Recent Advancements in Selenium-Based Cathode Materials for Lithium Batteries: A Mini-Review

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Abstract: Selenium (Se)-based cathode materials have garnered considerable interest for lithium-ion batteries due to their numerous advantages, including low cost, high volumetric capacity (3268 mAh cm\(^{-3}\)), high density (4.82 g cm\(^{-3}\)), ability to be cycled to high voltage (4.2 V) without failure, and environmental friendliness. However, they have low electrical conductivity, low coulombic efficiency, and polyselenide solubility in electrolytes (shuttle effect). These factors have an adverse effect on the electrochemical performance of Li-Se batteries, rendering them unsuitable for real-world use. In this study, we briefly examined numerous approaches to overcoming these obstacles, including selecting an adequate electrolyte, the composition of Se with carbonaceous materials, and the usage of metal selenide base electrodes. Furthermore, we examined the effect of introducing interlayers between the cathode and the separator. Finally, the remaining hurdles and potential study prospects in this expanding field are proposed to inspire further insightful work.

Keywords: Li-Se batteries; cathode; interlayers; electrochemical performance; shuttle effect; Se/C composites; separators

1. Introduction

The energy production from non-fossil fuel sources such as solar and wind has garnered considerable interest because the energy generated from fossil fuels contributes to global contamination and climate catastrophe. Because of their enormous storage capacity and excellent conversion efficiency, rechargeable batteries are the best energy storage technologies for storing energy from renewable sources [1,2]. The market is dominated by lithium-ion batteries (LIBs) due to their high energy density and outstanding cycling performance [3–7]. Traditional LIBs, on the other hand, are unable to meet energy requirements in terms of energy density as the need for battery capacity for grid energy storage and electric vehicles escalates. Various solutions are suggested to enhance the LIB’s performance, including using a high-performance cathode and substituting a graphite anode with a high-capacity lithium anode. However, because of their limited capacity, high cost, and low energy density, researchers have made a substantial effort to develop low-cost, high-energy-density cathode materials to replace existing cathode materials (LiCoO\(_2\), LiNi\(_{1-x-y}\)Co\(_x\)Al\(_y\)O\(_2\), and LiNi\(_{1-x-y}\)Mn\(_x\)Co\(_y\)O\(_2\)) [8].

Because of its numerous qualities, such as earth abundance, low cost, high-energy density [9–13], and non-toxic nature, lithium-sulfur (Li-S) has received a lot of interest as a future generation metallic battery. However, due to the insulating nature of sulfur (S) and the soluble nature of polysulfides during cycling, the shuttle effect occurs, resulting in poor cycling performance and low coulombic efficiency [14–18]. Preparing a cathode with a high energy density, strong cycling stability, and high electrical conductivity is a significant challenge. Selenium (Se), a group XVI element, is a promising candidate with the same
chemical properties as S but a higher electrical conductivity. Although Se’s theoretical capacity (675 mAh g\(^{-1}\)) is not as great as S’s (1675 mAh g\(^{-1}\)), it is still more than double that of the transition metal compounds (e.g., LiCoO\(_2\) (275 mAh g\(^{-1}\))). Table 1 compares the electrochemical properties of Se, S, and LiCoO\(_2\) cathodes. Se has a 20-fold higher electrical conductivity than S and is comparable to LiCoO\(_2\) [19–23]. Additionally, Se-based cathodes performed well in carbonate-based electrolytes, which are less expensive and have higher voltage stability than ether-based electrolytes [24]. Furthermore, Se has a more excellent melting point than S, resulting in a safer battery at elevated temperatures. Moreover, the polyselenides shuttle effect is weaker than that of polysulfides [25]. With these superiorities, Se-based cathodes are potential alternatives to S and LiCoO\(_2\). Table 1 compares the electrochemical properties of carbide-derived carbon-Se (Se-CDC) and carbide-derived carbon-S (S-CDC). The loading of the Se and S is the same (60%), which shows the good electrochemical performance of Se-CDC, suggesting Se as a promising cathode material [26].

