Electronic Supplementary Information

EXPERIMENTAL SECTION

Synthesis: The pristine single-crystal $LiNi_{0.83}Co_{0.12}Mn_{0.05}O_2$ cathode materials (SC-P) were prepared by sintering the mixture of $LiOH·H_2O$ (Ganfeng Lithium Co Ltd) and $Ni_{0.83}Co_{0.12}Mn_{0.05}(OH)_2$ (Ronbay New Energy Co Ltd) at 875 ℃ (12h) with the molar ratio of 1.05:1. The powder of SC-P was dispersed in the alcoholic solution of stoichiometric $In(NO_3)_3·3H_2O$ (In:Me=0.004, Me=Ni+Co+Mn), then the mixture was dried at 90℃ for 5h under vacuum. The In-modified cathode (SC-In) was synthesized by calcining powder mentioned above at the temperature of 700℃ for 7 hours.

Preparation of Electrodes: The electrodes of SC-P and SC-In were prepared by the same procedure. The acetylene black (SP), active material, and polyvinylidene fluoride (PVDF) were mixed with the mass ratio of 1:8:1. The 1.2mL N-methyl-2-pyrrolidone (NMP) was added to the 0.5g mentioned mixture to make up the emulsion. The emulsion was pasted onto the Al foil. The Al foil was then punched into several 14mm diameter discs. The metallic lithium with 100μm thickness was used as the anode electrode.

Cell Assembly and Testing: The electrolyte was 1 M $LiPF_6$ with EMC(32%), EC(32%), and DMC(32%), VC(2%) and DTD(1%). The CR2016-type coin half-cells were assembled in the nitrogen gas atmosphere glove (O_2 and H_2O < 0.01 ppm). Land Test System was used to test cyclic and rate performances (1 C = 180 mAh g^{-1}) in the range of 2.5-4.25 V or 4.5 V. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) are both tested at the Princeton VersaSTAT MC electrochemical workstation. CV was hold with scan rate at 0.1 $mV·s^{-1}$. EIS was tested from the frequency of 0.01 Hz to 100 kHz. with the amplitude of the AC signal of ±5 mV.

Instrumentation: Transmission Electron Microscopy (TEM, FEI Talos F200S) was applied to observe the phase structure of surface tested. The scanning electron microscope (SEM) was used to characterize the morphologies by JEOL JSM-7900F. X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha) was utilized to acquire the binding energy of elements. The information of oxygen vacancies was obtained electron paramagnetic resonance (EPR) is measured Bruker EMXplus-6/1. Gas evolution was evaluated by differential Electrochemical Mass Spectrometry (DEMS) (HPR-40, Hiden) via 0.75 mL/min He (99.999%). MCT-211(Shimadzu) is used to hold micro-compression testing. The loading speed is set as 20 (0.67mN/s) with holding time of 5 s. The test sample is prepared by dispersed in the alcohol to a hard alloy table.

