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Anode Materials for Aqueous Ammonium-Ion Batteries: Recent Developments and Energy Storage Applications

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Abstract. Aqueous ammonium-ion batteries (AAIBs) have drawn attention for safe, low-cost electrochemical storage, since aqueous electrolytes are chemically stable and NH_4^+ transport can be promoted by hydrogen-bond networks. Although many high-performance anodes have been reported, their practical performance is still restricted. Heteroatom doping, interlayer expansion, and composite design are commonly used to tune the electronic structure and diffusion pathways of anode materials. These approaches improve conductivity and ion mobility, but low energy density, active-material dissolution, and cycling instability remain unresolved. This review discusses recent work on AAIB anodes in four material families: metal compounds, two-dimensional/porous materials, carbon-based materials, and organic materials. The corresponding NH_4^+ storage mechanisms and present challenges are summarized to support the further development of high-performance AAIB systems.

Keywords: Aqueous ammonium-ion batteries, Anode materials, Electrochemical energy storage

1. Introduction

With continued economic and social development, reliance on conventional fossil fuels has increased carbon emissions and placed greater pressure on the environment. To mitigate this problem, industrial energy systems are being adjusted toward cleaner utilization and toward the targets of "carbon neutrality" and "net-zero emissions." Renewable sources, especially wind and solar energy, have therefore developed rapidly [1-3]. Their large-scale and continuous use, however, is restricted by site dependence, output fluctuation, and intermittency [4]. Rechargeable batteries, which combine high energy-conversion efficiency with flexible deployment, have consequently attracted extensive interest for household and grid-scale energy storage [5-7].

Lithium-ion batteries are already widely commercialized, but their high cost, flammable organic electrolytes, and associated environmental concerns remain difficult to avoid [8, 9]. In contrast, aqueous non-metallic ion batteries offer several advantages. Among them, aqueous ammonium-ion batteries (AAIBs) are particularly attractive because NH_4^+ has good chemical stability (resistance to H_2 and N_2), low molecular weight, and a small hydrated ionic size [10-12]. Unlike spherical metal ions, the tetrahedral NH_4^+ ion tends to undergo oriented intercalation, and its storage process is governed mainly by hydrogen-bonding interactions, which support rapid ion diffusion and reversible

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redox kinetics. In addition, ammonium salts dissociate easily in water, providing high ionic conductivity [13-15]. Nevertheless, it remains difficult to obtain anode materials with high NH_4^+ storage capability. Poor electrical conductivity, sluggish ion diffusion, limited interlayer spacing, material dissolution, and irreversible phase transitions can all shorten cycling life. This review therefore summarizes AAIB anode materials and aims to guide both rational material design and mechanistic understanding.

2. Classification of ammonium-ion battery anode materials

Figure 1 places the currently reported AAIB anodes into four material families. In this review, they are discussed as metal compounds, novel two-dimensional/porous materials, carbon-based materials, and organic materials.

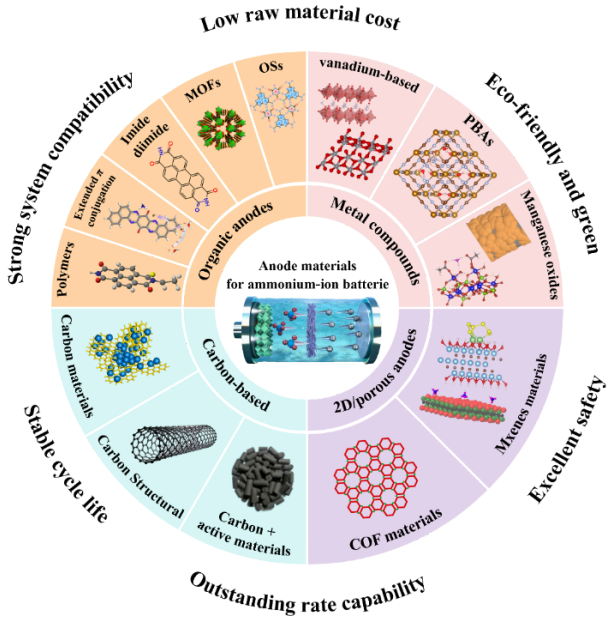


Figure 1. Classification of anode materials

2.1. Metal compounds

Metal compounds provide a broad compositional space, adjustable crystal chemistry, and usually high theoretical capacities. Their performance can be further tuned through cation/anion coupling, interfacial control, and kinetic regulation, which helps improve ammonium-storage capacity, cycling life, and adaptability under demanding conditions.

