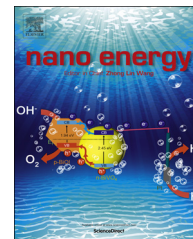


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COMMUNICATION

Organic electrode for non-aqueous potassium-ion batteries



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Abstract

Potassium-ion (K-ion) batteries can be an attractive alternative for grid-scale energy storage due to the abundance of K resources. For the first time, we designed and fabricated a non-aqueous K-ion battery (KIB) by employing 3,4,9,10-perylene-tetracarboxylic acid dianhydride (PTCDA) as the cathode. The PTCDA electrode exhibits a high capacity of 131 mA h g^{-1} in the potential range of 1.5–3.5 V vs. K/K⁺, which corresponds to the insertion of two K ions per PTCDA molecule. The PTCDA cathode also shows great cycling performance, with a capacity retention of 66.1% over 200 cycles. Furthermore, K₁PTCDA can be obtained when discharging PTCDA to 0.01 V vs. K/K⁺, which delivers a high capacity of 753 mA h g^{-1} . The excellent electrochemical performance makes PTCDA a promising organic electrode based on K-ion chemistry for grid-scale energy storage.

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Introduction

Due to the ever-increasing global population and detrimental environmental effects associated with fossil fuel usage, green and sustainable forms of energy are becoming

increasingly important [1]. In turn, energy-related research projects, especially on developing electrical energy storage (EES) technologies, are attracting great attentions. Among various EES technologies [2–8], lithium-ion batteries (LIBs) have been widely applied in portable electronic devices, e.g. smart phones and laptops. Recently, LIBs are pursued for electric vehicles and grid-scale energy storage due to their high energy density and long cycling life. However, there is a rising concern on the affordability of LIBs due to

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the shortage of Li resources and its uneven global distribution. Therefore, research efforts have shifted to developing other EES technologies.

Due to similar electrochemical principles, Na-ion [9-13] and Mg-ion [14-18] batteries have been increasingly investigated. For example, room temperature Na-ion batteries (NIBs) have gained broad interests in both academia and industry, due to the abundance and wide availability of Na resources. Apart from Na-ion based systems, K-ion battery technology also offers an attractive LIB alternative due to the resources' low cost and natural abundance as well as its lower redox potential compared to Na (-2.93 V for K/K^+ compared with -2.71 V for Na/Na^+ , all vs. standard hydrogen redox potential) [19,20]. In the past few years, researchers focused on aqueous K-ion batteries (KIBs) and achieved excellent K-ion kinetics [21-23]. However, the energy density of aqueous batteries are significantly limited by low cell voltages [24]. Most recently, the Wu group and the Chen group have proved that K-oxygen batteries [25] and K-sulfur batteries [26], respectively, can deliver excellent performance. Unfortunately, many challenges remain for metal-oxygen and metal-sulfur systems. Therefore, developing and understanding the fundamentals of room temperature KIBs are highly desirable.

Like LIBs or NIBs, exploiting electrode materials holds the key for developing high-performance KIBs. The current cathode materials for LIBs are mainly layered oxides or transition-metal phosphates, while the anodes are carbonaceous materials. Based on the intercalation mechanisms of these electrode materials, the energy density of LIBs or NIBs is often small due to the limited amount of space used to accommodate Li or Na ions. On the other hand, organic electrodes provide a new avenue to develop batteries with high energy densities by extending the traditional intercalation limit [27]. Moreover, the low cost, environmental friendliness, and recyclability of organic electrodes have resulted in heightened battery-related research endeavors. For example, organosulfur compounds, free radical compounds, carbonyl compounds, non-conjugated redox polymers, and layered compounds have been widely studied for LIBs, where exciting progress has been made [28-31]. Among these organic electrodes, 3,4,9,10-perylene-tetracarboxylic acid-dianhydride (PTCDA), an organic pigment (Figure 1a), has been studied as a cathode for both

