Construction of 3D Electronic/Ionic Conduction Networks for All-Solid-State Lithium Batteries

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High and balanced electronic and ionic transportation networks with nanoscale distribution in solid-state cathodes are crucial to realize high-performance all-solid-state lithium batteries. Using Cu2SnS3 as a model active material, such a kind of solid-state Cu2SnS3@graphene-Li7P3S11 nanocomposite cathodes are synthesized, where 5–10 nm Cu2SnS3 nanoparticles homogenously anchor on the graphene nanosheets, while the Li7P3S11 electrolytes uniformly coat on the surface of Cu2SnS3@graphene composite forming nanoscaled electron/ion transportation networks. The large amount of nanoscaled triple-phase boundary in cathode ensures high power density due to high ionic/electronic conductions and long cycle life due to uniform and reduced volume change of nano-Cu2SnS3. The Cu2SnS3@graphene-Li7P3S11 cathode layer with 2.0 mg cm⁻² loading in all-solid-state lithium batteries demonstrates a high reversible discharge specific capacity of 813.2 mAh g⁻¹ at 100 mA g⁻¹ and retains 732.0 mAh g⁻¹ after 60 cycles, corresponding to a high energy density of 410.4 Wh kg⁻¹ based on the total mass of Cu2SnS3@graphene-Li7P3S11 composite based cathode. Moreover, it exhibits excellent rate capability and high-rate cycling stability, showing reversible capacity of 363.5 mAh g⁻¹ at 500 mA g⁻¹ after 200 cycles. The study provides a new insight into constructing both electronic and ionic conduction networks for all-solid-state lithium batteries.

1. Introduction

All-solid-state lithium batteries using nonflammable inorganic solid electrolytes have attracted increasing attention due to its high safety and reliability.[1–4] Sulfide electrolytes are considered to be one of the most promising solid-state electrolytes due to high ionic conductivity and excellent processability as well as low elastic modulus.[5–9] The high ionic conductivities of glass-ceramic sulfide electrolytes such as Li10GeP2S12 (1.2 × 10⁻² S cm⁻¹),[8] Li7P3S11 (1.7 × 10⁻² S cm⁻¹),[5] and Li54Si74P144S117Cl6 (2.5 × 10⁻² S cm⁻¹)[9] are comparable with that of the liquid carbonate electrolytes. However, the electrochemical performance of all-solid-state lithium batteries using sulfide electrolytes is still inferior to that of commercial liquid electrolyte based lithium-ion batteries. The challenges are attributed to (1) poor electronic/ionic conduction networks in cathodes due to the limited triple solid–solid contact interface and the poor interface compatibility, and (2) high stress/strain due to volume change of active materials and incompressible solid electrolytes.[10–12]

