Supporting Information

# In situ induced surface reconstruction of single crystal lithium-ion cathode towards effective interface compatibility

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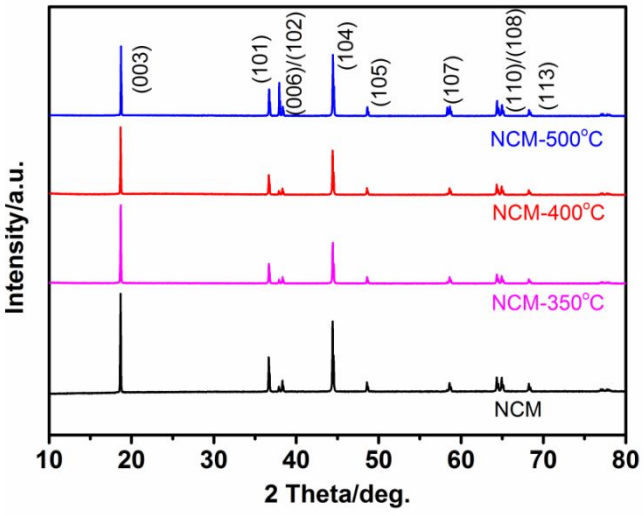


**Figure S1.** XRD patterns of NCM, NCM-350°C, NCM-400°C and NCM-500°C.

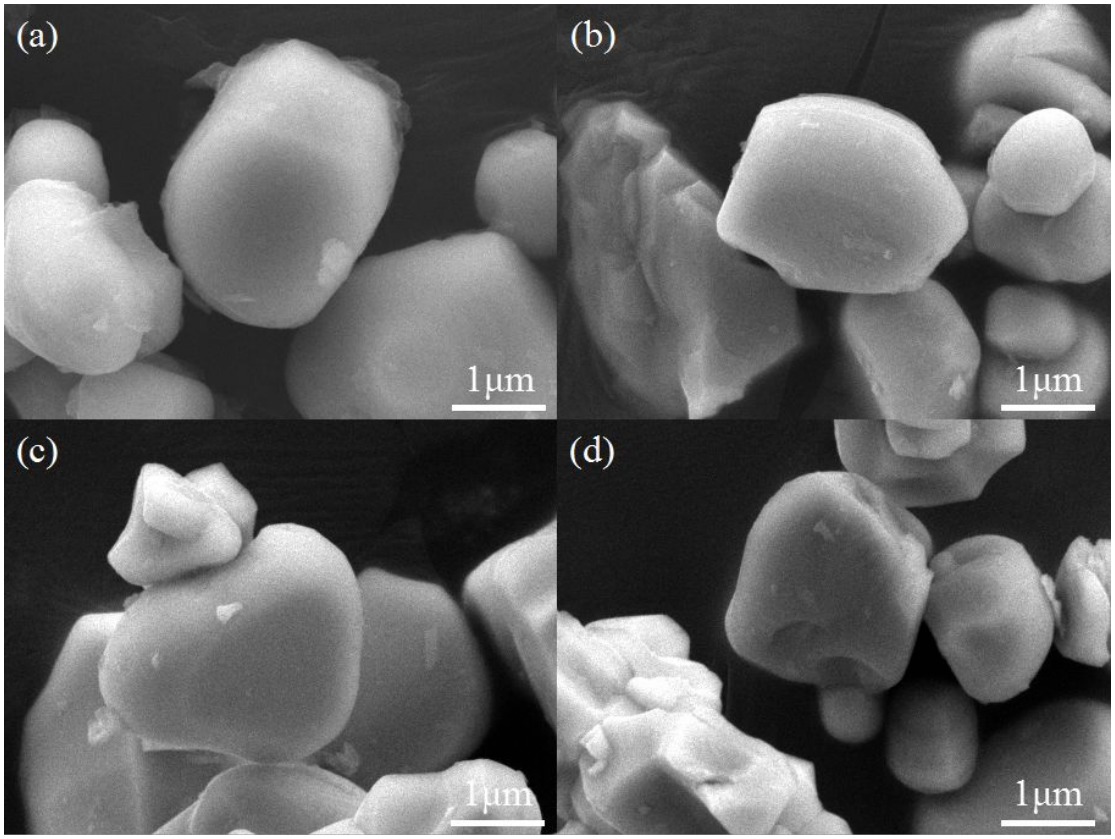


