



Formulating energy density for designing practical lithium–sulfur batteries

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The lithium–sulfur (Li–S) battery is one of the most promising battery systems due to its high theoretical energy density and low cost. Despite impressive progress in its development, there has been a lack of comprehensive analyses of key performance parameters affecting the energy density of Li–S batteries. Here, we analyse the potential causes of energy loss during battery operations. We identify two key descriptors (R_{weight} and R_{energy}) that represent the mass- and energy-level compromise of the full-cell energy density, respectively. A formulation for energy density calculations is proposed based on critical parameters, including sulfur mass loading, sulfur mass ratio, electrolyte/sulfur ratio and negative-to-positive electrode material ratio. The current progress of Ah-level Li–S batteries is also summarized and analysed. Finally, future research directions, targets and prospects for designing practical high-performance Li–S batteries are proposed.

Lithium-ion batteries (LIBs) are the dominant energy storage technology to power portable electronics and electric vehicles. However, their current energy density and cost cannot satisfy the ever-growing market demand^{1–3}. The Battery500 Consortium has proposed the need to reach a cell-level specific energy of 500 Wh kg^{−1} with a pack-level cost lower than US\$100 (kWh)^{−1} for electric vehicles⁴. Exploring new battery chemistries beyond conventional LIB systems is therefore necessary and urgent^{5,6}.

Table 1 compares the gravimetric energy density, corresponding driving distance and cost for several commonly used rechargeable battery systems, such as lead–acid, nickel–cadmium (Ni–Cd), nickel–metal hydride (Ni–MH) batteries, LIBs, advanced LIBs and lithium–sulfur (Li–S) batteries. Current LIBs with a cell-level energy density of 150–250 Wh kg^{−1} offer a drive range of 300 to 600 km for electric vehicles (for example, LIBs in Tesla electric vehicles have cell-level energy densities of ~250 Wh kg^{−1}), which is insufficient for achieving a 500-mile drive distance per charge with a reasonable battery pack size to alleviate range anxiety. This is due to the relatively low capacity (≤ 220 mAh g^{−1}) and heavy weight of conventional lithium–transition metal oxide (LMO) cathodes, which limit the energy density of Li metal–LMO full cells (future LIBs) to be hardly higher than 500 Wh kg^{−1}. Owing to multi-electron redox reactions of the sulfur cathode, Li–S batteries afford a high theoretical specific energy of 2,567 Wh kg^{−1} and a full-cell-level energy density of ≥ 600 Wh kg^{−1}. Along with the low cost and abundance of sulfur⁷, Li–S batteries offer great potential for next-generation battery systems for long-range electric vehicles⁸.

Considerable research efforts have been made to resolve the material challenges in Li–S batteries to boost electrochemical performance. These efforts include using porous carbon/polar hosts for mitigating polysulfide dissolution^{9–11}, three-dimensional cathodes for enhancing electronic/ionic conductivity and accommodating volume change^{12,13}, host and artificial solid electrolyte interphase design for protecting Li anodes^{14,15}, and modifications to electrolytes, separators, binders and current collectors^{6,16–18}. Despite great

advances, most reported studies were conducted using coin-cell configurations under low sulfur mass loadings, excessive amounts of electrolytes and uncontrolled Li anode conditions. A large proportion of inactive materials greatly offsets the high-energy advantage, thus limiting their practicality. High sulfur utilization and electrochemical performance are more easily achieved with excessive electrolytes and Li metals, which conceals the real challenges in Li–S batteries. The current cycle life of Ah-level Li–S pouch cells reaches approximately several tens of cycles, mainly due to the powdering of the Li metal anode and depletion of both electrolytes and Li. To realize high-energy-density Li–S batteries, all these key parameters need to be carefully taken into consideration.

Despite several previous reports^{19,20} discussing the importance of key design parameters (such as sulfur loading and cathode weight ratio) for realizing high-energy-density Li–S batteries, there has not been a thorough quantitative analysis of how the cell-level energy density is determined by those parameters. In particular, comprehensive analyses regarding the influence of all battery components on the full-cell energy density are lacking.

By considering all key parameters for designing practical Li–S battery technologies, here we propose two descriptors (R_{weight} and R_{energy}) to analyse the mass- and energy-level compromise on the full-cell energy density. An explicit formulation summarizing critical parameters for realizing high-energy-density Li–S batteries is provided accordingly. This is followed by discussions on the laboratory- to industrial-scale production of Li–S batteries. We also validate our energy density formulation using Li–S battery data reported in the literature. Future perspectives on the development of high-energy Li–S batteries are also described.

Formulation for high-energy-density Li–S batteries

A real Li–S battery consists of one or two current collectors for electron transport, a separator for electronically insulating electrodes, an electrolyte for ion transport and a package for shielding the battery core from air. We first summarize these battery components in Fig. 1a, where we distinguish the electrochemically

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Table 1 | Comparison of key parameters in different batteries

| Battery type | Energy density (Wh kg ⁻¹) | Driving distance (km) | Cost range (\$ kWh ⁻¹) |
|-----------------|--|---|--|
| Pb acid | 25–45 (ref. 43); 38–60 (ref. 44) | 47–84 ^a ; 71–112 ^a | 150–500 (ref. 43); 70–160 (ref. 44) |
| Ni–Cd | 50–75 (ref. 43) | 93–140 ^a | 800–1,500 (ref. 43); 400–2,400 (ref. 43) |
| Ni–MH | 70–80 (ref. 43); 42–110 (ref. 44) | 130–149 ^a ; 78–205 ^a | 200–729 (ref. 43); 210–365 (ref. 44) |
| Li-ion | 168–193 (ref. 45); ≈250 (ref. 46) | 300–600 (ref. 45) | ≈130 (ref. 46); 70–250 (ref. 44) |
| Advanced Li-ion | 269–350 (ref. 44); 343–433 (ref. 45); ≤500 (ref. 47) | 466–653 ^a ; 640–808 ^a ; ≤933 ^a | ≈130 (ref. 46) |
| Li–S | 350 (ref. 25)–609 (ref. 33) | 653–1,136 ^a | 36–130 (ref. 44) |

Advanced Li-ion batteries are defined as Li-ion batteries using advanced anode materials (such as silicon and metallic lithium, as opposed to graphite only) to pair with high-capacity lithium-transition metal oxide cathodes (such as lithium nickel manganese cobalt oxides). ^aDriving distance was calculated on the basis of the energy density of each battery type, using the calculation method listed in ref. 45.

active materials from the inactive ones. The mass of the cathode (m_{cathode}) includes any conducting agent, sulfur host and binder that may be used to boost the utilization of sulfur. Moreover, an excess of Li for prolonging the cycle life is also needed beyond the active Li used during battery operation. The mass ratio of the negative-to-positive electrode material ($R_{\text{N/P}}$) depicts the excessive amount of Li anode (m_{anode}). The mass of the electrolyte and separator is defined as $m_{\text{electrolyte}}$ and $m_{\text{separator}}$, respectively. To reflect the mass distribution of the real battery, we introduce a descriptor (R_{weight}) to depict the total mass percentage of both S and active Li.

