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High-performance lithium-ion batteries based on polymer/graphene hybrid cathode material

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Organic and carbon-based lithium-ion batteries possess abundant resources, nontoxicity, environmental friendliness, and high performance, and they have been widely studied in the past decades. However, it remains a challenge to construct such batteries with high capacity, high cycling stability, and high conductivity simultaneously. Here, we elaborately design and integrate organic polymer (p-FcPZ) with graphene network to create a hybrid material (p-FcPZ@G) for high-performance lithium-ion batteries (LIBs). The bi-polar polymer p-FcPZ containing multiple redox-active sites endows p-FcPZ@G with both remarkable cycling stability and high capacity. The porous conductive graphene network with a large surface area facilitates rapid ions/ electrons transportation, resulting in superior rate performance. Therefore, the half-cell based on p-FcPZ@G cathode exhibits simultaneously high capacity (~250 mA h g⁻¹ at 50 mA g⁻¹), excellent cycling stability (retention of 99.999% per cycle for 10,000 cycles at 2,000 mA g⁻¹) and superior rate performance. Additionally, the graphene-based full cell assembled with p-FcPZ@G cathode and graphene anode also demonstrates comprehensively high electrochemical performance.

lithium-ion batteries, organic electrode, graphene hybrid material, high performance, full cells

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1 Introduction

Lithium-ion batteries (LIBs) have attracted great interest and been widely used for decades [1-4]. Most traditional LIBs are constructed by inorganic electrode materials, such as LiCoO₂ cathode and graphite anode [5]. However, these lithium transition metal oxides cathode materials usually face challenges of scarce sources, toxicity and pollutions [6]. Therefore, it is essential to find other battery materials with abundant resources, nontoxicity and environmental friendliness to meet the increasing energy demands. From this perspective, organic materials are promising candidates for the future energy storage systems [7–9].

Numerous organic small molecules and polymers electrode materials for LIBs have been extensively studied and exhibited remarkable electrochemical performance [10,11]. Polymers with multiple redox-active moieties and minimized redox-inactive moieties are particularly favorable, and constructing redox-active linkages during polymerization is an effective method to realize balanced high capacity and cycling stability [12]. However, organic/polymer electrode materials generally exhibit low intrinsic conductivity, leading to poor kinetics and limited rate performance [13]. To address this issue, organic/carbon hybrid materials with

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improved conductivity should be delicately designed and fabricated to achieve synergistic benefits for high-performance LIBs [14–16].

Carbon-based materials, such as graphene, typically exhibits high electronic conductivity, making them ideal for rapid transportation of electrons [17,18]. Graphene also has large surface area and multidimensional transportation channels, which is beneficial for rapid diffusion of ions and electrons [19]. Additionally, the chemical stability and mechanical strength of graphene allow it to accommodate large volume variations during the charge/discharge process and further improve the cycling stability [18,19]. Moreover, graphene oxide (GO) or reduced GO (rGO) can bind diverse species and *in-situ* integrate with polymers to construct functional polymer/graphene hybrid materials [14].

With these in mind, we designed and prepared a ferrocenebased polymer/graphene hybrid material (named p-FcPZ@G) with high capacity, cycling stability and conductivity at the same time for LIBs through an in-situ fabrication strategy. Bi-polar polymer (p-FcPZ) with redoxactive C=N linkage can be generated by condensation between 1,1'-ferrocene dicarboxaldhyde (Fc) and 2,5-pyrazinediamine (DAPZ) without sacrificing too much of the capacity. By introducing GO into the polymerization system, we in-situ fabricated hybrid material p-FcPZ@G, which features both a redox-active polymer and a conductive graphene network. Consequently, p-FcPZ@G possesses multiple redox-active moieties and stable porous conductive framework, facilitating reversible electrochemical reaction, rapid ions/electrons diffusion and high stability, resulting in high capacity, excellent cycling stability and superior rate performance simultaneously [20].

