Batteries & Supercaps
Editorial
doi.org/10.1002/batt.202200467
Chemistry Europe
European Chemical Societies Publishing
www.batteries-supercaps.org

Special Collection

Lithium-Sulfur Batteries: Current Achievements and Further Development

Stefan Kaskel,*[a, b] Jia-Qi Huang,*[c] and Hikari Sakaebe*[d]

In this Editorial, Guest Editors Stefan Kaskel, Jia-Qi Huang, and Hikari Sakaebe introduce the Special Collection of *Batteries & Supercaps* on Lithium–Sulfur batteries. They discuss the challenges that lithium-ion batteries currently face and how they can be solved using lithium-sulfur batteries using various interesting approaches from scientists around the world.

The transition of our society from petroleum-based energy infrastructure to one that is sustainable and based on renewable energy necessitates improved and efficient energy storage technologies. Lithium-ion batteries (LIBs) are predominant in the current market due to their high gravimetric and volumetric energy density since their first commercialization in 1991.[1] However, the maximum energy density that LIBs can theoretically achieve is still lower than the requirement of future energy-intensive technologies used in grid-scale energy storage and transportation applications. Therefore, the development of new battery systems beyond LIBs is imperative, affordable, and environmentally responsible.

One of the most promising battery systems that can fulfill the requirement is the lithium-sulfur (Li–S) battery. The theoretical specific energy of Li–S batteries is 2600 $Wh\,kg^{-1}$, which is about five times higher than the current standard (430–570 $Wh\,kg^{-1}$) for LIBs such as LiC_6–$LiCoO_2$.[2] Besides, sulfur is abundant, affordable, and non-toxic. These properties make Li–S systems an attractive solution for the achievement of high-energy-density battery technologies. Despite the above attractive advantages, the practical application of Li–S batteries is hampered by major scientific hurdles,[3] such as the low conductivity of the sulfur element and discharge product lithium sulfide, the shuttle effect of lithium polysulfides in the electrolyte, sluggish electrochemical reaction kinetics of the sulfur cathode, and the huge volume changes for both electrodes upon lithiation and delithiation, which results in a significant loss of active materials, a short cycle life, and irreversible capacity fading. Under the intervention of concentrated lithium polysulfides on the interfacial reaction of Li anode, the repeated rupture and regeneration of the solid–electrolyte interface and uncontrolled growth of lithium dendrites increase the risk of battery failure and safety problems.

Technological breakthroughs in Li–S batteries are accomplished through the development of nanotechnology. Nazar and colleagues opened the era of rapid research and development of Li–S batteries by melting mesoporous carbon and elemental sulfur in 2009.[4] This Special Collection brings together the latest research progress on high-performance Li–S batteries, including 2 reviews and 14 research papers, involving electrode structure design, separator modification, electrolyte regulation, as well as advanced characterization technologies for Li–S battery systems.

In the past decades, the mechanistic understanding and performance improvement of Li–S batteries have greatly progressed. In this collection, Weret et al. discuss the working principles and challenges of Li–S batteries and review the strategies aiming to overcome the Li–S issues regarding the electrode design and modification, the development of novel electrolyte, the separator modification/functional interlayer insertion, and the protection of the lithium anode. Despite significant progress achieved in battery performance and chemistry understanding, the commercialization of Li–S batteries still needs to bridge the gap between laboratory results and practical conditions, which highly relies on the improvement of electrolyte systems. Donato et al. review the state-of-the-art electrolytes, highlight the different strategies undertaken with liquid and solid electrolytes, and provide future research directions for Li–S batteries from laboratory to practical applications.

