

Polyquinoneimides as Organic Cathodes for Advanced Potassium-Ion Batteries

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Abstract: Due to abundant raw materials, low cost and high theoretical energy density, organic potassium ion batteries (OPIBs) have attracted more and more attention from researchers as next promising battery system. However, some challenges still need to be solved. For example, most small-molecule organic electrode materials faced serious dissolution problem in liquid electrolytes, resulting in poor cycle stability. In particular, there are relatively few reports on the current cathodes with really excellent comprehensive performance (such as: high specific capacity, high working voltage, long life-span and excellent rate performance) for OPIBs. In this article, two polyanthraquinoneimide polymers were successfully synthesized and used as organic cathode materials for potassium ion batteries. Attributed to the large molecular structure of the polymer, PPAQ and PNAQ both were successful to be insoluble in the electrolyte (1 mol/L KPF₆ in DME). After comparison, it was found that PNAQ exhibited better comprehensive electrochemical performance than PPAQ. At a current density of 100 mA/g, PNAQ achieved a stable discharge capacity of 138 mAh/g for 200 cycles and the retention rate was as high as 92%. At the same time, after 750 cycles at a high current density of 800 mA/g, the specific capacity of PNAQ still remained at 71 mAh/g with an average attenuation of only 0.03 mAh/g per cycle. Through an appropriate design process for the molecular structure, we proved that organic polymers could become excellent cathodes for potassium ion batteries.

Keywords: Polyquinoneimide, Organic Cathodes, Potassium-Ion Batteries

1. Introduction

In the past decades, lithium-ion batteries (LIBs) have developed rapidly due to their excellent energy density and long cycle life, occupying a dominant position in the power supply field of portable electronic devices, electric vehicles and other large energy storage devices [1, 2]. However, the scarcity, uneven distribution and increasing demand of lithium mineral resources have greatly hindered the application of lithium-ion batteries in large-scale electrical energy storage [3, 4]. Therefore, researchers were paying more and more attention to the development of alternative battery systems using abundant and low-cost materials, including sodium ion batteries (SIBs) and potassium ion batteries (PIBs) [5-8]. Obviously, the content of potassium in the crust was more abundant than which of lithium (2.09 wt% vs. 0.0017 wt%), closing to sodium (2.36 wt%) [9]. And it was worth noting that the redox potential of K⁺/K (-2.93 V vs.

NHE) was close to which of Li⁺/Li (-3.04 V), 200 mV lower than the Na counterpart (-2.71 V) [10]. Simultaneously, the weaker Lewis acidity of K⁺ among alkaline ions was in favor of the larger transfer number and higher mobility of K⁺, which offer faster kinetics for PIBs [11, 12]. Therefore, PIBs could be expected as a promising next-generation battery technology.

However, due to the large ion radius of potassium ions, the large-scale application of potassium ion batteries still faced serious challenges [13]. Fortunately, based on large void space between organic molecules, low-cost and environmental-friendly organic electrode materials have been proven by different researchers to reversibly store potassium ions with a large ion radius [14, 15]. Compared with inorganic materials, this was a clear advantage. For example, in 2015, 3,4,9,10-perylene-tetracarboxylic dianhydride (PTCDA) was reported for the first time as the first organic cathode for organic potassium ion batteries (OPIBs), and

possessed a theoretical specific capacity of 131 mAh/g [16]. In 2017, potassium terephthalate (K_2TP) and potassium 2,5-pyridinedicarboxylate (K_2PC) were first studied as the first organic anode for PIBs, expressing obvious and reversible charging and discharging platforms [17]. Meanwhile, the reported two anode materials could complete average discharge capacity of 181 mAh/g and 190 mAh/g for 100 cycles, respectively. Subsequently, several organic anodes (such as: K_4PTC [18], K_2BPDC [19], ADAPTS [20], vitamin K [21], etc.) and several organic cathodes (such as: PAQS [22], PPTS [23], PTCDI [24], PI [25], AQDS [26], etc.) were also reported for PIBs. Although good research progress has been seen, the commercial application of OPIBs was still in the early research stage, and there were some key electrochemical data parameters that need to be improved (such as: high specific capacity, high working voltage, long life-span and excellent rate performance) [27]. At present, the application of organic electrode materials in PIBs had an obvious limitation: most small-molecule organic electrode materials faced serious dissolution problems in liquid electrolytes, resulting in poor cycle stability. In particular, there are relatively few reports on the current cathodes with really excellent comprehensive performance for OPIBs [28].

