Journal of
Materials Chemistry A

PAPER

View Article Online
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Cite this: *J. Mater. Chem. A*, 2025, **13**, 14982

Received 22nd December 2024
Accepted 6th April 2025

DOI: 10.1039/d4ta09090j

rsc.li/materials-a

Introduction of a multifunctional percolated framework into Na metal for highly stable sodium metal batteries†

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Introducing highly sodiophilic skeletons is a highly efficient approach to mitigating the challenges of sodium metal anodes (SMAs). However, the limited functionality of skeletons and poor processability of bare sodium metal further obstruct the practical application of SMAs. Herein, a stable SMA with high processability is realized by introducing a percolated multi-functional NPC/Na_2Se framework throughout metallic Na using simple heating infusion and rolling/folding processes. This percolated framework provides mechanical strength to mitigate cracking and facilitates interconnected pathways for the rapid and even distribution of charges, reducing hotspots and promoting homogeneous Na deposition. Moreover, the Na_2Se in the NPC/Na_2Se framework produces a stable solid electrolyte interphase (SEI) for fast Na^+ diffusion. At the same time, the NPC acts as a 3D matrix to confine the Na and buffer the huge volume change. Consequently, the modified Na@NPC/Na_2Se anode demonstrates excellent performance in both low-cost carbonate (1200 h at 1.0 mA cm^{-2}) and ether-based (8000 h at 5.0 mA cm^{-2}) electrolytes with high coulombic efficiency (99.89% after 200 h of plating/stripping). More remarkably, the Na@NPC/Na_2Se||NVP full cell manifests unprecedented cycling (85 mA h g^{-1} at 20C after 4000 cycles) and excellent rate capability (~105 mA h g^{-1} at 50C). This electrode featuring a multi-functional framework creates new opportunities for the development of SMAs and can be extended to anode free batteries.

1 Introduction

The global shift towards electric vehicles and grid-scale energy storage, coupled with the rapid advancement of portable electronics, has intensified the demand for cost-effective, safe, and sustainable electrochemical battery systems. This urgency is further driven by the scarcity and high cost of lithium resources, necessitating the development of alternative energy storage solutions that can be deployed on a large scale.[1,2] As a competitive contender to lithium-based batteries, sodium (Na)-based battery systems, including Na-ion (SIBs), Na–air (Na–O_2), and Na–sulfur (Na–S) batteries, have been explored as promising options for power grids and electric vehicles.[3–6] This is attributed to the inherent advantages of the Na metal anode (SMA), including its low electrode potential (−2.71 V *vs.* the standard hydrogen electrode), high theoretical capacity (1160 mA h g^{-1}), and cost-effectiveness ($4 per kg).[7,8] However, the high reactivity of Na metal leads to undesirable parasitic reactions, causing uncontrolled dendritic growth and an unstable solid electrolyte interphase (SEI).[9] As a result, this leads to low coulombic efficiency (CE), a reduced Na inventory, rapid electrolyte consumption, and ultimately, short lifespans and potential safety hazards, hindering the viability of SMBs.[10]

Tremendous endeavors have been made to overcome these challenges, with surface and structural modifications of Na metal electrodes emerging as a widely recognized and effective strategy.[11,12] For example, surface protection layers on Na metal, including Na alloys (NaBi,[13] NaSn[14]), Na_3P,[15] Na halides (NaF,[16] NaBr,[17] and NaI[18]), Na chalcogenides (Na_2Se, Na_2S, and Na_2Te)[19–21] and organic protective layers, have been designed and established superb reversibility in SMBs.[22] However, these

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† Electronic supplementary information (ESI) available. See DOI: https://doi.org/10.1039/d4ta09090j

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