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Al₂S₃ Cathode for Rechargeable Aluminum-Sulfur Batteries with Improved Cycling Reversibility

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Rechargeable aluminum-sulfur (Al–S) batteries are regarded as the potential choice for next-generation energy storage system with advantages of high theoretical energy density (1340 Wh kg⁻¹), the earth abundance of the constituent electrode materials, and safe operation. Whereas the electrochemical performance of Al–S batteries is beset by its poor reversibility. In this article, we demonstrate a highly reversible Al–S battery with Al₂S₃ as the cathode. Mechanistic studies with electrochemical and spectroscopic methodologies identified

that the enhancement in reversibility by Al₂S₃ cathode is attributed to the circumvention of the sluggish conversion of polysulfides to sulfur. Al₂S₃ cathode undergoes a faster solid to liquid conversion reaction (Al₂S₃↔polysulfide) than that of the sulfur cathode that suffers from slow kinetic process of solid-to-solid conversion (S↔Al₂S₃). Thus, Al–S battery composed of Al₂S₃ cathode delivers a highly reversible electrochemical reaction with the average Coulombic efficiency >95% for 100 cycles.

Rechargeable aluminum batteries (RABs) have recently attracted great attention due to its excellent safety, natural abundance of aluminum and high theoretical capacity (2980 mAh g⁻¹ and 8050 mAh cm⁻³).^[1] Nowadays, most of RABs are composed of an Al-metal anode in conjunction with various insertion-type cathodes including metal oxides, conductive polymers and carbon-based materials in ionic liquid-based electrolyte, which mostly show only limited specific capacities (<100 mAh g⁻¹).^[2–4] Sulfur as the conversion-type cathode material possesses a high theoretical capacity of 1672 mAh g⁻¹. When couple with aluminum anode, a rechargeable aluminum-sulfur (Al–S) batteries can deliver a high theoretical energy density (1340 Wh kg⁻¹), which are much superior to those of Al-ion battery systems. However, the low reversibility of the electrochemical reaction between Al and S when operating the Al–S batteries makes it very difficult to achieve a high Coulombic efficiency (CE) if S cathode is used.

Since first reported in 2015, efforts are mostly focused on the electrolyte modification or engineering carbon/sulfur

composite to enhance the reversibility of rechargeable Al–S batteries.^[5–9] For example, Wang et al. demonstrated the first reversible Al–S batteries by enhancing the oxidation kinetics of AlS_x via encapsulating sulfur into microporous carbon matrix.^[5]

Li et al. found that [Al₂Cl₆Br]⁻ exhibited a lower energy barrier to release Al³⁺ than the [Al₂Cl₇]⁻, leading to an enhanced electrochemical reversibility and extended life span of the battery.^[6] Recently, our group demonstrated Co(II,III) as an electrochemical catalyst in the sulfur cathode that rendered a reduced voltage hysteresis and improved capacity retention for Al–S batteries.^[7] However, thus far almost all of these works have been limited to low CE (that is, <90%), which is still unsatisfactory comparing to those of the Li–S batteries. The low CE leads to poor sulfur utility and active material loss, consequently, a low energy density and short life-span in rechargeable Al–S batteries. Identifying the influence factors of the sulfur conversion reversibility and designing a cathode with high CE, therefore, become crucial parts in the realm of Al–S batteries.

The poor reversibility of Al–S batteries is ascribed to the difficulty of oxidizing AlS_x.^[5] The high energy barrier of polysulfide converts to sulfur is the main reason for the low CE, which is associated with poor cycling life and low energy efficiency of the Al–S batteries. In the case of lithium-sulfur (Li–S) batteries, the replacement of sulfur with lithium sulfide (Li₂S), when preparing the cathode, can function well with improved reversibility. In comparison to the lithiation of elemental sulfur through the sluggish solid-solid S↔Li₂S reaction, the delithiation of Li₂S can undergo a fast solid-liquid Li₂S↔polysulfide reaction.^[10] Thus, we believe that Al₂S₃ can also be applied to the Al–S batteries to achieve an enhanced electrochemical performance if the sluggish conversion of polysulfides to sulfur can be circumvented.

