

Enhanced Cycling Stability of All-Solid-State Lithium–Sulfur Battery through Nonconductive Polar Hosts

Tianwei Jin, Keyue Liang, Jeong-Hoon Yu, Ting Wang, Yihan Li, Tai-De Li, Shyue Ping Ong, Jong-Sung Yu,* and Yuan Yang*



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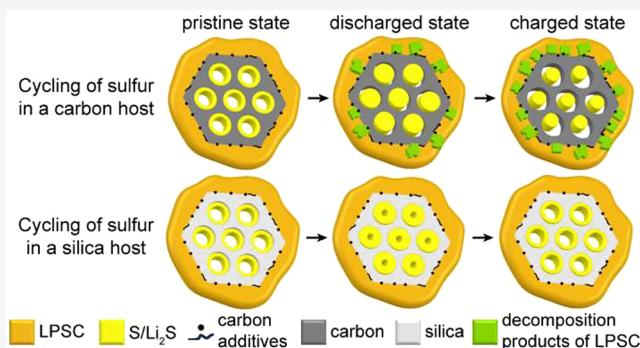
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ABSTRACT: All-solid-state lithium–sulfur batteries (ASSLSBs) are promising next-generation battery technologies with a high energy density and excellent safety. Because of the insulating nature of sulfur/Li₂S, conventional cathode designs focus on developing porous hosts with high electronic conductivities such as porous carbon. However, carbon hosts boost the decomposition of sulfide electrolytes and suffer from sulfur detachment due to their weak bonding with sulfur/Li₂S, resulting in capacity decays. Herein, we propose a counterintuitive design concept of host materials in which nonconductive polar mesoporous hosts can enhance the cycling life of ASSLSBs through mitigating the decomposition of adjacent electrolytes and bonding sulfur/Li₂S steadily to avoid detachment. By using a mesoporous SiO₂ host filled with 70 wt % sulfur as the cathode, we demonstrate steady cycling in ASSLSBs with a capacity reversibility of 95.1% in the initial cycle and a discharge capacity of 1446 mAh/g after 500 cycles at C/5 based on the mass of sulfur.

KEYWORDS: all-solid-state lithium–sulfur batteries, nonconductive hosts, polarity, silica, sulfur detachment



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Lithium–sulfur batteries (LSBs) have attracted substantial attention as a promising next-generation energy storage solution due to the ultrahigh specific capacity, high natural abundance, and environment friendliness of sulfur.^{1–3} However, the development of LSBs with liquid electrolytes is hindered by several key challenges. First, organic solvents in LSBs, which are highly flammable, dissolve intermediate lithium polysulfides, and the consequent shuttle effect results in low Coulombic efficiencies (CE) and short lifespan.⁴ Second, the electrically and ionically insulating nature of sulfur and Li₂S leads to sluggish cathode kinetics.⁵ Third, the large volume change of ~80% from sulfur to Li₂S causes them to lose contact with carbon additives and electrolytes, resulting in a substantial capacity loss.^{6,7} Recently, all-solid-state lithium–sulfur batteries (ASSLSBs) have attracted increasing attention as a potential solution.^{8,9} By adopting inorganic sulfide solid-state electrolytes (SSEs), such as Li₆PS₅Cl (LPSC) and Li₁₀GeP₅S₁₂ (LGPS) with high ionic conductivities (10^{-3} – 10^{-2} S cm⁻¹) and favorable mechanical properties, the shuttle effect can be eliminated together with greatly elevated battery safety.^{10–12}

To enhance the reaction kinetics in LSBs, high-surface-area conductive carbon hosts are often used.^{13,14} Unfortunately, sulfide electrolytes in ASSLSBs usually have narrow electrochemical stability windows, and Meng et al. have revealed that the oxidation and reduction decomposition of sulfides will be

accelerated when they are exposed to electronic conductors.^{15–17} Therefore, though nanoporous carbon hosts enhance the electron transfer to sulfur, their high electronic conductivities could also boost the decomposition of adjacent sulfide SSEs into insulating components (Figure 1a), which consequently impede the ion transport for the S/Li₂S redox.¹⁸ As a result, the cathode kinetics deteriorates over successive cycles and cell capacities accordingly diminish.^{19,20} Alternatively, various nanoporous oxides are nonconductive so that they are expected to suppress the decomposition of neighboring SSEs and retain the cycling stability, and the electronic conduction could be fulfilled by other strategies, such as reduced sulfur sizes and proper carbon additives with low specific surface areas (Figure 1b).

