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Journal of Energy Chemistry

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Review

The development in aqueous lithium-ion batteries

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ARTICLE INFO

Article history:

Received 31 January 2018

Revised 22 May 2018

Accepted 2 June 2018

Available online xxx

Keywords:

Li-ion batteries

Aqueous electrolyte

Cathode

Anode

Electrochemical performance

ABSTRACT

To meet the growing energy demands, it is urgent for us to construct grid-scale energy storage system than can connect sustainable energy resources. Aqueous Li-ion batteries (ALIBs) have been widely investigated to become the most promising stationary power sources for sustainable energy such as wind and solar power. It is believed that advantages of ALIBs will overcome the limitations of the traditional organic lithium battery in virtue of the safety and environmentally friendly aqueous electrolyte. In the past decades, plentiful works have been devoted to enhance the performance of different types of ALIBs. In this review, we discuss the development of cathode, anode and electrolyte for acquiring the desired electrochemical performance of ALIBs. Also, the main challenges and outlook in this field are briefly discussed.

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1. Introduction

Due to the formidable sustainability challenges, there are numerous needs for significant research in various disciplines, including the field of electrochemical energy storage derived from solar, wind and tidal power [1,2]. Substantial progress in battery technology is essential if we need to succeed in an energy transition towards a more carbon-neutral society. Since its

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commercialization in 1990, a rechargeable lithium ion (Li-ion) battery with organic electrolytes possess a lot of advantages of high energy density and lighter weight, making it possible to become an alternative energy source for large-scale applications in electricity storage stations, electric vehicles, and other portable electric devices [3–5]. However, the use of flammable and toxic organic electrolytes still hinders their wide application. Since development of the first aqueous Li-ion battery (ALIB) by Dahn's group in 1994, number of efforts have devoted on ALIBs in order to replace the flammable organic solvent with a more green and safe aqueous-base electrolyte, theoretically providing an average operating voltage of 1.5 V, with energy density (75 Wh kg^{-1}) larger than the Pb-acid batteries (75 Wh kg^{-1}) [6,7]. Nevertheless, the cycling of $\text{VO}_2/\text{LiMn}_2\text{O}_4$ aqueous lithium battery was very poor. Inspired by the pioneer work, $\text{LiV}_3\text{O}_8/\text{LiNi}_{0.81}\text{Co}_{0.19}\text{O}_2$, $\text{LiV}_3\text{O}_8/\text{LiCoO}_2$, $\text{TiP}_2\text{O}_7/\text{LiMn}_2\text{O}_4$, and $\text{LiTi}_2(\text{PO}_4)_3/\text{LiMn}_2\text{O}_4$ aqueous lithium-ion batteries have also been reported in the 2000s [8–10]. Similar to the $\text{VO}_2/\text{LiMn}_2\text{O}_4$ system, the cyclability of the problem was not fundamentally resolved. In the later 2007s, our group made a kind of ALRBs with superior cycling stability by combining LiMn_2O_4 cathode and Nasicon-type $\text{LiTi}_2(\text{PO}_4)_3$ in 1 M Li_2SO_4 aqueous solution [11]. Subsequently, our group found that the $\text{LiTi}_2(\text{PO}_4)_3$ anode in a discharged state can react with water and oxygen, resulting in capacity fading upon cycling. By eliminating oxygen, adjusting the pH values of the electrolyte and using carbon-coated electrode materials, $\text{LiTi}_2(\text{PO}_4)_3/\text{Li}_2\text{SO}_4/\text{LiFePO}_4$ ALIB exhibited excellent stability with capacity retention over 90% after 1000 cycles.

ALIBs, are the promising alternatives for large-scale applications, can address several challenges related to conventional Li-ion batteries. Firstly, it can replace highly toxic and flammable organic solvent. Secondly, the easy-to-use conditions and lower cost of electrolyte solvent than conventional Li-ion batteries is also an important progress. Finally, the ionic conductivity of the aqueous electrolyte is higher than those of organic electrolytes by two orders of magnitude, resulting in high round-trip efficiency and energy density even with bulky and scalable electrodes [12]. Despite the identical mechanism of ALIBs to that of conventional organic battery, the electrochemical reaction of Li^+ extraction/insertion in aqueous electrolyte is more complicated, thereby leading to a great influence on the selectivity of electrode materials. In aqueous solution, many side reactions from the O_2 and water involved, including the H_2/O_2 evolution reaction, and proton co-intercalation parallel to lithium ion intercalation, obviously affect the coulombic efficiency and cycling life of electrode systems. Therefore, the redox potential of Li^+ extraction/insertion should be within or near the electrolysis potentials of water. Over the past decade, a number of ALIBs employing various types of electrode materials have been explored to acquire better electrochemical performance batteries.

In this review, we present an overview of selected developments in the area of electrodes (both cathodes and anodes) and electrolyte materials for ALIB systems in the past few years. Mn-based oxides, layered lithium oxides and polyanionic compounds are extensively considered as the alternative cathode materials, and $\text{LiTi}_2(\text{PO}_4)_3$, TiP_2O_7 and vanadium oxides are exploited as anode materials ALIBs. In addition, the challenges and outlook for future ARLBs are also discussed.

