

Suppressing Zinc Dendrites: Enhancing Long-Term Stability of Aqueous Zinc-Ion Batteries through Metal-Organic Framework Surface Coating

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Abstract

A surging demand for electricity due to electric vehicles (EVs) and artificial-intelligence (AI) has been accelerating the early adoption of renewable energy. However, the nature of renewable energy is the intermittency of power sources such as wind and solar, which requires large-scale energy storage options. Aqueous Zinc-ion battery (AZIB) is a great candidate since it is affordable, safe, and scalable, but the zinc dendrite issue must be addressed before scale-up. In this work, the zinc metal surface was coated with one of the metal-organic frameworks (MOF-808), and characterized by scanning electron microscopy (SEM), X-ray diffraction (XRD), and electrochemical cycling tests. The MOF-808 coating effectively suppressed zinc dendrites and enabled a long cycle life of over 1400 cycles, compared to only 250 cycles for the uncoated zinc anode, with the MOF-coated anode also demonstrating a higher average capacity of 200 mAh/g versus 170 mAh/g for the bare zinc. Regarding the MOF-808 material optimization, it was shown that controlling the metal-to-ligand ratio is important to form the right material and to make uniform particles.

Keywords: Zinc ion battery, Zinc dendrite, Surface coating, Metal-organic framework

1. Introduction

To enable a net-zero society, it is important to deploy more renewable energy power plants around the world. One of the most critical hurdles in this regard is to tackle the energy source intermittency. We need safe, scalable, and cost-effective ways to store renewable energies (International Renewable Energy Agency, 2025). Aqueous Zinc-ion batteries (AZIBs) use zinc, an abundant and non-toxic element on earth, and they are fundamentally safe because the electrolytes are based on water (Fang et al., 2018). Despite their advantages, there are challenges like zinc dendrite formation that limit the battery's life (Cao et al., 2020). Dendrites are a common phenomenon in metal plating, and this type of metal growth results in non-uniform and porous plating, instead of a uniform and dense deposition. Once these dendrites grow, metal stripping during the reverse reaction also becomes non-uniform, and dendrites can detach. Consequently, the reversibility of the zinc anode decreases with each cycle. Furthermore, the continued growth of dendrites can lead to the penetration of the separator and a short-circuit with the cathode, which poses a critical safety and stability issue that must be resolved. The resulting short-circuit can cause rapid capacity fade, internal heating, and in severe cases, thermal runaway—making dendrite suppression essential not only for performance but also for the safe deployment of large-scale energy storage systems.

Since zinc dendrite formation is primarily a surface phenomenon, surface coating is a direct and effective method for regulating the metal surface and mitigating dendrites. Various coating materials have been adopted to modify the zinc surface, and compared to the bare surface, they showed a longer cycle life and a reduced likelihood of battery short-circuit. One reported example is to coat a mineral called montmorillonite on the zinc anode, which restrains zinc dendrite detachment from the surface and facilitates smooth transport of zinc ions (Wang et al., 2022). A polymer coating is also a surface protection method widely adopted in various fields. Although polymers are mechanically weaker compared to the ceramic mineral previously mentioned, they are reported to enhance the wetting property on

the zinc surface (Chen et al., 2021). This improved wetting allows the liquid electrolyte to be distributed more uniformly, thereby suppressing the initial formation of zinc dendrites.

Metal-Organic Frameworks (MOFs) are a class of hybrid materials that are often considered to bridge the gap between inorganic solids (like ceramics) and organic polymers (Li et al., 1999). MOFs have also been studied to mitigate the zinc dendrite issue. For example, a microporous Zn-based MOF, ZIF-8, was reported to promote uniform zinc plating and stripping (Wang et al., 2019). Separately, MOF-808 was coated on a battery separator and improved the uniformity of Zn ion flux during stripping/plating processes (Maebonruan et al., 2022). More recently, Akai et al. (2025) demonstrated a water-resistant redox-active MOF capable of stable charge storage in aqueous electrolytes, highlighting the expanding utility of MOF-based materials in next-generation aqueous battery systems. All these improvements were attributed to the MOF's unique nano-scale open structure, which allows small atoms and ions, such as zinc ions, to be transported through the material (Chai et al., 2023). In other words, a uniform supply of zinc ions to the zinc surface is effective to suppress zinc dendrite formation.

