Supporting Information

1-(2-cyanoethyl) pyrrole enables excellent battery performance at high temperature via the synergistic effect of lewis base and C≡N functional group

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1 Experimental section

1.1 Sample preparation

The lithium salt lithium hexafluorophosphate ($LiPF_6$) and solvents ethyl carbonate (EC), dimethyl carbonate (DMC), diethyl carbonate (DEC) were provided by Hubei Nuobang company. The electrolyte additive CP and cathode $LiFePO_4$ material (with ~20% carbon coating) were purchased from ALADDIN and Taiyuan Lishiyuan Chemical Company, respectively. The base electrolyte (E0) was prepared by dissolving $LiPF_6$ into the mixed solvent of EC+DMC+DEC (1:1:1, by volume) in a high purity argon-filled glove box with oxygen and water content below 0.1 ppm. The electrolyte containing 1 wt.% CP (E1) was also prepared in the glove box, and the electrolyte with CP additive appeared colorless and transparent without any precipitation after stored.

1.2 Electrochemical test

Cyclic voltammetry (CV) was determined on the CORRTEST electrochemical workstation with a scan rate of 0.1 $mV \cdot s^{-1}$. Electrochemical impedance spectroscopy (EIS) was also performed on the electrochemical workstation at frequencies ranging from 0.1 Hz to 100 kHz with a potential amplitude of 5 mV. A working electrode was prepared from 80 wt.% $LiFePO_4$ powder, 10 wt.% binder (PVDF) and 10 wt.% a conductive agent (carbon black). The batteries were charged and discharged at a constant current with a voltage range of 2.5 to 4.0 V in the

battery charge and discharge workstation (Wuhan Land company). The specific capacity of cathode material was calculated according to its mass load, and the active material on each electrode was about 2.0 $mg \cdot cm^{-2}$.

1.3 Material characterization

The active materials stored in the E0/E1 electrolyte at 80 °C for 48 h were characterized by Fourier Transform infrared spectroscopy (FTIR) and scanning electron microscopy (SEM, JSM-7100F, Japan), respectively. E0/E1 electrolytes stored at the high temperature (80 °C) were tested by NMR (31P and 19F). The recycled electrodes separated with anhydrous DMC in the glove box and dried in the oven at 45 °C for 24 h were investigated using Transmission electron microscopy (TEM, FEI TecnaiG20), X-ray diffraction (XRD, D8ADVANCE, BRUKER), X-ray photoelectron spectroscopy (XPS, Escalab250Xi Thermo Fisher Scientific). Inductively coupled plasma-optical emission spectroscopy (ICP-OES) was used to measure the content of main metals.

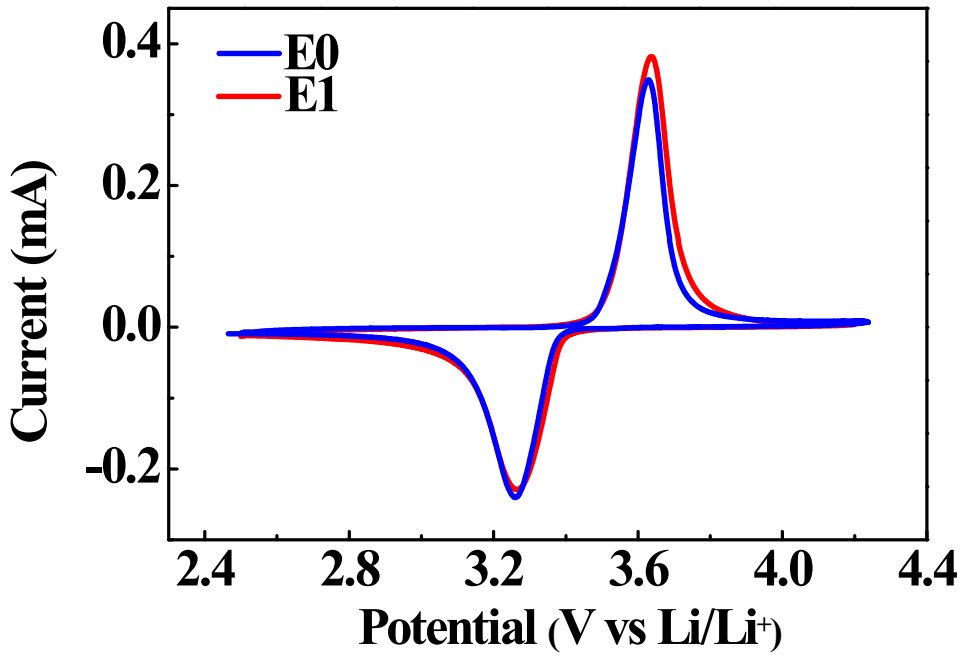

Figure S1 The cyclic voltammetry (CV) curves of batteries in the E0 and E1 electrolytes.

