

Supporting Information

Superior electrochemical properties of LiMn₂O₄ yolk-shell powders prepared by a simple spray pyrolysis process

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This file includes:

- (a) Coulombic efficiencies of yolk-shell LiMn₂O₄ powders at various current densities; the charge and discharge curves of (b) Yolk-shell LiMn₂O₄ powders post-treated at 650°C, (c) Yolk-shell LiMn₂O₄ precursor, and (d) Dense LiMn₂O₄ powders post-treated at 650°C at a current density of 3 C.
- SEM images of (a) Yolk-shell LiMn₂O₄ powders post-treated at 650°C, (b) Yolk-shell LiMn₂O₄ precursor, and (c) Dense LiMn₂O₄ powders post-treated at 650°C after 200 cycles at a current density of 3 C.

Detailed experimental procedure:

The precursor powders for LiMn_2O_4 yolk-shell materials were prepared by ultrasonic spray pyrolysis. A cylindrical quartz reactor (length, 1200 mm; diameter, 50 mm) was used. The reactor temperature was maintained at 700°C . The flow rate of the air used as the carrier gas was 10 L min^{-1} . An aqueous spray solution was prepared by dissolving lithium nitrate (LiNO_3 , Junsei) and manganese nitrate hexahydrate [$\text{Mn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, Junsei] in distilled water. The amount of lithium added to the spray solution was in 3 wt% excess of the stoichiometric amount required to facilitate the formation of LiMn_2O_4 powders. The total concentration of the Li and Mn components dissolved in distilled water was 0.2 M. The concentration of sucrose used as the carbon source material was fixed at 0.7 M. The precursor powders obtained by spray pyrolysis were post-treated at 650°C for 3 h in air atmosphere.

The crystal structures of the prepared cathode powders were determined using X-ray diffractometry (XRD, X'pert PRO MPD) with Cu $K\alpha$ radiation ($\lambda = 1.5418\text{ \AA}$). The morphological characteristics of the powders were investigated using scanning electron microscopy (SEM, JEOL JSM-6060) and high-resolution transmission electron microscopy (HR-TEM, JEOL-2100F). The surface areas of the powders were measured using the Brunauer–Emmett–Teller (BET) method with N_2 as the adsorbate gas.

The capacities and cycle properties of the powders were determined by using a 2032-type coin cell. The cathode electrodes were prepared from a mixture containing 80 wt% active material, 10 wt% activated carbon, and 10 wt% sodium carboxymethyl cellulose (CMC), the latter acting as a binder. Li metal and a microporous polypropylene film were used as the anode electrode and separator, respectively. The electrolyte was 1 M LiPF_6 in a 1:1 mixture (by volume) of ethylene carbonate/dimethyl carbonate (EC/DMC).