Table 1. Electrochemical properties of Se, S, and LiCoO\(_2\).

| Properties                           | Sulfur (S) | Selenium (Se) | LiCoO\(_2\) |
|--------------------------------------|------------|---------------|-------------|
| Theoretical gravimetric capacity (mAh g\(^{-1}\)) | 1675       | 675           | 274         |
| Electrical conductivity (S cm\(^{-1}\))      | \(5 \times 10^{-30}\) (25 \(^\circ\)C) | \(1 \times 10^{-5}\) (25 \(^\circ\)C) | \(1 \times 10^{-4}\) |
| Theoretical energy density (WhL\(^{-1}\))    | 2800       | 2528          | 1190        |
| Redox potential (V vs. Li\(^+\)/Li)         | 2.2        | 2             | 4.2         |
| Theoretical specific energy (Whkg\(^{-1}\))  | 2267       | 1155          | 420         |

Figure 1. Comparison of the electrochemical performance (a) cyclic stability and (b) charge–discharge profile of Se-CDC and S-CDC in the same electrolyte (5 M LiTFSI). Reproduced with permission from [26], Copyright 2015, Wiley Online Library. (c) Cycle performance at a current density of 0.2 C between 1.0 and 3.0 V and (d) rate capability at various current densities between 1.0 and 3.0 V. Reproduced with permission from [27], Copyright 2015, Elsevier BV.
Despite their numerous advantages, Li-Se batteries have additional obstacles that have hindered their commercialization. (1) The volume changes during the intercalation/deintercalation process, which pulverizes the Se-based electrode, resulting in a rapid capacity decline. (2) Typically, the Se is loaded to less than 50%, resulting in a low energy density. (3) As a result of creating the intermediary polyselenides, a shuttle effect occurs, resulting in a loss of active material and a reduction in battery life. Moreover, (4) they have inadequate conductivity, resulting in low electron and lithium-ion kinetics and thus poor electrochemical performance. Figure 2a shows the number of articles published in the last six years for lithium-selenium batteries, according to the Web of Sciences, while Figure 2b–d shows the problems Se-based cathodes faces. Many researchers have recently made significant efforts to solve the obstacles mentioned earlier for Li-Se batteries. Compositing the Se with carbonaceous materials is an efficient strategy to enhance the electrochemical performance of the Se-based cathode because carbon provides enough space for volume expansion during the charging/discharging process. For example, Zhang’s group studied the electrochemical properties of selenium nanowires that delivered a nearly zero capacity after 200 cycles. Se nanowires well-sealed with PANI layers of a thickness of ~20 nm, forming a core–shell structure, showed an improved capacity of ~500 mAh g⁻¹ after 200 cycles. Further treating this core–shell structure with graphene, the obtained composites are known as G@Se/PANI, showed an improved capacity of ~600 mAh g⁻¹ after 200 cycles. The electrochemical performance of Se nanowires, Se/PANI, and G@Se/PANI is shown in Figure 1c,d.

Figure 2. (a) According to the Web of Sciences database, articles on Li-Se batteries published in the last six years. The critical problems of Li-Se batteries: (b) the weak conductivity of solid Se or Li₂Se with low utilization, (c) the shuttle effect of polyselenides, and (d) significant electrode pulverization during charge and discharge processes. Reproduced with permission from [28]. Copyright 2021, Wiley Online Library.
In this study, firstly, the choice of the electrolytes for the Se base cathode is discussed. Then, recent advances in Li-Se batteries are reviewed in synthesis techniques and electrochemical performance. Specifically, compositing a Se cathode with carbonaceous materials, using a metal selenide base electrode and introducing interlayers between the cathode and separator are discussed. Finally, the challenges and prospects are presented, aiming to provide a possible pathway to create high-performance Li-Se batteries for the real world.