The other potential advantage of oxides over carbon as hosts for sulfur is their strong bonding with active materials. Carbon hosts have weak bonding with sulfur and Li₂S due to its nonpolarity, and previous works showed the active material would detach from the carbon hosts upon volume expansion

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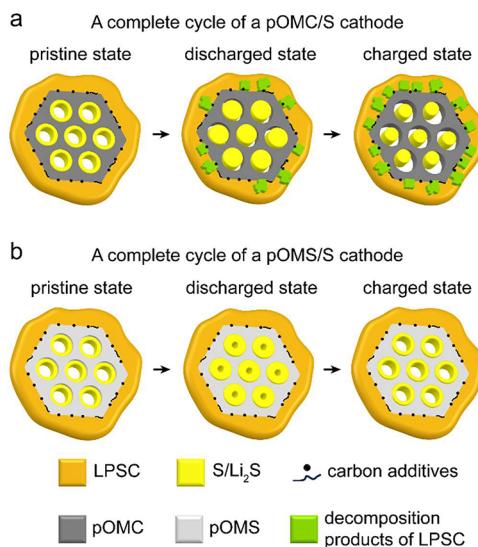


Figure 1. Schematics of cathodes in ASSLSBs with (a) a carbon host (platelet ordered mesoporous carbon, pOMC) and (b) a non-conductive polar host (platelet ordered mesoporous silica, pOMS), respectively. The highly conductive carbon host promotes the decomposition of adjacent electrolytes, and sulfur filled in the carbon host suffers from the detachment issue during cycling.²¹ In contrast, the polar silica host bonds strongly to the active materials, and its low electronic conductivity accordingly suppresses the electrolyte decomposition.

and contraction (**Figure 1a**), leading to loss in conduction pathway and capacity.^{21–24} In contrast, oxides are found to have strong chemisorption toward lithium sulfides out of their high polarities, making them good candidates as sulfur hosts or separator coatings in the LSBs with liquid electrolytes to mitigate the shuttle effect.^{25–32} Such superior bindings indicate that oxide hosts could potentially solve the sulfur detaching issue in carbon hosts and allow ASSLSBs to achieve better cycling performance.

Based on these hypotheses, herein, we propose a counter-intuitive concept that nonconductive polar hosts can substantially enhance the performance of ASSLSBs. As a demonstration, we synthesized a platelet ordered mesoporous silica (pOMS, **Figure 1b**) and a platelet ordered mesoporous carbon (pOMC, **Figure 1a**) as model systems for insulating oxide hosts and conductive carbon hosts, respectively. The platelet-shaped OMS and OMC feature structural advantages over conventional rod-like ones, such as much shorter mesopore nanochannels for homogeneous sulfur loading and shorter pathways for ions and electrons during cycling.³³

The pOMS/S composite with 70 wt % sulfur showed an excellent cycling performance, which presented a capacity reversibility of 95.1% in the initial cycle and delivered a steady discharge capacity of 1446 mAh/g after 500 cycles at C/5 at room temperature. In contrast, the pOMC/S composite with 70 wt % sulfur showed a poor capacity reversibility of 75.7% in the initial cycle, probably due to electrolyte decomposition and sulfur detachment, with a fast capacity decay from 1430 mAh/g in cycle 1 to 649 mAh/g in cycle 50 at C/10 at room temperature. Further postcycling characterizations based on FIB-EDS mapping and XPS revealed that the insulating SiO₂ host significantly suppresses solid electrolyte decomposition and enhances sulfur/host binding. With all of those findings, this work unveiled an unexpected strategy to accelerate the

ASSLSB development to achieve high-energy-density lithium batteries.

To study the effects of nonconductive polar hosts in ASSLSBs, pOMC and pOMS particles with similar morphology and pore structures were synthesized via a precipitation method based on our previous reports.^{26,34} Sulfur then infiltrated into the open mesopore nanochannels of the hosts by sulfur vapor deposition in a weight ratio of 70:30 (sulfur:host), named as pOMC/S70 and pOMS/S70. As scanning electron microscopy (SEM) images showed, pOMC/S70 (**Figure 2a**) and pOMS/S70 (**Figure 2e**) both have a thin hexagonal prism morphology with a thickness of ~250 nm and an edge length of 500–1000 nm. Transmission electron microscopy (TEM) images showed the open mesopores through the prism thickness of pOMC/S70 (**Figure 2b**) and pOMS/S70 (**Figure 2f**), and the corresponding high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) element mapping images confirmed the uniform sulfur infusion in pOMC/S70 (**Figure 2c,d**) and pOMS/S70 (**Figure 2g,h**).

X-ray diffraction (XRD) patterns in **Figure 2i** further showed that pOMC and pOMS were amorphous,^{35,36} and sulfur was amorphous or partially crystalline in pOMC/S70 and pOMS/S70. In addition, small-angle XRD results of pOMS (**Figure S1a**) and pOMC (**Figure S1b**) indicated the presence of a strong (100) and two weak (110) and (200) diffraction peaks, verifying a 2D hexagonal p6mm pore structure.³⁷ The corresponding spacing between pores is ~9.8 nm in both pOMS and pOMC. The diminishment of the (100), (110), and (200) peaks after the sulfur infusion denoted that sulfur indeed infiltrated into the pores.

In terms of pore structures, Brunauer–Emmett–Teller (BET) analysis (**Figure 2j**) showed that pOMC and pOMS have specific surface areas of 1643 and 864 m²/g, respectively. The corresponding pore size distributions centered at 4.2 nm for pOMC and 7.3 nm for pOMS, respectively (**Figure S2**). After loading with sulfur, the surface area of pOMC/S70 and pOMS/S70 sharply decreased to 45 and 15 m²/g (**Figure 2j**), respectively, with very low pore volumes (**Figure S2**), consistent with XRD confirmation. In addition, the 70 wt % sulfur loading was confirmed by thermogravimetric analysis (TGA) in **Figure S3**, a high value that is needed to increase the energy density of ASSLSBs.