In addition to the inactive mass-related issues (termed mass-level compromise), we also look at energy loss due to incomplete reactions and voltage polarizations (termed energy-level compromise). In an ideally completed electrochemical reaction (from two equivalents of Li and one equivalent of sulfur into final Li_2S), the reaction enthalpy is converted to electric energy that affords a theoretical specific capacity of 1,675 mAh g⁻¹ (on the basis of the mass of sulfur) at an average output voltage of 2.2 V (on the basis of the difference in their electrode potentials). However, in a real Li–S battery, this reaction can hardly be completed, to convert lithium and sulfur into Li_2S . For example, some sulfur materials are dissolved/diffused into electrolytes so that they cannot participate in subsequent electrochemical reactions, while some sulfur materials (such as Li_2S_2) remain in intermediate states during discharging rather than being completely converted to Li_2S . This degree of reaction completion can be directly reflected by the specific capacity of sulfur (C_{sulfur}) compared with its theoretical capacity of 1,675 mAh g⁻¹. On the other hand, voltage polarizations in various forms, such as ohmic, concentration and activation, inevitably occur during operation, which decreases the average output voltage (V_{average}) and results in a proportional loss of output energy. To quantify this energy-level compromise, we introduce another parameter as the energy utilization ratio (R_{energy}) of active materials, combining energy loss from both the incomplete electrochemical reactions (C_{sulfur} divided by the theoretical capacity) and the polarization-induced result (V_{average} divided by the theoretical voltage).

Having identified both the mass- and energy-level compromises, we now present equations below to calculate the gravimetric energy density of Li–S batteries on the basis of the R_{weight} and R_{energy} definitions that include all the parameters.

$$\text{Energy density} = 2,567 \text{ Wh kg}^{-1} \times R_{\text{weight}} \times R_{\text{energy}}, \quad (1)$$

where 2,567 Wh kg⁻¹ is the theoretical energy density of a Li–S full cell.

$$R_{\text{energy}} = \frac{C_{\text{sulfur}}}{1,675 \text{ mAh g}^{-1}} \times \frac{V_{\text{cathode}}}{2.2 \text{ V}}, \quad (2)$$

where 1,675 mAh g⁻¹ is the theoretical specific capacity of sulfur and 2.2 V is the theoretical output voltage of a Li–S full cell.

$$R_{\text{weight}} = \frac{\frac{M_{\text{Li}_2\text{S}}}{M_{\text{S}}} \times m_{\text{sl}} \times (1 - R_{\text{package}})}{\frac{m_{\text{sl}}}{R_{\text{cathode}}} + \frac{m_{\text{Al}} + m_{\text{Cu}}}{2} + m_{\text{separator}} + \rho_{\text{E}} \times R_{\text{E/S}} \times m_{\text{sl}} + \frac{2M_{\text{Li}}}{M_{\text{S}}} \times R_{\text{N/P}} \times m_{\text{sl}}} \quad (3)$$

where $M_{\text{Li}_2\text{S}}$, M_{S} and M_{Li} are the molar weights of Li_2S (45.947), sulfur (32.065) and lithium (6.941), respectively. The m_{sl} depicts the areal mass loading of sulfur in the cathode. R_{package} illustrates the weight ratio of the package in the whole full cell. R_{cathode} represents the weight ratio of sulfur in the cathode, which includes the sulfur, host, conducting agent and binder. m_{Al} , m_{Cu} and $m_{\text{separator}}$ are the areal masses of the Al current collector, Cu current collector and separator, respectively. ρ_{E} is the density of the electrolyte, $R_{\text{E/S}}$ is the ratio of electrolyte to sulfur (in $\mu\text{l mg}^{-1}$) and $R_{\text{N/P}}$ is the ratio of the theoretical areal capacity of the Li metal negative electrode to that of the sulfur positive electrode.

For illustration purposes, we consider a model Li–S pouch cell. To simplify the calculation, we use the following parameters commonly applied in real pouch cells. The package weight ratio, R_{package} , is 10 wt%. The thickness of the aluminium foil is 10 μm with an areal density of 2.7 mg cm⁻². The copper current collector is not included because the highly conductive Li metal foil can also be used as an anode current collector. The thickness of the separator is 10 μm with an areal density of 1.0 mg cm⁻², and the density of the electrolyte ρ_{E} is 1.1 g ml⁻¹. Regarding the mass of the current collectors, we use half of the current collector mass based on the double-side coating technique.

First, we inspect the relationship between R_{weight} and m_{sl} with $R_{\text{E/S}}$ when the other two key variables, R_{cathode} and $R_{\text{N/P}}$ are set to 90% and 2, respectively, which are commonly used in high-energy-density Li–S pouch cells. R_{weight} can be rewritten in equation (4) as:

$$R_{\text{weight}} = \frac{1.433 \times m_{\text{sl}} \times 0.9}{2.35 + (1.11 + 1.1 \times R_{\text{E/S}} + 0.433 \times 2) \times m_{\text{sl}}} \quad (4)$$

From this equation, the correlation result is demonstrated in Fig. 1b. Both reducing $R_{\text{E/S}}$ and increasing m_{sl} can improve R_{weight} . For instance, R_{weight} can be considerably improved from 9.5% to 35.1% by decreasing $R_{\text{E/S}}$ from 10 to 1 $\mu\text{l mg}^{-1}$, when m_{sl} is set at 4 mg cm⁻². However, R_{weight} shows less improvement, that is, from 24.1% to 29.4%, by increasing m_{sl} from 2 to 12 mg cm⁻², when $R_{\text{E/S}}$ is set at 2 $\mu\text{l mg}^{-1}$. These results suggest that decreasing $R_{\text{E/S}}$ is more effective for enhancing R_{weight} than increasing m_{sl} .