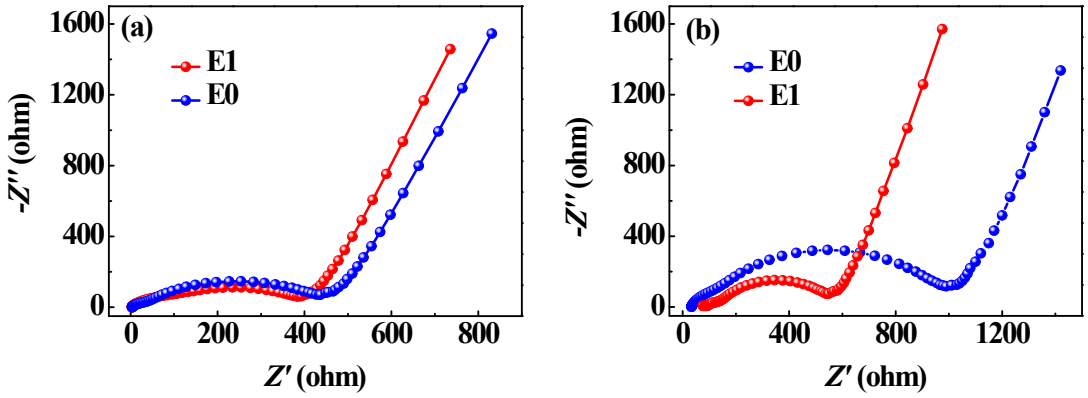

Figure S2 The electrochemistry impedance spectroscopy (EIS) of batteries in the E0 and E1 electrolytes (a) before and (b) after 200 cycles.

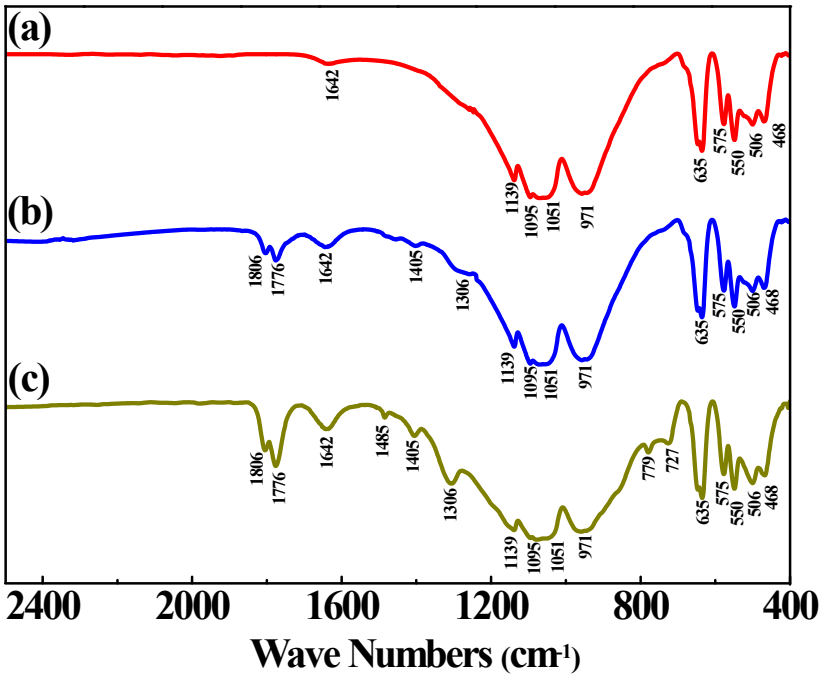

Figure S3 The FTIR spectra of $LiFePO_4$ before and after storage in the different electrolytes: (a) Fresh $LiFePO_4$; $LiFePO_4$ stored at 80 °C in the electrolytes (b) with CP and (c) without CP additive for 48 h.

