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International Journal of Materials Science and Technology Studies
ISSN: 3006-7693 (Print), ISSN: 3006-3744 (Online) | Volume 2, Number 3, Year 2024
DOI: https://doi.org/10.62051/ijmsts.v2n3.09
Journal homepage: https://wepub.org/index.php/IJMSTS/index

Progress on the Application of Modified Anode Materials for Aqueous Zinc Ion Batteries

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ABSTRACT

In the current developed electrochemical energy storage device system, drainage zinc ion batteries have attracted wide attention due to their safe operation, environmental friendliness and high theoretical specific capacity. However, when applied to the anode of drainage zinc ion batteries, uneven zinc deposition leads to the formation and growth of zinc dendrite, piercing the diaphragm and causing a short circuit in the battery. Zinc dendrites have mechanical rigidity and structural inhomogeneity, which is easy to fall off from the zinc negative electrode to form "dead zinc", leading to the reduction of active materials and the decline of battery capacity. In addition, hydrogen evolution reaction (HER), corrosion, passivation and other side reactions also occur, resulting in the stability of the battery. These adverse electrochemical reactions all occur at the zinc metal anode / electrolyte interface, which limits the development and application of drainage zinc ion battery. This paper summarizes the current carbon materials in stabilizing zinc metal anode and constructing some composite coatings to functionalize the surface of zinc anode to inhibit zinc dendrites and related side reactions, improve the stability of zinc metal anode, and then improve the electrochemical performance of drainage zinc ion battery.

KEYWORDS

Water system zinc ion battery; Zinc negative electrode; Carbon material; Composite functional materials; Research progress

1. INTRODUCTION

Due to their low cost, environmental protection and high safety, water-based zinc ion batteries (AZIBs) are considered that one of the most important preconditions for large-scale energy storage in the future is that zinc metal can be used as an anode, because of their high theoretical capacity and appropriate redox potential, making it possible to use water-based electrolytes. However, using zinc metal as an anode faces the challenge of zinc side reactions in a typical aqueous electrolyte, leading to anode corrosion, electrolyte consumption, low Coulomb efficiency, uneven Zn deposition, dendrite formation and short cell life. In view of these problems, the researchers have proposed many solutions, such as designing an artificial interface coating, adding electrolyte additives and designing a negative electrode substrate. Due to its rich structure and good stability, carbon materials are often used to stabilize the zinc anode in drainage zinc ion batteries. Among them, functional carbon material as cathode substrate can provide more load space for zinc deposition and adjust the deposition behavior of Zn^{2+}. Carbon material interface coating can avoid the electrolyte directly contact with zinc metal anode, thus reducing the occurrence of interface side reaction; Carbon material, as electrolyte additive, can evenly distribute the surface of zinc foil and inhibit the growth of dendrite. With the carbon material in the drainage system of zinc away

The research on stabilizing zinc cathode in batteries is increasing, so it is necessary to systematically summarize the latest progress of carbon materials in stabilizing zinc cathode, so as to provide some research ideas and reference for exploring carbon materials playing a greater role in the field of zinc ion batteries.

2. RESULTS AND DISCUSSION

2.1. Application Research of Carbon Materials in Zinc Metal Anode

2.1.1. Modification of zero-dimensional carbon materials in zinc metal anode

Graphene quantum dots (GQDs) are highly tunable, ultra-saturated zero-dimensional carbon materials that are used in ion sensing, bioimaging optoelectronics and electronic devices. Poor cycling performance occurs because PANI deprotonates in weakly acidic electrolytes. Ye group [1] Edge sulfated graphene quantum dots (SGQDs) were selected as dopants and PANI powder was prepared using chemical oxidation. Because the $-SO_3-$ group on SGQDs can serve as a local proton library to increase the protonation of PANI. In addition, it was found that the PANI electrodes doped with SGQDs showed excellent multiplier performance (115 mA h g^{-1} at 1 A g^{-1}) and good cycle stability (81% after 1500 cycles), far exceeding the performance of pure PANI. Zhang et al [2] Water-soluble graphene quantum dots (GQDs) were used as an electrolyte additive to improve the electrode – electrolyte interface. Density functional calculations (DFT) show that GQDs have a stronger adsorption energy for zinc ions relative to the zinc metal substrate, due to the oxygen-containing functional groups that can coordinate as Lewis base and zinc ions as Lewis acid. Therefore, GQDs adsorbed on the zinc anode surface can act as nucleation sites to inhibit the transverse two-dimensional diffusion deposition of zinc ions and reduce dendrite formation. (Figure 1) And the rich oxygen-containing functional groups can form hydrogen bonds with free water, thus alleviating the side reaction of water and inhibiting zinc corrosion and hydrogen evolution reaction.

