




New Avenues for Organic Redox Materials as Sustainable Lithium-ion Battery Cathodes

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Abstract As the demand for electrification of means of transportation and storage of electrical energy for later use is skyrocketing, rechargeable Li-ion batteries (LIBs) are at the heart of this revolution. Acknowledging the carbon footprints, environmental concerns and cost of the commercial cathode materials, this is the high time to advocate sustainable alternatives. This review aims at establishing the potential of organic redox-active molecules as a burgeoning class of sustainable solid cathode materials for LIBs. The materials are classified according to their structural features (molecules, metallo-organic complexes, and organic/metal-organic frameworks) and electrochemical performance to lay emphasis on practical applications and bottlenecks in commercialization. However, these materials are still in early stages of development, and new frontiers have been explored in the last five years.

Key words: batteries, Li-ion batteries, organic materials, energy storage

Introduction

While scientists, researchers, technocrats, and entrepreneurs are pondering over climate change, pollution, resource depletion and energy security, green energy storage technologies (ESTs) are providing a window of opportunity to take the bull by the horns.¹ Having a squint at the present ESTs, Li-ion batteries (LIBs) are leading from the forefront due to their wide range of applications, ranging from pocket devices to grid-scale installations.² Beginning with powering electronic gadgetry such as phones and computers a decade ago, at present LIBs are running our cars and homes with

plans to fuel air and space travel in future.³ Consequently, aggressive research activities on electrode materials and the surge in the LIB production have led to 85% reduction in prices over the last decade. As the demand for energy and its storage is increasing exponentially, the world energy storage market is estimated to grow as much as \$426 billion in the next decade where the LIB market has already touched the \$50 billion mark worldwide.⁴ Right from the beginning of the LIB age, it relies mostly on uninterrupted supply of expensive metals such as Co, Ni, Mn, Al, Fe, Cu and Li. On the flipside, the cathode materials based on these metals are now posing serious concerns related to availability of raw material, mining and synthesis cost, supply chain bottlenecks, geopolitical situations, and issues surrounding end of life such as toxicity and recycling.⁵ Therefore, the immediate challenge is not only to address these concerns but also to upgrade the storage capacity, the cell voltage and durability of the present LIBs to meet the future demands. This brings the concept of greener and sustainable batteries containing environmentally benign, economic, abundant, and safer organic electroactive materials. This review focuses on presenting an overview of state-of-the-art of organic cathode materials applied in various forms such as small molecules, metal complexes, and organic/metal-organic frameworks (MOFs) in LIBs over the period of last five years.

Organic materials are composed of earth-abundant elements such as H, C, N, O, S and P. Apart from lower environmental footprints, energy-economic synthesis, cost and recycling, one of the most attractive features of organic redox materials is high structure and property tunability (Figure 1).⁶ Recent efforts in developing organic materials were mainly aimed at structural modification of organic molecules containing the most common redox unit, i.e. carbonyls.⁷ This is because the charge/discharge potential, specific capacity, cycling stability and cycling rate of the Li-ion cells are dictated by the molecular structure of the material. For

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