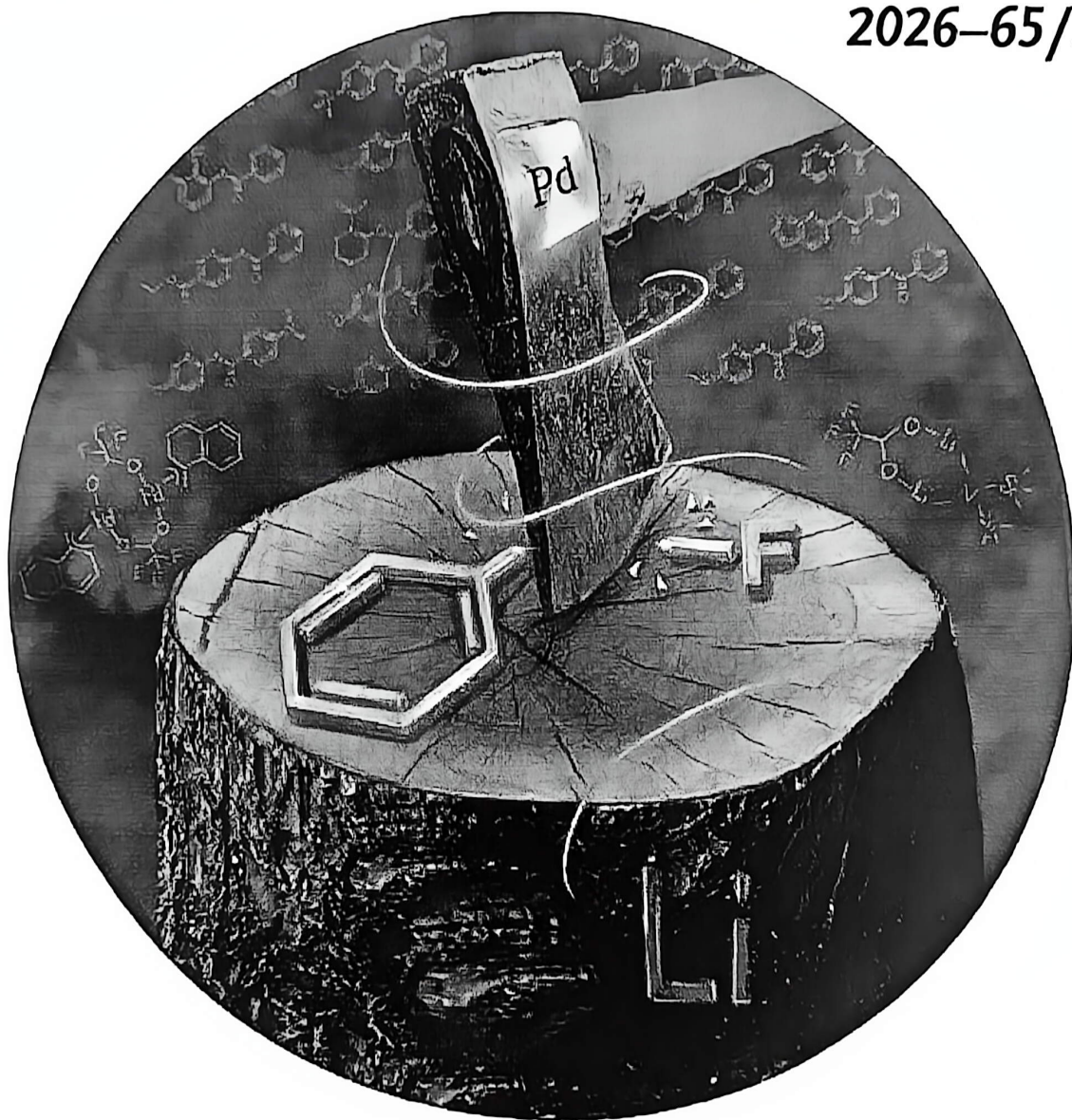


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The cover depicts a wooden trunk engraved with lithium, serving as the foundational base of the scene. An aryl fluoride rests on top, while a palladium axe hovers in a gentle, controlled swing rather than a forceful strike to break the strong bond. Beneath, the lithium cooperatively activates the substrate and thereby empowers the palladium axe, enabling smooth C–F bond activation under mild conditions. More in Communication (e9818208), Lukas J. Gooßen and co-workers.

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RESEARCH ARTICLE

A Sphere–Sheet Hetero–Interlayer With Mechanoadaptivity and Li⁺ Selectivity for High Performance Anode Free Lithium Sulfur Batteries

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ABSTRACT

Anode free lithium sulfur batteries (AFLSBs) can fully exploit the ultra-high energy density of Li–S systems. However, issues such as irreversible lithium loss caused by uneven solid electrolyte interphase (SEI) on anode during the charging/discharging process, along with severe volume changes leading to structural instability, are key factors limiting their lifespan far below practical application. Herein, a sphere–sheet hetero–interlayer (2DLP–PASf) is designed, which adapts to volume changes and exhibits Li⁺ selectivity. The interlayer is fabricated by electrophoretic deposition of –SO₃Li rich elastic ionomer emulsion (PASf) particles and laponite nanosheets (2DLP) onto a copper current collector. The elastic PASf particles support 2DLP to form a dispersed stress–buffering structure, which can accommodate volume changes during the charging/discharging process and suppress dendrite growth. Furthermore, rich –SO₃Li in densely packed PASf shells between 2DLP repel anions electrostatically through the Donnan effect, which facilitates efficient Li⁺ transport and flux homogenization, thereby inhibiting the polysulfide shuttle. Additionally, –C≡N and –CF₃ in PASf cores transform into a stable, Li₃N/LiF-rich secondary SEI on anode, which in turn synergizes with the tough 2DLP–PASf to further stabilize lithium deposition. As a result, the AFLSB assembled with 2DLP–PASf modified copper current collector and Li₂S cathode demonstrated significantly improved comprehensive electrochemical performance.

1 | Introduction

In recent years, the rapid development of electric vehicles, low-altitude aircraft and other electric vehicles have inspired the demand for energy storage devices with higher energy density [1–3]. Anode free lithium sulfur batteries (AFLSBs), composed of a Li₂S cathode and a bare Cu current collector are regarded as a promising configuration to fully exploit the ultrahigh specific energy of lithium–sulfur batteries. However, their insufficient

cycle life remains a major obstacle to further practical advancement [4–6]. Compared with conventional lithium sulfur batteries that primarily focus on issues such as insulation and shuttle effect [7, 8] of sulfur cathode, the anode free configuration presents more severe challenges: the irreversible lithium loss caused by the non-uniform solid electrolyte interface (SEI), [9, 10] together with the drastic volume changes of the lithium deposition during the charging/discharging process, [11, 12] readily leads to poor interfacial contact and rapid capacity fading. Therefore,