

Perspective

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Recent research on emerging organic electrode materials for energy storage

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Abstract

Due to the growth of the demand for rechargeable batteries in intelligent terminals, electric vehicles, energy storage, and other markets, electrode materials, as the essential of batteries, have attracted tremendous attention. The research of emerging organic electrode materials in batteries has been boosted recently to their advantages of low cost, environmental friendliness, biodegradability, and designability. This manuscript highlights and classifies several recent studies on organic electrode materials and lists their potential applications in various battery systems. Finally, the challenge and perspective of organic electrode materials are also summarized.

Keywords: Organic electrode materials, batteries, energy storage, low-cost electrode materials, environmentally friendly materials

INTRODUCTION

With more attention to green energy by society, wind power^[1], photovoltaic power^[2], tidal power^[3] generation are gradually squeezing out of traditional power generation. These power generation systems are unstable, and the power supply and user demand are inconsistent. Thus, power generation systems must be equipped with secondary batteries to store excess electricity^[4], leading to increased demand for energy



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storage batteries. There is also growing demand for energy storage batteries in other markets, such as smart electronic devices and new energy vehicles. These smart devices use lithium-ion batteries and polymer lithium-ion batteries, while Uninterrupted Power System and vehicle start-up systems commonly use lead-acid batteries^[5].

Early batteries were composed of metals with different electrochemically active potentials, such as the famous Voltaic Pile. Later, the battery developed into a rechargeable type. The electrode materials consist of metal oxides, metal salts, metals, *etc.*^[6-9], have been applied in rechargeable batteries such as the lead-acid battery (PbO₂-Pb), nickel-cadmium battery [NiO(OH)-Cd], lithium-ion battery (LiCoO₂-C, LiFePO₄-C, *etc.*). Most of the electrode materials of these batteries belong to inorganic materials, exhibiting high price, heavy metal toxicity, environmental pollution, and degradable recycling, which need further attention for energy storage applications^[10,11]. Now, lithium-ion batteries and lead-acid batteries currently have the highest market share^[12,13], and lead-acid batteries have achieved a closed loop of production-using-recycling-reproduction. In contrast, lithium-ion batteries have not yet formed an effective market-oriented recycling and reuse program, which leads to serious waste and pollution of electrode materials. Moreover, the resource of cobalt and lithium elements for the fabrication of lithium-ion batteries is unevenly distributed around the world, leading to the unstable cost of electrode materials^[14,15].

With the increasing demand for lithium-ion batteries in electric vehicles, subsequent production requires a large amount of lithium^[15], cobalt, nickel, manganese, and other elements. At the same time, after the service life of a large number of lithium-ion batteries in electric vehicles expires, there is no effective recycling procedure, and the batteries are directly discarded, causing serious pollution and waste of electrode materials. Therefore, it is crucial to look for novel, low-cost, non-toxic, biodegradable electrode materials that can be designed to meet our needs for next-generation batteries.

Organic electrode materials have changed this situation^[16]. Organic materials also have oxidation and reduction electrochemical activity and can be used as anode and cathode materials for rechargeable batteries. These materials can be derived from biomass, petroleum industry by-products, *etc.*, regardless of geographical restrictions. The cost of organic electrode materials is affected by the production process. Under the condition of mature and mass production processes, the cost becomes very low. At the end of the battery life, the organic electrode material can be automatically degraded or manually processed into non-toxic and pollution-free substances. At the same time, organic materials can be used in different battery systems by adjusting the length of the carbon chain, functional groups, and other structures. The advantages of emerging organic electrode materials and their applications are summarized as follows.

LOW COST

The low cost of organic electrode materials allows them to be used in various types of battery systems. Typically, Quinone materials have been successfully used in flow batteries (Huskinson *et al.*^[17], 2014). The electrode material was 9, 10-anthraquinone-2, 7-disulphonic acid [Figure 1A], which has a rapid and reversible redox reaction and showed a 0.6 W cm⁻² at 1.3 A cm⁻² power density.