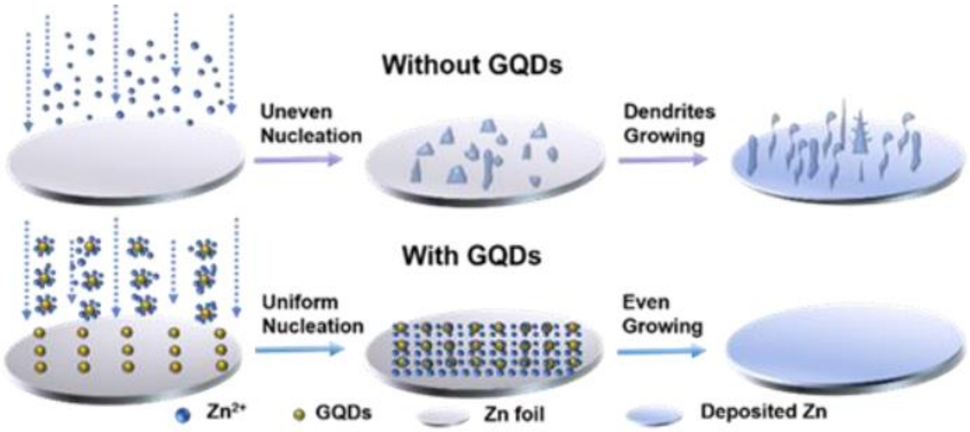

Figure 1. Schematic depiction of the effect towards suppressing dendrites growth.

In addition to zhang [2] A CDs containing rich aldehyde groups and cyanano functional groups were also constructed and a macro quantitative preparation of nitrogen-containing functional carbon points was realized. First, functional CDs with zinc-containing components have stronger binding energy to zinc, thus can reduce the nucleation overpotential and increase the nucleation site. Second, the quantum-sized CDs layer can uniformly distribute the electric field on the surface, and then induce uniform Zn deposition, and ultimately inhibit the disordered growth of the zinc dendrite. Finally, the use of the properties of CDs artificial SEI insoluble in water and negative surface charge avoids the excessive contact of Zn negative electrode and the electrolyte, while realizing the repulsion of SO_4^{2-}

with CDs, which helps to reduce Zn surface corrosion and HER. At the same time, it is shown that the half cell (Figure 2) and the whole cell (Figure 2) protected by CDC s layer at different current densities have good cycle stability and excellent rate performance.

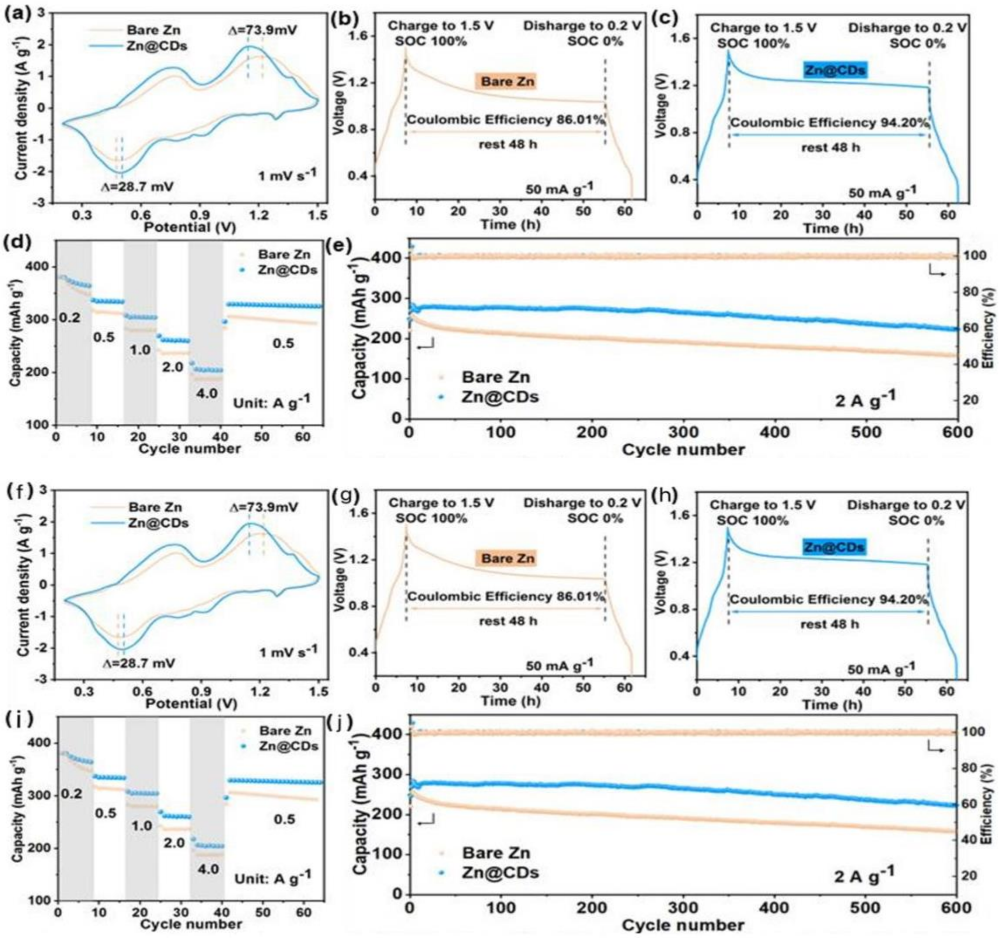

Figure 2. Cycling performance of symmetrical cells with bare Zn and Zn@CDs electrodes at (a) 1 mA cm^{-2}, (b) 4 mA cm^{-2} and (c) 6 mA cm^{-2}; Rate performance of (d) bare Zn and (e) Zn@CDs symmetric cell at various densities.(f) CV plots of Zn||NVO full cell; The self-discharge behavior of full cells with (g) bare Zn anode and (h) Zn@CDs anode; (i) Rate capability at different current densities from 0.2 to 4 A g^{-1}; (j) Long-time cycling performance at 2 A g^{-1}

