Supporting Information

Design and synthesis of high energy density heterostructure

$Na_{0.7}MnO_2$-$Li_4Mn_5O_{12}$ cathode material for advanced lithium

batteries

Xiujuan Gu,[a,b] Yanjun Cai,[a,b,*] Xiang Yao,[a,b] Hualing Tian,[a,b] Zhi Su,[a,b,*]

[a] College of Chemistry and Chemical Engineering, Xinjiang Normal University,

Urumqi, 830054, Xinjiang, China

[b] Xinjiang Key Laboratory of Energy Storage and Photoelectrocatalytic Materials,

Urumqi, 830054, Xinjiang, China

* Corresponding author. College of Chemistry and Chemical Engineering, Xinjiang

Normal University, Urumqi, 830054, Xinjiang, China.

E-mail address: suzhixj@sina.com (Z. Su).

The collected XRD patterns were analyzed by the Rietveld method using Rietica ver. 1.7.7. In the refinement, the optimized parameters included the background coefficients, zero-shift, peak shape parameters, phase lattice. The figures of merit for the refinement include the goodness-of-fit (c2), Bragg statistical reliability factor (R_B), and the weighted profile factor (R_{wp})

Table S1 Rietveld analysis of as-prepared samples

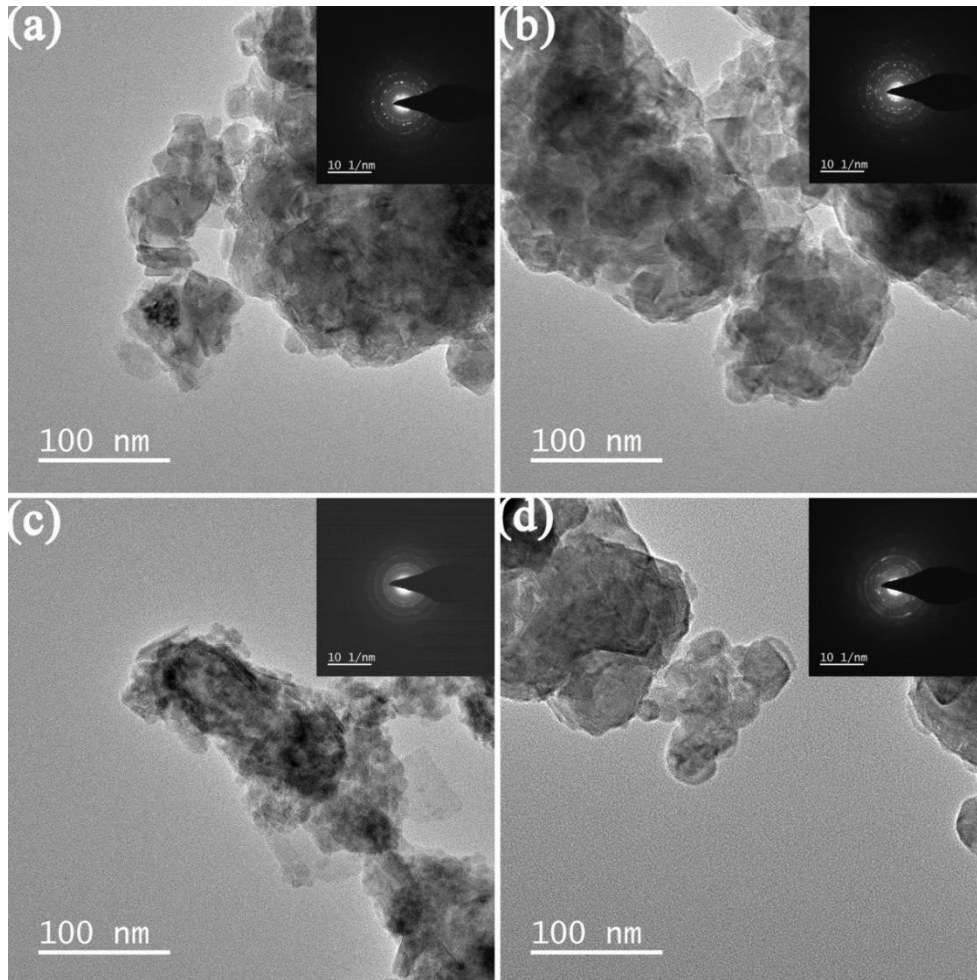| Samples / Phase	NL1	NL2	NL3	NL4	NL5	NL6
$Na_{0.91}MnO_2$	78.19%	85.39%	/	/	/	78.78%
$Na_{0.7}MnO_2$	17.38%	1.41%	60.17%	9.49%	/	21.22%
$Li_4Mn_5O_{12}$	(Mn_3O_4, 4.43%)	13.20%	39.83%	90.51%	100%	/

Fig.S1 TEM of the samples of (a) NL2, (b) NL3, (c) NL5, (d) NL6, insets contain the