2. Choice of the Proper Electrolyte for Li-Se Batteries

Maintaining the cycle life of a rechargeable battery requires the proper functioning of the active material. A battery usually has four parts: a cathode, an anode, a separator, and an electrolyte. An electrolyte, in addition to the other three components, is critical to a battery’s electrochemical performance. Electrolytes are currently divided into two categories: carbonate and ether-based electrolytes. Carbonate-based electrolytes appear to be preferable to ether-based electrolytes, particularly for Li-Se batteries, because carbonate-based electrolytes form Li$_2$Se in a single step without the creation of unstable intermediates that occur in ether-based electrolytes [29–34]. The loss of active material caused by the development of intermediate polyselenides results in poor cycling performance of Li-Se batteries. Amine et al. [25,35] used a high-energy ball milling technology to create an Se-based cathode (Se/MWCNs). They tested it for Li-Se batteries with carbonate-based electrolytes (1.2 M LiPF$_6$, dissolved in EC and EMC with a 3:7 ratio), which demonstrated a reversible capacity of 500 mAh g$^{-1}$ at 100 mA g$^{-1}$. They believe the excellent results are due to the single-phase conversion of Se to Li$_2$Se without creating intermediate polyselenides that dissolve in electrolytes and cause capacity fading. Lei et al. [33] synthesized microporous carbon spheres (MPCS) from glucose, utilizing a straightforward and effective hydrothermal synthesis approach followed by MPCS activation with KOH. Through a melt-diffusion process, selenium particles were homodispersed in the micropores of MPCS (Figure 3a,b). MPCS/Se composites prepared for use as the cathode of Li-Se batteries exhibit a nearly 45% Se loading, high capacities, and stable cycling performance in both the carbonate-based LiPF$_6$ electrolyte (565 mAh g$^{-1}$ at 1 C after 500 cycles) and the ether-based LiTFSI electrolyte (480 mAh g$^{-1}$ at 1 C after 500 cycles), as shown in Figure 3c,d. Notably, an exceptional rate capability was seen in the carbonate-based electrolyte, as well as coulombic efficiency of nearly 100% in both electrolytes. The microporous architecture of the high conductivity MPCS material adds to the material’s good electrochemical characteristics and rate capability, successfully mitigating volume change and inhibiting polyselenide dissolution. Caken et al. [29] investigated the in situ synthesis of intermediate polyselenides using four-electrode cycle voltammetry (CV) and three-electrode electrochemical impedance spectroscopy (EIS) to identify the presence of polyselenides in carbonate and ether-based electrolytes. In ether-based solvents, CV measurements demonstrate the presence of dissolved polyselenide intermediate forms, whereas no signals are detected in carbonate-based electrolytes. Figure 3e illustrates the discharge–charge curves of Li-Se batteries for EC-DMC and TEGDME. Huang et al. [36] used the carbonization of KOH and polypyrrole to create a Se-CP composite. They evaluated the materials for Li-Se batteries with carbonate and ether-based electrolytes, with a CV of 0.1 mV s$^{-1}$ within voltage windows of 1 and 3 V. There is only one reduction peak and one oxidation peak when using a carbonate-based electrolyte. However, there are several oxidation and reduction peaks when using an ether-based electrolyte. The presence of several peaks for the ether base electrolyte indicates that the Se to Li$_2$Se conversion occurs in multiple phases.
Nonetheless, Yi’s group [37] showed that direct conversion of Se to Li$_2$Se can occur in an ether-based electrolyte, but battery performance is still inferior to that of a carbonate-based electrolyte. Numerous plateaus in the CV curve may arise due to the structure of Se, with amorphous selenium showing multiple steps and trigonal selenium showing simple step conversion, according to Zhou’s group [38]. His subsequent research reveals that amorphous Se outperforms crystalline Se. Despite its superior performance, Se exhibits significant reactivity when used with a carbonate-based electrolyte. Se can be protected against severe side reactions with electrolytes by introducing host materials, resulting in a longer cycle life. In addition, the creation of stable SEI on a Se-based cathode can aid ion transport while preventing side reactions, ensuring excellent stability in carbonate-based electrolytes. Carbonate-based electrolytes are superior to ether-based electrolytes because they are less expensive and less dangerous. To address the electrolyte-dependent difficulties associated with Se cathodes, various hosts and diverse approaches have been used to enhance overall performance. On the other hand, solutions involving the separator, electrolyte, and cell layout have considerable promise. The following sections will cover the respective advancements in detail.