In this work, the direct modification of the zinc anode surface with MOF-808 was explored as an alternative to previous approaches modifying the battery separator. A series of experiments was conducted by controlling the reactant ratio and synthesis time to understand the effect of these conditions on zinc dendrite mitigation and battery performance. The objective of this study was to identify the optimal MOF-808 synthesis conditions for enhancing the stability and longevity of aqueous zinc batteries. It was found that the reactant ratio is the key parameter for synthesizing a stable MOF-808 and effectively mitigating the zinc dendrites.

2. Materials and Methods

2.1 Synthesis of MOF-808

MOF-808 was synthesized with a hydrothermal reaction method. 1,3,5-Benzenetricarboxylic acid (H3BTC) and $ZrOCl_2 \cdot 8H_2O$ were mixed in various molar ratios of 1:1, 1:3, and 1:5. The amount of H3BTC was fixed to be 0.525g, and $ZrOCl_2 \cdot 8H_2O$ amounts were 0.8, 2.4, and 4g, respectively. 20mL of acetone and 20mL of H_2O were added to the mixtures and sonicated for 10 minutes before transferring the mixture to a Teflon-lined autoclave. The hydrothermal reaction was conducted at 85°C for 12, 24, and 48 hours.

After cooling, the solution inside the autoclave was recovered, and the powder was collected through centrifuging at 12,000 rpm for 5 minutes. Subsequently, the supernatant was removed and the powder product was washed three times. The first wash was with H_2O , followed by 2 additional washes with ethanol. The remaining white solid (MOF-808) after washing was dried and ground evenly to achieve a uniform particle size.

2.2 Characterizations

Powder size and morphology of MOF-808 were characterized by scanning electron microscopy (SEM; Nova NanoSEM 450). X-ray diffraction (XRD; Rigaku, Miniflex 600) was used to confirm the successful synthesis.

For electrochemical characterizations, electrodes and coin-type battery cells were fabricated to verify if MOF-808 coating could protect the surface of zinc anode and thereby suppress zinc dendrites. 0.09g of MOF-808 and 0.2g of Polyvinylidene fluoride (PVDF) binder solution (5 wt.% concentration) with a small amount of NMP solvent were ball-milled until the slurry was sufficiently homogeneous. The MOF-808 slurry was cast on a zinc film and dried. Once completely dried, uniformly coated areas were selected and punched into 10 mm diameter electrode specimens. The resulting MOF-808 coating thickness was approximately 5–10 μm , with an areal loading of ~ 0.45 mg/cm^2 . Cell assembly proceeded with the electrodes and 2M $ZnSO_4$ electrolyte. For a full cell evaluation, I_2 (Iodine) cathode on carbon was used. Finally, the charge and discharge cycle data were collected by a battery cycler (WonATech WBCS 3000).

3. Results and Discussion

MOF-808 was synthesized with a hydrothermal method. Two reactants (H3BTC and $ZrOCl_2 \cdot 8H_2O$) were mixed with molar ratios of 1:1, 1:3 and 1:5. In parallel, reaction time was also controlled at 12, 24, and 48 hours. Figure 1

presents the MOF-808 powder morphology and size distribution as a function of reaction time, with the reactant ratio fixed at 1:3. The SEM images in Figure 1, corresponding to reaction times of 12 h, 24 h, and 48 h, show that no significant difference in powder morphology and size was observed. The average particle size remains consistent at approximately 1 micrometer. This result suggests that under the fixed 1:3 reactant ratio and 85°C condition, the particle growth is largely finished within 12 hours, suggesting that reaction time is not the determining factor for final particle size and morphology in this synthesis.

