Electronic Supplementary Information

A Facile and Generic Method to Improve Cathode Materials for Lithium-Ion Batteries via Utilizing Nanoscale Surface Amorphous Films of Self-Regulating Thickness

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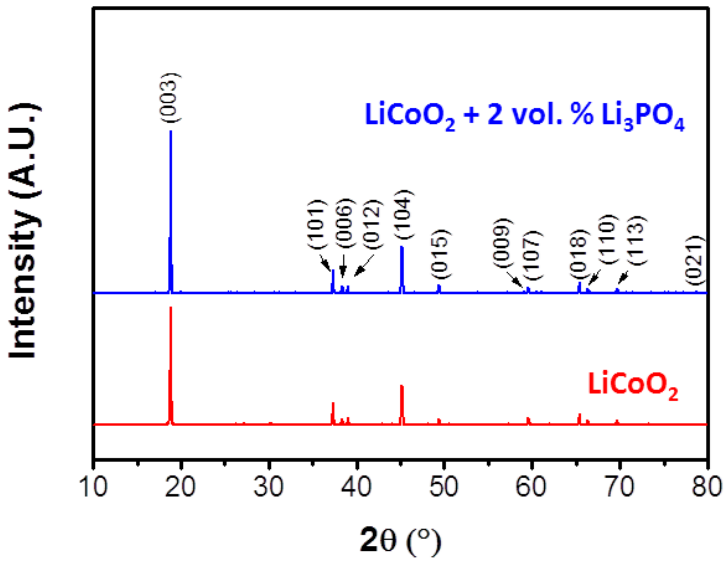

Fig. S1 XRD patterns of the $LiCoO_2$ and $LiCoO_2$ + 2 vol. % Li_3PO_4 specimens that were milled and subsequently annealed at 600 °C for 4 h.

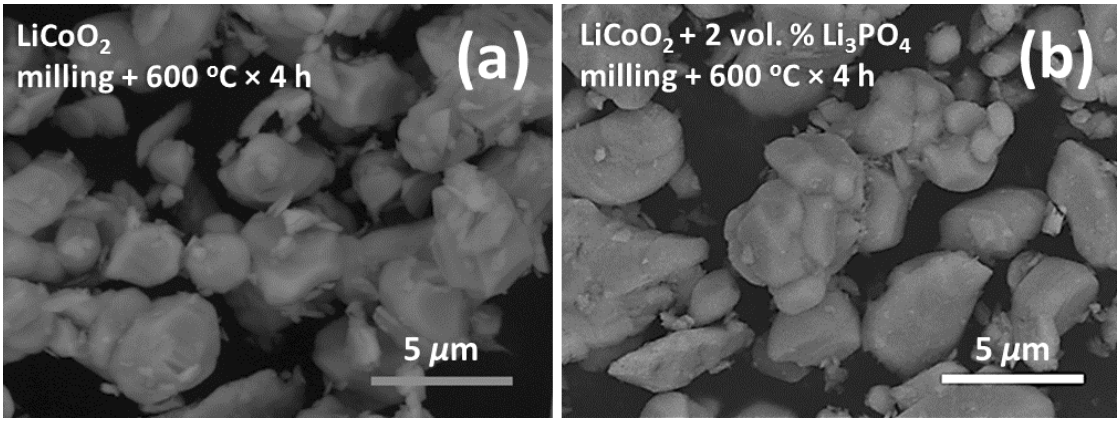

Fig. S2 Representative SEM images of the **(a)** $LiCoO_2$ and **(b)** $LiCoO_2$ + 2 vol. % Li_3PO_4 specimens that were milled and subsequently annealed at 600 °C for 4 h.