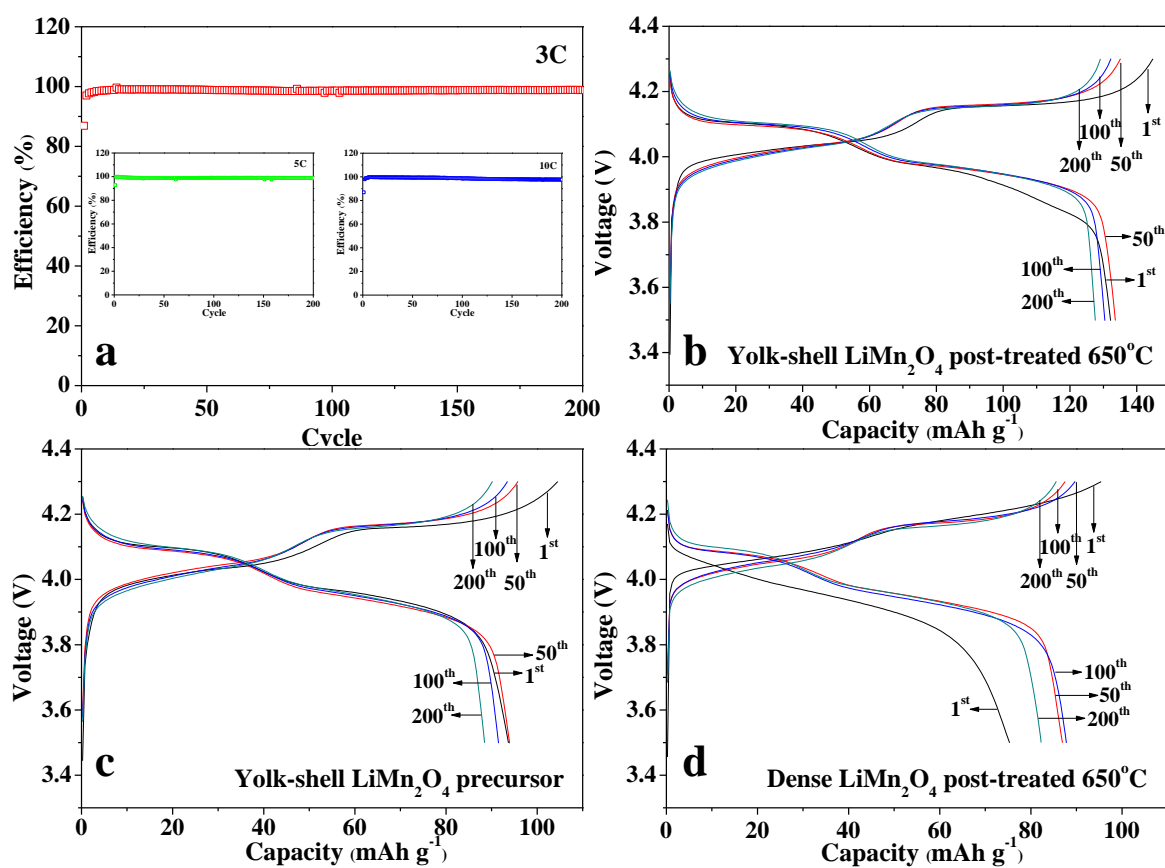


Fig. S1 (a) Coulombic efficiencies of yolk-shell LiMn_2O_4 powders at various current densities; the charge and discharge curves of (b) Yolk-shell LiMn_2O_4 powders post-treated at 650°C , (c) Yolk-shell LiMn_2O_4 precursor, and (d) Dense LiMn_2O_4 powders post-treated at 650°C at a current density of 3 C.

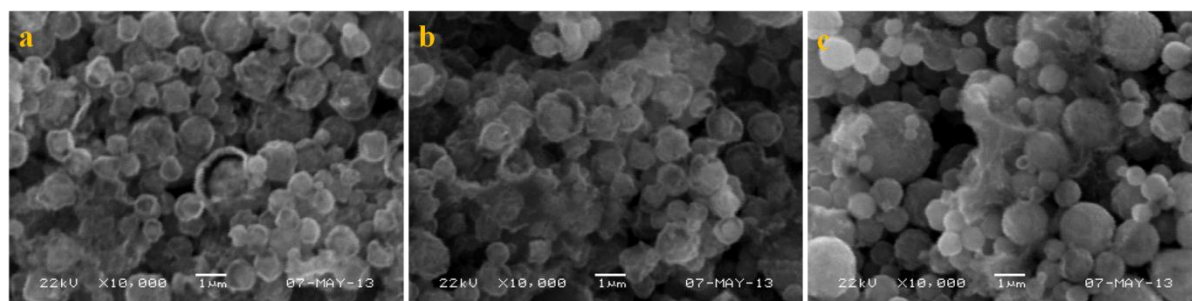


Fig. S2 SEM images of (a) Yolk-shell LiMn_2O_4 powders post-treated at 650°C , (b) Yolk-shell LiMn_2O_4 precursor, and (c) Dense LiMn_2O_4 powders post-treated at 650°C after 200 cycles at a current density of 3 C.