Though pOMS/S70 and pOMC/S70 present similar physical properties, they behave chemically distinctly in terms of the sulfur interaction, as revealed by X-ray photoelectron spectroscopy (XPS). pOMC/S70 displayed almost the identical S 2p spectrum as elemental sulfur, deconvoluted into a spin–orbit doublet (S 2p_{1/2} at 164.9 eV and 2p_{3/2} at 163.7 eV) with an intensity ratio of 1:2 (**Figure 2k**), indicating no or weak interaction between sulfur and the carbon host.³⁸ In contrast, the S 2p spectrum of pOMS/S70 in **Figure 2l** was remarkably different, and the new peaks at 163.2 and 164.4 eV were identified to be Si–S bond,^{39,40} and such interaction with sulfur was also observed in other polysulfide trapping hosts.^{41,42}

To elucidate the interfacial interactions between sulfur/Li₂S and the host, density functional theory (DFT) calculations were performed. In **Figure 2m**, the calculated adhesion energy for the C–Li₂S interface is −0.25 J/m² with a negligible interaction tendency observed at the interface. Conversely, the adhesion energy for the SiO₂–Li₂S interface is −0.93 J/m² with Si–S and Li–O distances of 0.22 and 0.20 nm,

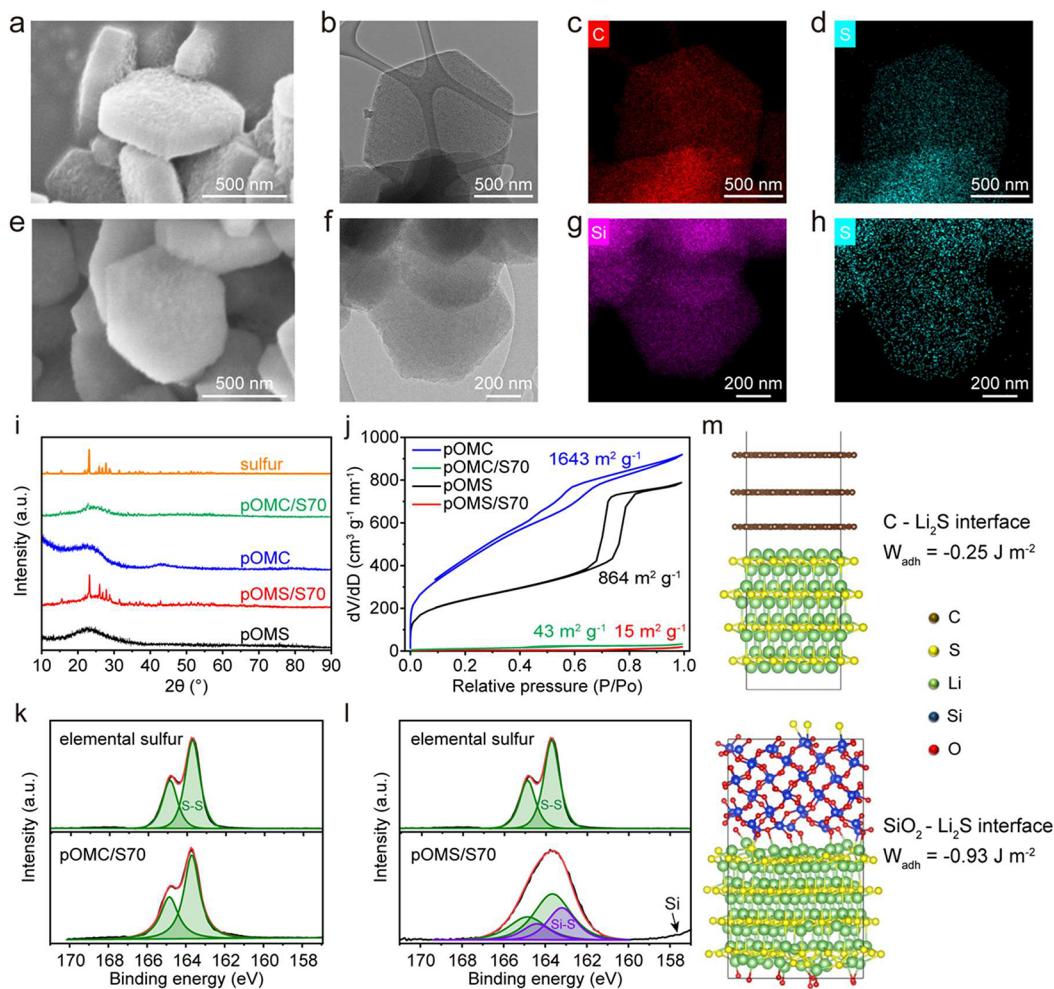


Figure 2. Physical and chemical properties of pOMS and pOMC hosts. (a–d) pOMC/S70: (a) SEM image, (b) TEM image, and (c, d) corresponding HAAD-STEM element mapping of (c) C and (d) S. (e–h) pOMS/S70: (e) SEM image, (f) TEM image, (g, h) corresponding HAAD-STEM element mapping of (g) Si and (h) S. (i) XRD patterns of elemental sulfur, pOMC, pOMC/S70, pOMS, and pOMS/S70. (j) Nitrogen adsorption/desorption isothermal profiles of pOMC, pOMC/S70, pOMS, and pOMS/S70. (k, l) Deconvoluted high-resolution S 2p XPS spectra of (k) pOMC/S70 and (l) pOMS/S70 with elemental sulfur as the reference. (m) DFT calculation of the interface structures of SiO₂–Li₂S and C–Li₂S.

