



Editorial

Stacking solid-state electrolyte and aluminum pellets for anode-free solid-state batteries



Sodium batteries (SBs) have obtained increasing attention due to their low cost and abundant resources [1]. Although SBs are usually viewed to sacrifice energy density, it is still possible for low-cost SBs to achieve a high energy density comparable to corresponding lithium batteries. The anode-free battery does not use anode-active materials, which relies on the electrochemical deposition of alkali metals directly onto the current collector surface. Thus, such kind of battery can achieve higher battery voltage, reduce battery costs, and also increase energy density (Fig. 1a) [2]. Nevertheless, anode-free batteries usually exhibit poor reversibility and cannot achieve sufficient reversible cycling.

Recently, Meng *et al.* from the University of California discovered an electrochemically stable sodium borohydride solid electrolyte ($\text{Na}_4\text{B}_{10}\text{H}_{10}\text{B}_{12}\text{H}_{12}$) that could achieve nearly perfect contact with a pelletized aluminum (Al) current collector (Figs. 1b and c) [3]. The stable and dense solid electrolyte, combined with the dense pelletized Al current collector, constructed a tight interface for suppressing the penetration of Na dendrites to ensure long-life cycling of Na anode-free all-solid-state batteries.

The sodium borohydride solid electrolyte has electrochemical stability, but the battery's Coulombic efficiency is very low when

paired with an Al foil current collector [4]. Na can only be stripped where there is a connection between the incoming Na^+ from the solid-state electrolyte and the electrons from the current collector since there is insufficient solid-solid contact between the solid electrolyte and the Al foil current collector. Incomplete Na stripping leads to poor reversibility. By pressing pelletized Al onto the solid electrolyte separator during cell fabrication, the solid-solid interfacial contact between the solid electrolyte and the current collector could be well improved to form the uniform and intimate contact with the solid electrolyte separator across the entire cell area. When cycled under the same conditions, the initial Coulombic efficiency of the half-cell is greatly improved to 93%. When using Al foil and cycling with a capacity of 4 mAh/cm², the critical current density was found to be 1.2 mA/cm² while the critical current density increased to 6.0 mA/cm² when using the Al pellet current collector (Fig. 2a). The Al particle current collector, combined with the sodium borohydride solid electrolyte, was used to assemble a Na anode-free all-solid-state battery with the low-cost NaCrO_2 and $\text{Na}_{0.625}\text{Y}_{0.25}\text{Zr}_{0.75}\text{Cl}_{4.375}$ as the cathode [5]. The entire battery could work at 40 °C with an initial Coulombic efficiency of 93%. Such a battery could maintain stable cycling over 400 cycles

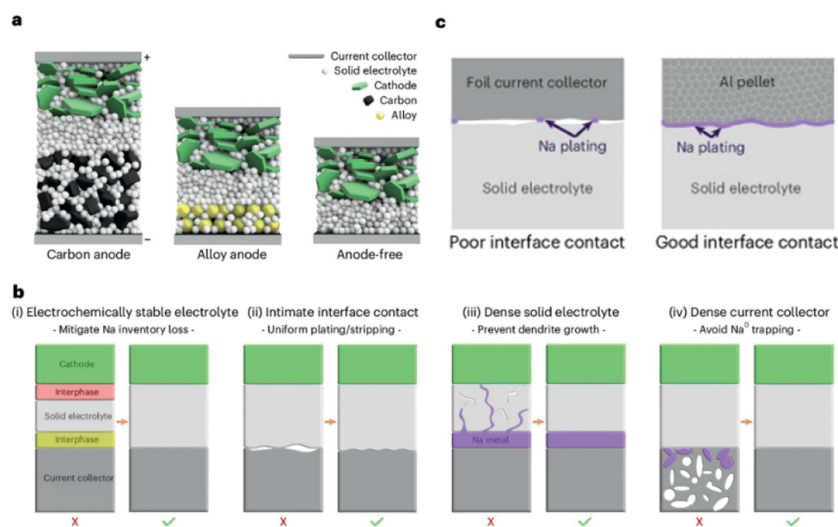


Fig. 1. (a) A schematic diagram of the three different anode configurations for a battery. (b) Al powder forms intimate contact with the solid electrolyte layer. (c) Four requirements for enabling an anode-free all-solid-state battery. Reproduced with permission [3]. Copyright 2024, Springer Nature.

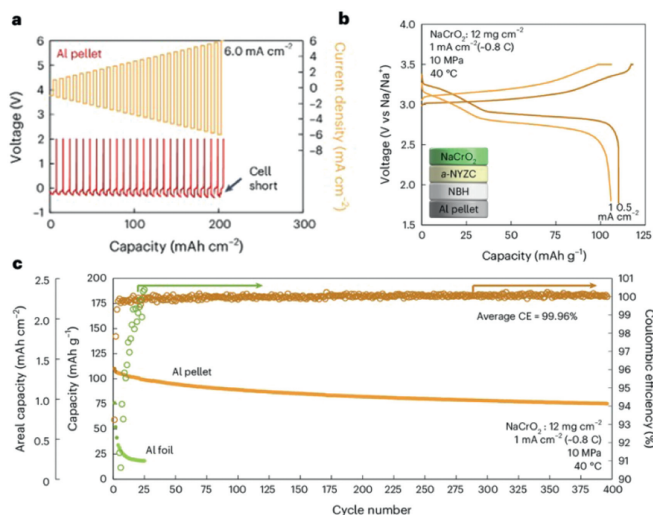


Fig. 2. (a) Critical current density when cycling 6 mAh/cm² capacity with Al pellet current collectors. (b) Voltage curves of a Na anode-free solid-state battery. (c) Cell capacity over 400 cycles. Reproduced with permission [3]. Copyright 2024, Springer Nature.

with a capacity retention of 70%, achieving a high average Coulombic efficiency of 99.96% under these cycling conditions (10 MPa, 40 °C) (Figs. 2b and c). The superior battery performance was attributed to the interfacial stability between solid electrolyte and Al pellets.

In summary, Meng's group achieved stable cycling of anode-free all-solid-state batteries, which can guide the research on other anode-free batteries. The pelletized Al current collector was proven to improve the solid-solid interfacial contact with the borohydride-based solid electrolyte for achieving higher current density cycling. By combining the electrochemically stable and dense solid electrolyte with the application of 10 MPa stack pressure, the practi-

cal application of anode-free all-solid-state batteries could be realized. The work reported by Meng's group will inspire us to explore anode-free all-solid-state batteries.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Jing Guo: Writing – review & editing, Writing – original draft, Visualization, Validation, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Jing Guo

School of Physics and Information Engineering, Shanxi Normal University, Taiyuan 031000, China

E-mail address: guojing@sxnu.edu.cn.

Received 12 September 2024

Revised 11 December 2024

Accepted 13 December 2024

Available online 13 December 2024

References

- [1] H.S. Hirsh, Y. Li, D.H.S. Tan, et al., *Adv. Energy Mater.* 10 (2020) 2001274.
- [2] J. Qian, B.D. Adams, J. Zheng, et al., *Adv. Funct. Mater.* 26 (2016) 7094–7102.
- [3] G. Deysheer, J.A.S. Oh, Y.T. Chen, et al., *Nat. Energy* 9 (2024) 1161–1172.
- [4] G. Deysheer, Y.T. Chen, B. Sayahpour, et al., *ACS Appl. Mater. Interfaces* 14 (2022) 47706–47715.
- [5] P. Ridley, L.H.B. Nguyen, E. Sebti, et al., *Matter* 7 (2024) 485–499.