In this article, we designed an appropriate molecular structure (acid anhydride amidation), based on 3,4,9,10-perylene-tetracarboxylic dianhydride (PTCDA), 1,4,5,8-naphthalenetetracarboxylic anhydride (NTCDA) and 1,4-diaminoanthraquinone (NH_2 -AQ- NH_2) monomer, successfully synthesized two organic polymers. The two polyanthraquinone imides were named PPAQ and PNAQ, respectively, as new cathode materials for PIBs. Due to the increase in molecular weight, PPAQ and PNAQ exhibited very insoluble properties for liquid electrolytes (1 mol/L KPF_6 in 1,2-dimethoxyethane (DME)). More importantly, each component unit (PTCDI, NTCDI and AQ) could express redox activity, endowing a high theoretical specific capacity (PPAQ: 180 mAh/g, PNAQ: 228 mAh/g). Subsequently, we explored the electrochemical properties of the two polymers and found that PNAQ had better comprehensive electrochemical properties. In the potassium-ion half-cell, PNAQ could achieve an ultra-stable average discharge specific capacity of 144 mAh/g for 200 cycles at a current density of 100 mA/g using 1 mol/L KPF_6 in DME as the electrolyte. Excitingly, under the high current density of 800 mA/g, PNAQ still exhibited a long life-span of 750 cycles and could maintain a high discharge specific capacity of 71 mAh/g. As far as we know, among all the cathode materials for organic potassium ion batteries that have been reported so far, the comprehensive electrochemical performance of PNAQ was relatively excellent.

2. Experimental Section

2.1. Synthesis of PPAQ

1,4-Diaminoanthraquinone (NH_2 -AQ- NH_2) (0.952 g, 4 mmol, Aladdin, 95%), 3,4,9,10-perylene-tetracarboxylic

dianhydride (PTCDA) (1.568 g, 4 mmol, Aladdin, 98%) and imidazole (50 mL, Aladdin, 99%) was added into 250 mL single mouth bottle, first magnetically stirred for 30 minutes, then sonicated for 30 minutes, then added zinc acetate ($Zn(OAc)_2$, 0.5 g, Aladdin, 96%). The obtained mixture was refluxed with magnetic stirring at 160°C for 3 days under the protection of a nitrogen atmosphere. Subsequently, the mixture was filtered for the precipitate and washed 5 times with water at room temperature. After drying at 110°C for 24 hours, the precipitated product was extracted by the Soxhlet's apparatus with the sequent solvents of CH_2Cl_2 , tetrahydrofuran (THF) and N,N-dimethylformamide (DMF), respectively. Each extraction process was operated for 3-5 days. Finally, 1.477 g of black product (PPAQ) was collected, yield: 62%.

2.2. Synthesis of PNAQ

1,4-Diaminoanthraquinone (NH_2 -AQ- NH_2) (0.952 g, 4 mmol, Aladdin, 95%), 1,4,5,8-Naphthalenetetracarboxylic anhydride (NTCDA) (1.072 g, 4 mmol, Aladdin, 98%) and imidazole (50 mL, Aladdin, 99%) was added into 250 mL single mouth bottle, then magnetically stirred for 30 minutes, then sonicated for 30 minutes, then added zinc acetate ($Zn(OAc)_2$, 0.5 g, Aladdin, 96%). The obtained mixture was refluxed with magnetic stirring at 160°C for 3 days under the protection of a nitrogen atmosphere. Subsequently, the mixture was filtered for the precipitate and washed 5 times with water at room temperature. After drying at 110°C for 24 hours, the precipitated product was extracted by the Soxhlet's apparatus with the sequent solvents of CH_2Cl_2 , tetrahydrofuran (THF) and N,N-dimethylformamide (DMF), respectively. Each extraction process was operated for 3-5 days. Finally, 2.09 g of black product (PNAQ) was collected, yield: 90%.

2.3. Materials Characterizations

The morphology images of the samples were collected by the field-emission scanning electron microscope (FE-SEM, Hitachi, S3400N). Energy dispersive X-Ray spectrometer (EDS) images were recorded on OXFORD ULTIM Max65.

2.4. Electrochemical Measurements

The PPAQ or PNAQ cathodes were prepared by grinding and stirring PPAQ or PNAQ (60wt%), conductive carbon additive (Ketjen Black, 30wt%) and LA133 (10wt%) for at least 45 minutes to obtain the mixture slurry. The mixture slurry was coated on a clean aluminum foil, then dried in the air at 110°C, and then punched to obtain electrode discs ($d=12\text{mm}$). The prepared electrode was dried in a vacuum oven at 110°C for 12 hours. For the PPAQ or PNAQ cathodes, the loading mass of the active material on the aluminum foil was about 2~3 mg/cm^2 . The electrolyte system was 1 mol/L KPF_6 in DME. The separate was Whatman's glass microfiber diaphragm (GF/F). In the half cells (CR2032), the counter electrode was a potassium foil, which was cut from a potassium metal block (Chengdu Kelong Inc.). These

batteries were assembled in a glove box filled with argon (a gas purity of up to 99.9999%). Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were tested using CHI instrument electrochemical workstation (650E).