respectively, indicating strong interaction and even bond formation. The distinct adhesion energies are in consistent with prior reports that Li₂S bonds weakly with carbon but strongly with silica, thereby implying the potential of silica hosts to mitigate sulfur detachment issues observed with carbon hosts.^{22,25,26,43}

Regarding interactions with sulfur (Figure S4), the adhesion energy for the C–S interface is determined to be 9.15 J/m². This positive and high value indicates a significant challenge for sulfur wetting on the carbon surface. On the other hand, the adhesion energy for the SiO₂–S interface is only 0.50 J/m². Although it remains positive, the significantly lower value suggests that sulfur exhibits a much stronger affinity for SiO₂ than for carbon. These DFT calculations suggest that silica hosts are capable of better fixing sulfur and Li₂S spatially to facilitate their robust contacts with SSEs and carbon additives during volume changes, thus enhancing the sulfur utilization and cycling stability of ASSLSBs relative to carbon hosts.

The discrepancy between the positive adhesion energy value for the SiO₂–S contact in the DFT calculation and the XPS result, indicating a strong Si–S bond, may be attributed to the presence of defects, such as oxygen vacancies, in silica. These

defects in practical scenarios, particularly on the surface, could facilitate the formation of Si–S bonds. However, in DFT calculations, interfaces are typically modeled between perfect materials at 0 K without any vacancies, which does not favor bond formation and can account for the positive adhesion energy value.

To illustrate the impact of polarity and nonconductivity of sulfur hosts on the electrochemical performances in ASSLSBs, 40 wt % pOMS/S70 and pOMC/S70 were respectively mixed with 40 wt % LPSC and 20 wt % carbon additives (8 wt % C65 carbon black, 8 wt % vapor-grown carbon fiber, and 4 wt % carbon nanotube) to build cathodes. The integration of such three carbon additives provided a sufficient electronic conduction network with a limited surface area to reduce electrolyte decomposition.¹⁵ The cathode was then paired with an LPSC electrolyte and an Li–In anode and cycled within 1.2–3.0 V vs Li/Li⁺. The corresponding cycling performance is displayed in Figure 3a, and the voltage profiles are shown in Figure 3b,c. Although pOMC/S70 exhibited a high specific capacity of 1430 mAh/g in the initial discharge at C/10 with 1C defined as 1000 mA/g for all cells, only 1082 mAh/g (75.7%) was recovered in the subsequent charge (Figure 3b).

