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Low-cost and safe hybrid aqueous zinc ion batteries using ethyl cellulose as binder

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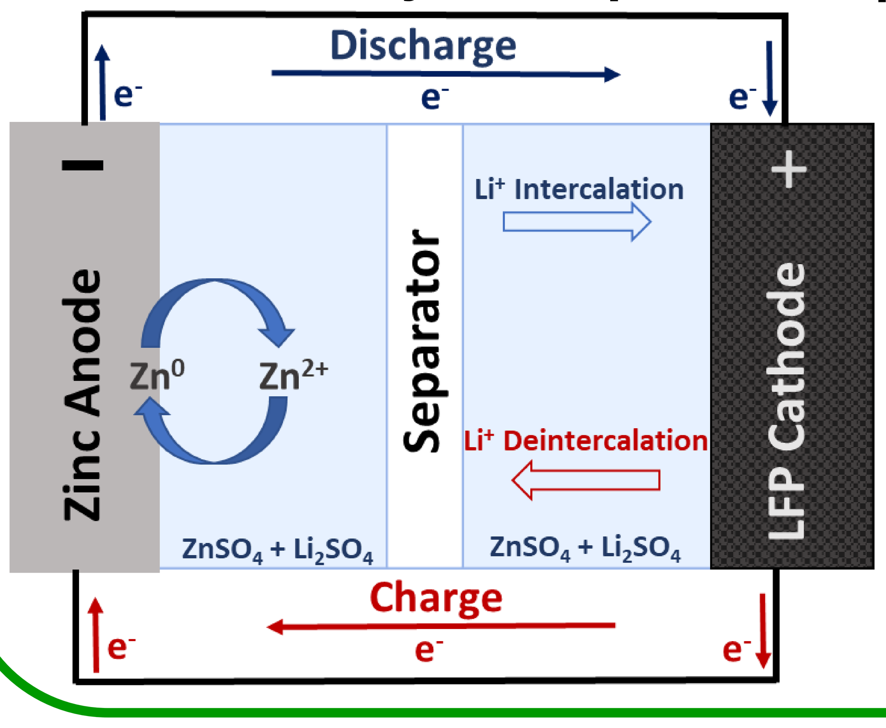
INTRODUCTION

Despite their inherently lower energy density than lithium-ion batteries (LIBs), aqueous zinc-ion batteries (**AZIBs**) have recently attracted interest as rechargeable energy storage devices due to their low cost and high operational and environmental safety. They are composed of metallic zinc as anode, an aqueous zinc salt electrolyte and a cathode capable of (de)intercalating Zn^{2+} ions upon its reduction (oxidation), similarly to LIBs cathodes. In this work we studied a hybrid AZIB in which a dual-ion electrolyte containing both Zn^{2+} and Li^+ ions is used in conjunction with a Li^+ ion intercalation cathode, i.e., $LiFePO_4$ (LFP), one of the most common, reliable and cheap LIBs cathodes. We demonstrate here that ethyl cellulose (EC) can be successfully used as the binder for the cathodic membrane thanks to its insolubility in water, while its solubility in alcohol allows using ethanol as solvent, avoiding toxic solvents. The Zn||LFP cells using EC as a binder deliver a discharge capacity of 153 $mAhg^{-1}$ at 0.1 Ag^{-1} during the 1st cycle, with a capacity retention of 86% after 50 cycles, demonstrating good stability and consistent performance.

