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3D Printing Technology for Enhancing the Performance and Application Prospects of Aqueous Zinc-Ion Batteries

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Abstract. Aqueous zinc-ion batteries (AZIB) have become the most powerful competitor for the next-generation energy storage devices due to their excellent safety, low cost and environmental friendliness. However, the traditional manufacturing process is difficult to deal with problems such as dendrite growth and electrode design. The significant design freedom, material savings and controllable structure of 3D printing make it possible to upgrade the performance of azib and achieve personalized manufacturing. This article not only summarizes the application of 3D printing in AZIB (cathode, anode, electrolyte, integral battery), but also provides a multi-dimensional discussion of key processes: direct Ink Writing (DIW), Fused Deposition Modeling (FDM). It discovered how 3D printing changes the specific capacity, cycle stability and flexible customization of batteries, and expounded on the issues and progress related to 3D printing. This research offers a brand-new structurally customized zinc-based energy storage device and also provides guidance on cutting-edge issues in fields such as porous structure electrode design and interface regulation.

Keywords: 3D printing, aqueous zinc-ion batteries, electrode structure, energy storage devices, flexible batteries

1. Introduction

The demand for flexible electronics, wearable devices and large-scale energy storage has been on the rise. Traditional lithium-ion batteries cannot well meet the requirements of safety and customization. They have quite a few problems. One is that organic electrolytes are prone to catching fire. Another one is the high cost. Aqueous zinc-ion batteries (AZIBs) use aqueous electrolytes. They won't catch fire. It is also non-toxic. Zinc resources are readily available. These features make them an excellent alternative to lithium-ion batteries [1]. However, Aqueous zinc-ion batteries (AZIBs) still face some challenges. For instance, the issue of zinc dendrite growth. There is also the issue of limitations in the design of the electrode structure. And the problem of low utilization rate of active substances. These problems have restricted the improvement of energy density and cycle life.

3D printing is an additive manufacturing technology. It builds complex three-dimensional structures by piling up materials layer by layer. It has quite a few advantages. For example, it has a

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high degree of design freedom. Less material waste is also a bit. It can also be customized. In recent years, the application of 3D printing in Aqueous zinc-ion batteries (AZIBs) has received increasing attention. It can precisely control the porosity of the electrode. It can also optimize the ion transport path. It is also conducive to the integrated manufacturing of flexible devices [2]. This article systematically reviews the relevant research on the application of 3D printing in the cathode, anode, electrolyte and full battery of Aqueous zinc-ion batteries (AZIBs). The article summarizes the process characteristics and performance improvement mechanisms. It also sorted out the future development direction.

2. Core processes of 3D printing technology in AZIBs

Among various 3D printing technologies, Direct Ink Writing (DIW) and Fused Deposition Modeling (FDM) are the two most widely used in the manufacturing of Aqueous zinc-ion batteries (AZIBs). Their material compatibility is very good. The process is also relatively simple. These characteristics have made them the key research directions.

2.1. Direct Ink Writing (DIW)

Direct-Ink-Washing (DIW) is a classic extrusion 3D printing technology. It takes advantage of the rheological properties of ink to deposit materials at specific positions on the substrate. Under pressure, it will extrude the high-viscosity material slurry from the printing nozzle. This can form continuous material patterns on the base [3].Direct-Ink-Washing (DIW) is highly suitable for printing slurries with high solid content. It is usually used to make various complex three-dimensional structures. It can precisely print out various ink materials. For example: ceramics, metals, polymers, nanomaterials, biomaterials, hydrogels and resins. It has good material adaptability. And its ability to establish three-dimensional structures is also very powerful.

Direct-Ink-Washing (DIW) technology has several advantages in electrode manufacturing. The first advantage is the ability to manufacture porous three-dimensional electrodes. This can increase the specific surface area. It can also shorten the ion transport path. The second advantage is that it supports simultaneous printing of multiple materials. This makes the integrated manufacturing of electrodes and electrolytes possible. The third advantage is that the entire process is relatively mild. It is applicable even when dealing with heat-sensitive materials or flexible substrates.

2.2. Fused Deposition Modeling (FDM)

FDM is a prevalent 3D printing methodology. This technique constructs three-dimensional objects by sequentially extruding and depositing layers of thermoplastic material through a nozzle onto a build platform, after heating the material to a molten state [4]. Each layer cools and solidifies, bonding firmly to the preceding layer until the entire object is formed. FDM permits the incorporation of functional fillers (such as conductive carbon nanotubes or graphene) into the printing filament to produce conductive composite filaments. These are utilised for printing support structures or conductive current collectors. As a rapid prototyping method, FDM is increasingly applied in electrode manufacturing due to its simplicity, efficiency, low cost, and capability for large-scale, high-precision forming.

