Supporting Information

Preparation and electrochemical properties of double-shell $LiNi_{0.5}Mn_{1.5}O_4$ hollow microspheres as cathode materials for Li-ion batteries

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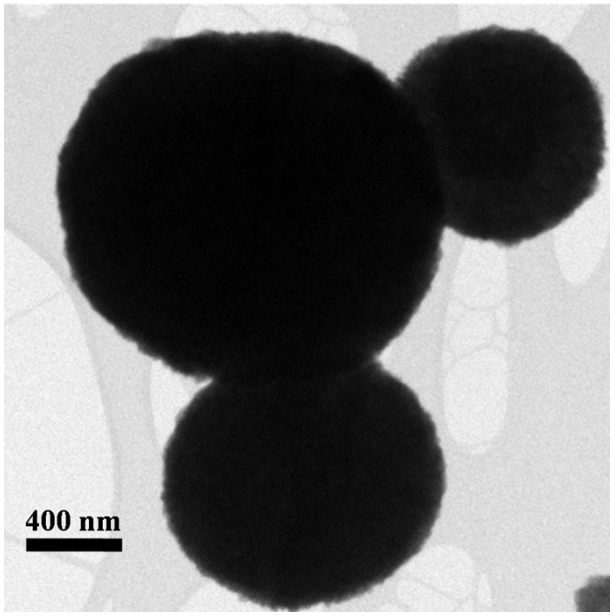

Fig. S1 TEM images of MnO_2.

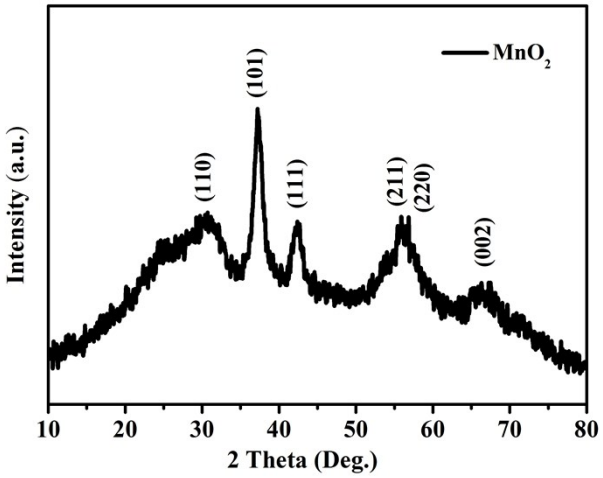

Fig. S2 XRD patterns of MnO_2 microspheres.

Fig. S2 shows the XRD pattern of MnO_2 microspheres. All of the diffraction peaks can be indexed to the tetragonal phase of β-MnO_2 (JCPDS, 24-0735), and no other characteristic peaks from impurities are detected in the spectrum.

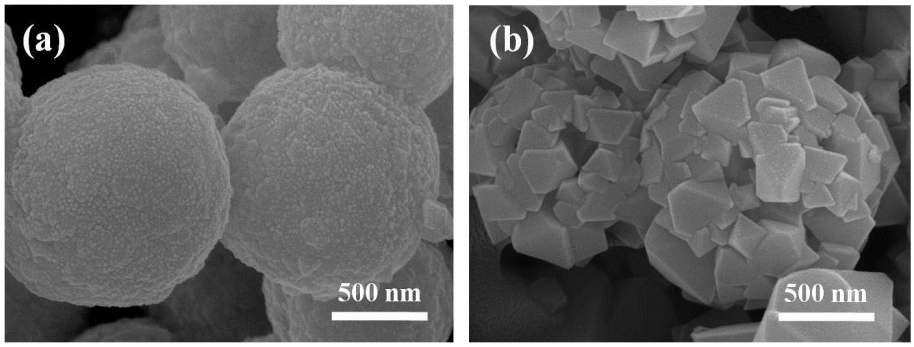

Fig. S3 SEM images of spherical LNMO heat-treated at 750 °C, 5 h (a), 15 h (b).

As can be seen from Fig. S3(a), the as-obtained LNMO spheres retain the morphology of MnO_2 template with the particle size of about 1μm after 5 h calcinations. Meanwhile, no broken spheres can be seen in this LNMO sample. Shown in Fig. S3(b) is the SEM image of LNMO powders obtained at 750 °C for 15 h. It can be found that the powders are still composed of spherical structure. However, the primary particles grew bigger than the LNMO-DS shown in paper and gradually broke into nanocrystallites.