As a result, the half-cell based on p-FcPZ@G cathode delivers a high reversible capacity of $\sim 250 \text{ mA h g}^{-1}$ at 50 mA g⁻¹. It also exhibits superior cycling stability with retention of 99.999% per cycle after 10,000 cycles at a high rate of 2,000 mA g⁻¹. The unique two-dimensional structure of graphene enables for storage of lithium ions on both sides, delivering a higher theoretical capacity than that of traditional graphite anode [21]. Because of its higher capacity and conductivity, the graphene-based full cells using p-FcPZ@G cathode and graphene anode are constructed. The graphene full cell exhibits a high capacity of $150.5 \text{ mA h g}^{-1}$ at 2,000 mA g^{-1} with a capacity retention of ~80.0% after 3,000 cycles, demonstrating excellent cycling stability. The graphene full cell also exhibits remarkable rate performance. achieving capacities of 232.2, 205.3, 179.1, 161.5, and 134.1 mÅ h g^{-1} at 100, 200, 500, 1,000, and 2,500 mA g^{-1} , respectively. All of the batteries assembled with our designed p-FcPZ@G hybrid material display remarkable electrochemical performance, revealing that p-FcPZ@G is a superior electrode material that is compatible with various anode systems. This demonstrates that p-FcPZ@G is well designed and our strategy could be effective to obtain battery materials with even better performance in future.

2 Results and discussion

2.1 Material preparation and characterization

The polymer p-FcPZ was prepared by polymerization between 1,1'-ferrocene dicarboxaldhyde (Fc) and 2,5-pyrazinediamine (DAPZ) in a mixed solvent of 1-butanol (BuOH) and 1,2-dichlorobenzene (DCB) (1:1, v/v) with a few drops of acetic acid (AcOH), and the reaction was kept at 120 °C for 72 h (Figure 1a). To in-situ fabricate the hybrid material p-FcPZ@G, Graphene oxide (GO) was mixed with these two monomers prior to polymerization. The successfully introduction of graphene network into p-FcPZ@G was supported by the obvious D and G bands of graphene in the Raman spectrum (Figure 1b) [22,23]. In the Fourier transform infrared spectroscopy (FT-IR) spectrum of p-FcPZ@G (Figure S1, Supporting Information online), a new peak for C=N bonds at $1,580 \text{ cm}^{-1}$ appears, suggesting the generation of C=N linkage during reaction [24]. The disappearance of peaks for -NH2 and -CHO groups of monomers indicates the completion of the polymerization [15,25]. Thermogravimetric analysis (TGA) demonstrates that p-FcPZ@G exhibits high thermal stability with a mass retention of 92.82 wt% at 300 °C under N₂ (Figure 1c). The weight loss below 150 °C could be attributed to the adsorbed solvents, and the oligomers could be removed before 300 °C. The mass decrement above 300 °C is caused by the carbonization of p-FcPZ@G [23]. It was calculated that p-FcPZ@G contains 22.10 wt% of graphene from the TGA results (Figure S2). The scanning electron microscopy (SEM) image of p-FcPZ@G shows porous and fluffy morphology (Figure 1d), while energy dispersive spectroscopy (EDS) images reveal the uniform distribution of C, N, and Fe elements for p-FcPZ@G (Figure S3). Furthermore, p-FcPZ@G displays a Brunauer-Emmett-Teller (BET) surface area of $237.39 \text{ m}^2 \text{g}^{-1}$ with porous features (Figure 1e and Figure S4). X-ray photoelectron spectroscopy (XPS) demonstrates the peaks of C 1s, N 1s, O 1s, and Fe 2p, also supporting the generation of p-FcPZ@G hybrid material (Figure S5).

2.2 Electrochemical performance of half-cells

The redox process of p-FcPZ is depicted in Figure 2a. Upon charging, Fe^{2+} in p-FcPZ is oxidized, generating p-FcPZ-PF₆ with Fe^{3+} [26]. Upon discharging, p-FcPZ-PF₆ is reduced to p-FcPZ, releasing PF₆⁻ first. Then, the C=N linkage of p-FcPZ is further reduced, and N atoms interact with Li⁺ from the electrolyte, forming p-FcPZ-2Li [25]. Both Li⁺ and PF₆⁻ ions could be stored in p-FcPZ@G, so the batteries made of p-FcPZ@G cathode are also Li-ion-based dual-ion batteries



Figure 1 Material synthesis and characterization. (a) Polymerization of p-FcPZ; (b) Raman spectrum of p-FcPZ@G; (c) thermogravimetric analysis (TGA) of p-FcPZ@G under N_2 from 30 to 1,000 °C; (d) SEM image of p-FcPZ@G; (e) Brunauer-Emmett-Teller (BET) isotherm of p-FcPZ@G at 77 K (color online).

