Achieving (002) Textured Zn Metal Anode With Dendrite-Free Morphology By Synergetic Leveling of the Reactive sites

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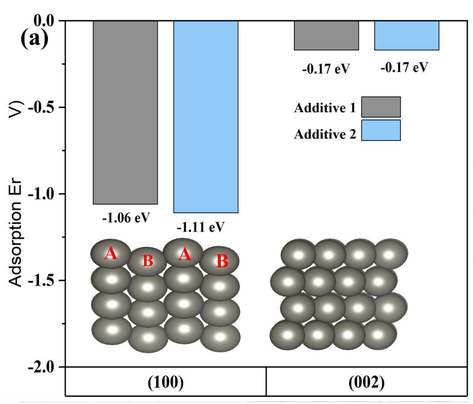
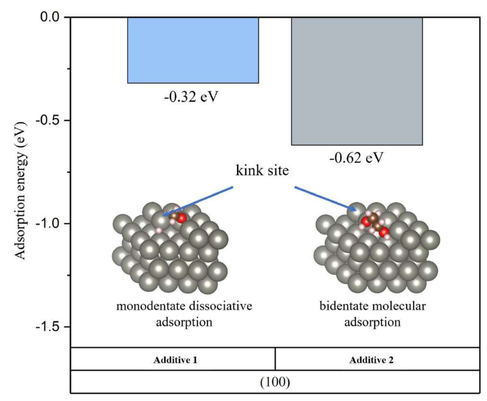
INTRODUCTION

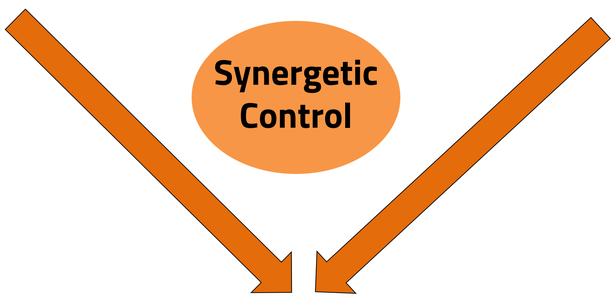
Aqueous zinc-ion batteries are safe and inexpensive energy storage candidates with tremendous potential for applications in both renewable storage and electricity grid. However, the practical progress has been restricted, largely owing to the poor Zn anode cyclability stemming from dendritic structure deposition and accompanying side reactions. Few studies paid considerations to the reactivity of the zinc sites on growing crystallites and the surface, which trigger dendritic deposition. Here, we identify those reactive sites to be on the corrugated $\{100\}_{hexagonal}$ surface of the nucleating zinc crystallites and around surface irregularities and demonstrate their effective leveling by a combination of two different electrolyte additives. The unique mechanistic advantage of the two additives and their synergism endow comprehensive control over the zinc morphology for durable cycling.

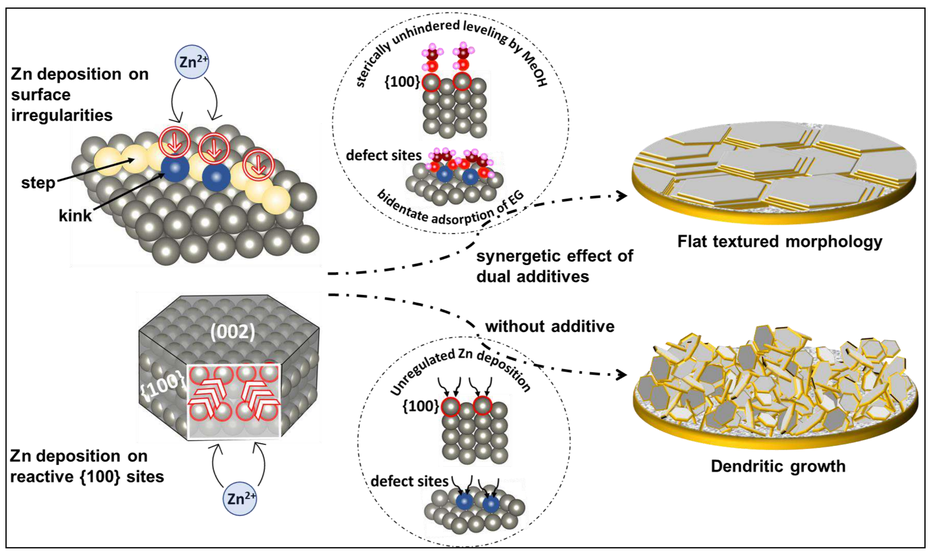
Synergetic Action of the two Electrolyte Additives: One Does Not Fit All