Second, we discuss the relationship between R_{weight} and R_{cathode} with $R_{\text{N/P}}$ and set the other two key variables, m_{sl} and $R_{\text{E/S}}$, to be 12 mg cm⁻² and 2 $\mu\text{l mg}^{-1}$, respectively, which are also commonly

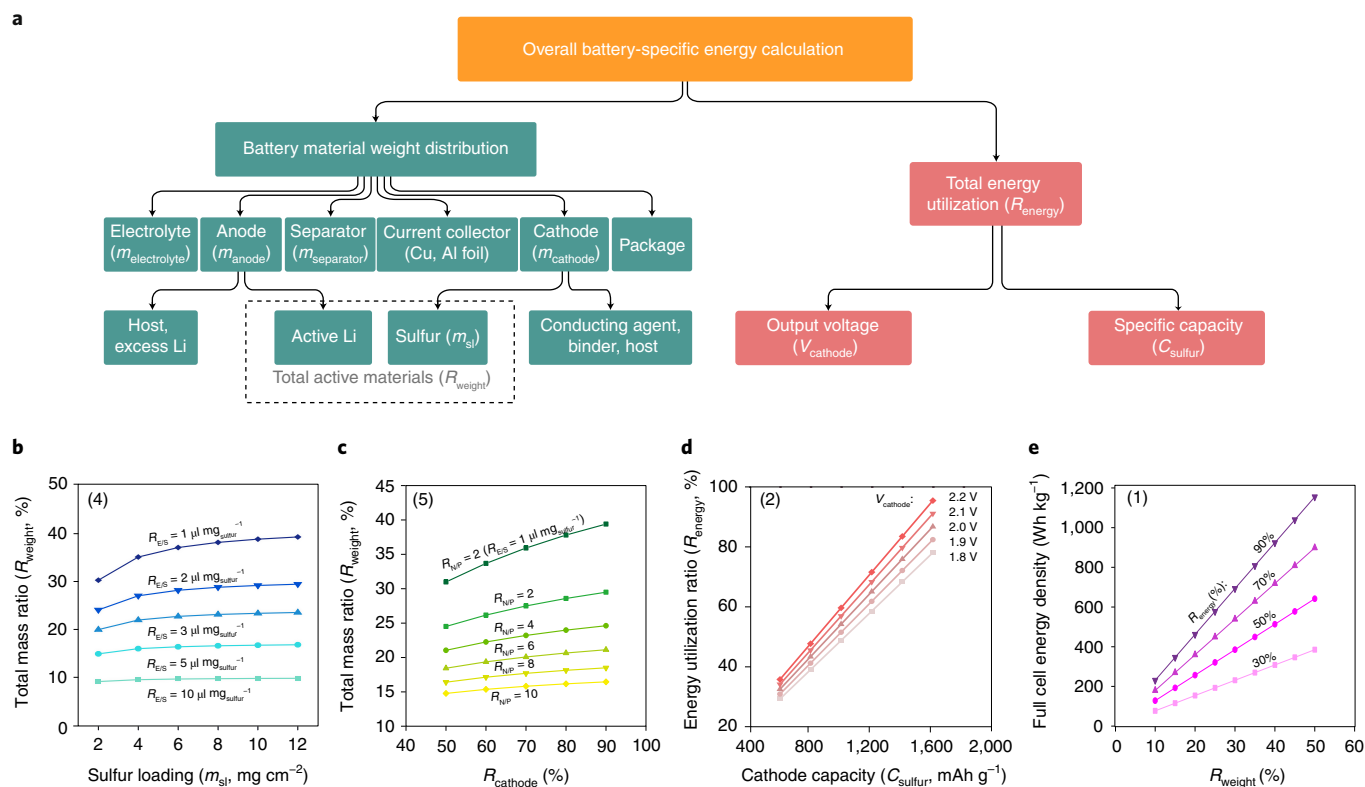


Fig. 1 | Formulation of energy density and effects from key parameters. **a**, Illustration of active and inactive components in the full cell that affect R_{weight} , the voltage and capacity parameters that influence R_{energy} and the final impact on the full-cell energy density. **b**, Calculation of total mass ratio in full cell with different $R_{E/S}$ and sulfur loading, assuming there is no Cu current collector (equation (4)). **c**, Calculation of total mass ratio in full cell with different $R_{N/P}$ and weight ratio of the sulfur in cathode (equation (5)). **d**, Calculation of energy utilization ratio with different capacity and voltage (equation (2)). **e**, Calculation of full-cell energy density with different R_{weight} and R_{energy} (equation (1)).

used in pouch cells in the literature. Then, R_{weight} can be described as:

$$R_{\text{weight}} = \frac{1.433 \times 12 \times 0.9}{\frac{12}{R_{\text{cathode}}} + 2.35 + 1.1 \times 2 \times 12 + 0.433 \times R_{N/P} \times 12} \quad (5)$$

The relationship between R_{weight} , R_{cathode} and $R_{N/P}$ is thus established, as shown in Fig. 1c. As R_{cathode} increases, R_{weight} slightly increases at various $R_{N/P}$ values. For example, even when changing R_{cathode} from 50 wt% to 90 wt%, R_{weight} only shows a small increase from 21.0% to 24.6% when $R_{N/P}$ is set at 4. In comparison, R_{weight} increases rapidly as $R_{N/P}$ decreases, especially at high sulfur contents. When R_{cathode} is set at 80%, R_{weight} can be improved from 16.2% to 28.6% by lowering $R_{N/P}$ from 10 to 2. These results suggest that decreasing $R_{N/P}$ and $R_{E/S}$ are much more effective for enhancing R_{weight} . In particular, if $R_{N/P}$ and $R_{E/S}$ are reduced to even lower values of 2 and $1 \mu\text{L mg}^{-1}$, respectively, then R_{weight} could reach 39.4%. Therefore, $R_{N/P}$ and $R_{E/S}$ are critical parameters for attaining an ultrahigh energy density of 800–1,000 Wh kg^{-1} in future Li–S batteries.

From equation (2) and Fig. 1d, increasing both the C_{sulfur} and V_{cathode} can considerably improve R_{energy} : for example, increasing C_{sulfur} from 600 to 1,200 and 1,600 mAh g^{-1} (assuming $V_{\text{cathode}} = 2.1 \text{ V}$) dramatically changes R_{energy} from 34.2% to 68.4% and 91.2%. Finally, the relationship of the final battery energy density with R_{weight} and R_{energy} is shown in Fig. 1e, as depicted by equation (1). For example, a Li–S battery designed with $R_{\text{weight}} \geq 28\%$ and $R_{\text{energy}} \geq 70\%$ can achieve an energy density of 500 Wh kg^{-1} ; an 800 Wh kg^{-1} battery may need the R_{weight} and R_{energy} parameters to be no lower than 37% and 85%,

respectively; and a 1,000 Wh kg^{-1} battery demands the R_{weight} and R_{energy} parameters be approximately 39% and 95%, respectively.