2.1.1. Vanadium-based materials

Vanadium-based compounds are valuable because vanadium can switch among several valence states and many compounds have layered motifs, features that favor reversible charge storage. In actual electrodes, however, vanadium oxides still suffer from low electronic conductivity, sluggish reaction kinetics, and dissolution. Researchers therefore rely on morphological control and framework modification to make NH_4^+ insertion/extraction more effective. Zhang et al. [16] introduced polyaniline (PANI) into V_2O_5 by thermal treatment to produce P-VO_x composites. The polymer expanded the interlayer distance from 1.15 nm to 1.55 nm, and the formation of V-N bonds

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and oxygen vacancies provided more favorable pathways for electron transport. The P-VO_x-3 electrode consequently delivered 223.5 mAh g^{-1} and retained 71.1% of its capacity after 100000 cycles. Vanadates, including $Zn_3V_3O_8$ and MnV_2O_4, are another important branch of vanadium-based anodes. Their particles may agglomerate during operation, blocking diffusion channels and favoring by-product formation. For this reason, morphology and structure tuning are also needed for vanadates. Wen et al. [17] compared well-dispersed $Zn_3V_3O_8$ nanorods with aggregated MnV_2O_4 nanoparticles as AAIB anodes. $Zn_3V_3O_8$ showed better electrochemical behavior, retaining about 82.6% of its initial capacity after long-term cycling. The full cell based on $Zn_3V_3O_8$ combined high energy density with relatively high peak power output. Ex situ analysis further captured the first-charge phase evolution of $Zn_3V_3O_8$ and the reversible hydrogen-bond changes during NH_4^+ insertion/extraction, giving evidence for the related phase-transition mechanism.

2.1.2. Prussian blue analogs

Prussian blue analogs (PBAs) have also been considered for AAIB anodes, mainly because their framework composition and defect structure can be adjusted without losing the open lattice. Xiong et al. [18] used ab initio molecular dynamics (AIMD) simulations and electrochemical kinetics to clarify NH_4^+ storage in PBAs. They found that connected anionic vacancies provide a permeation network that benefits storage. Modified HD-FePB retained 94.5% of its capacity over 10000 cycles, highlighting the importance of vacancy engineering. This study helped explain how NH_4^+ is stored in Prussian blue electrodes and suggested directions for PBA modification. Xing et al. [19] prepared K-V-Fe PBA nanocubes through a one-step hydrothermal route with oxalic acid as the reducing agent. The material couples the redox contribution of vanadium with the structural robustness of the Prussian blue framework, giving high discharge capacity and good cycling behavior. Vanadium introduction into a three-dimensional open framework thus offers a practical way to screen and optimize PBA-type storage materials.

2.1.3. Manganese oxides

Among metal compounds, manganese oxides (MnO_x) usually offer comparatively high specific capacity. The main difficulty is associated with Mn^{3+}-related Jahn-Teller activity. During charge/discharge, the Mn^{4+}/Mn^{3+} redox process can distort [MnO_6] octahedra, causing lattice strain, amorphization, phase conversion, and even Mn^{2+} dissolution [20]. Chen et al. [21] improved MnO_x electrodes by coating them with reduced graphene oxide (rGO). The electrode delivered 109 mAh g^{-1} at 5 A g^{-1} and retained 92.6% of its capacity after 1000 cycles. The rGO layer provided a conductive pathway and, at the same time, hindered Mn^{2+} migration, so the MnO_x-based anode became more stable. Yang et al. [22] reported amorphous manganese phosphate (MP-20, $(NH_4)_{0.27}MnO_{1.04}(PO_4)_{0.28}$), in which high capacity was linked to an NH_4^+/H^+ co-insertion mechanism. Its capacity reached 299.6 mAh g^{-1} at 1 A g^{-1}, suggesting that this phosphate is a strong candidate for AAIB anodes.