Organic cathode materials, along with low-cost anode materials (aluminium, zinc, *etc.*^[18]), can further reduce battery costs. In 2018, Kim *et al.*^[19] applied a redox-active triangular phenanthrenequinone-based macrocycle [Figure 1B] as cathode material into an aluminum battery. The large triangular structure of the material can allow the reversible embedding and detachment of aluminum ions in the redox reaction. This material has very good electrochemical properties, displaying a high revisable capacity of 110 mAh g⁻¹, and a long lifespan of up to 5000 cycles. The electrochemical performances of another two phenanthrenequinone-

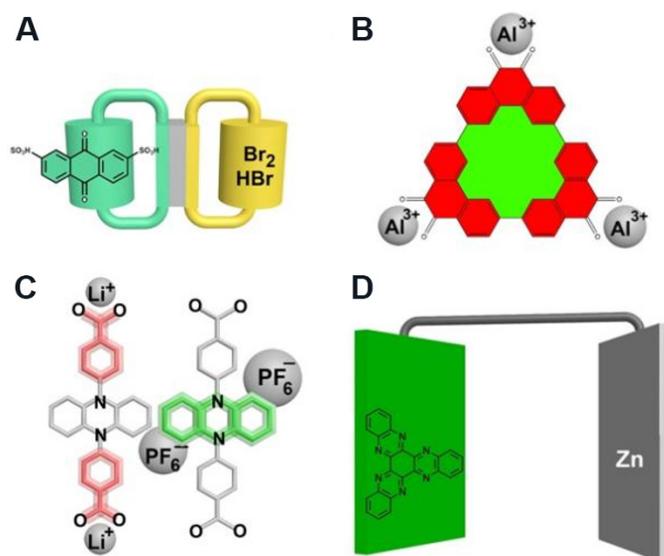


Figure 1. Structure formula of some low-cost organic electrode materials. (A) 9, 10-anthraquinone-2, 7-disulphonic acid for flow battery. (B) A redox-active triangular phenanthrenequinone-based macrocycle. (C) 4, 4'-(phenazine-5,10-diyl) dibenzoate. (D) The phenazine material for Zn-organic battery.

based materials with a monomer and linear trimer structure have also been studied. The triangular macrocycle structure showed the best result because of its layered architecture and the minimization of solvent effects.

In a symmetrical battery system, the low cost of organic electrode materials will be more obvious because the anode and cathode are the same. Dai *et al.*^[20] investigated a dual-ion organic symmetric battery system, which used a molecular anion of 4, 4'-(phenazine-5, 10-diyl) dibenzoate [Figure 1C]. For taking advantage of the phenazine and benzoate moieties parts and their electrochemically reversible redox reactions, an artificial bipolar molecular anion was designed and synthesized. The molecular anion had a high average discharge voltage of 2.5 V and an energy density of 127 Wh kg⁻¹ at 1 C discharge current.

Zinc-organic battery system is another hot area of organic electrode materials. Tie *et al.*^[21] studied a phenazine material [Figure 1D] in the aqueous Zn-organic battery system and got an initial discharge capacity of 405 mAh g⁻¹ at 100 mA g⁻¹. The capacity retention was 93.3% after 5000 cycles at 5 A g⁻¹.

Costs of an organic cathode material (p-chloranil) and two common inorganic materials were calculated [Table 1]. P-chloranil is a mature organic material that represents the cost of organic electrode materials when they are mass-produced. P-chloranil has been successfully used to the cathode in aqueous batteries by Yue *et al.*^[22]. In his work, the battery uses a p-chloranil material as a cathode and lead as an anode. The p-chloranil has a specific capacity of 200 mAh g⁻¹, and the battery discharge voltage platform is around 1 V. Considering the unit price of p-chloranil is 10 CNY (Chinese Yuan) kg⁻¹^[23], its watt-hour cost is:

$$\frac{10 \text{ CNY kg}^{-1}}{200 \text{ Ah kg}^{-1} \times 1 \text{ V}} = 0.05 \text{ CNY Wh}^{-1}$$

Table 1. Cost per watt-hour comparison of three cathode materials

Materials	Battery systems	Costs of cathode materials per Wh (CNY)
P-chloranil	P-chloranil-Pb	0.05
PbO ₂ (based on raw material lead ingot)	PbO ₂ -Pb	0.06
LiCoO ₂	LiCoO ₂ -graphite	0.22

In the PO₂-Pb lead-acid battery system, the specific capacity of positive electrode material (PbO₂) is about 100 mAh g⁻¹, and the battery discharge voltage platform is about 2 V. 207.2 kg Pb raw material corresponds to 239.2 kg PbO₂, and the unit price of the lead ingot is 13 CNY kg⁻¹[24]. The unit price of PbO₂ (based on raw lead ingots) per watt-hour is:

$$\frac{13.7\text{CNY kg}^{-1} \times \frac{207.2}{239.2}}{100 \text{ Ah kg}^{-1} \times 2 \text{ V}} = 0.06 \text{ CNY Wh}^{-1}$$

