

Recent progress in phosphorus based anode materials for lithium/sodium ion batteries



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ABSTRACT

Phosphorus has aroused growing concern as a promising anode material for both lithium and sodium ion batteries, owing to its high theoretical capacity and appropriately low redox potential. However, the poor electronic conductivity and large volume expansion of phosphorus during cycling lead to low electrochemical activity and unstable cyclability, which limits its practical application. Recently, various nanostructured phosphorus based anodes, which efficiently restrained the pulverization and supplied faster reaction kinetics, have been developed to solve these issues. This review aims to summarize the major progress of nanostructured phosphorus based electrode materials for lithium/sodium ion batteries. We first examine the most widely-used design strategy of compositing phosphorus with various carbon materials, ranging from 0D particles, 1D tubes or fibers, 2D sheets to 3D frameworks. And then, the progress of various metal phosphides and their composites is discussed, which mainly include Sn-P phosphides, Ni-P phosphides, Cu-P phosphides, Fe-P phosphides, Co-P phosphides, etc., and their composites. This is followed by a comparison of different compositing methods, which involve in preparing phosphorus-carbon composites and nanostructured metal phosphides or their composites. Finally, the challenges and perspectives regarding the phosphorus based anode materials are proposed.

1. Introduction

Energy issues have attracted great attentions during the past decade due to growing energy demand, exhausting refined fossil fuels and serious environmental issues caused by their usage. Various new and clean energy sources, i.e., solar, wind, hydro, tidal, and geothermal energies, are emerging rapidly. Therefore, a large-scale energy storage system is urgently required to store these renewable energies into the electrical grid to realize the peak shift. Lithium ion batteries (LIBs) have been presenting great promise, due to their fascinating characteristics, such as high energy conversion efficiency, stable cyclability, simple maintenance, adaptable power and energy features for different grid functions [1].

LIBs have been extensively used as the common power sources in the market of portable electronics since Sony realized their first commercial launch in early 1990s [2–4]. LIB technology has been experiencing great progress and commercialization in mid-size applications such as hybrid electric vehicles (HEVs) and electric vehicles (EVs) applications. Moreover, lots of grid-scale LIBs prototypes (approximately tens of megawatt-hours), used for storing renewable energy sources, have emerged on the market [5–7]. However, the

growing cost of LIBs due to the finite lithium resources ($\approx 0.0065\%$ as shown in Fig. 1a) would ultimately fail to satisfy the ever-increasing industrial demand, especially for HEVs, EVs, and large-scale renewable energy storage [8–10].

Alternatively, sodium ion batteries (NIBs) have attracted great attentions with the ever-growing demand for advanced rechargeable batteries, assigned to the abundance of sodium resources ($\approx 2.74\%$ as shown in Fig. 1a). Theoretically speaking, Na is heavier than Li, and NIBs may have a lower energy density than LIBs. However, the energy penalty is small because sodium has a suitable potential of -2.71 V (vs SHE). Therefore NIBs are much more suitable to a large grid stationary application, where the low cost and long cycle life of the batteries are more important for a whole system [11].

The electrochemical properties of the electrode materials are vital to the important performance characteristics of battery such as specific capacity and operation voltage. Therefore, the major challenge in advancing LIB and NIB technology lies in finding good electrode materials. However, the specific capacities of most cathode materials are low and it is difficult to greatly increase their specific capacities. The application of cathode materials with high redox potentials is also limited by the electrolyte which decomposes at high potentials.

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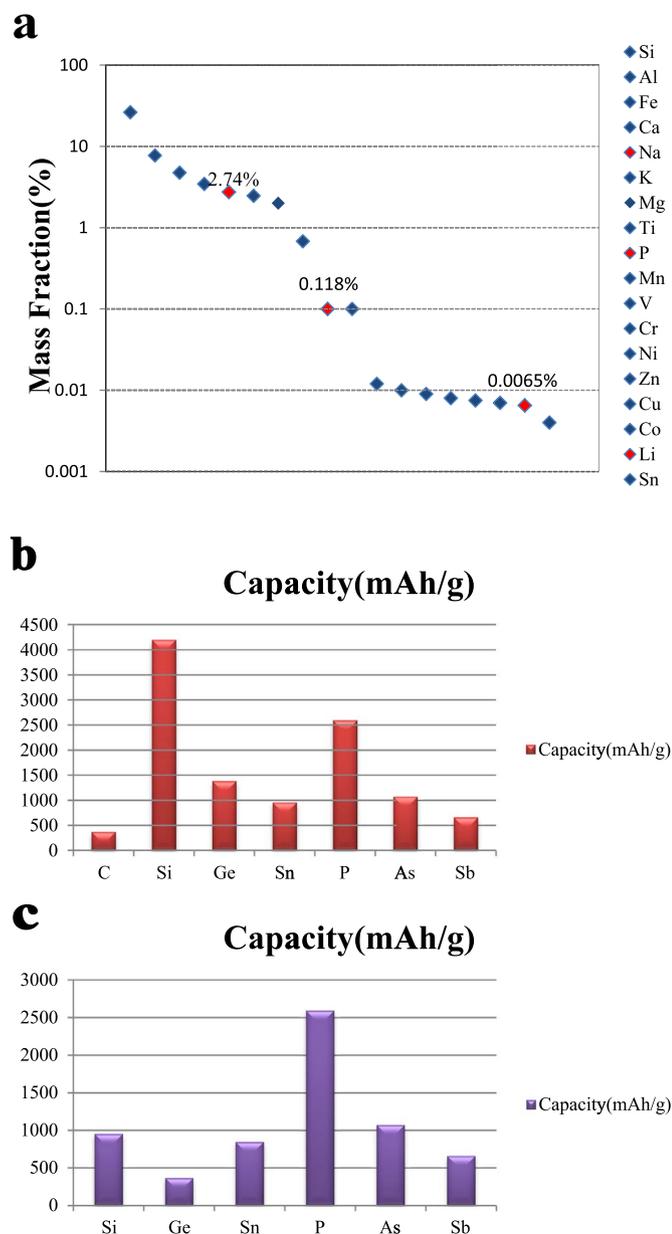


