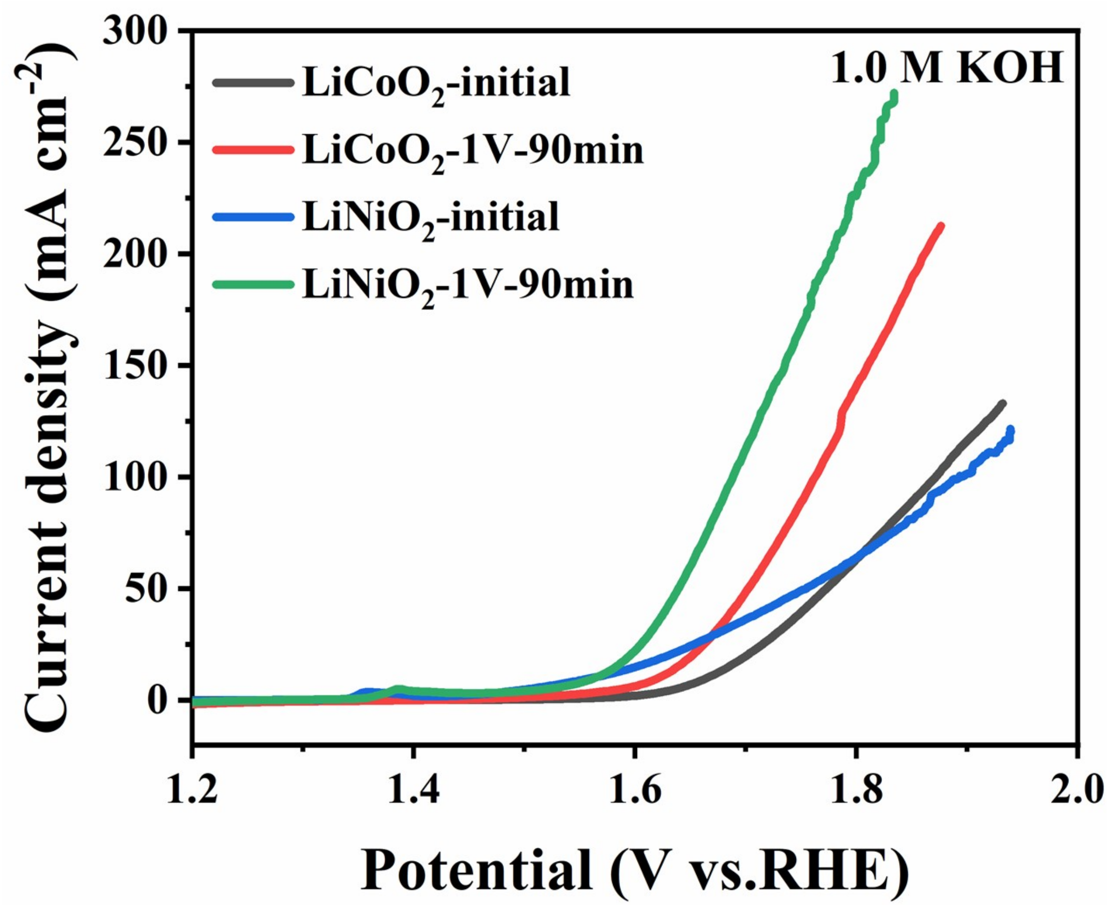

Fig. S16 LSV curves of $LiCoO_2$ and $LiNiO_2$ before and after electrochemical de-lithiation.

