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Recent Advances in the Use of Carbonyl Compounds as Active Components in Organic-Based Batteries

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ARTICLE DETAILS

Article history: Received 28 May 2018 Accepted 07 June 2018 Available online 17 June 2018

Keywords: Carbonyl Compounds Battery Energy

ABSTRACT

The quest for green and sustainable energy storage systems has brought about the need for a material system that can satisfy the following requirements; low cost of production, environmental benignity, flexibility, redox stability, renewability and structural diversity. Interestingly, organic compounds of carbonyl functionality have been identified as potential candidates to proffer solutions to the abovementioned challenges. This review therefore discusses various classes of carbonyl compounds as active components in some rechargeable batteries, highlighting their major strengths and drawbacks.

1. Introduction

The world's increasing energy demand has led to the exploitation of non-renewable energy supplies such as coal, oil, natural gas etc., [1, 2] which has led to the emergency problems of climate change, smog, impending exhaustion of fossils fuels, and constant threats of global warming. These have therefore resulted to a quest for a low-carbon society where intensive research efforts have been channeled towards the field of renewable energy. Several renewable energy options such as the solar, wind, biomass, hydropower and tidal energies were proposed and investigated with the hope that they can proffer solutions to mitigate the aforementioned crisis [3-6]. The efficient utilization of such intermittent energy sources requires effective and flexible storage systems [7-13]. On the one hand, battery systems (with a long battery life of > 10,000 cycles, high current density and fast charging) are identified as a viable solution [7, 8], while on the other hand, the cost of production of a conventional battery is far from ideal, owing to the fact that the fabrication of electrodes from limited ores increases battery cost and similarly the life cycle of the batteries are far from the stated value because their operations are driven by redox reaction of the metal elements [14, 15].

Furthermore, there are safety concerns with respect to the currently available types of batteries because they contain toxic and/or environmentally unfriendly components, such as toxic non-aqueous electrolyte in current Li-ion batteries, acid/alkaline electrolyte in Ni-MH or lead-acid batteries, and toxic electrode materials such as Pb in lead-acid batteries, Cd in Ni-Cd batteries, VOx in vanadium flow batteries, and Br2 in Zn-Br2 flow batteries. It is therefore essential that these toxic materials be replaced with non-toxic, biodegradable materials so that they can become more sustainable and less hazardous to the environment. Similarly, the growing markets for thin, small and lightweight portable devices also require battery systems of similar properties with flexibility of applications [16]. In order to satisfy such requirements, several rechargeable battery designs were invented and extensively studied in which inorganic-based batteries such as lithium-ion batteries (LIBs)

emerged as a technology of choice for portable electronics, power tools, and hybrid/full electric vehicles because it has an unmatchable combination of high energy and power density, slow self-discharge, and long cycle life [17, 18].

However, new requirements for secondary rechargeable batteries such as flexibility of applications, low cost of production and environmental friendliness has seen focus shifting from inorganic to organic-based batteries via the use of organic electrode materials particularly polymers because they have inherent advantages over their inorganic counterparts because they offer for high flexibility, ability to tailor their redox properties by adjusting their structures, they can be generated from renewable resources using low-temperature procedures, they are electroactive not only towards lithium but to any metal thereby making them applicable in the designs of magnesium, zinc, sodium and air batteries because their redox mechanism is based on conversion reaction. Design of such innovative electrode materials for practical applications must take full consideration of high gravimetric and volumetric density, superior power rates, long cycle life, low cost and good safety, and well balance of them [19].

In recent years, several promising approaches for organic material-based battery systems have been investigated [20-22], including extensive exploration of organic carbonyl compounds as high-energy cathode materials for rechargeable lithium batteries owing to their redox stability, structural diversity, high theoretical capacities, and infinite availability from biomass [23-35]. Generally, organic electrode materials can be categorized into different types based on their electrochemically active functional groups involved during redox reaction, which includes conjugated carbonyl compounds, conducting polymers, organosulfides and free radicals. Conjugated carbonyls compounds were first discovered for energy storage materials in 1969 [36] and can be stabilized with a variety of different substituent R-groups, i.e., R-(C=0)-R'. Quinone is one of the most well-known examples that have carbonyl groups and exhibit fast redox reaction.

Depending on the stabilizing mechanism/substituents, carbonyl compounds used as electrode materials are broadly classified into three groups:

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 Those that employ vicinal carbonyls to form stable enolates e.g. 1,2diones which can be further stabilized by neighbouring aromatic systems as in phenanthraquinones.

2. Those that contain aromatic carbonyl derivatives directly connected to an aromatic core, dispersing the negative charge by delocalization.

3. Those that contain quinonestructures whose stabilizing force is the formation of additional aromatic system upon reduction.