3. Carbon-Selenium Composites for Li-Se Batteries

Selenium can generate polyselenides that dissolve in the electrolyte, which results in low coulombic efficiency and capacity fading. To overcome this issue, confining Se in a carbon matrix is a good choice that prevents polyselenide dissolution and increases the Se
cathode’s electronic conductivity and structural stability, which results in the improved electrochemical performance of Li-Se batteries.

3.1. Porous Carbon/Se Composites

A porous structure with a high surface area and a large pore size could improve Se’s electrical conductivity and reduce the shuttle effect. Based upon the size of the pores, porous carbons are divided into different structures such as mesoporous carbon, microporous carbon, and hierarchical porous carbon.

3.1.1. Mesoporous Carbon/Se Composite

Mesoporous carbon with pore sizes ranging from 2 to 50 nm, a high surface area, and a unique structure design has several advantages, including a large surface area in contact with Se and a short diffusion length for the lithium ion. Liu et al. [39] used the spray drying hard template approach to produce MCMs/Se composites. The pore size of MCMs was regulated to 3.8, 5, 6.6, and 9.5 nm using an in situ polymerized colloidal silica template with varying pore sizes. The MCMs-3.8/Se with a 50% Se loading, with a surface area of 1554 m² g⁻¹, demonstrated an initial capacity of 513 mAh g⁻¹ and capacity retention of 300 mAh g⁻¹ over 100 cycles at 0.5 C, with a coulombic efficiency of 100%. Even at a higher rate of 2.5 C, the specific capacity is 210 mAh g⁻¹. MCMs-3.8/Se’s superior performance is likely due to its tiny, porous structure and interconnected mesoporous structure, which offers substantial Se confinement and mitigates the shuttle effect. At a scan rate of 0.2 mV s⁻¹, the CV curve exhibits two cathodic peaks at 1.9 and 2.1 V, corresponding to selenium conversion to soluble polyselenides and soluble polyselenides conversion to Li₂Se₂/Li₂Se, respectively. At 2.2 V, there is a single anodic peak, showing the process of overlap delithiation [35]. All consecutive scans overlap, indicating that the MCM-3.8/Se cathodes are stable during the repeated lithiation/delithiation process.

Zhang et al. [40] synthesized selenium-infused ordered mesoporous carbon composites via melt-diffusion from a ball-milled mixture of Se and CMK-3 (Se-CMK-3), which exhibits a high reversible capacity of 488.7 mAh g⁻¹ at 0.05 C after 100 cycles and a discharge capacity of 268.7 mAh g⁻¹ after 200 cycles, as illustrated in Figure 4a,b. The superior electrochemical performance could be ascribed to the CMK-3 matrix’s increased electronic/ionic conductivities and structural support. Xue et al. [41] used a simple annealing process to create a hollow carbon structure with linked mesopores to contain a Se composite. The Se/HMCS electrode performs admirably (as shown in Figure 4c,d). The superior electrochemical performance is attributed to HMCS’s distinctive hollow structure and a high concentration of selenium encapsulated within mesoporous material, which promotes electronic/ionic transit and provides additional buffer space to adjust the volumetric expansion of selenium and polyselenide during prolonged cycling. Wang’s group [42] produced Se/C composites by dissolving Se₁₂ in Se₈ and infusing it into mesoporous carbon. The resulting composite comprises 30% selenium and 70% mesoporous carbon. The composite has a large surface area of 467 m² g⁻¹ and pore size of 4.1 nm, enabling it to perform well electrochemically when tested with Li-Se batteries. Over 1000 cycles, the composite demonstrated a reversible capacity of 480 mAh g⁻¹ and high-capacity retention. Yang and coworkers [43] produced Se/CMK-3 composites through melt diffusion using CMK-3 and Se. After 50 cycles, the composite with a surface area of 17.2 m² g⁻¹ and a pore volume of 0.131 cm³ g⁻¹ demonstrated good electrochemical performance, with a specific capacity of 300 mAh g⁻¹. Lai’s group [44] synthesized a meso-C@Se composite from a specific metal-organic framework (MOF) with a reversible capacity of 280 mAh g⁻¹ at 2 C after 100 cycles. Xu et al. [45] generated a C@Se composite with a mesoporous carbon matrix formed from a unique metal–organic framework structure. After 100 cycles, the composite had a capacity of 417 mAh g⁻¹.
Figure 4. (a) Cyclic performances of Se-CMK-3 and Se/CMK-3 at 0.05 C and (b) high-rate cyclic performances at 0.5 C. Reproduced with permission from [40], Copyright 2020, American Chemical Society. (c,d) Rate performance of Se@HMCS at different rates and cycling performance at 0.5 A g\(^{-1}\). Reproduced with permission from [41], Copyright 2020, Elsevier BV.

