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Review

Two-dimensional organic cathode materials for alkali-metal-ion batteries

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ABSTRACT

With the increasing demand for large-scale battery systems in electric vehicles (EVs) and smart renewable energy grids, organic materials including small molecules and polymers utilized as electrodes in rechargeable batteries have received increasing attraction. In recent years, two-dimensional (2D) organic materials possessing planar layered architecture exhibit optional chemical modification, high specific surface area as well as unique electrical/magnetic properties, which have been emerging as the promising functional materials for wide applications in optoelectronics, catalysis, sensing, etc. Integrating with high-density redox-active sites and hierarchical porous structure, significant achievements in 2D organic materials as cathode materials for alkali-metal-ion batteries have been witnessed. In this review, the recent progress in synthetic approaches, structure analyses, electrochemical characterizations of 2D organic materials as well as their application in alkali-metal-ion batteries containing lithium ion battery (LIB), lithium sulfur battery (LSB), lithium air battery (LAB) and sodium ion battery (SIB) are summarized systematically, and their current challenges including cycling stability and electron conductivity for cathode materials in battery fields are also discussed.

Chao Zhang received his B.S. degree from Hefei University of Technology in June 2015. Then he joined Prof. Xinliang Feng's group at Shanghai Jiao Tong University and began master's studies in September 2015. He focuses on fabricated porous carbon materials and 2D materials for energy storage and conversion.

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Xinliang Feng, after obtaining his master's degree in organic chemistry from Shanghai Jiao Tong University in March 2004, joined the group of Prof. K. Müllen at the MPIP and obtained his Ph.D. in April 2008. He became a group leader at MPIP in December 2007. Since June 2010, he has been a professor at Shanghai Jiao Tong University and became a distinguished group leader at MPIP in 2012. In August 2014, he became a chair professor at Dresden University of Technology. His current scientific interests include graphene, two-dimensional nanomaterials, organic conjugated materials, and carbon-rich molecules and materials for electronic and energy-related applications.

1. Introduction

The technology of energy storage and conversion lights up various electronic devices, giving the comfortable life in modern society [1–4]. Electrochemical energy storage is considered as one of the most promising strategies for storing energy [3,5–7]. With the development of nanotechnology, the appropriate energy storage devices, including fuel cells [8–11], rechargeable alkali-metal-ion batteries [12–19] and supercapacitors [20–22], etc., have been designed rationally. Among them, rechargeable alkali-metal-ion batteries, featuring clean, portable and high energy density characters, have attracted much attention in the field of chemistry and material science [18,23,24]. For alkali-metal-ion batteries, the electrode materials are the one of most important component parts, yet they often suffer from short cycle life, slow charging-discharging rate and lower power density, which limits their wide applications in electric vehicles and smart renewable energy grids [25,26]. Thus, design and construction of electrode materials with controllable nanostructure is still the main challenge to gain high-performance alkali-metal-ion batteries [27–30].

The electrode materials in alkali-metal-ion batteries can be divided into negative electrode and positive electrode, termed anode and cathode, respectively. For the former one, tunable porous carbon frameworks decorated with metal oxides and metal sulfides as the anode materials exhibit high reversible capacity, long cycle life, and rate performance in the recent years [18,31–33]. On the other hand, layered lithium transition metal oxides represent the most successful category of cathode, e.g. $LiCoO_2$ or $LiFePO_4$, which can undergo oxidation to higher valences when lithium is removed [34–38]. However, the complex preparation methods with energy consuming and low cycling performance severely restricted the practical applications of these inorganic cathode materials [39,40]. Therefore, that the development of cathode materials could not reach to match with that of anode is the critical problem to construct high performance alkali-metal-ion batteries.

Recently, two-dimensional (2D) materials, featuring ultrathin thickness of few atoms and large lateral size, exhibit excellent characters, such as large surface area, high mechanical strength, and amplified electronic and optical properties, which make them promising candidates in next-generation energy devices applications [41–45]. Compared with inorganic 2D materials, such as transition metal dichalcogenide [46–49], metal oxides [50,51], and metal hydroxides [52,53], 2D organic materials with reversible redox activity have been considered as novel cathode materials, due to their tunable structures, light weight, flexibility, and good semiconducting properties [42,54]. As the typical example of 2D organic materials, graphene, a flat monolayer of carbon atoms arranged in a 2D honeycomb lattice, exhibits unique physical and electronic properties, including high specific surface area, high mechanical strength, long distance conductivity, and good accessibility for electrolytes [42,55–60]. To date, large amounts of graphene-based composite materials have been developed as cathode materials for high-performance alkali-metal-ion batteries [54,61–66]. Feng et al. reported that the macroporous graphene aerogel-supported iron(III) hydroxide phosphate dihydrate microspheres (GA/IHPDs) hybrids showed the excellent ultrahigh current densities and reversible specific capacity of 155 mA h g^{-1} after 300 cycles at a current density of 50 mA g^{-1}. These intriguing results could be attributed to the synergistic interaction between the IHPD and the graphene aerogel, in which the graphene not only provided a continuous pathway for electron transportation, but also reduced the volume changes of the IHPD during the charge–discharge cycles [67]. Unfortunately, pristine graphene gives the zero bandgap and low electrochemical activity, leading to its poor specific capacity [68]. In this regard, searching for new 2D organic materials with high conductivity and reversible electrochemical activity as the alternative key cathode components for applications in alkali-metal-ion batteries is highly desirable.

To meet the requirement of 2D organic materials as the cathode for the application in alkali-metal-ion batteries, incorporation of heteroatoms (e.g. boron (B), nitrogen (N), sulfur (S), phosphorus (P)) into the carbon backbone of 2D organic materials represents an efficient strategy to obtain high electrochemical performance for alkali-metal-ion batteries [69–72]. On the basis of efficient chemical synthesis, heteroatoms can be embedded into the frameworks of 2D organic materials with well-controlled architecture on molecule level by using appropriate building blocks and various linkages [73]. Over the past few decades, the remarkable achievements in design and synthesis of 2D heteroatom-embedded organic materials as cathode materials have been witnessed significantly. According to their structural size, these 2D organic materials can be classified as 2D polyaromatic molecules, 2D covalent organic frameworks, and 2D polymeric nanosheets. Considering the broad field of 2D organic materials, the construction and application of some branches of this field, like covalent organic frameworks (COFs), have been enough described in another review article [74–76]. However, a perspective review on the design of heteroatom-embedded 2D organic materials as cathode for the application in alkali-metal-ion batteries has not yet been documented so far. In this review, we will comprehensively summary the synthetic approaches, characterization, and electrochemical properties of heteroatom-embedded 2D organic materials as cathode materials in alkali-metal-ion batteries, including lithium ion battery, lithium sulfur battery, lithium air battery and sodium ion battery.