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Review

Metal–organic frameworks for energy storage: Batteries and supercapacitors

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ABSTRACT

Metal–organic frameworks (MOFs) are a class of porous materials that have attracted enormous attention during the past two decades due to their high surface areas, controllable structures and tunable pore sizes. Besides the applications in gas storage and separation, catalysis, sensor, and drug delivery, MOFs are receiving increasing research interest in the field of electrochemical energy storage. By focusing on recent advances, this review provides a broad overview of MOF-based or MOF-derived rechargeable lithium ion batteries and supercapacitors.

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1. Introduction

In the modern society, energy needs are ever-growing and the concerns about the depletion and the associated environmental pollution resulted from the burning of fossil fuels are becoming more and more serious. The pursuit of renewable energy sources and sustainable storage technologies has been a worldwide research target to resolve the energy and environmental crisis. Solar and wind energy are attractive renewable energy sources to produce electricity, but their intermittent nature makes the electricity

Abbreviations: 1,3,5-H_3BTC, 1,3,5-benzentricarboxylic acid; BDC, 1,4-benzenedicarboxylate; DEF, diethylformamide; DMF*N*,, *N*-dimethylformamide; NMP, 1-methyl-2-pyrrolidone; BET, Brunauer–Emmett–Teller; IM, imidazole; LiTFSI, lithium bistrifluoromethanesulfonimide; bbb, 1,4-bis(benzoimidazol-1-yl) butane; EC, ethylene carbonate; DMC, dimethyl carbonate; Super P, a type of commercial carbon black used as conductive agent in batteries.

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applicable and competitive only if effective and reliable energy storage technologies are developed. In the market of transportation, the development of hybrid electric vehicles (HEVs), plug-in hybrid vehicles (PHEVs) and full electric vehicles (EVs) raises the demand for the energy and power density of electrical energy storage (EES) devices. Emerging as the forefront of EES system, batteries and supercapacitors (SCs) are two major technologies [1,2]. However, neither of the two can possibly meet all the requests. Batteries have higher energy densities than SCs, but poorer power densities. Yet even when fully developed, the highest energy that can be stored in either batteries or SCs is still insufficient for such applications; rate performance, cycle life and safety of batteries need further improvements. The exploration of new materials with high performance and new chemistry for EES is a great challenge [3]; large research efforts are being devoted to the development of next-generation batteries and SCs.

Metal–organic frameworks (MOFs) are a class of porous materials first defined by Yaghi and co-workers in 1995 [4] and have attracted intense interest during the past two decades. These materials are crystalline and are assembled by metal-containing units (secondary building units (SBUs)) and organic linkers. By varying the SBUs and the functionalities of the organic linkers, more than 20,000 different MOFs have been created and this number is still growing [5–7]. In addition to exploring their versatile properties including gas storage and separation [8–13], sensor [14–17], catalysis [18–20], and drug delivery [21–23], the application of MOFs in electrochemical systems is currently an emerging field in recent years [24–28] (Fig. 1).

In this review, we provide a broad overview of recent investigations on the applications of MOFs and their derivatives in EES systems. Several early reviews have summarized the important applications of MOFs in electrochemistry [29–31]. They focus on the development of MOFs for clean energy applications, including hydrogen production and storage, fuel cells, LIBs, SCs and solar cells. Herein, we provide a detailed and broad review specifically on LIBs and SCs. We first present recent development in negative (anode) and positive (cathode) materials, and solid electrolytes for LIBs, followed by the results in the research of Li—S and Li—O_2 batteries, two of the most attractive battery technologies that are receiving broad interest for their high energy. We then discuss the applications of MOFs and their derivatives in SCs, which stand as the most