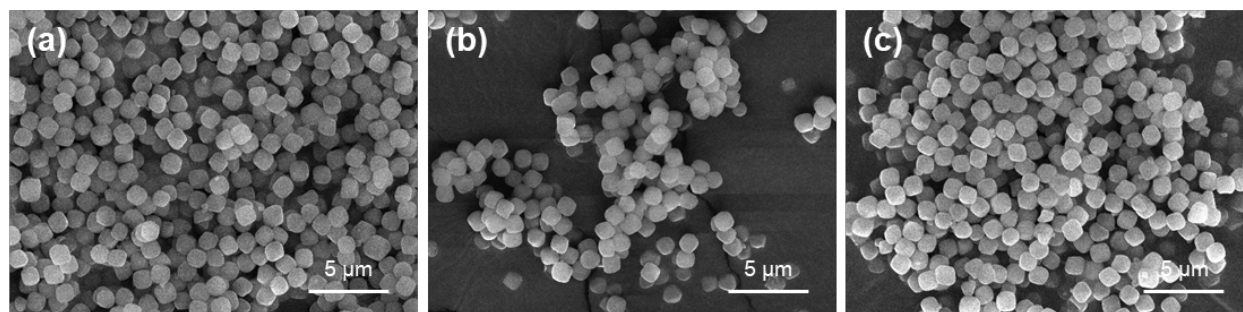


Figure 1. SEM images of MOF-808 particles synthesized at 85°C with different reaction times: (a) 12 h, (b) 24 h, and (c) 48 h.

However, when the reactant ratio is altered, there is a significant change in the powder morphology. As shown in Figure 2, for instance, the 1:1 reactant ratio resulted in random large particles, while the 1:5 ratio produced irregularly shaped, larger particles with an average size of 2~3 micrometers. This clearly indicates that the reactant ratio has a large influence on the final MOF-808 morphology. Particle size and morphology are critical for battery performance because smaller, more uniform particles enable a denser, more homogeneous coating layer on the zinc anode. A uniform coating promotes even ion transport across the electrode surface, which in turn reduces local current hotspots that initiate dendrite nucleation. Irregular or large MOF particles result in a non-uniform coating layer on the zinc anode surface. Such coating non-uniformity leads to an uneven distribution of zinc-ion flux during cycling, which undermines the effectiveness of dendrite suppression.

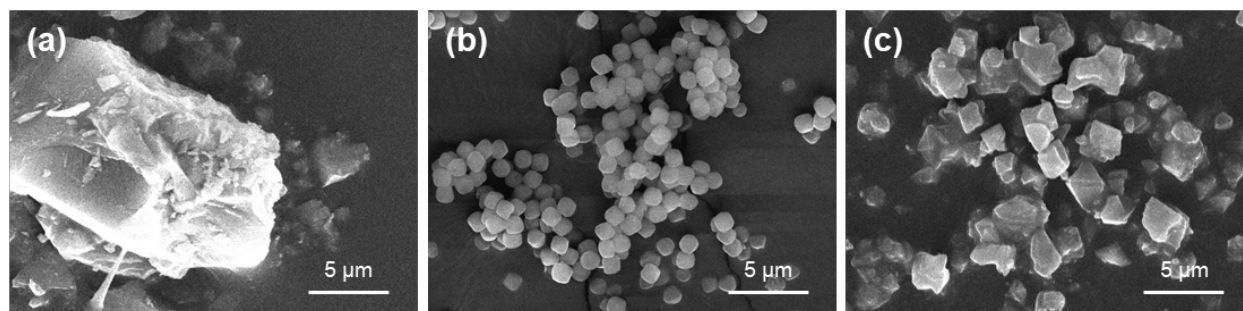


Figure 2. SEM images of MOF-808 particles synthesized with various reactant ratios (H3BTC and $ZrOCl_2 \cdot 8H_2O$): (a) 1:1, (b) 1:3, and (c) 1:5.