In this article, we demonstrate that the Al₂S₃ cathode possesses a high electrochemical reversibility with CE >95%

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for 100 cycles, which is considerably higher than previous reports (CE < 90%). The spectroscopic analysis reveals that the poor reversibility associated with the sluggish conversion of sulfide oxidation to sulfur is circumvented by applying Al_2S_3 cathode. Al_2S_3 cathode can be reversibly converted into polysulfides, which suffers a faster solid to liquid conversion reaction ($\text{Al}_2\text{S}_3 \leftrightarrow \text{polysulfide}$) than that of the sulfur cathode that undergoes a slow kinetic process of solid-state conversion reaction ($\text{S} \leftrightarrow \text{Al}_2\text{S}_3$).

To demonstrate the electrochemical properties of Al_2S_3 cathode, two types of electrodes consisting of elemental S and Al_2S_3 were prepared. The elemental S cathode and Al_2S_3 cathode consisted of 60 wt% S and 60 wt% Al_2S_3 powder, respectively, mixing with 20 wt% Ketjen Black, and 20 wt% poly(vinylidene fluoride) (PVDF) binder which coated on a Ta current collector with a mass loading of active materials of around 0.5 mg cm^{-2} . The mass loading of the normalized sulfur in Al_2S_3 cathode is about 0.3 mg cm^{-2} . The ionic-liquid electrolyte was made by mixing 1-ethyl-3-methylimidazolium chloride (EMIC) with AlCl_3 in a mole ratio of 1:1.3. The electrochemical performance of the S cathode and Al_2S_3 cathode are evaluated in Swagelok-type cells. Al_2S_3 is an ionic crystal with a highly stable hexa-coordinated structure that endows Al_2S_3 with strong bonding energy. Al_2S_3 cathode is therefore needs to be activated before cycling.^[11] Before cycling, the Al_2S_3 cathode was firstly charged to $> 2.0 \text{ V}$ versus Al/Al^{3+} for activation, then both the S cathode and Al_2S_3 cathode were cycled at 0.16 A g^{-1} began with discharge in the first cycle. As shown in Figure 1(a), the S electrode delivers a specific capacity of 1100 mAh g^{-1} (calculated based on the mass of element S), however, it fades sharply down to $< 200 \text{ mAh g}^{-1}$ in 10 cycles with CE of only $\sim 80\%$. We note that the CE of S electrode at the initial stage is above 100%, which is same as that in previously reports.^[5,7] This phenomenon is still unknown to the [EMIM]Cl: AlCl_3 electrolyte system and might involve electrostatics between Al^{3+} and sulfides anions or side reactions with the cationic aluminum species.^[6,12,13] In contrast, the Al_2S_3 electrode exhibits an initial specific capacity of 420 mAh g^{-1} (based on the mass of S in the cathode). The discharge capacity of the Al_2S_3 remains at 180 mAh g^{-1} with CE of $\sim 95\%$ for 100 cycles. (Figure 1a). This result reveals that Al_2S_3 possesses a higher CE and reversibility comparing to S electrode, but still suffers from a low active material utilization of less than 25% in consideration of the first discharge capacity divided to the theoretical capacity if Al_2S_3 is simply mixed with carbon black and PVDF. The low Al_2S_3 utilization was probably ascribed to its low electric conductivity similar to that of Li_2S cathode.^[11,14,15]