FDM offers rapid forming speeds, making it well-suited for mass production. It can also print large-scale devices, meeting the integration requirements of energy storage equipment. Furthermore, the materials used are cost-effective and readily recyclable.

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3. Application of 3D printing in key components of AZIB

3.1. Construction of high-capacity cathodes

The cathode is the core component determining the energy density of AZIBs. Cathodes prepared using traditional coating processes suffer from issues such as low active material utilisation and impeded ion diffusion. 3D printing can significantly enhance cathode performance through structural optimisation.

There are several application highlights of the DIW process in the production of negative electrodes. The DIW process design with a porous structure can print out layered or lattice-shaped anodes, which can expand the wetting area of the electrolyte and provide rich ion transport paths at the same time. For instance, in the research related to printing manganese dioxide (MnO_2) cathodes, the porosity and pore size distribution were altered, which increased the diffusion coefficient of zinc ions (Zn^{2+}) by 1-2 orders of magnitude. The capacity retention rate is 85% after 500 cycles at a current density of $1A \cdot g^{-1}$ [5].In addition, the traditional process of making thick electrodes often leads to problems such as cracks and peeling. DIW uses a layer-by-layer stacking method to manufacture thick electrodes, with a controllable thickness that can exceed 100μm, thereby effectively increasing the volumetric energy density. Research shows that the 3D-printed V_2O_5 thick electrode (200μm thick) has a reversible specific capacity of 475 $mAh \cdot g^{-1}$, which is 1.5 times that of traditional thin electrodes [6]. Furthermore, the multi-functional integration brought by the co-printing of multiple materials enables the embedding of current collectors or protective layers in the cathode. For instance, when printing the manganese dioxide (MnO_2) cathode, a silver nanowire grid is embedded simultaneously, which enhances the conductivity and inhibits the dissolution of active substances at the same time, ultimately extending the battery's cycle life to 1,000 times.

3.2. Design of highly stable anodes

Dendrites and side reactions form at the zinc anode, which is the main factor affecting the cycle life of Aqueous zinc-ion batteries (AZIBs). 3D printing can solve this problem by creating three-dimensional scaffolds and interface adjustments.

3D printing has several practical applications in the preparation and performance improvement of zinc-based anodes. The first is to produce three-dimensional porous skeleton zinc anodes using DIW or FDM technology. For example, zinc/nickel (Zn/Ni) lattice or zinc/carbon nanotube composites. They have a large specific surface area, which can reduce the local current density and make the electric field distribution more uniform. For instance, Zhang Xian 'an et al. successfully constructed a connected porous nickel-zinc lattice anode through face projection micro-stereolithography technology. The symmetrical battery fabricated with this anode did not form dendrites after cycling for 300 hours at a current density of 2 $mA \cdot cm^{-2}$, and its Coulombic efficiency remained above 98% [7]. In terms of alloying modification, the direct-injection wetting (DIW) method is adopted to add aluminum, bismuth, etc. to the zinc anode, which can adjust the zinc deposition process. In the experiment, zinc-bismuth (Zn-Bi) alloy anodes were prepared. After printing, this anode can generate a uniform intermetallic compound layer, which can effectively prevent the formation of dendrites. When this alloy is used as the material for symmetrical batteries, its service life can reach 4400 hours [8]. In addition, 3D printing can also form an interface protective layer on the zinc surface, for example, PVDF-based composite coating. The change in the content of the β phase will promote the ferroelectric effect and facilitate the uniform deposition of zinc ions (Zn^{2+}). The most representative one is the PVDF-MXene-coated zinc anode studied by Zhang Hongcheng et al. After

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cycling for 4,200 hours at a current density of 1 $mA \cdot cm^{-2}$ in a symmetrical battery, the overpotential remains below 50 mV [9].

The anode of aqueous zinc-ion batteries (AZIBs) is usually a zinc sheet or zinc foil. However, during the charging and discharging process, zinc dendrites will appear on the anode. These dendrites can produce "dead zinc", which can lead to a decrease in capacity. In severe cases, it may even cause the separator to be punctured, thereby causing a short circuit [10]. At the same time, side reactions such as hydrogen evolution can lead to electrode polarization and gas production. The introduction of 3D printing has two advantages. The first point is that zinc powder can be directly printed on a flexible substrate to prepare a three-dimensional porous structure with complex pores and networks. This enables active substances like zinc powder to be directly and evenly distributed on the substrate. This porous network structure helps to uniformly disperse the electric field, thereby reducing the local current density and preventing excessive accumulation in certain parts. This way, not so many dendrites will be produced. Side effects will also be suppressed. n addition, DIW can also be used for controllable doping of zinc alloys. Different ink components can be printed by adding a small amount of other metals, such as aluminum or nickel, to the zinc anode, resulting in a uniform zinc alloy layer. The structure of alloyed zinc is relatively stable, which greatly reduces the tendency of dendrite formation. Because the crystals formed during deposition are more stable, sharp dendrites will not grow [11]. The layer-by-layer deposition characteristic of DIW makes this doping process more uniform and controllable. This further effectively enhances the inhibitory effect on dendrites.