[26]. The capacity voltage profiles of p-FcPZ@G half-cell exhibits two voltage platform in the potential range of 3.2-3.8 and 1.8–2.4 V (vs. Li/Li^{+}), indicating the redox reactions of p-FcPZ@G based on p-type and n-type moieties, respectively (Figure 2b). The reversible capacity of ~250 mA h g⁻¹ could be obtained at 50 mA g⁻¹, which is close to its theoretical value, indicating a high utilization of redox-active sites in p-FcPZ@G. The practical capacity contribution from p-type and n-type moieties are 72.4 and 174.7 mA h g^{-1} , respectively. The cyclic voltammetry (CV) profile for p-FcPZ@G cathode at 0.3 mV s⁻¹ also shows two corresponding redox peaks (Figure 2c), which is consistent with capacity voltage profiles. Redox platform/peak at 1.8–2.4 V (vs. Li/Li⁺) is hard to be recognized in Figure 2b, c, possibly due to the large polymer system and porous conductive graphene framework that has capacity contribution in this potential range [15,27,28]. The redox platform/ peak of graphene may overlap and influence the platform/ peak of p-FcPZ@G, which usually happens in some carbon/ organic hybrid materials [27,28].

Additionally, under a larger current density of 2,000 mA g^{-1} , the p-FcPZ@G cathode exhibits a high capacity of 138.5 mA h g^{-1} after activation in the initial several cycles (Figure 2e). A specific capacity of 124.2 mA h g^{-1}

could retained after 10,000 cycles with retention of 99.999% per cycle, demonstrating ultra-high cycling stability. Furthermore, the p-FcPZ@G cathode displays remarkable rate performance with reversible capacities of 238.3, 208.8, 177.6, 155.4, and 130.6 mA h g^{-1} under current densities of 100, 200, 500, 1,000, and 2,500 mA g^{-1} , respectively (Figure 2d). This excellent cycle and rate performance can be attributed to the well-designed hybrid material that contains bipolar polymer active material and porous conductive graphene network.

2.3 Redox mechanism

Density functional theory (DFT) geometry optimization was applied on different redox states for the model dimer compound of p-FcPZ with pyrazine ring edges (2FcPZ) to study the redox mechanism. Figure 3a illustrates the most probable redox process for 2FcPZ. When charging, 2FcPZ is oxidized and combines two PF_6^- from electrolyte, forming 2FcPZ-2PF₆ [29]. During the discharging process, 2FcPZ-2PF₆ undergoes reduction and generates 2FcPZ first, followed by the further reduction of the four imine linkages (C=N) of 2FcPZ, generating 2FcPZ-4Li [25]. The optimized structures of 2FcPZ-2PF₆, 2FcPZ, and 2FcPZ-4Li were obtained through

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Figure 2 Electrochemical behaviors of half-cells based on p-FcPZ@G cathode. (a) Redox process of p-FcPZ; (b) capacity voltage profiles of p-FcPZ@G half-cell in the potential window of 1.2-4.0 V (*vs.* Li/Li⁺) at 50 mA g⁻¹ in the initial three cycles; (c) CV profile of p-FcPZ@G half-cell in the potential window of 1.2-4.0 V (*vs.* Li/Li⁺) at 50 mA g⁻¹ in the initial three cycles; (c) CV profile of p-FcPZ@G half-cell in the potential window of 1.2-4.0 V (*vs.* Li/Li⁺) with the scan rate of 0.3 mV s^{-1} ; (d) rate performance of p-FcPZ@G half-cell at current densities of 100, 200, 500, 1,000, and 2,500 mA g⁻¹, respectively; (e) long-cycle performance of p-FcPZ@G half-cell under 2,000 mA g⁻¹ for 10,000 cycles (color online).



Figure 3 Redox mechanism of 2FcPZ. (a) Redox process of 2FcPZ; (b) the geometry optimization of different redox states for 2FcPZ through DFT calculations (color online).

DFT calculations (Figure 3b). In the oxidized state (2FcPZ-2PF₆), PF_6^- locates near Fe³⁺ after charging. In the reduced state (2FcPZ-4Li), Li⁺ interacts with the nitrogen atoms of C=N linkages after discharging. More details of possible redox process and comprehensive analysis are given in Figure S6.

To confirm the redox process of p-FcPZ@G cathode, an

ex-situ attenuated total reflection (ATR)-IR analysis at different redox states during one charge/discharge cycle was conducted. Figure 4A shows the capacity-voltage profiles of p-FcPZ@G cathode with four different charge/discharge states for *ex-situ* experiments. State "a" represents the initial state of p-FcPZ@G cathode before cycling, while state "b" is the oxidized state, which is obtained after charging to 4.0 V

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Figure 4 (A) Capacity-voltage profiles of p-FcPZ@G cathode with different charge/discharge states for *ex-situ* XPS and ATR-IR measurements. (B) *Ex-situ* ATR-IR spectra of p-FcPZ@G cathode at different charge/discharge states. *Ex-situ* high-resolution XPS spectra of p-FcPZ@G cathode at different charge/discharge states for (C) P 2p, (D) F 1s, and (E) N 1s (color online).

