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A Detailed Discourse on the Epistemology of Lithium-Sulfur Batteries

The architecture of lithium-sulfur (Li-S) batteries can hold five times more charge capacity compared to Li-ion batteries. This review emphasizes the recent research findings on the desired loading of sulfur, the electrolyte-to-sulfur ratio, and a detailed view of the polysulfide shuttling effect. Problems with electrolyte stability are also discussed as well as the potential remedies they provided in various systems, as Li-S batteries have great potential to surmount these critical issues by understanding the mechanism. Future scopes of Li-S batteries can be progressively attained by optimizing the pore structure, designing highly conductive and strong sulfur confinement systems, and thereby pairing with anode materials to explore the possibility of innovative components for commercializing Li-S batteries.

Keywords: Advancements in Li-S batteries, Energy storage devices, Lithium-sulfur battery, Mitigation of polysulfides, Polysulfide shuttling

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1 Introduction

Universal anxiety about climate change and the ever-depleting fossil fuels has sparked the surfacing of green energy alternatives. With a surge in demand for energy, in this persistently evolving sphere, we look towards sustainable energy sources. One facet of this goal is to unravel solutions for energy storage and the subsequent synthesis of batteries. Among the available methods of energy storage, rechargeable batteries are one of the upcoming technologies, on account of their eco-friendly nature, high conversion efficiency, and flexibility. The contemporary and commonly used batteries are lithium-ion batteries, owing to their long cycle life; they are used in products such as wireless headphones, handheld power tools, and electric vehicles. The rechargeable batteries are an exciting substitute, in contrast to lithium-ion (Li-ion) batteries, for the next generation to be in tune with the expanding energy demand. These batteries are known to have an almost five times higher energy density than Li-ion batteries and are also cheaper and lighter in comparison [1].

The Ragone plot in Fig. 1 is a tool to compare various storage systems for energy, their power, and energy densities. The plot helps to determine the best Li-ion system with maximum efficiency and optimal discharge energy. Batteries have drawbacks such as limited lifetime and expensive maintenance, as seen from the plot. Lithium batteries, however, have some disadvantages such as a lower energy capacity compared to combustible fuels (e.g., petrol provides 13 kWh kg^{-1}, natural gas provides 15 kWh kg^{-1}, hydrogen provides 34 kWh kg^{-1}) and a prolonged recharging time. They are constrained to smaller vehicles with low mileage demands.

1.1 Recent Progress on New Materials

The third-generation rechargeable batteries consider the lithium-sulfur (Li-S) battery as a promising candidate for commercial purposes. As the next frontier, sustainable Li-S cells have been attractive due to their high theoretical capacity (1675 mAh g^{-1}) at a safe operating voltage (1.7–2.8 V) and the abundantly available, environmentally friendly sulfur with low cost. Although it has many advantages, the cycling stability is very poor due to the dendrite formation on Li-metal and depletion of the electrolyte. This leads to degradation at the anode surface with loss of active material and dissolution of lithium polysulfides (LiPS) in the electrolyte medium (shuttle effect) [2].

The recent improvements in designing electric vehicles require a cathode material with cell configuration and fast consumption of both the electrolyte and the active material. Most importantly, the cell parameters are designed according to the sulfur loading (mg cm^{-2}), sulfur content (wt %), and electrolyte-to-sulfur ratio (μL mg^{-1}) to achieve improvements in the

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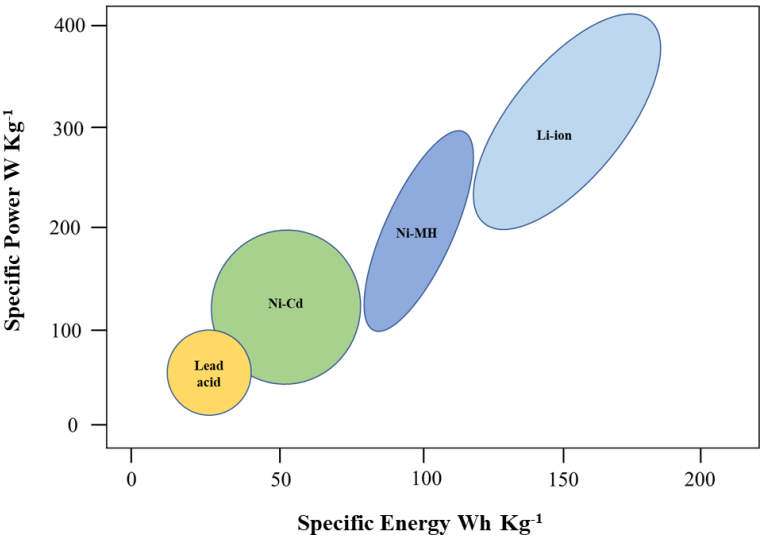

Figure 1. Ragone plot comparing multiple energy systems.

rate of cell performance with respect to cycle life, high capacity, energy density, and capacity retention. Recently, the research trend on Li-S addresses electrode modification and the design of sulfur-based cathode composites. Wu and Chung [3] devised a hot-pressed electrospun cathode with high loading of sulfur for a lean electrolyte cell. This design realizes low-weight conductive carbon, which provides a porous space for hosting the active material, attaining 8 mg cm^{-2}, and a high content of sulfur of 73 wt %. And they reported that the adoption of this method attains the high cell performance of 740 mAh g^{-1}, with outstanding rate performance of a prolonged cycle life of 200 cycles at 0.1–0.3 C.

The multifunctional self-supporting carbon nanobelt was introduced by the ion-beam sputtering deposition method. Hence, the biomass-derived hyphae carbon could enhance the loading to 4.6 mg cm^{-2} sulfur, which can deliver a retention capacity of 77 % for 400 cycles at 0.5 C. As an evidence of cycling performance, Li-S batteries have achieved a remarkable areal capacity of 9.8 mAh cm^{-2} at 0.1 C, and the capacity of 7.3 mAh cm^{-2} was maintained even after 60 cycles. This novel design strategy facilitates the redox kinetics of new sulfur species for long-life span Li-S batteries [4].

Recently, researchers have started to investigate the electrochemical characteristics of practical sulfur cathodes to achieve the high loading of sulfur of over 5 mg cm^{-2}, a sufficient sulfur content of 60 wt %, and a low electrolyte-to-sulfur ratio of 3–11 µL mg^{-1}. Cheng and Chung [5] revealed that chemical plating of a nickel nanoshell on the surface of sulfur particles traps the liquid-state active material and improves the electrochemical characteristics. ■pls check■ Electroless nickel-plated sulfur nanocomposites showed improved Li-S cell performance by their high sulfur content of 74 wt % and high loading of sulfur at 14 mg cm^{-1}, with a low electrolyte-to-sulfur ratio of 7 µL mg^{-1}. Therefore, this cathode attains high areal capacities of 7–14 mAh cm^{-2} and energy densities of 13–28 mWh cm^{-2}, greater than those of commercial Li-ion battery cathodes (2–4 mAh cm^{-2} and 10–14 mWh cm^{-2}).

2 Roadmap of Li-S batteries

The Li-S battery falls under the rechargeable batteries with remarkable specific energy and energy density. It has lithium as the anode and sulfur as the cathode. The lower atomic weight of the elements involved facilitates a wider range of applications due to the reduced overall weight of the battery. The batteries carry the additional advantage of reduced cost. The history of Li-S batteries, the developments in the field, and the never-ending extensive research on the topic have come a long way, as shown in Fig. 2, which describes the advancements in research on Li-S batteries. In 1962, Danuta Herbert and Juliuz Ulam patented a battery that contained lithium as its anode and sulfur as its cathode, both being employed in dry cells and storage batteries [6]. The electrolytes of choice were iodide, alkaline earth perchlorate, bromide, sulfonamide, or chlorate being dissolved in 1°, 2°, or 3° saturated aliphatic amine. An additional component to the existing topic was introduced in 1966 when Herbert patented an electrolyte that distinguished Li salts dissolved in amyl, butyl, or propyl amine as its components [7].