Li-ion and Na-ion batteries [32-34]. In LIBs, PTCDA showed a high capacity of 130 mA h g^{-1} at a current density of 100 mA g^{-1} , but it suffered from severe capacity fading [32]. Interestingly, PTCDA exhibited not only high capacity, but also good cycling performance in Na-ion batteries [33]. Inspired by these works, we report a novel non-aqueous KIB employing PTCDA as the electrode. For the first time, we demonstrate that PTCDA displays a high capacity of 131 mA h g^{-1} as a KIB cathode, which corresponds to the formation of K_2 PTCDA (Figure 1b). Interestingly, PTCDA can store about eleven K ions when discharging PTCDA to 0.01 V vs. K/K^+ , which indicates the possibility of this material as a high capacity KIB anode as well (Figure 1d).

Experimental section

Material characterization and electrochemical measurements

PTCDA was used as received from Tokyo Chemical Industry Co. Ltd. X-Ray diffraction (XRD) patterns were recorded with a Bruker D8 Advance using Cu $K\alpha$ radiation ($\lambda = 1.5406$ Å). Infrared (IR) absorption spectra were obtained using a Thermo Nicolet NEXUS 670 FTIR with an Attenuated Total Reflectance accessory for PTCDA powders. The morphology of PTCDA was observed with a Hitachi SU-70 field emission scanning electron microscope (FESEM). A JEOL JEM 2100 transmission electron microscope (TEM) was also used to characterize PTCDA morphology at an accelerating voltage of 200 kV. The electrochemical properties were investigated using CR2032-type coin cells. The electrodes were prepared by coating slurries containing PTCDA (70 wt%), carbon black (Super-P, 20 wt%), and polyvinylidene fluoride (PVDF, 10 wt%) dissolved in N-methyl-2-pyrrolidone (NMP) on current collectors (e.g. Al or Cu foil). Al foil and Cu foil were used when evaluating PTCDA at high and low potentials, respectively. After slurry coating, the electrodes were dried at 120 °C for 10 h under vacuum. The typical loading amount of the active material was 1.0 mg/cm². The cells were assembled in an Ar-filled glove box by employing K foil as the counter/reference electrode. The separator was a porous polyethylene membrane (Celgard 2425) and the electrolyte consisted of 0.5 M KPF_6 in an ethylene

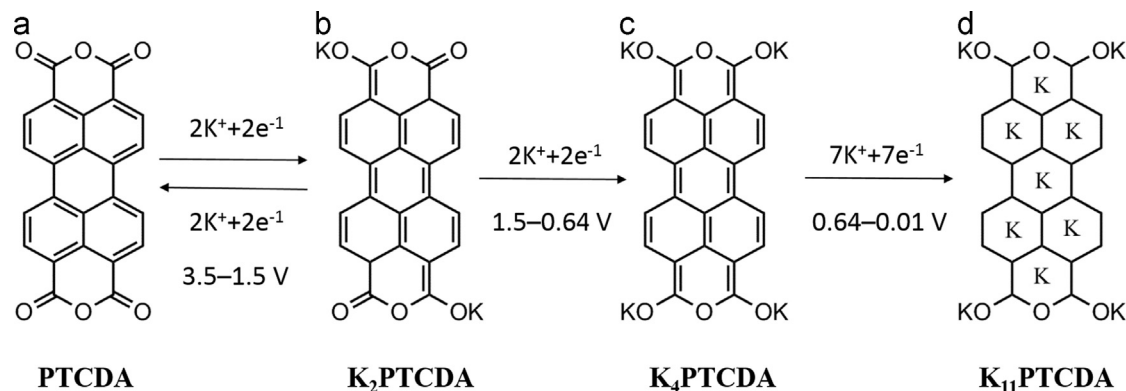


Figure 1 Schematic for the proposed electrochemical reaction mechanism of PTCDA in KIBs during the discharge/charge process.

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