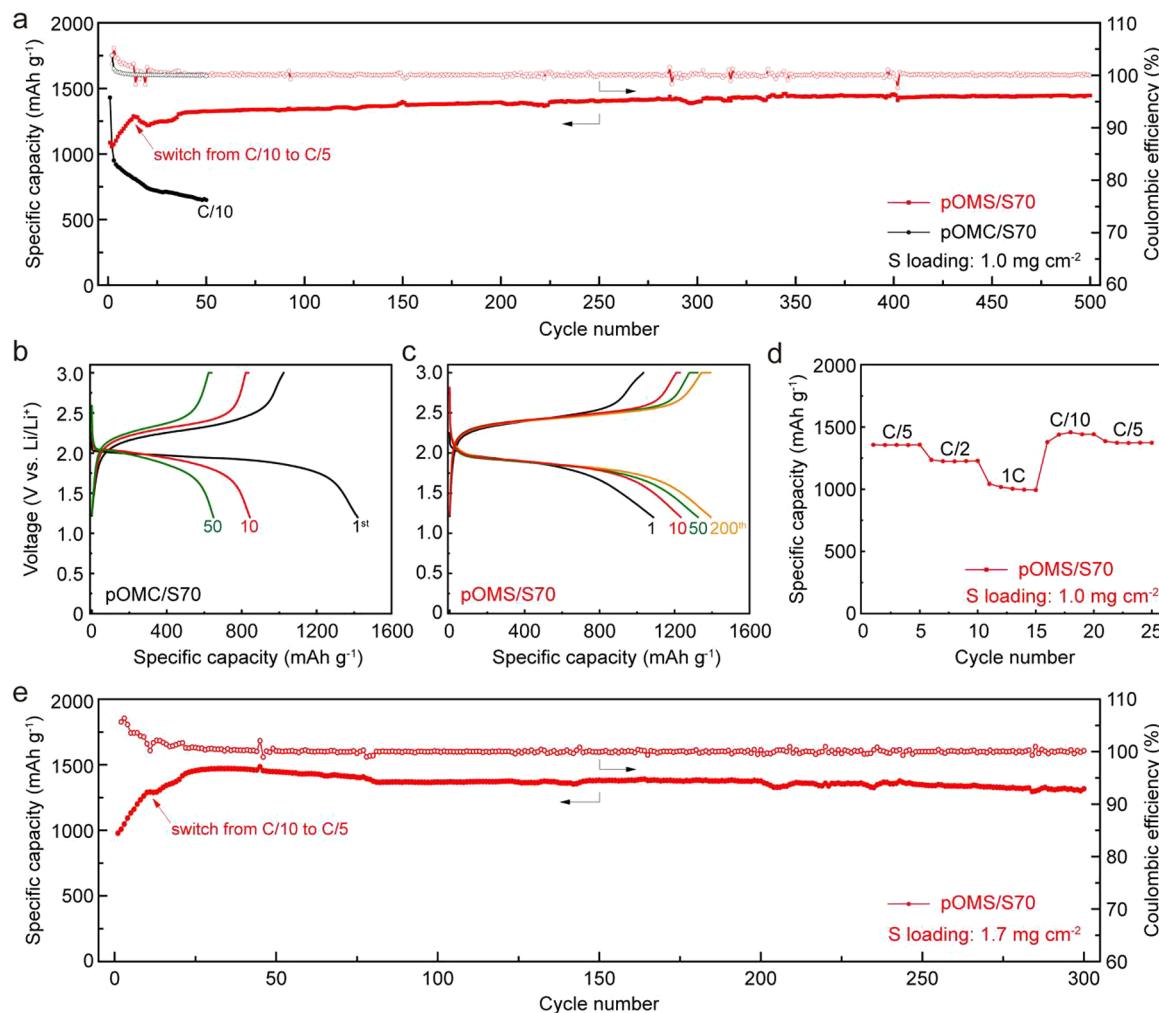


Figure 3. Electrochemical performance of all-solid-state Li–S batteries with a carbon host and a silica host, respectively. (a) Cycling performance of Li–In|LPSC|pOMS/S70 and Li–In|LPSC|pOMC/S70 cells with a mass loading of $1\text{ mg}/\text{cm}^2$ sulfur and (b, c) their corresponding voltage profiles for (b) pOMC/S70 and (c) pOMS/S70. (d) Rate performance of a Li–In|LPSC|pOMS/S70 cell after 150 cycles (see Figure S6 for the full cycling performance). (e) Cycling performance of a Li–In|LPSC|pOMS/S70 cell with a higher mass loading of $1.7\text{ mg}/\text{cm}^2$ sulfur. All cells underwent an initial cycle at C/20, followed by subsequent cycles at C/10 or C/5 with a constant-voltage step with a current cutoff of C/20. 1C is defined as 1000 mA/g for all cells based on the average specific capacity in the first cycle. All of the cells were cycled at room temperature.

Such a low reversibility could be caused by the electrolyte decomposition and sulfur detachment from the nonpolar carbon host, leading to a low sulfur utilization in the following cycles.²¹

In contrast, pOMS/S70 delivered a specific capacity of 1087 mAh/g in the initial discharge, and 1034 mAh/g was recovered in the subsequent charge (Figure 3c), corresponding to a reversibility of 95.1%. Such a high specific capacity also indicates that a conductive host is not the only option for ASSLSBs, and reasonable electronic transport can be achieved by a proper amount of carbon additives and reduced sulfur nanoparticle sizes to shorten the conducting pathway. As pOMS/S70 shares analogous morphological properties with pOMC/S70, the substantial enhancement in initial reversibility should not arise from morphological difference but could be attributed to the suppressed electrolyte decomposition and robust bonding of polar silica with sulfur/Li₂S, ensuring well-preserved electronic and ionic conduction of sulfur.

In addition to the improved reversibility in the first cycle, pOMS/S70 also demonstrated significantly enhanced long-term cycling performance compared to that of pOMC/S70.

pOMS/S70 exhibited a distinct activation stage where the specific capacity substantially increased from 1087 mAh/g in the first cycle to 1318 mAh/g in the 40th cycle and to 1446 mAh/g in the 500th cycle at C/5, whose cycling stability is among the best results in reported ASSLSB works (Table S2). In contrast, the specific capacity of pOMC/S70 rapidly decreased from 1430 mAh/g to 649 mAh/g after 50 cycles at C/10. Moreover, sulfur cathodes without nanoporous hosts showed markedly reduced specific capacities of less than 300 mAh/g (Figure S5), which arises from compromised kinetics and sulfur utilization due to a smaller contact area with LPSC/carbon and a longer transport pathway.²⁰

We think that the gradual capacity increase in pOMS/S70 arose from the increasing contacts between S/Li₂S and carbon additives/SSEs during volume expansion and contraction in cycling. Similar behaviors have also been reported in other works on ASSLSBs.^{44,45} Moreover, the insulating nature of SiO₂ also minimized the decomposition of sulfide electrolytes, and the strong bonding between S/Li₂S and SiO₂ anchored the active materials to avoid detachment from the hosts and the loss of conducting contact. On the contrary, the sulfur