The constant current charge and discharge performance was collected by the cell equipment (CT2001A, Wuhan LAND Electronic Co.) at a temperature of 25°C [29].

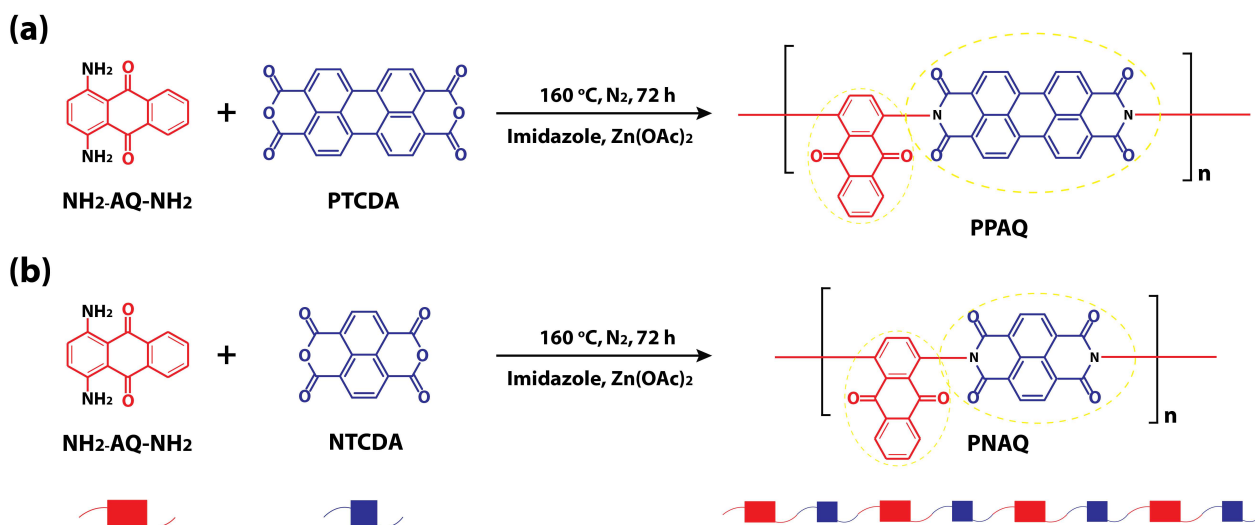


Figure 1. The synthetic routes for (a) PPAQ and (b) PNAQ.

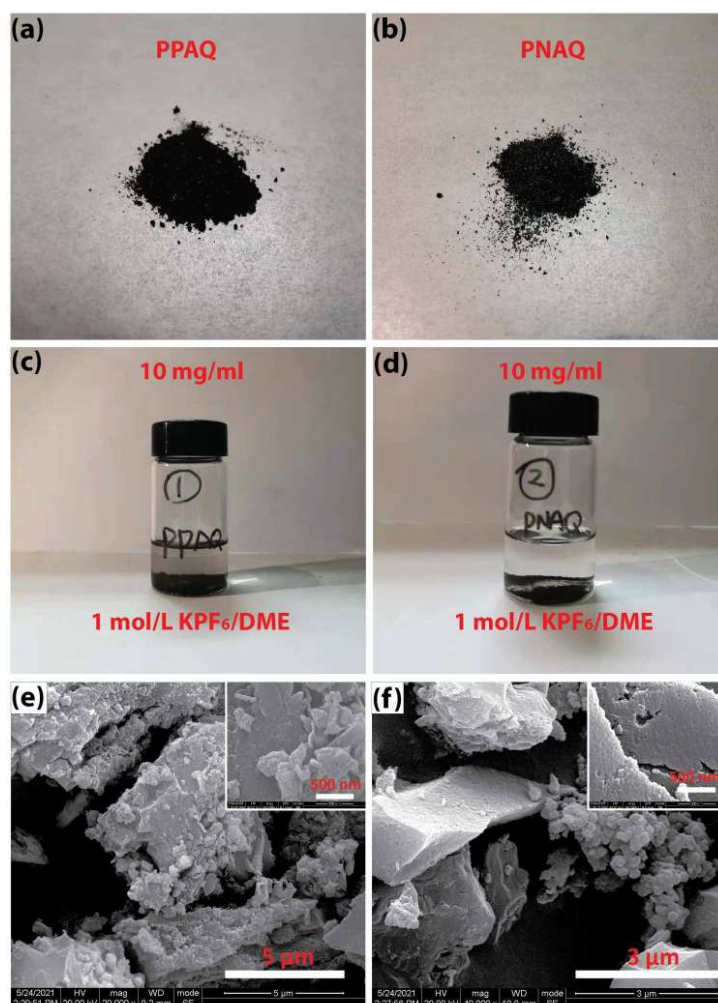


Figure 2. Digital photos of (a) PPAQ and (b) PNAQ; The solubility tests for (c) PPAQ and (d) PNAQ in 1 mol/L KPF₆/DME electrolyte. The SEM images for (e) PPAQ solid and (f) PNAQ solid.