The solvent to be used in combination was found to be isopropyl amine. In the same year, Rao [8] patented high-energy density metal-sulfur batteries. The electrolyte was examined to contain ammonium/light metal ions as cations and tetrafluoroborate or tetrachloroaluminate as anions. The battery exhibited a voltage ranging from 2.5 to 1.1 V. The standardized usage of aqueous electrolytes was challenged in 1970 by the findings of Moss and Nole [9] when they introduced and patented non-aqueous electrolytes. Technological improvements in the later years produced organic solvents such as propylene carbonate (PC), dimethyl sulfoxide (DMSO), and dimethylformamide (DMF), yielding a battery voltage of 2.3–2.5 V [10, 11]. The 1980s marked the introduction of rechargeable Li-S batteries employing ethers with 1,3-dioxolane (DOL) as the electrolyte solvent. In the following years, research regarding Li-S batteries was stunted due to the absence of reliable results, which subsequently hindered any progress in the application and advancement of the battery. A modification was brought to the status of Li-S batteries in 2009 when Linda F. Nazar published her work in *Nature Materials* [12]. This aided in attracting the interest of researchers towards the field of Li-S batteries and, hence, the topic was revisited. The growth in the development of Li-S batteries was proven by the 2500 papers that were published and 70 000 citations of the same, as demonstrated by the data on the Web of Science.

With their advantages, the Li-S batteries also brought the main cause of degradation and the greatest disadvantage: polysulfide shuttling. This is the phenomenon where LiPS (Li_2S_x, where the value of x ranges from 4 to 8) are formed in the cathode and proceed to leak. Due to their high solubility, they diffuse to the anode and, upon reaction, become short-chain polysulfides. Upon migration to the cathode, they get converted to long-chain polysulfides, thus leading to their deposition on the cathode. This causes low coulombic efficiency,

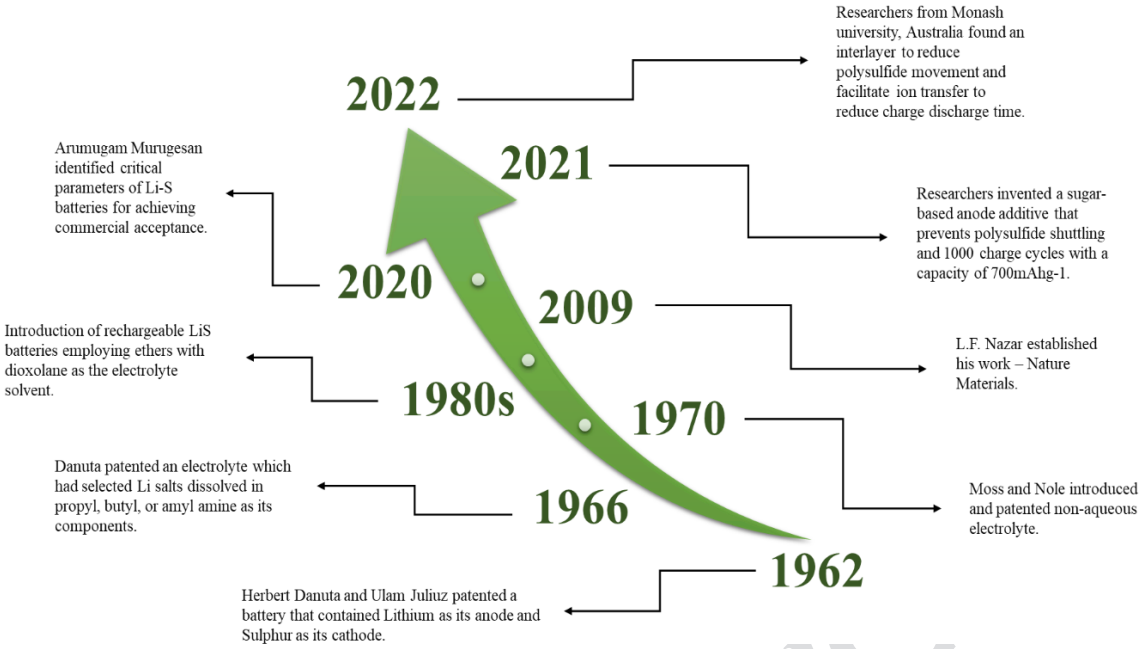

Figure 2. Progress in research on Li-S batteries.

lithium corrosion, and low battery life. This phenomenon was a barrier to the commercial application of Li-S batteries and kept numerous researchers busy to find a solution that overcomes the problem. Xu et al. [13] put forth a statement saying that polysulfides are beneficial for cell life. They further explain in *Advanced Energy Materials* how the issue of polysulfide shuttling can be contradicted by introducing electrolytes containing polysulfides, which leads to the precipitation and dissolution of Li-S polysulfides at the electrode interface. The most outstanding progress in the story of Li-S batteries is the identification of a particular electrolyte formation suitable for Li-S batteries [13].

The solvent-salt combinations were found to be sulfone based [14–16], DMSO [17–19], DMF [20], 1,2-dimethoxyethane (DME) [21], and polyethylene oxide (PEO) polymer electrolytes [22–26]. A solvent found to be widely applied is DOL [27]. Electrolytes can also be used in solid states, e.g. $LiS\text{-}SiS_2$ [28], $LiS\text{-}P_2S_5$ [29], and thio-lithium superionic conductor (LISICON) [30].

Another finding to battle polysulfide shuttling was the addition of additives such as $LiNO_3$ to the electrolyte to safeguard the Li anode species. It was proven via usage of an ether solvent that aids in the creation of a solid electrolyte interface (SEI), thus preventing any parasitic reaction between Li and LiPS [31]. After extensive research, the "four-electrode Swagelok cell" was used to enable in situ cyclic voltammetry for quantitative and qualitative analysis. It reveals the numbers of soluble polysulfides as far as quantitative measurements are concerned, and the sulfur discharge mechanism was studied for qualitative measurements [32–34]. Numerous researchers studied the performance and characteristics of the battery. Barchasz et al. [35] studied the discharge mechanism using high-performance liquid chromatography (HPLC), electron paramagnetic resonance (EPR) spectroscopy, and UV-Vis spectroscopy as characterization tools. Gao et al. [36] studied the influence of the electrolyte composition.

The results displayed that the solvents affected the Li-S electrochemical performance more than the salts. As another observation it was found that soluble polysulfides remain oxidized in ethereal solvents and get completely reduced in low-viscosity solvents. The performance of the battery and its characteristics can be predicted innovatively through modeling, which has emerged as a new research line. The years 2004 and 2008 marked the participation of Mikhaylik and Akridge [37] and Kumaresan et al. [38] in developing models of Li-S batteries to predict the cell life and state of health. Recent years have recorded appreciable advancements in Li-S batteries. Although modifications have been brought by many researchers, commercial application of the battery is still to be dealt with. In 2020, Bhargav et al. [39] identified critical parameters for achieving commercial acceptance. The critical parameters include a sulfur loading exceeding $5\,mg\,cm^{-2}$ along with a carbon content of less than 5 % plus an electrolyte-sulfur composition ratio lower than $5\,\mu L\,mg^{-1}$, and an electrolyte-to-capacity ratio lower than $5\,\mu L\,mAh^{-1}$ can promote the batteries for commercial application [39, 40]. In 2021, researchers invented a sugar-based anode additive that prevents polysulfide shuttling, and 1000 charge cycles with a capacity of $700\,mAh\,g^{-1}$ were observed [41]. In 2022, researchers from Monash University, Australia, found an interlayer to restrict polysulfide migration and enable ion transfer to decrease the charge-discharge time [42].

In the same year, researchers used aramid nanofibers designed into networks like a cell membrane structure to hinder the growth of dendrites. The phenomenon of ion selectivity tackled polysulfide shuttling by confining minute channels into the structure plus an electric charge [43].

The most recent finding was brought about by professors at Drexel University, in the form of a prototype of a Li-S battery

containing monoclinic gamma phase sulfur that did not degrade over 4000 cycles [44]. Constant and persistent research is being performed on enhancing the properties of Li-S batteries, overcoming the disadvantages and making them commercially applicable.

3 Mechanism of Li-S Batteries

Li-S batteries employ lithium as the anode and sulfur as the cathode, involving a discharge mechanism followed by a charging mechanism, and the reactions occurring at the electrodes can be seen in Fig. 3.

