Journal of Energy Storage 13 (2017) 103–122

Contents lists available at ScienceDirect

Journal of Energy Storage

journal homepage: www.elsevier.com/locate/est

Review

Recent advances in layered double hydroxides as electrode materials for high-performance electrochemical energy storage devices

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ARTICLE INFO

Article history:
Received 22 November 2016
Received in revised form 16 May 2017
Accepted 29 June 2017
Available online 19 July 2017

Keywords:
Layered double hydroxide
Nano-architectured
Energy storage devices
Supercapacitors

ABSTRACT

As we are facing increasing challenges of diminishing fossil fuel and global warming, there is increasing interest in developing advanced and cost effective electrochemical energy storage devices for diverse applications including mobile power supply to portable electronics, electric vehicles (EVs) or hybrid EVs (HEVs). To this end, layered double hydroxides (LDHs) have gained considerable attention in the past decade as a unique class of electrode materials due to their multiple cations, flexible ion exchangeability and tunable compositions. With abundant slabs and electrochemically active sites, the LDHs can used to produce energy storage devices with both the double-layer capacitance and Faradaic pseudo-capacitance. In this review, we summarize and discuss recent progress in the synthesis of LDHs based materials for electrochemical energy storage devices including lithium ion batteries and electrochemical supercapacitors.

Contents

1. Introduction

Adaptation to renewable energy is very important for the sustainable development of a country. The gradual growth in

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http://dx.doi.org/10.1016/j.est.2017.06.011
2352-152X/

energy consumption due to rapidly increasing population and industrialization coupled with the environmental concern all over the world necessitate the ecofriendly, efficient and cost effective energy conversion and storage (ECS) devices. Scientists and researchers are striving hard to build efficient energy storage systems/devices with improved performance and reliability. Among the various alternative energy storage technologies, electrochemical energy storage has the edge for its high efficiency, portability versatility and flexibility. Typical energy storage conversion and storage devices include photo-electrochemical water splitting, fuel and solar cells, Li-ion batteries [1,2] and supercapacitors [3–5].

The efforts put forth by the researchers to maximize the efficiency of existing bulk materials have been un-effective due to the inherently limited performance of these materials [6]. During the last two decades, nanostructured materials (e.g. nanoparticles, nanowires etc) have played a key role to improve the efficiency of energy storage devices by tailoring the materials properties at the nano-scale. Scientists have used various approaches to enhance the performance of materials such as downscaling of materials and their microstructure modifications. Composite formation and the development of self-standing vertical arrays of nanowires, nanosheets or nanotubes are also two effective methods to maximize the efficiency of active materials. All these approaches emphasized on increasing the surface area, reactivity, stability and conductivity of materials.

Among the electrochemical energy storage devices, lithium ion batteries (LIBs) promise high voltages, prolonged cycling life, high specific energy density, low self-discharge and low toxicity, whereas supercapacitors have attracted researchers because of their high power density that is an important requirement for applications in electric vehicles and hybrid electric vehicles [7–9]. The performance of LIBs and supercapacitors depend largely upon the electrode materials used. With the rapid developments in the synthesis techniques and characterizations, the performance of batteries and supercapacitors has increased a lot.

Recently, layered double hydroxides (LDHs) have been investigated for their potential usage in supercapacitors and LIBs because of their unique 2-D ionic lamellar structure that exhibit anionic exchange property [10], stability in the ambient atmosphere and alkaline resistivity.

LDHs can be symbolized by the general formula $[M^{2+}_{1-n}M^{3+}_{n}(OH)_2]^{n+}[A^{z-}]_{n/z}.\ mH_2O$, where M^{2+} denotes divalent cations (M = e.g. Fe, Cu, Mg, Ni, Zn or Co) and M^{3+} denotes trivalent (M = e.g. Al, Cr, Fe, Ga or Mn); the value of 'n' being the molar ratio of $M^{2+}/(M^{2+}+M^{3+})$ and is generally in the range 0.2–0.33; A^{z-} is an anion as shown in Fig. 1. By changing the molar ratio of M^{2+}/M^{3+}, nature of the metal cations, type of interlayer anions, a wide variety of physicochemical properties can be attained [10]. The hydroxide layers in LDHs possess positive charge formed by the isomorphous substitution of divalent cations with the trivalent cations. Anions present in between these layers act as charge balancing species. The hydrogen bonded water molecules occupy any free space between the layers. The structure stabilization is attributed to the hydrogen-bonding network between the water molecules, anions and the hydroxyl layers as well as the electrostatic forces present between the anions and the hydroxyl layers [11].

Benefiting from their unique structural and compositional merits such as layered structure low cost, high redox activity, tunable composition and environmentally friendly nature and the flexibility of incorporating mixed valence transition metal ions into the LDH structure in different compositions. LDHs have drawn extensive and intensive research attention during recent years in the field of electrochemistry due to their. Several reviews have been published about the synthesis methods and possible applications of LDHs, for example, Qiang Wang et al. [13] published a comprehensive review on the synthesis and applications of LDH nanosheets and Ma et al. [14] reviewed the field with emphasis on the exfoliation and delamination of LDHs. To the best of our knowledge there is no review on the LDHs with an emphasis on energy storage applications such as supercapacitors and Li-ion batteries. Therefore, in the current paper we will mainly focus on the synthesis of various LDHs and their applications in energy storage devices.

2. Synthesis of LDHs

For large-scale applications of any materials, sustainability in their synthesis methods is firstly required. It can be achieved if the methods are cost-effective, eco-friendly and easy to upgrade. The physico-chemical properties of materials such as surface properties, structure, catalytic activity and adsorptive properties etc. also depend upon the synthesis approach. A lot of attempts have been

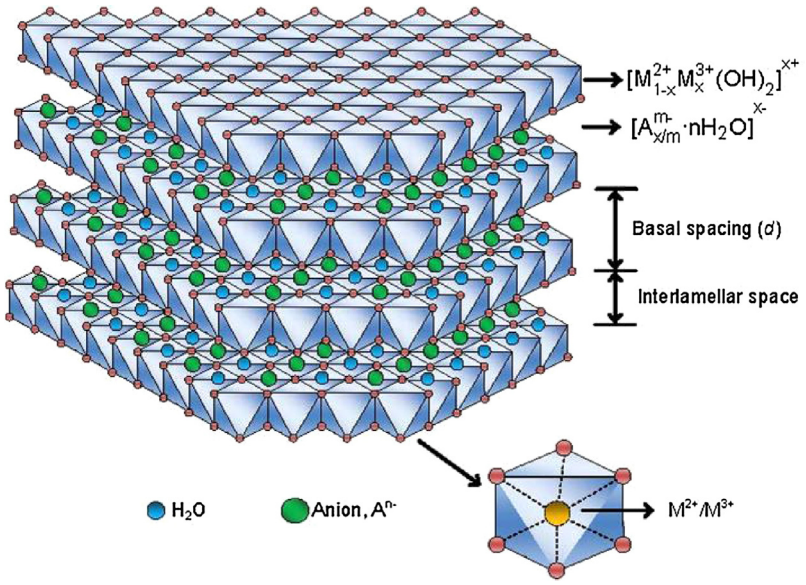

Fig. 1. Schematic illustration of the LDHs structures [12].