Stephen A et al. successfully fabricated flexible 3D zinc anodes with larger pore sizes and specific surface areas using DIW technology, overcoming the mechanical instability issues observed in conventional 3D-printed electrodes [12]. They used several different inks, including glycerin, to print out highly conductive three-dimensional zinc structures layer by layer on the substrate. The three-dimensional array structure reduces the overpotential required for zinc nucleation, making the electric field distribution on the anode surface more uniform. This battery offers A high specific capacity of 650$mAh \cdot g^{-1}$ under the condition of 0.2$A \cdot g^{-1}$. It can be seen from this that the 3D printed zinc anode not only successfully prevents the formation of zinc dendrites, but also greatly increases the diffusion rate of zinc ions.

3.3. Manufacturing of customised flexible full batteries

Flexible electronic devices put forward higher requirements for the shape matching and mechanical durability of batteries.

In terms of battery structure design and performance improvement, the DIW process has a relatively high degree of freedom. The first manifestation is that it can achieve non-planar structure integration, precisely printing electrodes and electrolytes on curved substrates such as ear models or flexible substrates, enabling seamless integration of batteries and devices. For instance, Ahn et al. printed Zn//MnO_2 batteries on parabolic surfaces, achieving a filling rate of 97%. Even in the case of bending, the battery capacity retention rate could still reach over 95% [13]. he second manifestation is that it can be formed in one piece. By using the DIW method, the positive electrode, electrolyte and negative electrode can be printed in sequence, eliminating the traditional packaging process. For instance, the gel electrolyte obtained through printing has good contact with the electrode, with an ionic conductivity of 1.2$mS \cdot cm^{-1}$ and a volumetric energy density reaching 50.5$mWh \cdot cm^{-3}$ [14]. Adjusting the printed track and ratio can also improve the mechanical properties of the battery, enhancing its tensile and bending resistance. Research has found that flexible batteries printed by

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mixing thermoplastic elastomers with electrodes still have a capacity loss of less than 5% after 100 bends [15].

The traditional two-dimensional thin sheet battery is generally composed of a positive electrode coated with slurry, a zinc foil negative electrode and a separator. It has a fixed two-dimensional shape and is not variable. It cannot be randomly attached to complex surfaces. Under the action of external forces, its mechanical stability will be damaged, which limits its application in complex electronic devices such as wearable devices and flexible robots [16]. The birth of 3D printing technology has brought a brand-new solution to this difficult problem. By using Direct Ink Writing (DIW) technology, Zinc-ion batteries (ZIBs) can be precisely manufactured on any curved surface without damaging the overall structure, and can also be perfectly combined with some irregular objects such as human ears and arms.

To achieve "customizable shape", the key lies in adjusting the properties of the printing ink so that the positive electrode, negative electrode and hydrogel electrolyte of the battery can be printed layer by layer without any gap on the same substrate, and finally form a complete battery cell [17]. Ahn et al. designed a non-planar 3D printing technology using DIW technology to manufacture the functional parts of ZIBs. By adjusting the colloid reaction in the ink (polymer viscosity, shear thinning) and the uniformity of nanoparticles, dual ion/electron conductive channels were formed. The ZIB prepared by this method has excellent instantaneous performance, with a volumetric energy density of 50.5mWh·cm^{-3}. Moreover, its capacity can still reach over 90% of the original value after 100 cycles, demonstrating good stability [18]. It is worth mentioning that in the manufacturing process of flexible full batteries by DIW technology, after precise model design and print route optimization for complex curves, the coverage area of electrodes and porous materials in complex non-planar regions has reached over 97%, and the total area utilization rate is as high as 99.2%. Compared with the traditional template printing process, it greatly improves the space utilization efficiency of materials and can also be compatible with the shapes of complex structures. Due to the good toughness and adhesion of the battery itself, the ZIBs printed by DIW have excellent mechanical endurance performance - under large bending conditions with a curvature radius of less than 5mm, the stability loss is negligible, and the cycle stability remains above 95% [19]. The experimental results show that the on-demand manufacturing flexibility of 3D printing enables it to match complex geometers arbitrarily without sacrificing battery performance, providing a foundation for the application of flexible energy storage devices in various fields.