3. Result and Discussion

3.1. Synthesis and Solubility Test

Figure 1 showed the synthesis processes of PPAQ and PNAQ. As shown in Figure 1a, $\text{NH}_2\text{-AQ-NH}_2$ could react with PTCDA to obtain polymer PPAQ through a simple one-step reaction under a relatively mild condition at 160°C . It should be emphasized that the reaction between $\text{NH}_2\text{-AQ-NH}_2$ and PTCDA was relatively difficult compared with 1,6-diaminoanthraquinone. The reason was that the steric hindrance between $\text{NH}_2\text{-AQ-NH}_2$ and the carbonyl group of PTCDA prevented the nucleophilic attack of the amino group of $\text{NH}_2\text{-AQ-NH}_2$ on the carbonyl dianhydride. Figure 2b showed the synthetic route of PNAQ. We used $\text{NH}_2\text{-AQ-NH}_2$ and NTCDA (derivative of PTCDA) to synthesize PNAQ polymer in one step under the same reaction conditions as PPAQ. The obtained products both were extracted by the Soxhlet's apparatus with the sequent solvents of CH_2Cl_2 , tetrahydrofuran (THF) and N,N -dimethylformamide (DMF), respectively. Meanwhile, each extraction process was operated for 3-5 days. Notably,

PNAQ and PPAQ had similar structural properties. The difference was that NTCDA, one of the monomers of PNAQ, had a lower molecular weight than PTCDA (one of the monomers of PPAQ), resulting in a better solubility and being easier to participate in the reaction. Therefore, PNAQ (180 mAh/g) possessed a higher theoretical specific capacity than PPAQ (228 mAh/g). Meanwhile, PNAQ (90%) could get a higher yield than PPAQ (62%) for a refined synthesis process. As shown in Figures 2a and 2b, PPAQ and PNAQ both were black solids. PPAQ and PNAQ solubility tests also were implemented. As shown in Figures 2c and 2d, the two polymers were immersed in the commonly used electrolyte (1 mol/L KPF_6/DME) for 1 week, and the two electrolytes were still clear and transparent, which proved that PPAQ and PNAQ were insoluble in the electrolyte, suggesting that the capacity decay caused by dissolution could be prevented. The micro morphology of the raw materials PPAQ and PNAQ was also characterized by SEM. As shown in Figures 2e and 2f, the particle size of PPAQ was about $5\mu\text{m}$, while the particle size of PNAQ was about $3\mu\text{m}$. In addition, the surface of PNAQ was flatter than that of PPAQ.

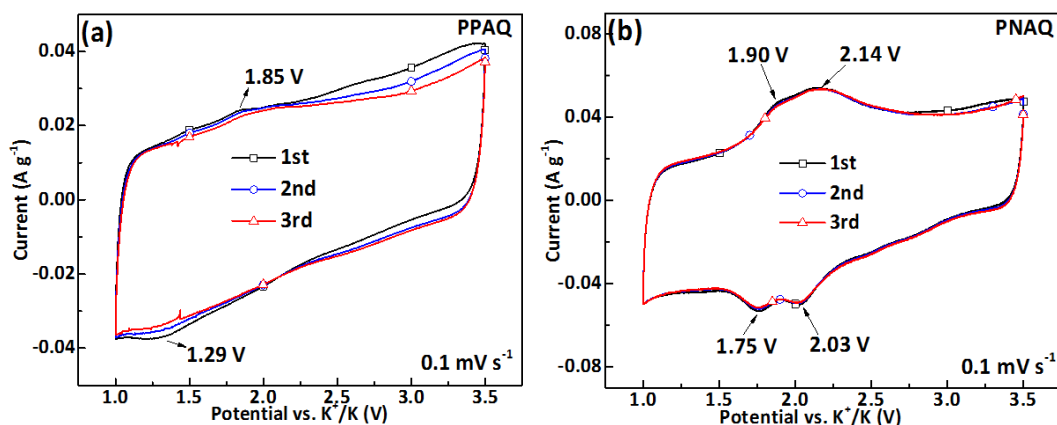


Figure 3. The multiple CV curves of (a) PPAQ and (b) PNAQ. Scan rate was 0.1 mV s^{-1} and the voltage window was 1.0-3.5 V vs. K^+/K .