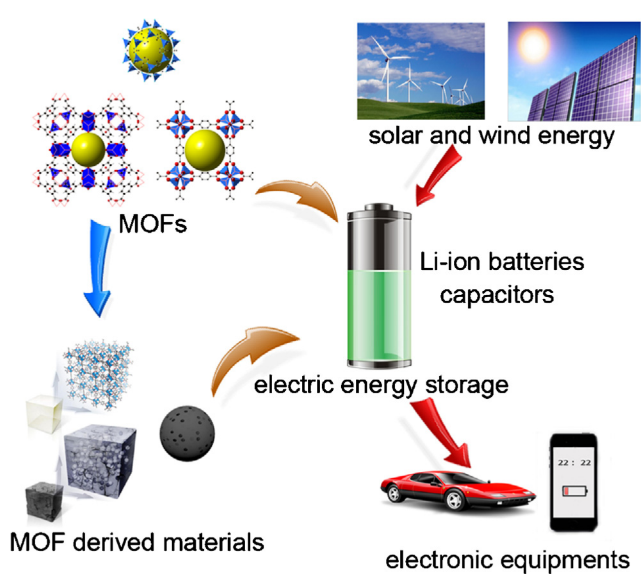

Fig. 1. The applications of metal–organic frameworks in electric energy storage. [Reprinted with permission from Ref. [27,32]. Copyright 2014, Royal Society of Chemistry, Copyright 2013, American Chemical Society.].

promising high power EES devices. We hope that this work could summarize pioneering studies and promote further development for MOF-based and MOF-derived EES systems.

2. Applications of MOFs in lithium batteries

Lithium ion batteries (LIBs) have found great commercial success for the energy supply of portable electronic devices. Yet when moving to large-scale EES applications, such as EVs and stationary EES, current LIBs cannot meet the requirements in terms of the energy density and power density. Going beyond the horizon of conventional LIBs needs innovative design and synthesis of new materials, finding new chemistry for Li storage and new EES technologies. As the most important three parts in a battery, anode, cathode and electrolyte have been studied extensively. But as we all know, none of the three primary components can meet the power demand of EVs; therefore there is large room for further improvement of their performances. MOFs are made by linking inorganic and organic units by strong bonds. Meanwhile, most of inorganic units are metal ions and metal oxides, which can serve as redox active sites during the electrochemical process. Thus MOFs can be potentially used as electrode materials for LIBs and fuel cells. Recently, MOFs have been investigated as anode, cathode, and electrolyte materials for LIBs. At the same time, many materials have been developed with the participation of MOFs and their derivatives [26]. Exciting results were presented in the past few years (Table 1).

2.1. Negative electrodes for Li-ion batteries

Graphite, which is the anode material in conventional LIBs, has a low capacity of 372 mA h g^{-1} and poor rate-performance. To satisfy the demand of high energy density and power density, developing new anode materials is necessary. Emerging anode materials, such as silicon and metal oxides, are under development, but still have a way to go before commercialization. MOFs have intrinsic permanent pores that can allow Li-ions to be stored and reversibly inserted/extracted, leading to insertion-type electrodes. Conversion-type electrodes can also be fabricated with properly selected metal centers and organic ligands if a reversible transformation/regeneration of the framework is ensured. More impressively, MOFs can be ideal templates for synthesizing metal oxides and carbon materials with unique nanostructures that can be hardly obtained through other methods. These fascinating properties provide numerous options for developing new anode materials.

2.1.1. Pristine MOFs

The first MOF using directly as an anode material for LIBs was MOF-177 reported in 2006 [33]. Li storage occurs through conversion reactions, with the destruction of the MOF structure and the formation of metallic Zn:

$$Zn_4O(BTB)_2 \cdot (DEF)_m(H_2O)_n + e + Li^+ \rightarrow Zn + Li_2O \quad (1)$$

$$DEF + H_2O + e + Li^+ \rightarrow Li_2(DEF) + LiOH \quad (2)$$

$$Zn + Li^+ + e \leftrightarrow LiZn \quad (3)$$

The irreversible degradation of MOF-177 leads to a poor cycling performance, which showed a high initial discharge capacity of 400 mA h g^{-1} under 50 mA g^{-1}, but dropped to 105 mA h g^{-1} in the second cycle. Albeit considerable drawbacks, this pioneering work opened new opportunities for researchers to explore new MOF materials as anodes for LIBs with better performance. After that, several other MOFs were studied, including $Zn_3(HCCO)_6$, $Co_3(HCOO)_6$, $Zn_{1.5}Co_{1.5}(HCOO)_6$ [34], Mn-LCP [35], $Co_2(OH)_2BDC$

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