1.1 Operating Principles of a Battery

Batteries, regardless of their chemistry (aqueous, non-aqueous, Li or Na-based) store energy within the electrode structure through charge transfer reactions. Such device architecture consists of several electrochemical cells that are connected in series and/or in parallel in order to provide the required voltage and capacity, respectively. Each cell is composed of a positive electrode referred to as the cathode which is usually contained within the battery and a negative electrode known as the anode which usually forms the outer case of the battery. These are terminals where the redox reactions take place i.e. oxidation takes place at the anode while reduction takes place at the cathode. The electrodes are separated by an electrolyte, usually a solution containing dissociated salts so as toenable ion transfer between the two electrodes i.e. cations which are positively charged ions move from the anode to the cathode and anions which are negatively charged ions move from the cathode to the anode. Once these electrodes are connected externally, the chemical reactions proceed in tandem at electrodes, liberating electrons and providing the current to be tapped by the user [17, 37, 38].

This review is therefore aimed at discussing the recent advances made in utilizing organic electrode materials especially different classes of carbonyl compounds as active components for secondary rechargeable batteries.

2. Working of Organic Carbonyl Electrodes

It has been well established in electrochemistry that for a rechargeable battery to function effectively, the following conditions must be satisfied:

- 1. Two electrode materials are required whose redox potential must not be the same.
- These two electrodes must be able to undergo redox reversible reactions.

The redox property of carbonyl compounds relies solely on the charge state of the carbon and oxygen atoms of the carbonyl functional group. In general, carbonyl compounds depending on the charge state of the functional atoms are reduced during redox reactions to negatively charged anions which have the capacity to produce one or more electron during charging or discharging process. The negative charge formed during the redox process is required to be balanced by a suitable counter ion derived from the electrolyte salt that will migrate back into https://doi.org/10.30799/jaec.054.18040105

the electrolyte upon re-oxidation. The salt must be suitable for both the cathode and the anode materials depending on the material systems employed as the counter electrode. Similarly, the salts to be used as electrolytes must possess the following properties; it must be inert towards the two electrodes, it must have low viscosity with high conductivity, and must have high boiling point [39, 40]. Therefore, salts of lithium, ammonium and sodium are often utilized for this purpose. Carbonyl compounds on their own rights are widely employed as cathodic materials while metals of either lithium or sodium are used as the anodes. Interestingly, some carbonyl compounds displaying low redox potential can be used as anode if the device architecture incorporates other materials such as metal alloys as cathodic materials [41, 42].

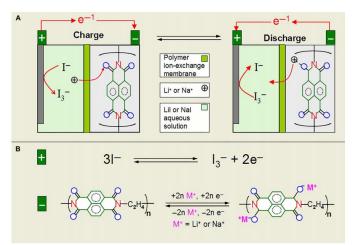


Fig. 1 Schematic illustration of (A) cell structure and (B) electrode reactions

${\it 2.1 Factors Affecting the Electrochemical Performance of Organic Carbonyl-based Batteries}$

Certain parameters of interest namely cell voltage and charge/discharge cycling stability of batteries are used performance indicators of a battery systems. However, the aforementioned parameters are influenced by some factors which reduce the theoretical efficiency of the battery. It can be recalled that the architecture of a battery requires two electrodes with different redox potentials; carbonyl compounds can have their redox potentials improved or lowered by adding substituents which can either have electron donating or withdrawal effect. These substituents that are not directly involved in the redox process decrease the theoretical capacity of the compound since it is a function of the molar mass of the compound and the number of electrons utilized during the reaction [43]. Also, charge/discharge cycling of the battery is dependent on several factors, some of which includes; solubility of the active materials in solution resulting to decrease in capacity, side reactions and/or instabilities of anions formed, structural modification during the charge/discharge leading to capacity loss on cycling. Finally, low conductivity of organic materials is also a limiting factor and this should be addressed by using conductive additive materials such as carbon nanotubes [44] and graphene [45, 46] in order to improve electron transport within the electrode.

3. Classes of Carbonyl-Containing Electrode Materials

This aspect of the review discusses the various classes of carbonylcontaining materials with different active sites depending on the type of functional groups present, which are employed as electrode materials in rechargeable batteries with emphasis placed on material activity and cycling stability.

3.1 Organic Anhydrides

This class of carbonyl compound was first studied as electrode material in rechargeable battery by Han and his co-workers in 2007. They utilized 3,4,9,10-perylenetetracarboxylicacidedianhydride as a functional material which yielded an initial capacity of 135 mAhg-1 with a cycling stability of 60% loss over 80 cycles due to dissolution in the electrolyte [25]. However, it showed improved performance when employed as cathode material in a sodium-ion battery [47]. The general mechanism for any aromatic anhydride is that it undergoes two-step electron reduction in which the reduced system is stabilized by enolation. Its application as an active component in energy storage systems owes to the fact that the stabilized carbonyl allows for reversible insertion of lithium ions at the oxygen atom of the anhydride functional group.

