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Supplementary Information

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Sample Surface Morphology

The surface morphology is one of the most important feature of the yolk-structured $LiMn_2O_4$ microsphere. To prove the micro-porosity shell which was formed through the oxidation reaction, the SEM images with larger magnification about its surface morphology is shown in **Fig. S1**.

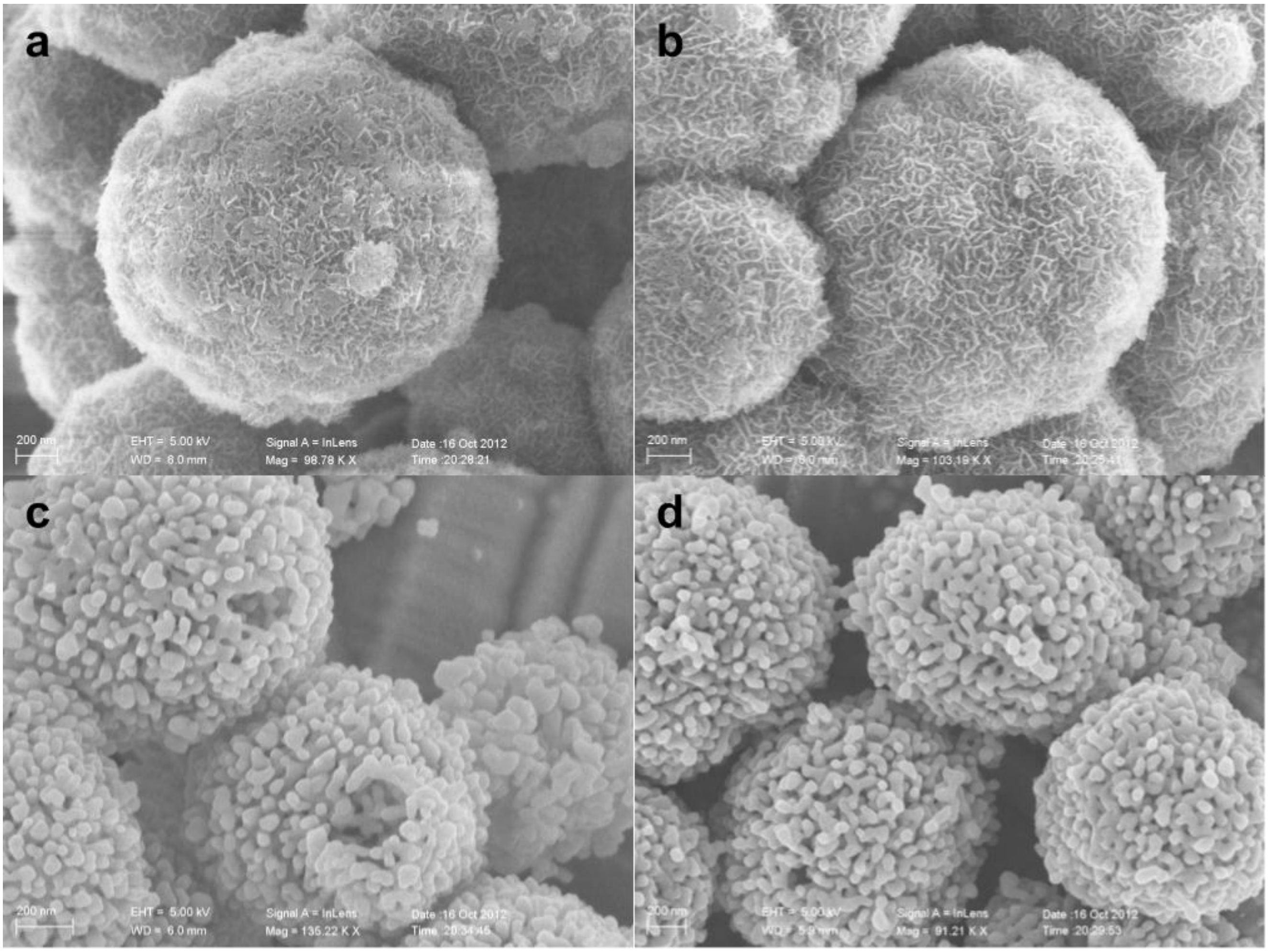

Fig. S1 SEM images of microspheres of (a) core-shell-structured $MnCO_3$-MnO_2, (b) yolk-structured $MnCO_3$ (core)-MnO_2 (shell), (c) yolk-structured Mn_2O_3 and (d) yolk-structured $LiMn_2O_4$.

As shown in **Fig. S1**, indeed there are some micropores on the surface of the yolk-structured $MnCO_3$ (core)-MnO_2 (shell). These micropores correspond exactly to the micro-porosity of the MnO_2 shell formed by the oxidation reaction product. Meanwhile, the particles (about 150 μm in diameter) on the surface of yolk-structured $LiMn_2O_4$ and the pores between them can also prove the existence of surface micropores.

Electrode Thickness Measurement

The three $LiMn_2O_4$ samples should have different tap densities due to their different particle morphologies. From macroscopic view, the three samples with same mass should have different volumes. To prove this point, we chose each electrode laminate with about same mass（each with about 6.2 mg active material）to compare their thickness.