**Figure S2.** SEM image of (a) NCM, (b) NCM-350$^{o}$C, (c) NCM-400$^{o}$C and (d) NCM-500$^{o}$C powder.

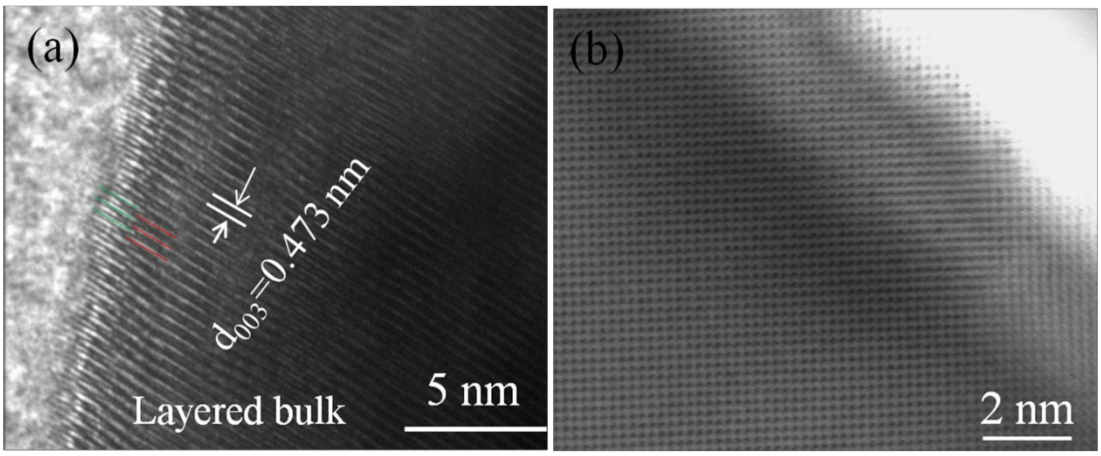


**Figure S3.** (a) HRTEM and (b) STEM image of NCM.

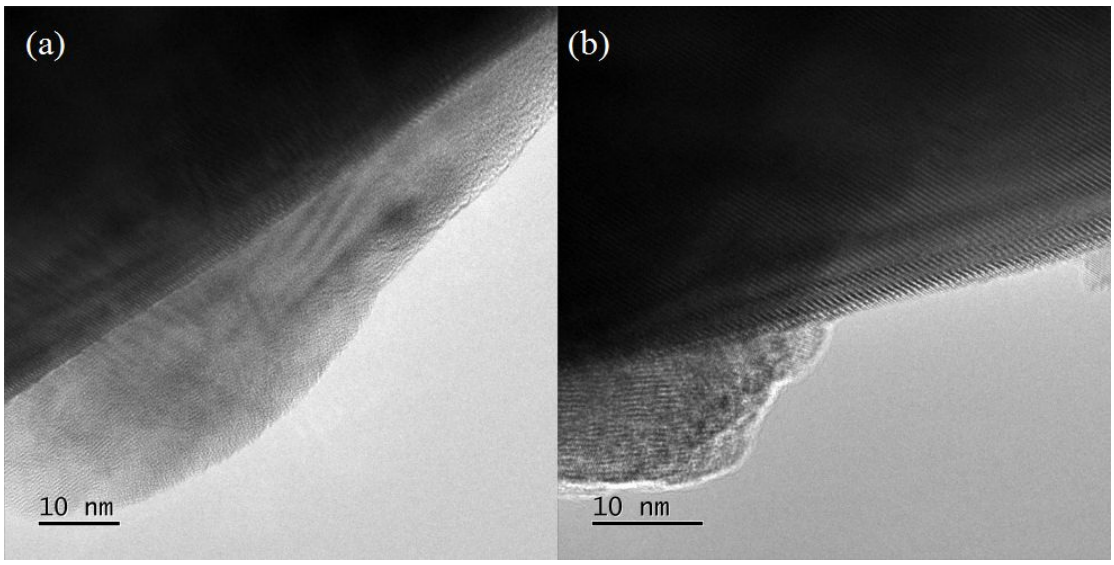


