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Lithium–Sulfur Batteries: Co-Existence of Challenges and Opportunities

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Lithium–sulfur (Li–S) batteries deliver a high theoretical energy density of 2600 Wh kg^{-1}, and hold great promise to serve as a next-generation high-energy-density battery system. Great progress has been achieved in the last decade in dealing with the intrinsic problems of Li–S batteries, while numerous challenges still exist towards the practical applications. Following the successfully concluded 1st symposium held in Tsinghua University, Beijing, China in June 12–13, 2016, the 2nd Symposium on Frontier of Lithium–Sulfur Batteries was held in June 17–18, 2017 at Institute of Metal Research, Chinese Academy of Sciences, Shenyang, China. The total number of attendees was over 400. We were able to bring together some of the most eminent scientists and young researchers to share their knowledge and experiences, which is of great importance to boost the growth of the community and the development of Li–S batteries as well. With the help of Dr. Yan Li, a former Deputy Editor of *Advanced Functional Materials*, we proposed this special issue based on selected presentations at the symposium and the invited submission from established research groups in this field for *Advanced Functional Materials*. This issue consists of 16 Feature Articles and Full Papers, covering the important aspects in the research of Li–S batteries from both the fundamental research point of view or the practical application perspective.

Regarding the fundamental understanding of the complex Li–S battery system, the rapid progress in mechanism understanding for Li–S batteries relies on the introduction of various advanced characterization techniques. Yu, Li, and co-workers summarized the applications of advanced characterization techniques in Li–S systems, and their important roles in promoting the mechanism understanding. Especially, the importance of multiple characterization technique combinations in ex-situ and in-situ Li–S system investigation is emphasized (article number 1707543). The electrochemical reactions of Li–S batteries involve complex redox processes. Rationally designing efficient mediators, as well as incorporating them into a working cell, emerges to be a promising method to construct high-energy-density Li–S batteries. Huang and co-workers review the mechanistic understanding of redox kinetics in Li–S reactions and recent advances in both heterogeneous and homogeneous mediator design (article number 1707536). The complex chemical interactions between polar materials and polysulfides are reviewed by Guo and co-workers (article number 1707520), especially the chemical polar-polar interaction, and how the intrinsic properties of polar materials affect the electrochemical performance of Li–S batteries.

Novel functional materials are crucial to further propel the cathode performance in Li–S batteries. Lu, Amine, and co-workers propose an advanced graphene nanocage structure with the capability of hosting cyclo-S_8 in the inner cavity and smaller sulfur molecules in the graphene shell of the cages (article number 1706443). Sun, Li, and co-workers reports a multifunctional carbon hybrid with metal–organic frameworks (MOFs)-derived nitrogen-doped porous carbon anchored on graphene serving as a sulfur host (article number 1707592). The high surface area and nitrogen-doping of the carbon nanoparticles enable effective polysulfide immobilization.

Xia and co-workers reported a novel porous carbon fibers/vanadium nitride (VN) arrays composite scaffold for the storage of sulfur (article number 1706391). The VN nanobelt arrays demonstrate a strong ability for chemically anchoring the polysulfides to retard the shuttle effect. Guo and co-workers report a new phase engineering strategy for making MXene/1T-2H MoS_2-C nanohybrids with plenty positively charged S-vacancy defects to boost the performance of Li–S batteries in terms of capacity, rate ability, and stability (article number 1707578). As one type of promising carbon materials to confine polysulfides within the cathode, the research progress on porous organic polymers (POPs) and POPs-derived carbon materials in Li–S batteries is summarized by Li, Wang, and co-workers in article number 1707597. POPs are promising to serve as sulfur host materials, interlayers, and separators in Li–S batteries. Lv, Zhang, Yang, and co-workers summarize that progress in tailored functional carbon to

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prevent the shuttling of polysulfides (article number 1800508). Carbon materials are classified into four specific functions, namely confining, trapping, blocking, and breaking up. The design principles of advanced carbons with synergistic functions is discussed.

New configurations of Li–S cells bring new opportunities. Park, Yang, and co-workers outline the recent progress in Li–S batteries based on novel configurations with functional interlayers/separators, and discuss the role of the configuration in Li–S batteries (article number 1707411). New configurations with lithium sulfide (Li_2S) instead of sulfur as cathode are of potential to work with anode materials other than lithium metal to eliminate serious safety concerns for conventional Li–S batteries. Wang and co-workers summarize the recent advances of Li–S batteries based on a Li_2S cathode with features of improved safety, high Coulombic efficiency, and high energy density (article number 1800154).

Chen and co-workers summarize the main efforts in the electrolytes of Li-S batteries, including liquid, solid state, and hybrid electrolyte systems. The development of functional electrolytes to alleviate the problems of Li-S batteries are highlighted (article number 1800919). The replacement of the organic electrolyte would bring a different chemistry to Li–S batteries. Suk, Park, and co-workers summarize the progress on aqueous electrolyte Li–S batteries, which is completely different from that in organic electrolytes, providing new routes to resolve the existing problems in conventional organic electrolyte systems (article number 1707593). The progress on applying various classes of solid-state electrolytes (SSEs) including gel, solid-state polymer, ceramic, and composite electrolytes to solve the issues of Li–S batteries is summarized by He and co-workers (article number 1707570). Guo and co-workers also review the recent achievements in all-solid-state Li–S batteries based on inorganic SSEs, especially focusing on the complexity of interfaces, including metallic lithium|SSEs, SSEs|SSEs, and composite sulfur cathode|SSEs. The potential approaches to deal with these interfacial issues are proposed (article number 1707533).

Towards the practical applications, reducing the amount of electrolyte is critical in achieving high energy density for Li–S batteries, while it is also challenging to deal with the complex redox process in lean electrolyte conditions. Shao, Liu, and co-workers report that a NH_4TFSI additive in the electrolyte solution greatly alleviates the passivation issue in Li–S batteries by propelling the dissociation of Li_2S for improved performance under lean electrolyte conditions (article number 1707234).

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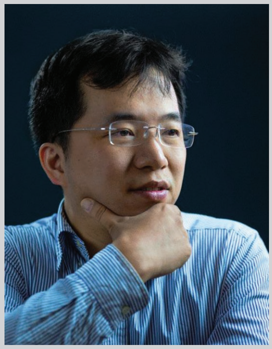

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