2.1.2. Modification of one-dimensional carbon materials in zinc metal anode

Liu group [3] A metal zinc anode with a functional interface modification layer and its preparation method and application are provided. The metal zinc anode includes a metal zinc matrix, and a functional interface modification layer containing a nanoscale carbon material coated on the surface of the metal zinc matrix. The nanoscale carbon material includes at least one of the carbon nanofibers, carbon nanotubes, nanocarbon spheres, and graphite. By using these nanoscale carbon materials, the interface modification layers with high specific surface area, porous structure, and excellent electrical conductivity can be prepared. This modified layer can improve the stability of the contact interface between the metal zinc anode and the electrolyte, inhibit the corrosion and dendrite effect of the metal zinc cathode, reduce the polarization of the electrode, improve the electrochemical performance of the battery, and effectively inhibit the growth of the zinc dendrite. Huang group [4] Hydroxylated carbon nanotubes (OH-CNT) and graphene oxide (GO) are filtered to obtain the carbon film, which is used as a buffer layer in the zinc ion battery. The symmetric cells with Zn / OH-CNT electrodes did not significantly change in morphology from before testing after 500 h of galvanizing and stripping, and had a lower overpotential (83 mV). The excellent electrical conductivity of carbon nanotubes (CNT) and the large specific surface area can provide rich active sites for the electrochemical reaction, and the stable structure makes it have a long cycle life in the process of electrochemical reaction. Ba et al [5] A high-performance SEI based on the cation -π interaction was constructed to protect the zinc anode in the AZIBs. The rGO layer in the TAA / rGO composite can construct conductive networks and uniform interfacial electric fields. At the same time, the sulfur-containing functional groups on the surface of graphene can not only provide nucleation sites for Zn^{2+} fast sedimentation, but also use its affinity for Zn^{2+} to induce rapid and uniform deposition of Zn^{2+}, and effectively inhibit zinc dendrite and side reactions.(Figure 3) In the TAA / $rGO_{1/2}$ composite, the symmetric battery has a cycle life of over 5000 h at the current density of 4.4 $mAccm^{-2}$ and 1 mAh cm^{-2}, and the polarization voltage is only 38.1 mV; At the high current density of 10 $mAccm^{-2}$ and 1 mAh cm^{-2}, the cycle life is up to 750 h. Full-cell TA /$rGO_{1/2}$ @ Zn //MnO_2 also showed excellent multiplier performance.

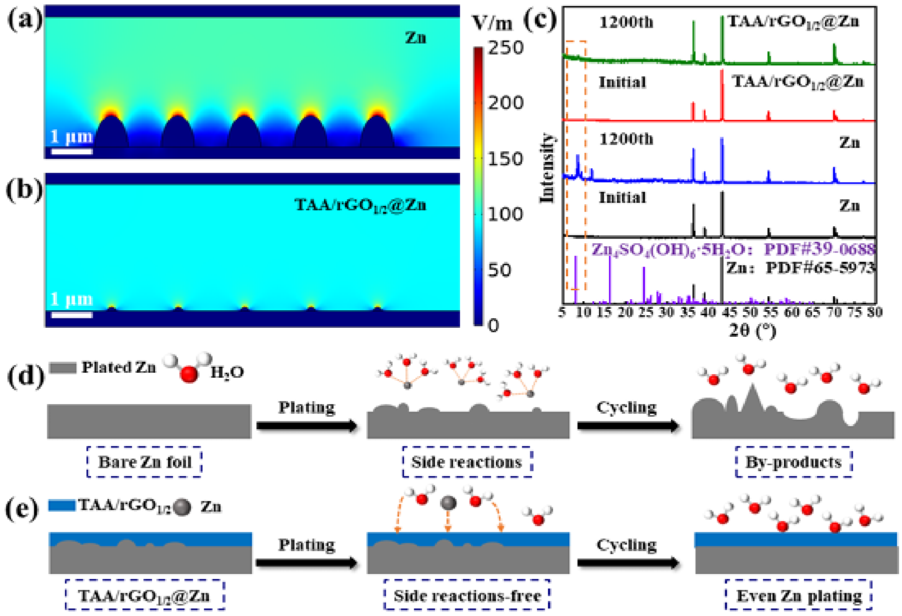

Figure 3. (a-b) The simulated electric field distributions of anodes, (c) XRD pattern, and (d-e) the schematic representation of Zn^{2+} deposition on anodes

Shi group [6] Vanadium dioxide (B) -multiwall carbon nanotubes (VO_2 (B) -MWCNTs) and zinc nanosheet electrodes were prepared through a series of processes including vacuum filtration, stripping and electroplating. VO_2 (B) -MWCNTs, as battery positive electrode, zinc nanoplate as negative electrode, trifluorthane sulfonate zinc-polyvinyl alcohol ($Zn(CF_3SO_3)_2$-PVA) hydrogel for electrolyte assembly into aqueous zinc ion microcell (ZIMB). The specific capacity of ZIMB is up to 314.7 μAh cm^{-2}, with an energy density of 188.8 μWh cm^{-2} and a power density of 0.61 mW cm^{-2}, with excellent flexibility (bending angle up to 150°) and good thermal stability.