**Figure S4.** TEM images of NCM cathode at different locations.

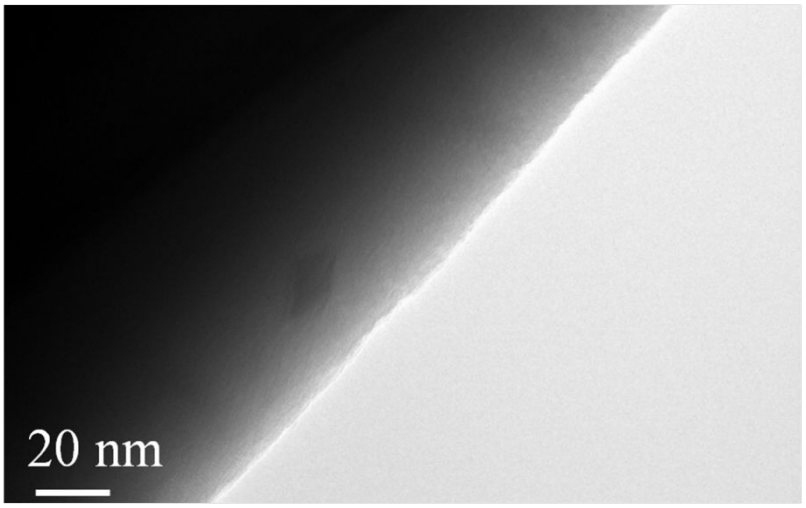


**Figure S5.** TEM image of NCM-400$^{o}$C cathode at large area region with small magnification.

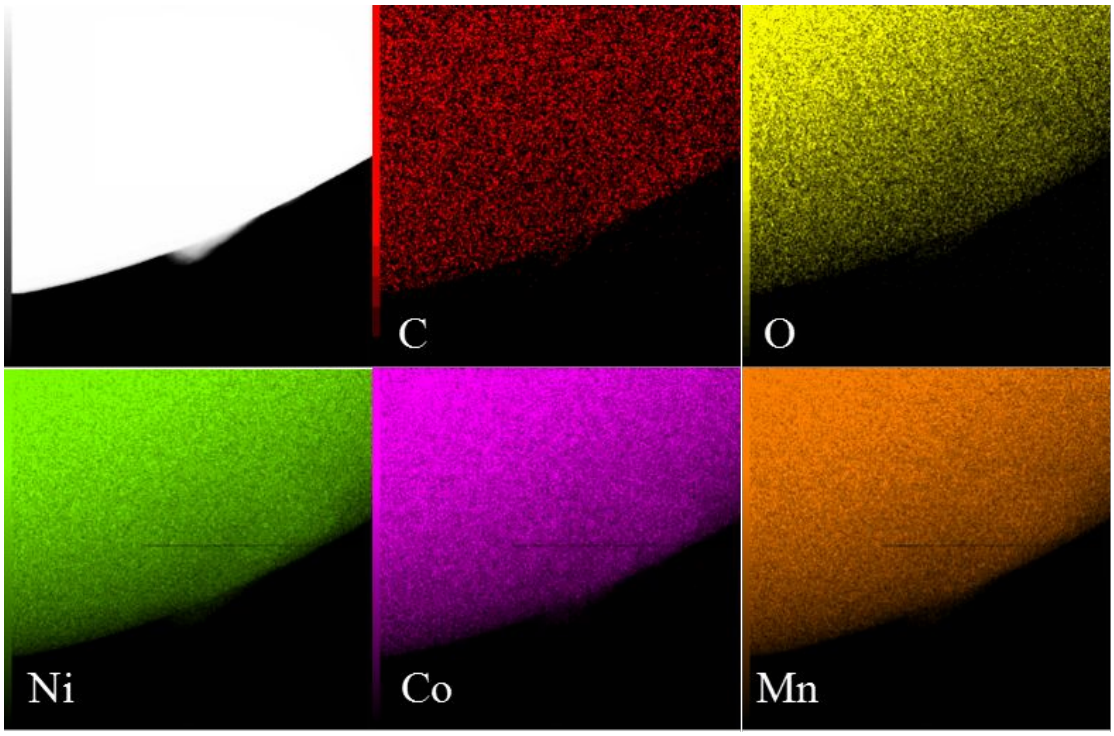


**Figure S6.** EDS elemental mapping of a NCM cathode particle.

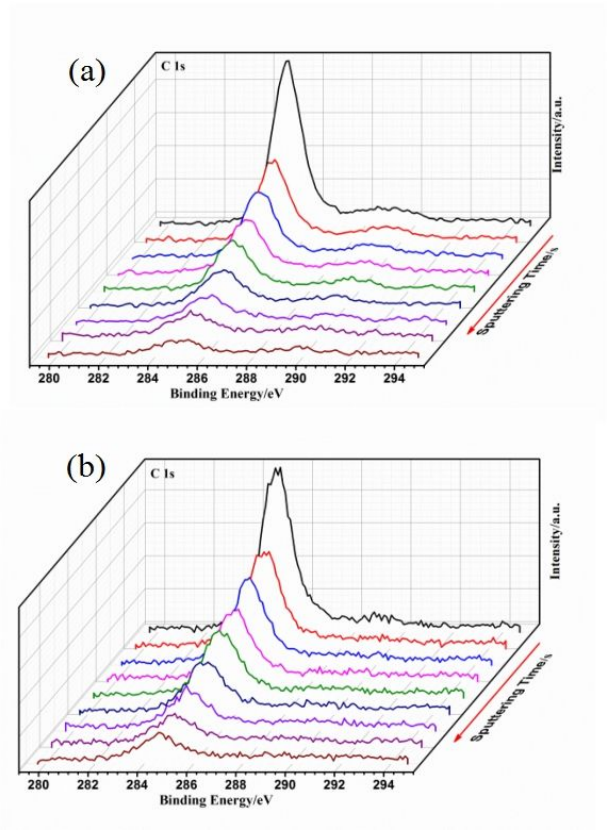