3.2. Redox Mechanism

In order to investigate the redox mechanism of PPAQ and PNAQ, the related CV test was carried out in the potassium ion half-cell. In the prepared half-cell, active material (PPAQ or PNAQ, 60%); conductive additive (Ketjen Black, 30%); binder (LA133, 10%) was coated on a 12mm diameter aluminum foil. The neat loading mass on foil was about $2\sim 3\text{ mg cm}^{-2}$ for PPAQ or PNAQ cathodes. The electrolyte system was 1 mol/L KPF_6 in DME and metal potassium was used as the anode (see the experimental section for the detailed preparation process). The CV scan curves of the first three cycles of PPAQ were shown in Figure 3a. PPAQ showed reversible a redox mechanism, but the redox peak was very weak, which mean no obvious voltage platform for constant current charge and discharge process. In Figure 3b, the first three cycles of the CV scan test of PNAQ also were carried out, and the scan rate was 0.1 mV/s . Excitingly, PNAQ exhibited the expected very clear redox behavior.

During the reduction scan for the first cycles, two very obvious reduction peaks appeared, located at 2.03 and 1.75 V (vs. K^+/K). Meanwhile, when back to the oxidation scan, two clear oxidation peaks appeared and located at 1.90 and 2.14V, respectively. In the subsequent scans, both the oxidation peak and the reduction peak were very stable. In addition, the scan curves almost overlapped, which mean the expected reversible redox activity for PNAQ. Based on the above CV test results, the redox mechanism of PPAQ with a maximum of 4 electrons (only two monomers were calculated) was shown in Figure 4a, endowing the theoretical specific capacity of 180 mAh/g for PPAQ. Meanwhile, the PNAQ cathode also showed a redox mechanism with a maximum of 4 electrons (based on only two monomers) in PIBs. As shown in Figure 4b, PNAQ possessed a theoretical specific capacity as high as 228 mAh/g.

In order to further observe the redox mechanism of PNAQ, the two-dimensional EDS spectrum for the reduced PNAQ was shown in Figure 5. It could be observed through the EDS

test that when the PNAQ cathode discharged to 1.0 V, K^+ could be evenly intercalate into the PNAQ electrode, which further proved the potassium-ion storage mechanism of PNAQ.

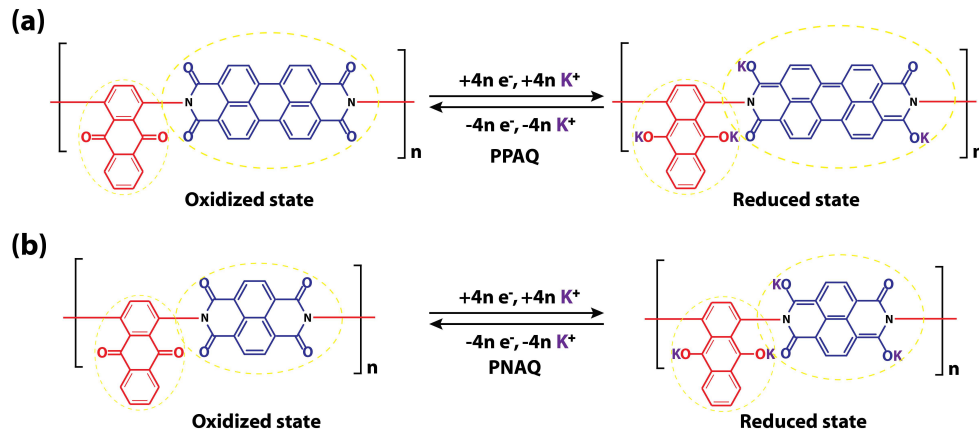


Figure 4. The proposed redox mechanisms for (a) PPAQ and (b) PNAQ in PIBs.