2. Electrodes for aqueous lithium ion batteries

2.1. Cathode materials

2.1.1. Mn-based oxides

Since the development of the ALIB by Dahn's group in 1994, VO_2 anode and LiMn_2O_4 cathode, numerous aqueous lithium-ion cathode materials have been explored. Among various cathode ma-

terials explored so far, spinel LiMn_2O_4 is the most widely researched aqueous cathode material. Subsequently, ALIBs, fabricated with spinel $\text{Li}_2\text{Mn}_4\text{O}_9$ as the anode and LiMn_2O_4 as cathode in 6 M LiNO_3 solution, can deliver $\sim 100 \text{ mAh g}^{-1}$ capacity at an average voltage of 1–1.1 V [13]. In 2007, Nakayama et al. investigated the electrochemical properties of LiMn_2O_4 thin film electrode in aqueous solution. The cyclic voltammograms of the film electrode conducted in aqueous solution were similar to the organic system, two identical coupled current peaks were observed, corresponding to the lithium-ion insertion/extraction reaction, as shown in Fig. 1(a) and (b) [14]. Also, the previous work reported by Eftekhari et al. has shown that the LiMn_2O_4 electrode is electrochemically stable in aqueous medium during potential cycling. In 2009, Tian and Yuan prepared nanostructured spinel LiMn_2O_4 material via a room-temperature solid-state grinding reaction [15]. Electrochemical measurements of the LiMn_2O_4 were conducted in different concentrated solutions, 1 M, 5 M and 9 M LiNO_3 with pH value of 5.71, 5.56 and 5.34, respectively. They have found that, the peak potentials obviously shifted positively with increasing concentration of LiNO_3 , but the two redox couples were distinct and current increased markedly in 5 M LiNO_3 , which may indicate better reversibility of the electrode reaction (Fig. 1(c)). Moreover, the electrode in 5 M LiNO_3 solution exhibited the best cycling stability with capacity retention of 71.2% after 600 cycles (Fig. 1(d)). Driven by the unsatisfactory cycling of LiMn_2O_4 material, Qu et al. reported porous LiMn_2O_4 consisting of nanograins as aqueous cathode material for ALIBs [16]. As shown in Fig. 2(a), the porous LiMn_2O_4 exhibits excellent rate behavior as compared to solid LiMn_2O_4 , a discharge capacity of approximately 108 mAh g^{-1} was obtained at 5 A g^{-1} , and up to 90.0 mAh g^{-1} was maintained at 10 A g^{-1} . More importantly, Fig. 2(b) shows better cycling stability of porous LiMn_2O_4 measured at 1 A g^{-1} , which can retain 93% capacity retention after 10,000 cycle, and whereas the discharge capacity of the solid LiMn_2O_4 electrode decreases rapidly in the first 2000 cycles.

In addition, some efforts have been devoted to doping metal ions to the LiMn_2O_4 crystal to modify the structural integrity. In the Yuan's work, Al-doped $\text{LiAl}_x\text{Mn}_{2-x}\text{O}_4$ samples with different dopant contents were prepared by calcining at different temperatures for different calcination times [17]. It was found in Fig. 3(a), the $\text{LiAl}_{0.1}\text{Mn}_{1.9}\text{O}_4$ calcined at $800 \text{ }^\circ\text{C}$ exhibits two distinct couple peaks like that of the ordinary LiMn_2O_4 , which can be ascribed to the high crystallinity of the $800 \text{ }^\circ\text{C}$ derived material. Further cycle performances concluded that the $\text{LiAl}_{0.1}\text{Mn}_{1.9}\text{O}_4$ derived from 12 h calcination remained higher capacity retentions of 75.6% after 8000 cycles even though it did not exhibit the highest initial specific capacity, as shown in Fig. 3(b). Similarly, Cvjeticanin et al. have also prepared $\text{LiCr}_{0.15}\text{Mn}_{1.85}\text{O}_4$ by a rapid glycine-nitrate method, which displayed delivered more stable cyclability as compared to the undoped LiMn_2O_4 (Fig. 3(c)). These improvements in stability may be attributed to the depressed Jahn–Teller distortion and stabilization of the octahedral sites [18]. Subsequently, Stojkovic et al. have investigated the influence of vinylene carbonate addition to aqueous electrolyte on the performance of doped $\text{LiCr}_{0.15}\text{Mn}_{1.85}\text{O}_4$ material [7]. It was found that vinylene carbonate can promote the stability of $\text{LiCr}_{0.15}\text{Mn}_{1.85}\text{O}_4$ in aqueous electrolyte (Fig. 3(d)). The vinylene carbonate additive produced a stable solid electrolyte interphase (SEI) layer on electrode materials, which can prevent water molecules from penetrating into aqueous electrodes [19–21].

2.1.2. Layered lithium oxides

Since layered LiCoO_2 has made huge commercial application as cathode materials in conventional LIBs, it has also been demonstrated in aqueous batteries. For example, in our previous work, we have studied the electrochemical behavior of LiCoO_2 in 1 M Li_2SO_4

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