Among all cathode materials, transition-metal sulfides have been considered the most promising cathodes for high-performance all-solid-state lithium batteries because they (1) show better interface compatibility with sulfide electrolytes than lithium transition-metal oxides, (2) have higher energy density and higher chemical stability than lithium transition-metal oxides,[13–16] and (3) possess better electronic conductivity and electrochemical stability than Li2S and sulfur.[17–20] However, the poor electronic/ionic conduction network and continuously increased solid–solid contact interface resistance during charge/discharge cycles are still challenges for solid-state transition-metal sulfide cathodes. One promising strategy is to coat nanosized transition-metal sulfides onto highly conductive carbonaceous materials to enhance the electronic conductivity.[21–23] The thickness of transition-metal sulfide should be less than few nanometers to reduce the stress/strain and interface resistance, and to allow the electrons conduction across the transition-metal sulfide nanolayer through tunnel effect to solid electrolyte for charge transfer reaction. The long cycle life of such active material/electron additive nanocomposite cathodes was demonstrated by us using nanolayer Cu2ZnSnS4 (or sulfur) deposited graphene as model cathodes.[21,22] The Cu2ZnSnS4/graphene nanocomposite prepared using hydrothermal approach shows a discharge capacity of 645.4 mAh g⁻¹ after 50 cycles at 50 mA g⁻¹.[22]
while 2 nm S-coated graphene can maintain a high and reversible capacity of 830 mAh g$^{-1}$ at 1.0 C for 750 cycles.$^{[24]}$ However, the ionic conduction of active material/electron additive is still a challenge due to poor contact between active materials with electrolyte, which limits the further enhancement of the electrochemical performance. To enhance the ionic conduction of cathode, we coated a thin layer of electrolyte onto active materials nanosheet to form active materials/electrolyte nanocomposite. All-solid-state lithium batteries employing cobalt sulfide-Li$_2$P$_2$S$_11$ nanocomposites in combination with the Li$_2$P$_2$S$_11$ electrolyte and Super P as the cathode and lithium metal as the anode showed reversible discharge capacity of 421 mAh g$^{-1}$ at 1.27 mA cm$^{-2}$ after 1000 cycles.$^{[25]}$ However, the poor electronic conduction in active materials/electrolyte nanocomposite cathode limits the electrochemical performance. Here, we reported a comprehensive strategy to simultaneously enhance both ionic and electronic conductions by formation of 3D electronic/ionic conduction networks in cathode. Use of Cu$_2$SnS$_3$ as a model active material, we first coated 5–10 nm Cu$_2$SnS$_3$ nanoparticles onto graphene to ensure high electronic conduction, and then coated additional sulfide electrolyte layer/particle to form nanoscaled 3D electronic/ionic networks (Figure 1). Such a solid-state nanocomposite cathode structure is similar to porous sulfide nanocomposite cathodes in liquid electrolyte. Due to immobility of solid sulfide electrolytes, the transition-metal sulfide nanoparticles anchored on graphene can effectively reduce the stress/strain and achieve a good electronic conduction. The thin electrolyte layer coating on nano-active material/graphene composite play a critical role in establishing the 3D ionic conduction network for high power density and long cycle life.

The reason to select copper tin sulfide (Cu$_2$SnS$_3$) as a model active cathode material is because it has a higher electrical conductivity than other copper-based multicomponent chalcogenide,$^{[26,27]}$ and higher energy density than reported Cu$_2$ZnSnS$_4$ in all-solid-state lithium batteries. The low energy density of Cu$_2$ZnSnS$_4$ is because the alloying reaction of Zn in Cu$_2$ZnSnS$_4$ begins below 0.5 V$^{[28]}$ that is beyond the range of the setting cut-off voltage (0.5–3 V) of Cu$_2$ZnSnS$_4$ cathodes.

In this work, Cu$_2$SnS$_3$@graphene nanocomposite were prepared using a hydrothermal reaction, and then a thin layer of Li$_2$P$_2$S$_11$ electrolyte was coated on Cu$_2$SnS$_3$@graphene via in situ liquid-phase reaction forming Cu$_2$SnS$_3$@graphene-Li$_2$P$_2$S$_11$ nanocomposites (Figure 1). An all-solid-state lithium battery employing Cu$_2$SnS$_3$@graphene-Li$_2$P$_2$S$_11$ as a cathode material, a lithium metal as an anode and a sulfide electrolyte bilayer as a solid electrolyte was fabricated and the electrochemical performances of Cu$_2$SnS$_3$@graphene-Li$_2$P$_2$S$_11$/Li$_{10}$GeP$_2$S$_{12}$/75%Li$_2$S-24%P$_2$S$_5$-1%P$_2$O$_5$/Li all-solid-state cell were evaluated and compared to other all-solid-state cells using Cu$_2$SnS$_3$, Cu$_2$SnS$_3$@graphene as cathode material. The results prove that the 3D nanoscaled ionic/electronic networks formed by coating electrolytes on active materials/graphene composite can significantly improve the cycling stability and rate capability of all-solid-state lithium batteries. The resultant Cu$_2$SnS$_3$@graphene-Li$_2$P$_2$S$_11$ nanocomposite based cathodes in all-solid-state lithium batteries exhibit capacity of 732.0 mAh g$^{-1}$ and 363.5 mAh g$^{-1}$ after cycling at 100 mA g$^{-1}$ for 60 cycles and 500 mA g$^{-1}$ for 200 cycles, respectively.