The CV profiles of the Al_2S_3 electrode and S electrode are presented in Figure 1(b). The Al_2S_3 cathode shows two anodic peaks at 1.8 and 1.5 V and one cathodic peak at 0.3 V, while the S cathode presents two anodic peaks at 1.8 and 1.7 V and one cathodic peak at 0.2 V. A larger polarization in the CV profile of the S electrode than that of Al_2S_3 cathode indicates a more sluggish kinetic response on S cathode. In Li-S battery, the amount of polysulfides dissolved in electrolyte was indicative of the kinetic response of the electrochemical conversion reactions of sulfur: the more polysulfides in the electrolyte, the

faster kinetic response of the electrode possessed.^[8] Therefore, after running the Al-S batteries with the S and Al_2S_3 cathodes for 5 cycles, we disassembled them under an argon atmosphere in a glove box to analyze the chemical contents in the electrolyte. The chemical contents of the electrolyte were analyzed by UV-vis spectroscopy performed on an electrolyte-soaked glass fiber separator. The UV-vis spectra of the electrolyte after cycling with the S cathode at the discharged and charged states were overlapped, illustrating sluggish kinetic response of the battery. Whereas, for the electrolyte cycled with the Al_2S_3 cathode, a large amount of S_6^{2-} was detected at the charged state. The polysulfide production could be attributed to the high reaction activity in the Al_2S_3 cathode. The kinetically accelerated Al-S battery assembled with Al_2S_3 cathode can be further supported by the electrochemical impedance spectroscopy (EIS) analysis. The batteries assembled with both the S cathode and Al_2S_3 cathode show similar resistance of $\sim 30 \Omega$ before cycling. After 5 discharge-charge cycles, the charge transfer resistance and interfacial resistance of the battery assembled with the S cathode significantly increased to over 500Ω in both charged and discharged states, which were much larger than those of the battery assembled with Al_2S_3 cathode. Therefore, an improved reaction kinetics in the battery is observed when Al_2S_3 is applied in the cathode.

To understand the difference in electrochemical activity between the Al_2S_3 cathode and S cathode, we performed in-situ Raman spectra analysis. As shown in Figure 2, the Raman spectra of pristine S cathode shows pronounced peaks (Figure S1) of elemental S at 157, 220 and 475 cm^{-1} . It should be noted that the Raman spectra does not show any evidence of polysulfides (S_3^- , S_4^{2-} , S_6^-) which is same as previous reports.^[16] The intensity of the peaks associated with S decrease during the first discharge process until undetectable at 0.3 V. The peaks associated with Al_2S_3 (252, 347 and 504 cm^{-1}), the final discharged product of Al_2S_3 battery was not observed even when the S cathode was discharged to 0.1 V in Figure 2(a). It could be attributed to the polysulfides layer covered on the S cathode, the thickness of which was larger than the detectable depth of Raman laser ($\sim 100 \text{ nm}$).^[5,17] During the charging stage, the characteristic peaks of sulfur appear at only 2 V or above. This result could be ascribed to the sluggish kinetics of the sulfur conversion from polysulfides to elemental sulfur that leads to high voltage hysteresis. For the Al_2S_3 cathode, the peaks of Al_2S_3 gradually disappear during the charge process and no peaks related to the sulfur were generated even when battery was charged to 2.2 V (Figure S2). The Al_2S_3 cathode could change forth and back reversibly in the next discharge process within voltage range of 0.1 to 1.8 V. These results support the notion that the low reversibility of sulfur cathode could be attributed to the inability to oxidize polysulfide to elemental sulfur. Whereas the electrochemical process of the Al_2S_3 electrode could be reversibly cycled between S^{2-} and polysulfides, which avoid the sluggish conversion to sulfur.