In addition to the gravimetric energy density, the volumetric energy density of Li–S batteries is also important but is affected by the porosity and tap density of the electrodes. A dense electrode with low porosity is required to minimize electrolyte uptake. Applied pressure can adjust the thickness and porosity of the as-prepared electrodes, which reduces the electrolyte amount for wetting and increases the volumetric energy density. Xue et al. established a good model based on a commercial battery configuration and proposed that cathode porosity is another key parameter for cell-level volumetric energy density²¹. However, the volume change in metallic Li and sulfur electrodes, as well as gas generated from electrolyte decomposition, will greatly change the battery volume during cycling, thus affecting the accuracy for both calculation and volumetric energy-density-based applications²².

Ah-level high-energy-density Li–S batteries

Current achievements in Li–S pouch cells. Research and commercial efforts on Ah-level Li–S pouch cells have realized energy densities of 300 to 600 Wh kg^{-1} (Fig. 2a), much higher than that of the current Li-ion cylinder cells used in the Tesla Model 3 electric car (246 Wh kg^{-1})²³. For example, scholars at the Beijing Institute of Technology (BIT) designed interconnected mesoporous ‘bubble-like’ carbon fabrics to anchor nanosulfur and form free-standing cathodes without inactive binders or current collectors, realizing 315.98 Wh kg^{-1} in pouch Li–S batteries²⁴. Investigators at the Dalian Institute of Chemical Physics (DICP) used 1,3-dioxolane as the electrolyte solvent and a large-surface-area carbon material-based

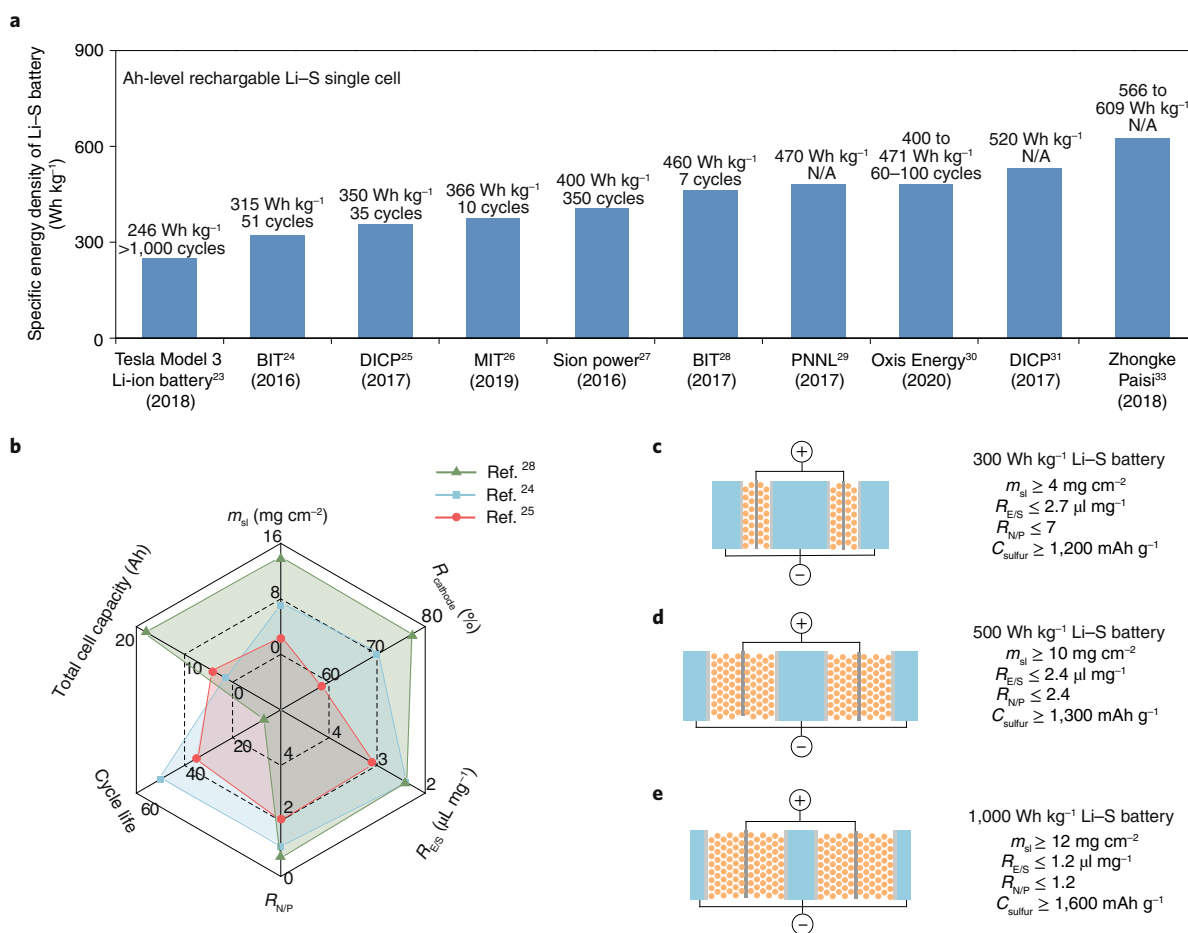


Fig. 2 | Representative Ah-level Li-S battery pouch cells with key design parameters. **a**, Comparisons of energy densities between several Li-S pouch cells reported in the literature and current Li-ion cylinder cell in Tesla electric car, developed after 2015. The energy density values of batteries from Tesla²³, Sion Power²⁷, Oxis Energy³⁰ and Zhongke Paisi³³ are from the commercial sources and public announcements. For convenience, the panel is arranged in the order of the energy density value. For the batteries of PNNL²⁹, DICP³¹ and Zhongke Paisi³³, no cycle numbers were provided in the original reports. **b**, Radar plot of the key parameters for fabricating high-energy-density Li-S pouch cells reported in the literature^{24,25,28}. **c–e**, Illustration of recommended key parameters for designing high-energy-density Li-S batteries (300 Wh kg⁻¹ (**c**); 500 Wh kg⁻¹ (**d**); 1,000 Wh kg⁻¹ (**e**)), calculated from equations (1)–(3).

