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Review article

## Stabilization strategies for zinc anode interfaces under high discharge depth

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## ABSTRACT

The practicalization of zinc-based energy storage devices is hindered by instability of zinc anodes and low depth of discharge (DOD). Significantly improving stability under high DOD is a must for achieving zinc-based energy storage devices with high energy density and long lifespan. However, this important scientific issue has only been emphasized in recent years, and the key to its solution lies in realizing a highly reversible zinc deposition/stripping process at the anode interface. Researchers have made breakthroughs in this problem through direct interfacial stabilization strategies with electrolyte additives, in-situ coatings, and in-situ quasi-solid-state electrolytes. In this review, we have summarized the deposition/stripping reactions and various interfacial stabilization strategies under high DOD, providing a reference for constructing high-energy-density and long-life zinc-based energy storage devices.

## 1. Introduction

With the over-consumption of non-renewable resources and the rapid development of artificial intelligence, more and more attention has been paid to the storage and utilization of energy. Electrochemical energy storage technology has the advantages of high energy conversion, high flexibility and high stability, which will surely dominate the field of energy storage in the future [1]. Among many electrochemical energy storage systems, aqueous zinc-based energy storage devices stand out thanks to the high theoretical capacity ( $820 \text{ mAh g}^{-1}$ ,  $5855 \text{ mAh cm}^{-2}$ ), low redox potential ( $-0.76 \text{ V vs. SHE}$ ), high safety, environmental friendliness and low cost [2]. It is widely considered to fully outperform the current commercialized aqueous lead-acid batteries in terms of energy density and power density, and is expected to totally replace them [3]. Moreover, it can be used as a candidate substitute for lithium-based and sodium-based energy storage devices, satisfying application scenarios of high safety and low energy density.

However, the current aqueous zinc-based energy storage devices have two major problems on zinc anodes. One is the instability (including dendrite growth, hydrogen evolution corrosion, passivation, etc.), which shortens the lifespan; and the other is the low DOD, which makes the energy density a performance bottleneck [4]. For the energy density of batteries, the capacity ratio of the anode to the cathode (N/P) close to 1 is the most ideal. But the low DOD of zinc anodes results in N/P far exceeding 1 and too much zinc cannot be effectively utilized. The

practical results show that DOD is limited to 10–50 %, implying 2–8 times oversized zinc anode relative to cathode capacities. Such a low active-material-loading of cathodes seriously induces the low energy density [5]. In the past decade, more and more researches focus on improving zinc utilization to reduce N/P (Fig. 1a). Fig. 1b shows the proportion of different strategies to increase zinc utilization, including strategies on the anode, electrolyte, cathode and separator. Unexpectedly, high DOD often further aggravates the instability of zinc anodes, which sharply shortens the lifespan. In recent years, more and more researchers have realized the necessity of improving the stability of anodes under high DOD and have already made some breakthroughs (Fig. 1c).

Since all the reactions of deposition and stripping occur at the anode-electrolyte interface, anode stabilization strategies directly targeting at the interface are the most efficient. Herein, we dissect the anode-electrolyte interface reactions and summarize various anode-electrolyte interface stabilization strategies, providing theoretical guidance for further improving the stability under high DOD.

## 2. Interfacial reactions of zinc anodes

## 2.1. Zinc deposition reaction

During charging,  $\text{Zn}^{2+}$  is reduced to Zn on the anode surface. The deposition involves three stages: the diffusion and adsorption of  $\text{Zn}^{2+}$ ,

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