In the LiCoO₂-graphite lithium-ion battery system, the practical, specific capacity of LiCoO₂ is about 140 Ah kg⁻¹, and the battery discharge voltage platform is about 3.8 V. The unit price of LiCoO₂ is about 110 CNY kg⁻¹[25]. Thus, the cost per watt-hour of lithium cobaltate (LCO) in a battery system is:

$$\frac{110 \text{ CNY kg}^{-1}}{130 \text{ Ah kg}^{-1} \times 3.8 \text{ V}} = 0.22 \text{ CNY Wh}^{-1}$$

SAFE AND GREEN

Organic materials can be designed to be non-toxic and environmentally friendly, which gives them advantages in terms of safety and greenery. As an anode material, zinc is also non-toxic and environmentally benign. Therefore, in addition to the low cost, the organic-zinc battery system is also safe and green, which is very suitable for application in wearable electronic devices. Guo *et al.*[26] studied a pyrene-4, 5, 9, 10-tetraone [Figure 2A] cathode and zinc anode battery system in a mild aqueous electrolyte. The organic cathode material exhibited a capacity of 336 mAh g⁻¹ and an energy density of 186.7 Wh kg⁻¹, and over 1000 cycles of life.

Biodegradability is another green advantage of organic electrode materials. Because organic materials can be designed into a polypeptide polymer structure, this structure can spontaneously degrade in nature, avoiding the environmental pollution problem. Nguyen *et al.*[27] synthesized polypeptide materials [Figure 2B] and designed a metal-free organic radical battery, which showed a capacity of 37.8 mAh g⁻¹. They also studied the degradation and toxicity of the materials, and found the cathode polypeptide was deemed non-toxic towards preosteoblast cells, mouse fibroblast cells, and bovine coronary venular endothelial cells, and could be degraded into small molecules.

Aromatic organic materials contain only carbon and hydrogen elements, without oxygen, nitrogen, sulfur, and other elements. Compared with heavy metals and nitrogenous organic matter, they are less toxic, and some of them are even non-toxic. If the materials are used in batteries, it will be very environmentally friendly. Traditional graphite anode materials also contain only carbon but are currently used only in lithium-ion batteries. The large size of sodium and potassium ions limits the use of graphite in the anode electrode of such batteries, while aromatic organic materials solve this problem. Eder *et al.*[28] assessed the