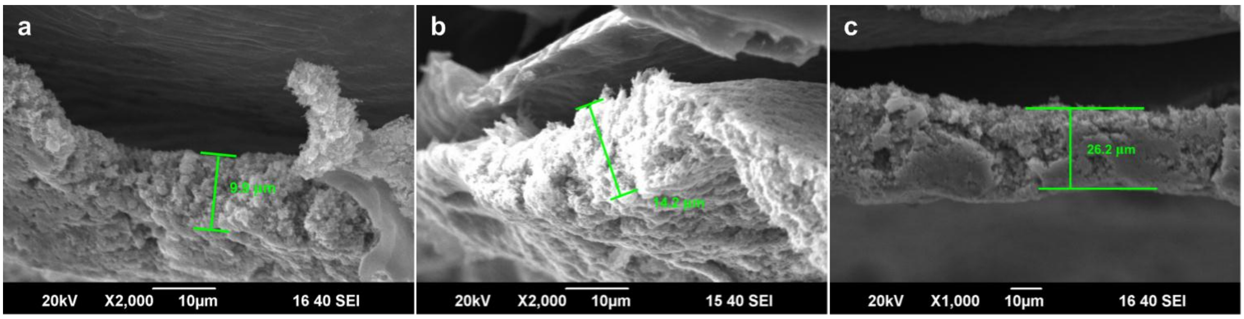

Fig. S2 SEM images of the electrode thickness of the three samples, (a) LMO-S, (b) LMO-Y, (c) LMO-H.

As shown in **Fig. S2**, the electrode thickness of the three samples is different one another. The LMO-S electrode is the thinnest (nearly 10 μm), because the solid structure has the highest density. At the same time, LMO-H electrode is the thickest (26.2 μm) due to its hollow structure, while the thickness of LMO-Y electrode is nearly 14.2 μm (middle of the three) which is also consistent with its particle-structure mentioned before.

Different Synthetic Conditions for Yolk-structured $LiMn_2O_4$

Sample Preparation and Characterization

To find out the well-optimized conditions for synthesizing the yolk-structured $LiMn_2O_4$ microspheres, different concentration of HCl and $KMnO_4$ (**Table S1**) is also tried during the synthetic procedures as a comparison.

Table S1 The different conditions in the synthetic process

	Concentration of $KMnO_4$ (mol L^{-1})	Concentration of HCl (mol L^{-1})
yolk-A	0064	0.6
yolk-B	0.032	0.6
yolk-C	0.032	1.2

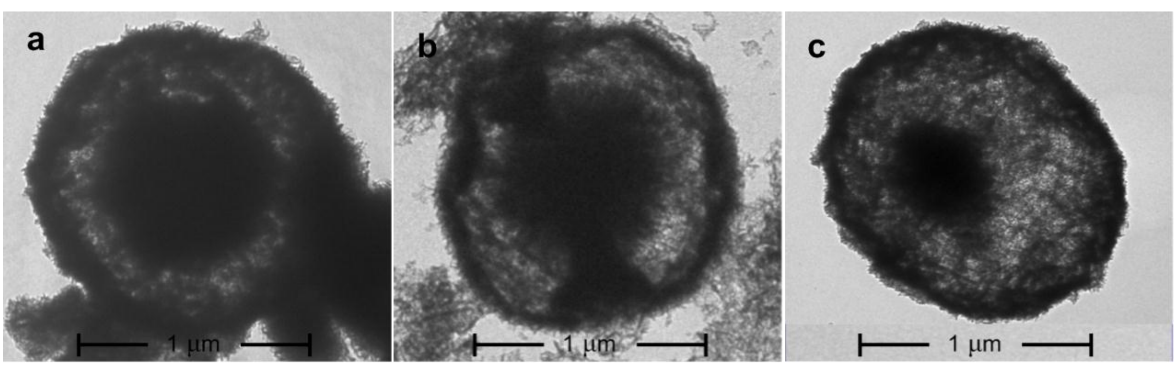

Fig. S3 TEM images of the $LiMn_2O_4$ microsphere synthetized in differert conditions, (a) yolk-A, (b) yolk-B, (c) yolk-C.

As it shown in **Fig. S3**, yolk-A own the thickest shell because the larger concentration of $KMnO_4$ is employed duration the oxidation process, while sample yolk-C performs the smallest core radius mainly depends on the larger concentration of HCl duration the procedure of etch.

Cell Test Results

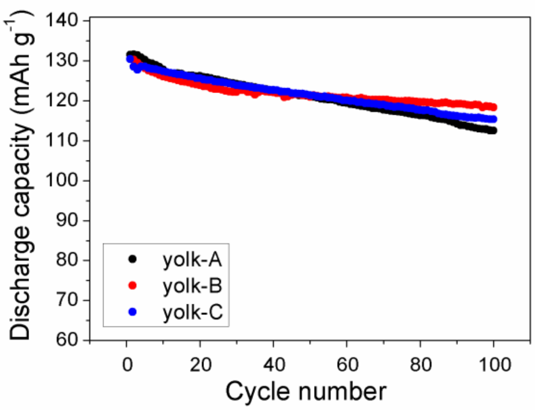

Fig. S4 Cycling performations of the three $LiMn_2O_4$ samples at 55°C at 1C rate

Fig. S4 reveals the cycle performance of the three yolk-structured $LiMn_2O_4$ samples at 55°C at 1C rate between 3.2 V and 4.35 V. Through the figure above, we can easily find out the yolk-B sample performs the best retetion of capacities after 100 cycles. Its excellent cycle performance at 1C rate and elevated tempertature presents the well-optimized conditions for synthesizing the yolk-structured $LiMn_2O_4$ microspheres.