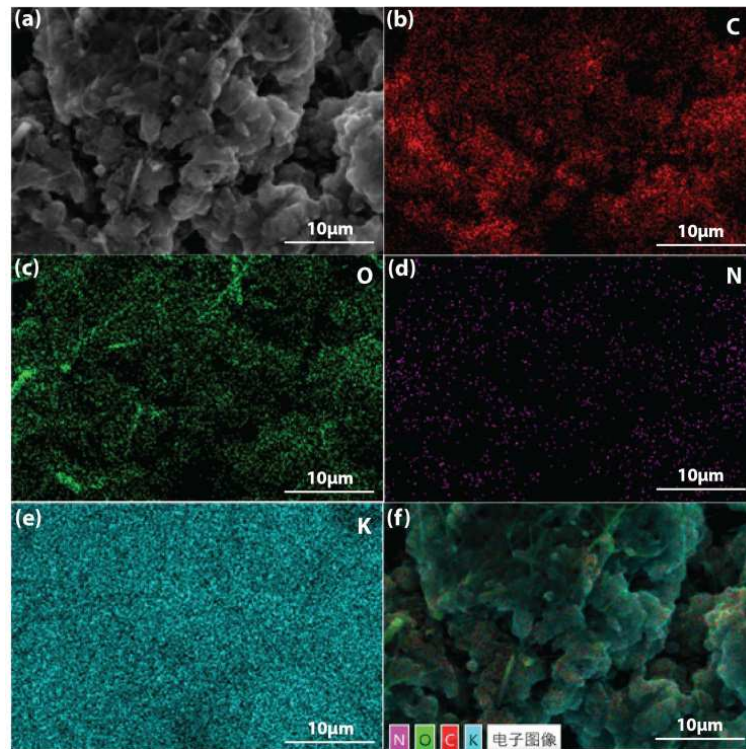
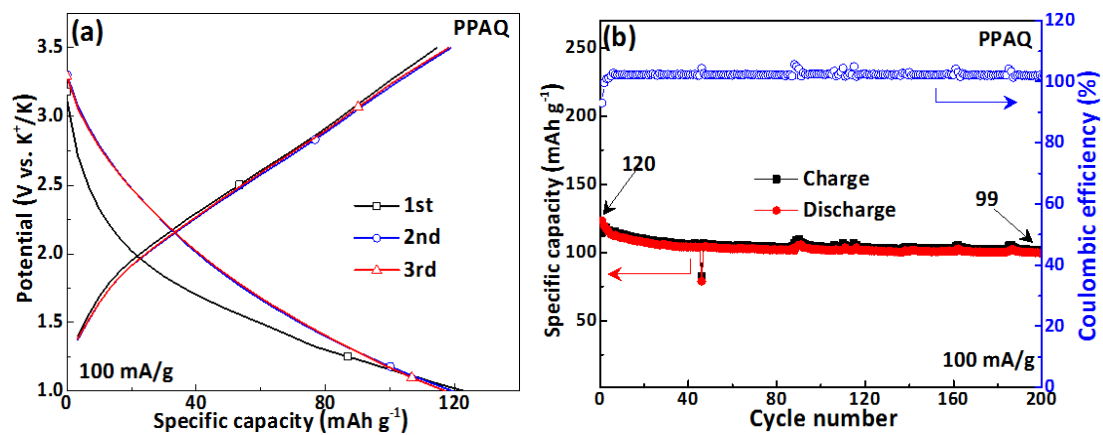


Figure 5. The (a) SEM image and (b-e) energy dispersive X-Ray spectrometer (EDS) images of C, O, K elements for PPAQ electrode after discharged to 1.0 V.



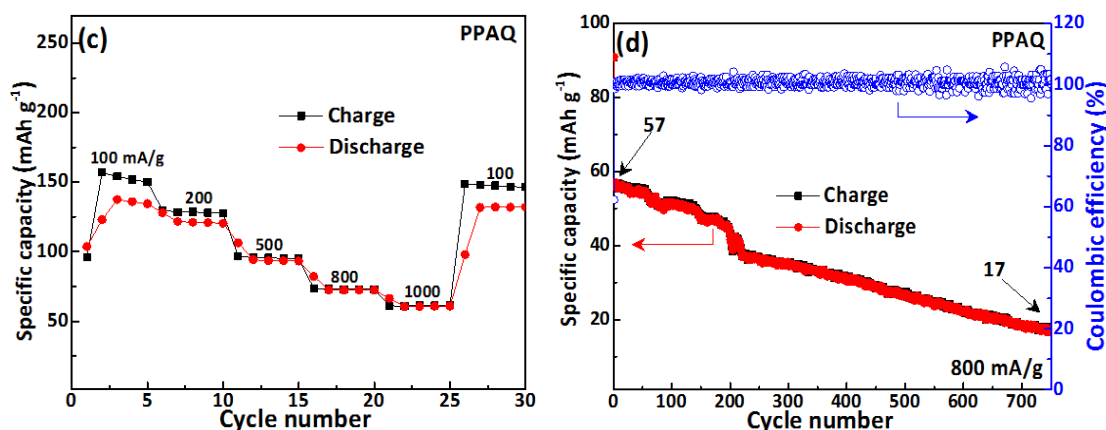


Figure 6. (a) The charge-discharge curves for PPAQ; (b) The long-cycle profile for PPAQ at 100 mA g⁻¹; (c) The rate performance for PPAQ; (d) The long-cycle profile for PPAQ at 800 mA g⁻¹.