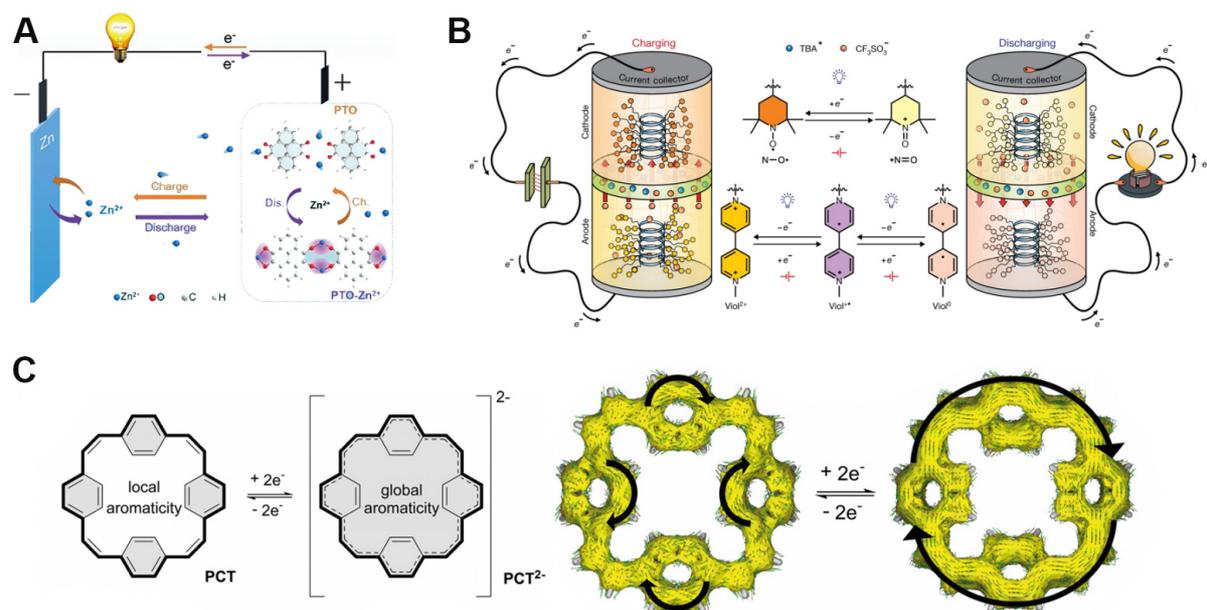


Figure 2. Structural formula of some environment-friendly organic electrode materials. (A) Pyrene-4, 5, 9, 10-tetraone^[26]. (B) Polypeptide anode and cathode polypeptide materials^[27]. (C) [2.2.2.2]paracyclophane-1, 9, 17, 25-tetraene^[28].

possibility of [2.2.2.2]paracyclophane-1, 9, 17, 25-tetraene [Figure 2C] in sodium-ion batteries, which showed a good potential value.

DESIGNABLE STRUCTURE

Organic molecules have strong design flexibility by adding and subtracting carbon chains and changing functional groups. These structural changes can alter the electrochemical properties of materials and expand the application fields. In particular, organic electrode materials consist of carbonyl and π conjugate structures. The carbonyl group is the site of the REDOX reaction, and the π conjugate structure provides high conductivity and overall material stability. For example, when p-benzoquinone and 3, 4, 9, 10-perylene-tetracarboxylic dianhydride were used as cathode materials, their electrochemical reaction mechanisms are shown in Figure 3A. The mechanism of functional group REDOX of organic materials is different from the interlayer embedding and exhalation mechanism of layered inorganic materials. For example, in sodium-ion batteries and potassium-ion batteries, because the ionic radius of sodium-ion and potassium-ion is much larger than that of lithium-ion, the traditional lithium-ion battery electrode materials cannot be directly applied to sodium-ion batteries and potassium-ion batteries. Organic electrode materials can be directly applied to lithium-ion batteries, sodium-ion batteries, and potassium-ion batteries with the same storage mechanism. Cong *et al.*^[29] investigated the redox performance of four aromatic hydrocarbons, including biphenyl, naphthalene, triphenylene, and phenanthrene [Figure 3B]. The redox potentials of these anode materials were 0.35, 0.42, 0.45, and 0.46 V vs. K/K⁺, respectively. It's found that the potassium biphenyl showed the best electrochemical performance as an anode in K-O₂ batteries. By adding the electron-donating methyl group to the benzene ring, the biphenyl redox potential can be reduced from 0.35 V to 0.29 V vs. K/K⁺.

3, 4, 9, 10-perylene-tetracarboxylic acid-dianhydride (PTCDA) is another organic cathode material used in lithium, sodium, and potassium ion batteries. Tong *et al.*^[30] polymerized PTCDA monomers with different short alkyl chains [Figure 3C] and studied their electrochemical properties and redox kinetic differences in potassium-organic batteries. They achieved an energy density of 113 Wh kg⁻¹, power of 35.2 kW kg⁻¹, and