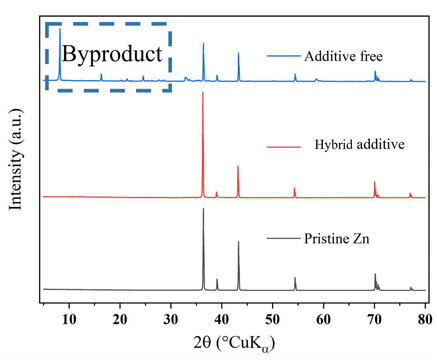
- Although both additives strongly adsorb on {100} sites, additive 1 allow most effective control of the reactive A sites owing to the low steric influence
- Additive 2 provide most effective blocking of the surface irregularities (e.g., kinks) through bidentate binding

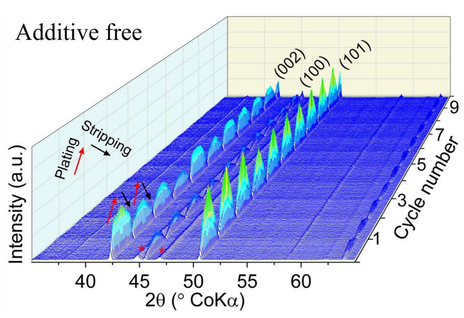


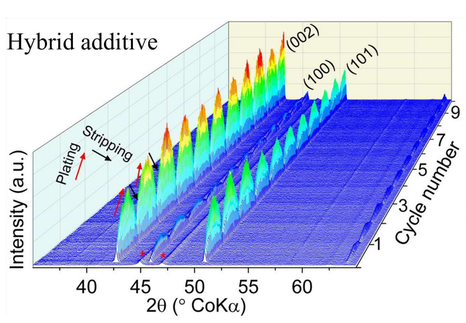


- In the absence of the electrolyte additive, the growth follows the addition of zinc on the {100} planes that are incidentally corrugated with zinc sites of uneven reactivity , triggering the dendritic deposition. This issue is further aggravated by the surface irregularities.
- With the two electrolyte additives in an optimal ratio, both types of active sites are blocked, and an effective morphology control is achieved.

Electrodeposited Zinc Morphology

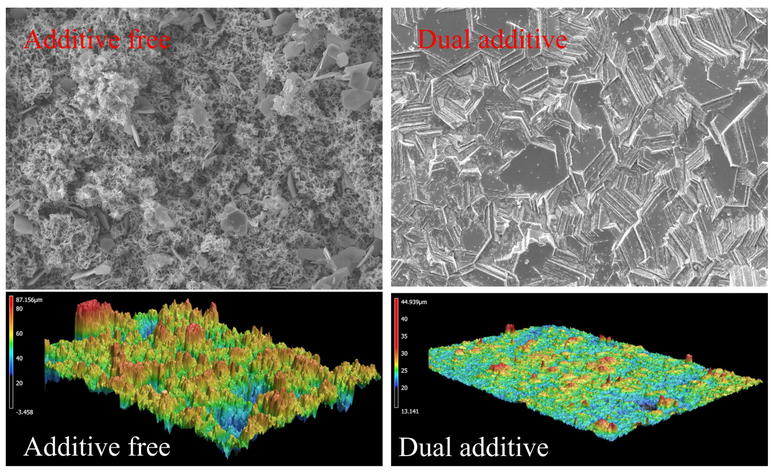






- The dual additives can suppress the byproduct formation after cycling
- The maximum (002)/(001) intensity ratio can be achieved in optimized hybrid additives after 20th plating.
- The synergism of the two additives in the deposition control take effects from the very first cycle.