2.1.3. The modification of two-dimensional carbon materials in the zinc metal anode

Graphene is a two-dimensional ordered carbon material with unique physicochemical properties, excellent electrical properties, and extremely high specific surface area. The introduction of functional group modified graphene protective layer on the surface of metal zinc anode can not only use graphene to regulate the uniform deposition of Zn^{2+}, inhibit side reactions such as zinc dendrite and hydrogen evolution, and improve the stability of the battery, but also use the anchoring of its surface functional group on Zn^{2+}. Ininduced rapid Zn^{2+} deposition and improved the cycling stability and multiplier performance of AZIBs. Xie et al [7] NMC @ CuB DC composite material was prepared by using NMC carbon material and zinc-philic CuBDC MOF as the anode protective layer of the drainage zinc ion battery to achieve stable and high-performance zinc anode. NMC @ CuB DC can effectively limit the 2 D diffusion of Zn^{2+}, and its copper species can reduce the zinc deposition barrier, induce uniform zinc deposition, and further inhibit the dendrite growth. (Figure 4) NMC @

CuB DC <NMC is maintained at different current densities, indicating that the introduction of conductive nitrogen doping NMC and NMC @ CuBDC can reduce the zinc core barrier. Meanwhile, the zinc-friendly Cu species in copper-based MOF material makes the core barrier lower and promotes uniform zinc deposition.

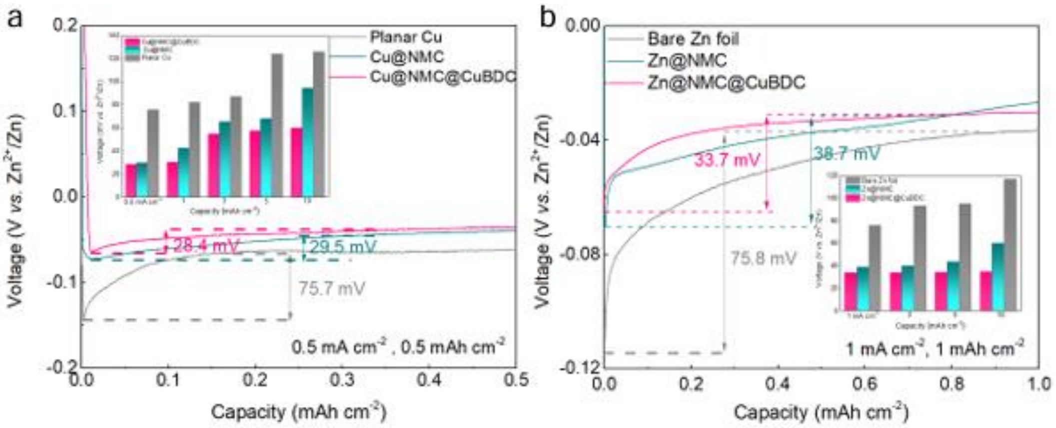

Figure 4. (a, b) are the nucleation overpotentials in the first cycle of Cu||Zn battery and Zn||Zn battery respectively. Inset: The overpotential distributions of the corresponding batteries under different current densities.

Zhou group [8] Using a simple solution casting technology, a layer of graphene oxide (GO) was coated on the surface of the zinc negative electrode with a layered structure, serving as a functional protective layer to inhibit the growth of the zinc dendrite.(Figure 5), the GO layer can not only promote the penetration of the electrons through the ZnO insulating layer formed by oxidation on the surface of the zinc cathode, but also reduce the dissolution of metal zinc intermediates in the charge and discharge process, so as to improve the utilization rate and cycle stability of the zinc cathode. Compared with the unmodified Zn anode, the Zn @ GO composite zinc negative electrode coated with 1.92 wt% GO showed superior cycling performance, with an area discharge capacity reaching 128% of the unmodified Zn anode. In addition, the O_2 positive electrode combined with the Zn @ GO negative electrode is expected to be used in a wide range of fields, from electric vehicles to grid-level energy storage.