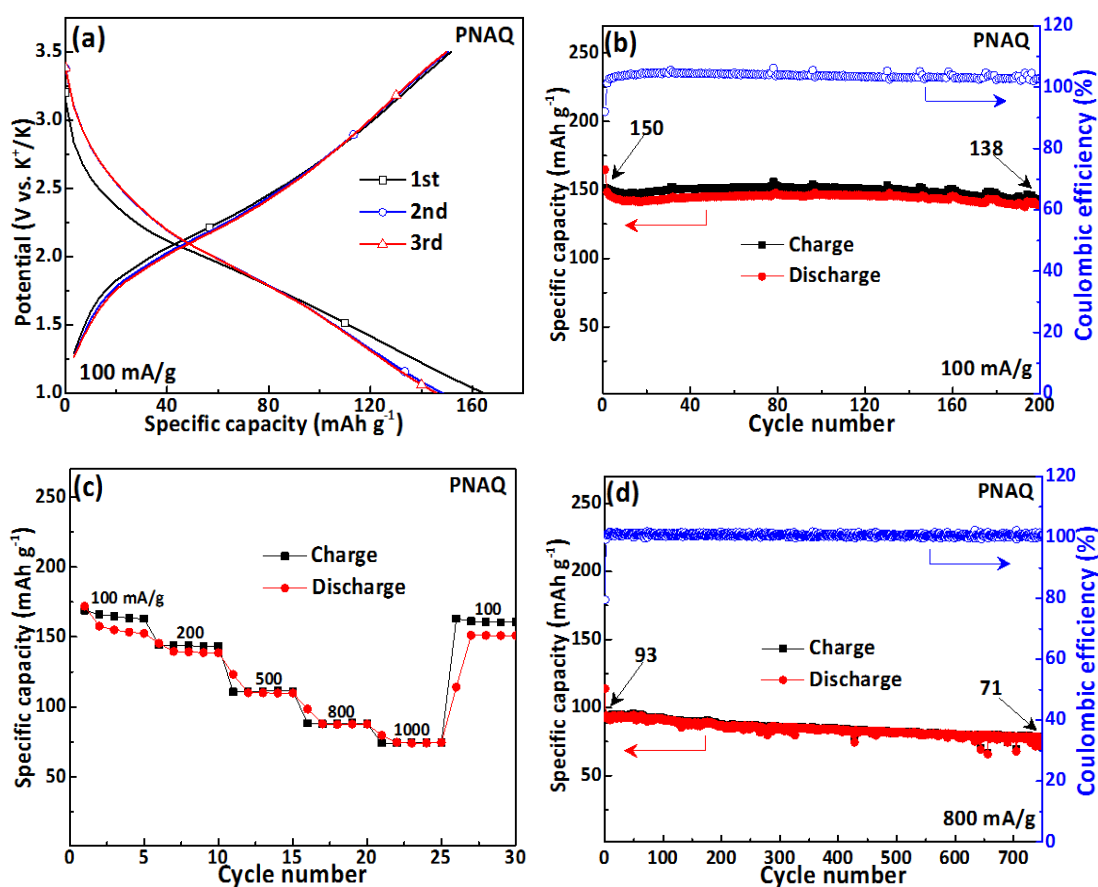


Figure 7. (a) The charge-discharge curves for PNAQ; (b) The long-cycle profile for PNAQ at 100 mA g⁻¹; (c) The rate performance for PNAQ; (d) The long-cycle profile for PNAQ at 800 mA g⁻¹.

3.3. Electrochemical Performance

After verifying the redox mechanism of the two polymers PPAQ and PNAQ, the electrochemical performance was initially evaluated in K-ion half cells. As shown in Figure 6a, PPAQ exhibited the obvious discharge slope at the range of 1.0–3.5 V in the 1st cycle. Meanwhile, PPAQ showed a specific discharge capacity of 120 mAh/g with initial Coulomb efficiency (ICE) of 93% in the first cycle. The cycle profile for PPAQ was shown in Figure 6b. PPAQ

could achieve a stable cycle of 200 cycles with the capacity retention of 82.5% and the coulombic efficiency of 100% at a current density of 100 mA/g. The rate performance of PPAQ was shown in Figure 6c. At a high current density (800/1000 mA/g), PPAQ could achieve a specific discharge capacity of 73/61 mAh/g. The long-cycle profile for PPAQ was shown in Figure 6d. After 750 cycles, the specific capacity of PPAQ decayed from 57 mAh/g to 17 mAh/g at a high current density of 800 mA/g, showing poor long-cycle stability.