cathode to inhibit the polysulfide shuttle, stabilize the Li anode and maintain sulfur utilization, reporting 350 Wh kg⁻¹ Li-S soft package batteries²⁵. Researchers at Massachusetts Institute of Technology designed dense intercalation-conversion hybrid cathodes by combining Mo₆S₈ and sulfur, reducing the cathode porosity to 55% and increasing the energy density to 366 Wh kg⁻¹ (ref. ²⁶). The Sion power company reported large Li-S batteries realizing an energy density of 400 Wh kg⁻¹ with 350 cycles²⁷. In addition, at BIT, sulfur-impregnated oval-like carbon microstructures (OLCM/S) were assembled with isotropic electron transportation for sulfur utilization and large electrolyte/electrode interface features for lean electrolyte wetting, producing pouch cells with a specific energy capacity of 460.08 Wh kg⁻¹ (ref. ²⁸). At Pacific Northwest National Laboratory (PNNL), a soft PEO₁₀LiTFSI polymer was developed to immobilize the electrolyte and confine polysulfides under lean electrolyte conditions, realizing 470 Wh kg⁻¹ in pouch Li-S batteries²⁹. Oxis Energy announced >15 Ah Li-S battery products with energy densities as high as 400 Wh kg⁻¹, and Li-S battery prototypes at an energy density of 471 Wh kg⁻¹ (ref. ³⁰). DICP³¹ and Institution of Chemical Defence (ICD)³² also reported rechargeable Li-S pouch cells with high energy densities of 520 and 605 Wh kg⁻¹, respectively. Zhongke Paisi³³ announced a rechargeable Li-S battery with an ultrahigh energy density of 609 Wh kg⁻¹, and their

20 Ah-level pouch cell with an energy density of 566 Wh kg⁻¹ was identified through the China National Light Industry Council (in ref. ³³ the Chinese language certification is translated as ‘through third-party testing, their 20 Ah Li-S battery has an energy density of 566 Wh kg⁻¹). Impressively, investigators at DICP demonstrated an ultrahigh energy density of 916 Wh kg⁻¹ achieved in a Li-S primary battery²². These achievements in single Ah-level Li-S batteries mostly rely on smart designs with interconnected and well-ordered porous carbon materials^{24,28} as major hosts, cathodic electrocatalysts²⁶ with high adsorption ability, good conductivity and high catalytic activity to ensure the fast conversion of polysulfides, and advanced electrolytes^{25,29} for stabilizing Li anodes and suppressing polysulfide dissolution. As will be illustrated in the next section, these design strategies provide both high R_{energy} and R_{weight} , leading to their high energy densities.

In addition to these single battery achievements, researchers have also demonstrated large Li-S battery packs composed of tens of batteries for practical kWh-level energy consumption applications. For example, at Sion power, a 12 Li-S battery pack was fabricated for higher voltage output³⁴, and at Oxis energy, a battery management system containing a 16-cell pack was designed that is capable of measuring the temperature, current and voltage of individual batteries during operation³⁵. Impressively, Chen et al. demonstrated

a 1 kWh-level Li-S battery pack (22 V, 50 Ah)³¹, with a pack-level energy density as high as 330 Wh kg⁻¹. Based on this progress on both high-energy-density single batteries and large packs, Li-S batteries have proved their advantages in multiple energy storage applications, such as successfully supporting the 11-day non-stop flight of drones achieved by Airbus and Oxis Energy companies³⁶.

Key parameters and formulation validation. On the basis of these works, the key battery parameters for designing high-energy-density Li-S pouch cells can be summarized for future reference. For example, Chen et al. reported key parameters on their OLCM/S cathode-based pouch cells²⁸, including an ultrahigh m_{sl} of 14 mg cm⁻², an R_{cathode} of 77.4% (based on the mass ratio of 87:5:8 for OLCM/S:carbon black:binder), an $R_{\text{E/S}}$ of 2.45 $\mu\text{m g}^{-1}$ and an $R_{\text{N/P}}$ of 0.72. In terms of energy utilization, C_{sulfur} attained approximately 1,100 mAh g⁻¹ (0.05C), while V_{average} reached 2.05 V. Applying these parameters in equations (1) to (3), the R_{weight} , R_{energy} and energy-density values of their pouch cell are calculated to be 28.8%, 61.2% and 452 Wh kg⁻¹, respectively. This calculated energy density, based on our equation, is very close to their experimental energy density of 460 Wh kg⁻¹, suggesting the accuracy of our energy density calculation equation and the importance of R_{weight} and R_{energy} . In addition to this OLCM/S work, most of several other Ah-level Li-S pouch cell results with energy densities higher than 300 Wh kg⁻¹ also emphasize the following key parameters with regard to R_{weight} , such as $m_{\text{sl}} \geq 7 \text{ mg cm}^{-2}$ on each side of the cathode, $R_{\text{cathode}} > 70\%$ in the cathode, a relatively low $R_{\text{E/S}} \leq 3.5 \mu\text{m g}^{-1}$ and a low $R_{\text{N/P}} < 5$ (Fig. 2b). This detailed information gives guidance on future battery design, consistent with our proposed energy-density calculation formulation.

On the basis of these validated equations, recommended battery parameters can be proposed for designing high-energy-density Li-S batteries. For instance (Fig. 2c-e), a sulfur loading of 4 mg cm⁻², an E/S ratio $\leq 2.7 \mu\text{m g}^{-1}$ and an N/P ratio ≤ 7 are recommended to achieve a cell-level energy density of 300 Wh kg⁻¹, in addition to a C_{sulfur} of $\sim 1,200 \text{ mAh g}^{-1}$ (see Methods). However, for fabricating higher energy density Li-S pouch cells ($> 500 \text{ Wh kg}^{-1}$), the recommended parameters include $m_{\text{sl}} \geq 10 \text{ mg cm}^{-2}$, $R_{\text{E/S}} \leq 2.4 \mu\text{m g}^{-1}$, $R_{\text{N/P}} \leq 2.4$ and $C_{\text{sulfur}} \geq 1,300 \text{ mAh g}^{-1}$ (see Methods). Notably, all these recommended parameters can be dynamically adjusted to reach the desired energy density in the full cell, when several parameters listed in equations (1) to (3) exceed the recommendations (that is, a C_{sulfur} and a m_{sl} higher than recommended numbers, or a $R_{\text{N/P}}$ and a $R_{\text{E/S}}$ lower than recommended numbers), but several other parameters do not reach the recommendations. For example, a Li-S battery can still reach 500 Wh kg⁻¹ when its C_{sulfur} and m_{sl} only reach 1,200 mAh g⁻¹ and 6 mg cm⁻² (which are lower than the recommended parameters of 1,300 mAh g⁻¹ and 10 mg cm⁻²), respectively, but $R_{\text{E/S}}$ and $R_{\text{N/P}}$ reach 2.1 $\mu\text{m g}^{-1}$ and 2.1, respectively (which are lower than the recommended parameters of 2.4 $\mu\text{m g}^{-1}$ and 2.4, respectively). For 18650-type cylindrical cells, these requirements should be much higher considering the high weight ratio of the stainless-steel-cell shell.

