Supplementary data

Intergrown Li₂FeSiO₄·LiFePO₄/C nanocomposites as high-capacity cathode material for lithium-ion batteries

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Experimental

(1-x)Li₂FeSiO₄·xLiFePO₄/C (x = 0, 0.02, 0.04, 0.08, 0.10 and 1.00) nanocomposites were synthesized by tartaric acid assisted sol-gel method. stoichiometric amounts of tetraethyl orthosilicate (TEOS) and tartaric acid were first dispersed in ethanol, then a deionized water solution of stoichiometric amounts of LiNO₃·H₂O and Fe(NO₃)₃·9H₂O was added under continuous stirring, NH₄H₂PO₄ dissolved in deionized water was added followed by. The mixture was stirred in a water bath at 60 °C for 3 h and then evaporated at the same temperature to form a transparent gel. After being dried at 80 °C for 10 h, the xerogel was grounded and sintered at 350 °C for 4 h, 600 °C for 8 h in Ar atmosphere.

Material Characterization

The structures of the as-synthesized samples were characterized by powder X-ray diffraction (XRD, Rigaku MiniFlex600, Cu Kα radiation). The morphologies of the

samples were observed by scanning electron microscopy (SEM, JEOL, JSM-7500F). The high-resolution transmission electron microcopy (HRTEM, tecnai G2 F20) equipped with an energy dispersive X-ray detector (EDX) were employed to investigate the particle distribution and surface morphologies of the synthesized samples. The specific surface area and pore size distribution were analyzed by Brunauer–Emmett–Teller (BET) nitrogen adsorption–desorption measurement (Japan, BELSORP-Mini). The weight ratio of the coated carbon of the sample was tested by using elemental analyzer (German, Vario EL CUBE).

The working electrodes were made from a mixture of 75 wt. % of the active material, 15 wt. % of the conducting agent (Super P), and 10 wt. % of the polyvinylidene difluoride (PVDF) binder. The assembled cells were cycled at different charge-discharge rates in the voltage range of 1.5 – 4.8 V on a CT2001A cell test instrument (LAND Electronic Co.). The specific capacity was calculated on the basis of the amount of the active material, excluding the mass of the coated carbon. The cyclic voltammograms (CV) were performed at different scan rates with a Parstat 263A electrochemical workstation (AMTECT Company).



Fig. S1. SEM images of (1-x)Li₂FeSiO₄·*x*LiFePO₄ nanocomposites with x = 0, 0.02, 0.04, 0.08, 0.10, and 1.0.



Fig. S2. The EDS image of $0.96Li_2FeSiO_4 \cdot 0.04LiFePO_4/C$ nanocomposites measured on Cu grid showing the presence of C, O, Fe, Si, P and Cu.



Fig. S3. XPS spectrum of $(1-x)Li_2FeSiO_4 \cdot xLiFePO_4$ nanocomposites with x = 0, 0.04, and 1.00.



Fig. S4. The charge-discharge curves of $(1-x)Li_2FeSiO_4 \cdot xLiFePO_4$ nanocomposites at 0.2 C, 0.5 C and 1 C (a — e). The cyclic performance of sample with x = 0 and 0.04 tested at 45 °C (f).



Fig. S5. The cyclic performance of samples with x = 0 and 0.04 tested at 0.2 C at 15

°C.



Fig. S6. SEM images of electrode surface of samples with x = 0 and 0.04 before (a and b) and after 100 cycles (c and d).

x = 0.02

x = 0.04

x = 0.08

x = 0.10

3.095+

3.047@

3.130@

3.093e

discharge of sample with $x = 0.02$).					
	Sample∉	Charge Potential/V $_{\!$	Discharge Potential/ V_{e^j}	$\Delta V/V_{*^3}$	$D_{Li}^+/cm^2 s^{-1}$
	$x = 0_{*}$	3.296¢	2.155+	1.141.	3.7×10 ⁻¹⁵ ,

2.534+

2.646

2.630+

2.589+

0.561₽

0.401

0.500+

0.504@

 1.07×10^{-14}

 $2.36 \times 10^{-14_{e^2}}$

 $1.98 \times 10^{-14_{e^2}}$

 $1.08 \times 10^{-14_{e^2}}$

Table S1. Potential differences at 0.5 C for charge-discharge process (50% depth of discharge of sample with x = 0.02).