Based on the Li-S redox process, the entire electrochemical reaction is

$$16Li + S_8 \rightleftharpoons 8Li_2S \quad (1)$$

Reaction at the anode [41]:■Is this [45]?■

$$Li \rightarrow Li^+ + e^- \quad (2)$$

Reaction at the cathode:

$$2Li^+ + S + 2e^- \rightarrow Li_2S \quad (3)$$

Lithium ions from the anode react with sulfur to generate Li_2S as the end result. This is also known as the reduction of sulfur to Li_2S; however the entire reaction process comprises several distinct stages. During the discharge mechanism, lithium ions disintegrate from the negative electrode (anode), then diffuse via the electrolyte and migrate to the positive electrode (cathode).

$$S_8 + 2e^- + 2Li^+ \rightarrow Li_2S_8 \quad (4)$$

$$3Li_2S_8 + 2e^- + 2Li^+ \rightarrow 4Li_2S_6 \quad (5)$$

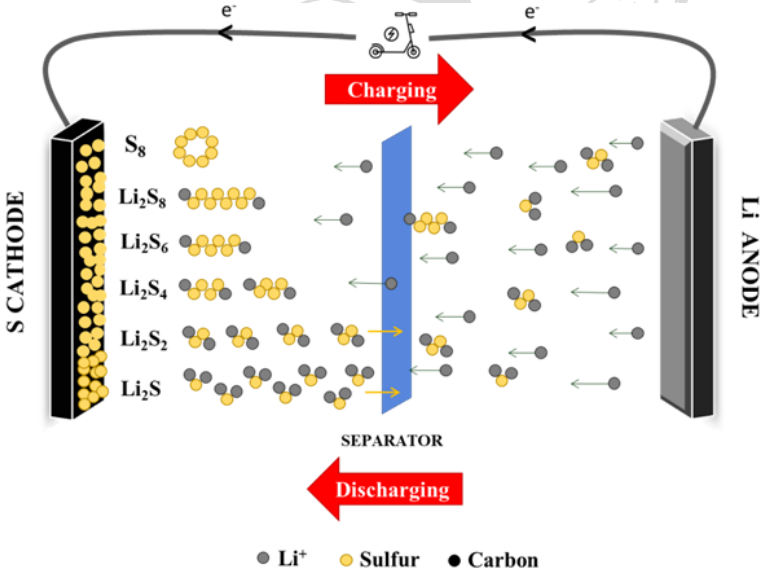

Figure 3. Mechanism of Li-S batteries.

$$2Li_2S_6 + 2e^- + 2Li^+ \rightarrow 3Li_2S_4 \quad (6)$$

$$2Li_2S_4 + 4e^- + 4Li^+ \rightarrow 4Li_2S_2 \quad (7)$$

$$4Li_2S_2 + 8e^- + 8Li^+ \rightarrow 8Li_2S \quad (8)$$

Sulfur initially gets reduced to form LiPS (Li_2S_x, $1 \le x \le 8$) [46], a rather threatening compound that impacts the performance of the battery and the overall functioning. A few hypotheses on the discharge mechanism show that LiPS are produced gradually after the first dissolution of the solid-state (S_8) elemental sulfur. Organic solvents are the most commonly used electrolytes, wherein LiPS of higher order (Li_2S_x, $4 \le x \le 8$) get soluble, whereas intermediate products like lower-order LiPS are insoluble in the electrolyte. As the cell discharges, sulfur is reduced in organic solvents as various chemical species such as polysulfide anions and radicals (S_8^{2-}, S_7^{2-}, S_6^{2-}, S_5^{2-}, S_4^{2-}, S_3^{2-}, S_2^{2-}, S^{2-}, $S_3^{-\bullet}$, $S_2^{-\bullet}$, and $S^{-\bullet}$), which weakens the polysulfide chain and results in the formation of lithium sulfide [47].

During the charge cycle, oxidation of sulfur occurs at the cathode and, upon charging, the Li^+ ions move towards the anode. At the anode, the reduction of Li-ions takes place. The dissolution of Li^+ ions and the following electrodeposition gradually promote unpredictable development of the SEI. This creates active sites for the occurrence of nucleation and enables the dendritic development of lithium. Short circuits occur in Li-S batteries due to the expansion of lithium dendrites, which finally causes the death of the battery itself [48]. Also, the polysulfide gets deposited on the electrodes and restricts the movement of Li^+ ions, thus negatively affecting the functioning of the battery.

4 Traits of Li-S Batteries

Certain major and most promising traits of Li-S batteries are their elevated energy density, low cost, and the bountiful nature of sulfur material. The edge over others listed is possible when Li-S batteries use sulfur in its elemental form as active material in the cathode and allow the sulfur to proceed towards the theoretical capacity with minimum process cost [49–57].

Sulfur is the 17th most plentiful element in the Earth's crust. The melting point of sulfur is 112.8 °C (rhombic) and 119 °C (monoclinic); it has a gravity density of 2.07 g cm^{-3} (rhombic) and 1.957 g cm^{-3} (monoclinic) at 20 °C and undergoes sublimation effortlessly. It is a fragile, odorless, pale yellow solid, insoluble in H_2O but soluble in CS_2 [54]. A multitude of issues arise concerning the sulfur cathode in batteries as it is an insulator of electricity: a higher quantity of carbon material is required for utilization of the active material [57, 58]. The major issue with lithium metal as the anode in Li-S batteries is the lower coulombic efficiency, corrosion, and the lithium plating, ■pls check deletion of "analysis of"■ which leads to exploitative reactions [59–61]. Requirements for

electrolytes include elevated ionic conductivity, a large electrochemical window, reliability, safety, and low viscosity [62].

Fig. 4 represents the elementary components, namely: cathodes, anodes, separators, and electrolytes, and their respective roles in Li-S batteries.

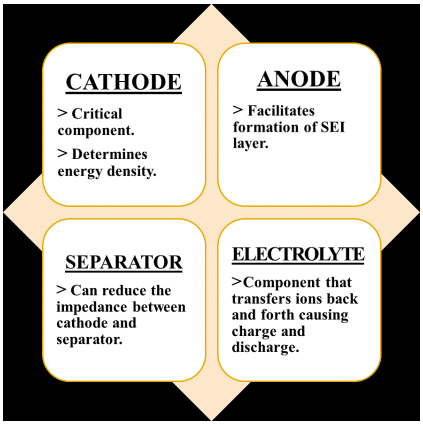

Figure 4. Components and their roles in Li-S batteries.

5 Roles of the Components in Li-S Batteries

5.1 Role of the Cathode Materials

In Li-S batteries, lithium plays a vital role as the anode and sulfur takes the role of the cathode material. The electrode materials can be modified by introducing silicon, tin, and metal oxides for the former and Li_2S for the latter [63].

Sulfur as a cathode material is advantageous as it contributes to an elevated energy density and a very high theoretical specific energy; it is economical and environmentally considerate. In parallel, it is considered disadvantageous due to the non-conducting nature of the active material, polysulfide formation, dissolution, and the shuttling effect, and to the volumetric expansion of sulfur. The undesired characteristics of sulfur can be battled by combining the material with a carbon material, such as porous carbon [64–68], graphene [69–73], carbon nanotubes [74–77], and carbon spheres [78], and conductive polymers [79–81], where metal organic frameworks (MOF) and their usage in battling polysulfides are also up and coming [82, 83]. The choice of the cathode material plays a significant role as it influences the conductivity, the shuttle effect, and the anchoring of polysulfides. Carbon materials are employed to present an electron and an ion with an adequate space for the reaction of non-conductive sulfur. The carbon-based materials should possess a proper electron/ion migration path and contain sufficient volume to house the volumetric expansion of sulfur in the electrochemical reaction. The key parameters and the aid provided by the cathode materials are listed as sulfur content, discharge capacity, and coulombic efficiency, and the capacity retention of various materials such as polyaniline (PANI), graphene oxide (GO)-S, and reduced graphene oxide (rGO)/S/PANI in Tab. 1.

5.1.1 Conductivity

Research has been performed with various carbon materials to study their effects on the conductivity of a battery. As stated, graphene, carbon nanotubes, and carbon spheres are used. Generally, graphene is integrated with carbon nanotubes to build electron/ion transport channels of various ranges in length [90], and carbon materials derived from biomass are also used [91–95].

A study of the role of carbon materials revealed that they initially improve the conductivity but do not provide an optimal solution since a reduction in energy density was also observed. Scientists [39] regarded it important to reduce the carbon content in cathodes where the ideal and desirable quantity is less than 5 %. Carbon as one of the cathode materials aids in the betterment of the porosity, wettability, and interaction with polysulfides.

5.1.2 Mitigation of the Shuttle Effect

Polysulfide shuttling is a phenomenon that scientists have been battling against through extensive research. It includes the formation of polysulfides in the battery and the growth of the same into long-chain polysulfides upon charge and discharge. This negatively affects the parameters and functioning of the battery. Various research efforts on cathode materials, electrolytes, and catalysts can help in the physical anchoring

Table 1. Empirical parameters associated with various cathode materials for Li-S batteries.

