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Vanadium Oxide–Modified Cathodes for Aqueous Zinc-Ion Batteries: A Review

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Abstract. Aqueous zinc-ion batteries (AZIBs) are gaining traction as a safe, low-cost alternative to lithium-based systems, not least because of their high ionic conductivity. Among the candidate cathode materials, vanadium oxides stand out for their layered/open frameworks, multiple valence states, and high theoretical capacities—V_2O_5 in particular offers a combination of facile synthesis and a two-electron redox chemistry that gives a respectable 589 $mAh \cdot g^{-1}$. In practice, though, its poor electronic conductivity, sluggish Zn^{2+} diffusion, structural degradation upon repeated cycling, and active-material dissolution remain serious bottlenecks. This review summarizes the four main classes of AZIB cathodes and the three prevailing energy-storage mechanisms, then focuses on recent modification strategies for V_2O_5—namely, preparation routes, Li^+-storage/deintercalation behavior, morphology engineering, and conductive composite design—and discusses how each strategy improves electrochemical performance. Despite these advances, long-term structural stability and rate capability at high current densities still fall short of practical requirements. The aim here is to offer a reference for developing better vanadium-based cathodes in the future.

Keywords: aqueous zinc-ion batteries, V_2O_5, vanadium oxides, cathode materials, material modification

1. Introduction

1.1. A brief introduction to chemical power sources

Economic growth and technological progress have pushed energy demand steadily upward, and the search for better storage solutions has turned attention back to electrochemical power sources.

Batteries are generally classified as primary, secondary, or fuel cells; the secondary category includes nickel-cadmium, lead-acid, lithium-ion, flow batteries, and the now-emerging aqueous multivalent-metal systems.

Among the latter, magnesium-, zinc-, and aluminum-based cells are under active investigation [1]. Aqueous dual-ion batteries (ADIBs) have drawn particular interest because the constituent metals are earth-abundant. Zinc, in particular, reacts mildly with water, which simplifies cell assembly—hence the growing focus on aqueous zinc-ion batteries (AZIBs).

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1.2. Overview of AZIBs

Since Yamamoto's first work on neutral secondary Zn–MnO_2 cells in 1986 [2], interest in rechargeable AZIBs has grown considerably. With a theoretical capacity of 820 $mAh \cdot g^{-1}$, they promise substantial energy output for applications ranging from smart devices to small EVs and grid-scale storage. The aqueous electrolyte is not only more environmentally benign than organic alternatives but also offers higher ionic conductivity and better safety. The zinc anode potential (−0.763 V vs. SHE) is low enough to allow reversible Zn stripping/plating in water—something that is not straightforward for many other metal anodes—and the system shows fast ion-transfer kinetics and intrinsic safety [3]. These features make AZIBs strong competitors among the various secondary-battery chemistries.

The cell configuration resembles that of a conventional lithium-ion battery: a cathode with an open crystal lattice, a zinc-salt aqueous electrolyte, metallic zinc foil as the anode, and a glass-fiber separator (Figure 1).

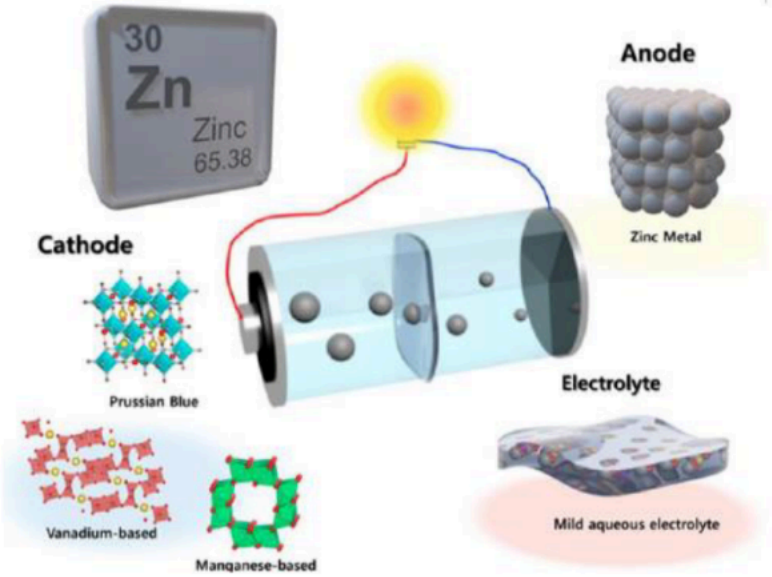


Figure 1. Schematic of an aqueous zinc-ion battery [3]

2. Cathode materials for AZIBs

Despite their promise, AZIBs face practical hurdles. The large radius and high charge density of Zn^{2+} cause strong electrostatic interactions with the host lattice during intercalation, often leading to structural collapse over repeated cycling. Choosing the right cathode material is therefore critical if AZIBs are to become commercially viable [4]. Currently, the main cathode families are manganese oxides, cobalt oxides, Prussian blue analogues, and organic compounds.