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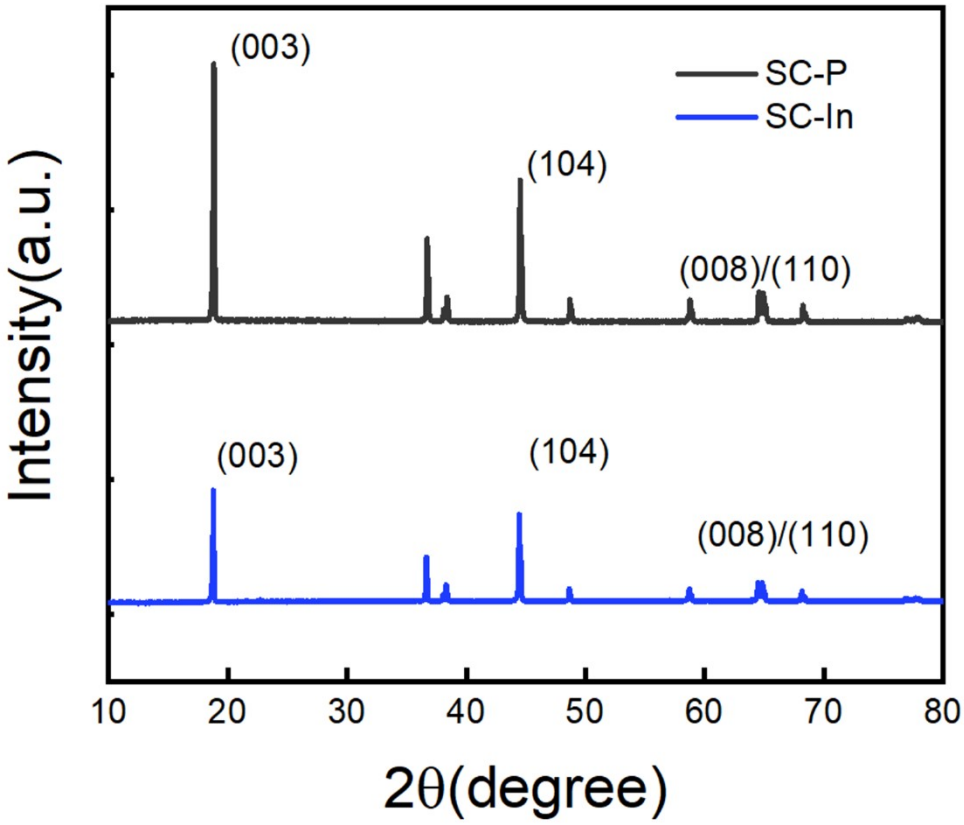

Fig.S1 The XRD patterns of SC-P and SC-In.

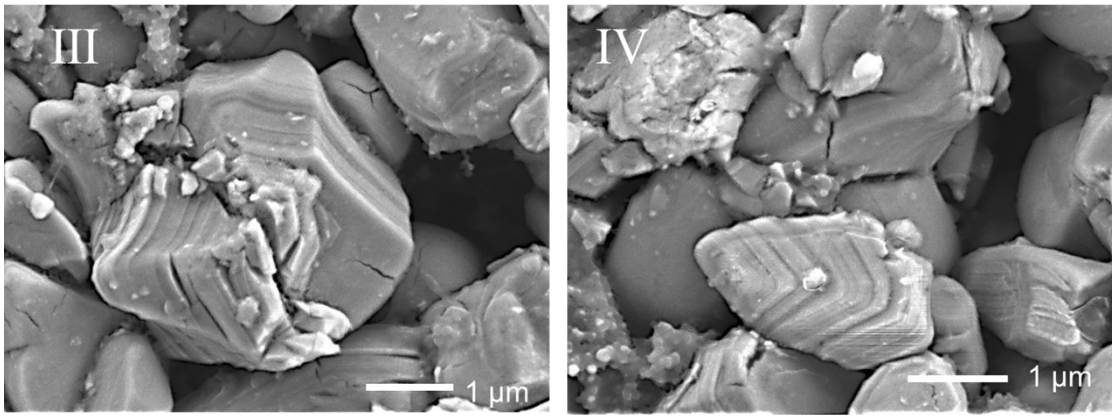

Fig.S2 The FESEM images with different cracks of SC-P.