Material	Empirical data	Ref.
PANI@S/C	Sulfur content 43.7 wt %, discharge capacity 635.5 mAh g^{-1} at 10 C rate, coulombic efficiency 91.7–103.2 %, capacity retention 60 % after 200 cycles	[84]
GO-S	Sulfur content 66 wt %, discharge capacity 1320 mAh g^{-1} at 0.02 C rate, coulombic efficiency 96.4 %, capacity retention 60 % after 200 cycles	[85]
rGO/S/PANI	Sulfur content 51 wt %, discharge capacity 809 mAh g^{-1}, capacity retention 74 %	[86]
GO@S/C	Sulfur content 51 wt %, discharge capacity 1433 mAh g^{-1}, capacity retention 37 %	[87]
CMK-3/S/PEDOT:PSS	Discharge capacity achieved 1140 mAh g^{-1}, coulombic efficiency 93–97 %, capacity retention 80 % after 100 cycles	[88]
CMK-3/S@PANS@TPS	Discharge capacity 1246 mAh g^{-1} at 0.25 C rate, coulombic efficiency 98.2 %, capacity retention 89 % after 100 cycles	[89]

of polysulfides and the chemical anchoring of functional groups.

In recent studies, a promising way emerged to mitigate the shuttle effect in Li-S batteries by single-atom catalysts whose catalytic efficiency is higher in the conversion of lithium polysulfides. A novel S@Co-P cluster/NC cathode (NC, N-doped carbon) is a dual-atom site whose catalytic mechanism is coupled with moderate chemical absorptivity which significantly accelerates the conversion of lithium polysulfides. This innovative material demonstrated good cycling performance of roughly 200 cycles at a low rate of 0.2 C, with a reversible capacity of 1015 mAh g^{-1} and capacity retention of 81.8 % at an elevated sulfur content of 6.2 mg cm^{-2} and high areal capacity of 6.5 mAh cm^{-2} [96].

Shuttling effect mitigation enhances the cycle lifespan of the Li-S batteries, which would use encapsulating polysulfide electrolytes (EPSE) to suppress the parasitic reactions occurring in the battery. Using 1,1,2,2-tetrafluoroethyl-2,2,3,3-tetrafluoropropyl ether (HFE), a superior outer shell solvent, those EPSE can be constructed. These HFE-EPSE-based Li-S batteries showed an extended lifespan from 54 to 135 cycles with a sulfur cathode of about 4.4 mg cm^{-2} loading and an ultrathin-film lithium-metal anode. This study sets forth the importance of polysulfide solvation chemistry for the construction of improved EPSE and highlights the role that weakening the solvating power of the outer shell solvents plays in the development of viable Li-S batteries [97]. Analogously, a redox co-mediator, dimethyl diselenide (DMDSe), helps to facilitate the sulfur redox kinetics in Li-S batteries with EPSE. The efficacy of DMDSe lies in reducing the anode parasitic reactions of LiPS while it improves the liquid-liquid and liquid-solid conversion kinetics. As an end result, a Li-S pouch cell with a steady 37 cycles and a high energy density of 359 Wh kg^{-1} is achieved [98].

When the cathode material and liquid electrolytes help in the mitigation process, an organic-based SEI also has its equal part in the conversion of polysulfides. The restricted cycle life time of Li-S batteries is caused by parasitic interactions between the LiPS and the Li metal anodes; therefore, construction of an organic-based SEI using 1,3,5-trioxane, a reactive co-solvent, is vital. This solvent gets distributed ■pls check■ on the surface of the Li metal anode and protects it from interactions of the LiPS parasitic reactions, thus enabling a long cycle life span of Li-S batteries. As a result, the organic-rich SEI increases the cycle life of Li-S coin cells with 50-μm Li anodes and 4.0-mg cm^{-2} sulfur cathodes from 130 to 300 cycles. Additionally, a 3-Ah-level Li-S pouch cell attained a high energy density of 400 Wh kg^{-1}, owing to the organic-rich SEI [99]. Another research work using a Nafion protective layer showed a good cycle lifespan, which was doubled to 92 cycles and aided in maintaining the lithium metal anodes in functioning Li-S batteries [100].

Although the Li-S battery has huge potential as an energy storage device and it is considered to enhance the cycle life, capacity retention, and general performance of Li-S batteries, the shuttle effect must still be minimized. To make these batteries more useful for a wide range of applications, such as electric vehicles and grid energy storage, researchers are still investigating these techniques and creating novel materials.

5.1.3 Physical Anchoring of Polysulfides

Pore size reduction of the carbon material in the cathode used helps prevent the return of sulfur to its elemental state (S_8) but keeps it converted to S_4 or S_2, and thus long-chain polysulfides cannot be formed. Xin et al. [101] performed an experiment where carbon with a micropore size of 0.5 nm was used in place of a sulfur host in the cathode. This displayed outstanding results with a capacity retention rate of 200 cycles at 0.1 C and a specific capacity of 800 mAh g^{-1} at a higher rate (5 C). Microporous carbon is said to restrict sulfur to its S_2 and S_4 forms whereas mesoporous carbon provides sufficient space for S_8 molecules, thus extensively preventing long-chain polysulfide formation [102].

Moving to total sulfur loading, the size of the micropores in carbon materials has a lower significance for and influence on the performance and capacity of the battery. In certain cases, the particle size may enable appreciable contact between the active components and encourage the movement of electrons or ions, which subsequently increases ■pls check■ the utilization of sulfur. This may lead to dangerously high contact with the electrolyte, thus causing excessive dissolution of polysulfides. There exists a particle size that is known as the golden particle size, in the case of ZIF-8, a MOF. It is approximately equal to 200 nm and brings sulfur utilization and polysulfide dissolution into equilibrium [103].

5.1.4 Chemical Anchoring of Functional Groups

The polarity of functional groups used can be altered to alleviate the shuttle effect as a chemical reaction between the groups, and polysulfides can control the shuttle effect. Cathode materials that possess good conductivity can have their surface modified to control the polysulfide migration to the electrodes. The conductivity can be modified as demonstrated by Qiu et al. [104] who treated reduced graphene with ammonia. In another case, Luo et al. [105] employed a carbon matrix with sufficient oxygen-based functional groups in the place of the sulfur host. This increased the stability of the sulfur due to its interaction with oxygen and over 2000 cycles produces a reversible capacity of 500 mAh g^{-1}.

5.2 Role of the Anode Materials

Li-S batteries are greatly impaired by poor coulombic efficiency and shortened lifespan, leading to polysulfide shuttling and out-of-control growth of lithium dendrites. The increase in dendrite creation needs to be suppressed and the interconnection between dissolvable polysulfides and lithium must be hindered, which is crucial for a sheltered and effective functioning of the anode material and also for elevated capacity in Li-S batteries. Tab. 2 represents the functions of a variety of anode materials, namely, Li-B, nanostructured lithium sulfide, Si-O_x-based anodes, and carbon materials.

The carbon material utilized in Li-S batteries acts as conductive additive but also as preventer of the shuttle effect, spatial protector, and anode protector [110].

Table 2. Various anode materials and their functions.

Anode material	Functions	Ref.
Li-B alloy as anode	Restraining the creation of dendritic growth, bringing down the blockage in the interlayers of electrode	[106]
Nanostructured lithium-sulfide materials	Restraining the creation of dendrites, bringing down the electrode interface blockade	[107]
SiO_x-based anodes	Superior charge-discharge properties	[108]
Carbon materials	High availability of sulfur, low cost, and sustainability with regard to the environment	[109]

For more than a century, technologies such as Pb-acid and Ni-metal batteries have been used in various applications like automobiles and drones [111–113]. Li-ion batteries may have taken over the portable electronics market, despite their limiting charge storage capacity and rapid attainment of theoretical ranges; but other devices have been explored for higher energy density, cost-effectiveness, and good cycle life [114–116].

Metal lithium is a highly important component in Li-S batteries as it pushes for high energy because of its low gravimetric density, quick recharge ability, low density, and electrochemical potential [117].

The depreciation of lithium metal as an anode is a huge issue that combines the consequences of reactivity in the anode, the operating conditions, and the type of electrolytes used. The development and amplification of lithium microstructures on the surface of lithium during cycling is a regular occurrence when the electroplating of metals occurs at a high current [118–120]. Electrochemical deposition of lithium and the creation of dendrites have been evaluated in the past, and models based on theory showing the deposited lithium microstructures have been explored [121–123]. The most important consequence of the unchecked growth of lithium microstructures is the presence of dead lithium, which may lead to safety problems and capacity loss [124]. Electronic and ionic entrapment of the dead lithium occurred throughout the creation and growth of a robust solid electrolyte interphase during repetitive cycles. Inactive lithium obstructs the movement in the metal anode, accelerates the creation of dendrites and reduces the capacity of the device [125].

