Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

Influence of HDI as cathode film-forming additive on the performance of $LiFe_{0.2}Mn_{0.8}PO_4$/C cathode

1. Experimental Section
2. Table S1 The simulated resistance R_{ct} of simples.
3. Figure S1 XRD patterns of the $LiFe_{0.2}Mn_{0.8}PO_4$/C materials with different C-coatings.
4. Figure S2 TEM image of the $LiFe_{0.2}Mn_{0.8}PO_4$/C cathode with C-coating 2 wt%.
5. Figure S3 SEM image of the $LiFe_{0.2}Mn_{0.8}PO_4$/C cathode with C-coating 2 wt%.
6. Figure S4 Cyclic and rate performances of the $LiFe_{0.2}Mn_{0.8}PO_4$ cathodes.

1. Experimental Section

1.1 Preparation of $LiFe_{0.2}Mn_{0.8}PO_4$/C electrode and electrolyte

Propylene carbonate (PC), dimethyl carbonate (DMC), polyvinylidenefluoride (PVdF), Super P (SP) and $LiPF_6$ were provided by Zhangjiagang Guotai-Huarong New Chemical Materials Co. Ltd., battery grade. All of the metal salts, hexamethylene diisocyanate (HDI, 99 %), dimethyl sulfoxide(DMSO, AR) and N-methyl-2-pyrrolidone (NMP, AR) were provided by Aldrich and used as received without further purification. A first solution was obtained by mixing 0.8 M $MnSO_4 \cdot H_2O$, 0.2 M $FeSO_4 \cdot 7H_2O$, and 1 M H_3PO_4 in DMSO. Then 3.0 M $LiOH \cdot H_2O$ solution was mixed with the first solution by controlling the pH value at 10.2-10.3, the Li:(Mn+Fe):P molar ratio of 3:1:1. After stirring for half an hour under Ar atmosphere, the mixture was heated at 130 ℃ for 6 h under a nitrogen atmosphere, then cooled to room temperature and filtered. The precipitates were washed with distilled water for several times and dried under vacuum at 80℃ for 12 h to obtain the phase-mixed $LiMn_{0.8}Fe_{0.2}PO_4$ (LMFP). The carbon coating was performed on the LMFP powers with a solution of sucrose with the ratio of 2 wt%, 3 wt%, 5 wt%, 10 wt% separately. The mixture was dried at 60 ℃ under vacuum, followed by sintering in a pipe furnace at 600 ℃ under a gas mixture of H_2:Ar 5:95.

The base electrolyte was 1.0 mol L^{-1} $LiPF_6$ in PC/DMC (1:1, v:v), and the investigated electrolytes were different concentrations of HDI added in base electrolytes. All of the electrolytes were prepared in an argon-filled glovebox with water and O_2 content <0.1 ppm.