**Figure S7.** XPS spectra of C 1s for (a) NCM and (b) NCM-400$^{o}$C with different sputtering time (0, 5, 10, 20, 30, 60, 120, 180, 240 s).

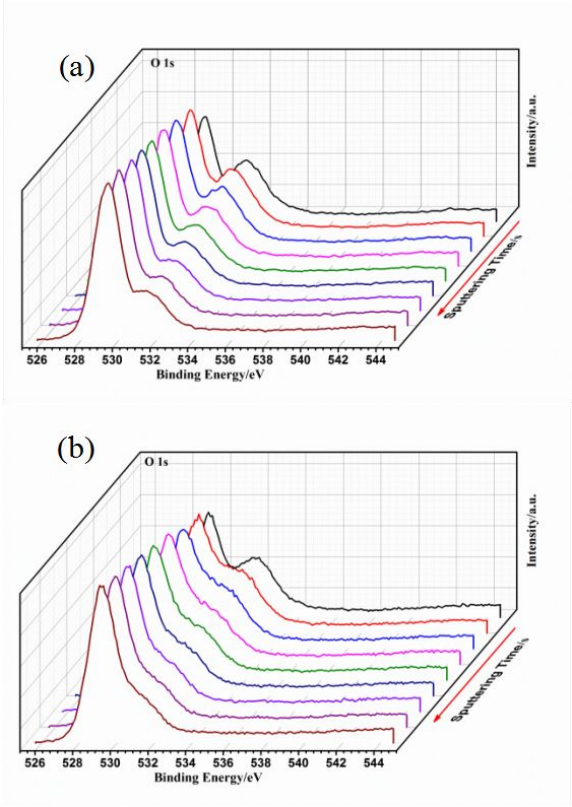


**Figure S8.** XPS spectra of O 1s for (a) NCM and (b) NCM-400$^{o}$C with different sputtering time (0, 5, 10, 20, 30, 60, 120, 180, 240 s).