In EMI-based ionic liquid system, the electrophilic polysulfide intermediates could react with the nucleophilic C2 carbon in the imidazole ring of EMI⁺ during cycles.^[6,18,19] Wang's group also discussed this issue in their previous research and

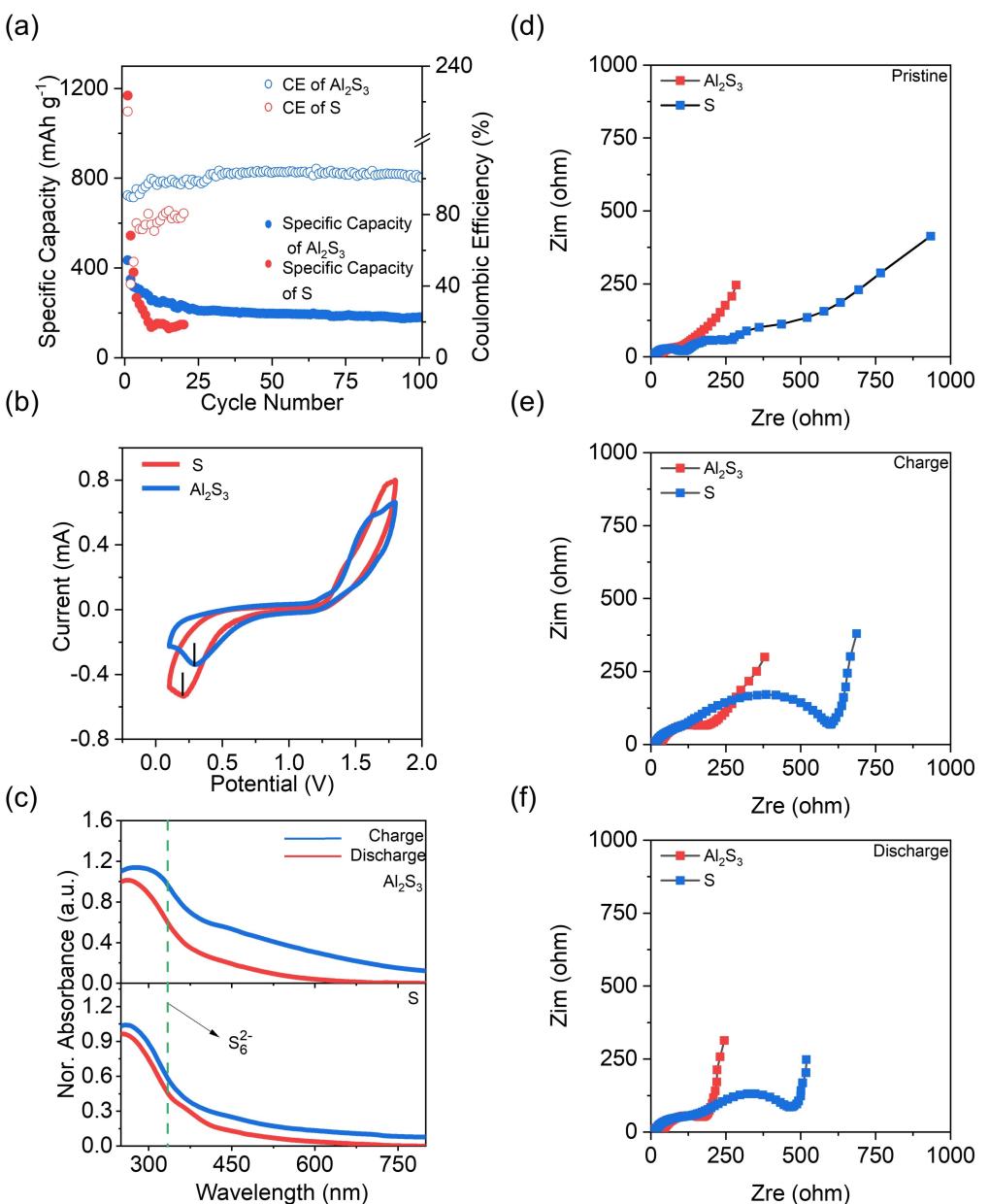


Figure 1. a) Specific capacity of S and Al_2S_3 cathodes as a function of cycle number under current density of 0.16 A g^{-1} . b) Cyclic voltammetry (CV) curves of S and Al_2S_3 cathode under scan rate of 0.05 mVs^{-1} . c) UV-vis spectra of the electrolyte after 5 discharge-charge cycles in the Al-S battery assembled with S and Al_2S_3 cathodes. D-f) Nyquist plots of the S cathode and Al_2S_3 cathode, respectively.