WORKING PRINCIPLES

Schematic illustration of the hybrid aqueous Zn||LFP battery



***Charge** / **Discharge** reactions*

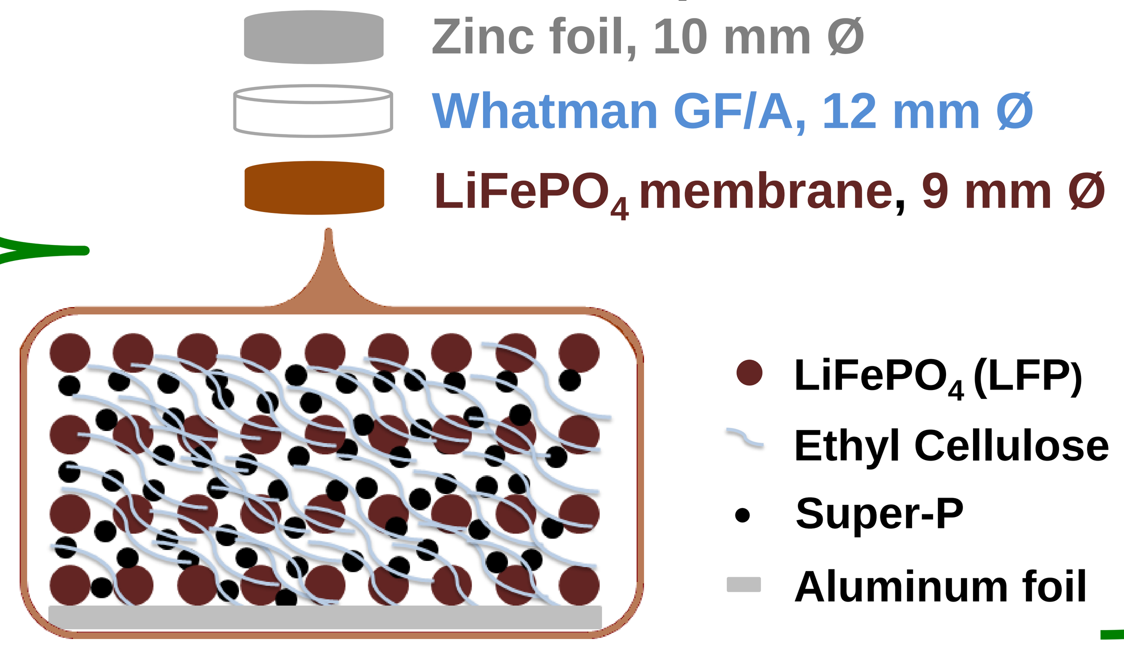
$Zn^{2+} + 2e^- \leftrightarrow Zn$