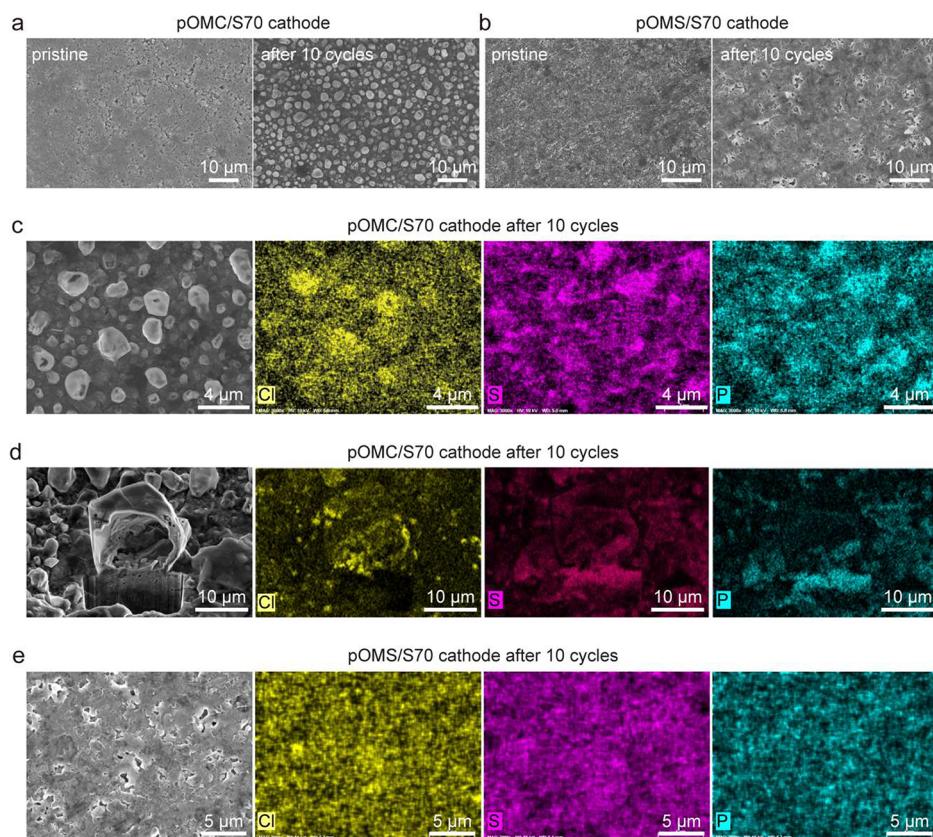


Figure 4. Morphological and chemical evolution of pOMS/S70 and pOMC/S70 cathodes. (a) SEM images of the pOMC/S70 cathode before and after 10 cycles. (b) SEM images of the pOMS/S70 cathode before and after 10 cycles. (c) SEM image of the pOMC/S70 cathode after 10 cycles and the corresponding EDS elemental mapping of Cl, S, and P. (d) FIB-SEM image of the pOMC/S70 cathode after 10 cycles and the corresponding EDS elemental mapping of Cl, S, and P. (e) SEM image of the pOMS/S70 cathode after 10 cycles and the corresponding EDS elemental mapping of Cl, S, and P. All cathodes after cycling were retrieved from all-solid-state cells at 3 V vs Li/Li⁺ with a mass loading of 1 mg/cm² sulfur, cycled at C/20 for the first cycle and at C/10 for the following nine cycles at room temperature.

utilization in the carbon host would deteriorate at each cycle due to the decomposition of electrolyte and detachment from carbon, resulting in capacity decay.

Besides the steady long-term cycling performance, pOMS/S70 also exhibited satisfying rate performance at current rates from C/10 to 1 C (Figure 3d). pOMS/S70 exhibited an average discharge capacity of 1356 mAh/g at C/5, 1228 mAh/g at C/2, 1011 mAh/g at 1 C. The capacities recovered to 1431 mAh/g at C/10 and 1375 mAh/g at C/5, highlighting its outstanding rate performance and stability (see Figure S6 for the full cycling performance). It further validates that the electronic conduction can be satisfied in an insulating host with reduced sulfur sizes and a limited amount of carbon additives.

Based on all benefits of silica hosts shown above, we further demonstrated a long cycle life with a higher loading of 1.7 mg/cm² sulfur. As shown in Figure 3e, the cathode delivered a specific capacity of 978 mAh/g in the first cycle, which increased to 1467 mAh/g after 40 cycles and was maintained at 1317 mAh/g after 300 cycles with an average CE of 100.3%. To demonstrate the reproducibility of our proposed design, more Li-InLPSCl-pOMS/S70 cells with similar cycling performance are shown in Figure S7. Cycling performance of a cathode with a higher sulfur content of 35 wt % is also demonstrated (Figure S8), illustrating that the cycling is still stable with higher sulfur contents thanks to the nonconductive polar host. All cycling results above show that nonconductive

polar sulfur hosts can enable the excellent performance of ASSLSBs.

It is worth mentioning that though nonconductive polar hosts could allow a better cycling life via suppressing SSE decomposition and sulfur detachment, the cathode kinetics was inferior to those with a conductive carbon host, as the voltage hysteresis of pOMS/S70 was larger than pOMC/S70 (0.59 V vs 0.36 V in the initial cycle, Figure 3b,c). It suggests that the host conductivity should be carefully designed to be moderate to simultaneously meet the criteria of both power capability and cycling life of ASSLSBs, which may also allow higher sulfur contents for high energy densities.

