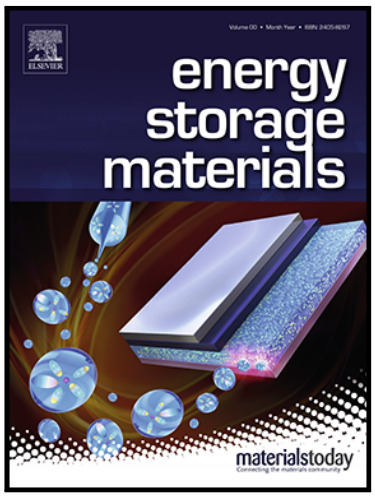

Achieving ultra-long lifespan Zn metal anodes by manipulating desolvation effect and Zn deposition orientation in a multiple cross-linked hydrogel electrolyte

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Achieving ultra-long lifespan Zn metal anodes by manipulating desolvation effect and Zn deposition orientation in a multiple cross-linked hydrogel electrolyte

Highlights

1. An ultra-long cycle life of 5000 h with a high coulombic efficiency of 99.5%;
2. Reducing the desolvation energy barrier (62.31 kJ mol^{-1} to 36.42 kJ mol^{-1});
3. Guiding the preferential orientation of Zn deposition simultaneously;
4. Use a simple one-pot synthesizing method.

Abstract

Aqueous zinc-ion batteries (ZIBs) have received extensive attention due to the intrinsic advantages of high safety, low cost and environmental friendliness. But detrimental side reactions and dendrite problems resulting from high desolvation penalty and inhomogeneous Zn^{2+} flux at the interface severely hindered their practical applications. Herein, a novel polyacrylamide-poly (ethylene glycol) diacrylate-carboxymethyl cellulose (PMC) hydrogel electrolyte is designed to overcome these obstacles through reducing the desolvation energy barrier and guiding the preferential orientation of Zn deposition simultaneously. Theoretical calculation and experimental results reveal that the desolvation activation energy of the PMC hydrogel electrolyte (36.42 kJ mol^{-1}) is substantially lower than that of conventional $ZnSO_4$ aqueous electrolyte (62.31 kJ mol^{-1}) due to the regulated Zn^{2+} solvation structure. Based on the high binding energy between amide groups and Zn^{2+}, polymer chains in PMC also serve as facile Zn^{2+} transport channels to guide the uniform deposition behavior on the Zn (002) crystal surface, which is verified by the grazing incidence XRD analysis. Consequently, an ultra-long cycle life of 5000 h with a high coulombic efficiency of 99.5% is achieved by using the PMC hydrogel electrolyte. Considering the simple one-pot synthesizing method, the PMC hydrogel electrolyte provides new opportunities for developing practical ZIBs.

Keywords: zinc ion battery, multiple cross-linked hydrogel electrolyte, ultra-long lifespan, desolvation effect, preferred orientation deposition

1. Introduction

With the vast consumption of fossil energy and the severe environmental pollution, the demand for low-cost and sustainable clean energy has been increasing rapidly. Because of the high energy density and excellent cycling stability, lithium-ion batteries (LIBs) have been widely used in portable devices and electric vehicles. However, the potential safety issues and high cost of LIBs have restricted their application in large-scale energy storage fields.[1-4] Due to the relatively high specific capacity of zinc metal (820 mAh g^{-1}) and the non-flammability of aqueous electrolytes, aqueous zinc-ion batteries (AZIBs) bring many unique benefits over commercial LIBs to be worthy of being considered as promising candidates for large-scale electrochemical energy storage applications.[5-7] Nowadays, the most commonly-used electrolyte in the AZIBs is zinc sulfate. However, it would result in the poor cycle stability and low coulombic efficiency of zinc anodes, which can be attributed to the following two reasons: i) the increased internal resistance caused by the generation of by-products (the generation of $Zn_4(OH)_6SO_4 \cdot xH_2O$);[8-10] ii) the internal short circuit caused by zinc dendrites formed disorderly on the anode surface.[11, 12] These problems have seriously hindered the practical application of AZIBs.[8, 9, 13, 14]

So far, many efforts have been made to improve the electrochemical performance of Zn metal anodes, such as alloying of zinc metal,[15, 16] surface coating[11, 17-19], high-concentration electrolyte, and electrolyte additives[20-23]. In addition to these commonly-used strategies, designing hydrogel electrolytes to replace traditional