3.2 Aromatic Diimides

This class of carbonyl compounds has proven to be an electrode material in energy storage systems because it has a unique ability to undergo two-step redox reactions involving two electrons. This is possible because stability is achieved on the carbonyl molecules during the first stage of the reaction by inducing the formation of stabilized radical anions on the electron deficient ring, rendering the second step inappropriate because it results in the destruction of the redox system. Aromatic imides have been successfully employed in battery systems because of its high specific theoretical capacity (248 mAhg-1). The first success story with respect to aromatic imides was recorded by Renault and his co-workers in 2011 [48]. They investigated the charge/discharge properties of dilithium pyromelliticdiimide used as an electrode material in a rechargeable battery, they found out that the material exhibited an activity of 220 mAhg 1 which slightly dropped even after 25 charge/discharge cycles to about 200 mAhg-1. Another aromatic diimides employed for the same purpose is naphthalene diimide [49], which exhibited material activity of 140 mAhg-1 but drops to 80 mAhg-1after 100 cycles.

Even though these classes of carbonyl compound have shown promise, their full cycling stability has not been achieved yet because their practical application as an active component in rechargeable batteries is hampered by their solubility in electrolytes. This can be partly resolved by the formation of metal salts of the corresponding imide which have lower solubility in most of the electrolytes used in rechargeable batteries or alternatively, by using aqueous electrolytes which comes with its own drawbacks.

3.3 Polyamides

Similarly, polyamides because of their redox-active properties are also considered as candidates for secondary energy storage systems. These possessed similar two-step redox reaction involving two electrons exhibited by aromatic diimides with the first reversible reaction utilized in storage systems. So, the theoretical capacity of the materials is a function of the size and the aromatic unit between the two diimide molecules. The smaller the aromatic unit, the lower the redox potential and the higher the theoretical capacity of the material. To this end, several polyamides have been intensively studied as electrode materials in both lithium and sodium batteries, some of which include; polyimides synthesized by polycondensation of 1,4-phenylenediamine pyromellitic anhydride 6 and 4,4'-oxydiphthalic anhydride polyimides synthesized by polycondensation reactions of pyromellitic anhydride and 1,4,5,8-naphthalenetetracarboxylic dianhydride with 1,4phenylenediamine and 1,2ethanediamine were investigated [51]. Dense polyimide networks were obtained bypolycondensation of 1,3,5-tris(4aminophenyl)benzene with pyromelliticdianhydride14,1,4,5,8naphthalenetetracarboxylicdi anhydride and 3,4,9,10-perylene tetracarboxylicdianhydride, respectively [52]. These materials showed varying degree of success in terms of material activity, redox potential and charge/discharge cycling stability. However, the application of polyamides as cathode materials in both lithium and sodium batteries is still hampered by two major drawbacks which are; low operating voltage, usually 1.5 to 2.3V vs. Li+/Li and deep discharging can lead to the destruction of the active material through irreversible reactions because the polyamide system has little protection against the second reduction of the diimide system.

3.4 Conjugated Carboxylates

Organic conjugated carboxylic acids undergo a reversible two electron redox reaction, whereby the resulting dianion is stabilized by a conjugated, preferably aromatic system. The loss of resonance or aromaticity in the core of the molecules brings the redox reaction to very low potential. The first example of carboxylates in organic sodium-ion batteries (OSIBs) was disodium terephthalate (Na2TP) which was simultaneously reported by two research groups [53, 54]. Na2TP contains two carbonyl groups that allow inserting or extracting Na-ions, corresponding to a theoretical capacity of 255 mAhg-1. Similarly, organic lithium batteries based on dilithium terephthalate electrodes displayed an extraordinary performance. At a cell potential of 0.8 V a capacity of 300 $mAhg^{-1}$ (100% active material) was observed at the first charge/discharge cycle. Upon cycling over 50 charge/discharge cycles, the battery still exhibited a capacity of 234 mAhg-1 (78% material activity) [55]. Several other carboxylates have been tested positive as electrode materials in a rechargeable batterIS which include 4,4'-tolanedicarboxylate [56], (2,5dilithium-oxy)-terephthalate [57] and 2,5-pyrazinedicarboxylate [58].