The underlying reaction mechanism is still debated and appears to depend on electrolyte pH. Three widely accepted models are illustrated in Figure 2.

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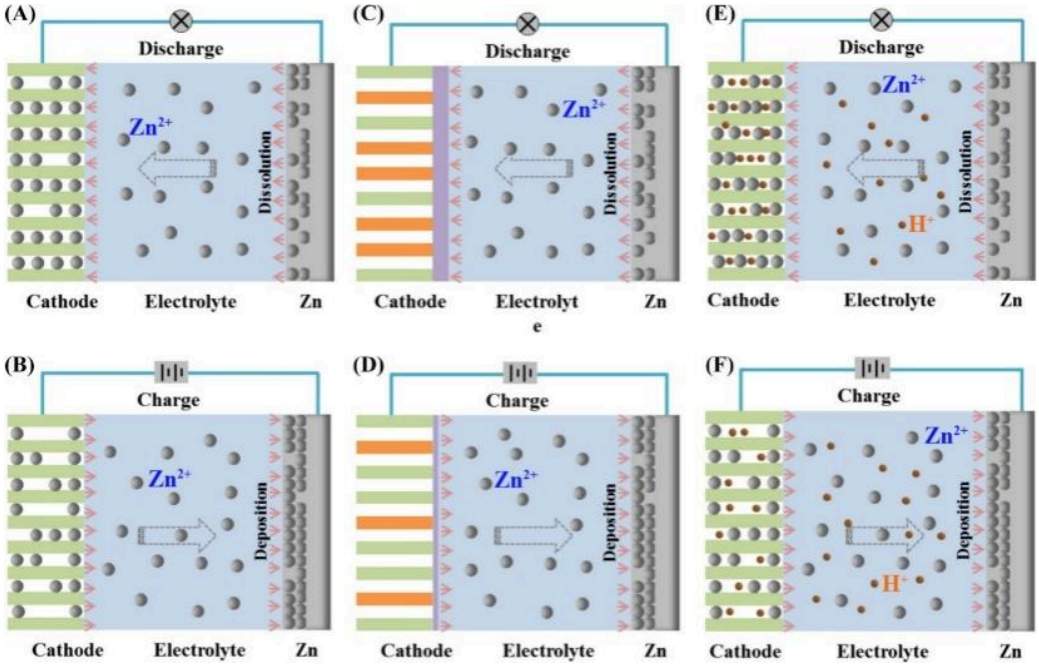


Figure 2. Schematic diagram of zinc-ion battery storage mechanisms; (a) and (b) Zn^{2+} intercalation/deintercalation mechanism; (c) and (d) chemical reaction mechanism; (e) and (f) Zn^{2+}/H^{+} co-intercalation/deintercalation mechanism [4]

(1)Zn^{2+} intercalation/deintercalation

In neutral or weakly acidic media, the dominant process is direct Zn^{2+} insertion/extraction. During discharge, the zinc anode oxidizes to Zn^{2+}, which migrates through the electrolyte and is reduced at the cathode. Upon charging, the reverse occurs.

(2)Co-intercalation of Zn^{2+} and protons

Because H^{+} is much smaller, it can enter the host lattice alongside Zn^{2+}—a phenomenon observed in some Mn- and Co-based oxides. In principle, this co-insertion boosts both capacity and cycling stability, but it is highly sensitive to pH and electrolyte additives.

(3)Chemical conversion

In this case, the cathode undergoes a phase transformation to a new compound during cycling, enabling Zn^{2+} (and sometimes H^{+}) storage. Yong et al. [5], for example, reported reversible conversion between MnO_2 and MnOOH in a $Zn/ZnSO_4/\alpha\text{-}MnO_2$ system, with concomitant proton uptake.:

$$Anode: Zn \leftrightarrow Zn^{2+} + 2e^{-} \tag{1}$$

$$Cathode: H_2O \leftrightarrow H^{+} + OH^{-} \tag{2}$$

$$Mn0_2 + H^{+} + e^{-} \leftrightarrow MnOOH \tag{3}$$

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$$Zn^{2+} + 2OH^- + \frac{1}{3}\,ZnSO_4 + \frac{x}{3}\,H_2O \leftrightarrow \frac{1}{3}\,ZnSO_4[Zn(OH)_2]_3 \cdot xH_2O \quad (4)$$

2.1. Manganese-based oxides

Manganese is abundant, cheap, and relatively non-toxic, and its multiple valence states (+2, +3, +4, +7) allow flexible redox behavior.