2.2. Novel two-dimensional / porous anode materials

2.2.1. MXene-based electrode materials

For MXene-based electrodes, the layered structure, high conductivity, and fast ion transport are especially useful for AAIB anodes. Wang et al. [23] created a heterostructure by forming Ti-N bonds

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in MXene and confining about 10 nm In_2Se_3 nanoparticles between the layers; the interlayer spacing increased from 9.95 Å to 15.4 Å. The enlarged gallery and interfacial structure promoted ion movement and improved electrochemical response. The electrode reached 1776.1 F g^{-1} at 1 A g^{-1}, and 98.84% of the capacitance remained after 6000 cycles, owing to strengthened hydrogen-bond-assisted NH_4^+ adsorption. Bao et al. [24] studied V_2CT_x MXene and confirmed that NH_4^+ storage proceeds mainly through a pseudocapacitive mechanism. At 1 A g^{-1}, V_2CT_x delivered a relatively high charge-storage capacity, indicating that MXene electrodes can combine rapid transport with stable operation.

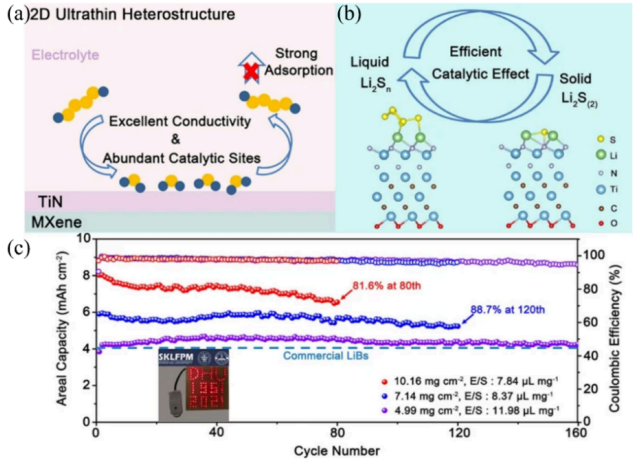


Figure 2. (a) Adsorption and catalytic mechanism of MX-TiN toward LiPSs species. (b) Optimized configurations of Li_2S_6 on MX-phase MX-TiN and $Ti_3C_2T_x$ -type MXene. (c) Cycling performance of the S/MX-TiN cathode at different sulfur loadings [23]

2.2.2. Covalent organic frameworks/ porous organic frameworks

Covalent organic frameworks (COFs) and related porous organic frameworks are useful for ammonium-ion storage because their pore size and chemical environment can be designed in advance, while their rigid skeletons limit dissolution. Tian et al. [25] synthesized the nitrogen-doped HATP-PT COF as an aqueous AAIB anode. Its stable framework and hydrogen-bond coordination enabled NH_4^+ storage across 0.3-1.0 V vs. SCE. The electrode delivered approximately 74 mAh g^{-1} at 1.0 A g^{-1} and remained stable after 20000 cycles. In a full cell with a Prussian blue cathode, 89% of the initial capacity was retained after 20000 cycles. These results show that COF-based anodes can offer both structural robustness and long-term cycling stability for AAIBs.

2.3. Carbon-based anode materials

Carbon-based anodes usually store NH_4^+ by intercalation and surface adsorption, and defect-rich or heteroatom-doped carbons may supply a small additional contribution through weak redox reactions. Because NH_4^+ has a small ionic radius (143 pm), the accessible spacing, pore network, and surface functional groups of carbon materials strongly affect storage behavior [14, 26]. Hydrogen bonding is particularly important in aqueous carbon electrodes; it also differentiates ammonium storage from the storage of $Li^+/Na^+/K^+$ in carbon hosts [26]. Graphite, hard carbon/porous carbon, and heteroatom-doped carbon therefore show different storage mechanisms [27]. The electrolyte also matters, since aqueous and non-aqueous systems can lead to different

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ammonium-ion storage behavior [28]. In practice, carbon anodes often work through several coupled processes rather than a single pathway.

2.3.1. Direct use of "carbon materials" as anodes

In an ammonium-ion hybrid supercapacitor, Wang et al. [28] adopted hierarchical carbon as the anode. The device showed 238.3 F g^{-1} at 2.0 V for 10000 cycles, with an energy density of 66.2 Wh kg^{-1}. Chen et al. [29] used activated carbon cloth in an aqueous hybrid supercapacitor; it delivered 1550 mF cm^{-2} and 861.2 μWh cm^{-2}, while 72.2% of the capacity was retained after 5000 cycles.