1.2 Measurements

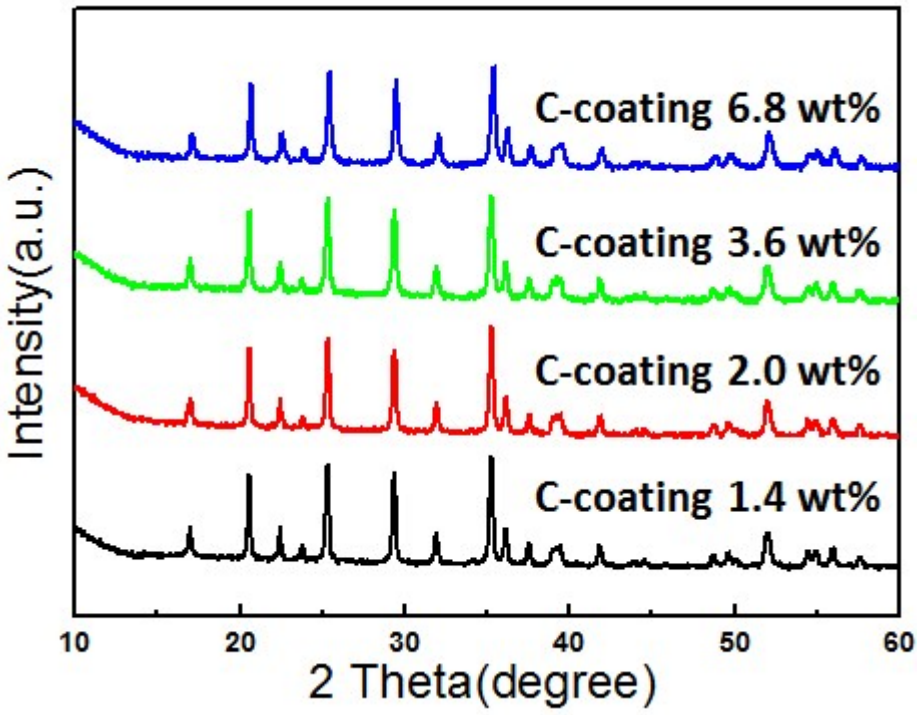
The 2032-type coin cells of Li/$LiFe_{0.2}Mn_{0.8}PO_4$/C with microporous membranes (Celgard 2550) as the separator were assembled in the argon-filled glove box. Electrochemical behaviors of cells constructed with and without HDI in the electrolytes were determined by cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) on an electrochemical workstation (FRA 1455A, Solartron Metrology). The cells were cycled on a Land cell tester (CT2001A, Wuhan Jinnuo Company) with charge cutoff voltages were set at 4.4 V(*vs.* Li^+/Li), discharge cutoff voltages were set at 2.2 V(*vs.* Li^+/Li), room temperature(RT), and the capacities were measured in $LiFe_{0.2}Mn_{0.8}PO_4$/C.

The cathodes were taken from the cycled cells after disassembled carefully in an argon-filled glove box, washed with DMC, wiped with filter papers, and vacuum dried overnight in small chamber of glove box. Surface analysis was conducted with a PHI 3056 X-ray photoelectron spectroscopy (XPS) which was excited by an Mg Ka radiation at a constant power of 100W (15 kV and 6.67 mA), analyzed with GASA, and the morphologies of samples were observed with scanning electron microscopy (SEM, HitachiS-4800) and microstructures were examined by transmission electron microscopy (TEM) on a FEI Tecnai F20 equipped with a field emission gun at an accelerating voltage of 200 kV. X-ray diffraction (XRD) was performed on a Bruker D8 Advanced diffractometer with Cu Kα (λ = 1.5406 Å) radiation. The samples were scanned from 10 ° to 90 ° with the step of 0.2 ° and the step duration of 0.2 s.

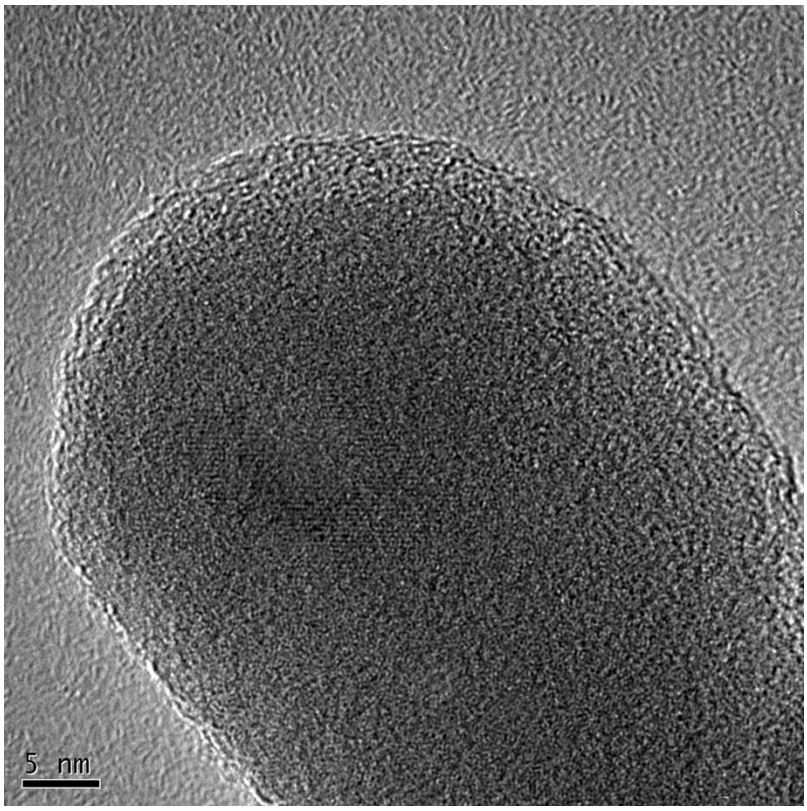
2. Table S1. The simulated resistance R_{ct}(ohm g^{-1}) of simples. All the simples were charged at 4.4 V(*vs.* Li^+/Li) and tested at open circuit voltage, room temperature.

