Covalent Heterojunctions Enhance Bi_2S_3/Reduced Graphene Oxide (rGO) Nanocomposite Performance as Aqueous Zinc Ion Battery Material

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Abstract

The shortage of lithium resources, safety and recycling difficulty has focused attention on alternative energy storage devices in recent years. The aqueous zinc-ion battery (ZIB) stands out against such a background because of its earth abundance, safety, and environmental friendliness.[1] However, the limited choice of cathode materials hinders the development of advanced high-energy-density aqueous ZIBs. At present, manganese oxide[2] and vanadium oxide[3] are the two most widely studied zinc-ion battery cathodes, but the migration of Zn^{2+} in these materials is limited by the strong electrostatic interaction with lattice oxygen ions, resulting in poor reversible capacity. Metal sulfides, instead, may effectively improve the electrochemical performance reversibility of ZIBs. Layered metal sulfides have been extensively studied in monovalent cation (Li^+, Na^+, K^+) rechargeable batteries.[4] However, although limited studies with Bi_2S_3[5,6] as ZIB cathode material exist, their detailed electrochemical charge storage and transfer mechanisms are not well understood.

In this work, we explore the effect of covalent anchoring Bi_2S_3 on reduced graphene oxide (rGO) on the stability and cycling performance as a cathode for aqueous ZIBs. During the hydrothermal synthesis, the reduced graphene oxide serves as the nucleation substrate enabling the formation of fine and uniformly sized Bi_2S_3 grains, Figure 1 (a). Raman and X-ray photoelectron spectroscopy (XPS) confirm the formation of Bi-O-C heterojunctions during hydrothermal synthesis. These oxygen bridges serve as efficient electron transfer channels in the Bi_2S_3/rGO composite for rapid charge compensation during Zn^{2+} incorporation/extraction. As a result, Bi_2S_3/rGO composite shows notably better rate performance and cycling stability compared with pristine Bi_2S_3. The specific capacity of Bi_2S_3-rGO8 composite is ~186 mAh g^{-1} at the current density of 500 mA g^{-1} after 150 cycles, considerably higher than unsupported Bi_2S_3. Additionally, the Bi_2S_3 nucleated on GO with smaller particle sizes can shorten the transport path of zinc ions, which is beneficial for fast charge transfer. Therefore, Bi_2S_3-rGO8 can deliver more than 100 mAh g^{-1} at 10 A/g charge/discharge current density, Figure 1 (b).

Also, the zinc storage mechanism was analyzed by X-ray diffraction spectroscopy (XRD) and XPS, indicating a reversible conversion reaction of Zn^{2+} in the Bi_2S_3-rGO framework. During discharging, Zn^{2+} is embedded in Bi_2S_3-rGO frame to form ZnS and Bi wrapped in rGO. The process is accompanied by the dissolution of bismuth into electrolyte and the formation of $(ZnSO_4)[Zn(OH)_2]_3 \cdot 5H_2O$ (ZHS) on the electrode surface. Inhibition of these two processes may further increase the cycle stability of Bi_2S_3-rGO. Rotating ring disc electrode (RRDE) measurements, in which we detect dissolved Bi, indicate that Bi dissolution in the electrolyte during charging/discharging is mitigated in Bi_2S_3/rGO electrode, compared to pristine Bi_2S_3.

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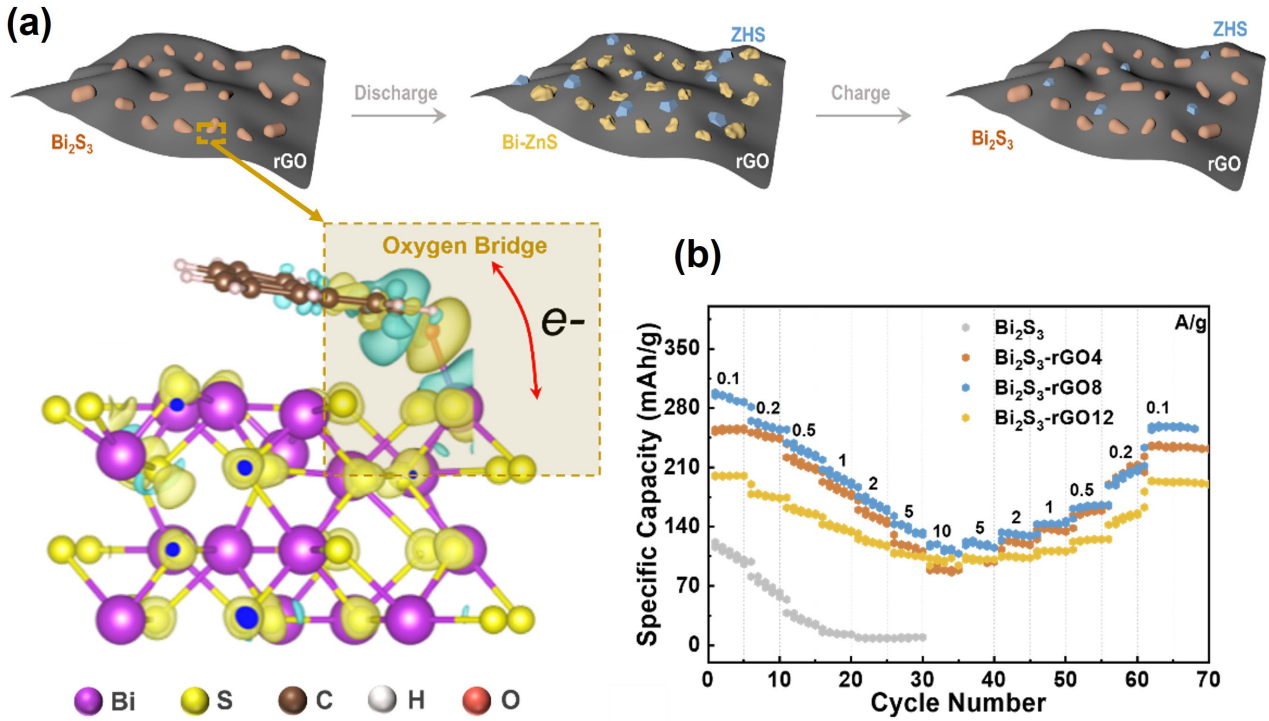

Figure 1. Sketch of Bi_2S_3/rGO nanocomposites during (dis)charging, interface charge-density difference, and battery rate performance.

Figure 1

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