demonstrated that the by-products needed to be removed by ion-milling to avoid the influence of surface residual by-products.^[5] Thus, to further deconstruct the products generated when cycling the Al_2S_3 and S cathodes, Gas Cluster Ion Beam X-ray photoelectron spectroscopy (GCIB-XPS) depth profiles after different Ar sputtering durations are acquired (Figure 3). The etching rate is about 8 nm per minute by using silicon wafers as etching depth calibration. Thus, the thickness of the polysulfides on the electrode surface can be roughly estimated by multiplying the etching rate and the etching time.

At the discharged state, the signal of S^{2-} is hardly detected from the S cathode, and the valance state of sulfur species

increases along with the etching depth (Figure 3a). The characteristic peak signal of S^0 appears after 60 min of etching, indicating that a 480 nm thick polysulfides covering on the sulfur electrode, and the elemental S still remains even when the electrode is discharged. When the S cathode is charged to 1.8 V at a charged state, the signals of polysulfides are detected after 40 min of etching, equivalent to 320 nm of the polysulfides layer (Figure 3b). These results infer that only at most one-third of polysulfides convert to S^0 after charging. The electrochemical reaction of the S cathode is far from completion. On the other hand, the signals of S^0 are not observed from the Al_2S_3 cathode of charged state during the whole etching process (Figure 3c). Meanwhile, the peaks that corresponds to

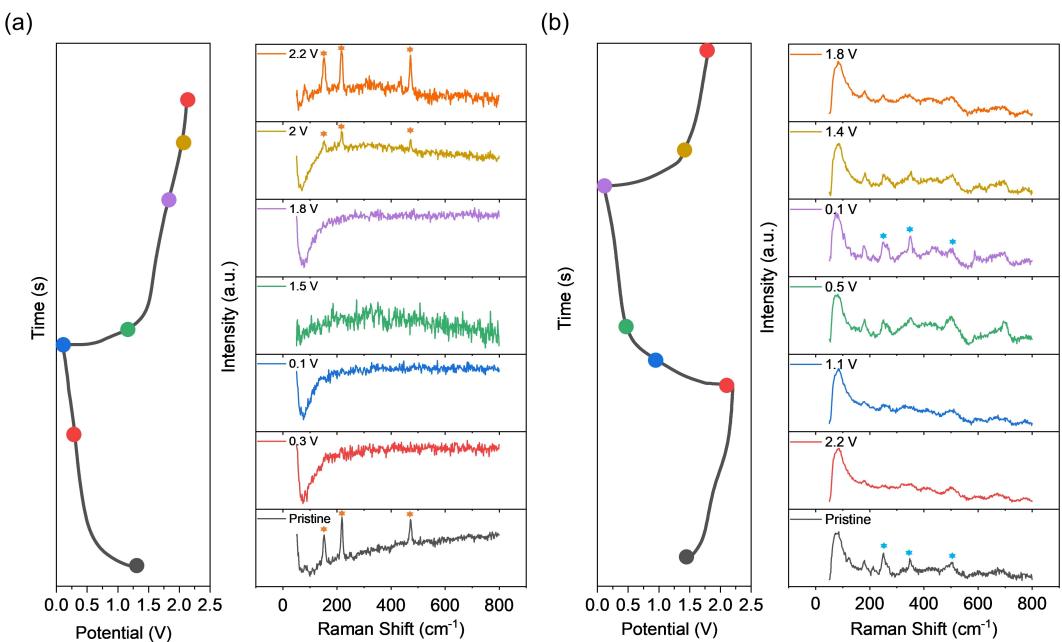


Figure 2. In-situ Raman spectra of a) the S cathode and b) Al₂S₃ cathode acquired at different time points during a cycle, respectively.

polysulfides can be observed even after etching for more than 50 min from the discharged Al₂S₃ cathode, illustrating better reversibility of Al₂S₃ cathode than that of S cathode (Figure 3d).