$2LiFePO_4 \leftrightarrow 2Li^+ + 2FePO_4 + 2e^-$

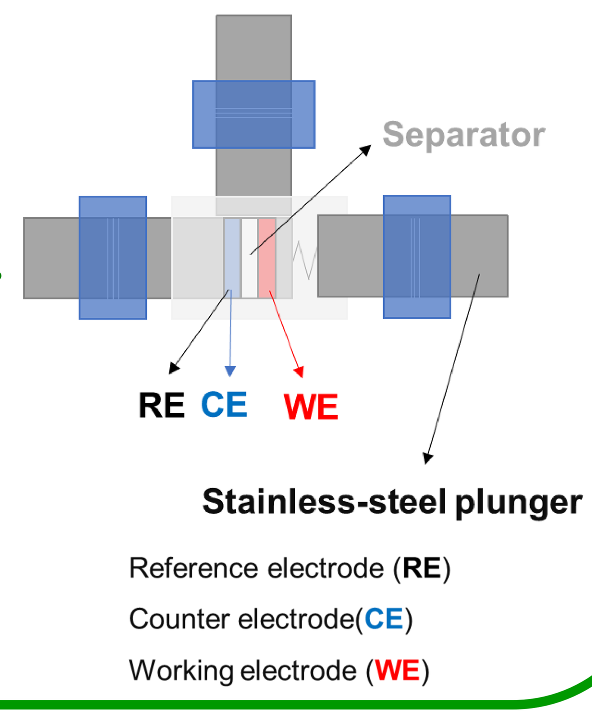
Advantages

- Safe operation
- Abundant, low-cost materials
- Long Lifetime
- Ecofriendly

Cell Core and Positive membrane composition