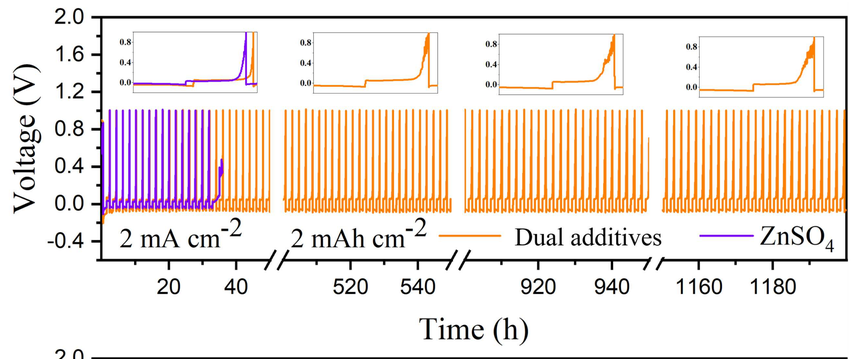
Comact deposition, flatter and smooth surface is achieved with dual additives in the electrolyte.

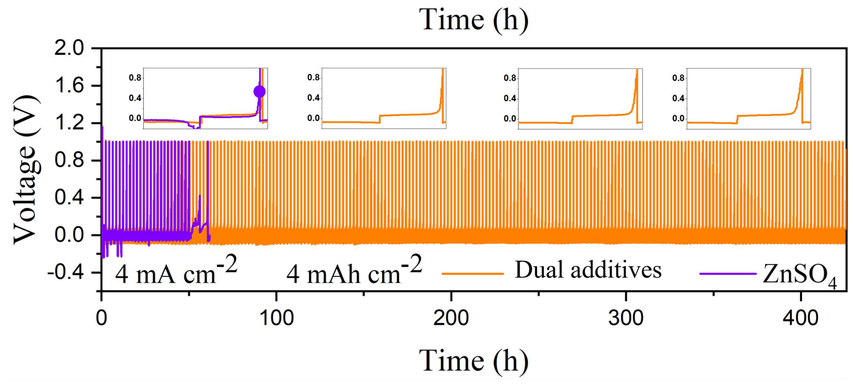


The final morphology is strongly correlates with the Zn anode cycling performance

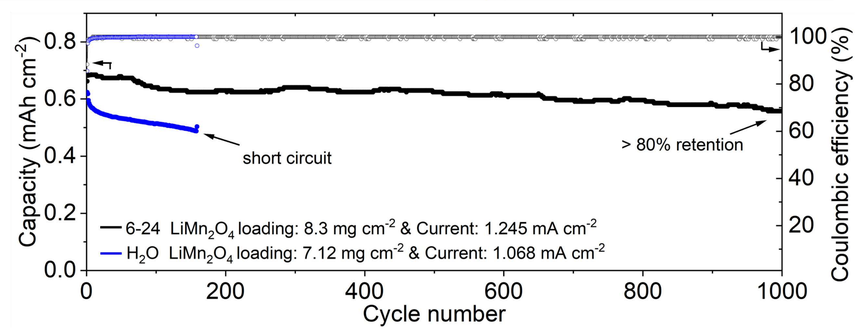
Electrochemical Performances

The dual additives endow the Zn anode with ultra-stable reversibility and cyclability at different practical cycling condition.





The stability of Zn cycling translates to the long-term full cell cyclability: 80% capacity retention after 1000 cycle under high cathode loading.



Conclusion

The dual additives in the electrolyte can :

1. simultaneously passivate the reactive atoms and irregular surface sites during cycling;
2. significantly suppress the dendritic growth and byproduct formation;
3. endow the Zn anode with densely stacked morphology with (002) texture;
4. deliver exceptionally stable cyclability in Zn-Ti half cell and $LiMn_2O_4$/Zn full cell

ACKNOWLEDGEMENTS