Based on the GCIB-XPS analysis, the conversion process of sulfur species in the S and Al₂S₃ cathodes are further schematically illustrated in Figure 3(e). For the S cathode, the conversion between polysulfides (S_n²⁻) and S⁰ has sluggish kinetic response as there is still a large amount of S⁰ remained under the polysulfides layers in the discharge and recharged electrodes. The insulating S⁰ layer that is sandwich between the current collector and polysulfides impedes electron and ion transport during charge and discharge, resulting poor reaction kinetics and reversibility. For the Al₂S₃ cathode, the electrochemical reactions occur only between the Al₂S₃ and polysulfides, avoids the generation of S⁰, thereby, demonstrating a higher reaction reversibility and hence a higher CE. Combined with the results of UV-vis spectrum performed on an electrolyte-soaked glass fiber separator that the main oxidation product of Al₂S₃ is S₆²⁻ at the charged state, therefore, the S_n²⁻ is the possible destination of oxidation. These advantages lead to an improved electrochemical performance of the Al₂S₃ cathode comparing to sulfur cathode.

From the results above, the Al₂S₃ cathode shows its potential as an ideal choice for the cathode in Al–S system. However, the application of Al₂S₃ cathode is also confronted by the low electric conductivity, which leads to low active material utilization and consequently, low specific capacity. In previous research, we demonstrate that carbon network can improve the conductivity of cathode and metallic cobalt have catalytic effect on the electrochemical conversion reactions of polysulfide towards sulfur containing cathode in Al–S battery.^[7,20] Thus, we prepare Co decorated nanostructured Al₂S₃–carbon composite (Al₂S₃@ZIF-67-C) to further show the advantages of

Al₂S₃ cathode in Al–S system (Figures S3–S6). The mass loading of active material in Al₂S₃@ZIF-67-C cathode is about 1 mg cm⁻², corresponding to 0.3 mg cm⁻² of sulfur in the cathode (normalized to the mass of sulfur). The preparation and characterization of cathode materials are listed in Supporting Information in detail.

As shown in Figure 4(a), the voltage hysteresis of Al₂S₃@ZIF-67-C is reduced to 0.8 V (measured at a current density of 0.16 mA g⁻¹) in comparison to 0.9 V for Al₂S₃ cathode and 1.3 V for sulfur cathode, respectively. The EIS results in Figure 4(b) also confirm the enhanced charge transfer process of Al₂S₃@ZIF-67-C cathode comparing with Al₂S₃ cathode and sulfur cathode, which exhibits a fastest charge transfer process. In Figure 4(a), the Al₂S₃@ZIF-67-C shows an initial capacity of ~560 mAh g⁻¹ corresponding to an active materials utilization of ~40%, which is much higher than that of pure Al₂S₃ cathode (~25%). Moreover, the Al₂S₃@ZIF-67-C exhibits an improvement of cycling performance over S electrode and Al₂S₃ cathode, which displays a capacity of 400 mAh g⁻¹ with CE > 95% at current density of 160 mA g⁻¹ after 100 cycles. We also prepared two types of cathodes assembled with S@ZIF-67-C and pure ZIF-67-C. The ZIF-67-C contributed to a negligible capacity of 30 mAh g⁻¹. It was interesting to note that the cathode made from a mixture of elemental S and ZIF-67-C exhibited an initial capacity of ~900 mAh g⁻¹ and decayed very rapidly to 250 mAh g⁻¹ after 20 cycles (Figure S7). These results indicated that the Al₂S₃ played a critical role in circumvention of the sluggish conversion of polysulfides to sulfur. Interestingly, we found the Al₂S₃@ZIF-67-C cathode can also be run under large current density of 1 A g⁻¹ (Figure S8), it preserves ~460 mAh g⁻¹ after 200 cycles with CE > 97%. Thus far most of Al–S works have been measured at low current density (that is, < 250 mA g⁻¹).^[5,6,9,21] It is believed that the poor reaction kinetics