aqueous electrolytes has been also proved to be effective in restraining Zn dendrite growth and side reactions.[24-26] These designs include: 1. Tuning ions flux and inducing uniform deposition in the interface[24, 27, 28]; 2. Restricting the Two-dimensional diffusion of zinc ions by electrostatic interactions and functionalized groups; [12, 27] 3.Reducing the activity of water molecules by restricting its movement in gel toward suppressing the side reactions.[29] Besides, the quasi-solid hydrogel electrolyte formed by polymer has high stability and certain flexibility, which is also suitable for assembling flexible aqueous zinc ion batteries (FAZIBs). In practical applications, benefit from the superior mechanical strength , the flexible zinc-ion batteries have better resistance to the external impact and damage than traditional aqueous zinc-ion batteries.[30-32] At present, it has a wide range of application prospects in smart wear,[31] biological implantable electronic devices,[33] and extreme environments.[34, 35] Despite the great progress, two underlying points usually has not been paid enough attention for the development of high-performance hydrogel electrolytes for AZIBs. One is that the desolvation process of $[Zn(H_2O)_6]^{2+}$ consumes a huge amount of energy with zinc deposition in an aqueous electrolyte, which may trigger hydrogen evolution reaction (HER). This will increase the pH value at the reaction interface and lead to the formation of by-products ($Zn_4(OH)_6SO_4 \cdot H_2O$). The electronic insulation and non-electroactive properties of the by-products will further increase the uneven distribution of the electric field on the surface of the negative electrode. Although HER can be to some extent reduced due to the decrease of water activity in the hydrogel electrolyte, the

desolvation pentalty still remains formidable. The other crucial factor that should be carefully considered is the crystal orientation of deposition Zn, which is closely associated with dendrite growth. Typically, metallic Zn tends to grow along the (101) plane, that is mostly prone to the formation of vertical Zn dendrites. Recent studies indicate that the binding energy of zinc on the (002) crystal plane is much lower than that on the (101) crystal plane, which is more resistant to the formation of dendrite than the Zn(100) and Zn(101) crystal planes.[36] Accordingly, various interface modification approaches such as introducing graphene coating layer and using more (002) basal plane texture exposed Zn foil have been adopted to affect the crystal orientation of deposition Zn.[7, 37] However, how to guide the (002) crystal plane preferred Zn plating/stripping behavior in the hydrogel electrolyte is rarely involved. Therefore, it is of a great challenge to develop a novel hydrogel electrolyte that can reduce the activation energy of desolvation and guide the deposition of zinc ions on the Zn (002) crystal surface simultaneously.

Herein, we designed a simple one-pot method to prepare a hydrogel electrolyte PAM-PEGDA-CMC (PMC) with the ability to reduce the activation energy of desolvation process, and guide the preferential orientation of zinc ions deposition. In a rational design, the free radicals generated by APS through thermally initiated, which allow the polymerization of acrylamide (AM) monomers and poly(ethylene glycol) diacrylate (PEGDA) as a cross-linking agent to generate covalent bonds , and finally bridge to form the basic structure of the 3D network.[38, 39]The carboxymethyl cellulose (CMC) molecular chains shuttle through the 3D network to form a

semi-interpenetrating network (Figure Sx).[40, 41] Through electrochemical impedance spectroscopy (EIS) and grazing incidence XRD (GIXRD) analysis, it is found that the amide group of the PAM molecule in the formed 3D hydrogel plays a key role in reducing the desolvation activation energy and achieving the dendritic-free and highly prefer orientation deposition/stripping of the zinc anode. The symmetric Zn/Zn battery assembled by the PMC electrolyte has an ultra-long cycle time (~5000 h), and the Zn/Cu half-battery has highly reversible Zn deposition/exfoliation (99.5% average Coulomb efficiency). Meanwhile, the flexible battery assembled using PMC hydrogel electrolyte shows excellent safety and durable, which can still work normally under the harsh conditions of punching and bending.

2. Results

Synthesis and characterization of materials

The method of synthesis PMC hydrogel electrolyte is shown in Figure 1a. AM, PEGDA, CMC and APS were completely dissolved in 2.0 M $ZnSO_4$ solution to obtain a clear and transparent solution. The mixture was poured into the template and heated to 40 °C on the heating plate. During the heating process, free radicals released from APS acted on the carbon-carbon double bond and initiated the polymerization. Constant temperature for 30 min and cooled down to room temperature to obtain PMC hydrogel electrolyte. As shown in Figure 1b, the hydrogel electrolyte is colorless and transparent. The thickness of the PMC hydrogel electrolyte controlled by the mold is almost the same as the commercially available glass fiber (Figure S2). This makes that the thickness of the battery assembled in the subsequent experiment

is the same and ensures the accuracy of the experiment. At the same time, we found that PMC has good flexibility (Figure S3), which shows that the gel has the prospect in flexible batteries. The tensile strengths of pure PAM gel, PAM-PEGDA and PAM-PEGDA-CMC (PMC) were measured by an electronic tensile testing machine (Figure 1d). It is found that after adding the PEGDA (the cross-linking agent), PAM and PEGDA polymerize to form a 3D structure, which increase the tensile strength of the hydrogel. Furthermore, after adding CMC, the tensile strength of the hydrogel is further improved significantly. This is because CMC and PAM-PEGDA form a semi-interpenetrating three-dimensional network structure, which further increases the mechanical properties of the hydrogel.[42, 43] The tensile strength of the PMC gel electrolyte reached 2.25 MPa. Symmetric batteries were assembled with PMC as the electrolyte, and cycled for 0, 20, and 50 hours, and then disassembled to find that the PMC gel remained intact (Figure S4). This shows that the excellent mechanical properties of PMC can keep it stable during cycling. As shown in Figure 1c, the Fourier transform infrared spectroscopy analysis of PMC in different states, tells that there is no obvious characteristic peak of C=C bond (purple area) in PMC, which indicates that PMC is a covalent cross-linking induced by free radicals obtained by polymerization. The dark green area represents the characteristic peak of zinc sulfate. The light green area is the characteristic peak of Zn-O bond. In order to further prove the reliability of the Zn-O bond, we also performed the XPS test on the PMC. It can be seen from Figure S5 that there are obvious characteristic peaks of the Zn-O bond in the Zn2p analysis pattern.[44] The corresponding characteristic peaks of Zn-O bonds