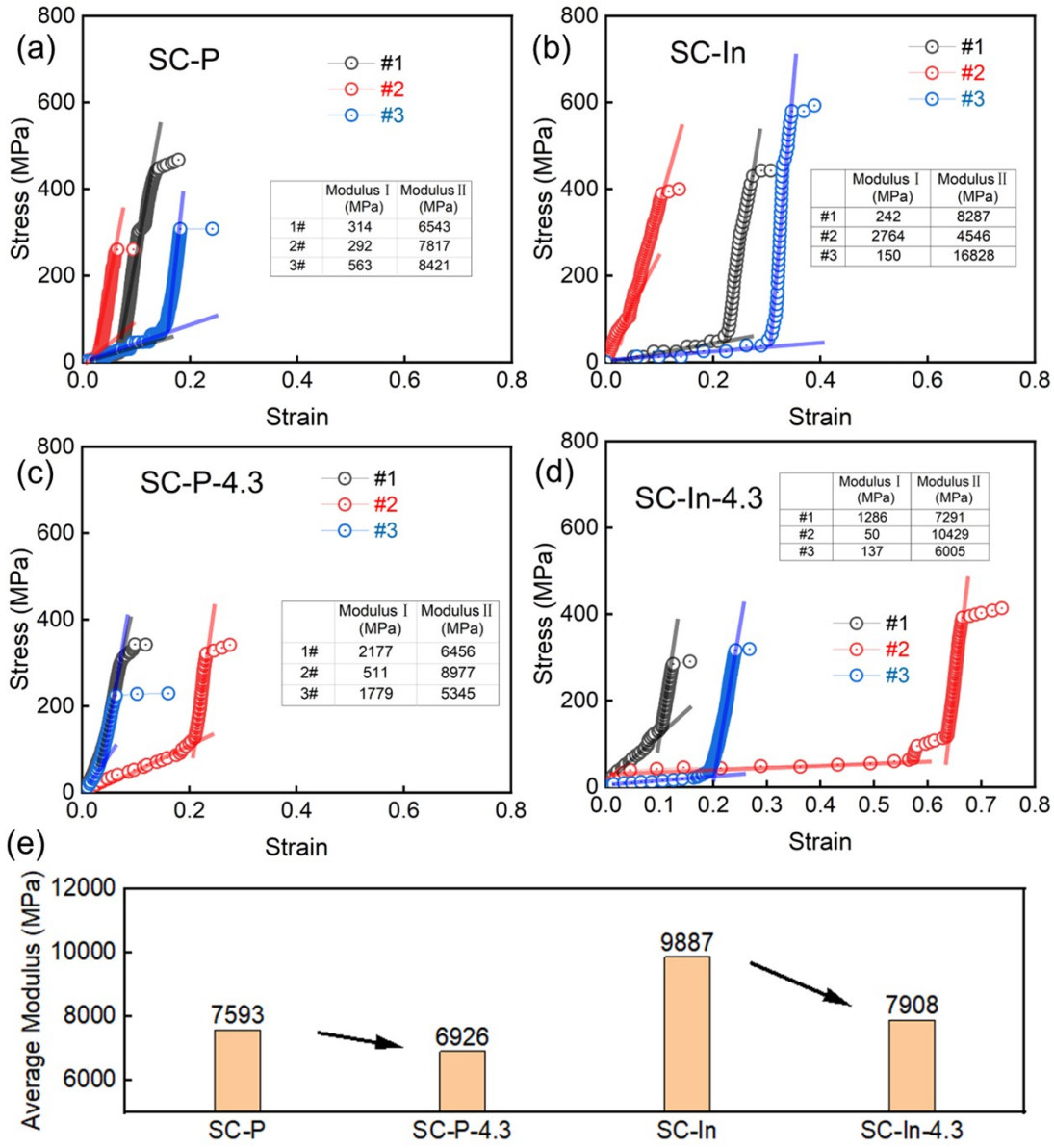

Fig.S3 The fitting results of modulus for (a, b) SC-P, SC-In and (c, d) SC-P-4.3, SC-In-4.3. (e)The average modulus at the latter stage.