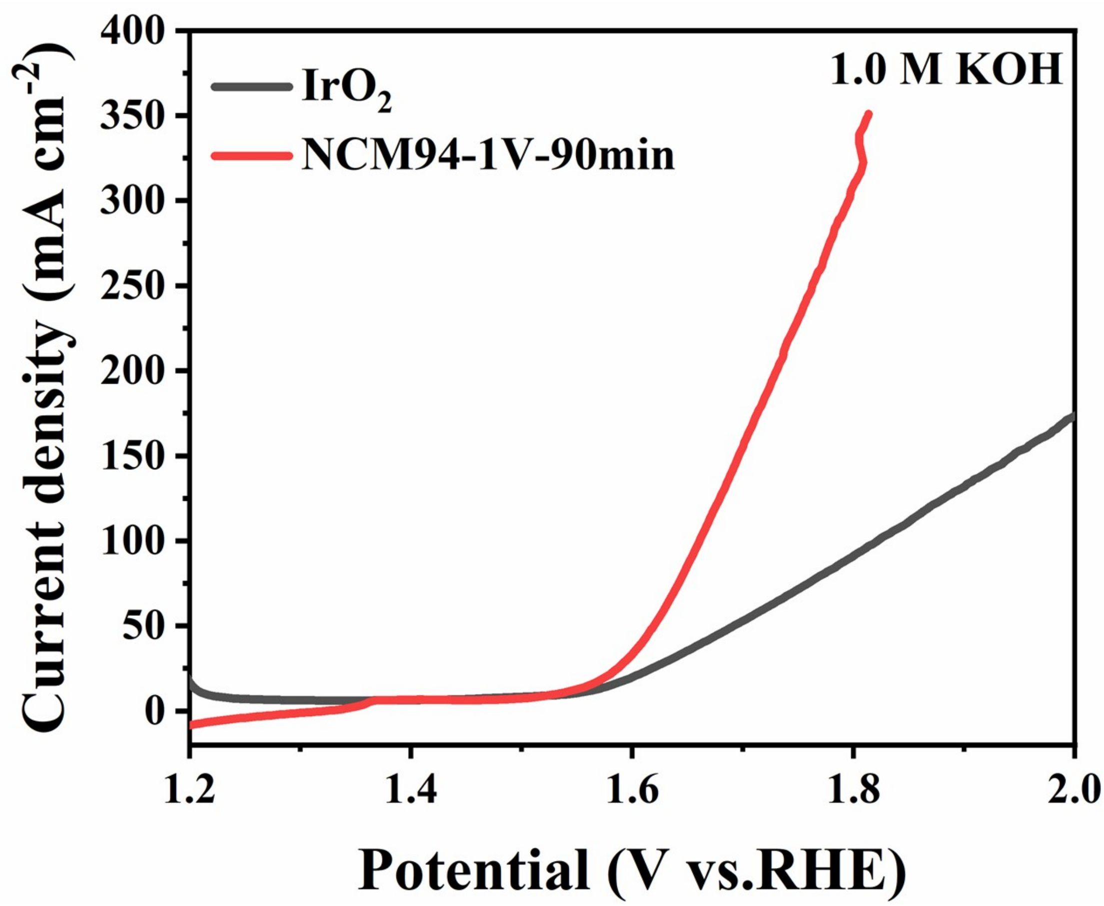

Fig. S17 LSV curves of NCM94-1V-90min and commercial IrO_2.