Accordingly, we propose ‘four high’ (‘4H’) and ‘four low’ (‘4L’) criteria for building high-energy-density (for example, 400 to 500 Wh kg⁻¹) and long cycling life Li-S batteries. 4H refers to a C_{sulfur} value of $> 1,200 \text{ mAh g}^{-1}$, an m_{sl} of $> 8 \text{ mg cm}^{-2}$, an R_{cathode} of $> 70 \text{ wt}\%$ and a Coulombic efficiency of $> 99.9\%$, and 4L suggests a porosity of $< 60\%$, an $R_{\text{N/P}}$ of < 3 , an $R_{\text{E/S}}$ of $< 3 \mu\text{m g}^{-1}$ and a minimal inactive material. Although it is challenging to simultaneously achieve these values, these are the target values that we suggest for the community to make Li-S chemistry practically feasible.

Target to achieve practical Li-S batteries. From the above analyses, the necessary parameters for designing practical 400 to 500 Wh kg⁻¹ Li-S full cells are summarized in Table 2, along with

the research status and general solutions. The recommended key parameters can be dynamically adjusted; for example, a Li-S full cell with a low R_{energy} (such as 64.9%) can still reach an energy density of 500 Wh kg⁻¹ when a high R_{weight} (30.0%) is used.

Nevertheless, the specific energy is not the only parameter of importance for practical Li-S batteries. Another important problem yet to be solved is the cycle life issue. Current LIBs can retain at least 80% of their initial capacity after more than 500 cycles³⁷. In comparison, current Li-S pouch cells are only capable of retaining less than 80% capacity after no more than 100 cycles. Considering the excellent cycling performance ($> 1,000$ cycles) of various well-designed sulfur cathode materials in half-cells (using excessive amounts of anodes and electrolytes)³⁸, this insufficient cycle life issue of Li-S full cells should be attributed to the rapid depletion of Li anodes and electrolytes, especially under practical conditions where a minimal amount of anodes and lean electrolytes are typically used. As the Li stripping/plating reversibility only reaches approximately 99% in ether-based electrolyte systems (1% loss of utilized Li metal in the anode during each cycle)³⁹, the Li-S pouch cell with an $R_{\text{N/P}}$ of 2 only demonstrates 120 cycles before 20% capacity loss. Zhang and co-workers disassembled the 5 Ah Li-S pouch cell after 10 cycles and found severe powdering of the Li metal anode due to a side reaction between the Li metal and electrolytes²². Additionally, the depletion of electrolytes will lead to loss of ionic conduction between electrodes, which increases polarization and even damages the full cell. In this regard, developing more stable electrolytes, Li metal anodes and solid electrolyte interphase will be of critical importance for future practical long-cycle-life and high-energy-density Li-S battery systems.

In addition to the cycle life issue, other challenges need to be resolved. First, the current rate capability of Li-S pouch cells (commonly slower than 0.2C) is unsatisfying, mainly due to the highly electrically insulating nature of sulfur and lithium (poly) sulfide active materials. Second, the current material cost of Li-S batteries (US\$400 kWh⁻¹) is much higher than that of current LIBs (US\$150–250 kWh⁻¹), mostly because of the high price of bis(trifluoromethane)sulfonimide lithium salt (LiTFSI)-based electrolytes and thin Li metal films (see Methods)^{22,24,40}. The target price of Li-S batteries should be below US\$100 kWh⁻¹ to compete with LIBs. This calls for new electrolytes and thin Li manufacturing technologies⁴¹. Finally, the highly combustible microscale porous Li metal anode (after cycling) and ether-based electrolytes in Li-S battery systems are much more likely to cause explosions and safety hazards than conventional LIBs, especially when the battery core is short-circuited or exposed to air. Fire-extinguishing protection layers would be possible for addressing this issue, such as a fire-retardant current collector⁴². Future modifications to high safety and low-cost liquid electrolytes, additives, thin Li anodes and other components are also essential for practical applications of Li-S batteries in markets.

Conclusions

The Li-S battery is one of the most promising energy storage systems on the basis of its high-energy-density potential, yet a quantitative correlation between key design parameters and battery energy density is lacking in the field. Through the identification and analysis of mass- and energy-level compromises, we present mathematical connections between the battery energy density and the primary design/performance parameters (such as sulfur loading, negative-to-positive electrode capacity ratio and cathodic specific capacity based on sulfur). We suggest achieving a low ratio of electrolyte to sulfur, a low ratio of areal anode capacity to areal cathode capacity and a high specific capacity based on sulfur to be of the highest priority for high-energy-density Li-S batteries. We also summarize the current development of Ah-level high-energy Li-S batteries. By analysing the detailed parameters of these