[a] *Prof. S. Kaskel*
Department of Inorganic Chemistry I
Technische Universität Dresden
Bergstraße 66, 01069 Dresden (Germany)
E-mail: Stefan.Kaskel@tu-dresden.de
[b] *Prof. S. Kaskel*
Fraunhofer IWS,
Winterbergstr. 27, 01277 Dresden (Germany)
[c] *Prof. J.-Q. Huang*
Advanced Research Institute of Multidisciplinary Science
Beijing Institute of Technology
Beijing 100081 (P. R. China)
E-mail: jqhuang@bit.edu.cn
[d] *Prof. H. Sakaebe*
Research Institute of Electrochemical Energy (RIECEN)
Department of Energy and Environment
National Institute of Advanced Industrial Science and Technology (AIST)
1-8-31 Midorigaoka, Ikeda, Osaka 563-8577 (Japan)
E-mail: hikari.sakaebe@aist.go.jp

An invited contribution to a Special Collection dedicated to Lithium-Sulfur Batteries.

The design of carbon-based sulfur host materials constitutes an effective approach to relieve the issues raised by low electronic conductivity and the polysulfide shuttle effect. Jiménez-Martín et al. synthesized two tailored nanomaterials based on conductive reduced graphene oxide and mixed them with different porous carbon structures. The relationship between the properties of different carbon materials and battery performance was elaborated. Won et al. further presented nanoconductive carbon additives with minimized interfacial energy for lithium polysulfides, which controls the shuttle effect by confining the dissolved lithium polysulfide within the cathodes.

Except for the nanocarbon materials, metal composites and metal nanoparticles are also mainstream host materials for sulfur cathodes due to additional adsorption and catalytic sites for lithium polysulfides. Mei et al. developed a facile synthesis of carbon-coated Ti_4O_7 hollow nanoparticles to function as sulfur host materials and verified the electrochemical performance on the pouch-cell scale. Besides, Cheng et al. applied electroless nickel plating to achieve a nickel-sulfur nanocomposite to improve the conductivity of active solid-state materials and inhibit the diffusion of lithium polysulfides. On the other hand, homogeneous redox mediators are effective promotors to propel the redox kinetics of the cathode. Peng et al. proposed a strategy of mixed redox mediators for the full-range mediation of the sulfur redox reactions in working Li–S batteries.

Separator modification presents another way to synchronously suppress the shuttle effect of soluble polysulfides, electrocatalyze the sulfur redox reactions, and homogenize lithium ion transmission. Yue et al. designed a ferromagnetic Fe_3O_4 nanoparticle-modified separator for Li–S batteries to enhance energy storage performance. Besides, Liu et al. directly coated oxidized carbon nanotubes decorated with nickel-iron oxide on the commercial separator to verify the performance advantages. Except for the cathode-side modification of separators, Xiao et al. synthesized $La_2Zr_2O_{7-x}$ nanotubes with surface oxygen vacancies to serve as Janus coating layers, which is expected to tackle the shuttle effect and lithium dendrite issues due to efficient adsorption and accelerated lithium-ion transport.

Sparingly solvating electrolytes have been reported to reduce the electrolyte content in practical Li–S batteries. Li et al. investigated the performance of Li–S cells using cathodes with a relatively high sulfur loading under lean electrolyte conditions of sparingly solvating electrolyte. Furthermore, Kensy et al. systematically studied the structure-activity relationship be-

Stefan Kaskel studied chemistry and received his Ph.D. degree in 1997 from the Eberhard Karls University, Tübingen (Germany). As a Alexander von Humboldt fellow he worked with J. Corbett at Ames Laboratory, USA (1998–2000) on intermetallic compounds. He was a group leader at the Max-Planck-Institut für Kohlenforschung in Mülheim a.d. Ruhr (2000–2004) and after his habilitation at the Ruhr University (Bochum, 2004) in the area of heterogeneous catalysis, he became full professor for Inorganic Chemistry at Technical University Dresden. Since 2008 he is also technology field leader at the Fraunhofer IWS, Dresden. His research interests are focused on porous and nanostructured materials (synthesis, structure, function) for applications in energy storage, batteries, catalysis, and separation technologies. He was recognized as a Highly Cited Researcher multiple times since 2016.