(vs. Li/Li⁺). State "c" corresponds to the reduced state after discharging to 1.2 V (vs. Li/Li⁺), and state "d" is obtained when charged to 3.0 V (vs. Li/Li⁺) again after one cycle, which possesses the same potential as its initial state (state "a").

Ex-situ ATR-IR spectra of p-FcPZ@G cathode at different states display reversible changes during one charge/discharge cycle (Figure 4B). Peaks of PF_6^- at 833 and 555 cm⁻¹ arise when charging (state "b"), and these two peaks decrease after discharging, indicating reversible insertion/de-insertion of PF_6^- into/from p-FcPZ@G cathode [26]. Moreover, when discharged to 1.2 V (vs. Li/Li⁺), a peak attributed to N–Li bond at 1,301 cm⁻¹ appears [30], suggesting the intercalation of Li⁺ at reduced state (state "c"). This peak disappears when charging again (state "d"), indicating ideal reversibility.

To further confirm the redox mechanism of p-FcPZ@G cathode, *ex-situ* XPS spectra were studied. After immersion in the electrolyte (state "a"), the high-resolution P 2p spectrum for the p-FcPZ@G cathode shows peaks of LiPF₆ and PO_x at 137.4 and 134.7 eV, respectively (Figure 4C) [31]. The high-resolution F 1s spectrum exhibits peaks at 688.0 (LiPF₆ and Li_xPF_y) and 685.8 (LiF) eV (Figure 4D) [32,33]. In the high-resolution N 1s spectrum, peaks of C–N and C=N can be observed at 401.0 and 400.0 eV, respectively (Figure 4E) [30]. After charging, the peaks of LiPF₆ in the high-resolution P 2p and F 1s spectra increase, revealing the insertion of PF₆⁻ into the p-FcPZ@G cathode (state "b", Figure

4C, D). When discharging, the two peaks of LiPF₆ decrease, indicating de-insertion of PF₆⁻ from the p-FcPZ@G cathode (state "c"). Additionally, in the high-resolution N 1s spectrum, the peak of N–Li at 399.3 eV appears with the decreased peak of C=N, indicating the reduction of C=N bonds and the interaction between Li⁺ and N atoms (state "c", Figure 4E) [30]. When charged to 3.0 V (vs. Li/Li⁺) (state "d"), the high-resolution spectra of P 2p, F 1s, and N 1s recover to their initial states (state "a"), revealing good reversibility of the redox reaction for p-FcPZ@G cathode [34]. Moreover, *ex-situ* XPS spectra for high-resolution Li 1s and Fe 2p are also examined (Figure S7), showing the reversible insertion of Li⁺ and transformation from Fe²⁺ to Fe³⁺, respectively, which further supports the proposed redox mechanism of p-FcPZ@G cathode.

2.4 Kinetic analysis

Electrochemical impedance spectroscopy (EIS) studies were conducted for half-cell based on p-FcPZ@G cathode before and after cycling (Figure 5a). The as-prepared half-cell made of p-FcPZ@G cathode shows a charge transfer resistance ($R_{\rm CT}$) of 250.85 Ω . After 100 cycles at 2,000 mA g⁻¹, the $R_{\rm CT}$ reduced significantly to 20.15 Ω , suggesting the electrochemical activation process during cycling [12]. The Li⁺ diffusion coefficients ($D_{\rm Li^+}$) of the p-FcPZ@G cathode were obtained through galvanostatic intermittent titration technique (GITT) analysis, and was found to be as high as 2.96×10^{-9} and 2.09×10^{-9} cm² s⁻¹ during the discharge and charge process, respectively (Figure 5b and Figure S8). These ideal $R_{\rm CT}$ and $D_{\rm Li^+}$ values indicate fast ions and electros diffusion, resulting in excellent rate performance [28].

