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## Ultrafast Nano-Spherical Single-Crystalline of LiMn<sub>0.792</sub>Fe<sub>0.198</sub>Mg<sub>0.010</sub>PO<sub>4</sub> Solid-Solution Confined among Unbundled Interstices of SGCNT

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Fig. S1 Raman spectrum for the pristine super growth (single-walled) carbon nanotube (SGCNT) with the  $I_D/I_G = 0.40$ . The measurement was conducted on the bundled position of SGCNT by Raman spectroscopy systems (Horiba Jobin-Yvon, LabRam HR evolution).



Fig. S2 [**TOP**] XRD patterns for LiMn<sub>0.76</sub>Fe<sub>0.19</sub>Mg<sub>0.05</sub>PO<sub>4</sub>/SGCNT composite. The major diffraction peaks of these composites ((101), (111), (211), and (311)) are well indexed to the orthorhombic structure of LiMnPO<sub>4</sub> and no peaks for possible impurities were found, as same as other two LiMn<sub>0.8</sub>Fe<sub>0.2</sub>PO<sub>4</sub>/SGCNT and LiMn<sub>0.792</sub>Fe<sub>0.198</sub>Mg<sub>0.010</sub>PO<sub>4</sub>/SGCNT (shown in Fig. 1 right A and B). [**BOTTOM**] Plots of three refined lattice (a-, b-, and c-) parameters and the volume of LiMn<sub>0.8</sub>(1-z)Fe<sub>0.2</sub>(1-z)PO<sub>4</sub> with respect to Mg dosage (z = 0-0.05). The lattice parameters and volume were calculated from the Rietveld analysis on XRD patterns shown in Fig. 1 Right (A), (B), and Fig. S2 top. A decrease in each three parameters and volume with an increase of Mg dosage is due to the smaller lattice size of LiMgPO<sub>4</sub> than other phosphates; LiFePO<sub>4</sub> and LiMnPO<sub>4</sub>. The obtained linearity in each four figures suggests that the Mg atom was successfully doped into the LiMn<sub>0.8</sub>Fe<sub>0.2</sub>PO<sub>4</sub>/SGCNT nano-crystals.



Fig. S3 HRTEM images for the synthesized LiMn<sub>0.8</sub>Fe<sub>0.2</sub>PO<sub>4</sub>/ supergrowth (single-walled) carbon nanotubes (denoted as SGCNT) composite, which was prepared under ultra-centrifuging (UC) treatment with the same dosage condition of Mn/Fe (= 4) for the LiMn<sub>0.792</sub>Fe<sub>0.198</sub>Mg<sub>0.01</sub>PO<sub>4</sub>/SGCNT except Mg doping (see experimental section). (a) Lower magnification HRTEM image shows that the spherical LiMn<sub>0.8</sub>Fe<sub>0.2</sub>PO<sub>4</sub> (10-40 nm of diameter), which is highly dispersed and encapsulated within the SGCNT interstices. (b) Higher magnification HRTEM image of the LiMn<sub>0.8</sub>Fe<sub>0.2</sub>PO<sub>4</sub> nanoparticle with clear lattice fringes. These images (LiMn<sub>0.8</sub>Fe<sub>0.2</sub>PO<sub>4</sub>/SGCNT composite) and Fig. 2a-d (LiMn<sub>0.792</sub>Fe<sub>0.198</sub>Mg<sub>0.010</sub>PO<sub>4</sub>/SGCNT composite) clearly indicate that the UC treatment enables the *in-situ* synthesis of nano-crystalline LiMn<sub>0.8</sub>Fe<sub>0.2</sub>PO<sub>4</sub> or LiMn<sub>0.792</sub>Fe<sub>0.198</sub>Mg<sub>0.010</sub>PO<sub>4</sub> particles in the presence of SGCNT, simultaneously achieving such a highly unbundled state of the SGCNT.



Fig. S4 Electron diffraction patterns for  $LiMn_{0.792}Fe_{0.198}Mg_{0.010}PO_4/SGCNT$  (their HRTEM images are shown in Fig. 2), indicating that those patterns are identical to  $LiMnPO_4$  and graphite attributed to the basal plane of SGCNT.



Fig. S5 (a) Charge-discharge curves for pure SGCNT electrode. Charge-discharge tests were performed using a 2032-typed coin cells composed of Li metal and SGCNT electrodes. Used electrolyte composition was 1M LiPF<sub>6</sub>/EC + DEC (EC/DEC =50/50, v/v). Charge-discharge performance was conducted under the CC-CV (charge) and CC (discharge) mode between 2.5 and 4.5 V, at constant charge current density of 0.1C and at different discharge current densities ranging from 0.1C to 100C (assuming  $1C = 40 \text{ mA g}^{-1}$ . The electrode composition was 90 wt% of SGCNT and 10 wt% of polyvinylidene difluoride. (b) Summarized results of Fig S5a: discharge capacity for SGCNT electrode with respect to current density.