Among the various Mn oxides—MnO, MnO_2, Mn_2O_3, Mn_3O_4—most work has centered on MnO_2 because of its layered or tunnel structures. The α-MnO_2 polymorph, with its 4.6-Å tunnels, offers a theoretical capacity of ~200 mAh·g^{-1} and an average voltage of 1.4 V, both favorable for Zn storage.

Jia et al. pre-intercalated Zn^{2+} into MnO_2 to lower the oxygen-adsorption barrier; the modified cathode delivered 127.3 mAh·g^{-1} after 1000 cycles at 1 A·g^{-1} and retained >90% capacity over 100 cycles across a range of temperatures (Figure 3) [6].

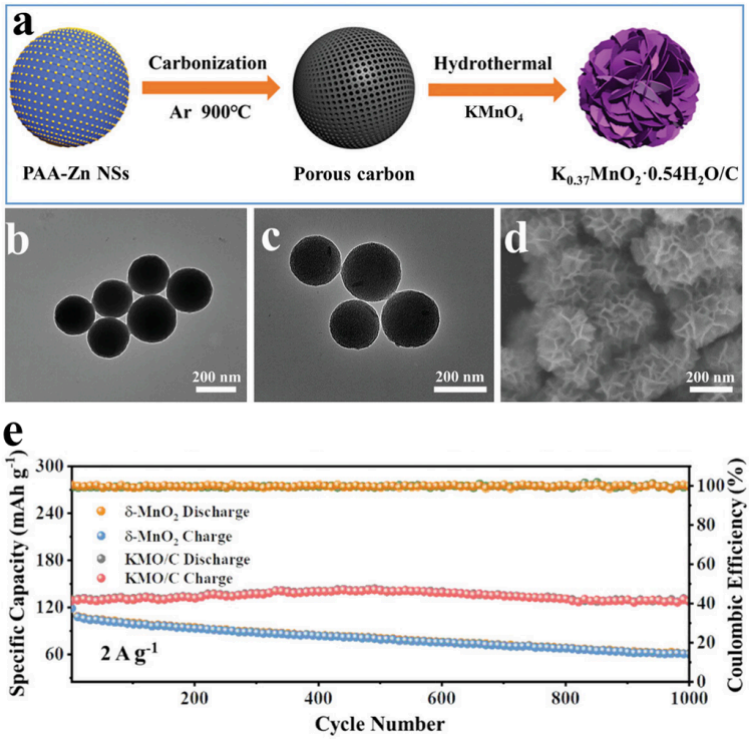


Figure 3. (a) Synthesis of KMO/C; (b,c) TEM of PAA-Zn NSs and CNSs; (d) SEM of KMO/C; (e) long-term cycling of δ-MnO_2 vs. KMO/C [7]

2.2. Organic electrodes

Compared with inorganics, organic materials offer structural tunability, high theoretical capacity, and sustainability.

Examples include conductive polymers such as PANI and PPy, whose extended π-systems facilitate rapid electron transfer.

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2.3. Prussian blue analogues (PBAs)

PBAs—hexacyanometalates with an open, face-centered cubic framework—can accommodate various cations reversibly with minimal volume change. Their large interstitial channels support fast ion diffusion over hundreds of thousands of cycles, and they are easy to synthesize, inexpensive, and environmentally friendly. The electrochemical properties can also be tuned by varying the transition-metal composition [8] (Figure 4).

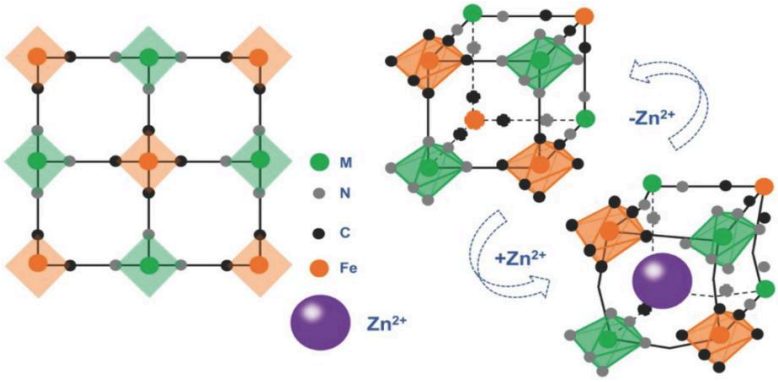


Figure 4. Schematic of Zn^{2+} intercalation into $MFe(CN)_6$ (M = Fe, Co, Ni, Cu, Mn) [8]