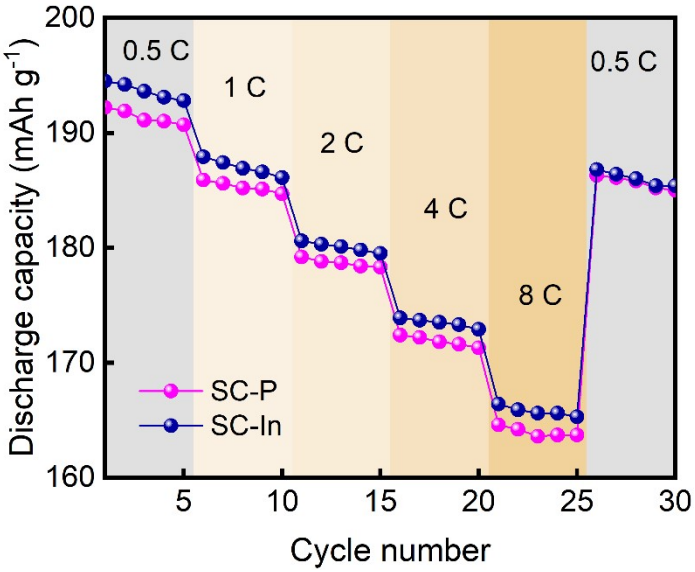

Fig.S4 The rate performances of SC-P and SC-In.

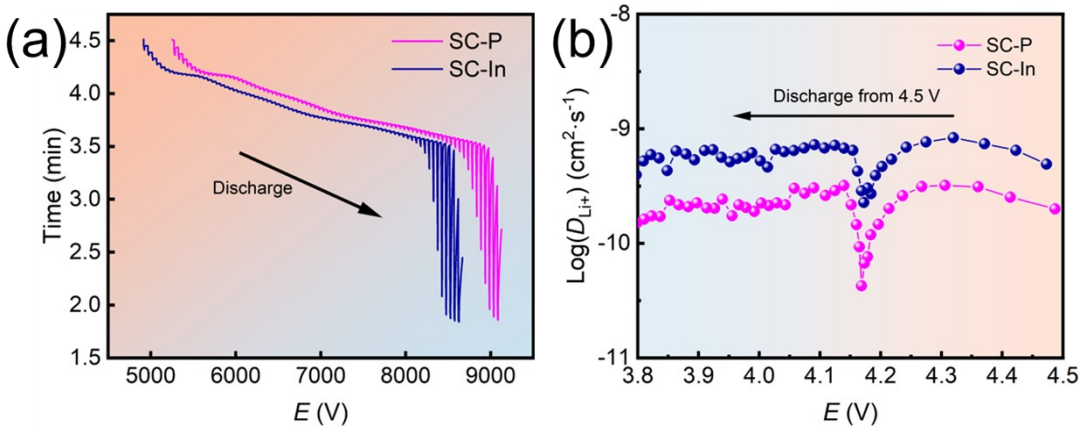

Fig.S5 The GITT curves of SC-P and SC-In.

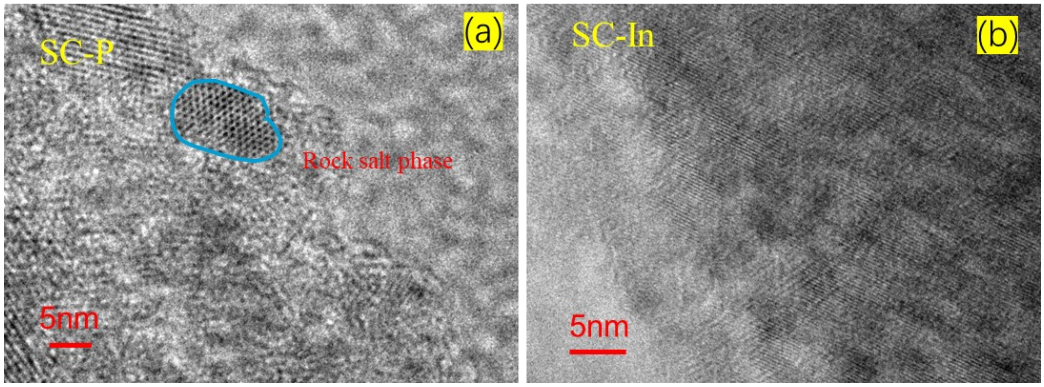

Fig.S6 TEM images of (a) SC-P and (b) SC-In after 100 cycles at 4.5V.