Figure 3 shows X-ray diffraction patterns of the synthesized MOF-808 powder. As shown in Figure 2, the reactant ratio strongly influences whether MOF-808 can be successfully synthesized. The 1:1 reactant ratio does not form MOF-808 and shows broad peaks, which is consistent with the large powder chunks in Figure 2. It suggests that the 1 $ZrOCl_2 \cdot 8H_2O$ to 1 H3BTC ratio may not have enough Zr and cannot form nanocrystal structure. When the 1:3 and 1:5 reactant ratios are compared, the 1:5 ratio shows broader diffraction peaks, which indicates more impurities present in the powder due to excess amount of $ZrOCl_2 \cdot 8H_2O$. Overall, the 1:3 reactant ratio condition makes a good MOF-808 material without detectable impurities, and the reaction time does not have much influence on the final powder properties. Therefore, this sample was finally selected for further

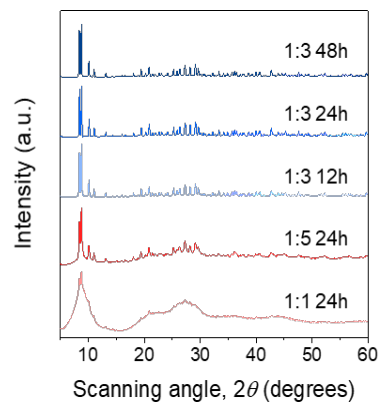


Figure 3. XRD patterns of the synthesized MOF-808 powders.

characterization with battery tests.

MOF-808 was uniformly coated on the zinc anode (MOF/Zn) as shown in Figure 4, and it was compared with a bare zinc anode. Symmetric coin cells were fabricated using Zn metal on both the cathode and anode. MOF/Zn

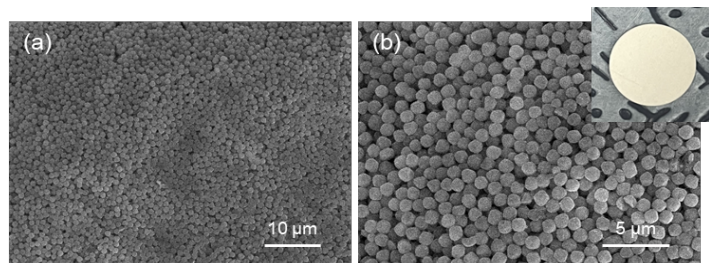


Figure 4. Top-view SEM images and digital image of the MOF/Zn anode.

symmetric cells were also made for comparison. The symmetric cells were charged and discharged with a current density of 1 mA/cm² for 1 hour, where Zn metal was alternately plated and stripped during each charge and discharge cycle. Figure 5 shows voltage curves of Zn and MOF/Zn symmetric cells during electrochemical cycling.

It is clearly shown that the bare Zn symmetric cells exhibit numerous voltage spikes in the curves, which is indicative of irregular and unstable electrochemical reactions caused by Zn dendrites. These voltage spikes are a direct consequence of dendrite formation: as dendrites grow unevenly, they create localized high-resistance regions and sudden short-circuit pathways that manifest as abrupt deviations in the voltage signal. In comparison, the MOF/Zn symmetric cells display smooth and continuous voltage curves without sudden voltage spikes. This suggests that the MOF-808 coating successfully stabilized the Zn plating and stripping reactions, enabling stable cycling.

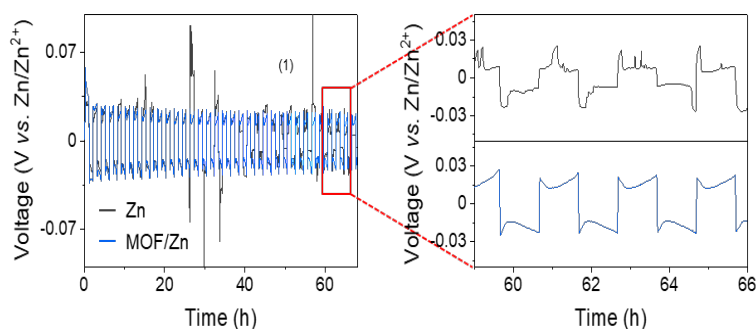


Figure 5. Cycling performance of Zn-symmetric cells with Zn and MOF/Zn anodes. The cells were cycled at 1 mA/cm² with an areal capacity of 1 mAh/cm².