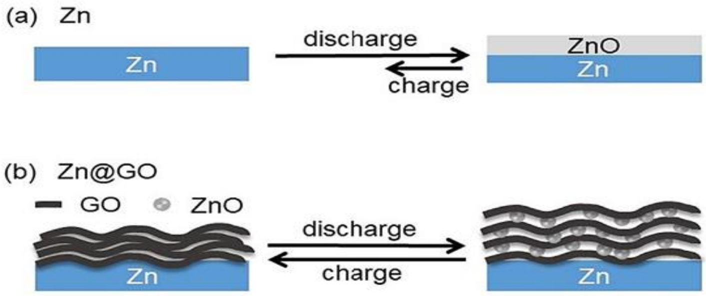

Figure 5. Schematic representation of morphological changes of Zn anode during electrochemical cycling process

Peng et al [9] The thioacetamide / reduced graphene oxide (TAA / rGO) composite was prepared by reaction of thioacetamide (TAA) with GO by hydrothermal method. The rGO layer in the TAA / rGO composite can construct conductive networks and uniform interfacial electric fields. At the same time, the sulfur-containing functional groups on the surface of graphene can not only provide nucleation sites for Zn^{2+} fast sedimentation, but also use its affinity for Zn^{2+} to induce rapid and uniform deposition of Zn^{2+}, and effectively inhibit zinc dendrite and side reactions. In addition, Zhao et al [10] The conductive skeleton expansion graphite EG was synthesized by high temperature expansion method through mauffle furnace, and combined with vanadium sulfide by hydrothermal treatment to prepare VS_2 / EG composite material. The effects of different EG additions (25 mg, 50 mg and 75 mg) on the electrochemical properties of the electrodes were compared. The results showed that all the composites with EG addition significantly improved the electrochemical stability of VS_2 compared to the original VS_2. In particular, when the amount of EG addition is 50 mg, the specific capacity can reach 169 mAh g^{-1} at the current density of 0.1 A g^{-1}; at the current density of 2.0 A g^{-1}, after 1000 cycles, the structural stability of VS_2 / EG increases significantly compared with VS_2, and the capacity retention rate reaches 66% of the initial value. Using these composites in series into energy storage devices can light the light belt for more than 10 minutes. These results fully confirm the feasibility of introducing a conductive skeleton. The addition of EG enhances the ionic conductivity of the electrodes and accelerates the electron transfer. The stable structural skeleton helps to protect VS_2 from structural damage during the cycle, thus further improving the electrochemical stability of the material.

2.1.4. Modification of 3 D carbon materials in the zinc metal anode

A stable interface layer in the surface layer of zinc anode can effectively block the direct contact between zinc metal and electrolyte, and then alleviate the occurrence of side reactions. Zhang group [11] The low temperature plasma technology was used to modify the carbon cloth, and the electrode was constructed., In order to improve the performance of the negative electrode of the drainage zinc ion battery. The modified carbon cloth surface forms a loose and porous deposition layer, which is conducive to the penetration of electrolyte and the utilization rate of zinc. At the same time, it can accommodate zinc in the process of charge and discharge, and inhibit the formation of zinc dendrite, so as to prolong the service life of the battery. The experimental results show that (Figure 6) the symmetric battery life using modified carbon cloth as the negative electrode was significantly improved (Zn @ CFs as modified carbon cloth, Zn @ CF unmodified carbon cloth), and the whole battery assembled with the activated carbon positive electrode also showed high cycle stability and capacity retention rate.

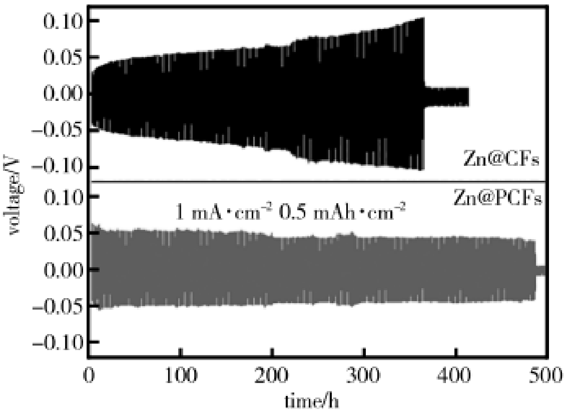

Figure 6. Time-voltage curves of Zn @ CFs / / Zn @ CFs and Zn @ PCFs / / Zn @ PCFs symmetrical cells at current density of 1 mA cm^{-2} and charge-discharge capacity of 0.5 mAh cm^{-2}

Carbon materials with 3D porous structures are widely used in the battery field for the preparation of 3 D structured electrodes. These carbon materials themselves can simultaneously have a large specific surface area, excellent electrical conductivity, and electrochemical inert. The 3 d carbon network carrier zinc anode can effectively evenly distribute the electric field, reduce the zinc nucleation overpotential and size, thus alleviating the growth of zinc dendrite.3 D carbon nanotube network, graphene foam, carbon fiber, tin-modified C 3 D carbon felt were used as the zinc cathode matrix to improve the deposition behavior of zinc metal. For example, Zeng et al [12] People use highly conductive carbon nanotube frames for dendrite-free zinc deposition (Figure 7). First, a disordered carbon nanotube (CNT) array was constructed on a carbon cloth by chemical vapor deposition method, and then the Zn / CNT composite negative electrode was obtained by electrochemical deposition. The electrochemical results show that the Zn / CNT anode significantly improved the electrochemical performance of the zinc anode compared with the pure carbon cloth (Zn / CC) deposition, indicating that the carbon nanotube frame plays a key role in the zinc deposition performance.

