

Fig. S1 HRSEM images of different $K_{0.25}Mn_2O_4$ nanomaterials show nanofibers of (a) KMO-80; (b) KMO-100; (c) KMO-110; (d) KMO-120, with the inset showing still higher magnification.

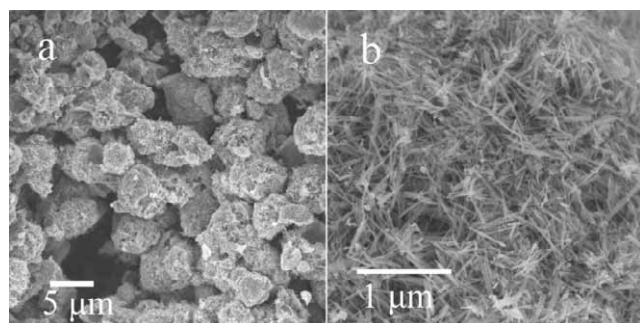


Fig. S2 SEM images of KMO-130 material. The KMO-130 was prepared by using the same hydrothermal method, with the reaction temperature of 130 °C. The SEM image (Fig. S2a) of KMO-130 shows that the sample has diverse microspherical structures. The uniform microspherical morphologies which were found in KMO-80, KNO-100, KMO-110, and KMO-120 have disappeared. Fig S2b shows high resolution SEM (HRSEM) images of the sample.

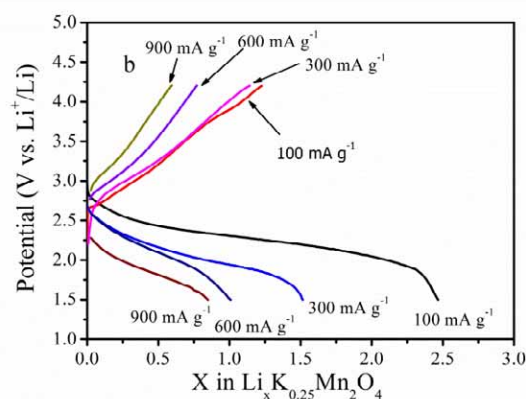
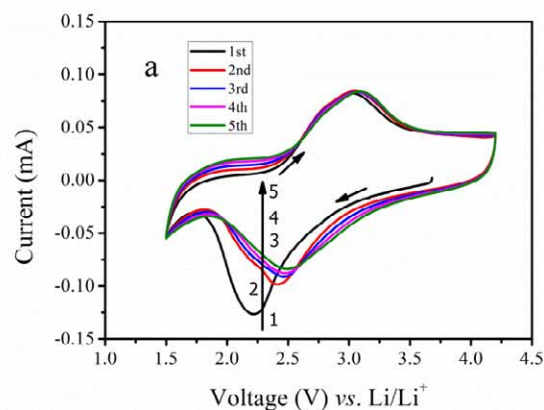


Fig. S3 Electrochemical measurements on the $K_{0.25}Mn_2O_4$ electrode: (a) cyclic voltammograms for the first 5 cycles at scan rate of 0.1 mV s⁻¹; (b) voltage vs. the amount of Li insertion in the $K_{0.25}Mn_2O_4$ electrode at different current densities.

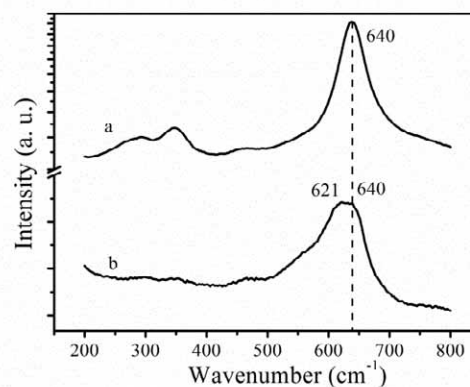


Fig. S4 Raman spectra of $K_{0.25}Mn_2O_4$ nanofibers: a) before cycling, b) after 100 cycles at 100 mA g⁻¹.

Raman spectroscopy was used to investigate vibrational information on the manganese-oxygen (MnO_6) octahedra in the $K_{0.25}Mn_2O_4$. A representative Raman spectrum of fresh $K_{0.25}Mn_2O_4$ electrode is shown in Fig. S4a. The sample shows a strong wide peak at approximately 640

cm^{-1} and some weak shoulder bands around 291, 350, 395, and 460 cm^{-1} . The high-frequency Raman band at 640 cm^{-1} , which indicates well developed tetragonal structure with an interstitial space consisting of (2×2) tunnels, is assigned to the A_g mode.^{S1-S3} The observed Raman frequencies in the 200-500 cm^{-1} range can be assigned to the Mn-O stretching of MnO_6 octahedra.^{S4-S7} A second Raman spectrum of this sample, collected after 100 charge-discharge cycles at 100 mA g^{-1} , is shown in Fig. S4b. The Raman frequencies in the 200-500 cm^{-1} range remain unchanged, except for a slight shift. However, the strongest characteristic peak can be divided into two peaks, at 640 and 621 cm^{-1} , indicating the elongation of the Mn-O bond, as evaluated from the results in the literature.^{S7-8} The change has occurred because the surrounding environment of the Mn was changed by the presence of more Li ions.

1. Y.-L. Ding, J. Xie, G.-S. Cao, T.-J. Zhu, H.-M. Yu, X.-B. Zhao, *Adv. Funct. Mater.* 2011, 21, 348.
2. S.-J. Hwang, C.-W. Kwon, J. Portier, G. Campet, H.-S. Park, J.-H. Choy, P. V. Huong, M. Yoshimura, M. Kakihana, *J. Phys. Chem. B* 2002, 106, 4053.
3. T. Gao, M. Glerup, F. Krumeich, R. Nesper, H. Fjellvåg, P. Norby, *J. Phys. Chem. C* 2008, 112, 13134.
4. E. K. Nyutu, C.-H. Chen, S. Sithambaram, V. M. B. Crisostomo, S. L. Suib, *J. Phys. Chem. C* 2008, 112, 6786.
5. W. Y. Hernández, M. A. Centeno, F. Romero-Sarria, S. Ivanova, M. Montes, J. A. Odriozola, *Catal. Today* 2010, 157, 160.
6. R. Baddour-Hadjean, J.-P. Pereira-Ramos, *Chem. Rev.* 2010, 110, 1278.
7. L. Sun, Q. Cao, B. Hu, J. Li, J. Hao, G. Jing, X. Tang, *Appl. Catal. A: Gen.* 2011, 393, 323.
8. F. D. Hardcastle, I. E. Wachs, *J. Phys. Chem.* 1991, 95, 5031.