2.4. Vanadium-based compounds

After manganese-based oxides, vanadium-based oxides are the next explored as AZIBs cathode materials. Because vanadium-based oxides' layered and tunnel structures, namely flexible and diverse open frameworks provide larger space for Zn^{2+} intercalation/deintercalation, favorable for cathode material stability. Compared with manganese oxides, vanadium oxides in aqueous electrolytes can provide higher specific capacity. But operating voltage is relatively low (relative to zinc is 0.8 V), and its common point with manganese element is possessing multiple redox valence states, abundant resources and low price, therefore vanadium-based oxides are also considered possessing application prospects as aqueous zinc-ion battery cathode materials.

V-O polyhedra through multiple connection modes form different vanadium oxides. The most commonly used for aqueous zinc-ion battery cathode materials are layered or tunnel-type vanadium oxides, for example layered V_2O_5, V_3O_7 and tunnel-structure VO_2, V_2O_3, V_6O_{13} and other materials.

(1)VO_2

VO_2 because of its unique tunnel structure design and relatively high electrode potential, is ideal candidate material for cathode materials. Its wide tunnel structure and stable structure provide superior conditions for Zn^{2+} rapid (de)intercalation. And, VO_2 exhibits up to 646 mAh g^{-1} theoretical specific capacity [9].

(2)V_2O_3

V_2O_3 exhibits corundum-type crystal structure, in which oxygen atoms in close-packed manner arrange into hexagons, while vanadium atoms occupy these hexagons between octahedral interstices' 2/3. In this structure, two VO_6 octahedra through shared edges connect with adjacent octahedra, constructing a tunnel-like three-dimensional network. This structural characteristic provides favorable conditions for ion deintercalation, helps battery performance enhancement.

(3)V_2O_5

V_2O_5 possesses high theoretical specific capacity (589 mAh g^{-1}), can undergo two-electron redox reactions, realizing Zn^{2+} therein rapid electron gain and loss, and possesses easy synthesis, low price

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and special layered structure and other advantages. Therefore, is regarded as a promising aqueous zinc-ion battery cathode material possessing development potential. The above series of advantages make V_2O_5 can serve as aqueous zinc-ion battery's excellent cathode material.

(4)V_3O_7

V_3O_7 possesses special microstructure, it is formed by VO_6 octahedra unique corner-edge connection, and closely connected with VO_5 square pyramids, thus constituting unique two-dimensional V_3O_8 layers. H_2O is located at two-dimensional V_3O_8 layers' both sides, with VO_5 square pyramids in V atoms forming hydrogen bonds, connecting adjacent two V_3O_8 layers [10]. Excellent layered structure provides channels for Zn^{2+} migration, but large-scale application still needs to improve $V_3O_7 \cdot H_2O$ slow diffusion kinetics.

(5)V_6O_{13}

Possessing perovskite-like framework structure monoclinic V_6O_{13} is composed of a series of distorted VO_6 octahedra arranged in zigzag layout, these octahedra through shared edges or vertices form single-layer and double-layer structures. This unique structure provides ample channels for ion diffusion, exhibits remarkable ion diffusion performance. At the same time, V_6O_{13} at room temperature not only exhibits relatively high electrochemical activity, but also possesses good conductivity. However, its intrinsic conductivity is relatively low, which to some extent hinders electron rapid transfer, thereby affects electrochemical reaction effectiveness.

In summary, above vanadium-based oxides due to special layered or tunnel-type structures, are favorable for Zn^{2+} therein shuttling and energy storage, are very promising aqueous zinc-ion battery cathode materials. Among the many vanadium oxides V_2O_5 possesses two-electron redox mechanism and relatively high theoretical specific capacity (589 mAh g^{-1}), and is easy to synthesize, inexpensive, is cathode material with development prospects, but its more flexible open framework during cycling is prone to collapse, thereby causing cathode material dissolution [11]. In addition, conductivity and diffusion kinetics sluggishness and other problems have relatively large influence on cycling stability. Therefore, still need to modify V_2O_5, to improve its structural stability and conductivity.

3. V_2O_5 cathode material research status

From previous section can be seen, in aqueous zinc-ion battery cathode materials, currently widely adopted metal oxides mainly include manganese-based oxides and vanadium-based oxides, their special layered or tunnel-type structures provide channels for Zn^{2+} intercalation.