Table 2 | Research targets, status and solutions to achieve 400 to 500 Wh kg⁻¹ Li-S battery

| Parameters | Recommended parameters | Status | General solutions | |
|---------------------|--------------------------|--|---|---|
| R_{energy} | V_{cathode} | $\geq 2.1 \text{ V}$ | Some reports have achieved this requirement during ultraslow discharging rate ($\leq 0.1\text{C}$). | (1) High electric conducting host materials can decrease electric resistance-induced overpotential. (2) Uniform distribution of sulfur nanoparticles, efficient catalyst and fast ion transportation can mitigate the redox reaction barrier-induced overpotential. (3) Good polysulfide confinement can promote the stability of V_{cathode} after repeated cycling. |
| | C_{sulfur} | $\geq 1,200 \text{ mAh g}^{-1}$ | Some reports have reached $\sim 1,200 \text{ mAh g}^{-1}$ during ultraslow discharging rate ($\leq 0.1\text{C}$). | (1) High electric conducting host materials can improve the utilization of active material during operation. (2) Nanosizing sulfur particles, efficient sulfur redox reaction catalyst and fast ion transportation can promote active material utilization during operation. (3) Good polysulfide confinement can suppress the degradation of C_{sulfur} after repeated cycling. |
| R_{weight} | m_{sl} | $\geq 8 \text{ mg cm}^{-2}$ | Some reports have achieved this requirement by increasing coating thickness and decreasing inactive component in the cathode. Further improving the m_{sl} is not very meaningful considering its low contribution in improving R_{weight} . | (1) Low weight yet high electric conducting, high sulfur philicity/bonding hosts can promote good cathode performance under high sulfur loading conditions. (2) Effective binder, conducting agent materials and three-dimensional host design can help high loading cathode fabrication. |
| | R_{cathode} | $\geq 70\%$ | Some reports have achieved this requirement by decreasing the weight of hosts, binders and conducting agents. Further improving the R_{cathode} is meaningful considering its moderate contribution in improving R_{weight} . | (1) Effective yet low-weight hosts, binders and conducting agent material design can decrease the amount of inactive materials in the cathode. (2) Three-dimensional or layer-by-layer electrode structural design can increase sulfur loading. |
| | $R_{\text{E/S}}$ | $\leq 3 \mu\text{L mg}^{-1}$ | Some reports have achieved this requirement by using low surface area and highly wettable host material design. Decreasing this $R_{\text{E/S}}$ parameter is important based on its high contribution in improving R_{weight} . | (1) Low material porosity (by low surface area host or effective calendaring process) and good electrolyte wettability in the cathode are essential for reducing the electrolyte amount. (2) Developing stable electrolyte against side reaction and evaporation is essential for maintaining good electrolyte wetting and ionic conduction after repeated cycling under lean electrolyte condition. |
| | $R_{\text{N/P}}$ | ≤ 3 | Some reports have achieved this requirement by using thin Li foil. Decreasing this $R_{\text{N/P}}$ parameter is important based on its high contribution in improving R_{weight} . | (1) Using thin Li metal foil or increasing m_{sl} can considerably reduce the $R_{\text{N/P}}$. (2) Developing more stable Li anodes and electrolyte against side reaction is essential for retaining anode and full-cell cyclability under lean Li anode conditions. |
| | Other inactive materials | $R_{\text{package}} \leq 10\%$; other material as light weight as possible. | Some reports use a thin Al current collector in the cathode, absence of Cu collector in the anode and around a $10 \mu\text{m}$ thick separator. Some reports build large battery core to reduce the mass ratio of package. | (1) Using thin and low-weight inactive materials (such as sulfur hosts, Li hosts, binders, conducting agents and tabs) is important. (2) Developing three-dimensional, self-standing and high electric conducting electrode material can avoid using current collectors. (3) Developing ultrathin yet stable solid-state electrolyte can reduce the weight of separator and electrolyte. (4) Building large battery core can reduce the ratio of package mass. |
| Summary | R_{weight} | Around 28% | Some reports have achieved this requirement in coin cells and pouch cell configurations. | All the solutions to achieve desired parameters in R_{weight} as listed above, need to be considered. |
| | R_{energy} | Around 70% | Some reports have achieved this requirement in coin cells. Few reports have achieved this requirement in pouch cells for multiple reasons, such as insufficient electronic/ionic conductivity in materials, electrolyte wettability and manufacture technologies. | All the solutions to achieve desired parameters in R_{energy} as listed above, need to be considered. |

Ah-level cells, we propose a ‘four high–four low’ principle, which is not only consistent with our quantitative equation for the energy density, but also provides design guidelines for the development of high-performance Li–S batteries. The energy density calculation and design protocols presented in this work are applicable not only to Li–S batteries, but also can be extended to other energy storage and conversion technologies.

Methods

Calculation for 300 Wh kg⁻¹ Li–S full cell. In a Li–S pouch cell (Fig. 2c), a sulfur loading of 4 mg cm⁻², an $R_{\text{cathode}} \geq 90\%$, an $R_{\text{E/S}} \leq 2.7 \mu\text{g mg}^{-1}$ and an N/P ratio ≤ 7 are recommended to achieve a cell-level energy density of 300 Wh kg⁻¹, in addition to the specific cathodic capacity of $\sim 1,200 \text{ mAh g}^{-1}$ (equations (6)–(8)). This is demonstrated below.

$$R_{\text{weight}} = \frac{\frac{M_{\text{LiS}}}{M_{\text{S}}} \times m_{\text{S}} \times (1 - R_{\text{package}})}{\frac{m_{\text{S}}}{R_{\text{cathode}}} + \frac{m_{\text{Al}} + m_{\text{Cu}}}{2} + m_{\text{separator}} + \rho_{\text{E}} \times R_{\text{E}} \times m_{\text{d}} + \frac{2M_{\text{Li}}}{M_{\text{S}}} \times R_{\text{N}} \times m_{\text{d}}} \quad (6)$$

$$= \frac{1.433 \times 4 \times 0.9}{\frac{4}{0.9} + 1.35 + 1 + 1.1 \times 2.7 \times 4 + 0.433 \times 7 \times 4} = 16.75\%$$

$$R_{\text{energy}} = \frac{C_{\text{sulfur}}}{1,675 \text{ mAh g}^{-1}} \times \frac{V_{\text{cathode}}}{2.2 \text{ V}} = \frac{1,200 \text{ mAh g}^{-1}}{1,675 \text{ mAh g}^{-1}} \times \frac{2.15 \text{ V}}{2.2 \text{ V}} = 70.01\% \quad (7)$$

$$\text{Energy density} = 2,567 \text{ Wh kg}^{-1} \times R_{\text{weight}} \times R_{\text{energy}} \quad (8)$$

$$= 2567 \times 16.75\% \times 70.01\% = 301.0 \text{ Wh kg}^{-1}$$

Calculation for 500 Wh kg⁻¹ Li–S full cell. In a representative Li–S pouch cell, a sulfur loading of 10 mg cm⁻², an $R_{\text{cathode}} \geq 90\%$, an $R_{\text{E/S}} \leq 2.4 \mu\text{g mg}^{-1}$ with an N/P ratio ≤ 2.4 are recommended to achieve a cell-level energy density of 500 Wh kg⁻¹, in addition to the specific cathodic capacity of $\sim 1,300 \text{ mAh g}^{-1}$ (Fig. 2d and equations (9)–(11)):