Due to a low reduction potential, unfavorable interactions between the lithium metal, electrons on the surface, and neighboring electrolyte species occur during the assembly of the cell, which leads to the creation of a SEI with versatile mechanical properties [126, 127]. Ideally, SEI layers have high electrical resistance and ionic conductivity, a wide temperature stability range, and low electrolyte solubility [128–130]. Typically, SEI films are fragile due to their instability, which is a crucial issue. The porosity and randomness of the layer allow the phenomenon of side chain reactions at particular locations, leading to the deposition and dissolution of lithium [131–134].

There are methods towards the protection of lithium metal anodes through electrolyte modification-based approaches like liquid electrolytes, additives, ionic liquids, polymers, and inorganic solid-state electrolytes [135–144].

5.3 Role of Separators

Desirable characteristics of a separator include high stability, longer life span, high efficiency, electrochemical, dimensional, thermal, and mechanical stability, and high permeability towards the electrolyte [145–147]. Commonly used separator materials are polyethylene, polypropylene, and their combination [148]. Functional separators are responsible for the separation of the anode and the cathode by restricting the physical contact between them while enabling ion transport in the cell [147], and their classification is shown in Fig. 5.

There is no electrochemical activity involved between the membrane and the electrolyte. Physical features that define a separator are its thin and porous design. Functional separators are an improvised design where a functional group is introduced as a coating on the separator and is expected to provide additional properties to tackle existing issues in a battery [146–148]. The design of advanced functional separators involves three primary parameters. Firstly, the separator thick-

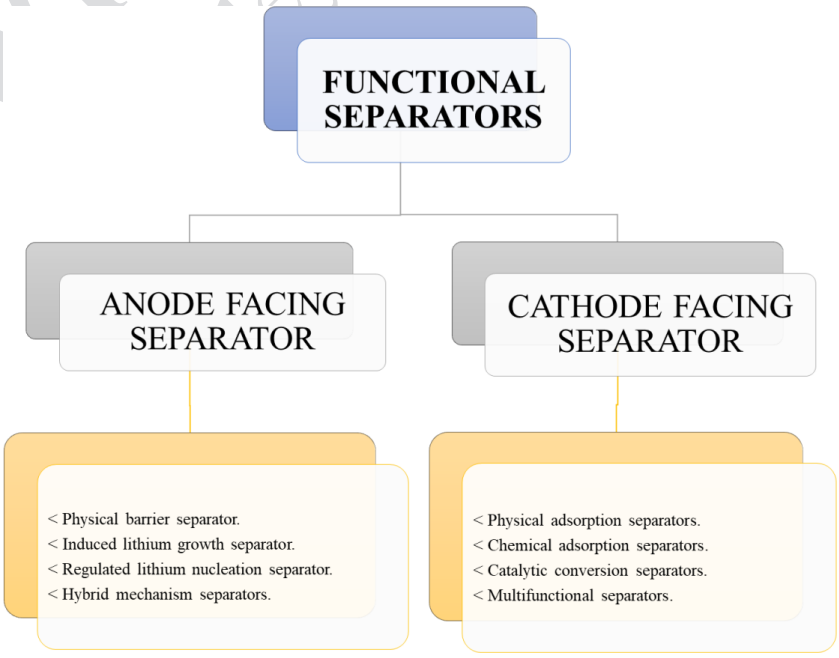

Figure 5. Classification of functional separators.

ness influences the ion transport and rate capability. Secondly, the separator weight influences the energy density. And thirdly, the porosity of the separator affects the electrolyte uptake and thus affects the energy per unit volume along with the cycle life of the battery [147].

Researchers have invested efforts in improvising functionalized separators in terms of design, the construction material, and the preparation process. Noticeable improvements in this regard include the "plasma-functionalized carbon-layered separators for enhanced performance of Li-S batteries". The membrane is initially irradiated with CO_2 plasma; is it then coated using a thin carbon layer and is finally treated with plasma to provide modifications to the carbon layer. The positive results observed were increased wettability, electrical conductivity, the presence of active sites that enable redox reactions and transport of ions, and decreased polysulfide shuttling. The initial capacity was observed to be 1204 mAh g^{-1}, with a remaining capacity of 802 mAh g^{-1} [146].

Necessary parameters such as initial capacity, remaining capacity, discharge capacity, fade rate, and capacity retention are compared in Tab. 3 for various separators. Continuous efforts are being made to improve on this matter and control well-known issues such as polysulfide shuttling, inadequate sulfur utilization, and lower cyclability, and hence enable the industrialization of Li-S batteries.

5.4 Role of the Electrolytes

The electrolyte is one of the constituent parts of a battery; it transfers ions between the two electrodes, which causes the cycle of both charging and discharging in batteries. Electrolytes can have billions of combinations of salts, solvents, and additives. The electrolyte functions as a catalyst to make the battery conductive by pushing for the movement of ions between the electrodes [53]. Tailoring of electrolytes in Li-S batteries plays a major role in battery chemistry. When the cathode hosts' capacity for adsorption is insufficient to anchor the LiPS, they will separate from the cathode surface and move towards the anode under the influence of their concentration gradient. Therefore, evading the generation of polysulfides with novel electrolytes (both liquid and solid electrolytes) could mitigate the shuttle effect.

Liquid electrolytes, such as ether-based electrolytes, are commonly used to reduce the solubility of LiPS. The assembled batteries with fluorinated diether (FDE; 1,3-(1,1,2,2-tetrafluoroethoxy)propane) electrolyte exhibited a specific capacity of 701 mAh g^{-1} at 0.5 C and retained an average of 99 % coulombic efficiency even after 200 cycles. Furthermore, insertion of fluorinated ethers into the lithium bis(fluorosulfonyl)imide (LiFSI)/DME electrolyte could minimize the solubility of LiPS by delivering superior performance of 775 mAh g^{-1} at 0.05 C after 150 cycles, with the formation of a LiF-rich SEI film on the lithium metal surface [151].

Solid electrolytes are introduced in all-solid-state lithium-sulfur batteries (ASSLB) as they have tremendous prospects for newer-generation storage systems because of their elevated specific capacity, eco-friendly nature, and lower cost. This includes the use of solid electrolytes, which are inorganic, polymeric, and composite electrolytes [152]. Quasi-solid-state gel polymer electrolytes (GPE), which involve the integration of liquid electrolytes into solid polymer matrices, have been explored, for which outstanding ionic transport and stunted interfacial resistance provided by the GPE are parts of the progress made in Li-S batteries. There are challenges that restrict the renewal of organic carbonate-type electrolytes with $LiPF_6$ as a conducting salt. The development of stable electrolytes that work in high voltage ranges such as 5 V is a challenge that has been tackled by the advanced systems of electrolytes involved in film-forming high-voltage additives and new solvents [153].

6 Polysulfide Adsorption Test

Lithium-sulfur batteries possess an appreciably high theoretical capacity of 2500 Wh kg^{-1}, whereas Li-ion batteries possess a theoretically calculated capacity of 420 Wh kg^{-1} [154–156]. Along with this quality, the low cost and abundance of sulfur showcase Li-S batteries as an attractive innovation. There exists a hindrance to the wide application of Li-S batteries: polysulfide shuttling and the resulting degradation of the batteries. Polysulfides are the intermediates formed in the redox reaction where sulfur is converted to lithium sulfide [157–160]. Multiple innovations have been put forth to encounter the formation of polysulfides. The findings include the installation of hosts with higher surface area and a porous structure such as hollow carbon spheres [161, 162]. The hosts exhibited satisfying performance over the first few hundred cycles and ceased to do so over the rest of the cycles, when the familiar degradation and polysulfide presence were observed. This was because of poor interactions between the nonpolar host material and the polar polysulfides. Further, chemical reactions were sought after to overcome this issue of insufficient interaction, where metal oxides, such as, e.g., Al_2O_3, SiO_2, TiO_2, and MnO_2, were considered along with metal sulfides and metal nitrides [163–173]. The efficiency of these hosts can be measured by estimating the

Table 3. Various separators employed in Li-S batteries and their experimental results.

Separator	Experimental results	Ref.
Plasma-functionalized carbon-layered separators	Initial capacity 1204 mAh g^{-1}, remaining capacity 802 mAh g^{-1} (cycles: 100, rate 0.2 C)	[146]
Multiwall carbon nanotubes	Discharge capacity 1324 mAh g^{-1}, fade rate 0.14 % per cycle	[149]
PMIA/octaphenyl-POSS membrane (HPPS)	Initial capacity 900 mAh g^{-1}, capacity retention 67.8 % after 500 cycles	[150]

polysulfide adsorption capacity, which can be assessed by a set procedure, as explained below.