Compared with manganese-based materials, vanadium-based materials because possess more stable structures and oxidation state changes, alleviate element dissolution problem, and vanadium element possesses abundant reserves, relatively small relative atomic mass, possesses mixed multivalent states and other characteristics, is very promising cathode material, currently cathode material aspect many researches also concentrate on vanadium-based oxides. But vanadium-based oxides as cathode materials' performance is not perfectly flawless, they also face many self-problems:

(1)Most vanadium-based compounds are semiconductors, material itself conductivity is not good, affecting electron transport rate, causing during cycling process electrode polarization degree increase and capacity decay.

(2)Zn^{2+} effective charge is relatively large, during cathode layered material migration process will with V, O and other atoms produce relatively large electrostatic interaction forces, causing Zn^{2+} diffusion or migration kinetics sluggish, simultaneously reducing cathode material cycling stability;

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(3) Aqueous electrolyte in layered vanadium-based compounds will occur active material dissolution, especially under high current density, active material dissolution rate is faster, this condition significantly reduces cathode material cycling stability, and influences its material utilization efficiency.

In summary, to cope with cathode material faced many problems, current research has widely adopted multiple strategies to optimize cathode material performance, including regulating cathode material morphology, enhancing structural stability and improving material utilization rate, thereby elevating battery overall performance.

3.1. Guest particle pre-intercalation

Because Zn^{2+} insertion/extraction process ion diffusion kinetics suppression can lead to structure degradation, most pure oxide ions' reversible capacity is low, cycling performance poor, in layered structures introducing cations or structural water molecules to expand layer spacing is one of ways to improve cathode material stability.

(1) Metal Cation Pre-intercalation

In V_2O_5 interlayers introducing Na^+, Zn^{2+} and Ca^{2+} and other metal ions is effective strategy to improve V_2O_5 structural stability. Yang et al. [12] utilized hydrothermal reaction to introduce Fe^{2+}, Co^{2+}, Ni^{2+}, Mn^{2+}, Zn^{2+} and Cu^{2+} and other transition metal ions into V_2O_5 interlayers, and found metal ion pre-intercalation can elevate battery cycling stability, CuVO-300 initial discharge capacity was 307 mAh g^{-1}, obviously decreased to 204 mAh g^{-1}, then in 10000 cycles maintained relatively good stability.

V_2O_5 layers in crystal structure transformation depends on pre-intercalated metal ion types and quantities, this to improving battery capacity, cycle life and rate capability also plays important role. Guan et al. [13] adopted a simple hydrothermal method to pre-intercalate Al^{3+} into V_2O_5 interlayers, prepared nanobelt-like $Al_{0.15}V_2O_5{\cdot}1.01H_2O$ cathode material, at 10 A g^{-1} current density, after experiencing 10000 cycles, still could maintain 144 mAh g^{-1} specific capacity (Figure 5).

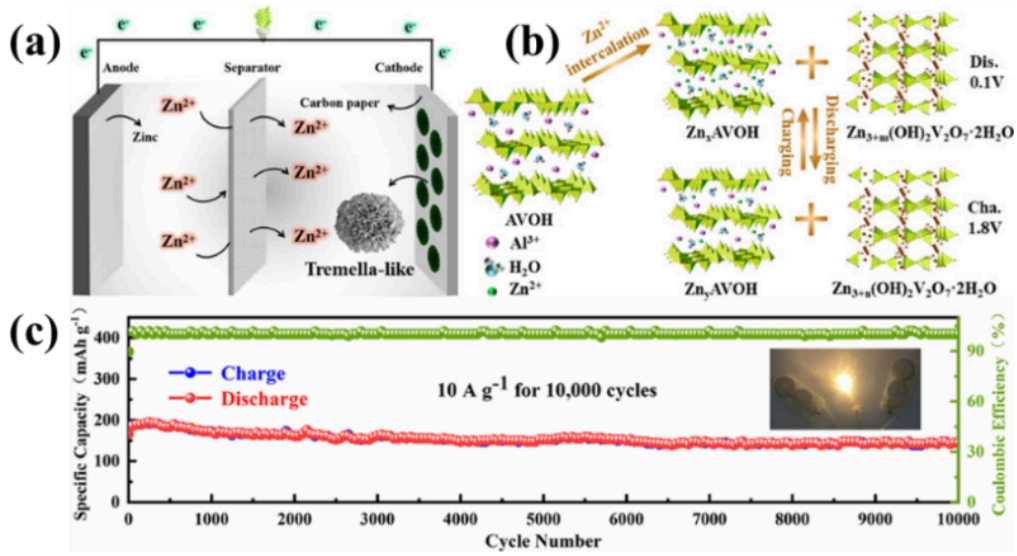


Figure 5. (a) With A-VOH material as electrode ZIBs working principle (b) A-VOH electrode and reversible Zn^{2+} during charge-discharge process accumulation/insertion schematic diagram; (c) 10 A g^{-1} current long-cycle performance [13]