3.1.2. Microporous Carbon/Se Composite

Microporous carbon materials with pore diameters of less than 2 nm and a high specific surface area are considered ideal hosts for Se impregnation. The microporous structure’s high conductivity results in exceptional electrochemical properties and rate capability, effectively decreasing volume expansion and managing polyselenide dissolution. Lei’s group [33] developed MPCS with a large surface area of 782 m\(^2\) g\(^{-1}\) and pore size of 0.552 nm by utilizing a simple hydrothermal procedure and the melt diffusion approach. A Se/MPCS composite with a surface area of 9.32 m\(^2\) g\(^{-1}\) and a Se loading of 45% was prepared. When used as a cathode for Li-Se batteries, Se/MPCS demonstrated excellent electrochemical performance with both carbonate-and ether-based electrolytes. The composite demonstrated a cycling stability of 565 and 480 mAh g\(^{-1}\) after 500 cycles with a coulombic efficiency of 100% for carbonate and ether-based electrolytes. Shiraz et al. [46] described the effects of a lithium-ion selenium battery with a lithiated graphite/silicon anode and a microporous carbon/se (MPC/Se) cathode in a carbonate electrolyte comprising fluoroethylene carbonate (FEC). Due to the decreased dissolution of polyselenides into the electrolyte, the FEC additive facilitated the formation of a stable MPC/Se cathode. It enhanced the mechanical stabilization of the graphite/silicon anode, leading to a stable solid electrolyte interphase (SEI) layer on both the cathode and anode surfaces. As a result, lithium-ion selenium batteries demonstrated exceptional specific capacity and stability. Figure 5 illustrates the SEM images and electrochemical performance. Liu et al. [47] synthesized microporous carbon with a vast surface area of 890 m\(^2\) g\(^{-1}\) and a pore diameter of 1.1 nm using polyhedral ZIF-8 as a precursor [48]. Se was injected into it using the melt diffusion process, resulting in a Se-C composite with a Se loading of 51 wt%. When employed as a cathode for Li-Se batteries, the obtained composite demonstrated an excellent cycling performance of 240 mAh g\(^{-1}\) after 3000 cycles. Bao’s group [49] synthesized Se/MCNF by transforming Se particles into Se\(_n\) and chemically bonding them to the inner surface of MCNFs. When employed as the cathode for Li-Se batteries, the composite demonstrated an outstanding cycling performance of 420 mAh g\(^{-1}\) over 3000 cycles at 5 C. Huang et al. [37] synthesized MiPCSs with an 853 m\(^2\) g\(^{-1}\) surface area by hydrothermally treating Pluronic F127 and phenolic resol and then carbonizing them. After loading with Se, the resulting
composite Se/MiPCSs demonstrated a cycle stability of 700 mAh g\(^{-1}\) after 1200 cycles. Zhou’s group [50] generated a Se/MC composite by embedding Se in microporous carbon obtained from MOFs with 51% Se content. When utilized as the cathode for Li-Se batteries, the composite demonstrated a capacity of 511 mAh g\(^{-1}\) at 5 C after 1000 cycles in the electrolyte LiDFOB/EC-DMC-FEC. The excellent performance results from the electrolyte’s compatibility and the confinement of Se in ultra-small microporous carbon.