Seven metal oxides, seven respective metal sulfides, and a metal nitride were chosen as subject materials for the test [174, 175]. The Brunauer-Emmett-Teller (BET) theory was applied to the materials for normalization of the surface area. Preparation of the sample was carried out as follows. Candidate materials with a surface area of 0.5 m^2 were added to Li_2S_6 with a molarity of 3 mM in DOL and DME solution of 4 mL for a duration of 3 h. Li_2S_6 is a long-chain polysulfide formed when sulfur is subjected to a chemical reaction with Li_2S in a solution of DOL/DME. For BET assessment, the candidate material of 200 mg was heated to 120 °C and then degassed for a period of 10 h, and the surface area and porosity were then analyzed. For the adsorption test, the Li_2S_6 prepared was stirred at a temperature of 70 °C overnight in an air-filled glove box to provide a Li_2S_6 solution of a brownish red color and a concentration of 0.25 M, which was then diluted to 3 M ■3 M is too high■ for the test. The candidate samples were dried overnight at 80 °C. For UV-Vis spectroscopy, 2 mL of the prepared solutions were extracted and restricted from contact with other entities. UV analysis was performed using an Agilent Cary 6000i UV instrument. For inductively coupled plasma atomic emission spectroscopy (ICP-AES) measurements, $CuSO_4.5H_2O$ and $LiNO_3$ were dissolved in 200 mL deionized water for calibrating S and Li for their intensities to account for offsets. Li_2S_6 solutions of concentrations varying from 0.5 to 3.0 mM were diluted in 10 mL deionized water. The solutions prepared for the polysulfide adsorption tests were also in 10 mL deionized water. The aforementioned solutions were analyzed in the ICP-AES measurement to figure out the intensities of S and Li. For the X-ray photoelectron spectroscopy (XPS) measurement, the candidate materials were centrifuged for easy separation. The desired subjects were placed in a vacuum chamber. A PHI Versa probe 1 scanning XPS microprobe system was used to perform the XPS analysis [176]. The results of the tests and analyses demonstrated the strong polysulfide adsorption capacity of MnO_2 and V_2O_5. Different host materials show different percentages of polysulfide adsorption, as seen in Fig. 6.

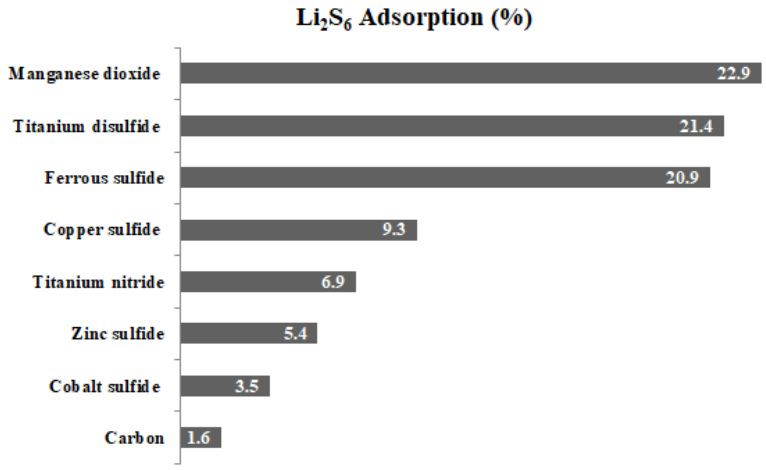

Figure 6. Polysulfide adsorption percentage on various host materials.

7 Li-S Batteries: Challenges and Solutions

Li-S batteries are a type of rechargeable battery that offers a multitude of benefits such as reduced raw materials cost, increased safety features, and reduced burden to the environment. But there are also problems associated with these batteries, like the full utilization of the active material, upkeep of the electrode structure, and a good cycle life with efficiency, as discussed in Fig. 7. These issues limit their usage for commercial purposes [177].

7.1 Flammability of Organic Electrolytes

Li-S batteries attract remarkable attraction due to their substantial theoretical capacities and energy densities. In spite of outstanding progress in the field, there are concerns about the safety during the usage of organic electrolytes. There are concerns about their flammability, toxicity, liquid leakage, and volatilization due to the presence of liquid organic electrolytes. There is also the possibility of dendrite formation during cycling, which may result in internal short-circuiting, leading to combustion and even the explosion of cells.

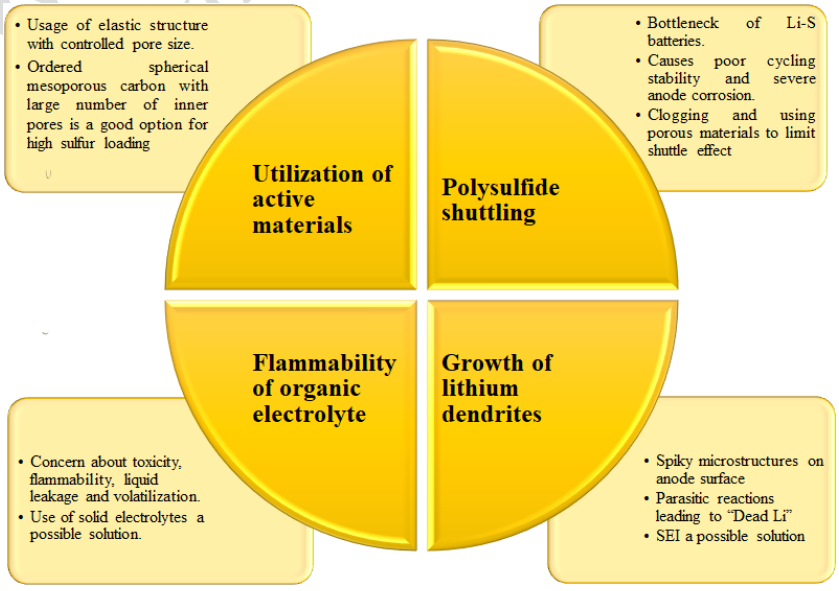

Figure 7. Drawbacks of Li-S batteries.

Of the solutions being explored, a potential answer is the usage of solid electrolytes, by substituting the conventional electrolytes with solid electrolytes, i.e.
- oxide-based solid electrolytes
- sulfide-based solid electrolytes
- Unfortunately, there is also a fair share of problems with solid electrolytes such as:
- low conductivity
- poor chemical stability in the presence of air
- unfavorable electrode/electrolyte interfaces

The preparation of an electrolyte-electrode surface with a large area of contact is described in [152, 153].

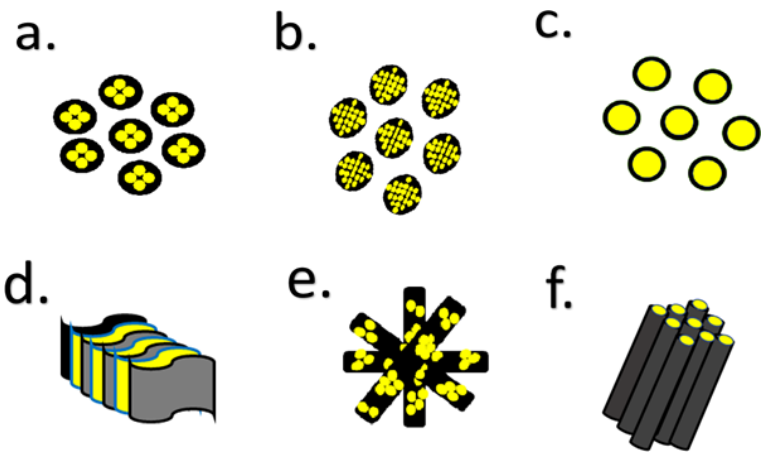

Figure 8. (a) Carbon materials in spherical shape (10^{-6}), ■pls explain 10^{-6}■ (b) carbon nanoparticles (uniform, 2–50 nm in diameter), (c) carbon (pores present, hollow), (d) sheets of GO, (e) carbon nanofibers (pores present), and (f) carbon nanofibers to accommodate sulfur (hollow).

7.2 Utilization of Active Materials

Consumption of the entire volume of active materials is a crucial challenge to overcome. The usage of a structure that is elastic in combination with a constrained pore size could be used to aid the changes in the volume of the active material.

Major attempts to boost the electrochemical performance of Li-S batteries are based on improving the carbon-based sulfur composite. Tab. 4 lists a number of composites synthesized by different methods to explore solutions. An ideal situation for a carbon-sulfur composite would have
- the affinity for hosting sulfur
- small pores without the presence of large outlets to accommodate polysulfides
- high electrical conductivity
- interaction of the liquid electrolyte and the active material

An ordered spherical carbon material, which is mesoporous, is a good variable to function as the matrix to contain higher amounts of sulfur with no effect on the performance. Carbon nanofibers, which are porous and hollow nanofibers, are amazing substrates to be integrated with sulfur. They exhibit outstanding cyclability over 100 cycles, as seen for the different hollow, porous structures in Fig. 8 [177, 178].