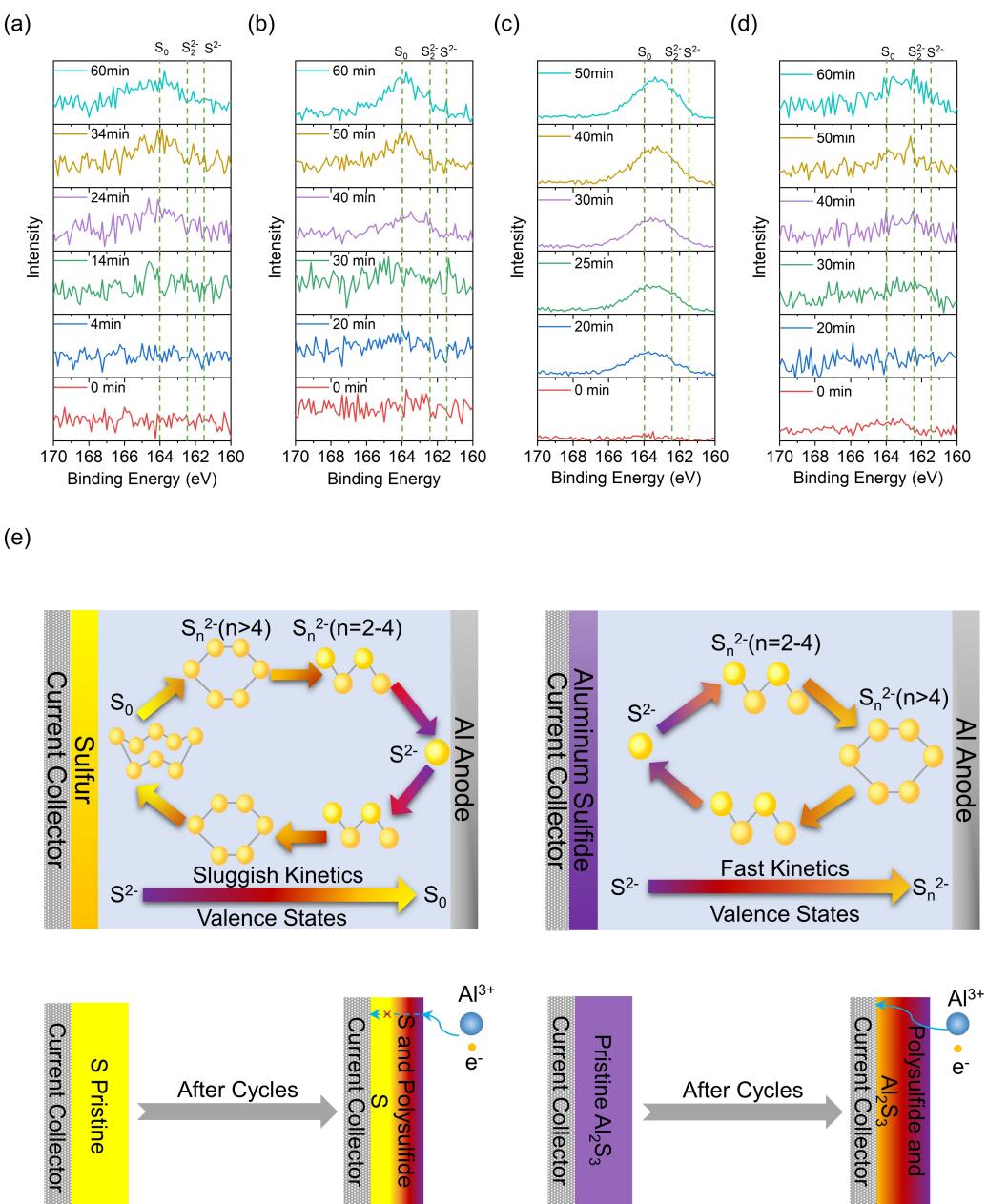


Figure 3. GCIB-XPS spectra of a) discharged and b) charged state of S cathode after 5 discharge-charge cycles. GCIB-XPS spectra of c) charged and d) discharged state of Al₂S₃ cathode after 5 discharge-charge cycles. e) The illustration of the process of sulfur species conversion in S and Al₂S₃ cathode.