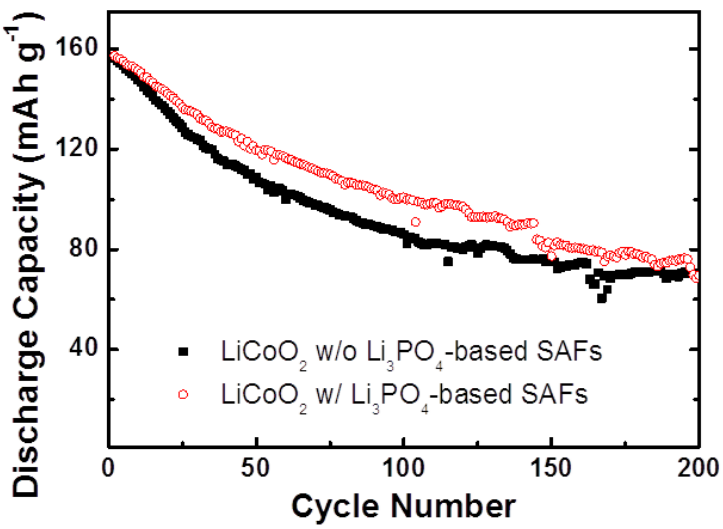

Fig. S3 Cycling stability of $LiCoO_2$ specimens with and without Li_3PO_4-based SAFs that were cycled between 3 V and 4.5 V (*vs.* Li/Li^+) at the charge/discharge rate of 1C at room temperature. The capacity fading is appreciably lower in the specimen with ~ 2.5 nm thick SAFs. It is possible that the SAFs disintegrated after ~140 cycles due to the strain accumulation resulted from cycling at the extended voltage range of 3 V to 4.5 V.

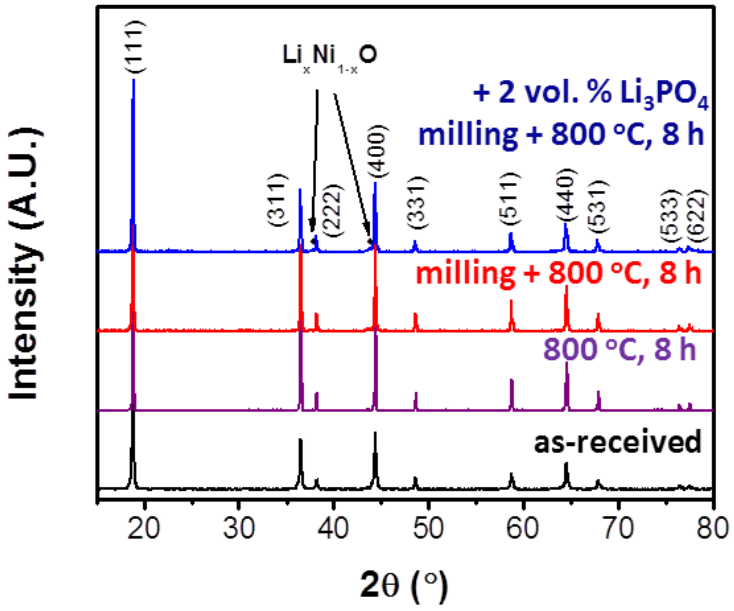

Fig. S4 XRD patterns of various $LiMn_{1.5}Ni_{0.5}O_4$ specimens. Minor amounts of a secondary crystalline phase of $Li_xNi_{1-x}O$ were detected in the milled and annealed specimens.