2. Results and Discussion

Figure 2 displays the X-ray powder diffraction (XRD) patterns of Cu$_2$SnS$_3$, Cu$_2$SnS$_3$@graphene, and Cu$_2$SnS$_3$@graphene-Li$_2$P$_2$S$_11$ nanocomposite. The main characteristic diffraction peaks of pure Cu$_2$SnS$_3$ at 28.6°, 32.7°, 47.6°, and 56.7° correspond to the diffraction from the (111), (200), (220), and (311) planes of the standard cubic phase Cu$_2$SnS$_3$ (JCPDS card no. 01-089-2877).$^{[26]}$ The obtained Cu$_2$SnS$_3$@graphene and Cu$_2$SnS$_3$@graphene-Li$_2$P$_2$S$_11$ nanocomposite show similar XRD patterns to that of Cu$_2$SnS$_3$, indicating the introduction of graphene oxide during hydrothermal synthesis process and the electrolyte deposition during in situ liquid-phase reaction.

![Figure 1. Schematic illustration of the fabrication procedure of Cu$_2$SnS$_3$@graphene-Li$_2$P$_2$S$_11$ nanocomposite, where hydrothermal method was applied to synthesize Cu$_2$SnS$_3$@graphene composite and then Li$_2$P$_2$S$_11$ electrolyte was coated on Cu$_2$SnS$_3$@graphene by an in situ liquid-phase reaction followed by an annealing treatment.](Image)

![Figure 2. XRD patterns of Cu$_2$SnS$_3$, Cu$_2$SnS$_3$@graphene, and Cu$_2$SnS$_3$@graphene-Li$_2$P$_2$S$_11$ nanocomposite.](Image)
do not influence the phase of Cu$_2$SnS$_3$. For Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ nanocomposite, no obvious peaks related to graphene and electrolyte are observed, which might due to the low amount of graphene and electrolyte in the nanocomposites. The amount of graphene in the Cu$_2$SnS$_3$@graphene can be determined by carbon sulfur analyses and the results reveal that the graphene content in the as-prepared Cu$_2$SnS$_3$/graphene is about 9.6%. The reduction of graphene oxide during hydrothermal reaction was confirmed by Raman spectroscopy, as shown in Figure 3. Two peaks at about 1350 and 1592 cm$^{-1}$ are observed, which attributes to the D and G bands from graphene. The peak at about 1592 cm$^{-1}$ (G band) is related to the vibration of the sp$^2$-banded carbon atoms in a 2D hexagonal lattice, while the peak at about 1350 cm$^{-1}$ (D band) is related to the defects and disorder in hexagonal graphitic layers.[29] The intensity ratio ($I_D/I_G$) of the D band to G band is 1.027 for Cu$_2$SnS$_3$/graphene that is higher than that of graphene oxide ($I_D/I_G=0.911$), due to reduction of graphene oxide during hydrothermal reaction[27,30] and decrease in the average size of the sp$^2$ domains.[29,31] After coating Cu$_2$SnS$_3$@graphene with Li$_7$P$_3$S$_{11}$ electrolyte, the intensity ratio ($I_D/I_G$) of the D band to G band (1.024) for Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ nanocomposite is similar to that of Cu$_2$SnS$_3$@graphene, suggesting that the introduction of electrolytes does not change the electronic conduction property of graphene.

The morphology of three Cu$_2$SnS$_3$ cathode materials was observed using a scanning electron microscope (SEM). As shown in Figure 4, the pure Cu$_2$SnS$_3$ has particle size of 5–10 nm and obvious aggregations. However, the Cu$_2$SnS$_3$ nanoparticles in Cu$_2$SnS$_3$@graphene composite anchor homogeneously on the graphene nanosheets without aggregation. After Li$_7$P$_3$S$_{11}$ coating on the Cu$_2$SnS$_3$@graphene, more particles are distributed on the graphene nanosheets, which implies that Li$_7$P$_3$S$_{11}$ are deposited on the surface of Cu$_2$SnS$_3$@graphene forming Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ nanocomposite.