caused by the slow solid-state sulfur conversion reaction should be the major reason. Further increase the cycling performance of Al-S battery under high current density requires systematic studies including the development of advanced conductive matrix, catalysts, and electrolyte. The combination of high reversible capacity, life-span and CE is critical to practical application, and our Al₂S₃@ZIF-67-C shows superior combination of gravimetric capacity, life-span and CE to state-of-the-art Al-ion battery (Figure S9). The enhanced reversibility in Al₂S₃@ZIF-67-C can be ascribed to the confinement of the nanostructured Al₂S₃ in the carbon matrix, which effectively facilitates sulfide oxidation kinetics owing to its ready electron

access, large electrochemical reaction area, and shortened Al³⁺ diffusion length.

In summary, we compared the electrochemical properties of Al₂S₃ and S as the initial material for the cathode of Al-S batteries. The Al₂S₃ cathode illustrates a much higher electrochemical reversibility by circumventing the sluggish conversion between polysulfides and elemental sulfur. The Al₂S₃ undergoes a faster solid (Al₂S₃) to liquid (polysulfides) conversion than that of the S cathode because of the insulating elemental S that remained between the current collector and polysulfide coating even at the fully discharge state. The Al₂S₃, when mixed with high porous carbon materials, presents an average CE > 95% and life span > 100 cycles with specific capacity of

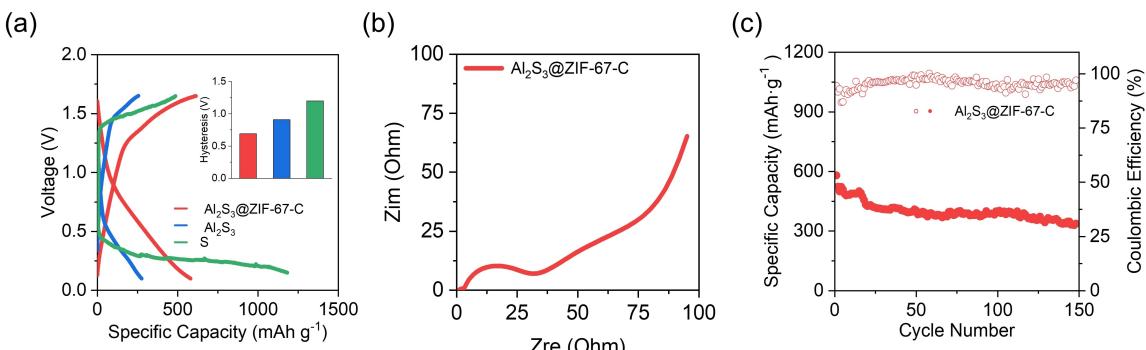


Figure 4. (a) Galvanostatic discharge–charge curves and their voltage hysteresis of the Al–S battery composed of Al₂S₃@ZIF-67-C, Al₂S₃ and S cathode at a current density of 160 mA g⁻¹, respectively. (b) EIS image of Al₂S₃@ZIF-67-C cathode. (c) Cycle performance of Al₂S₃@ZIF-67-C cathode under current density of 160 mA g⁻¹.

>400 mA h g⁻¹. This work sheds light on a new strategy to develop low-cost and viable Al–S batteries for electrochemical energy storage and energy conversion systems.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords: aluminum sulfide · aluminum-sulfur battery · Coulombic efficiency · GCIB-XPS · in-situ Raman spectra

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