7.3 Polysulfide Shuttling

The shuttle effect happens when the sulfur species added reaches the negative side of the electrode surface and undergoes reduction. This is considered to be a bottleneck of the Li-S batteries. This effect results in poor cycling stability and severe corrosion at the anode of Li-S batteries. The diffusion movement of polysulfides back and forth between the anode and the cathode is called the shuttle effect [64].

At present, there are two approaches to limiting the shuttle effect:
- clogging the polysulfide migratory route by modifying the separator or by insertion of interlayers
- using porous materials to avoid the diffusion of polysulfide into electrolytes by anchoring the polysulfide on the cathode surface by adsorption [179]

An analytical method to combat polysulfide shuttling is the analysis of the adsorption capability of host materials used in the batteries.

7.4 Growth of Lithium Dendrites and Ways to Suppress Them

The anode is one of the crucial components of Li-S batteries responsible for the extended cycle stability of the battery. The main issue with lithium metal cells is that, during charging, spiky microstructures are formed in an irregular manner. Without the addition of organic compounds, which will result in brighter and smoother metal surfaces, the metal surface is dendritic and powdery. Formation of the dendrites is based on

Table 4. Composites synthesized by various methods can also be used to overcome the problem.

Method	Description	Integration of S and C	Features
Mixing	Integration of contents by stirring magnetically	Poor	–
Ball-milling	Usage of new/modified binder material	Moderate	Energy intensive
Thermal treatment	One- and two-step heating	Robust	Good mesoporous carbon

the mass transportation of metal cations that advance from the bulk electrolyte to the outer limit of the double layer, which is followed by electro-adsorption. The cations are then reduced to adsorbed atoms, which then diffuse and become incorporated into the metal lattice. With regard to Li-S batteries, the development of lithium dendrites causes safety issues and a low cycle life. A surface energy model has been proposed to explain the complex interrelation between the Li metal anode and the electrolyte. The breaking up of Li dendrites causes the phenomenon of "dead Li". There is also the possibility of exploding of the Li metal anode because of short-circuiting due to Li dendrites. The charge-induced growth model and the SEI model have been used to overcome this issue.

The charge-induced growth model is based on the Nernst equation.

$$E_{\mathrm{Red}} = E^{*}_{\mathrm{Red}} - \frac{RT}{nF} \ln\left(\frac{\alpha_{\mathrm{Red}}}{\alpha_{\mathrm{Ox}}}\right) \quad (9)$$

Other methods explored for Li metal protection are to permit Li to react with other materials to create a soft film such as a SEI film in order to prevent dendrite formation. Additives are added to electrolytes to prevent or retard dendrite formation. Nanostructures are used to bring about the deposition of Li by ultralow current density.

The lithium metal ions are subject to the Lorentz force because of electromagnetic fields which are pushed into a spiraling motion, which results in the magnetohydrodynamics (MHD) effect, which efficiently promotes the transfer of mass and the distribution of ions to inhibit the growth of dendrites and obtains the uniform deposition of lithium. Results show that electrodes within the magnetic field exhibit good cycling and rate performance in Li-S batteries [180–183].

8 Recent Advancements in Li-S Batteries

8.1 Novel Binders and Binder-Free Methods

The working of high-performance batteries depends on the optimization of their components, from electrolytes to binder systems. A variety of polyvinyl imidazolium-based nanoparticles are being used as binder components in cathodes of Li-S batteries. This results in a highly increased specific capacity and also an outstanding long-term electrochemical durability for 1000 charge-discharge cycles.

Carboxymethyl cellulose lithium (CMC-Li) can be used as a novel binder in Li-S batteries. CMC-Li is a novel water-based binder prepared by utilizing cotton as raw material. In comparison to the polyvinylidene fluoride (PVDF) binder, the battery with CMC-Li binder seems to retain 97.8 % of the initial reversible capacity after the first 200 cycles at 176 mAh g^{-1} [184–186].

The capacity specifications of binders such as MPVDF, CMC, and rGO/boron nitride (BN) and their descriptions are given in Tab. 5.

Monolayers of SnO_2 nanoparticles can also be used as a binder-free method, by uniformly stacking on the exterior surfaces of carbon nanotubes and bundles within the sheets, which are stacked across each other.

Novel polymers may be of use as binders that are conductive in nature for elevated-capacity anodes, which contain ■pls check■ silicon in Li-S batteries. This is done to explore issues such as quickly fading capacity and the impaired cycle life of silicon anodes because of their massive volume variation during continued cycling. The abundance of the carboxyl groups in the polymer chains can effectively increase the performance of the binding force to the silicon nanoparticles. The polymer binders have been classified as cellulose binders, conductive binders, and self-healing binders [188].

A composite of the novel polymer type called MPVDF has also been developed, by implanting maleic anhydride-grated PVDF into PVDF as a binder for lithium cobalt oxide in Li-S batteries. Comparative investigations of MPVDF and PVDF as binders have been done with scanning electron microscopy (SEM), X-ray diffraction (XRD), and electrochemical measurements.

Composite films such as rGO/BN have been created by filtration in vacuum, which is facile in nature, followed by thermal treatment. The rGO/BN composite film with a boron nitride content of 2 wt % is an anode material that is binder free, with an elevated reversible specific capacity of 278 mAh g^{-1} at a high current density of 100 mA g^{-1} and a very high rate capability and capacity retained over the first 200 cycles. The improved performance of the composite film is because of the unique structure and synergistic effects between the graphene and the layered boron nitride [189–192].

Table 5. Novel binders in Li-S batteries.

Novel binder	Description	Capacity specification	Ref.
Polyvinyl imidazolium-based binder	Nanoparticles	8 % capacity loss	[183]
CMC-lithium	Water based, using cotton as raw material	4.49 % capacity loss	[186]
SnO_2 nanoparticle monolayers	Stacked uniformly on exteriors of carbon nanotubes and bundles	Severe capacity fading, rapid aggregation of Sn particles, huge change in volume (over 300 %)	[187]
MPVDF	Implanting maleic anhydride-grated polyvinylidene fluoride in PVDF	Discharge capacity increases by 38.5 %, capacity retention improved from 84.5 % to 90.2 %	[188, 189]
rGO/BN composite films	Designed by vacuum method and followed by thermal treatment	After the first 200 cycles, capacity retention seems to be high	[190, 191]

8.2 New Modified Electrolytes

Novel electrolytes, their description, and advantages are compared in Tab. 6. Ceramic and polymeric electrolytes supply benefits in the context of design simplicity and safety during operation but have lower conductivities than organic electrolytes. Solid electrolytes remove the need for containment of liquid electrolytes, which improves durability. $Li_{1+x}Al_xGe_{2-x}(PO_4)_3$ (LAGP) and PVDF-co-hexafluoropropylene (PVDF-HFP) have the highest conductivities but are disadvantageous due to their mechanical strength and electrode compatibility [193].

Surface-modified inorganic salts are used as nanofillers in polymer electrolytes to improve their properties and their application in both non-rechargeable and rechargeable cells. The nanofillers consist of alumina (Al_2O_3) and titania (TiO_2) powders with super-acidic groups, which are made to enter the surface. They are then added to high- and low-molecular-weight PEO and $LiClO_4$. In this way, various composites can be obtained that exhibit good stability and a high lithium transference number. The use of polymer electrolytes is considered in order to solve safety matters due to the lithium dendrite growth in the electrolyte [194, 195].

An altered silyl-terminated polymer electrolyte by a new fabrication procedure has been proposed. Due to the three-dimensional (3D) network-like structure, the amorphous phase in the polymer electrolyte has an elevated ionic conductivity (0.36×10^{-3} S cm^{-1}), an increased thermal stability ($T = 379$ °C) and also a very high lithium ion transference number (0.65). This has become a promising candidate for realistic processes [196, 197].

The major matter in question with liquid electrolytes is the high level of solubility of the intermediates in liquid organic electrolytes, leading to the shuttling effect and an alarming loss of capacity. In comparison to electrolytes of a solid nature, conventional liquid electrolytes are the preferred choice in Li-S battery commercialization. There has been a focus to develop various host materials for sulfur as the cathode, safeguarding the lithium metal anode by the usage of SEI coatings and the advancement of the structure of Li-S batteries [196]. The current research findings based on liquid electrolytes in Li-S batteries are focused on designing a lean electrolyte cell, to ensure a high impact on the gravimetric capacity. As a result, further efforts have been made to design a 3D cathode made of a tunable carbon nanofiber network with limited porous substrate, which can deliver a high reversible discharge capacity of 802 and 607 mAh g^{-1}, with a capacity retention of 82 % after 200 cycles and a long shelf life of 3 months at rates of 0.2 and 0.1 C. Hence, in a lean electrolyte cell, limited nanopores prevent the utilization of the electrolyte, thereby achieving high electrochemical efficiency and stability to attain high areal and gravimetric capacities of 11.5 and 9.6 mAh cm^{-2} and 569 and 476 mAh g^{-1}, respectively 198].