Table S1 FTIR peak frequencies of $LiFePO_4$ and their assignments after storing in the different electrolytes.

Peak frequencies (cm^{-1})	Peak assignments	Peak frequencies (cm^{-1})	Peak assignments
1806	$\nu_{as}C=O$	1405	ν-C-C
1776	$\nu_{s}C=O$	1306	δ-CH_3
1642	δ-H_2O	779	δ-O-CO_2
1485	δ-CH_2	727	δ-CH

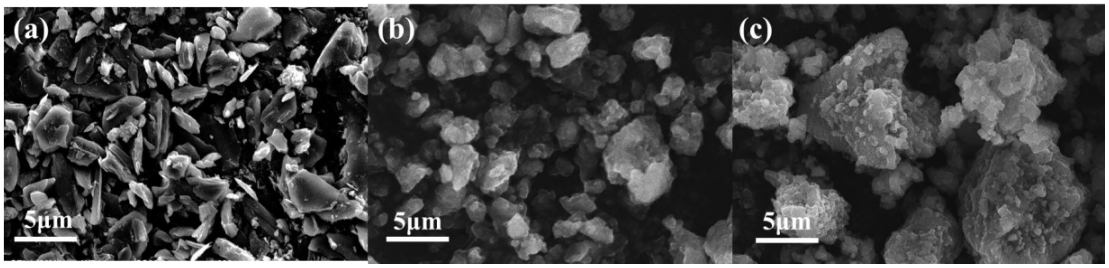

Figure S4 SEM images of the $LiFePO_4$ stored in the electrolytes at 80 °C after 24 h: (a) Fresh $LiFePO_4$; $LiFePO_4$ stored in the electrolytes (b) with CP and (c) without CP additive.

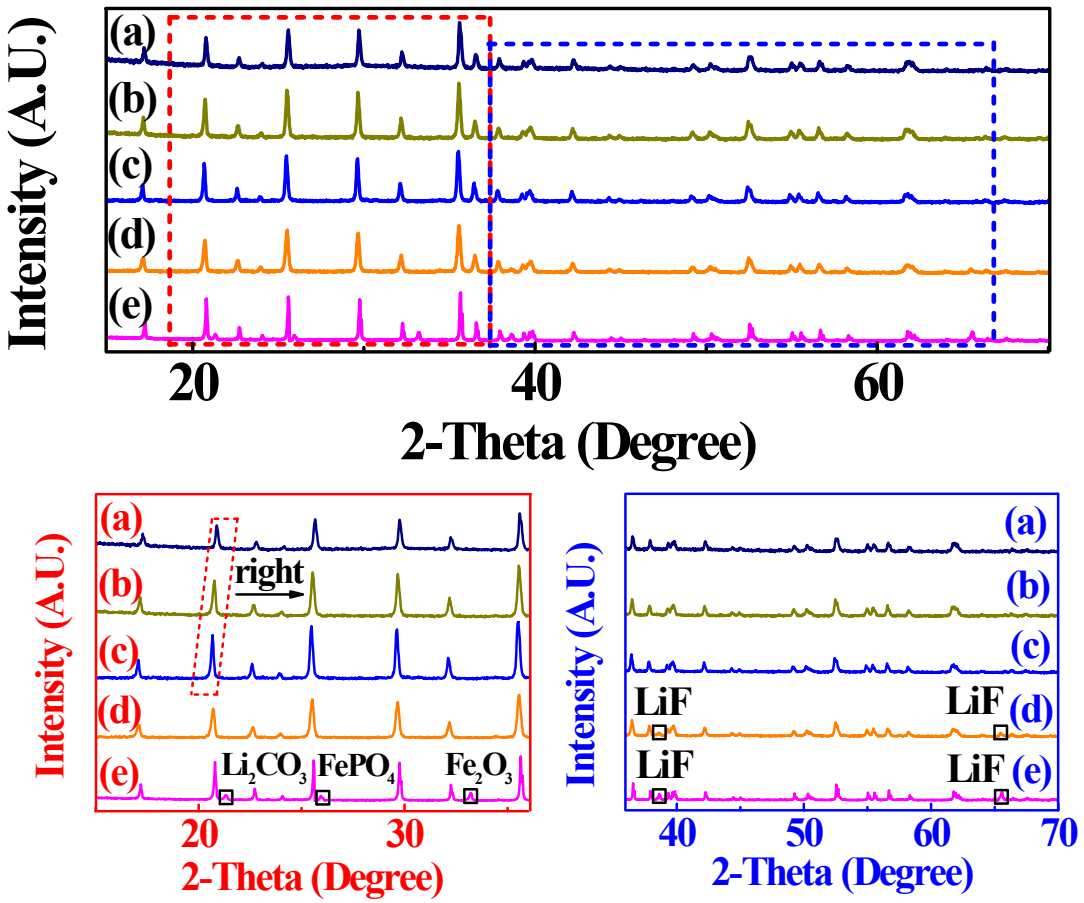

Figure S5 The XRD patterns of the $LiFePO_4$ electrode material: stored at 80 °C in the electrolytes (a) without and (b) with CP additive for 48 h; (c) Fresh $LiFePO_4$; The electrode materials after 200 cycles in the electrolytes (d) with and (e) without CP at 60 °C.

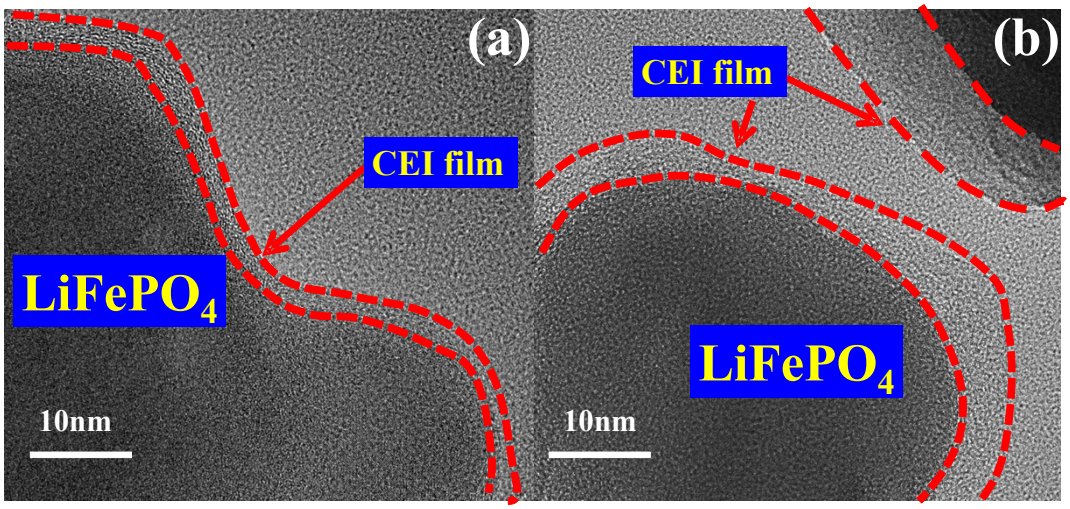

Figure S6 TEM images of $LiFePO_4$ electrodes after 200 cycles at 60 °C: (a) in the E1 electrolyte, and (b) in the E0 electrolyte.