2.3.2. Carbon-based "structural skeleton/conductive framework" anodes

Huang et al. [30] grew one-dimensional conjugated MOFs on porous carbon nanofibers to form HPCNFs@Ni-BTA, where the conductive carbon framework facilitated NH_4^+ diffusion. The electrode gave 678.5 F g^{-1} at 0.5 A g^{-1} and 220.1 F g^{-1} at 10 A g^{-1}. The assembled supercapacitor further delivered 156 F g^{-1} at 0.3 A g^{-1}, and its peak energy density reached 48.8 Wh kg^{-1}.

2.3.3. "Carbon + active materials" composite anodes

Combining a high-capacity active component with a conductive carbon matrix is another way to balance capacity and electron transport. Gong et al. [31] grew VO_2 directly on graphene to obtain a free-standing VO_2/rGO aerogel. This composite anode achieved 655 mAh g^{-1} at 0.5 A g^{-1}.

2.4. Organic anode materials

Organic anodes rely mainly on reversible redox reactions at carbonyl, imine, and related functional groups to store NH_4^+. They are attractive because their molecular structures can be adjusted, allowing capacity, conductivity, ion transport, and reaction kinetics to be tuned together. Proper molecular design is therefore critical for improving rate performance and cycling life [32].

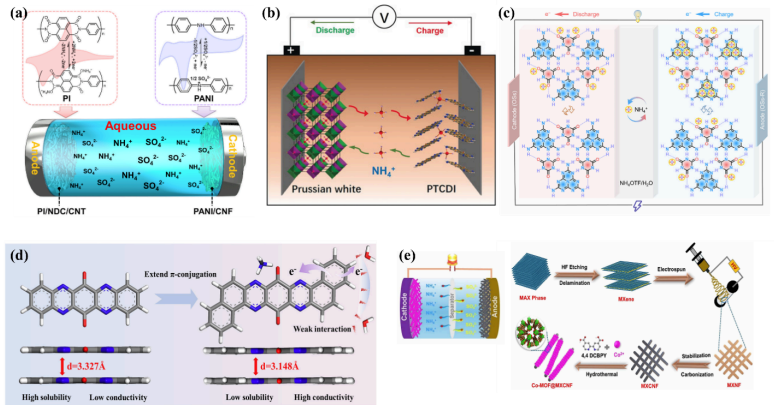


Figure 3. (a) Schematic of the full organic aqueous dual-ion battery with PI/NDC/CNT anode and PANI/CNF cathode [33] ; (b) Structure schematic of the aqueous ammonium-ion battery using ammonium-based Prussian blue analogs as the cathode and PTCDI as the anode [34] ; (c) Structure schematic of OSs||OSs-R battery [35] ;(d) Schematic representation of the effect of extended conjugation on the electrochemical behavior of organic molecules [36] ; (e) Schematic of Co-MOF@MXCNF//MXCNF asymmetric supercapacitor and its synthesis process [37]

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2.4.1. Polyimide/polymer-based materials

Polyimide (PI), obtained from naphthalene tetracarboxylic acid dianhydride precursors, can reversibly host NH_4^+ and is therefore a representative polymer anode for NH_4^+ batteries. Zhang et al. [38] prepared PI anodes through solution polymerization and showed that NH_4^+ storage proceeds through a two-step enolization process. The electrode delivered 157.3 mAh g^{-1} at 0.5 A g^{-1} and 107.7 mAh g^{-1} at 10 A g^{-1}, while retaining 86.4% capacity after 10000 cycles. Zhou et al. [33] reported PI/NDC/CNT nanofiber composites, which achieved 161 mAh g^{-1} at 0.5 A g^{-1}, kept 87.9% capacity after 5000 cycles, and still maintained about 60% of the capacity at 10 A g^{-1}.