Fig. 1. (a) Elemental abundance in the earth's crust. (b) Theoretical specific capacity of the C, Si, Ge, Sn, P, As and Sb elements for LIBs; (c) Theoretical specific capacity of the Si, Ge, Sn, P, As and Sb elements for NIBs.

Therefore, it is a desiring approach to develop anode materials which own high specific capacities and relatively low redox potentials. Using conversion chemistry such as alloying materials is one of most used strategy for storing a large number of ions. Si-based material is the representative example, for which 4.4 Li can react with one Si to form Li-Si alloy, providing the highest theoretical specific capacity of 4200 mAh/g among LIB anode materials (Fig. 1b) [12–15]. However, for NIBs, it can store only one Na per Si, delivering a theoretical specific capacity of 954 mA h/g [16–18]. While phosphorus can not only react electrochemically with lithium to form Li_3P , but also store three Na at attractive potentials with a high theoretical specific capacity of 2596 mAh/g, which significantly exceeds that of any other NIB anode presently available (Fig. 1c) [19–30]. In addition, phosphorus has the advantages of low cost, abundance ($\approx 0.118\%$ in Fig. 1a) and easy availability, which provide great potential for its practical applications for LIBs and NIBs.

In this review, we will present the recent advances in phosphorus based anodes for LIBs/NIBs, with a focus on phosphorus carbon

composites and metal phosphides or their composites. This article covers the development of new promising phosphorus based anodes for LIBs/NIBs, lithium-storage mechanisms of metal phosphides and many efforts to enhance the electrochemical performance of phosphorus based anodes. In addition, methods for the synthesis of phosphorus-carbon composites and metal phosphides or their composites are summarized. Finally, the challenges and opportunities for the phosphorus based anodes of LIBs/NIBs are suggested.

2. Phosphorus based anodes for LIBs/NIBs

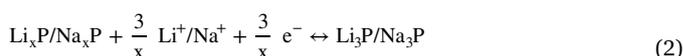
As an element of the fifth group in the periodic table, phosphorus possesses four main allotropes: white phosphorus, red phosphorus, violet phosphorus and black phosphorus (Fig. 2a) [31]. White phosphorus is volatile and toxic, and it bursts into flames if exposed to the natural atmosphere, enabling it unsuitable for electrode materials. Violet phosphorus has been rarely investigated in the past decades. However, its 2D layered violet phosphorene has drawn increasing research interest recently [32,33]. Alternatively, red phosphorus and black phosphorus have been studied commonly as the anode materials because they are chemically stable at room temperature and atmosphere. Red phosphorus is commercially available with ease, but its low conductivity ($\sim 10^{-14}$ S/cm) results in poor reversibility of electrochemical reaction. Crystalline black phosphorus as anode materials shows substantially improved reversibility relative to red phosphorus (Fig. 2b) [19]. However, due to the nature of poor electronic conductivity of phosphorus, there is a far way between its experimental capacity and theoretical value. It has been demonstrated that the conductivity of phosphorus based electrode can be improved by doping red phosphorus with iodine, and the electrodes for LIBs exhibited much higher specific capacity (1868 mAh/g at the second cycle) and much longer cycling life (1562 mAh/g at 520 mA/g after 150 cycles) than that of red phosphorus, even than black phosphorus (Fig. 2c) [34]. However, the electrochemical performance of most phosphorus based electrodes has been improved by forming different type of phosphorus carbon composites and metal phosphides or their composites as discussed in the following sections.

2.1. Phosphorus carbon composites

The intrinsic outstanding conductivity and diversity in architecture of carbon material make it the most popular material preferred for phosphorus based anodes. Various strategies have been developed to realize a perfect compositing between phosphorus and carbon materials. Carbon materials can be summed as particles, e.g., carbon black and graphite [19,25,31,35–41]; one-dimensional materials, e.g., nanotubes and nanofibers [42–49]; two-dimensional materials, e.g., graphene and reduced graphene oxide [53–64]; and three-dimensional materials, e.g., aerogels and mesoporous carbon [65–70,74]. In this section we will give a summary of the recent progress in phosphorus carbon composites.

2.1.1. Lithium/sodium-storage mechanism

The general reactions of phosphorus carbon composites with lithium/sodium are summarized as follows:



During lithiation/sodiation process, phosphorus reacts with lithium/sodium to form the compounds of $\text{Li}_x\text{P}/\text{Na}_x\text{P}$, with the final products of $\text{Li}_3\text{P}/\text{Na}_3\text{P}$. The delithiation/desodiation process involves a stepwise lithium/sodium ion extraction from the fully lithiated/sodiated $\text{Li}_3\text{P}/\text{Na}_3\text{P}$, corresponding to several plateaus in voltage profile, as well as the several cathodic peaks in the cyclic voltammogram.

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