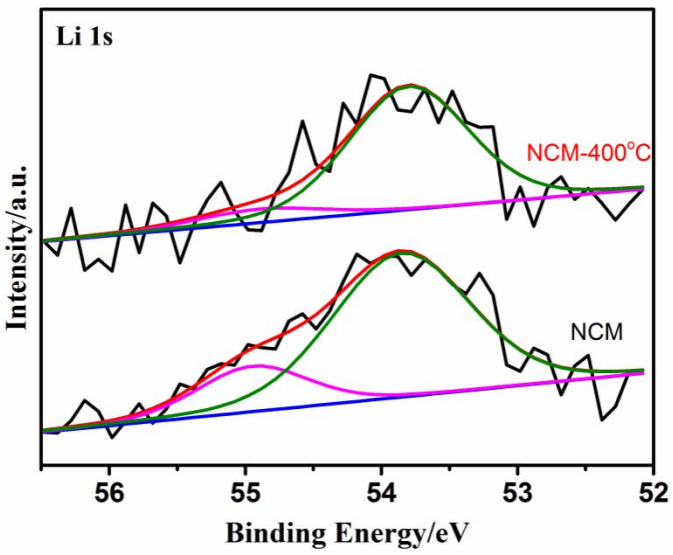


**Figure S9.** XPS spectra of Li 1s for NCM and NCM-400°C surface.

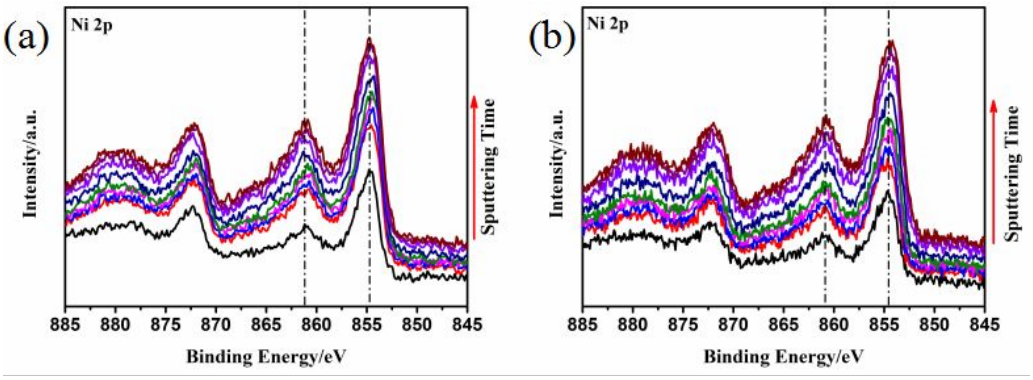


**Figure S10.** XPS spectra of Ni 2p for (a) NCM and (b) NCM-400$^{o}$C with different sputtering time.

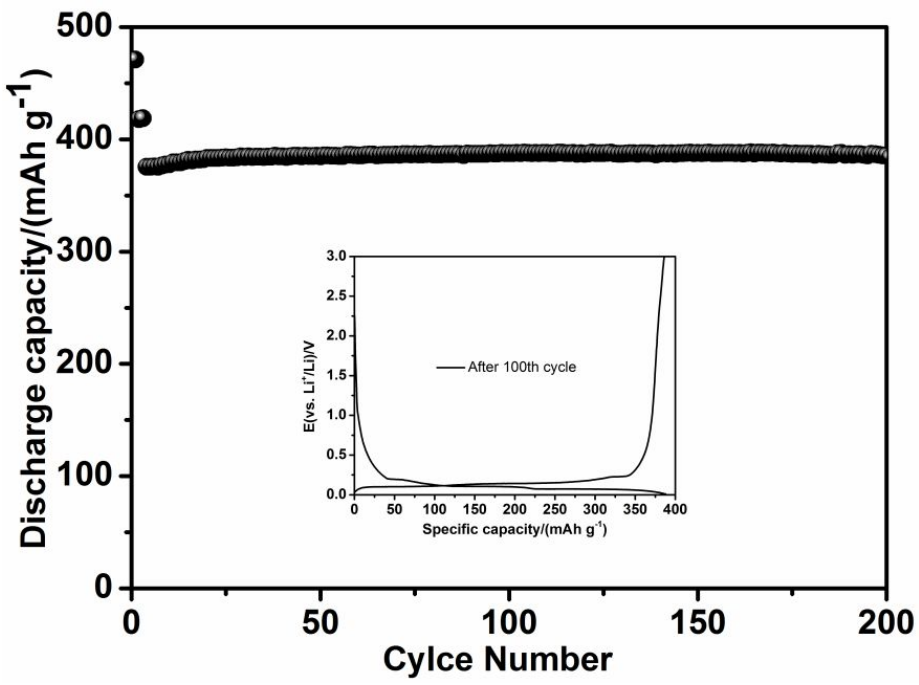


**Figure S11.** Cycling performance of graphite at 0.5 C and 25°C within a voltage range of 0.01-3.0 V in a half cell against lithium anodes.

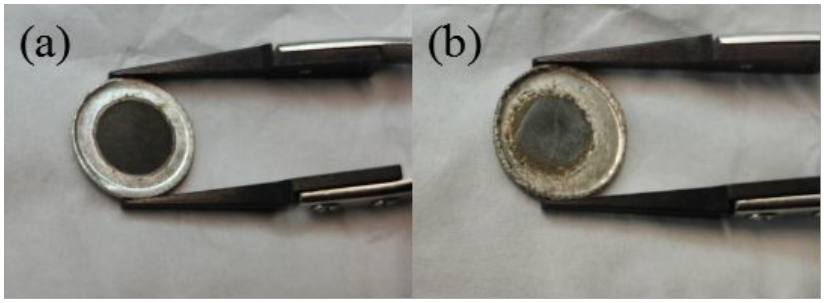


**Figure S12**. Digital images of lithium harvested from the disassembled cycled cells after 500 cycles at 5 C for (a) NCM and (b) NCM-400$^{o}$C.

