Synergistic Dual Doping of Graphene Oxide: A New Route to High-Capacity Anodes for Sodium-Ion Batteries

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Sodium-ion batteries (NIBs) have emerged as promising alternatives to lithium-ion batteries (LIBs) for large-scale energy storage systems, owing to the abundance, low cost, and geographic availability of sodium [1]. Despite these advantages, the relatively large ionic radius of Na^+ and its weak interaction with the graphite lattice hinder its intercalation, rendering conventional graphite anodes nearly inactive in NIB systems. This fundamental limitation is primarily due to the insufficient interlayer spacing in graphite, which fails to accommodate Na^+ ions effectively. To address this challenge, various carbonaceous materials—such as amorphous carbon, hard carbon, and doped graphene derivatives—have been explored [2]. These materials possess high electronic conductivity, tunable pore structures, and chemical stability, making them suitable hosts for sodium storage. Among them, heteroatom-doped graphene-based materials have shown considerable potential due to their enhanced Na^+ adsorption sites, expanded interlayer distances, and defect-rich structures that facilitate ion transport [3,4]. Dual doping, in particular, has gained attention as a strategy to synergistically tailor the electronic structure and surface chemistry of graphene. Nitrogen atoms can introduce active sites and improve electronic conductivity, while sulfur doping can enlarge the interlayer spacing and enhance Na^+ binding through polar interactions. Together, these dopants can promote reversible sodium storage by stabilizing intermediate states and reducing energy barriers for ion insertion and extraction. Additionally, the choice of electrolyte plays a critical role in enabling effective Na^+ intercalation. Ether-based electrolytes such as diglyme have been reported to facilitate the co-intercalation of solvated Na^+ ions into carbon matrices, a process not feasible with conventional carbonate-based systems. This co-intercalation mechanism can mitigate structural degradation and improve the long-term stability of the anode.

In this study, we investigate nitrogen-sulfur dual-doped graphene oxide (N-S-GO) as a standalone anode material for NIBs, in combination with an ether-based (diglyme) electrolyte. Without requiring additional conductive additives or composite components, N-S-GO exhibits stable cycling performance, enhanced sodium-ion diffusion, and improved reversibility. The structural advantages provided by dual doping, combined with the favorable ion transport properties of the ether-based electrolyte, make this approach a promising candidate for future sodium-ion battery technologies. To the best of our knowledge, this is the first report demonstrating the direct use of N-S dual-doped graphene oxide with an ether-based electrolyte system for sodium-ion batteries. This work contributes to the development of high-performance, scalable, and cost-effective anode materials for next-generation energy storage systems.

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