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(2) Ammonium Ion Pre-intercalation

Compared to other vanadates, ammonium vanadate due to its smaller density and molecular weight, possesses higher volumetric capacity and gravimetric energy density, this to improving battery performance has important significance, and pre-intercalated NH_4^+ acts as V_2O_5 interlayers' "pillar", relieving Zn^{2+} migration process caused cathode material collapse. Zhao [14] research group found pre-intercalated ammonium ions made material layer spacing effectively enlarged, from initial 7.2 Å expanded to 11.8 Å, improved Zn^{2+} repeated deintercalation time cathode cycling stability, at same time also improved specific capacity, new cathode material $(NH_4)_xV_2O_5{\cdot}nH_2O$ (NVO) at 500 mA g^{-1} current density, possessed 391 mAh$\cdot g^{-1}$ high specific capacity. To pursue better cycling stability and higher specific capacity, on basis of intercalated ammonium ions again carrying out alkali metal ions introduction, Wang [15] research group into NVO intercalated alkali metal sodium ions, this research first prepared $NH_4V_4O_9$ material, then into it doped sodium ions, new material after testing found, at 10 A$\cdot g^{-1}$ after 2000 cycles, NVO-Na cathode at 0.1 A$\cdot g^{-1}$ current density possessed high specific capacity (400.2 mAh g^{-1}) and capacity retention (97.2%).

In summary, ammonium ion insertion can expand layer spacing and improve specific capacity, with alkali metal (e.g. sodium ions) pre-intercalation joint action, battery performance improvement effect is more remarkable.

(3) Water Molecule Pre-intercalation

Pre-intercalated water molecules in cathode material play extremely important roles, specifically manifested in following aspects. First, water molecules' presence significantly increases V_2O_5 crystal layer spacing, this for Zn^{2+} deintercalation widens channels, thereby improving ion transport efficiency. Second, because layer spacing expanded, interlayer interaction forces correspondingly weakened, prompting Zn^{2+} diffusion rate to be significantly elevated. Finally, water molecules in interlayers shield Zn^{2+} partial effective charge amount, this action weakens Zn^{2+} and vanadium oxide layers between electrostatic interactions, further improving Zn^{2+} in material migration behavior. Kundu et al. [16] first reported a kind of nanobelt-structured $Zn_{0.25}V_2O_5{\cdot}nH_2O$ material, these intercalated water molecules through their water molecules reversible intercalation action, realized layer spacing dynamic adjustment. This design made aqueous zinc-ion batteries exhibit excellent kinetic performance and rate performance, providing new ideas for related field research and applications.

3.2. Defect engineering

Defect engineering is a rational and effective strategy, used to regulate materials' electronic structure and lattice arrangement. Through introducing defects, can from internal to external comprehensively optimize material structure, regulate material layer spacing, thereby improve electrode performance. In recent years, defect engineering in improving vanadium-based compounds as aqueous zinc-ion battery cathode materials application has gradually increased.

The oxygen-deficient V_6O_{13} (O_d-VO) designed by Liao [17]'s research group, from the perspective of microstructure, possesses alternating single and double vanadium oxide layers, is rich in oxygen vacancies, and can generate more divalent cation insertion sites, thereby increasing capacity. Utilizing surface electron density difference analysis to understand the modulation mechanism of O_d-VO in Zn^{2+} storage/release, it was found that V_6O_{13} (p-VO) exhibits uniform charge distribution, which can uniformly induce Zn^{2+} to migrate and gain/lose electrons within the material; from the perspective of the migration process, when oxygen atoms are extracted from the vanadium oxide lattice, defect sites with excess electrons are formed, and these defect sites

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subsequently lead to the electron cloud delocalization of O_d-VO, thereby achieving an increase in capacity.