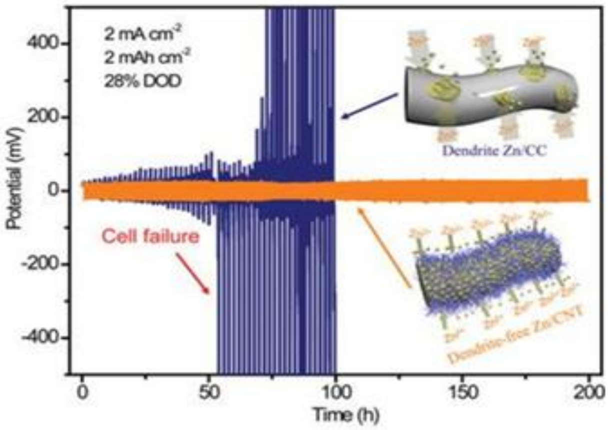

Figure 7. The Zn/CNT|Zn/CNT symmetric battery exhibits a stable voltage profile with a low voltage hysteresis of approximately 27 mV for 200 h under a current density of 2 mA cm^{-2} and a limited capacity of 2 mA h cm^{-2}. In contrast, the voltage profile of Zn/CC|Zn/CC shows significant voltage fluctuations after 50 h.

Yu group [13] Three-dimensional porous carbon material (3D-HPC) with high surface area, rich graded porous structure and high conductivity of graphitization and coconut shell was prepared by hydrothermal carbonization and KOH activation. In the preparation process, sugarcane bagasse containing cellulose and hemicellulose components can not only give 3D HPC of large specific surface area, but also have different types of pore structures such as micropore, mesopore and macropore; the coconut shell rich in lignin structure can bring higher graphitization to 3D-HPC, thus improving the conductivity. ZHSCs assembled with 3D-HPC as positive and zinc foil as negative were able to have a specific capacity of 305 mAh g^{-1} at 0.1 Ag^{-1} and maintain an initial capacity of 94.9% after 20,000 cycles. Although the electrochemical energy storage mechanism of porous carbon materials in ZHSCs has not been clearly understood, the performance of ZHSCs can be significantly improved by regulating the specific surface area and the pore structure suitable for Zn^{2+} and anion diameter in the electrolyte. In addition, Cui et al [14] 3 D crosslinking porous carbon fibers were prepared by the $ZnCl_2$ crosslinking / poration strategy, and the optimal preparation protocol was determined through a series of characterization and tests (carbonization temperature of 800℃, 10% addition of zinc chloride).(figure 8) 3D-PA NC-10% using $ZnCl_2$ cross-linking and pore making, three-dimensional crosslinking and hierarchical porous structure, three-dimensional crosslinking structure effectively improve the conductivity and stability, hierarchical porous structure provides a good ion transport channel and storage site, thus effectively improve the specific capacity and ratio of zinc ion mixed capacitor performance.

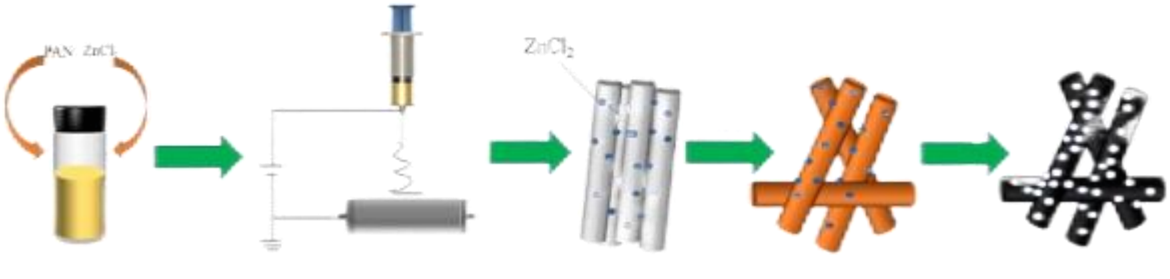

Figure 8. A Schematic representation of the preparation process of the 3D-PANC

2.2. Modification of Zinc Anode By Functional Materials

Zhang group [15] The quasi-single ionic conductor is constructed by using a β -cyclodextrin polymer (β -CD polymer-P-CD) to coat and protect the zinc anode. This P-CD coating prevents the corrosion of the zinc anode and prevents the side reaction of the metal anode. More importantly, the cyclodextrin unit forms quasi-single ion conductors through host-guest interaction and hydrogen bonding, thus increasing the migration number of zinc ions (from 0.31 to 0.68), inhibiting the formation of space charge regions, and thus stabilizing the coating / stripping of zinc ions. (Figure 9) The results show that the cycle life of the Zn / / Zn symmetrical cell coated with P-CD is increased by 70.6 times to 600 hours at a high current density of 10mA cm^{-2} and 10mAh cm^{-2}. Furthermore, the Zn / / K_1 coated with P-CD. The V_3O_8 (KVO) full battery capacity retention rate reached 94.5% after 1000 cycles at high active substance mass load (20mg cm^{-2}) and low N / P ratio (1.46). These results provide a new perspective on the control of solid electrolyte interfaces to stabilize the zinc cathode, and provide a practical strategy to improve the drainage of zinc ion batteries (AZIBs).