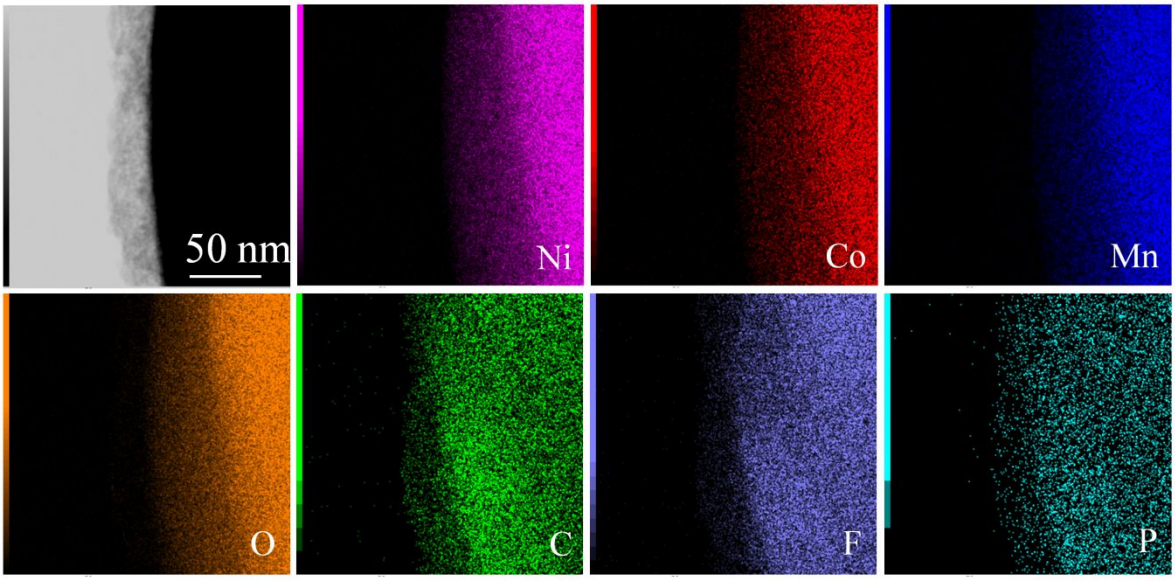


**Figure S13.** STEM-EDX elemental mapping of cycled NCM cathode particle.

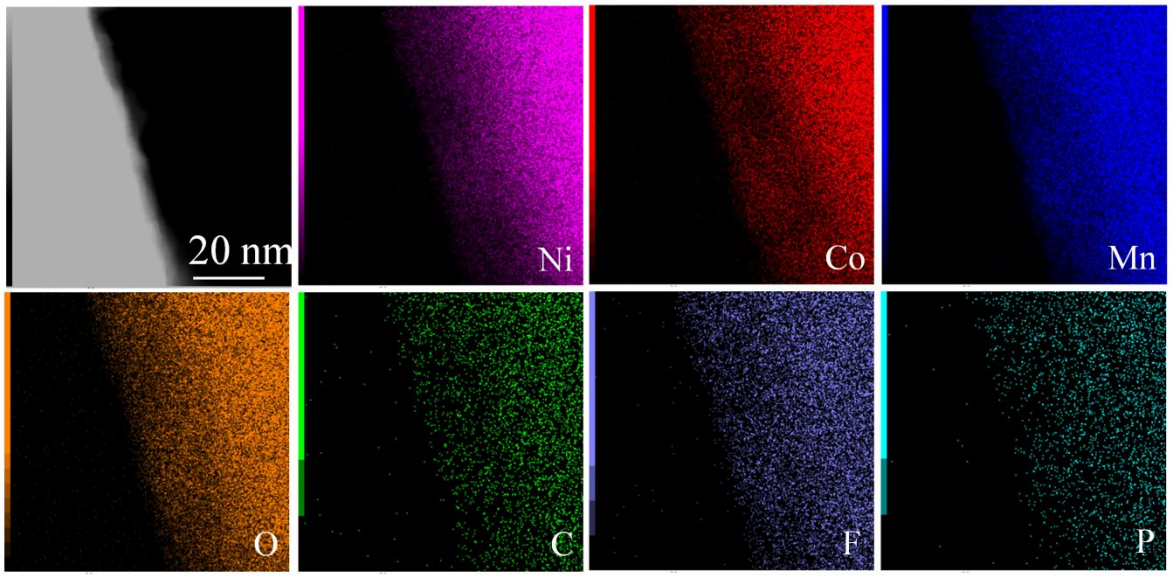


**Figure S14.** STEM-EDX elemental mapping of cycled NCM-400$^{o}$C cathode particle.