can also be fitted in the O1s spectrum. The PMC hydrogel electrolyte was dried in an oven at 80 °C, and the water content (free water) was 63.97%. The PMC gel electrolyte was frozen in liquid nitrogen to obtain a relatively flat section. After drying the moisture, scanning electron microscope (SEM) images and Zn EDS element map show that the zinc element is evenly distributed in the PMC hydrogel electrolyte (Figure S6 a&b).

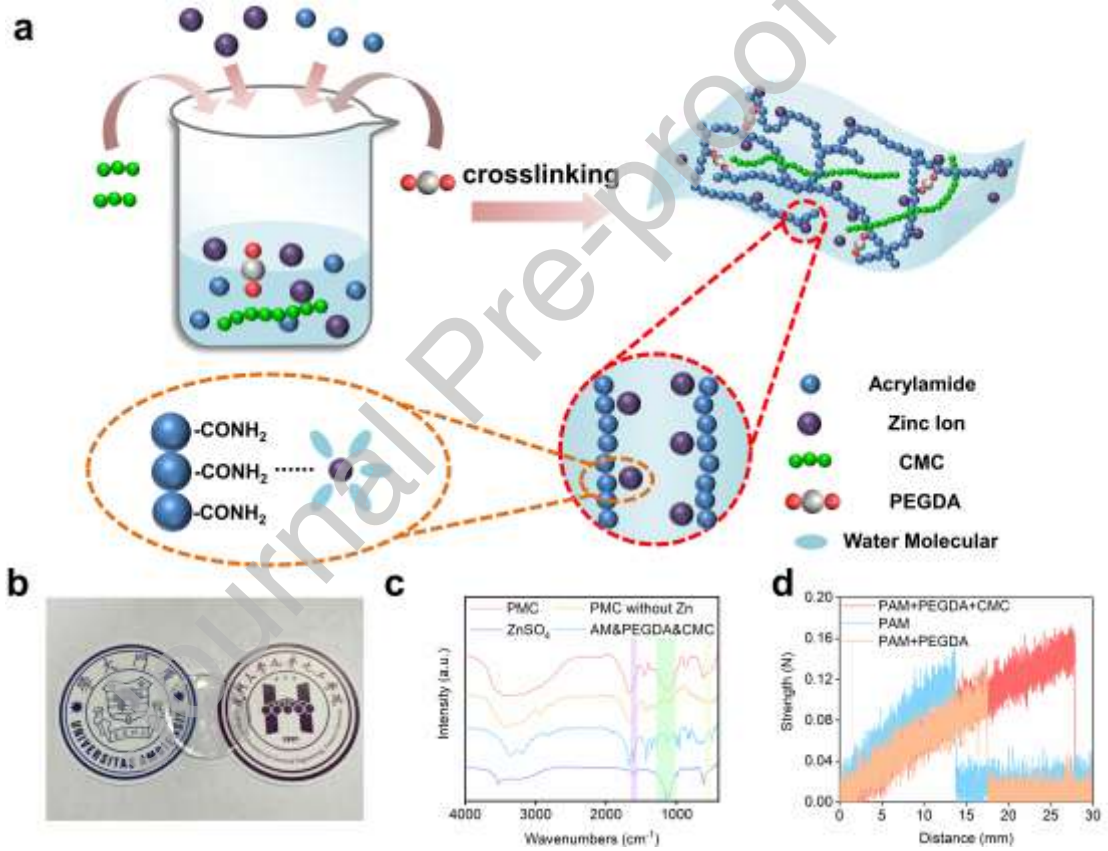

Fig 1. Synthesis and characterization of PMC gel electrolyte. (a) Synthetic schematic diagram of PMC hydrogel electrolyte; (b) Optical photograph of PMC hydrogel electrolyte; (c) Infrared image of PMC hydrogel electrolyte; (d) Tensile strength test (PAM+PEGDA+CMC, PAM and PAM+PEGDA).

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Pengxiang Lin: conceptualization, methodology, investigation, experiment, writing-original draft preparation; **Jianlong Cong:** design of experiments, partial work of the study; **Jiyang Li** and **Minghao Zhang:** discussion of involved mechanism; **Pengbin Lai:** DFT calculation; **Jing Zeng:** supervision; **Yang Yang** and **Jinbao Zhao:** reviewing and editing.

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: