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Preview

Two functions, one site: Accelerating lithium-sulfur chemistry

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This preview highlights a study by Bi and co-workers overcoming the adsorption-catalysis trade-off in lithium-sulfur batteries. Using *sp*-hybridized nitrogen dopants in lightweight graphdiyne hollow multishelled structures, they couple polysulfide trapping and accelerated conversion on identical atomic sites, enabling exceptional sulfur utilization under practical, lean-electrolyte conditions.

Lithium-sulfur (Li-S) batteries offer one of the highest theoretical energy densities among next-generation rechargeable systems (~2,600 Wh kg^{-1}) and utilize abundant, low-cost sulfur, positioning them as a premier sustainable alternative to conventional lithium-ion batteries.[1] However, their practical implementation remains severely hindered by the notorious polysulfide shuttle effect, sluggish redox kinetics, and the requirement for substantial host materials that add dead weight and dramatically compromise gravimetric performance.[2] Recent strategies employing nitrogen-doped (N-doped) carbon hosts have primarily focused on enhancing polysulfide adsorption via polar interactions; however, these approaches often require additional catalytic components or heavy, complex architectures, creating a persistent trade-off between trapping efficiency, conversion kinetics, and net energy density.[3]

To circumvent these limitations, Bi and co-workers, in a recent study published in *Nature Sustainability*,[4] report a spatially coupled approach that seamlessly integrates robust chemical adsorption and efficient electrocatalysis within lightweight, *sp*-hybridized nitrogen-doped graphdiyne (*sp*-N GDY) hollow multishelled structures (HoMSs). This elegant design achieves exceptional sulfur utilization with minimal inactive mass, delivering highly competitive pouch-cell performance even under lean electrolyte conditions.

The concept of *sp*-hybridized nitrogen (*sp*-N) doping, first realized in graphdiyne at acetylenic linkages in 2018,[5] features a unique orbital overlap with adjacent *sp*-carbon atoms. This configuration induces strong charge redistribution and rapid electron transfer—electronic properties previously exploited in metal-free electrocatalysis[6] and now strategically leveraged for polysulfide management in Li-S chemistry. As illustrated in Figure 1A, these specialized *sp*-N sites act as dual-functional active centers that simultaneously anchor lithium polysulfides (LiPSs) and catalyze their conversion on the exact same atomic sites.

Crucially, the full power of this electronic motif is realized by embedding it within an advanced hollow multishelled architecture (HoMS).[7] The hierarchical HoMS morphology provides effective multi-scale confinement, abundant internal voids to accommodate high sulfur loading, omnidirectional ion/electron transport pathways, and robust mechanical buffering against the large volume expansion inherent to sulfur lithiation. This combination creates a highly integrated, spatially coupled system wherein polysulfides are dynamically trapped and converted within the multi-shelled framework, drastically minimizing solid-state diffusion distances and eliminating unnecessary inactive mass overhead.

Density functional theory calculations coupled with *in situ* spectroscopic analysis (Raman, infrared, UV-vis, and electrochemical impedance) provide compelling mechanistic insights into this behavior. Both the central (cent-*sp*-N) and side (side-*sp*-N) *sp*-N configurations exhibit markedly higher LiPS binding energies (Figure 1B) and more favorable reaction thermodynamics (Figure 1C)—particularly for the typical rate-limiting $Li_2S_4 \leftrightarrow Li_2S_2$ conversion step—compared to conventional pyridinic nitrogen, graphitic nitrogen, or undoped GDY. These findings reveal why this specific architectural design succeeds where traditional adsorption-dominant hosts often falter: strong chemical binding no longer compromises conversion kinetics because both trapping and catalytic functions are electronically and spatially coupled at these ultra-lightweight atomic sites.

The synergy between sp-N doping and HoMS architecture delivers striking electrochemical performance. At a high sulfur content of 93.9 wt %, the cathode achieves ~1,462 mAh g^{-1} calculated based on the total mass of the sulfur and the host framework—approaching theoretical utilization with minimal inactive mass overhead (Figure 1D). This performance substantially outperforms the control materials (undoped GDY HoMSs, N-GDY HoMSs, and *sp*-N GDY hollow spheres), underscoring the value of this dual-engineering strategy. Furthermore, high sulfur loadings up to 8.5 mg cm^{-2} are successfully demonstrated. Crucially, pouch cells evaluated under technologically relevant conditions—featuring high sulfur loading (≥4 mAh cm^{-2}) and lean electrolyte conditions (E/S ratio <3)—sustain an energy density of ~350 Wh kg^{-1} for more than 160 cycles (Figure 1E), demonstrating the practical viability of the strategy.[8]

By overcoming the conventional adsorption-catalysis trade-off at the atomic

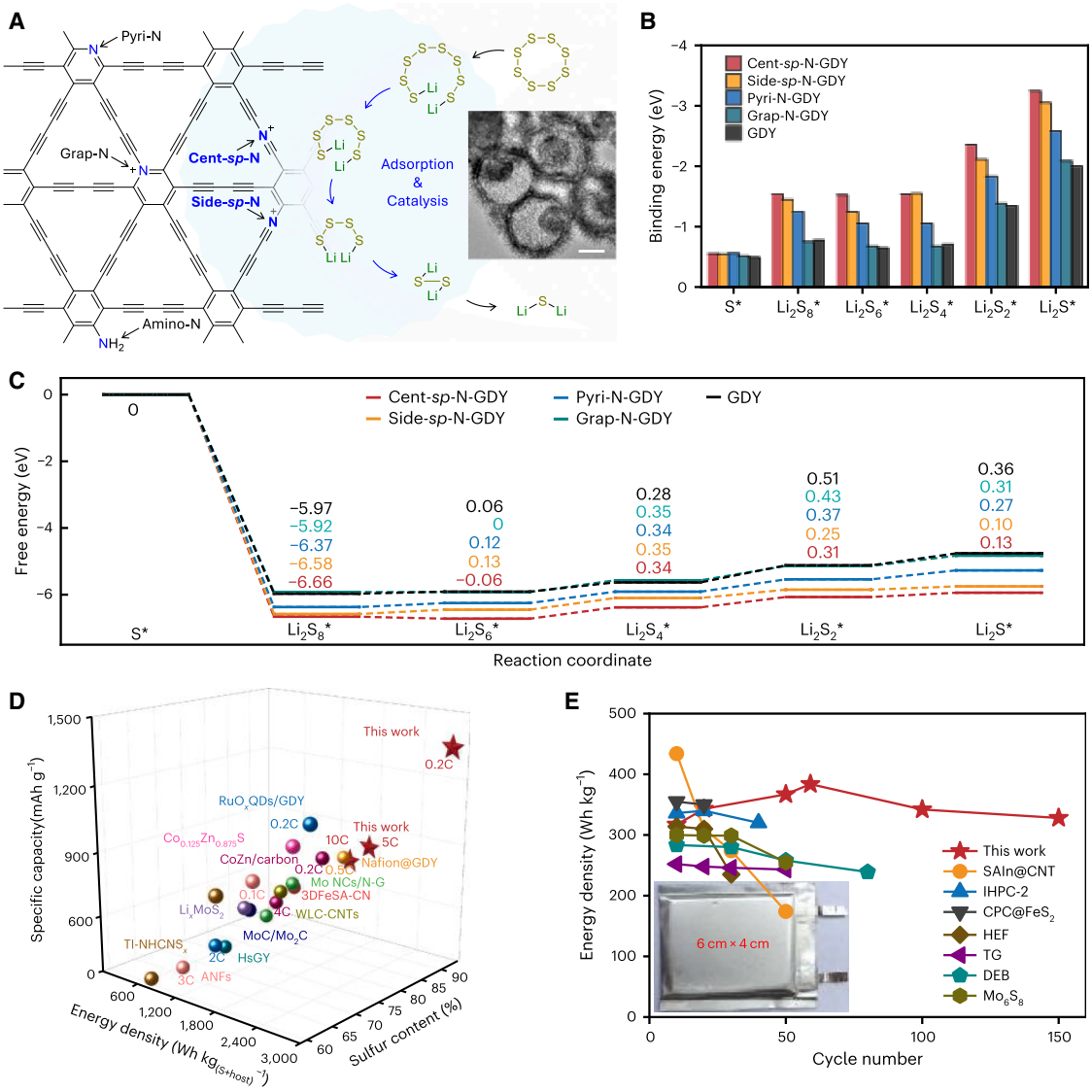

Figure 1. Comprehensive illustration of the *sp*-N GDY HoMS strategy for Li-S batteries

(A) Schematic of *sp*-N-doped GDY enabling spatially coupled polysulfide adsorption and catalytic conversion. Inset: transmission electron microscopy image of sp-N GDY HoMSs (scale bar: 500 nm).

(B) Binding energy comparison of sulfur species on different active sites.

(C) Calculated free energy diagrams for the discharge process from S_8 to Li_2S on various active sites.

(D) Rate performance comparison (specific capacities at different current densities) with representative reported Li-S batteries. Specific capacities are calculated using the combined mass of sulfur and host.

(E) Gravimetric energy density comparison of S@*sp*-N GDY HoMS pouch cells versus other reported Li-S systems. Inset: photo of the featured Li-S pouch cell.

(B)–(E) and the insets in (A) and (E) were reproduced from Bi et al.[4] under a Creative Commons license.

level while preserving an exceptionally lightweight host matrix, this work represents a significant milestone toward viable, high-energy-density Li-S batteries. It demonstrates that strategic electronic tailoring of carbonaceous framework can simultaneously mitigate chemical, mechanical, and gravimetric bottlenecks—propelling Li-S technology closer to commercial viability in weight-sensitive sectors such as long-range electric vehicles, electric aviation, and premium portable electronics. Furthermore, this approach also aligns closely with broader sustainability goals by maximizing the use of earth-abundant sulfur and minimizing dependence on critical or transition metals.

Nevertheless, key technical hurdles remain. The sophisticated multi-step synthesis of *sp*-N GDY HoMS architecture currently restricts its scalability. Translating this dual-function *sp*-N concept into simpler, continuously manufacturable carbon frameworks and morphologies will be essential for widespread industrial adoption. If successful, this foundational design principle—spatially and electronically coupling robust adsorption and accelerated catalysis within an ultralight host—could substantially accelerate the

commercialization of Li-S and inspire analogous strategies in other conversion-based battery systems.

In summary, this work establishes *sp*-N-doped GDY HoMSs as a powerful material platform that elegantly resolves a long-standing dilemma in Li-S electrochemistry. By merging atomic-level electronic precision with hierarchical architectural control, the work establishes new performance benchmarks under challenging high-sulfur-loading and lean-electrolyte conditions, offering a clear, resource-efficient blueprint for next-generation battery materials design.

DECLARATION OF INTERESTS

The authors declare no competing interests.

REFERENCES

1. Zhou, G., Chen, H., and Cui, Y. (2022). Formulating energy density for designing practical lithium–sulfur batteries. Nat. Energy *7*, 312–319.
2. Chen, S., Miao, K., and Zhou, J. (2025). Advances in lithium-sulfur batteries for commercialization. Sustain. Mater. Technol. *45*, e01500.
3. Yari, S., Conde Reis, A., Pang, Q., and Safari, M. (2025). Performance benchmarking and analysis of lithium-sulfur batteries for next-generation cell design. Nat. Commun. *16*, 5473.
4. Bi, R., Wang, J., Wan, J., Zhang, L., Sun, M., Huang, B., Li, B., Liang, Z., Zhao, Y., Li, L., et al. (2026). Spatially coupled adsorption and catalysis for sustainable lithium–sulfur batteries. Nat. Sustain. *9*, 763–773.
5. Zhao, Y., Wan, J., Yao, H., Zhang, L., Lin, K., Wang, L., Yang, N., Liu, D., Song, L., Zhu, J., et al. (2018). Few-layer graphdiyne doped with sp-hybridized nitrogen atoms at acetylenic sites for oxygen reduction electrocatalysis. Nat. Chem. *10*, 924–931.
6. Liu, B., Zhan, S., Du, J., Yang, X., Zhao, Y., Li, L., Wan, J., Zhao, Z., Gong, J., Yang, N., et al. (2023). Revealing the Mechanism of sp-N Doping in Graphdiyne for Developing Site-Defined Metal-Free Catalysts. Adv. Mater. *35*, 2206450.
7. Xu, W., Bi, R., Yang, M., Wang, J., Yu, R., and Wang, D. (2023). Hollow multishelled structural TiN as multi-functional catalytic host for high-performance lithium-sulfur batteries. Nano Res. *16*, 12745–12752.
8. Kakiage, K., Yano, T., Uehara, H., and Kakiage, M. (2024). Ultra-lightweight rechargeable battery with enhanced gravimetric energy densities >750 Wh kg^{-1} in lithium–sulfur pouch cell. Commun. Eng. *3*, 177.