2.4.2. Small molecule carbonyl/quinone-based organic materials (extended conjugation)

Small organic molecules are prone to dissolution in aqueous electrolytes, so stabilizing their molecular structure is a key concern. One effective method is to extend the conjugated skeleton, which strengthens intermolecular interactions and improves cycling durability. Tang et al. [36] designed TABQ-NQ (benzobenzo[a]benzo[7,8]quinolinyl[2,3-i]phthalimide-8,17-dione), an extended-conjugation molecule that enhances π-π stacking and NH_4^+ binding. The electrode provided 190 mAh g^{-1} at 0.2 A g^{-1} and retained 146 mAh g^{-1} after 650 cycles, corresponding to 76.8% capacity retention.

2.4.3. Imidazole/di-imidazole-based organic materials

Imidazole- and di-imidazole-based organic electrodes have attracted interest because their carbonyl redox reactions can couple with hydrogen bonding and proton transfer. This chemistry matches the small, strongly hydrated NH_4^+ ion, which tends to form hydrogen bonds. Wu et al. [34] built a rocking-chair AAIB using PTCDI as the anode; the cell reached 43 Wh kg^{-1} and retained 67% capacity after 1000 cycles. In concentrated NH_4Ac electrolyte, Tsai et al. [39] showed that PTCDI-rGO delivered 165 mAh g^{-1} at 0.5 A g^{-1} and maintained 72% capacity after 1000 cycles.

2.4.4. Metal-organic frameworks

Metal-organic frameworks (MOFs) and MOF-derived materials offer well-defined pores, abundant active sites, and flexible structural design. These features are useful for addressing slow kinetics in AAIB anodes [40]. By adjusting pore size, interlayer spacing, and surface functional groups, MOFs can accommodate NH_4^+ and increase capacity through combined hydrogen-bonding and redox pathways [41]. Their hierarchical pores also shorten ion/electron transport distances and improve rate performance. Hussain et al. [37] prepared an oxygen-rich Co-MOF with an (O_4-CoN_2) coordination geometry, which enhanced stability, active-site exposure, and electrochemical performance. They further integrated Co-MOF nanoflowers with $Ti_3C_2T_x$ MXene carbonized nanofibers (MXCNF) to obtain a porous Co-MOF@MXCNF heterostructure. The resulting capacitor delivered 980 F g^{-1} at 1 A g^{-1}.

2.4.5. Multiple hydrogen bond self-assembled organic superstructures

Ordered organic superstructures assembled through weak intermolecular interactions provide another route for improving active-site utilization and cycling stability. Liu et al. [35] constructed organic superstructures (OSs) for all-organic ammonium-ion batteries (AOBs) by pairing six-electron melamine units with three-electron cyanuric acid units that act as hydrogen-bond acceptors.

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Intralayer hydrogen bonding and interlayer π-π stacking maintained the ordered structure. As a result, the material delivered 213 mAh g^{-1} and survived 60000 cycles, indicating multi-active-site NH_4^+ storage with a low energy barrier.

3. Conclusion and outlook

Research on anode materials for aqueous ammonium-ion batteries (AAIBs) has expanded the range of usable material systems and has also improved their electrochemical behavior. Even so, AAIB anodes are still far from practical use, because the link between material structure, electrolyte environment, and NH_4^+ storage behavior is not yet fully understood. Problems such as limited energy density, dissolution of active species, side reactions at the electrode surface, sluggish transport, and relatively poor intrinsic conductivity are still frequently observed. Therefore, future anode design needs to give equal attention to capacity, structural stability, dissolution resistance, and continuous ion/electron transport. In situ/ex situ characterization, theoretical calculation, and data-assisted analysis should be used together to identify the real storage process and to guide the design of more reliable electrodes, interfaces, and full cells.

Several directions deserve further attention. Inorganic, organic, and composite anodes should continue to be developed with emphasis on low cost, long cycling stability, high capacity, and possible synergistic storage mechanisms. At the same time, mechanism-based studies are needed to follow NH_4^+ insertion, adsorption, coordination, and transport during cycling, so that material optimization is not only based on capacity values. Electrode-electrolyte interface engineering also remains important, since a stable interface can reduce side reactions, improve kinetics, and slow down the loss of active materials. For full-cell construction, better matching between anodes, cathodes, and electrolytes is required to improve energy density, safety, and service life. In addition, machine learning and high-throughput screening may be useful supporting tools for selecting electrode materials and optimizing cell matching, but their predictions still need careful experimental verification.

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