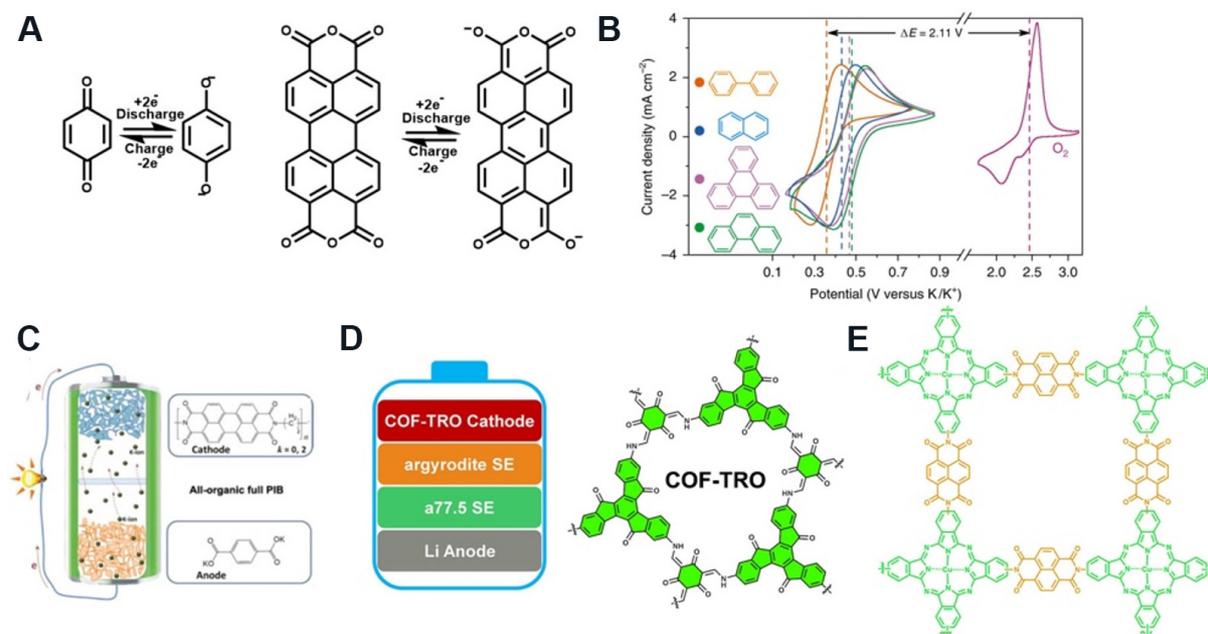


Figure 3. Structural formula and REDOX mechanism of some designable organic electrode materials. (A) REDOX mechanism of p-benzoquinone and 3, 4, 9, 10-perylenetetracarboxylic dianhydride. (B) From top to bottom are biphenyl, naphthalene, triphenylene, and phenanthrene^[29]. (C) Polymerized PTCDA monomers with different short alkyl chains^[30]. (D) The truxenone-based covalent organic framework material^[31]. (E) The conjugated microporous polymers material^[32].

excellent performance at an ultrahigh discharge current density up to 147 C. The battery capacity loss was near 0 after 1000 cycles at a current density of 7.35 C.

Covalent organic framework materials have a tunable chemical structures, which they can be used in different batteries for various purposes. Yang *et al.*^[31] reported a truxenone-based covalent organic framework (COF-TRO) material [Figure 3D] in all-solid-state lithium-ion batteries as the cathode. The material showed a capacity of 268 mAh g⁻¹, and 99.9% capacity retention after 100 cycles at a 0.1 C rate. The electrode material was synthesized from TRO and 1, 3, 5-triformylphloroglucinol by polymerization under vacuum heating.

Conjugated microporous polymers (CMPs) materials also have redox-active properties and can be tunable. Wang *et al.*^[32] synthesized and studied the CMPs by integrating copper (II) tetraaminephthalocyanine (CuTAPc) and 1, 4, 5, 8-naphthalenetetracarboxylic dianhydride (NTCDA) units into the RCMPs (CuPcNA-CMP, Figure 3E). They obtained a capacity of 245.3 mAh g⁻¹ at the current density of 0.1 A g⁻¹, and 89% capacity retention after 500 cycles. The rate performance was also outstanding, achieving 125.1 mAh g⁻¹ at 5 A g⁻¹. And this electrode material was prepared by the dissolution of CuTAPc and NTCDA into DMAc, and then condensation polymerization under vacuum heating. Similar to covalent organic frame materials, CMPs are also organic polymer materials. Their specific capacities are very similar. The theoretical and practical capacities of the same kind of organic electrode materials depend on the number of REDOX functional groups and the molecular weight of the molecule. Therefore, a higher number of electrochemically active functional groups and lower relative molecular weight usually predict higher specific mass capacity.

CRYOGENIC SUPERIORITY

Organic electrode materials have been widely used in cryogenic batteries because of their easy bonding and detachment to protons. Organic materials that have REDOX activity by binding and releasing protons become a good choice for low temperature battery electrode materials. Based on this principle, Tie *et al.*^[21] successfully prepared enhanced zinc-organic batteries with diquinoxalino [2,3-a:2',3'-c] phenazine (HATN) as an electrode material. Tie *et al.*^[33] also summarized the design strategy of a high-performance aqueous Zn/organic battery. Because aqueous solutions containing acids have a much lower freezing point, and the apparent movement rate of protons in water is very fast, the proton aqueous solution becomes one of the best low temperature electrolytes. Sun *et al.*^[34] studied alloxazine [Figure 4A] material as the anode in aqueous proton battery. This battery delivered a high specific energy density of 110 Wh kg⁻¹ at a power density of 1650 W kg⁻¹ at -60 °C. When the temperature is lower to an extremely low temperature (-90 °C), the material could also exhibit a discharge capacity of 85 mAh g⁻¹.

