

# The next generation of batteries

## Ab-initio Simulations for the Lithium-Sulfur Batteries and Sodium-Sulfur Batteries

**M.K. Afshar, E. Nadimi, I. Frank**, Institute of Physical Chemistry and Electrochemistry, Leibniz Universität Hannover

### In Short

- Exploring dual-atom catalysts (DACs) for Li-S batteries
- Investigating single-atom catalysts for Na-S batteries
- Based on first-principles calculations

The global transition toward renewable energy technologies has intensified the demand for high-performance and cost-effective energy storage systems. Lithium-sulfur (Li-S) and sodium-sulfur (Na-S) batteries are promising candidates due to their high theoretical capacities (1675 mAh/g for both Li-S and Na-S), low material cost, and environmental sustainability. However, practical implementation is challenged by the dissolution and migration of polysulfides (shuttle effect), sluggish redox kinetics, poor electrical conductivity of sulfur and its reduction products, and significant volumetric expansion during cycling. These limitations result in capacity fading, reduced cycle life, and safety concerns [1,2]. As shown in Fig1, the sodium-sulfur (Na-S) battery operates through a multi-step redox reaction mechanism similar to lithium-sulfur (Li-S) systems. During discharge, sodium ions ( $\text{Na}^+$ ) produced at the sodium anode migrate to the sulfur cathode, forming a sequence of soluble polysulfide intermediates ( $\text{Na}_2\text{S}_8$ ,  $\text{Na}_2\text{S}_6$ ,  $\text{Na}_2\text{S}_4$ ) followed by insoluble species ( $\text{Na}_2\text{S}_3$ ,  $\text{Na}_2\text{S}_2$ ) and finally  $\text{Na}_2\text{S}$  [2].

During the previous project period, our team employed first-principles computational methods, including density functional theory (DFT) and ab initio

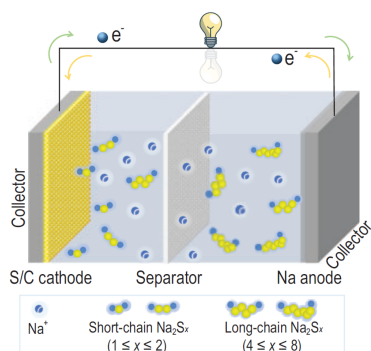


Figure 1: Schematic of the Na-S Battery[2].

molecular dynamics (AIMD), to investigate single-atom catalysts (SACs) and defect-engineered cathode hosts for Li-S batteries. Our high-throughput screening of SACs at divacancy and Stone-Wales defects revealed that Fe- and V-based SACs offer optimal adsorption energies and catalytic activity, effectively facilitating lithium polysulfide conversion and suppressing the shuttle effect. Furthermore, single-atom Chromium embedded in nitrogen-doped graphene ( $\text{Cr-N}_4$ ) demonstrated enhanced polysulfide binding, improved cycling stability, and a high initial capacity of 1136.4 mAh/g at 0.2 C [3]. AIMD simulations revealed that  $\text{Fe-N}_4$  and  $\text{Ti-N}_4$  hosts catalyze sulfur dissociation, promote  $\text{Li}^+$  accessibility, and prevent polysulfide migration more effectively than pristine graphene. In addition, PV-doped  $\text{MoS}_2$  effectively anchors Li-S clusters, further reducing shuttle effects and improving structural stability during lithiation. These studies generated multiple publications and a manuscript currently under review, providing a robust computational foundation for future investigations.

In this follow-up project, we aim to extend our research in two key directions. First, we will explore dual-atom catalysts (DACs) for Li-S batteries, as shown in fig2, to harness possible synergistic effects between neighboring metal centers. DACs are expected to enhance adsorption and conversion of both long- and short-chain polysulfides, increase active site density, and mitigate the shuttle effect more effectively than single-atom systems [4–6]. We will systematically screen homometallic and heterometallic DAC configurations via high-throughput DFT calculations, identify the most promising candidates, and simulate their lithiation behavior using AIMD to understand their catalytic mechanisms.

Second, we will investigate single-atom catalysts for Na-S batteries. Na-S batteries operate through similar multi-step redox reactions as Li-S batteries, facing comparable challenges such as polysulfide dissolution, sluggish kinetics, and electrode insta-

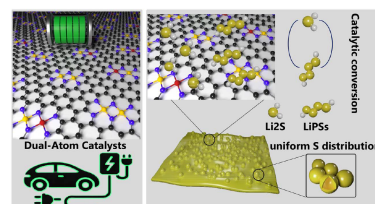


Figure 2: Schematic illustration of dual-atom catalysts (DACs) for Li-S batteries lit4.

bility. By evaluating a broad range of 3d and 4d transition metal SACs, we aim to identify efficient catalysts that immobilize sodium polysulfides, accelerate conversion reactions, and enhance cycle stability. AIMD simulations will complement DFT calculations to capture realistic lithiation processes and dynamic interactions at the atomic level.

Overall, this project combines high-throughput screening, adsorption and conversion energy analysis, and AIMD simulations to systematically explore optimal catalyst designs for Li-S and Na-S batteries. The results will provide critical atomic-level insights into polysulfide interactions and electrochemical mechanisms, guiding the rational design of high-performance, durable battery materials. The integration of DACs and SACs is expected to significantly improve energy density, cycling stability, and reaction kinetics, advancing the development of next-generation energy storage technologies.

### WWW

<http://www.zib.de>

### More Information

- [1] K. Mahankali, S. Nagarajan, N. K. Thangavel, S. Rajendran, M. Yeddala, L. M. R. Arava, *J. Catalysts* **10**, 1137 (2020). doi: 10.3390/catal10101137
- [2] L. Wang, T. Wang, L. Peng, Y. Wang, M. Zhang, J. Zhou, M. Chen, J. Cao, H. Fei, X. Duan, others, *J. National Science Review* **9**, nwab050 (2022). doi:10.1093/nsr/nwab050
- [3] F. Bettels, D. Rashidi, Z. Lin, L. Schenk, T. Li, H. Wu, I. Frank, E. Nadimi, Y. Liu, C. Zhang, F. Ding, L. Zhang, *J. Batteries & Supercaps* , 2500200 (2025). doi:10.1002/batt.202500200
- [4] S. Maiti, M. T. Curnan, K.-W. Kim, S. Subhalaxmi, J. Hur, R. Narayan, K. Maiti, J. K. Kim, *Journal of Materials Chemistry A* **13**, 31829-31868 (2025). doi:10.1039/d5ta03508b
- [5] Y. Zhang, Y. Qiu, L. Fan, X. Sun, B. Jiang, M. Wang, X. Wu, D. Tian, X. Song, X. Yin, Y. Shuai, N. Zhang, *Energy Storage Materials* **63**, 103026 (2023). doi: 10.1016/j.ensm.2023.103026
- [6] L. Shen, Y.-W. Song, J. Wang, C.-X. Zhao, C.-X. Bi, S.-Y. Sun, X.-Q. Zhang, B.-Q. Li, Q. Zhang, *Small Structures* **4**, 2200205 (2023). doi:10.1002/sstr.202200205

### Funding

DFG project FR 1246/15-1

### DFG Subject Area

327-01 Electron Structure, Dynamics, Simulation