Figure 5. SEM image of (a) MPC and (b) MPC/Se. (c) Cycling performance and (d) coulombic efficiency of MPC/Se cathode in a half-cell configuration with 1.3 M LiPF\(_6\), EC:DEC electrolyte with 0–15% FEC additive. The testing was performed at 0.1 C in the first 10 cycles and 0.5 C afterward (1 C = 678 mA g\(^{-1}\)). Reproduced with permission from [46], Copyright 2020, IOP Science.

3.1.3. Hierarchical Porous Carbon/Se Composites

Hierarchical porous carbon contains pores of different sizes, including micropores that help stabilize polyselenides and accommodate active materials, macropores that act as rapid channels for electrolyte, thus further improving electrochemical performance, and mesopores that assist in the transfer of ions and electrons and improve active material utilization. Song et al. [51] synthesized a nitrogen-doped core-shell hierarchical porous carbon (NCCSHPC) from metal–organic frameworks (MOFs) with linked meso/micropores to successfully encapsulate Se for a high-performance Li-Se battery. Micropores form at the intersection between the ZIF-8 core and ZIF-67 shell, whereas mesopores form at the core–shell interface following pyrolysis of the core–shell ZIF 8@ZIF 67 precursor. This unique hierarchical porous structure advantageously restricts selenium and polyselenides, preventing them from dissolving in the pores and minimizing volume change. The in situ nitrogen doping of NCSHPC enhances not only the electrical conductivity of Se but also the chemical adsorption of Li\(_2\)Se, as shown by density functional theory calculations. The Se/N CSHPCII (molar ratio of Co source to Zn source is 1.0 in the core–shell ZIF8@ZIF67 precursor) demonstrates a high reversible capacity because of the dual physical confinement and strong chemisorption. Zhao’s group [52] used a carbonization and KOH-activation procedure to create hierarchically microporous activated carbon (AC) from leftover coffee grounds. Despite its simplicity of synthesis, the optimized AC (AC-700) exhibited a huge surface area of 1355 m\(^2\) g\(^{-1}\) and a substantial micropore volume of 0.52 cm\(^3\) g\(^{-1}\). After 100 cycles at 0.1 C, the Se/AC-700 cathode demonstrated a reversible capacity of 655 mAh g\(^{-1}\) in Li-Se batteries with a carbonate electrolyte. Additionally, the Se/AC-700 cathode displayed su-
uperior cycling performance over 400 cycles without any capacity degradation. The superior battery performance was due mostly to rapid electron transfer and Li-ion diffusion in Se contained within the AC-700’s microporous carbon (Figure 6a–e). MHPC was synthesized by Liu et al. [53] through a hydrothermal and carbonization process. Melt diffusion was used to create a composite of Se and MHPC. The huge surface area and large pore size of MHPC (1695 m$^2$ g$^{-1}$, 0.21 cm$^3$ g$^{-1}$) enable the storage of a large amount of Se (48.68%). When used as the cathode for Li-Se material, it demonstrated a capacity of 588.2 mAh g$^{-1}$ at 0.5 C and 99.9% coulombic efficiency. The hollow and mesopore structures of the composite contribute to its excellent electrochemical performance. Jiang et al. [54] used a melt diffusion technique to produce hierarchical porous carbon aerogels (HPCA). To obtain the Se/HPCA composite, Se was distributed into the pores. When used as a cathode for Li-Se batteries, the composite demonstrated excellent electrochemical performance, retaining a coulombic efficiency of 50% after 50 cycles in an ionic-based (PYR14TFSI) electrolyte and capacity retention of 309 mAh g$^{-1}$ after 100 cycles at 0.5 C in a LiNO$_3$ modified electrolyte. Zhang’s group [55] constructed a Se-NCHPC composite with 56.2% Se content using a simple template-assisted and chemical activation procedure. When employed as the cathode for Li-Se batteries, the composite demonstrated a reversible discharge capacity of 305 mAh g$^{-1}$ at 2 C after 60 cycles. The existence of micro/meso/macropores is ascribed to their high electrochemical performance. Yushin et al. [26] synthesized a 60% Se-infiltrated ordered meso-microporous silicon carbide-derived carbon (OM-SiC-CDC). When used as a cathode for Li-Se batteries, the composite demonstrated a reversible capacity of 500 mAh g$^{-1}$ after 150 cycles at 0.2 C in a 5 M LiTFSI electrolyte. Shen’s group [36] created an MMPCb using a KOH mixture and carbonization of pomelo pericarp and loaded it with Se to produce an MMPb/Se composite with a high reversible capacity of 467 mAh g$^{-1}$ at 0.2 C after 300 cycles when used as a cathode for Li-Se batteries. The excellent performance is a result of the MMPb’s bimodal pores and hierarchically porous structure.