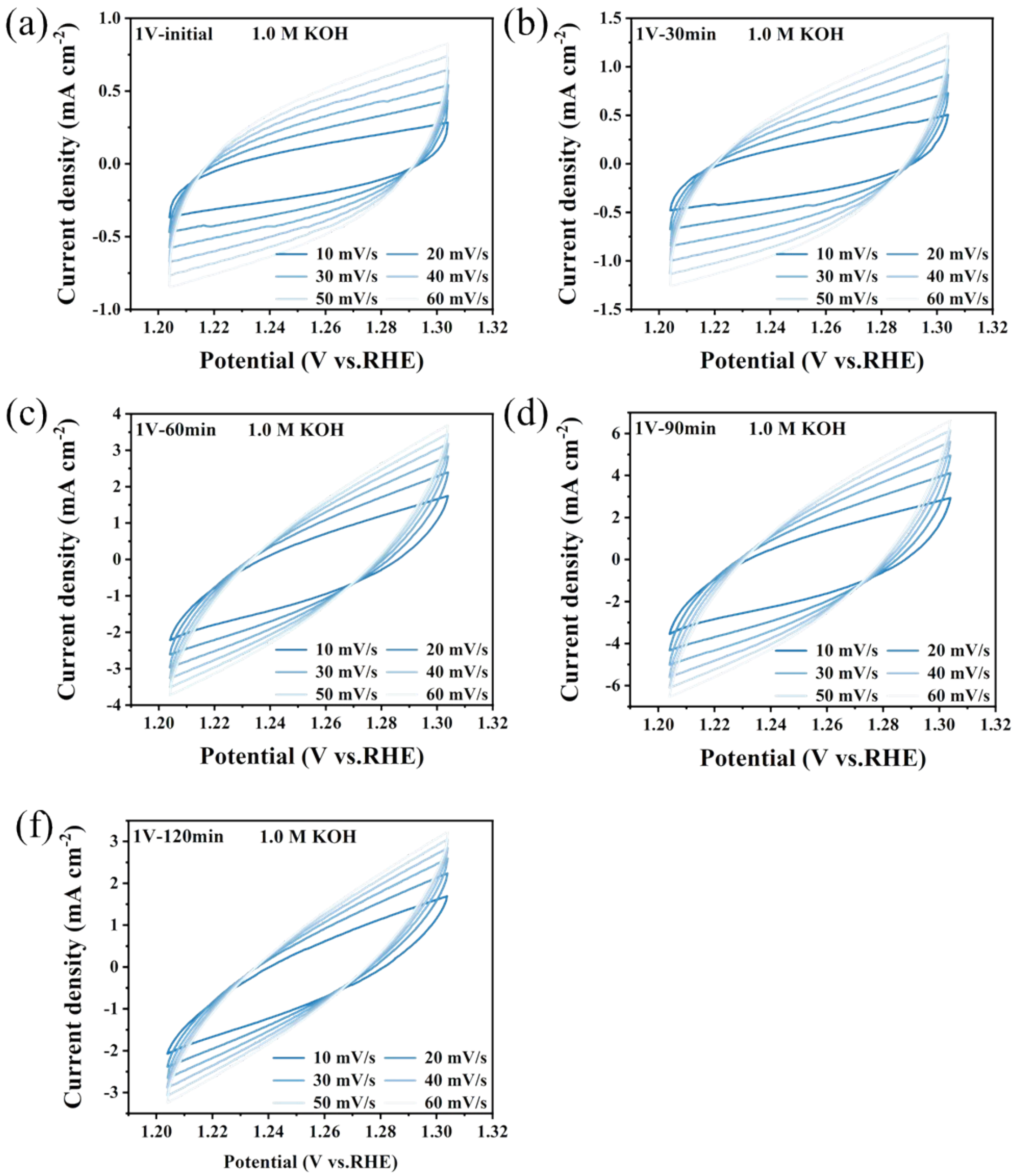

Fig. S18 CV curves of de-NCM94 after varied electrochemical de-lithiation times.

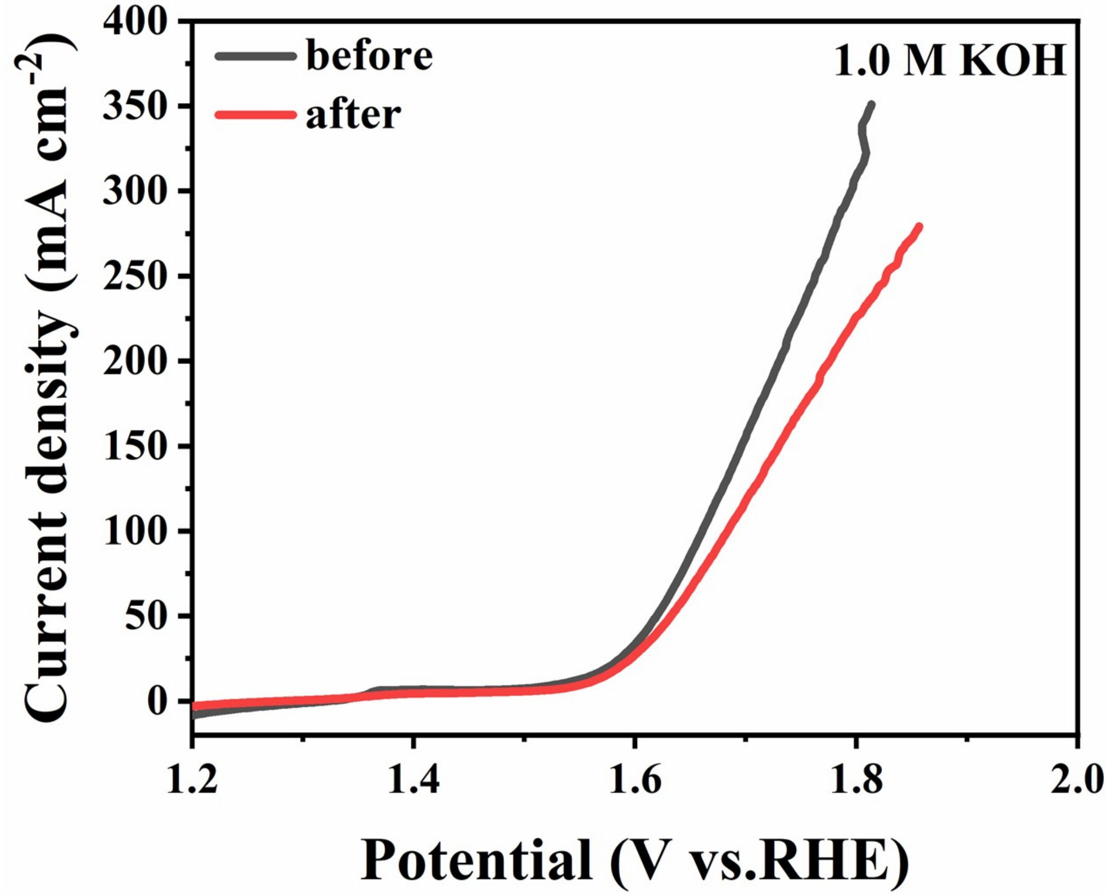

Fig. S19 LSV curves of NCM94-1V-90min before and after stability testing of 300 h.