$$R_{\text{weight}} = \frac{\frac{M_{\text{LiS}}}{M_{\text{S}}} \times m_{\text{S}} \times (1 - R_{\text{package}})}{\frac{m_{\text{S}}}{R_{\text{cathode}}} + \frac{m_{\text{Al}} + m_{\text{Cu}}}{2} + m_{\text{separator}} + \rho_{\text{E}} \times R_{\text{E}} \times m_{\text{d}} + \frac{2M_{\text{Li}}}{M_{\text{S}}} \times R_{\text{N}} \times m_{\text{d}}} \quad (9)$$

$$= \frac{1.433 \times 10 \times 0.9}{\frac{10}{0.9} + 1.35 + 1 + 1.1 \times 2.4 \times 10 + 0.433 \times 2.4 \times 10} = 25.67\%$$

$$R_{\text{energy}} = \frac{C_{\text{sulfur}}}{1,675 \text{ mAh g}^{-1}} \times \frac{V_{\text{cathode}}}{2.2 \text{ V}} = \frac{1,300 \text{ mAh g}^{-1}}{1,675 \text{ mAh g}^{-1}} \times \frac{2.15 \text{ V}}{2.2 \text{ V}} = 75.85\% \quad (10)$$

$$\text{Energy density} = 2,567 \text{ Wh kg}^{-1} \times R_{\text{weight}} \times R_{\text{energy}} \quad (11)$$

$$= 2,567 \times 25.67\% \times 75.85\% = 499.8 \text{ Wh kg}^{-1}$$

Calculation for 1,000 Wh kg⁻¹ Li–S full cell. The 1,000 Wh kg⁻¹ Li–S full cell is much more challenging and demands much higher R_{weight} and R_{energy} . A sulfur loading of 12 mg cm⁻², an $R_{\text{cathode}} \geq 90\%$, an $R_{\text{E/S}} \leq 1.2 \mu\text{g mg}^{-1}$ with an N/P ratio ≤ 1.2 are recommended to achieve a cell-level energy density of 1,000 Wh kg⁻¹, in addition to the C_{sulfur} of $\sim 1,600 \text{ mAh g}^{-1}$ (Fig. 2e and equations (12)–(14)):

$$R_{\text{weight}} = \frac{\frac{M_{\text{LiS}}}{M_{\text{S}}} \times m_{\text{S}} \times (1 - R_{\text{package}})}{\frac{m_{\text{S}}}{R_{\text{cathode}}} + \frac{m_{\text{Al}} + m_{\text{Cu}}}{2} + m_{\text{separator}} + \rho_{\text{E}} \times R_{\text{E}} \times m_{\text{d}} + \frac{2M_{\text{Li}}}{M_{\text{S}}} \times R_{\text{N}} \times m_{\text{d}}} \quad (12)$$

$$= \frac{1.433 \times 12 \times 0.9}{\frac{12}{0.9} + 1.35 + 1 + 1.1 \times 1.2 \times 12 + 0.433 \times 1.2 \times 12} = 40.99\%$$

$$R_{\text{energy}} = \frac{C_{\text{sulfur}}}{1,675 \text{ mAh g}^{-1}} \times \frac{V_{\text{cathode}}}{2.2 \text{ V}} = \frac{1,600 \text{ mAh g}^{-1}}{1,675 \text{ mAh g}^{-1}} \times \frac{2.2 \text{ V}}{2.2 \text{ V}} = 95.52\% \quad (13)$$

$$\text{Energy density} = 2567 \text{ Wh kg}^{-1} \times R_{\text{weight}} \times R_{\text{energy}} \quad (14)$$

$$= 2,567 \times 40.99\% \times 95.52\% = 1005 \text{ Wh kg}^{-1}$$

All these above recommended parameters can be dynamically adjusted based on the previous equations. For example, a Li–S full cell can still achieve 1,000 Wh kg⁻¹ if its C_{sulfur} only reaches 1,500 mAh g⁻¹ (lower than the recommended 1,600 mAh g⁻¹) but its $R_{\text{E/S}}$ reaches 1 $\mu\text{g mg}^{-1}$ (better than the recommended

1.2 $\mu\text{g mg}^{-1}$). The energy density of a full cell designed with adjusted parameters can be verified using the proposed equations from this perspective.

Weight and cost calculation of raw materials in a Li–S battery. For a 1.55 Ah Li–S pouch cell (with $R_{\text{N/P}}$ of 2.36 and $R_{\text{E/S}}$ of 2.27 $\mu\text{g mg}^{-1}$)²⁴, the weight ratio²⁴ and cost (\$ per kg) of each component are as follows: carbon black, 3.4% and 5 (ref. 22); binder, 1.7% and 5.7 (ref. 22); sulfur, 12.1% and 0.143 (ref. 22); aluminium foil, 10% and 4 (ref. 22); ether electrolyte, 30.3% and ≥ 242 ; metallic Li, 12.4% and $\geq 1,000$ (ref. 40); separator with tab and tape, 9.6% and around 0.714 (ref. 22); package, 20.2% and 2.57 (ref. 22). The cost of ether electrolyte was calculated on the basis of its components: LiTFSi, Solvay, 22.1 wt%, US\$300 kg⁻¹; 1,2-dimethoxyethane, Sigma-Aldrich, 33.3 wt%, US\$177 kg⁻¹; 1,3-dioxolane, Sigma-Aldrich, US\$261 kg⁻¹, 40.8 wt%; lithium nitrate, Sigma-Aldrich, US\$245 kg⁻¹, 3.8 wt%. Thus, the price of the Li–S full cell was calculated to be US\$198.6 kg⁻¹ and US\$397.2 kWh⁻¹ if the battery achieves 500 Wh kg⁻¹. The electrolyte and thin Li metal film are the highest cost (36.9% and 62.4%) amongst all the components in the Li–S battery. With large-scale production in the future, the cost of ether-based electrolyte and thin Li metal film are speculated to decrease to a similar order of magnitude to carbonate electrolyte (US\$10 kg⁻¹) and bulk Li (US\$100 kg⁻¹) as they share similar chemical composition, synthesis procedures and manufacturing processes. Thus, the material cost of Li–S batteries is proposed to be lower than US\$100 kWh⁻¹ in the future.

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Competing interests

Y.C. is the founder and a board director of Amprius Inc., which develops Si anodes. He owns shares in Amprius. G.Z. and H.C. declare no competing interests.

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