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Editorial

Constriction-susceptible lithium support for fast cycling of solid-state lithium metal battery

Recently, lithium metal batteries (LMBs) have been intensively explored owing to its high capacity and low potential of Li as the anode [1]. However, challenging problems for LMBs easily occur during cycling process [2], *e.g.*, the growth of Li dendrites, the formation of unstable electrode-electrolyte interphases and side reactions inside the battery [3]. These problems can lead to reduced battery performances and short circuit [4], which are tried to be solved by some strategies including various three-dimensional (3D) supports for Li deposition, electrolyte additives and Li alloys. However, it is still difficult to solve the stability of Li cycling effectively. 3D supports are demonstrated to be effective for tolerating the volume change during Li deposition. Especially, active supports could be used for robustly fusing Li roots. However, the complete lithiation of active support also affects the robust Li deposition. Fortunately, Li's group recently revealed that the constriction-susceptible micron-sized Si could act as the active three-dimensional skeleton to carry thick Li layers for rapid plating and stripping [5]. They found that the behavior of micron-sized Si in organic electrolyte was markedly different from solid-state batteries, where the alloy reaction was limited at the shallow surface of Si particles through a reaction strain-induced diffusion confinement process (Figs. 1a-c). Before discharge, there were pores between the Si particles of the original anode based on Li/graphite-$Li_{5.5}PS_{4.5}Cl_{1.5}$(LPSCl1.5)–LGPS–LPSCl1.5-silicon/graphite (Li/G-SEs-SiG) asymmetric battery, which was further confirmed by focused-ion beam-scanning electron microscopy (FIB–SEM) and energy-dispersive spectrum (EDS) mapping techniques (Figs. 1b(i) and c(i)). There is neither cracks nor significant expansion for micro-sized Si particles (Figs. 1b(ii) and c(ii)) when the voltage is close to 0 V. At −0.2 V, the significant Li deposition could be observed between Si particles (Figs. 1b(iii) and c(iii)). When the capacity was near 5000 mAh/g, the short circuit appeared. In the battery with organic electrolyte, micron Si particles exhibit normal plateau at voltages of 0.5 V and 0.3 V (relative to Li/Li^+) with the lithiation capacity of ∼ 1800 mAh/g (Fig. 1d) with the obvious pulverization (Fig. 1e). Clearly, the Si particles could act as the active 3D Li scaffold to tolerate volume change for fast-cycling solid-state LMBs.

In addition, Li's group achieved superior cycling performance of Li anodes in coin cells and pouch cells in the help of the constriction-susceptible effect of Si when paired with $LiNi_{0.83}Mn_{0.06}Co_{0.11}O_2$ (NMC83) cathode at high cathode loads or high area capacity. They designed the multilayer electrolyte configuration based on Si-based Li anodes for solid state batteries. In Fig. 2a, the capacity retention still maintains up to 80% after 1500 cycles at 6 C (19.5 mA/cm^2) with 110 mAh/g when the cathode load was 22 mg/cm^2. The multi-layer designs in Fig. 2b could provide higher capacity than single-layer designs with the area capacity of ∼2–3 mAh/cm^2. Li-NMC83 solid-state pouch cells could deliver the initial capacity of about 125 mAh/g with the capacity retention of about 80% after 6000 cycles (Figs. 2c and d). In addition, they predicted the materials located in a unique region in the space of lithiation voltage versus constriction susceptibility (*e.g.*, Si, Ag and Mg alloys) could prefer to Li plating for even distribution of local current density at the anode through high-throughput calculations (Fig. 2e). This demonstrates that micro-sized Si is not the individual example for the active support for Li deposition and stripping, which will provide the possibility to use more constriction-susceptible materials for other solid-state metal batteries.