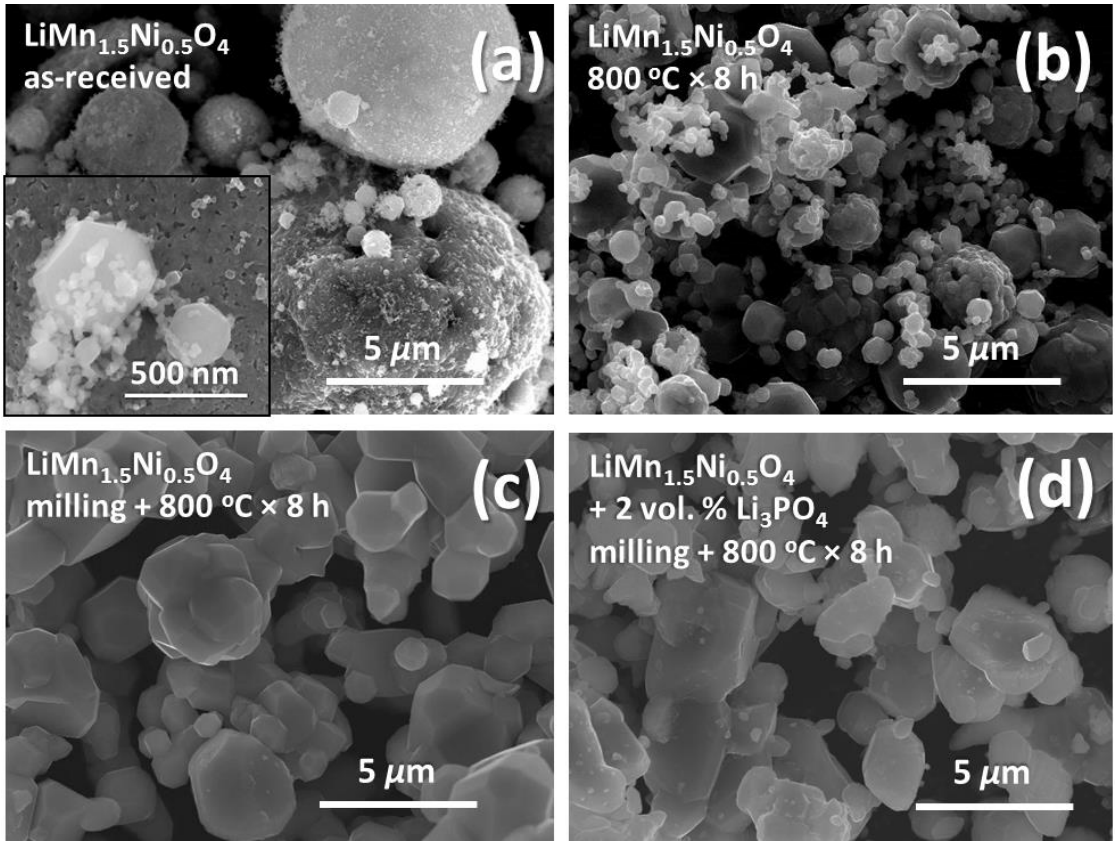

Fig. S5 Representative SEM images of **(a)** the as-received $LiMn_{1.5}Ni_{0.5}O_4$ specimen and **(b)** the reference $LiMn_{1.5}Ni_{0.5}O_4$ specimen annealed at 800 °C (without ball milling), as well as the **(c)** $LiMn_{1.5}Ni_{0.5}O_4$ and **(d)** $LiMn_{1.5}Ni_{0.5}O_4$ + 2 vol. % Li_3PO_4 specimens that were milled and subsequently annealed at 800 °C for 8 h.

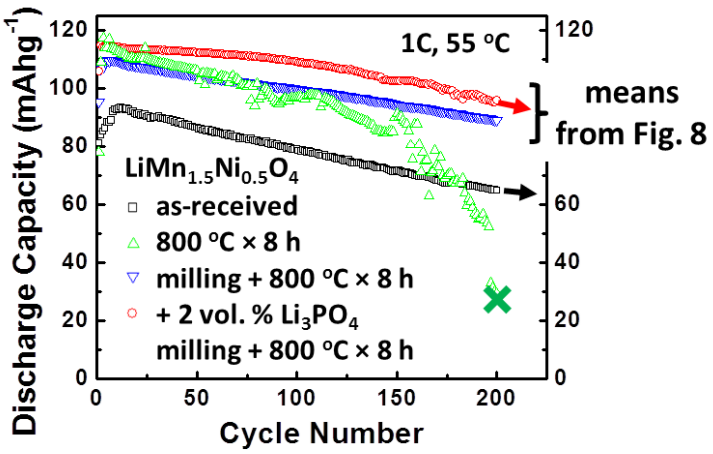

Fig. S6 Comparison of the cycling performances of the as-received $LiMn_{1.5}Ni_{0.5}O_4$ specimen and the reference $LiMn_{1.5}Ni_{0.5}O_4$ specimen annealed at 800 °C without ball milling, along with the two means of the $LiMn_{1.5}Ni_{0.5}O_4$ specimens with and without Li_3PO_4-based SAFs that were milled and annealed at 800 °C, which were averaged from the data shown in Fig. 8 (noting that those uncoated specimens that failed before 200 cycles were excluded for obtaining the mean). All fresh cells were charged and discharged at 1C at 55 °C. "X" indicates that the battery died.