To elucidate the origin of the distinct electrochemical performance of pOMS/S70 and pOMC/S70 cathodes, SEM and energy dispersive spectroscopy (EDS) were employed to analyze their morphological and chemical changes after cycling. As shown in Figure 4a, numerous micrometer-sized agglomerates appeared in the pOMC/S70 cathode after 10 cycles, which was also reported in other ASSLSB studies.⁴⁶ Further EDS mapping elucidated that P signals overlapped well with S but complementary to Cl, whose signals concentrated in the agglomerate areas (Figure 4c). Such a separation of Cl from P and S agreed well with the previous studies that LPSC decomposes into LiCl, P₂S₅, Li₂S, P during cycling, indicating a severe LPSC decomposition in the pOMC/S70 cathode after 10 cycles.¹⁷ The fine structure of the agglomerates was further revealed by focused ion beam (FIB) milling. As shown in

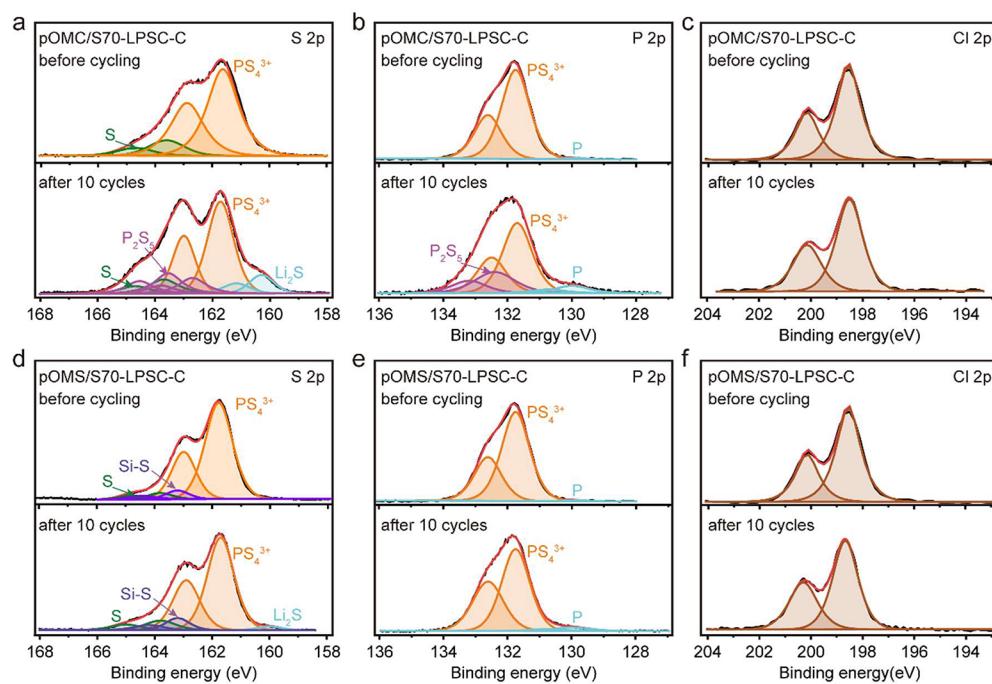


Figure 5. Deconvoluted high-resolution XPS spectra of the pOMS/S70 and pOMC/S70 cathodes. (a) S 2p, (b) P 2p, and (c) Cl 2p XPS spectra of the pOMC/S70 cathodes before and after 10 cycles. (d) S 2p, (e) P 2p, and (f) Cl 2p XPS spectra of the pOMS/S70 cathodes before and after 10 cycles. All cathodes after cycling were retrieved from all-solid-state cells at 3 V vs Li/Li⁺ with a mass loading of 1 mg/cm² sulfur, cycled at C/20 for the first cycle and at C/10 for the following nine cycles at room temperature.

Figure 4d, the agglomerates had a shell structure with the EDS mapping of P overlapping with S but complementary to Cl, and scattered $\sim 1 \mu\text{m}$ Cl-rich domains were observed. Such results suggested that the LPSC decomposition, which would expand in volume, in the pOMC/S70 cathode might cause chemical redistribution, and the electronic and ionic conduction pathways of sulfur could be blocked when pOMC/S70 particles were surrounded by the insulating LiCl and P_2S_5 .^{16,47}

In contrast, LPSC decomposition was well alleviated in the pOMS/S70 cathode. As shown in Figure 4b, the morphology of the pOMS/S70 cathode remained intact after 10 cycles, and the corresponding EDS elemental mapping in Figure 4e also showed a uniform distribution of Cl, P, and S without separation, indicating that the LPSC remained essentially intact. The stable chemical distribution in the cathode ensured robust electronic and ionic conduction in the cathode and excellent electrochemical performance.

X-ray photoelectron spectroscopy (XPS) was further employed to elucidate chemical changes of cathodes in ASSLSBs. Figure 5a–c shows the XPS spectra of S, P, and Cl in pOMC/S70 cathodes before cycling and after 10 cycles. The S 2p spectrum of the pristine pOMC/S70 cathode had peaks at 161.8 and 163.70 eV with their spin-orbit doublets ($\text{S } 2p_{1/2}$ and $2p_{3/2}$), corresponding to the PS_4^{3+} in LPSC electrolyte and elemental sulfur, respectively (Figure 5a).^{38,48} However, a strong new peak at 160.1 eV with its spin-orbit doublet from Li_2S and appreciable new peaks at 162.70 and 163.50 with their spin-orbit doublets from P_2S_5 appeared at the charged state after 10 cycles,^{48,49} which were from the irreversible lithiation of sulfur during cycles and LPSC decomposition.²¹ The P 2p spectrum of the pristine pOMC/S70 cathode had a peak at 131.70 eV with its spin-orbit doublet from the PS_4^{3+} in LPSC and very weak elemental P

peak at 130.0 eV which came from the electrolyte impurity (Figure 5b). After 10 cycles, significant peaks at 132.40 eV from P_2S_5 and 130.0 eV from P with their spin-orbit doublets were observed, which were the LPSC decomposition products.^{15,48} The Cl 2p spectrum had no change after 10 cycles because the Cl bonding energy in LPSC and LiCl were the same (Figure 5c).⁵⁰