More detailed morphology and structure analysis of Cu$_2$SnS$_3$, Cu$_2$SnS$_3$@graphene and Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ nanocomposites were further conducted using transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM). As shown in Figure 5, the pure Cu$_2$SnS$_3$ nanoparticles (Figure 5a) with a particle size distribution of 5–10 nm aggregate into a large secondary particles, while the Cu$_2$SnS$_3$ nano-particles in Cu$_2$SnS$_3$@graphene (Figure 5c) and in Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ nanocomposite (Figure 5e) distribute homogenously on graphene nanosheets without aggregation, which is in accordance with the SEM results. These results imply that the presence of graphene will restrict aggregation of nanoparticles during the hydrothermal process. In the HRTEM images of Cu$_2$SnS$_3$ (Figure 5b), Cu$_2$SnS$_3$@graphene (Figure 5d), and Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ (Figure 5f), the lattice fringe of about 0.314 nm corresponds to the (−2 −1 1) lattice plane of triclinic Cu$_2$SnS$_3$ structure[27] and the 0.299 nm can be indexed to (3 1 1) lattice plane of crystal.
Li$_7$P$_3$S$_{11}$ electrolyte. As shown in Figure 5f, the lattice planes of Cu$_2$SnS$_3$ and Li$_7$P$_3$S$_{11}$ are interlaced, suggesting that Cu$_2$SnS$_3$ nanoparticle is partially coated by Li$_7$P$_3$S$_{11}$ electrolyte, demonstrating the successful formation of large amount of nanoscaled 3D triple phase in Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ nanocomposite. These intimate and stable solid-solid contact triple-interfaces between Cu$_2$SnS$_3$ nanoparticles, graphene, and electrolytes will significantly enhance the cycle life and rate performance. Furthermore, the scanning transmission electron microscopy (STEM) element mapping of Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ nanocomposite in Figure 5g also confirms that the Cu$_2$SnS$_3$ nanoparticles were coated with Li$_7$P$_3$S$_{11}$ electrolyte as demonstrated by overlapping of P element with Cu, Sn elements.

The graphene in Cu$_2$SnS$_3$@graphene and Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ nanocomposites not only enhances the electronic conduction, but also significantly enhances reaction area as demonstrated by the high surface area obtained using Brunauere–Emmette–Teller (BET) nitrogen absorption/desorption measurement. The specific surface area of Cu$_2$SnS$_3$@graphene is 84.0795 m$^2$ g$^{-1}$, which is five times higher than that of neat aggregated Cu$_2$SnS$_3$ nanoparticles (17.1361 m$^2$ g$^{-1}$). The large surface area of Cu$_2$SnS$_3$@graphene provides more sites for the deposition of Li$_7$P$_3$S$_{11}$ electrolyte during the in situ liquid-phase synthesis of Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ nanocomposite, which is beneficial for the construction of continuous ionic/electronic network. In addition, the introduction of graphene can also accommodate the volume expansion/contraction of active materials during discharge-charge processes, suppressing agglomeration of active materials. Therefore, long cycling stability and high-rate capability are expected for Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$ nanocomposite cathodes.

The electrochemical performances of three active material based cathodes (Cu$_2$SnS$_3$, Cu$_2$SnS$_3$@graphene, and Cu$_2$SnS$_3$@graphene-Li$_7$P$_3$S$_{11}$) were evaluated in all-solid-state lithium cells using cyclic voltammogram (CV). As shown in Figure S1a, Supporting Information, during the first cathodic scan (Li ion insertion) of pure Cu$_2$SnS$_3$ cathode, two broad peaks at 1.881 and 0.907 V are observed, corresponding to the electrochemical reduction of Cu and Sn ions into metallic Cu and Sn nanoparticles. After
first lithiation/delithiation activation cycle, the reduction peak at around 1.881 V shift to 1.966 V. The Cu$_2$SnS$_3$@graphene (Figure S1b, Supporting Information) and Cu$_2$SnS$_3$@graphene-Li$_3$P$_3$S$_11$ (Figure S1c, Supporting Information) cathodes display an identical electrochemical reaction mechanism as Cu$_2$SnS$_3$ cathode but have a less polarization between lithiation and delithiation due to the enhanced electronic conductivity for Cu$_2$SnS$_3$@graphene cathode and both high ionic and electronic conductivity for Cu$_2$SnS$_3$@graphene-Li$_3$P$_3$S$_11$ cathode. After first activation cycles, the CV curves of all three Cu$_2$SnS$_3$ cathodes are almost overlapped, indicating the high reversibility of Cu$_2$SnS$_3$ in the all-solid-state lithium cell.