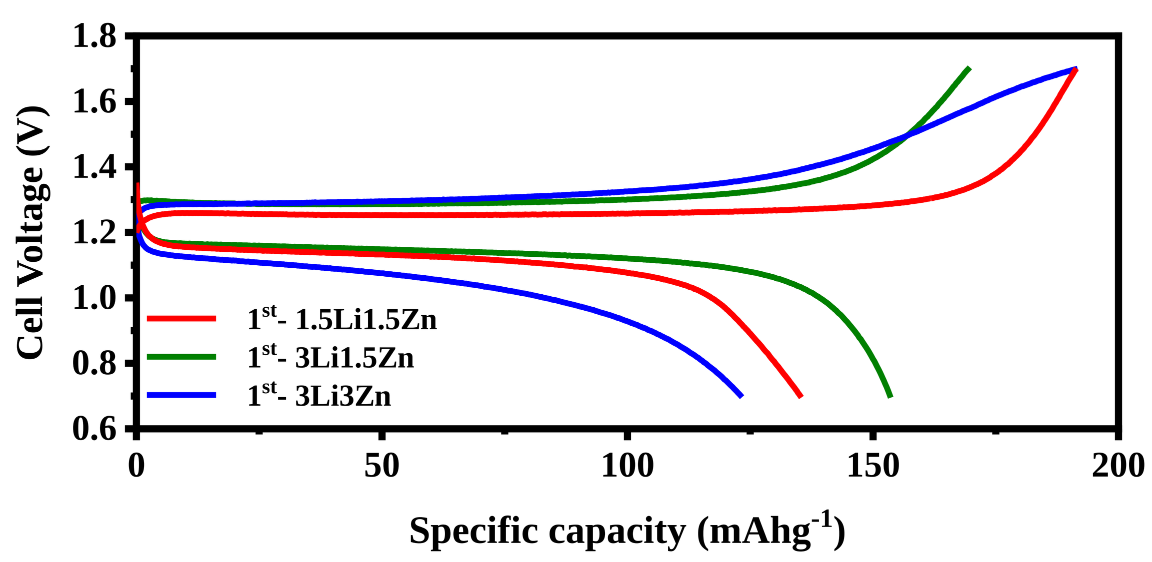


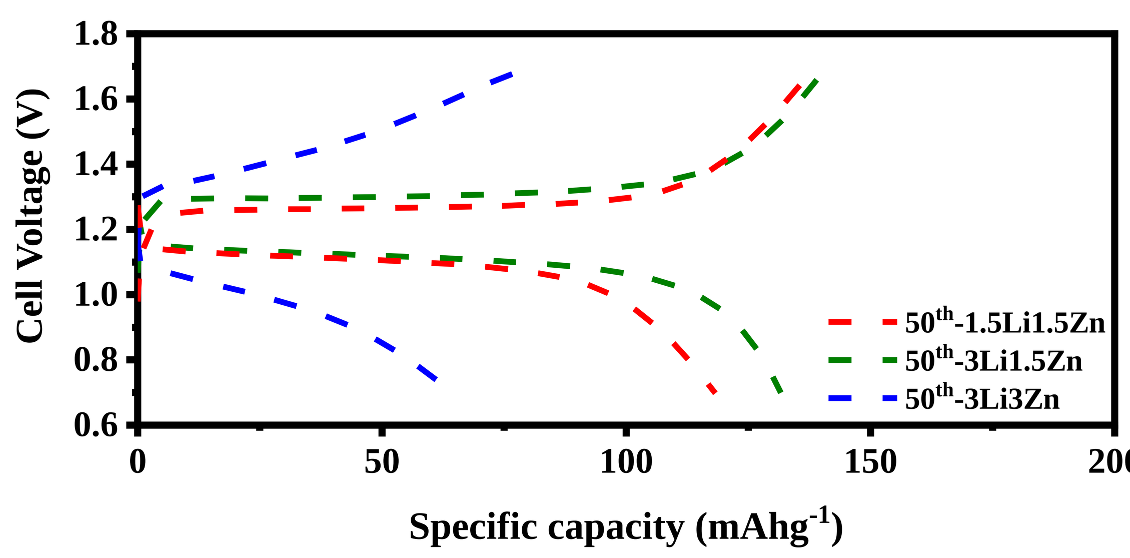
Swagelok T-Cell

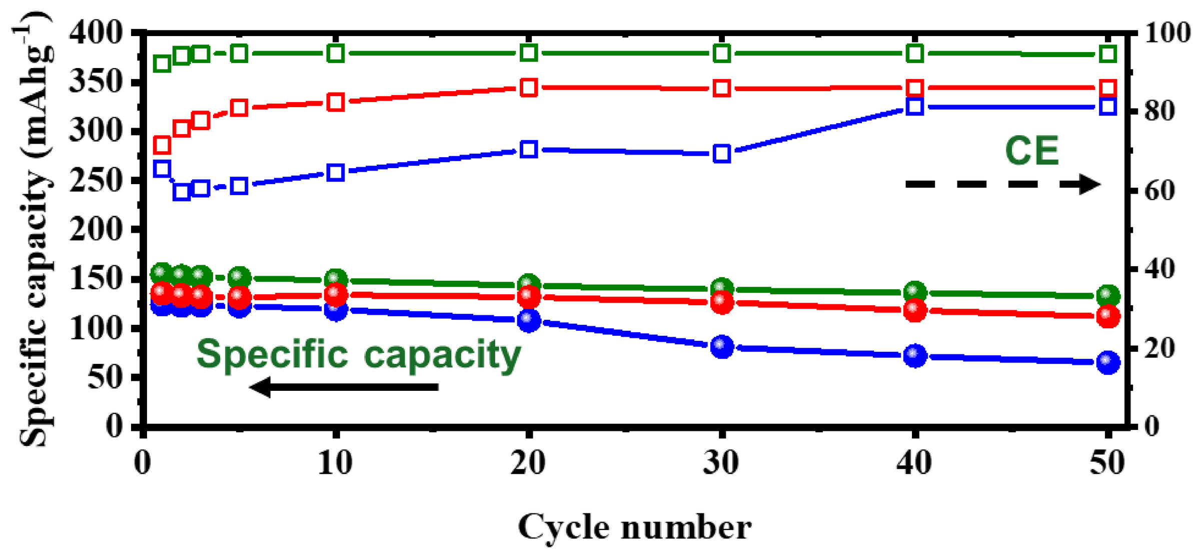


ELECTROCHEMICAL CHARACTERIZATION

GALVANOSTATIC CHARGE/DISCHARGE





