Topological chitosan framework enables reversible columnar array anode for high-performance aqueous zinc batteries

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ABSTRACT Eco-friendly aqueous zinc batteries can replace lead-acid batteries in scenarios that balance safety and energy density. However, the synergistic deterioration between structural collapse and kinetic failure of zinc anodes at high discharge depth and high current densities restricts the actual energy and power densities. Herein, we propose a strategy for the in situ integration of topological chitosan framework (D-CTS) on current collectors by regulating phase separation kinetics during multistage coordination-neutralization electrophoresis. The vertical through-hole array is formed by the coupling of instantaneous and delayed phase separation. Then, the columnar zinc array is mediated by D-CTS to construct the integrated component (D-CTS-Zn) of vertical through-hole separator and array anode. The embedded interconnected nanonetworks within the through-hole wall enable the dynamic equilibrium of columnar zinc array by lateral ion compensation mechanism. As a result, the Zn||Zn symmetric cell with D-CTS-Zn stably cycles over 3000 cycles at 200 mA cm^{-2} under 60% discharge depth. The assembled D-CTS-Zn||MnO_2 battery delivers an energy density of 83 Wh kg^{-1} at an ultrahigh power density of 9.25 kW kg^{-1}. This work provides a constructive strategy for chitosan phase separation regulation and separator-induced reversible metal array anodes.

Keywords: Topological chitosan framework, Delayed phase separation, Instantaneous phase separation, Zinc array anodes, Aqueous zinc batteries

INTRODUCTION

Benefiting from the high specific capacity of zinc (820 mAh g^{-1}) and the intrinsically safe aqueous electrolyte [1-3], rechargeable aqueous zinc batteries (AZB) can replace environmentally harmful lead-acid batteries in scenarios that balance safety and energy density [4-7]. However, the synergistic deterioration between structural collapse and kinetic failure of zinc anodes at high depth of discharge (DOD) and high current densities results in an order of magnitude difference in the AZB lifespan compared to the commercial target (>1000 cycles) [8-10]. This compels current AZB to adopt high N/P ratio (negative/positive capacity ratio) design and low-rate charge/discharge protocol (especially under high cathode active material loading conditions), compromising the expected actual energy density and power density [11,12]. Therefore, improving zinc anode stability at high DOD and high current densities is most critical for the large-scale application of AZB.

To improve the stability of zinc anodes under high current densities or high DOD, some chemical strategies, such as electrolyte additives, can stabilize zinc anodes by regulating the chemical microenvironment during zinc plating and stripping [13-15]. However, it is challenging for single chemical strategy to break through the intrinsic limitations of the structure. It is worth mentioning that structural strategies can modulate electrochemical performance on a larger scale from another dimension and are compatible with chemical strategies. The typically propose strategy is the structural design of separator and electrode structure [15-22]. On the one hand, the separators with vertical through-hole most significantly reduce mass transfer resistance for reducing concentration polarization [23-26]. Chen et al. fabricated an anisotropic through-hole separator via directional freezing to accelerate ion transport [26]. However, such isolated vertical through-holes with dense walls face challenges to maintain the overall uniformity of ion distribution. On the other hand, various array structures are fabricated to reduce local current density and improve the plat-

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Dear Prof. Wang:

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