

## **Riga flow batteries**

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a ESI-HRMS spectrum of the Zn(PPi)26-, the peak found at m/z = 606.5764 is assigned to [K5ZnP4O14]-(calcd: 606.5715). b 31P NMR of 3 M K4PPi, 0.3 M and 0.8 M Zn(PPi)26-, respectively. c Raman spectra of 0.8 M ZnCl2, 3 M K4PPi, 0.3 M and 0.8 M Zn(PPi)26-, respectively. d ATR-FTIR spectra of 0.8 M ZnCl2, 3 M K4PPi and 0.8 M Zn(PPi)26-, respectively. e The chelated process of Zn(PPi)26- ions.

3D snapshot of a 0.2 M ZnBr2 system and d 0.2 M ZnCl2-K4PPi (1:3) system obtained from MD simulations. RDFs for b ZnBr2 and e ZnCl2-K4PPi system collected from MD simulations. The optimized molecular structures and corresponding binding energy of c Zn(H2O)62+ and f Zn(PPi)26-. ESP-mapped molecular van der Waals surface of g Zn(H2O)62+ and h Zn(PPi)26-54. i The LUMO and HOMO isosurfaces of Zn(H2O)62+ (left) and Zn(PPi)26- (right), respectively.

a CV curves of 0.1 M Zn(PPi)26- and 0.1 M ZnBr2 solution on a carbon paper electrode at 50 mV s-1, respectively. b Tafel plots for Zn plating/stripping in 0.2 M Zn(PPi)26- solution at 0.1 mV s-1. c CV curves of 0.1 M Zn(PPi)26- at various scan rates ranging from 10 to 50 mV s-1. d Linear relationship between reduction peak current densities (ipc) with square root of the scan rate (n1/2) derived from c.

a GCD profiles of the ZIFBs at 40 mA cm-2 using 0.2 M Zn(PPi)26- negolyte or 0.2 M ZnBr2 negolyte in the first cycle. The charge process ended with a cutoff voltage of 1.9 V and 1.6 V, respectively, while the discharge process ended with a cutoff voltage of 0.2 V. b Rate performance of 0.2 M Zn(PPi)26- based ZIFB with a charging capacity of 20 mAh cm-2 at various current densities, the discharge process ended with a cutoff voltage of 0.2 M Zn(PPi)26- based ZIFB cutoff voltage of 0.2 V. c Cycling performance of 0.2 M Zn(PPi)26- based ZIFB at 40 mA cm-2. The charging capacity was controlled to 20 mAh cm-2, while the discharge process ended with a cutoff voltage of 0.2 V.

Where g is the surface energy of the Zn-electrolyte interface, Vm is the molar volume of Zn, F is Faraday's constant, and i is the NOP.

a, b Laser confocal scanning morphology of Zn deposits obtained by charging 0.2 M Zn2+ negolyte and 0.2 M Zn(PPi)26- negolyte in an unsymmetrical ZIFB with a JCM-D CEM, respectively. c, d SEM morphology of Zn deposits obtained by charging 0.2 M Zn2+ negolyte and Zn(PPi)26- negolyte in a symmetrical ZFB with a filter paper separator, respectively. e PXRD patterns of carbon felts for Zn(PPi)26- negolyte with deposition capacities ranging from 40 to 180 mAh cm-2. f Binding energy of H2O molecule and PPi4- ion on the surface of Zn (101) crystalline plane. g, h The proposed Zn deposition process for Zn2+ negolyte and Zn(PPi)26-



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negolyte, respectively.

Analytical grade Potassium iodide (KI, 99%) and Potassium pyrophosphate (K4PPi, 99%) are purchased from Bide pharmatech Co., Ltd. Other reagents were purchased from Sinopharm Chemical Reagent Co. Ltd. All the reagents were used without further purification.

K4PPi (39.64 g, 120 mmol) was dissolved in 25 mL of deionized water. Then, ZnCl2 (5.452 g, 40 mmol) dissolved in 40 mL of deionized water was added dropwise to the K4PPi solution. The resulting chelated Zn(PPi)26- solution was stirred continuously until the solution became transparent, and then concentrated to 45 mL under reduced pressure at 50 ?C. A low concentration of Zn(PPi)26- solution was obtained by diluting the saturated solution with deionized water.

Cyclic voltammetry (CV) curves and linear scanning voltammetry (LSV) curves were tested using an DH 7001 electrochemical workstation (Jiangsu Donghua Analytical Instrument Co., Ltd.) with a three-electrode system. A graphite rod (3 mm in diameter) and an Ag/AgCl electrode (pre-soaked in 3 M KCl solution) served as the counter electrode and reference electrode, respectively.

A commercial Zn foil (0.2 cm x 1 cm) was used as the working electrode (polished by the sandpaper). CV curves of the different chelated solutions were measured at a scan rate of 0.1 mV s-1. The concentration of Zn2+ is 0.05 mM, and the concentration ratio of PPi4- and Zn2+ ranges from 10:1 to 30:1. In the case of a very slow sweep rate, it can be considered that the Zn(P2O7)m2-4m solution and zinc electrode were kept in dynamic equilibrium, and the equilibrium equation could be described as follows:

The complex-ratio m can be calculated from the Nernst equation described as follows:

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