Basic ionic liquids and Li-salt molten complexes are superb candidates for electrolytes, given their ability to greatly subdue the LiPS dissolution. Molten compounds do not readily dissolve solutes of an ionic nature, leading to promising operation of Li-S batteries for more than 400 cycles with a discharge capacity greater than 700 mAh g^{-1} sulfur and coulombic efficiencies greater than 98 % throughout the cycles. In addition to this, a fluorinated nonflammable solvent does not break the solvent structure, which greatly advances the power density of Li-S batteries [197].

GPE intermediates separating solid- and liquid-based electrolytes function not only as an electrolyte but also as a separator, which reduces the leaking of liquid (aqueous) electrolytes and increases the interfacial bulk resistance of solid electrolytes. In Li-S batteries, modification of manifold substrates such as PEO, PVDF, PVDF-HFP, poly(*m*-phenylene isophthalamide) (PMIA), and poly(methyl methacrylate) (PMMA) alleviates the severity of the polysulfides shuttling effect and hinders the enlargement of dendrites [199].

A newly designed gel-ceramic multilayer electrolyte serves as the electrolyte and is separated ■specific?■ for Li-S batteries. The Li-S cells, not restricted by the shuttle effect, have a higher electrochemical performance and an initial discharge capacity at a maximum of 725 mAh g^{-1} [200].

Preliminary studies have shown that the prepared composite gel polymer electrolytes (CGPE) can be implemented as likely electrolytes for Li-S batteries. Inclusion of plasticizers has significantly improved the ionic conductivity of the gel electrolytes [201, 202].

GPE can help lower the dissolution and diffusion of intermediary polysulfides of lithium, elevating the cycle life and reducing the severity of the self-discharge of Li-S batteries. A really improved stability can be obtained regarding a capacity

Table 6. Novel electrolytes and advantages.

Novel electrolytes	Advantages	Ref.
Ceramic and polymeric electrolytes	Design simplicity and safe operation	[193]
Organic carbonate-type electrolytes with $LiPF_6$	Advanced systems, film-forming high-voltage additives and new solvents	[194]
Salt-in polymeric electrolytes	Good stability and high lithium transference number	[195]
New altered silyl-terminated polymer electrolyte	Very high lithium ion transference number, elevated ionic conductivity	[196, 197]
Liquid electrolytes	Increase ionic conductivity, lead to better capacity retention, suppress polysulfides dissolution	[198]
Gel polymer electrolytes	Reduce leaking of liquid electrolytes, hinder enlargement of dendrites	[199]
Gel-ceramic multilayer electrolyte	Higher electrochemical performance, unconfined shuttle effect	[200]

retention of about 72 % at a current density of 835 mA g^{-1} after 100 cycles [202].

8.3 Design of Permselective Membranes

Membranes in batteries generally play the role of preventing short circuits and contact between electrodes, causing ion flow, and displaying stability of different forms. The separators in Li-S batteries enable ion transfer and simultaneously pose as a barricade between the electrodes; yet, these characteristics are not sufficient to control polysulfide shuttling. To amend this inability, permselective membranes are introduced in the batteries; they can be defined as separators that enable selective ion transfer while satisfying other functions of its kind: the movement of electrolytes with the desired ions is enabled while the transport of undesirable ions is restricted. A permselective membrane is said to have 100 % efficiency if the transport of undesirable ions is entirely constricted [203–206].

Various advancements have been made in the design of membranes relating to the negative charge of multiple functional groups, as a pathway to tackle polysulfide shuttling. In this, Nafion and graphene oxide have presented themselves as go-to options for the mentioned type of membrane. Among the pair, Nafion, a cation-selective membrane, has gained greater attention due to its SO_3^- functional group, appreciable Li^+ conductivity, and great stability [155]. Fabrication of the Nafion cation-selective membrane is carried out by performing lithium-ion transfer on a film of Nafion 212 ionomer in a solution containing 1.0 M LiOH in a mixture of water and ethanol (1:1 wt/wt) [207]. A Li-S battery employing a Nafion membrane was observed to have 1185 mAh g^{-1} initial discharge capacity and a coulombic efficiency above 97 %. A comparison between the Nafion membrane and the Celgard 2400 separator showed that the latter displayed a retention of 69 % across 50 cycles and the former displayed a retention of only 46 % across 50 cycles, although both separators began with the same initial capacity. The Nafion ionomer has $-OCF_2CF(CF_3)OCF_2$-CF_2SO_3Li side chains, and when the Li ions dissociate from the chains, a negative charge is obtained by the SO_3 groups [208–212]. This allows the movement of Li^+ and entirely restricts polysulfides, which are negatively charged. The clustered structure of the film ensures the prevention of the transport of polysulfides. Permselective membranes thus solve an impeding issue: polysulfide shuttling, which has been the greatest hindrance to the commercialization of Li-S batteries.

9 Outlook and Scope

Since the introduction of Li-S batteries in the 1960s, there have been high expectations for the propitious elevated-energy density battery system. The last few decades have seen increased progress in the working of the Li-S batteries. More focus has been on bettering the Li-S batteries for a more practical approach in regard to preparing a new lineage of the sulfur cathode, establishing a kinetic promoter, bringing about a particular solvation of ions in the electrolyte, and a means to safeguard the lithium metal anode [213].

As we have almost exhausted all types of applications of Li-ion batteries, there is an emphasis on research regarding Li-S batteries that might depend on reversible redox reactions within sulfur and lithium. Carbon-based materials, organic materials, metal oxides, MOF, metal hydroxides, metal sulfides, metal nitrides, metal carbides, metal phosphides, and metal borides are being used as host materials for Li-S batteries [214].

Particular solutions of electrolytes have been used to overcome reactions across the surface of the lithium metal anode, by the formation of good passivation films on the surface. To increase the scope of Li-S batteries, issues affecting the anode material such as polysulfide shuttling must be ameliorated or completely eradicated, and efforts must be undertaken to come up with high-performance sulfur cathodes. Usage of liquid ionic solutions, electrolyte additives with mediators, substitute anodes, and silicon-lithium-sulfur are considered [215]. It is good to see that recent research work is more focused on the mitigation of polysulfides and parasitic reactions by using novel single-atom catalysts, and ether-based electrolytes, and on redox co-mediators for EPSE, an organic-based solid electrolyte interphase, and an effective Nafion protective layer for stabilizing lithium metal anodes [96–100]. These research studies have achieved polysulfide conversion and showed a good number of cycles while charging and discharging, but there is still a need to link the gap between research on Li-S batteries and realistic applications of the batteries [216].

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Abbrevations

BET	Brunauer-Emmett-Teller
BN	boron nitride
CGPE	composite gel polymer electrolyte
CMC	carboxymethyl cellulose
DME	1,2-dimethoxyethane
DMF	dimethylformamide
DMSO	dimethyl sulfoxide
DOL	1,3-dioxolane
EPR	electron paramagnetic resonance
EPSE	encapsulating polysulfide electrolyte
GPE	gel polymer electrolyte
HPLC	high-performance liquid chromatography
LiPS	lithium polysulfide
LISICON	lithium superionic conductor
MOF	metal-organic framework
PANI	polyaniline
PC	propylene carbonate

PEO	polyethylene oxide
PMIA	poly(*m*-phenylene isophthalamide)
PMMA	poly(methyl methacrylate)
PVDF-CO-HFP	poly(vinylidene fluoride-co-hexafluoropropylene)
rGO	reduced graphene oxide
SEI	solid electrolyte interface

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Review Article: After an overview of the history behind Li-S batteries, this review summarizes the recent research findings on their crucial parameters and bottlenecks. Future challenges and possible solutions are evaluated and further steps towards new-generation Li-S batteries with advanced components are outlined.

A Detailed Discourse on the Epistemology of Lithium-Sulfur Batteries

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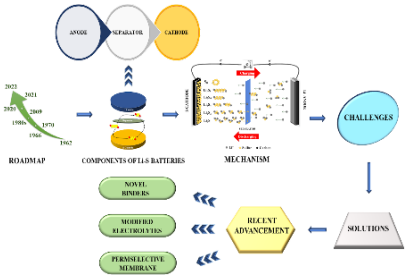

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