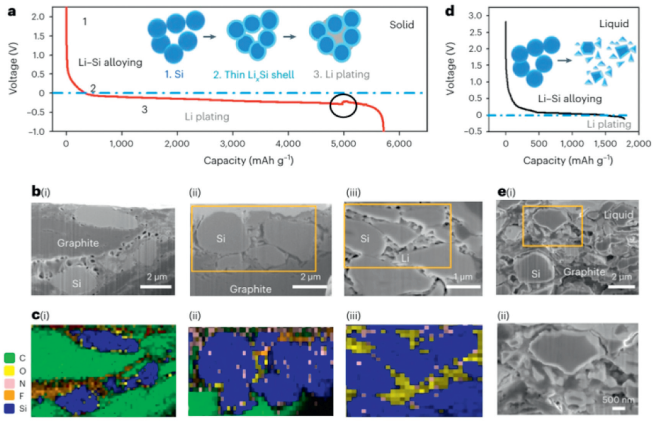

Fig. 1. (a) Discharge profile of Li/G-SEs-SiG battery. (b) FIB–SEM images and (c) SEM-EDS mapping of the corresponding SiG anode at different discharge states. (d) Discharge profile of Li||SiG battery with liquid electrolyte. (e) FIB–SEM image of SiG anode following discharge in Li||SiG battery. Reproduced with permission [5]. Copyright 2024, Springer Nature.

In conclusion, Li's group has realized the fast cycling of solid-state LMBs at high areal capacity in merit of anode materials sensitive to mechanical constraints as the active support for Li deposition and stripping. The work also successfully predicted the dynamic interaction between the surface lithiation and Li plating of suitable anode materials for fast cycling. This work provides the

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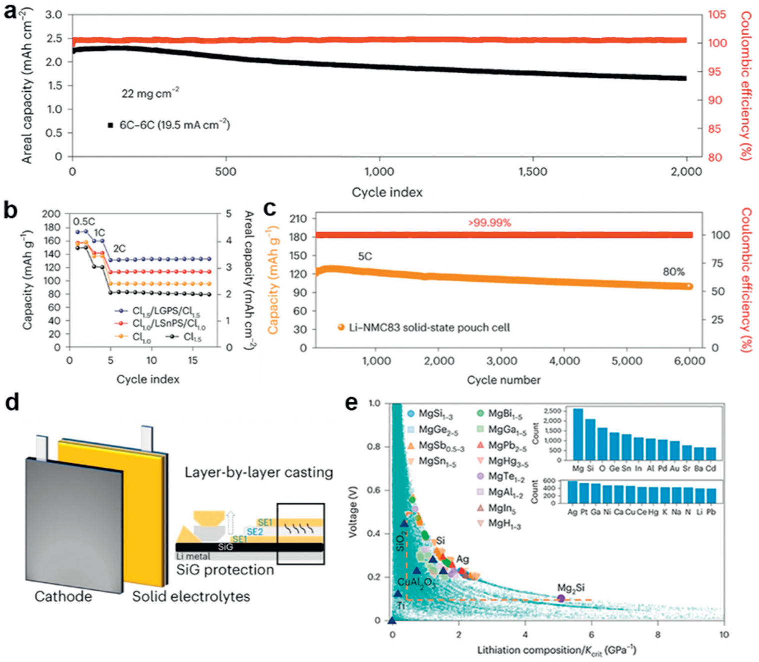

Fig. 2. (a) Cycling performance of Li||NMC83 coin cells with constriction-susceptible anode. (b) Rate performance of Li||NMC83 coin cells with different single- and multi-SE-layer. (c) Cycling performance of Li||NMC83 solid-state pouch cell. (d) Illustrative scheme of pouch cell and layer-by-layer casting. (e) Calculated anodic reaction *versus* Li anode for 59,524 materials. Reproduced with permission [5]. Copyright 2024, Springer Nature.

strength for the commercialization of solid-state LMBs. The meaningful findings are expected to be used for other solid-state metal batteries in future.

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

Dong Sui: Writing – review & editing, Writing – original draft, Supervision, Investigation, Funding acquisition, Conceptualization. **Jiayi Liu:** Visualization, Validation, Investigation.

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