4. Optimisation of printing parameters tables

The 3D printing parameters directly determine the structure and performance of the electrode. The key parameters are: In the 3D printing of AZIBs electrodes, there are many important process parameters that need to be noted. Firstly, nozzle size and printing speed: Although choosing a small-diameter nozzle (50-100μm) increases printing accuracy, small-diameter nozzles are more prone to clogging and require low-viscosity ink; Too high a printing speed will cause porous electrodes, while too low a printing speed will lead to material accumulation. The actual adjustment range is usually between 5 and 20mms^{-1} [20]. Secondly, the layer thickness and porosity must be controlled. Increasing the layer thickness reduces the electrode resolution, but minimizes the interface resistance. Porosity is mainly determined by the printing spacing. The optimal porosity of the electrodes of AZIBs is approximately 40% to 60% [21]. Finally, the curing conditions need to be controlled. If photosensitive ink is used, the intensity of ultraviolet light and exposure time must be carefully adjusted to avoid incomplete curing or excessive cross-linking. For thermal curing, the

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curing temperature must be lower than the decomposition temperature of the ink material, with a maximum of 150°C [22].

5. Innovation in material systems

There are two effective ways to prepare raw materials for 3D printed batteries: First, ink system design: Adding nanoparticles such as MXene and bismuth nanosheets to the ink can enhance rheological properties and also improve conductivity. For instance, after doping $Zn(OTf)_2$ in the ink of PVDF-HFP, the content of the β phase increased from 30% to 70%, and the zinc ion conductivity doubled [23]. Secondly, multifunctional material blending: Mixing conductive polymers and MOFs together can simultaneously enhance the transmission efficiency of electrons and ions. The capacity retention rate of the PANI/Mn-MOF cathode material at 5 $A \cdot g^{-1}$ is 78% [24].

D printing technology endows AZIBs electrodes with a special structure, mainly improving battery performance in three aspects. The first is the structural gain effect: The fabricated three-dimensional porous structure significantly increases the specific surface area of the electrode. On the one hand, it provides more reaction sites; on the other hand, it effectively shortens the diffusion distance of ions, ultimately achieving a simultaneous improvement in battery capacity and rate performance [25].The second is the stress release effect: The three-dimensional porous structure has a very good space utilization rate, which can slow down the volume change of electrode materials during charging and discharging, prevent electrode cracking and peeling, and thereby improve the cycle stability of the battery [26]. The third is the interface control effect: By using 3D printing to fabricate protective layers or alloy layers, the nucleation sites of zinc can be precisely controlled, reducing the formation of zinc dendrites and various other side reactions, thereby enhancing the coulombic efficiency of the battery [27].

6. Challenges and outlook

6.1. Existing challenges

Constraints on material compatibility necessitate heightened and stricter requirements for ink viscosity and solid content across most 3D printing technologies. This not only limits the application of highly reactive materials but also risks reactions between certain printing materials (such as photosensitive resins) and aqueous electrolytes, thereby further compromising battery stability.

The trade-off between precision and efficiency results in slower speeds and higher costs for high-precision printing (<50 μm), making it difficult to meet mass production demands. Large-scale printing also risks structural deformation.

Consistency issues persist, such as temperature and pressure fluctuations during printing potentially causing uneven electrode performance. Yield rates in batch production require improvement.

6.2. Future development directions

Utilising novel printing technologies such as exploring the application of Stereolithography (SLA) and Selective Laser Sintering (SLS) in AZIBs to broaden material selection; developing multi-material co-printing techniques to achieve integrated manufacturing of electrodes-electrolyte-encapsulation.

Innovating functional inks, for instance designing self-healing and smart responsive inks, to enhance battery mechanical stability and environmental adaptability; Developing low-toxicity, high-

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solid-content inks offers advantages in cost reduction and enhanced safety.

Combining finite element simulation with machine learning to optimise electrode structural parameters (e.g., porosity, pore size distribution) for precise performance prediction and process parameter control.

Expanding application scenarios to advance 3D-printed AZIBs in wearable medical devices, flexible displays, IoT sensors, and other fields, broadening customisable energy storage solutions.

7. Conclusion

3D printing technology offers novel avenues for optimising performance and structural innovation in aqueous zinc-ion batteries. By constructing porous electrodes, suppressing dendrite growth, and enabling flexible customisation, it significantly enhances the battery's energy density, cycling stability, and mechanical properties. Although challenges remain in material compatibility and process efficiency, the iterative advancement of printing techniques and innovation in material systems position 3D-printed AZIBs for commercial application in flexible electronics and large-scale energy storage. Future efforts should focus on overcoming multi-scale manufacturing bottlenecks to accelerate the industrialisation of laboratory breakthroughs, thereby injecting fresh momentum into the development of new energy devices

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