3.5 Ketones

Ketones can be electrochemically reduced to the corresponding alcohols and reoxidized to their original state, realizing a one-electron redox reaction. Several compounds of ketones have been tested positive as an electrode material some of which included isodium salt of rhodizonate [59], which has a theoretical capacity of approximately 500 mAhg-1 and has been extensively studied in sodium ion battery. It yielded a capacity of 150 mAhg-1 which drops to 18 mAhg-1 after 40 cycles. Similarly, tri-carbonyl-based tris-N-salicylideneanthramine(TSAA) and (TSAQ) tris-N-salicylideneanthraquinoylamine showed excellent reversible transformation between C=O and C-O-Na because the reactivity of the α -carbon radical intermediates was largely suppressed by an electronic resonance effect and steric hindrance from the substitution groups [60]. TSAQ delivered capacities of 370 and 220 mAhg-1 at 50 mAg-1 and 1 Ag⁻¹, respectively, and showed no obvious capacity degradation after more than 2500 cycles at 1 Ag-1.

3.6 Quinones

The quinone materials are regarded as the most important electrode materials in the carbonyl electrode materials for secondary batteries owing to their higher redox potentials and higher theoretical capacities compared with other imides and conjugated carboxylate-based electrodes. This is because they are resource abundant and have environmental benignity. They can be extracted or refined from many plants. The quinone electrode materials can be categorized into two different groups; small molecule quinone electrodes (e.g. multi-carbonyl quinones, fused hetero aromatic quinones, substituted quinones, oxocarbon salts and carboxylate quinones) and quinone polymer electrodes. Small molecule quinones with high theoretical capacities, such as benzoquinone, anthraquinone and phenanthrene quinone [61, 62], can generate multi-electron transfer reversible reaction. benzoquinone as the basic structural unit, they can be derived from multicarbonyl quinone compounds [63, 63]. For the fused hetero aromatic quinones [64], it should be emphasized that the introduction of O, S, N elements on hetero aromatic quinone can obviously enhance discharge potential of the quinone electrodes production on an industrial level. The quinone compounds that are n-type organic compounds have the oxidative ability and enable reversible electron transfer reaction. This can be extended to multi-electron reactions if more carbonyl groups are introduced on quinones, which can form multivalent charged anions, leading to a high theoretical capacity for the quinone materials. Taking benzoquinone as an example, during the discharge process, one of carbonyl groups accepts one electron and forms a free radical anion, and the other carbonyl group continues to obtain one electron, producing divalent anions. The redox mechanism of the quinone electrode materials is based on electron transfer reaction therefore they can not only be applied in Lithium ion batteries/sodium ion batteries but also can be extended to magnesium, zinc batteries and supercapacitors. The first application of quinone material in a battery system was reported by Williams in 1970 [36] where they used dichloroisocyanuric acid (DCA) as the active component. The DCA electrode showed an irreversible electrochemical redox reaction in an organic non-aqueous electrolyte. Chloranil as an electroactive material reported by Alt et al. was used in the first secondary battery [65]. The quinone electrode displayed 95%retention of capacity after 50 cycles at a current rate of 0.5 C in both aqueous acid and organic electrolytes. In general, the quinone electrode materials showed the advantages of high theoretical capacity and high resource availability. Although the three main challenges of the organic materials such as the dissolution of the quinone electrodes in organic electrolytes, poor electronic conductivity and low discharge plateaus need to be solved before practical application in secondary batteries. One approach to overcome the dissolution problem of small molecule quinoide structures in the electrolyte and the consequent capacity loss is the incorporation of the quinone structure into a polymeric moiety. This approach is accompanied with challenges. The choice of the appropriate polymerization technique is difficult because of both the radical scavenging properties of quinones and their electrophilic and nucleophilic moiety. Generally speaking, high yield polymers depend on polymerization techniques.

4. Conclusion and Future Perspective

Application of carbonyl compounds as active components in rechargeable batteries over the last three decades have come a long way owing to their environmental benignity and resource availability. This study reveals that various classes of carbonyl compounds can be employed

as electrode materials with high theoretical capacity but their electrochemical performance is dependent on the functionality of the carbonyl compounds themselves. Of all the classes of carbonyls considered, quinone structures in our opinion, are the most promising class of carbonyls to be explored in this regard but the progress achieved hitherto is far from ideal, owing to the fact that quinone just like most other organic compounds, is soluble in the system electrolyte, has low electronic conductivity and discharge potential. However, these challenges could be addressed by modifying their structures with carbon-based materials such as graphene, carbon nanotubes and porous carbon. This can not only improve the conductivity but will also inhibit dissolution of the quinone themselves.

From a sustainable point of view, carbonyl compounds can be obtained from a wide range of renewable sources including plants, and various extraction techniques have been revealed on how to utilize this important class of organic compounds in energy storage, it still remains a major challenge for carbonyl compounds to achieve high energy, power density and material activity. There are still many possibilities to improve the electrode kinetics and the capacity of organic carbonyl compounds to discover even more efficient electroactive structures.

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