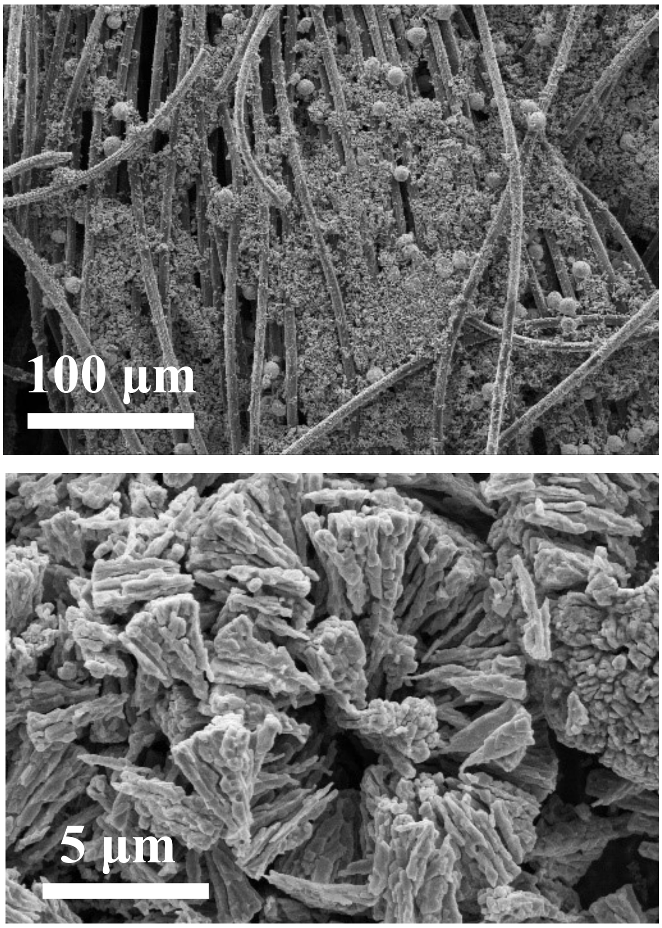

Fig. S20 SEM images of NCM94-1V-90min after stability testing.

Supplementary Tables

Table 1. Rietveld refinement results of de-NCM after varied de-lithiation time.

samples	a=b (Å)	c (Å)
Initial	2.8800(4)	14.1994(0)
1V-30min	2.8774(5)	14.2201(3)
1V-60min	2.8753(7)	14.2285(1)
1V-90min	2.8740(8)	14.2311(0)
1V-120min	-	-

Table 2. ICP-MS result of electrolyte after 90 min of electrochemical de-lithiation under 1 V.

	Li	Ni	Co	Mn	de-lithiation degree
concentration	2.31 μg/mL	24.15 ng/mL	0.18 ng/mL	＜0.1 ng/mL	38%

Table 3. Comparison of overpotential of recycling spent LIBs cathode materials for OER electrocatalysts at 10 mA cm^{-2}.

spent lithium-ion battery materials	OER catalysts	recycling method	overpotential (at 10 mA cm^{-2})	reference
$LiNi_{0.94}Co_{0.05}Mn_{0.01}O_2$	**de-NCM94**	**electrochemical de-lithium**	**270 mV**	**This work**
$LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$	NiCoMnB	boriding process	271 mV	1
$LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$	NiCoMnB	boriding process	263 mV	1
$LiFePO_4$	Ni-$LiFePO_4$	electrochemical transformation	285 mV	2
Spent LIBs	$MnCo_2O_4$	chemical leaching, calcination	400 mV	3
$LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$	NiMnCo-AC	thermal radiation	340 mV	4
$LiCoO_2$	CP4($LiCoO_2$)	calcination	550 mV	5
$LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$	$Ni_{0.5}Mn_{0.3}Co_{0.2}(OH)_2$	mechanochemical activation process	280 mV	6
$LiCoO_2$	CoN-graphene	calcination, hydrothermal	280 mV	7

References

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