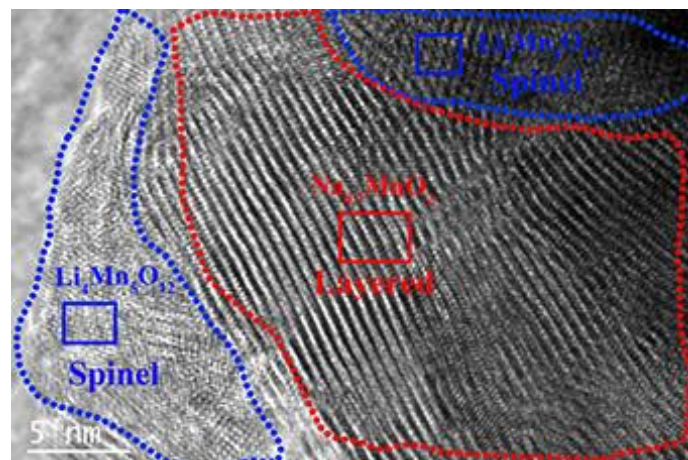corresponding selected area electron diffraction (SAED) images.

Fig.S2 HRTEM of NL3.

Fig.S2 is HRTEM of as-prepared samples of NL3. HR-TEM images further

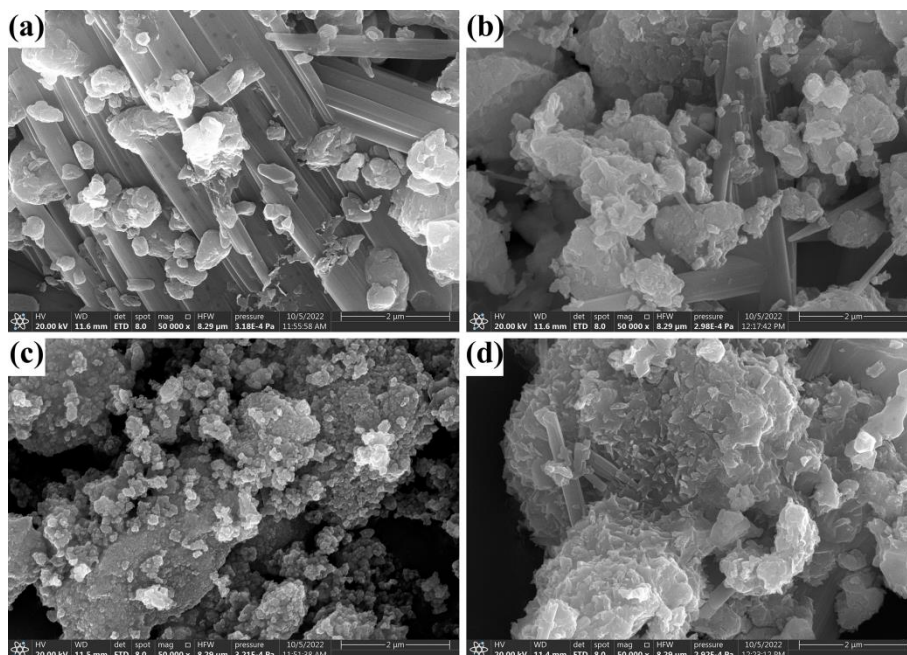confirm the layered-spinel heterostructure was formed.

Fig. S3 SEM images of the as-prepared materials (a) NL2, (b) NL3, (c) NL5, (d) NL6.

The as-prepared samples were tested by scanning electron microscopy, as shown in Fig. S3. It can be observed from the Fig. S3a and Fig. S3b the samples of NL2 and NL3 are composed of layered sheets and particles. The NL5 ($Li_4Mn_5O_{12}$) shows particle morphology and the NL6 shows layered morphology. According to the results of scanning electron microscopy, the samples of NL2 and NL3 have formed heterostructure.
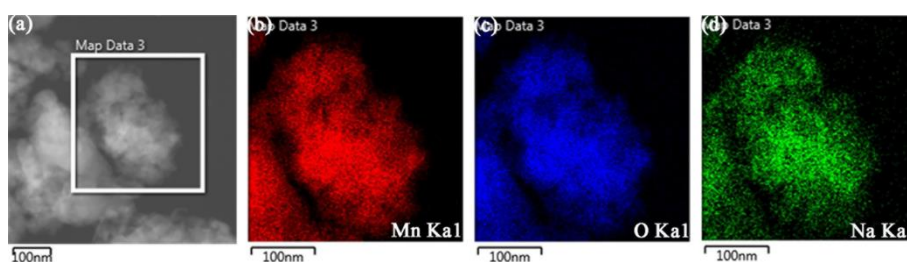

Fig.S4 SEM of NL2(a), and the EDX element mapping of (b) Mn, (c) O (d) Na on NL2.

TableS2 Data of each electrochemical component of as-prepared samples

Samples	W-T	W-P	CPE-T	CPE-P
NL1	4650.00	4.23	0.57	6.05×10^{-5}
NL2	230.00	7.93	0.37	2.44×10^{-5}
NL3	120.50	19.00	0.31	5.90×10^{-5}
NL4	195.30	0.02	0.28	1.55×10^{-5}
NL5	3.50×10^{-5}	2.46×10^{-17}	0.24	3.82×10^{-5}
NL6	157.70	0.11	0.31	1.96×10^{-4}

W_s: Warburg impedance, CPE: constant phase angle element.