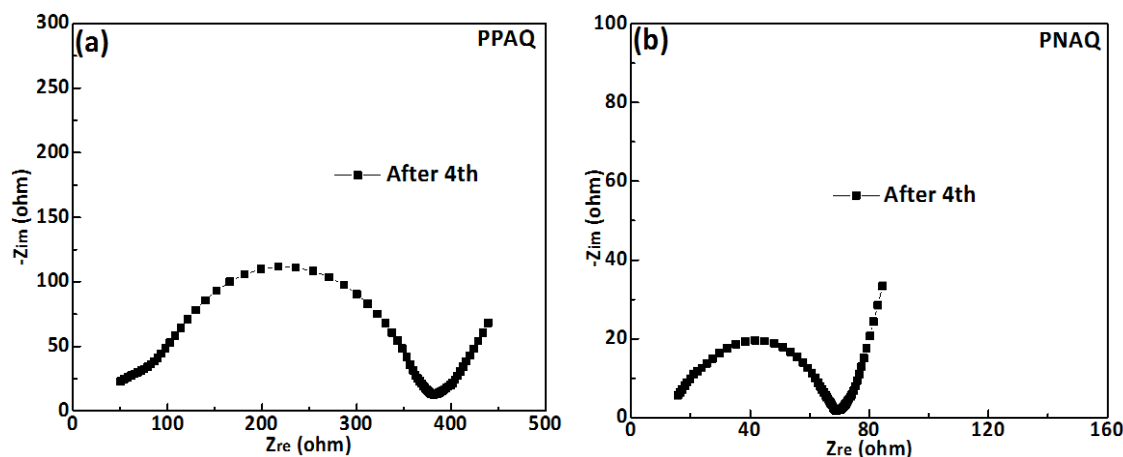


Figure 8. The EIS data of (a) PPAQ and (b) PNAQ electrodes after 4th cycles.

Subsequently, we continued to test the electrochemical performance of PNAQ in PIBs. As shown in Figure 7a, PNAQ also exhibited the obvious discharge slope at the range of 1.0-3.5 V in the 1st cycle. The difference was that the slope of the discharge curve was smaller and closer to the discharge plateau, corresponding to the redox peaks of the CV resulting. Excitedly, PNAQ could achieve a high specific capacity of 162 mAh/g in the first week, with an ICE of 92% in the first cycle. In addition, the cycle profile for PNAQ was shown in Figure 7b. At a current density of 100 mA/g, PNAQ could achieve a very stable cycle for 200 cycles and remained at 138 mAh/g with the retention rate of 92%. The rate performance of PNAQ was shown in Figure 7c. At high current density (800/1000 mA/g), PNAQ still finished a high specific capacity of 95/88 mAh/g. Meanwhile, when the current density returned to 100 mA/g, the specific capacity for PNAQ also recovered to 151 mAh/g. The long-cycle profile of PNAQ was shown in Figure 7d. After 750 cycles at a high current density of 800 mA/g, the specific capacity of PNAQ still remained at 71 mAh/g with an average attenuation of only 0.03 mAh/g per cycle. Compared with PPAQ cathode, PNAQ not only showed higher specific capacity, but also possessed super stability. As confirmed by the EIS test in Figure 8, the charge transfer resistance (R_{ct} : 55 Ω) for PNAQ cathode became smaller than that (R_{ct} : 330 Ω) for PPAQ after 4 cycles. As far as we know, the comprehensive electrochemical performance of PNAQ was excellent among the reported cathodes for organic potassium ion batteries (Table 1).

Table 1. A summary for non-aqueous OPIBs for the reported.

Cathode	Voltage ^a	Capacity retention ^b	Ref.
PNAQ	1.0-3.5	76%@750 th cycle (800 mA/g)	This work
PTCDA	1.5-3.5	77%@200 th cycle (50 mA/g)	[15]
PI	1.4-3.5	83%@500 th cycle (100 mA/g)	[25]
PTCDI	1.4-3.8	89%@600 th cycle (4 A/g)	[24]
PIM	1.5-3.4	78%@1000 th cycle (2 A/g)	[30]
PQ-1,5	1.2-3.2	91%@200 th cycle (250 mA/g)	[31]

^a The unit is V.

^b The ratio of the last-cycle capacity value to the peak value (current density).

4. Conclusion

In summary, the two organic polymers PPAQ and PNAQ were synthesized by a simple one-step reaction and used as new organic cathodes for K-ion batteries. PPAQ and PNAQ owned the high theoretical specific capacity of 180 mAh/g and 228 mAh/g, respectively. Due to the large molecular structure of the polymer, PPAQ and PNAQ both were successful to be insoluble in the electrolyte (1 mol/L KPF₆ in DME), suggesting the excellent cycle stability. After comparison, it was found that PNAQ had better comprehensive electrochemical performance than PPAQ. At a low current density of 100 mA/g, PNAQ could achieve a very stable cycle for 200 cycles and remained at 138 mAh/g with the retention rate of 92%. Meanwhile, the specific capacity of PNAQ still remained at 71 mAh/g for 750 cycles at a high current density of 800 mA/g, with an average attenuation of only 0.03 mAh/g per cycle. Through an appropriate design process for the molecular structure, we proved that organic polymers could become excellent cathodes for potassium ion batteries.

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