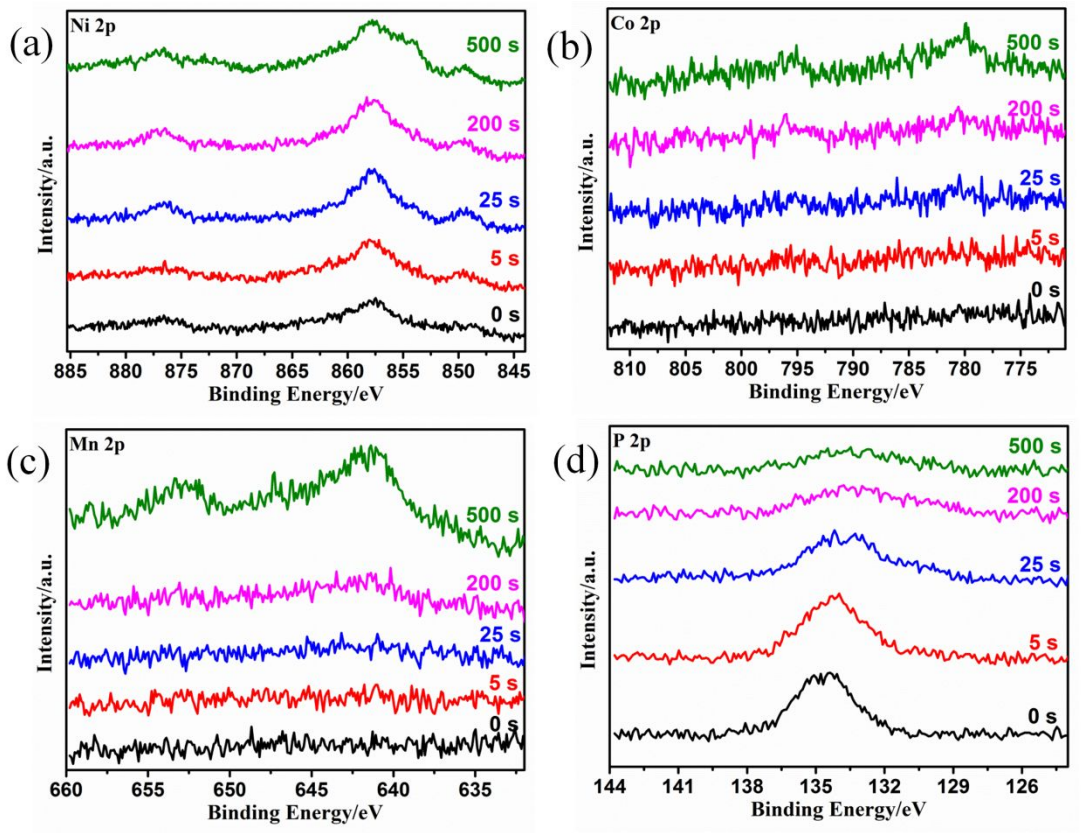


**Figure S15.** XPS spectra in argon atmosphere of (a) Ni 2p, (b) Co 2p, (c) Mn 2p and (d) P 2p for NCM after cycling at 5 C for 500 cylces.