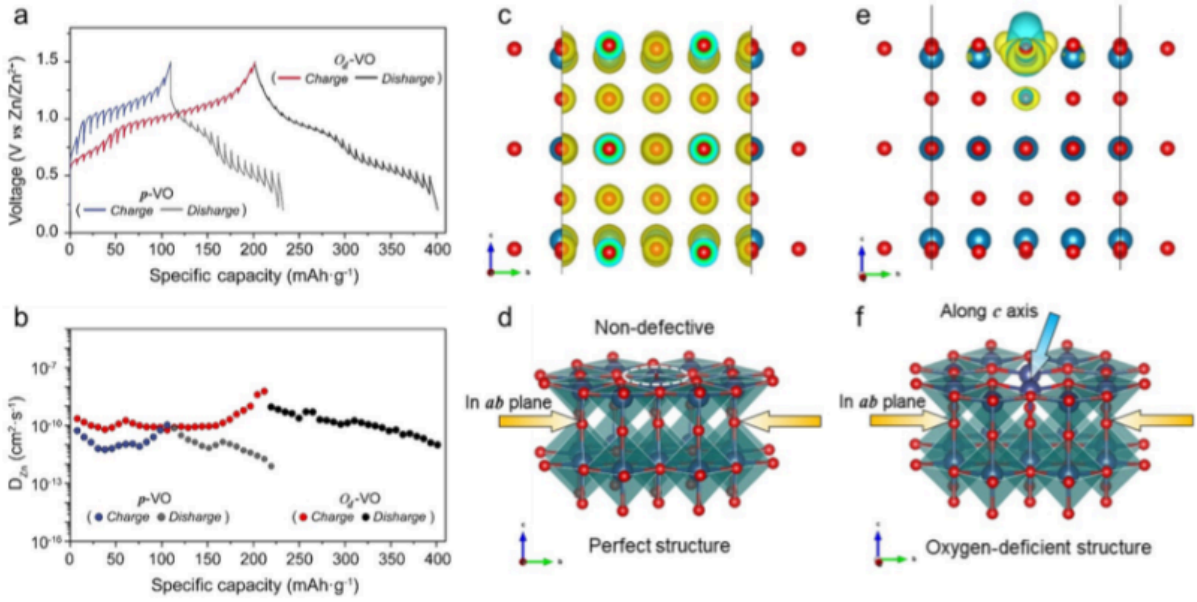


Figure 6. (a, b) respectively are p-VO and O-d-VO electrodes' GITT curves and Zn^{2+} diffusion coefficients; (c, d) p-VO charge distribution and corresponding structure; (e, f) O-d-VO charge distribution and corresponding structure [17]

In summary, constructing oxygen defects from microstructure and mechanism aspects and other aspects has great help for improving battery overall performance, its application still needs more attempts and research.

3.3. Morphology control

Besides through intercalating ions or molecules to expand interlayer spacing to achieve cathode material stability improvement, can also through certain specific experimental conditions, realize cathode material microstructure morphology regulation. Structure regulation can be roughly divided into microstructure size nanosizing, porous hierarchical structure and special morphology regulation, etc.

Carrying out material size nanosizing can increase its surface area, shorten ion transport paths, during charge-discharge process can achieve fast Zn^{2+} migration and obtain better effect, this method is mostly used after material pretreatment completion, carrying out secondary processing on material, to further improve material performance. Yang et al. introduced Co into V_2O_5 interlayers, and under surfactant sodium dodecyl sulfonate (SDS) action composited with graphene oxide to prepare CoVO composite material. SDS addition significantly affects composite material morphology, from results can find, with SDS increase different proportions of nanopolyhedra gradually transform into nanorods, from three-dimensional morphology transform into two-dimensional morphology, as shown in Figure 6, increased material specific surface area, can significantly improve material active sites, carrying out Zn^{2+} intercalation and deintercalation.