The first three discharge-charge voltage profiles of pure Cu$_2$SnS$_3$, Cu$_2$SnS$_3$@graphene, and Cu$_2$SnS$_3$@graphene-Li$_3$P$_3$S$_11$ in all-solid-state lithium batteries between 0.5 and 3.0 V at the current density of 100 mA g$^{-1}$ and room temperature are shown in Figure 6a-c. Three Cu$_2$SnS$_3$-based cathodes show similar voltage profiles, where the obvious discharge plateau at around 2.0 V and a slop between 0.9 and 1.5 V derive from the reduction of Cu and Sn species to metallic Cu and Sn, respectively, and the slop between 1.7 and 2.1 V, and a plat plateau at 2.3 V in the charge curves are related to the reversible oxidation reaction of Sn and Cu nanoparticles, respectively, which are in accordance with the results shown in CV

![Figure 6. Galvanostatic discharge/charge profiles of a) pure Cu$_2$SnS$_3$ nanoparticles, b) Cu$_2$SnS$_3$@graphene, c) Cu$_2$SnS$_3$@graphene-Li$_3$P$_3$S$_11$. d) Cyclic performance of as-prepared samples under a current density of 100 mA g$^{-1}$. e) High-rate cyclic performance of as-prepared samples at 500 mA g$^{-1}$.](image-url)
curves (Figure S1, Supporting Information). The firstoulomeric efficiency of Cu2SnS3@graphene-LiP3S11 electrode is 82.9%, which is higher than that of pure Cu2SnS3 (66.1%) and Cu2SnS3@graphene (72.2%) cathodes. The lowoulomeric efficiency for pure Cu2SnS3 and Cu2SnS3@graphene cathodes might be caused by the inferior triple contacts among the discharge products, electrolyte, and electronic additive due to volume expansion of active material after discharge process, leading to electrochemically inaccessibility of partial noncontact discharge products.[25] Besides, after the first cycle, the voltage separation between the charge and discharge plateaus is the smallest for Cu2SnS3@graphene-LiP3S11 nanocomposite, suggesting the nanoscaled intimate triple contact structures, where electrochemical reaction occurs, are still well maintained after cycling, further confirming the introduction of both graphene and LiP3S11 electrolyte can increase the reversibility of Cu2SnS3. Figure 6d compares the cyclic performance of pure Cu2SnS3, Cu2SnS3@graphene, and Cu2SnS3@graphene-LiP3S11 cathodes at a low current density of 100 mA g−1. Cu2SnS3@graphene-LiP3S11 electrode exhibits a stable capacity of 732.0 mAh g−1 for 60 cycles, corresponding to a high energy density of 410.4 Wh kg−1 based on the total mass of cathode layer. However, the discharge specific capacities of Cu2SnS3@graphene and Cu2SnS3 cathodes decreases to 541.2 and 111.8 mAh g−1 after 60 cycles, respectively. The long cycling stability was measured at a high current density of 500 mA g−1 (Figure 6e). After 200 cycles, the discharge specific capacity of Cu2SnS3@graphene-LiP3S11 electrode still retains at about 363.5 mAh g−1, while the discharge capacities of Cu2SnS3@graphene and Cu2SnS3 cathodes are rapidly decreased to 78.9 and 36.4 mAh g−1, respectively. These results indicate that Cu2SnS3@graphene-LiP3S11 nanocomposite cathode has long cycling stability in all-solid-state lithium batteries due to unique cathode architecture with 3D ionic/electronic network and small/uniform volume change.

The rate performances of pure Cu2SnS3, Cu2SnS3@graphene, and Cu2SnS3@graphene-LiP3S11 electrodes were also evaluated. Figure S2, Supporting Information, shows the galvanostatic discharge/charge curves of three Cu2SnS3-based cathodes. The Cu2SnS3@graphene-LiP3S11 nanocomposite can provide discharge specific capacities of 964.5, 813.2, 706.8, 540.3, 466.9, and 359.5 mAh g−1 at current densities of 50, 100, 200, 500, 1000, and 2000 mA g−1, respectively, while the Cu2SnS3@graphene cathodes can only provide discharge specific capacities of 829.7, 739.5, 584.5, 462.5, 219.5, and 96.6 mAh g−1, respectively. Cu2SnS3 cathode shows the worst rate performance with capacities of 828.1, 660.6, 543.1, 436.3, 202.8, and 67.2 mAh g−1 at the same step currents, respectively.