Jia-Qi Huang studied chemical engineering and received his B.E. (2007) and Ph.D. (2012) degrees from Tsinghua University (China). After working as assistant and associate research fellow at Tsinghua University, he joined the Advanced Research Institute of Multidisciplinary Science (ARIMS) at Beijing Institute of Technology (China) as associate professor in 2016. In 2019, he was promoted to full professor at Beijing Institute of Technology. His research interests focus on advanced high-energy-density batteries such as lithium-sulfur batteries and lithium-metal batteries, especially on the chemical phenomena in the formation and evolution of electrode interface. He was recognized as a Highly Cited Researcher by Clarivate since 2018 in materials science and chemistry.

Hikari Sakaebe is a Prime Senior Researcher at Research Institute of Electrochemical Energy, Department of Energy and Environment, National Institute of Advanced Industrial Science and Technology (AIST, Japan). She received her Ph.D. (1996) degree in Industrial Chemistry from the Graduate School of Engineering, Kyoto University (Japan). Her main research interests focus on the development of materials for high-energy batteries including metal polysulfides for lithium-sulfur batteries, metal fluoride conversion electrodes, metallic Li anode, among others. She also works as a professor at the Institute for Materials Chemistry and Engineering, Kyushu University (Japan) since June 2021.

Chemistry Europe
European Chemical Societies Publishing

tween the carbon host porosity and the conversion mechanism of the sulfur cathode in sparingly solvating electrolyte.

Instead of the widely used ether-based electrolyte, carbonates provide another promising choice as electrolyte for Li–S batteries when combined with a sulfur-ultra-microporous composite cathode. Para et al. explored the electrochemical behavior of ultra-microporous carbon with infiltrated sulfur in a carbonate-based electrolyte and demonstrated the kinetic and chemical advantages of the cathode electrolyte interface (CEI) formed on the surface of the cathode. Vanadium polysulfide (VS_4) is also a potential cathode material for lithium-sulfur batteries matched with a carbonate-based electrolyte. Yoshii et al. investigated the electrode behavior of a low-crystalline VS_4 cathode with different electrolytes and analyzed the effect of fluoroethylene carbonate additives on the battery cycling performance.

Advanced electrode morphology and structure characterizations are crucial for exploring the performance boundaries and failure causes of Li–S batteries under different operating conditions. Chien et al. demonstrated the impact of compression on battery performance through X-ray computed tomography and kinetic analysis based on operando X-ray diffraction. In addition, Shateri et al. explored the effect of temperature on the cycling life of Li–S cells using system identification and X-ray tomography.

This Special Collection groups together the latest research on the development of Li–S battery technology. To thoroughly understand the Li–S chemistries and promote the practical application of Li–S batteries, multidisciplinary collaborative research on interface chemistry, materials science, and surface engineering is highly required. Moreover, advanced characterization techniques and high-throughput theoretical calculations are expected to help understand the mechanism and practical progress of Li–S batteries. We would like to particularly thank all the contributing authors for their great work and the reviewers for their support in the preparation of this Special Collection. We sincerely hope that it can promote the development of Li–S battery technology and bring more interest and inspiration to the readers of *Batteries & Supercaps*.

Keywords: electrolytes · lithium-sulfur batteries · metal composites · structural characterization · sulfur host materials

[1] B. Scrosati, *J. Solid State Electrochem.* **2011**, *15*, 1623–1630.
[2] Z. X. Chen, M. Zhao, L. P. Hou, X. Q. Zhang, B. Q. Li, J. Q. Huang, *Adv. Mater.* **2022**, *34*, 2201555.
[3] S. Dörfler, H. Althues, P. Härtel, T. Abendroth, B. Schumm, S. Kaskel, *Joule* **2020**, *4*, 539–554.
[4] X. L. Ji, K. T. Lee, L. F. Nazar, *Nat. Mater.* **2009**, *8*, 500–506.

Manuscript received: October 26, 2022
Version of record online: October 31, 2022