On the contrary, the LPSC degradation in pOMS/S70 cathodes after 10 cycles was significantly suppressed. As shown in Figure 5d, the S 2p spectrum of the pOMS/S70 cathode remained basically the same as the pristine state except for a small peak of Li_2S that might come from the irreversible lithiation of the sulfur during cycling. The P 2p spectrum in Figure 5e also showed that the only decomposition product was a small amount of elemental P at a much lower level than that of the pOMC/S70 cathode after cycling. The Cl 2p spectrum also remained the same as shown in Figure 5f.

Besides after 10 cycles, we further studied the XPS spectra of the pOMS/S70 cathode after 300 cycles (Figure S9), which showed that the electrolyte decomposition catalyzed by the carbon additives is substantially slower and less than the decomposition catalyzed by pOMC/S70. This indicates that an appreciable amount of the electrolyte decomposition in the pOMC/S70 cathode is caused by the carbon mesoporous hosts and thus is adjacent to S/ Li_2S , which will severely block the conduction pathway of Li⁺ and e⁻ to S/ Li_2S and lead to capacity decay. In contrast, decomposition from carbon additives (e.g., VGCF) is less and has a smaller impact on the conduction pathways. In summary, the XPS results above illustrated that the electrolyte decomposition was successfully restrained by using a nonconductive SiO₂ host compared to carbon hosts.

In conclusion, we innovatively proposed a new design concept for the sulfur hosts in ASSLSBs that low conductivities

could suppress the adjacent electrolyte decomposition and high polarities could mitigate the sulfur detachment issue, thereby enhancing the cycling performance of ASSLSBs. The adequate electronic conduction of sulfur can be realized with reduced sulfur nanoparticle sizes and proper choices of carbon additives rather than merely relying on conductive hosts. Accordingly, a silica host for sulfur which has strong bonding with sulfur and Li₂S was demonstrated. The SiO₂/S composite delivered a discharge capacity of 1087 mAh/g in cycle 1, 95.1% of which was reversible in the following charging and 1446 mAh/g after 500 cycles at C/5. In contrast, C/S composite delivered a discharge specific capacity of 1430 mAh/g in cycle 1, 75.7% of which was reversible in the following charging, and only 649 mAh/g after 50 cycles at C/10. The excellent performance of the SiO₂/S composite is attributed to reduced electrolyte decomposition and better attachment between SiO₂ and active materials, which are confirmed by FIB-SEM-EDS and XPS analysis. This study unveils that nonconductive polar hosts could be attractive for ASSLSBs because of the mitigated electrolyte decomposition, stabilized cycling, and satisfying power density.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.4c01210>.

Experimental section, additional figures of material characterizations, DFT calculations, and cycling performance (Figures S1–S9), and tables of DFT information and cycling performance comparison with literature (Tables S1 and S2) ([PDF](#))

■ AUTHOR INFORMATION

Corresponding Authors

Jong-Sung Yu – Department of Energy Science and Engineering, Daegu Gyeongbuk Institute of Science and Technology, Daegu 42988, Republic of Korea; Energy Science and Engineering Research Center, Daegu Gyeongbuk Institute of Science and Technology, Daegu 42988, Republic of Korea; orcid.org/0000-0002-8805-012X; Email: jsyu@dgist.ac.kr

Yuan Yang – Program of Materials Science and Engineering, Department of Applied Physics and Applied Mathematics, Columbia University, New York, New York 10027, United States; orcid.org/0000-0003-0264-2640; Email: yy2664@columbia.edu

Authors

Tianwei Jin – Program of Materials Science and Engineering, Department of Applied Physics and Applied Mathematics, Columbia University, New York, New York 10027, United States; orcid.org/0000-0002-4355-5474

Keyue Liang – Program of Materials Science and Engineering, Department of Applied Physics and Applied Mathematics, Columbia University, New York, New York 10027, United States

Jeong-Hoon Yu – Department of Energy Science and Engineering, Daegu Gyeongbuk Institute of Science and Technology, Daegu 42988, Republic of Korea

Ting Wang – Department of NanoEngineering, University of California, San Diego, La Jolla, California 92093-0448, United States

Yihyan Li – Program of Materials Science and Engineering, Department of Applied Physics and Applied Mathematics, Columbia University, New York, New York 10027, United States

Tai-De Li – Nanoscience Initiative at Advanced Science Research Center, Graduate Center of the City University of New York, New York, New York 10031, United States; Department of Physics, City College of New York, City University of New York, New York, New York 10031, United States

Shyue Ping Ong – Department of NanoEngineering, University of California, San Diego, La Jolla, California 92093-0448, United States

Complete contact information is available at: <https://pubs.acs.org/10.1021/acs.nanolett.4c01210>

Notes

The authors declare no competing financial interest.

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