To understand the mechanism for superior rate capacity and long cyclability of the Cu2SnS3@graphene-LiP3S11 cathodes, the impedance of Cu2SnS3@graphene-LiP3S11 cathodes at a fully charged state at first and 100 cycles was measured using electrochemical impedance spectroscopy (EIS) and compared them to the impedances of other two cathodes. As shown in Figure S3, Supporting Information, the Nyquist plots mainly consist of ohmic resistance (R0) in the high-frequency range, which stems from the resistance of the electrode and solid electrolyte layers, a compressed semicircle in the middle-frequency region, which corresponds to the charge transfer resistance (Rct), and a sloped line in the low-frequency region, which is related to Li-ion diffusion into the bulk electrode (Zw). Constant phase element (CPE) represents the nonideal capacitance of the double layer. The fitted results obtained from the equivalent circuit model are shown in Table S1, Supporting Information. After the first discharge/charge cycle, the EISs of three cells show a similar slope line, and Rct values are almost the same due to the employment of the same bilayer electrolyte. After 100 cycles, the Cu2SnS3@graphene-LiP3S11/Li cell shows the smallest ohmic resistance of 160.3 Ω and charge transfer resistance of 148.4 Ω. While the Cu2SnS3 cathode shows the largest ohmic and charge transfer resistance of 783.9 and 661.9 Ω, respectively (Table S1, Supporting Information). The super stable Cu2SnS3@graphene-LiP3S11 cathodes are attributed to uniformly distributed nanoscaled Cu2SnS3 on graphene and LiP3S11 electrolyte on Cu2SnS3@graphene, which ensures uniform electronic/ionic conduction networks, thus resulting in the homogenous and fast reaction of Cu2SnS3 as well as small and uniform volume change. The uniform volume change will generate small stress/strain, thus less resistance increase during charge/discharge cycles and long cycle life.[32] In addition, graphene can also accommodate the expansion and contraction of Cu2SnS3, leading to enhanced structural integrity of the solid-state Cu2SnS3@graphene-LiP3S11/Li cells. In all-solid-state lithium batteries fabricated by mixing and compressing, the contact between active material and electrolyte is point-to-point contact. The coating LiP3S11 electrolytes on Cu2SnS3 precipitated graphene change the point-contact to 3D intimate contact in the obtained cathode, which enhance the reaction kinetics and retains integrated morphology and structural robustness of the cathode. Therefore, the best electrochemical performance is realized in all-solid-state lithium batteries using Cu2SnS3@graphene-LiP3S11 based cathodes.

3. Conclusion

Cu2SnS3@graphene-LiP3S11 nanocomposite cathode material with 3D ionic/electronic conduction networks were synthesized by two-step reactions: a hydrothermal process to homogeneously coat 5–10 nm Cu2SnS3 particle on graphene nanosheet followed by in situ liquid-phase deposition of LiP3S11 electrolyte onto Cu2SnS3@graphene. The Cu2SnS3@graphene-LiP3S11 nanocomposite cathode material can maintain the structural integrity, enhance the contact area between electrode material and electrolyte, leading to the decrease of interfacial resistance, and thus endowing the batteries with excellent cycling stability and rate capability. The discharge specific capacity of Cu2SnS3@graphene-LiP3S11 cathode materials in all-solid-state Li batteries retains at about 732.0 mAh g−1 at 100 mA g−1 for 60 cycles, corresponding to a high energy density of 410.4 Wh kg−1 based on the total mass of cathode layer. At a high current of 500 mA g−1, Cu2SnS3@graphene-LiP3S11 cathodes can maintain 363.5 mAh g−1 for 200 cycles. Besides, the Cu2SnS3@graphene-LiP3S11 nanocomposite also demonstrates extraordinary rate capability of 964.5, 813.2, 706.8, 540.3, 466.9, and 359.5 mAh g−1 at current densities of 50, 100, 200, 500, 1000, and 2000 mA g−1, respectively. This study provides a new
route for optimizing the interfacial structure of all-solid-state lithium battery with good rate capability and cycling stability.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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