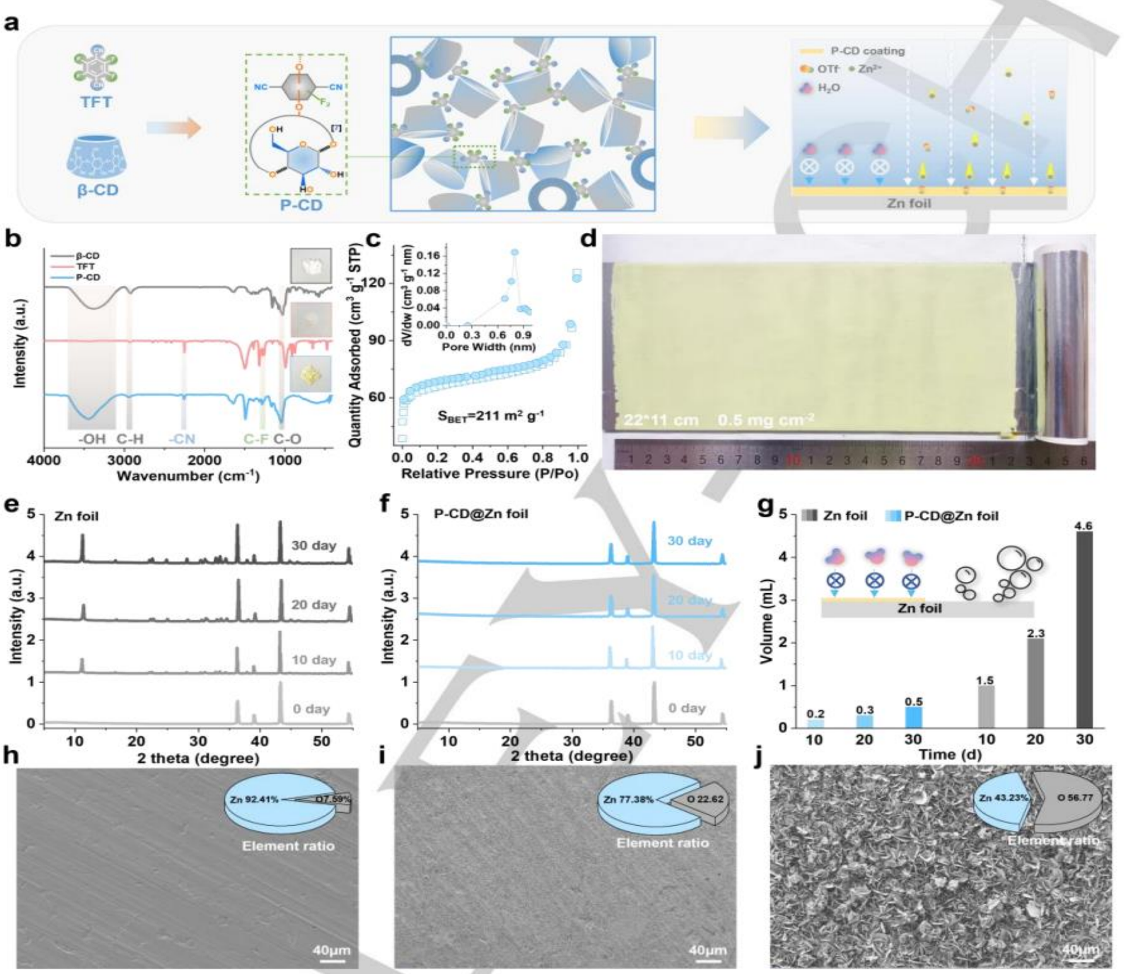

Figure 9. (a) Working mechanism of the synthesis of polymer P-CD and polymer P-CD on Zn foil.(b) Fourier transform infrared spectrum (FTIR) spectra of precursors and products, and the embedded images are optical photographs of P recursors and products.(c) N_2 adsorption (square) and desorption (circular) isotherm of P-CD, embedded in the volume map of Shorvath-Kawazoe differential holes.(c) Optical photograph of the P-CD amplified coating on the zinc foil. X-ray diffraction (XRD) profile of uncoated / coated zinc foil (e) after different interference in bag cells; (g) corresponding hydrogen produced in bag cells. Scanning electron microscope (SEM) images and element ratios on the surface of (h) commercially available zinc foil and (i) / (j) uncoated zinc foil were maintained in bag cells for 30 days.

Meng group [16] By coating PVA solution on Zn metal surface to forming a high polymer surface layer, the growth of zinc dendrite was effectively inhibited and improved the electrochemical properties. The polar group of PVA interacts with Zn^{2+}, facilitating its rapid diffusion and avoiding parasitic reactions, achieving uniform deposition. PVA coating also alleviates the ion concentration difference and inhibited zinc dendrite. Thus, the PVA composite anode has high capacity at 0.1A g^{-1} current density and maintains high capacity retention and Coulomb efficiency after 1000 cycles. (Figure 10) Research shows that the PVA protective layer effectively improves the performance of zinc ion batteries, and this strategy is also of reference value for other battery systems. As an organic interface material, PVAs protective layer reduces the non-uniform accumulation of Zn^{2+} in the process of charge and discharge, improves the ion transmission efficiency, and makes the composite zinc anode have excellent electrochemical properties. At 0.1 A g^{-1} current density, PVA @ Zn anode exhibits high discharge capacity and good multiplier performance, as well as high cycle stability and Coulomb efficiency at 1.0 A g^{-1}.