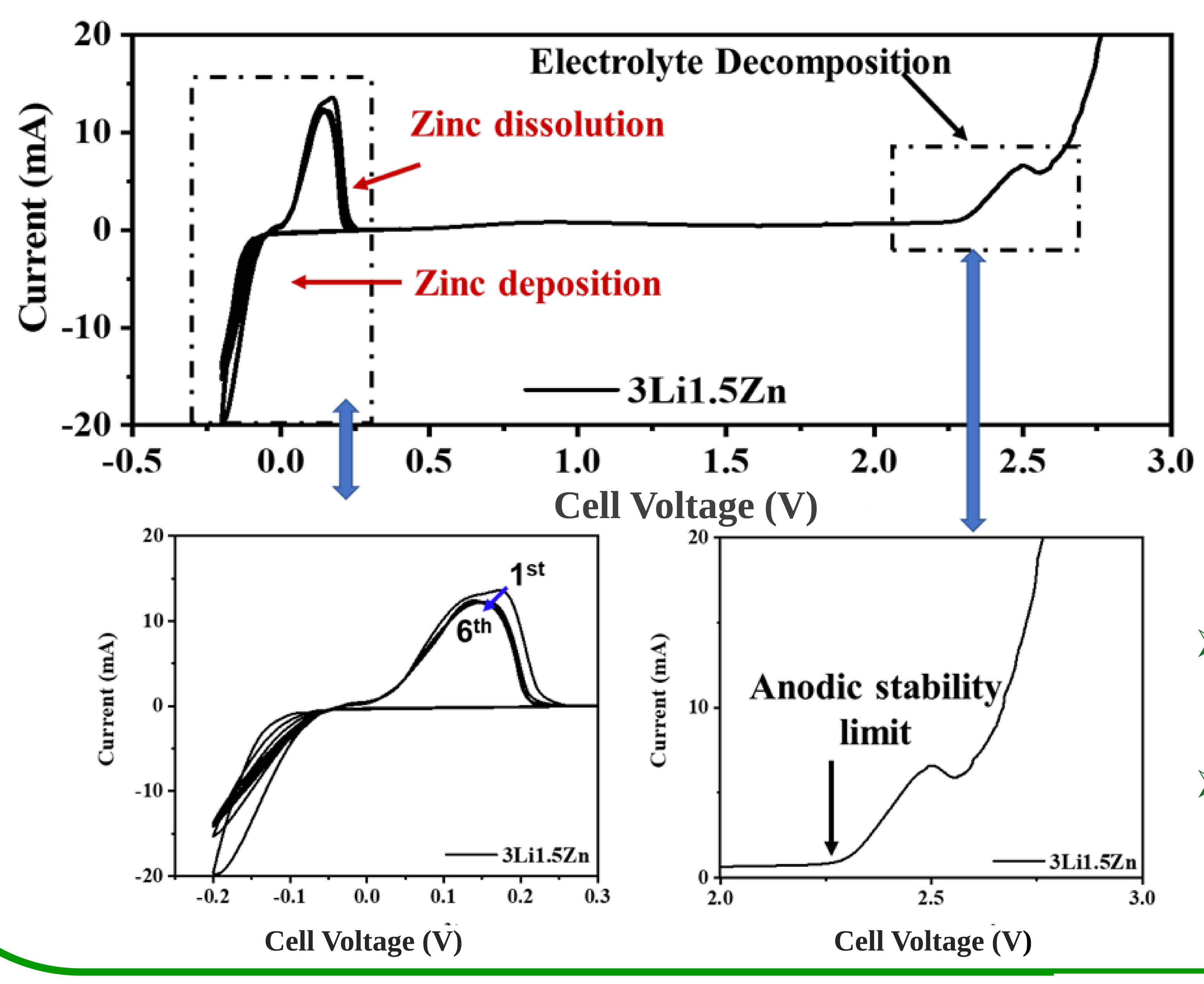


Electrolyte formulation: 1.5 M of Li_2SO_4 and 1.5 M of $ZnSO_4$ (**1.5Li1.5Zn**), 3 M of Li_2SO_4 and 1.5 M of $ZnSO_4$ (**3Li1.5Zn**) and 3 M of Li_2SO_4 and 3 M of $ZnSO_4$ (**3Li3Zn**)