It was necessary to examine the Zn anode surface after cycling to verify the effect on Zn dendrites. After 50 cycles, the symmetric cells were disassembled, and the surfaces of Zn and MOF/Zn anodes were characterized with SEM (Figure 6). The MOF-808 coating was carefully removed to observe the underlying Zn surface and Zn dendrites. The bare Zn surface turned porous and rough after cycling, whereas the Zn surface underneath MOF-808 was relatively smooth and dense. It indicates that the surface-coated Zn anode has notably fewer Zn dendrites during cycling, which is consistent with the stable cycling data in Figure 5. It was also observed that the MOF-808 coating layer turned porous and showed some cracks after cycling. This may be because the Zn metal was repeatedly plated and stripped underneath the MOF-808 coating, and the top coating layer became unstable during the cycling test. However, despite the cracks, the surface coating stays in place and has no visible contamination.

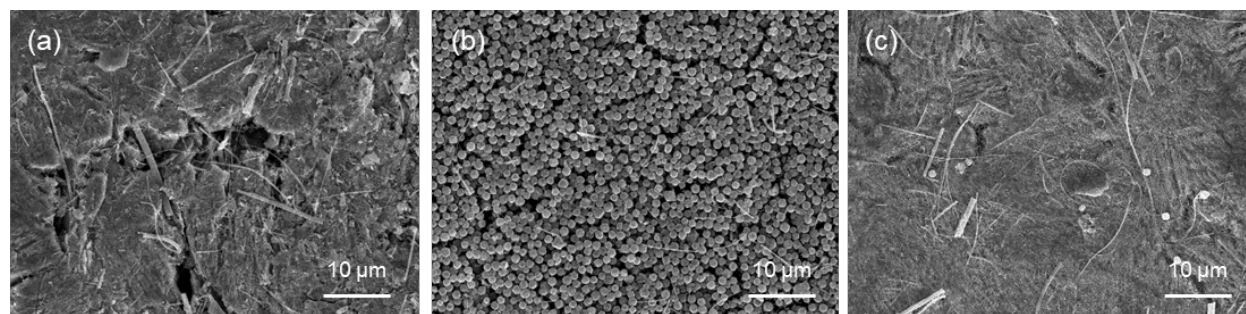


Figure 6. Top-view SEM images of the cycled anodes of Zn-symmetric cells after 50 cycles: (a) Zn anode, (b) MOF/Zn anode, and (c) Zn surface underneath the MOF layer.

The Zn metal surface was further characterized with XRD to track any changes during cycling. Figure 7 compares X-ray diffraction patterns of the bare Zn surface (black line) and the Zn surface from the MOF/Zn sample (blue line).

It shows a signal from $\text{Zn}_4\text{SO}_4(\text{OH})_6 \cdot x\text{H}_2\text{O}$ (red triangles) that is precipitated from the aqueous electrolyte. There is a clear contrast in the precipitate signal, where the bare Zn surface has stronger diffraction peaks and more surface contaminations. It can be interpreted that a rougher surface morphology shown in Figure 6a provides more surface area for precipitation. The smooth Zn surface of the MOF/Zn sample not only has less surface area for precipitation, but also limits direct contact between the liquid electrolyte and the Zn surface. The surface contamination can negatively affect uniform Zn plating and stripping reactions. For example, if the precipitates exist randomly, those affected areas may block Zn plating and stripping, and irregular Zn dendrites grow easily. This provides further evidence that the surface coating with MOF-808 can improve reversible Zn plating and stripping reaction for many cycles.

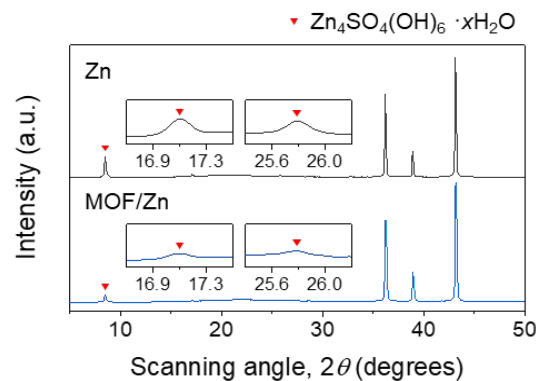


Figure 7. XRD patterns of Zn and MOF/Zn anodes of the Zn-symmetric cells after 50 cycles.