C-coating(wt%)	1.4	2.0	3.6	6.8
R_{ct} of simples in base electrolyte (ohm g^{-1})	142.6	97.1	85.5	95.9
R_{ct} of simples with 10 mM HDI added (ohm g^{-1})	76.7	71.4	78.5	80.6

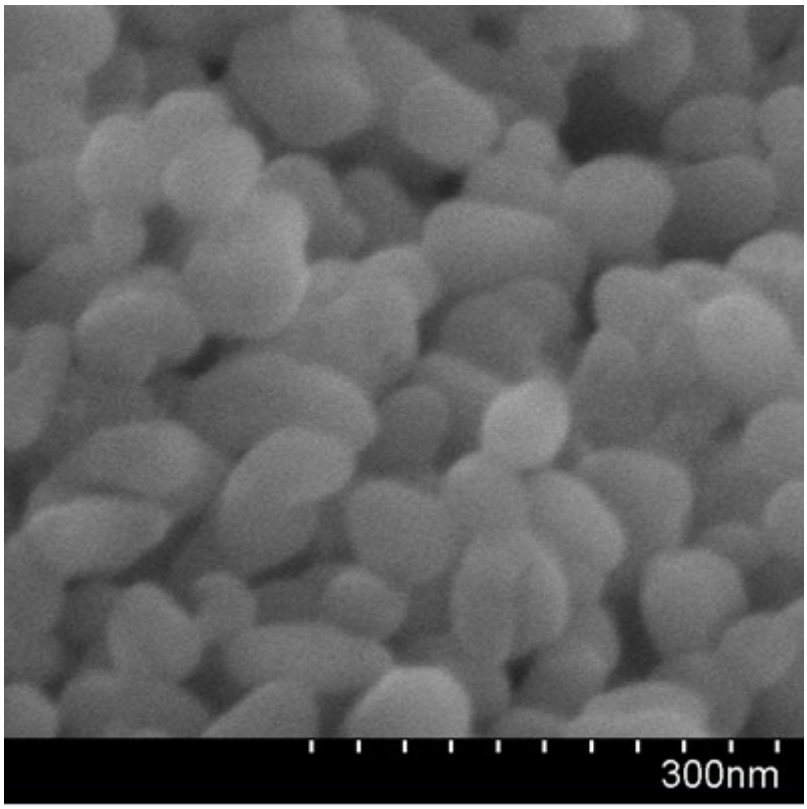
3. Figure S1. XRD patterns of the $LiFe_{0.2}Mn_{0.8}PO_4$/C materials with different C-coatings.



4. Figure S2. TEM image of the cathode surface with C-coating 2 wt%.



5. Figure S3. SEM image of the cathode surface with C-coating 2 wt%.



6. Figure S4. Cyclic(a) and rate(b) performances of the $LiFe_{0.2}Mn_{0.8}PO_4$ cathodes in base and HDI added electrolytes.