2.5 Electrochemical performance of the full cells

Due to the remarkable electrochemical performance of the half-cells made of p-FcPZ@G cathode, full cells based on p-FcPZ@G cathode are fabricated and investigated. First, a full cell was prepared using p-FcPZ@G cathode and graphite anode, which exhibits similar capacity voltage profiles and CV curves with those of half-cell (Figures S9 and S10), demonstrating the bi-polar feature of our cathode material. The full cell using graphite anode operates in the voltage window of 1.2–4.0 V with average working voltage of 2.4 V. The full cell with graphite anode delivers a capacity of

~250 mA h g⁻¹ at 50 mA g⁻¹, of which ~200 mA h g⁻¹ could be remained after 100 cycles (Figure S11). A reversible capacity of 144.0 mA h g⁻¹ was achieved at 2,000 mA g⁻¹, and a capacity of 114.4 mA h g⁻¹ could be retained after 3,000 cycles (Figure S12). It can be observed that the full cell based on p-FcPZ@G cathode and graphite anode exhibits simultaneously high capacity and cycling stability. Besides, the full cell also shows good rate performance, rate capacity of 200.8, 178.3, 157.2, 145.3, and 129.2 mA h g⁻¹ could be realized under current densities of 100, 200, 500, 1,000, and 2,500 mA g⁻¹, respectively (Figure S13).

Graphene possesses high conductivity and unique layer structure, of which both sides could store Li^+ , delivering a higher theoretical capacity of 744 mA h g⁻¹ (LiC₃) compared with traditional graphite anode (372 mA h g⁻¹, LiC₆) [21]. Therefore, a full cell made of p-FcPZ@G cathode and graphene anode was further fabricated. This graphene full cell contains graphene in both cathode (conductive network and



Figure 5 Kinetics analysis of p-FcPZ@G half-cell. (a) EIS for p-FcPZ@G half-cell from 0.01 to 10,000 Hz with amplitude of 0.05 mV; (b) Li^+ diffusion coefficients (D_{Li^+}) for p-FcPZ@G half-cell through GITT experiment (color online).



Figure 6 Electrochemical behaviors of the graphene full cell based on p-FcPZ@G cathode and graphene anode. (a) Capacity voltage profiles of the graphene full cell in the voltage range of 1.2-4.0 V at 50 mA g⁻¹ in the initial three cycles; (b) rate performance of the graphene full cell at current densities of 100, 200, 500, 1,000, and 2,500 mA g⁻¹, respectively; (c) cycle performance of the graphene full cell under 2,000 mA g⁻¹ for 3,000 cycles (color online).

framework) and anode (active material). The graphene full cell also displays similar capacity voltage profiles with that of half-cell and graphite full cell, indicating that the bi-polar p-FcPZ@G dominates the redox reaction as expected (Figure 6a). The graphene full cell works from 1.2 to 4.0 V with average output voltage of 2.4 V. As anticipated, the graphene full cell exhibits better rate performance than that of the graphite full cell. Specific capacity of 232.2, 205.3, 179.1, 161.5, and 134.1 mA h g^{-1} could be obtained at 100, 200, 500, 1,000, and 2,500 mA g^{-1} , respectively, revealing excellent rate performance (Figure 6b). A high capacity of ~180 mA h g^{-1} could be delivered under 500 mA g^{-1} , and the stable cycle could continue for hundreds of cycles (Figure S15). An initial capacity of 150.5 mA h g^{-1} was achieved under a current of 2,000 mA g^{-1} , of which ~80.0% could be maintained after 3,000 cycles, presenting a capacity retention of 99.993% per cycle, and demonstrating superior cycling stability (Figure 6c).

As a result, both the full cells, made of graphite anode or graphene anode exhibit remarkable electrochemical performance, demonstrating the effectiveness of our well-designed p-FcPZ@G hybrid material and its compatibility with different anode materials.

3 Conclusions

In conclusion, we present and report an organic polymer/ graphene hybrid material by integrating the redox-active bipolar polymer with conductive graphene network. The polymer p-FcPZ possesses multiple redox-active sites and minimized redox-inactive moieties, exhibiting high capacity and cycling stability at the same time. The porous morphology and high conductivity of graphene network facilitate fast diffusion of electrolyte and rapid transportation of ions/ electrons, resulting in excellent rate performance. All the half-cells and full cells incorporating the p-FcPZ@G cathode deliver simultaneously high capacity, high cycling stability, and outstanding rate performance, suggesting the overall remarkable performance and compatibility with different anode systems. These results indicate that our strategy offers a possible strategy for the design and preparation of nextgeneration carbon-based battery materials with comprehensively excellent electrochemical performance.

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Conflict of interest The authors declare no conflict of interest.

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