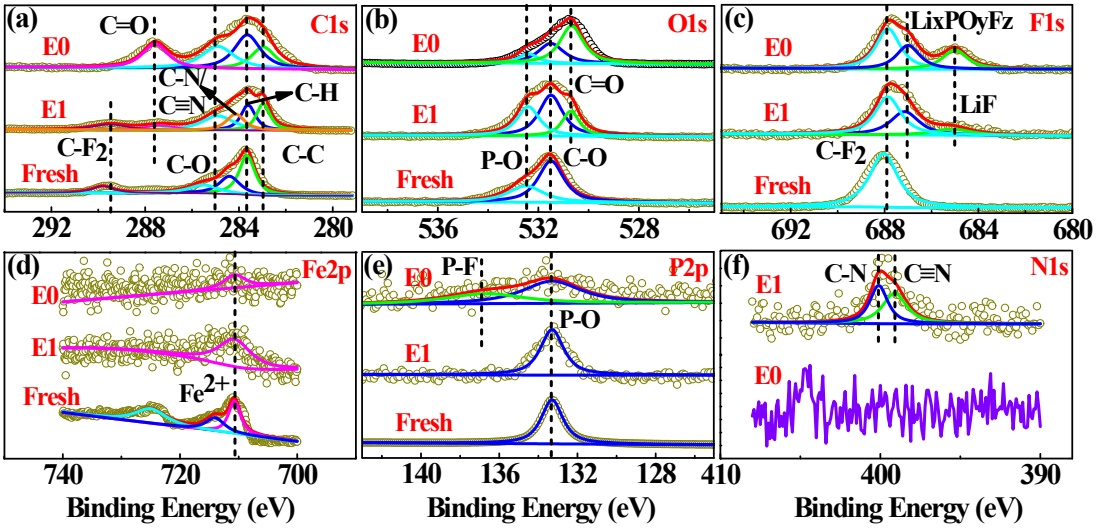

Figure S7 XPS spectra of electrode materials before and after cycling (E1: with 1 wt.% CP; E0: No additive)

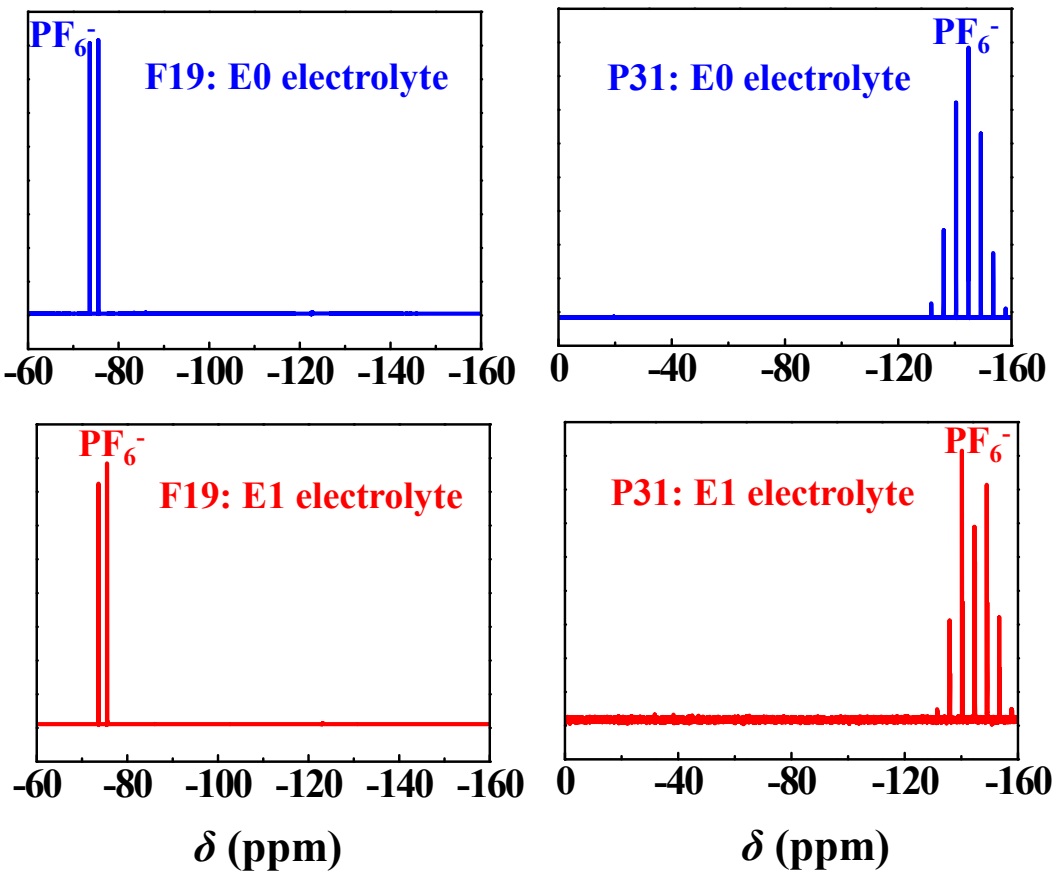

Figure S8 NMR spectra of the electrolytes with and without CP additive before stored at 80 °C for 48 h.

Table S2 The calculated E_{HOMO}/E_{LUMO} and activity degree of DMC, DEC, EC, CP.

Molecules	E_{HOMO} (au.)	E_{LUMO} (au.)	Activity degree*
DMC	-0.37839	-0.00144	++
DEC	-0.38411	-0.01390	+
EC	-0.37276	-0.00271	++
CP	-0.33556	-0.01116	+++

*The more signs of +, the higher of activity degree.