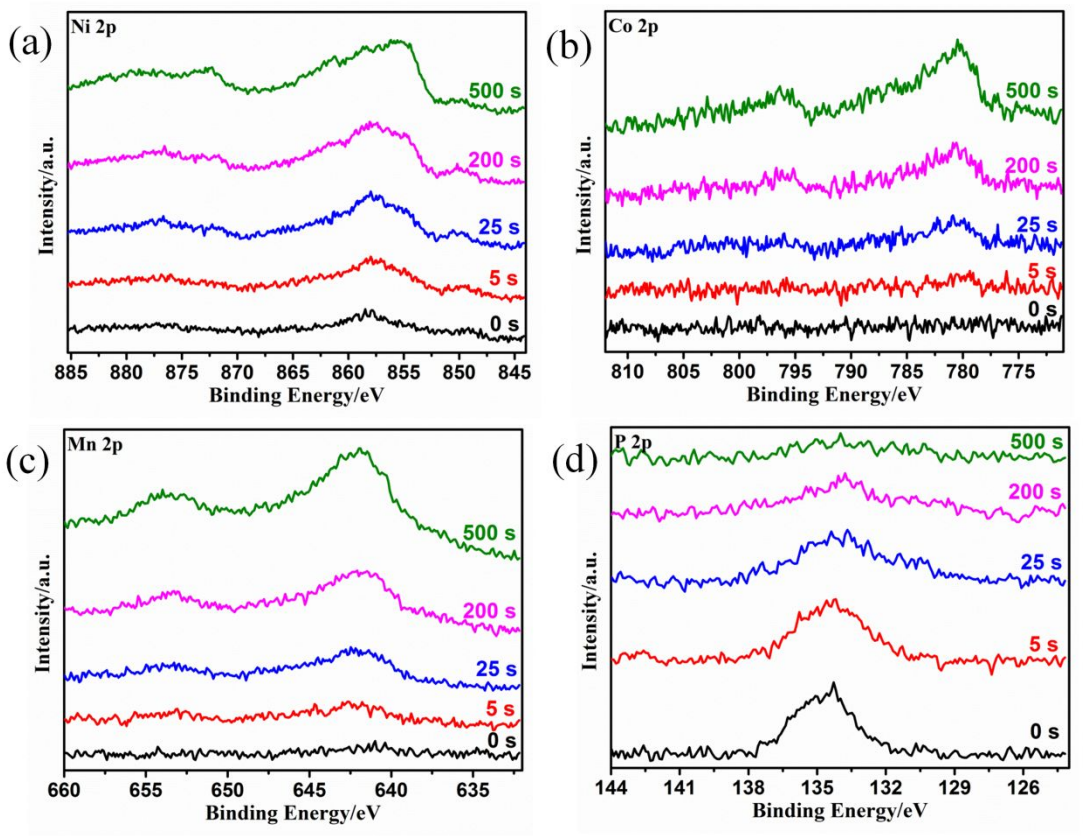


**Figure S16.** XPS spectra in argon atmosphere of (a) Ni 2p, (b) Co 2p, (c) Mn 2p and (d) P 2p for NCM-400$^{o}$C after cycling at 5 C for 500 cylces.

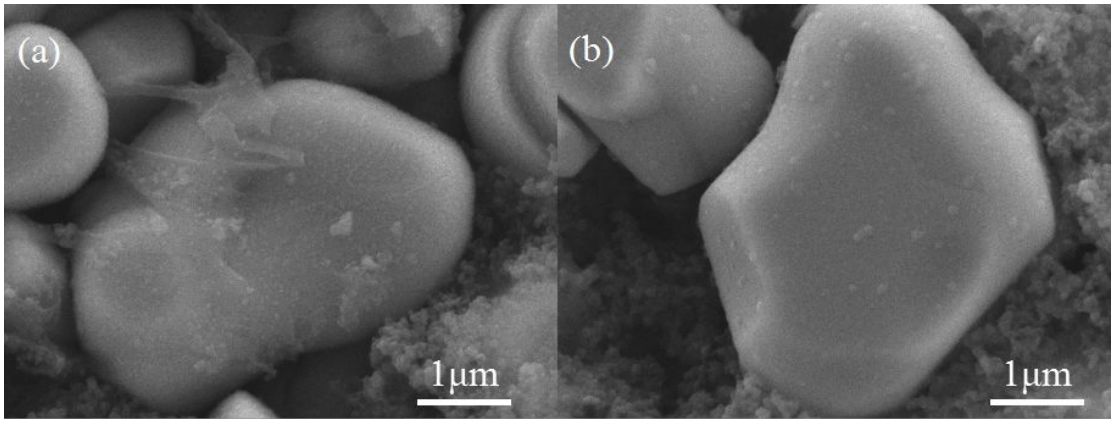


**Figure S17**. SEM image of cycled (a) NCM, and (b) NCM-400$^{o}$C.