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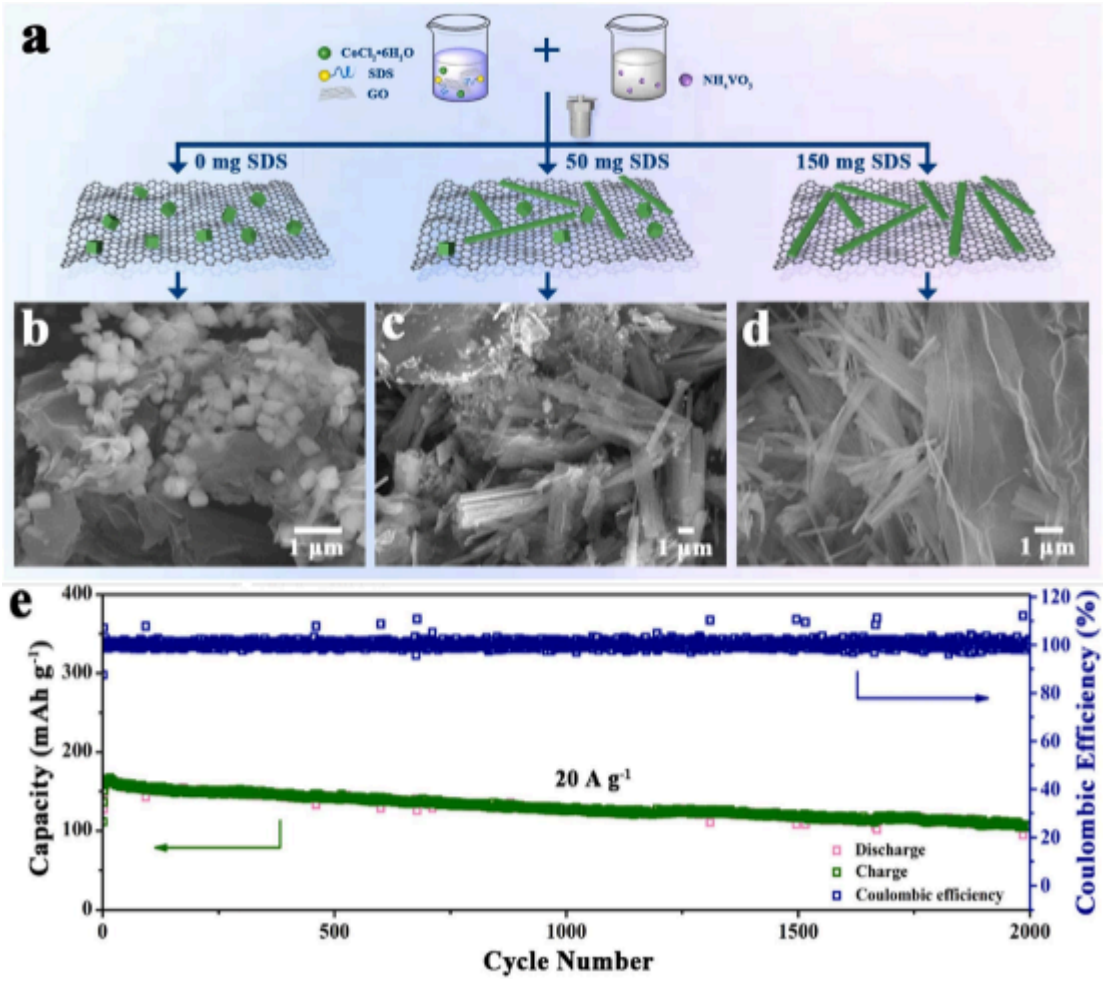


Figure 7. (a) CoVO composite material synthesis scheme; different SDS addition amounts under SEM images; (b) 0 mg; (c) 50 mg; (d) 150 mg; (e) CoVO-150 at 20 A g^{-1} long-term cycling performance [18]

3.4. Constructing composite materials

Cathode materials' low conductivity hinders electron transport, will cause electron collection efficiency decrease, thereby causing relatively low specific capacity. Therefore compositing vanadium-based oxides and titanium-based oxides type cathode materials with conductive materials can improve cathode material conductivity, thereby improving battery capacity..

Wang et al. through ion adsorption method treated ultrathin amorphous V_2O_5 uniformly grown on graphene both sides, forming ultrathin flat nanosheet composite material (A-V_2O_5/G). This material due to its nanometer size small, therefore possesses extremely high ion diffusion paths, abundant active sites, high conductivity and excellent structural stability, proved high reversible aqueous ZIBs (A-V_2O_5/G-ZIBs) possess unprecedented high capacity, high rate capability, long cycling performance and excellent safety [19].

4. Conclusions and outlook

This paper reviews various chemical power sources development status, constrained by lithium resource shortage, organic electrolyte safety defects, aqueous zinc-ion batteries by virtue of high

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ionic conductivity, safety, low cost, environmental friendliness and other advantages, become lithium-ion batteries extremely promising alternative energy storage devices. Currently exist manganese-based, organic, Prussian blue, vanadium-based four types of mainstream cathode materials, as well as Zn^{2+} intercalation, Zn^{2+}/proton co-intercalation, chemical conversion three types of storage mechanisms, and compares various materials' electrochemical performance. Vanadium-based materials possess open layered, tunnel frameworks and high theoretical capacity, among which V_2O_5 due to two-electron redox, easy synthesis characteristics receives much attention, but intrinsic conductivity weak, zinc ion transport kinetics poor, cycling structure collapse and active material dissolution and other defects limit its application. The paper summarizes guest pre-intercalation, defect engineering, morphology control, carbon-based composite four types of modification strategies, can significantly improve V_2O_5 electrochemical performance. But single modification is difficult to balance comprehensive performance and mass production requirements, follow-up can develop defect-carbon composite synergistic modification process, accelerate vanadium-based zinc-ion batteries practical application process.

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