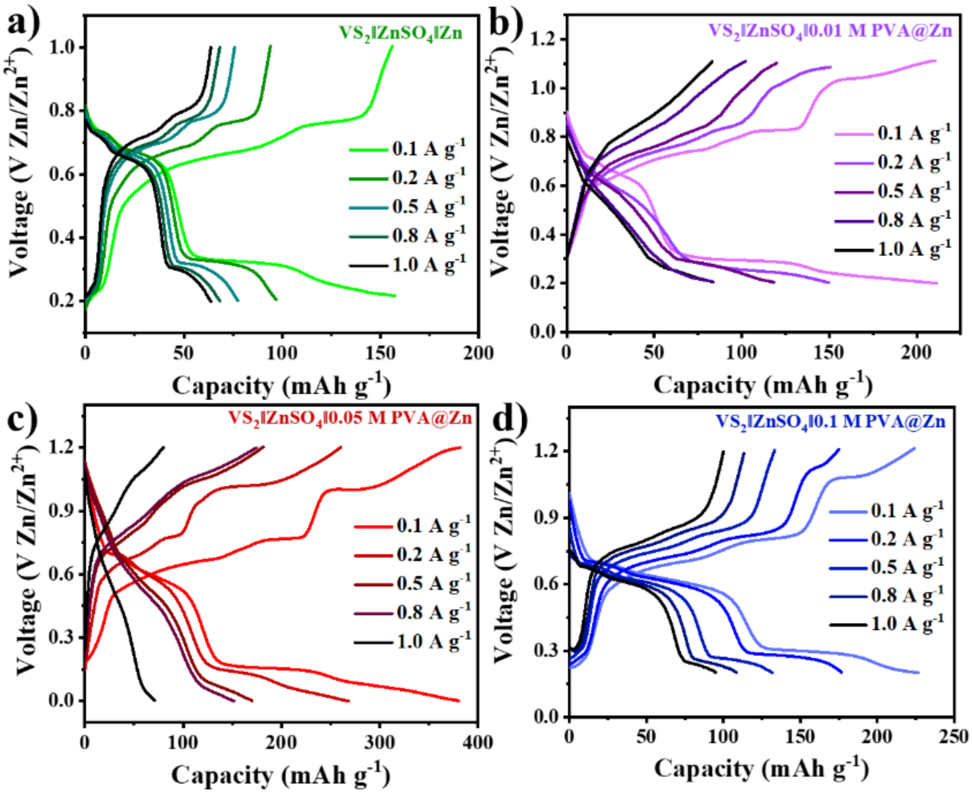

Figure 10. Charge and discharge curve of the whole cell assembled with 0.1) 0 M, (b) 0.01 M, (c) 0.05 M and (d) 0.1 M at 0.1 A g^{-1}, 0.2A g^{-1},0.5 A g^{-1}, 0.8A g^{-1} and 1.0A g^{-1}

3. CONCLUSION

Water system zinc ion battery has become one of the best choices in the field of new energy storage system in recent years because of its advantages of high energy density and long cycle life. Nevertheless, the drainage zinc ion batteries still face many challenges in the practical process, especially the problem that the zinc metal anode is easy to corrosion. This inherent limitation seriously hinders the further development of the drainage zinc ion battery. In the rapid development of energy storage technology, carbon materials with excellent electrical conductivity and diverse structure have become the key materials to stabilize the zinc anode, and remarkable results have been achieved in practical application. In this paper, the application of a variety of carbon materials, such as carbon quantum dots, carbon nanotubes, carbon nanofibers, graphene and carbon materials derived from metal-organic frameworks, in the field of drainage zinc ion batteries. These carbon materials, in different forms and mechanisms of action, positively affect the stability of the zinc anode. As the electrolyte additive, the carbon material can effectively adsorb on the surface of zinc metal to guide the orderly deposition of Zn^{2+}; as the negative interface layer, the carbon coating not only protects the zinc metal anode, avoids direct contact with the electrolyte, but also can uniform ion flow, significantly reduce metal corrosion and ensures the growth of zinc metal; as the negative substrate, the frame structure of carbon material can evenly distribute the current density, induce the uniform nucleation of metal zinc, and then optimize the deposition behavior of zinc. The application of these carbon materials in drainage zinc ion batteries not only opens up a new way for the efficient utilization of functional materials and the development of sustainable energy, but also provides innovative ideas and methods for the research and development of other electrochemical energy storage equipment. Through these studies, we have a deeper understanding of the role of carbon materials in zinc-ion batteries, which will help to promote advances in future energy storage technologies and the reality of environmentally friendly energy solutions

DECLARATION OF COMPETING INTEREST

The autours declare no competing financial interest.

ACKNOWLEDGEMENTS

This work was financially supported by the Undergraduate Innovation Training Program Project of Xuzhou University of Technology (No. xcx2024001)

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