Another cathode material, p-chloranil, was successfully applied in an ultralow temperature aqueous battery system. Yue *et al.*^[22] reported the p-chloranil [Figure 4B] organic material composite with reduced graphene oxide (PCHL-rGO) used in aqueous Pb-quinone batteries. The Pb/PCHL-rGO batteries exhibited a discharge capacity of 87 mAh g⁻¹ at a current of 0.1 A g⁻¹. And the cycle retention was 97% after 500 cycles at 0.5 A g⁻¹ under -70 °C. This battery system not only has good electrochemical performance at low temperatures, but also is very inexpensive for practical application.

CONCLUSION AND OUTLOOK

Compared with traditional inorganic electrode materials, organic electrode materials have many advantages in cost, safety, environmental friendliness, design diversity, and low temperature battery applications. On the other hand, organic electrode materials also have disadvantages, mainly in poor conductivity, solubility, and low voltage platform. Researchers are constantly working to overcome these shortcomings.

Not all organic electrode materials conduct electricity well, so many organic electrode materials must be mixed with a large number of conductive carbon materials when used in batteries, which affects the overall volume energy density and mass energy density of the battery. Therefore, it is crucial to enhance the intrinsic electronic conductivity of organic materials so that the electrochemical properties can be boosted. Conjugate π structures combined with various functional groups may be an important direction in the future because of their excellent electrical conductivity.

Furthermore, solubility in the electrolyte is another serious problem for organic electrode materials, especially smaller organic monomers. In lithium-ion, sodium-ion, and potassium-ion batteries, organic materials such as small molecules tend to dissolve in the organic electrolyte during charge-discharge cycles. Combining organic materials with a small amount of highly conductive material (such as graphene^[15], carbon nanotubes, MXene, *etc.*) seems to be an effective way to solve the problem of poor electronic conductivity as well as the solubility of the electrode active materials. Polymerization of organic monomers into polymers can also solve the problem of material dissolution, but the method of polymerization needs to be explored by researchers. At present, many polymerization methods use organic solvents, some of which are often expensive or harmful to organisms. Finding a method using water polymerization or solvent-free polymerization has become an effective solution.

Compared with cathode materials such as LiCoO₂ and ternary materials, organic cathode materials have a lower voltage platform. Lithium-ion batteries made of LiCoO₂ have an open circuit potential above 4 V, while the voltage of most organic lithium-ion batteries is between 2 V and 3 V. Due to the decrease of the

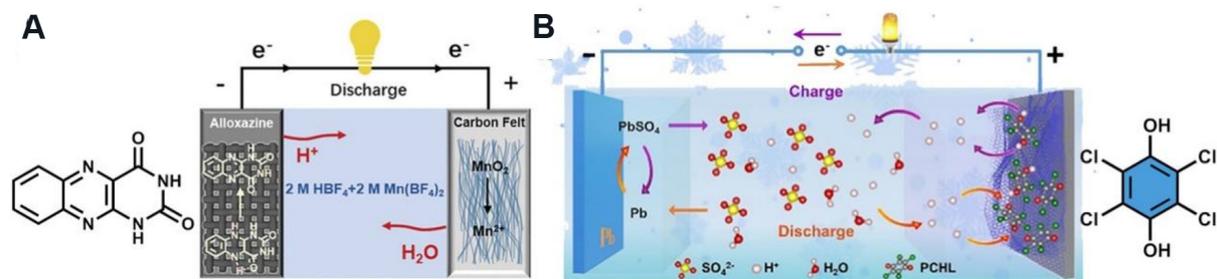


Figure 4. Structural formula of organic electrode materials in cryogenic batteries. (A) Alloxazine^[34]. (B) P-chloranil^[22].

voltage platform, the energy density of the battery decreases, which affects the cost advantage of the application of organic materials in the battery. This requires researchers to develop novel REDOX groups and combine them with organic host structures.

Although there are still many challenges in developing high-performance organic electrode materials, it is no doubt that the electrochemically active organic electrode materials will attract more attention in the future. With the increasing demand for rechargeable batteries, the abandonment of traditional battery materials and environmental pollution problems, and the further reduction of the cost of organic materials, the large-scale application of organic electrode materials in various commercial batteries is promising.

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Authors' contributions

Selected the topic and conceived the structure of this paper: Huang T, Liu H, Wang G

Searched references and wrote the draft: Huang T, Long M, Xiao J

Polished this manuscript: Liu H, Wang G

The paper was revised by all authors.

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Conflicts of interest

Not applicable.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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