Finally, zinc-iodine (Zn-I_2) batteries with the Zn and MOF/Zn anodes were electrochemically characterized. The Zn and I_2 combination exhibits an average operating voltage of 1.2V, as shown in Figure 8.

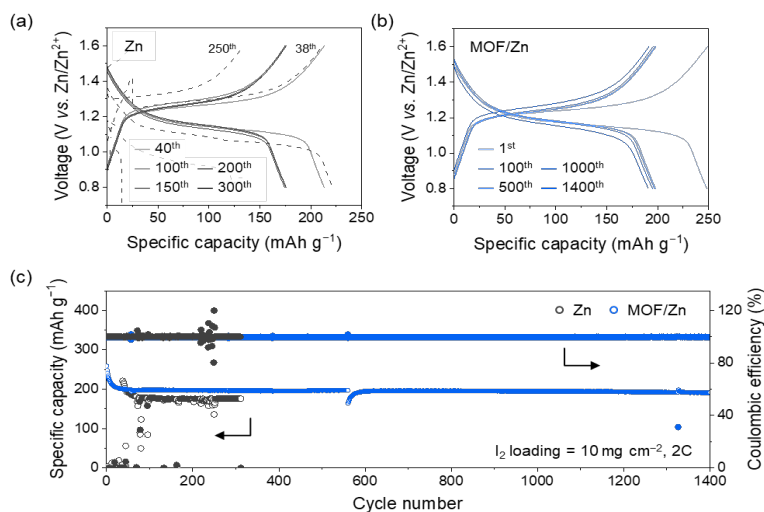


Figure 8. Charge/discharge voltage profiles of the Zn-I₂ full cells with (a) Zn and (b) MOF/Zn anodes. (c) Capacity retention values and Coulombic efficiencies of the full cells with different anodes.

The Zn-I₂ battery with bare Zn anode shows capacities of 170 mAh/g and becomes unstable after 200 cycles. It eventually stopped after 250 cycles due to a short-circuit. In contrast, the battery with MOF/Zn demonstrated stable cycling with average capacities of 200 mAh/g and lasted for 1400 cycles without notable capacity loss. The voltage curves are reproduced consistently and do not show any signs of degradation, such as voltage spikes. The MOF-808 surface coating stabilizes Zn anode, suppresses Zn dendrites, and delivers high-capacity and long-lasting battery performance.

4. Conclusion

To overcome the challenging zinc dendrite issue in aqueous zinc-ion batteries (AZIBs), MOF-808 was selected and successfully synthesized by controlling various reactant ratios. With an optimal reactant ratio, the resulting nano-structured MOF-808 demonstrated effective Zn dendrite suppression, as confirmed by electrochemical characterizations. The MOF coating was found to limit direct contact between the liquid electrolyte and the Zn metal, which maintains a clean and smooth Zn surface. This significant reduction in surface contamination resulted in uniform Zn plating and stripping cycles. These results suggest that modification of the Zn metal surface is an essential strategy to enable the large-scale application of AZIBs for renewable energy storage.

The success of MOF-808 as a surface coating material highlights the broader potential of porous hybrid frameworks for stabilizing metal anodes in aqueous battery systems. By providing a chemically compatible, ion-permeable barrier, MOF coatings offer a versatile approach that could be extended to other metal anodes or battery chemistries beyond Zn-I₂. Future research should investigate the long-term mechanical durability of the coating beyond 1400 cycles, as well as the electrochemical behavior under higher current densities and elevated temperatures

relevant to real-world applications. Additionally, strategies such as thicker coatings or composite MOF–polymer layers may further improve coating stability and mitigate the cracking observed after extended cycling. From a practical standpoint, scaling up the hydrothermal synthesis and slurry-coating process to larger electrode areas will be critical for commercial feasibility.

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