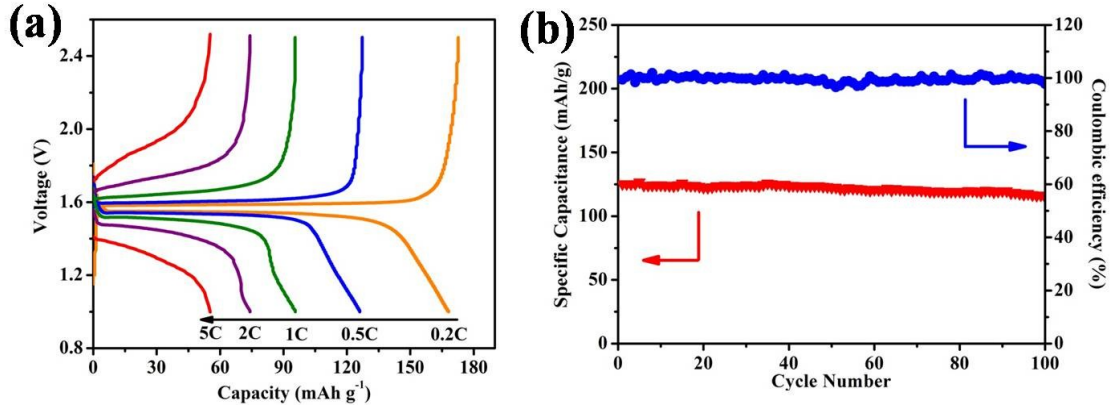

Fig. S4 Charge and discharge curves of LTO at different rates (a), Cycle performance and Coulombic efficiency of LTO (b). [1]

Fig. S4(a) depicts the first charge/discharge curves of the LTO electrode at the current rates from 0.2 C to 5 C [1]. At the initial lower rate of 0.2 C, the LTO electrode shows a flat discharge voltage plateau at the potential of ~1.55 V and exhibits a discharge capacity of 168 mAh g^{-1}, which is very close to the theoretical capacity of 175 mAh g^{-1}. The gap between the charge and discharge is only 0.1 V, indicating no obvious polarization. It is notable that the capacity retention rate at 5 C still maintain nearly 33% of its capacity compared with 0.2 C. For evaluating the cycling stability of the LTO sample, the charge/discharge at a current rate of 0.5 C for 100 cycles is shown in Fig. S4(b). It can be observed that the LTO sample shows a stable discharge capacity. The first discharge capacity is 125 mAh g^{-1}, and even after 100 charge/discharge cycles, its capacity also remains as 115 mAh g^{-1}. Furthermore, as is shown in Fig. 5, the Coulombic efficiency in this long cycle period still remained almost at 99%. The results show that the prepared LTO electrode has good capacity retention which is suitable to be a negative electrode in LNMO-DS//LTO full-cell.

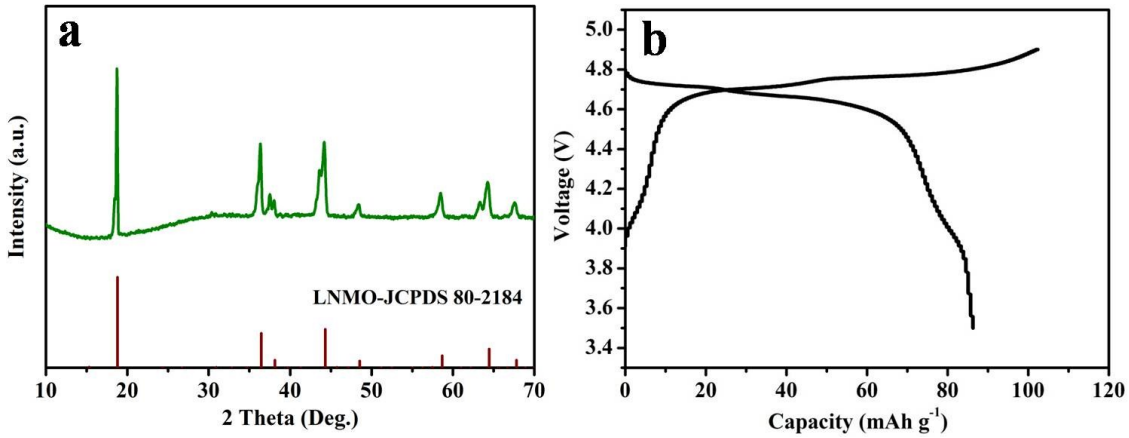

Fig. S5 XRD patterns of bulk LNMO with heat-treated at 750 °C for 10 h (a), galvanostatic charge/discharge curves of bulk LNMO (750 °C for 10 h) at 1st cycles at 0.5 C (b).

Fig. S5 shows the XRD and electrochemical performance of bulk LNMO annealing at 750 °C for 10 h. According to the pattern in Fig. S5 (a), the bulk LNMO did not show pure phase when annealing under 750 °C for 10 h, which demonstrated impurity phases in XRD patterns. Furthermore, it delivers an initial discharge capacity of 86.3 mAh g^{-1} with a Coulombic efficiency of 84.6%, which are much lower than the corresponding electrochemical performance of LNMO-bulk (950 °C for 18 h) in this paper.

References

1. S. Deng, J. Li, S. Sun, H. Wang, J. Liu and H. Yan, *Electrochimica Acta*, 2014, **146**, 37-43.