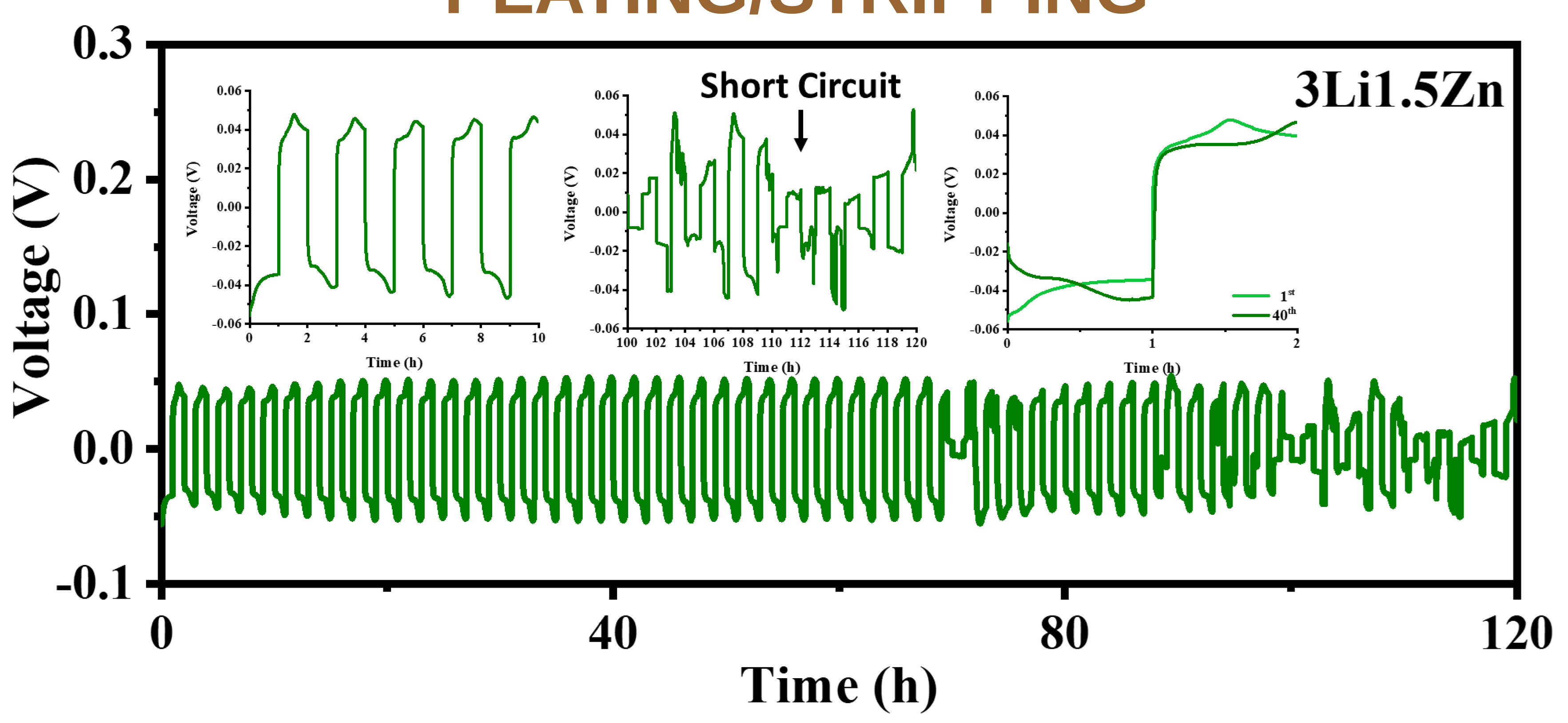
- The Discharge-specific capacity of Zn||LFP cell using 3Li1.5Zn electrolyte was 153 $mAhg^{-1}$, higher respect to 135 and 123 $mAhg^{-1}$ of 1.5Li1.5Zn and 3Li3Zn.
- The Coulombic efficiency (CE) of 3Li1.5Zn (96 %) was higher with respect to 1.5Li1.5Zn (85 %) and 3Li3Zn (80 %)

3Li1.5Zn Best performance

CYCLIC VOLTAMMETRY: Anodic Limit



PLATING/STRIPPING



- The 3Li1.5Zn electrolyte shows an anodic stability up to ca. 2 V vs. Zn^{2+}/Zn^0 which is sufficient for operation with the LFP cathode material.
- The long-term cycling test of Zn||Zn symmetrical cells, using the 3Li1.5Zn as an electrolyte at a current of 0.5 mA lasting 2 h for each cycle (1 h of plating and 1 h of stripping), demonstrating a rather stable response over 70 h.

CONCLUSIONS

- The utilization of EC is demonstrated to be a good alternative to the use of other binders, consenting the use of non-toxic solvents.
- The Zn||LFP cell with the 3Li1.5Zn dual-ion electrolyte shows a good initial discharge capacity of 153 $mAhg^{-1}$ at 0.1 Ag^{-1} with a capacity retention of 86 % after 50 cycles.
- The observed capacity fading in the Zn||LFP cell is primarily due to the oxidation of the aluminum foil in the aqueous medium. Replacement of Al with SS is expected to significantly reduce capacity fading

ACKNOWLEDGEMENTS

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