Figure 6. SEM images of waste coffee grounds (WCG) (a) as received, (b) after carbonization, and (c) after activation (AC-700). (d) Cycling stability, (e) long cycling performance of Se/AC-700 measured at 0.5 C. Reproduced with permission from [52], Copyright 2019, Elsevier BV.
3.2. CNTs-CNFs/Se Composites

Due to the excellent electrical conductivity and high strength of carbon nanotubes and carbon nanofibers, they are well suited for use as Se hosts. Wang et al. [57] used ethylenediamine as the solvent to produce a Se/MWCNT-S composite. Se nanoparticles (100 nm) are placed uniformly on MWCNTs. It showed a reversible capacity of 468.7 mAh g\(^{-1}\) at 4 C when employed as the cathode for Li-Se batteries. The superior performance is related to the reduced size of Se particles and their dispersion within a carbon matrix. Balakumar and Kalaiselvi [58] synthesized MWCNTs encased in porous carbon using a hard template synthesis method (TTC). The resulting structure has a large surface area of 1131 m\(^2\) g\(^{-1}\), enabling 70% Se loading. When employed as the cathode for Li-Se batteries, Se/TTC provided an initial capacity of 625 mAh g\(^{-1}\) at a C/5 rate and 400 mAh g\(^{-1}\) after 800 cycles. The excellent performance results from the synergistic impact of porous carbon and MWCNT. Zhang et al. [59] prepared PCNFWs by carbonizing PPy and activating them with KOH. PCNFWs have a large surface area of 719.2 m\(^2\) g\(^{-1}\) and a pore volume of 0.4 m\(^3\) g\(^{-1}\) and can encapsulate Se to generate Se/PCNFWs. The composite demonstrated good electrochemical performance when used as a cathode for Li-Se batteries. At 0.2 C, it delivered an initial discharge capacity of 563.9 mAh g\(^{-1}\) and a reversible discharge capacity of 414.5 mAh g\(^{-1}\) that was retained after 100 cycles. The excellent performance results from the advantageous characteristics of the linked porous nanostructure, the large channel structure, and the high specific surface area.

3.3. Se/C Core-Shell Composite and Free-Standing Carbon/Se Composite

The core-shell construction is an effective method of enhancing the electrochemical performance of Li-Se batteries. Because they have sufficient space, they can buffer volume variations while charging and discharging, reduce polyselenide dissolving into the electrolyte, and maintain high utilization of active material during the lithiation/delithiation processes. Utilizing the core–shell structure, Guo’s group [60] synthesized a Se/C core-shell structure using a simple hydrothermal procedure, which demonstrated good electrochemical performance when used as a cathode material for Li-Se batteries yielding 350 mAh g\(^{-1}\) after 100 cycles and high coulombic efficiency of 95%. The high-performance results from the flexible and homogenous core–shell structure, which remains intact even after 100 cycles and helps limit polyselenide dissolution and capacity fading. Zhang et al. [61] synthesized a Se/C core-shell structure by a hydrothermal technique and demonstrated that when used as a cathode for Li-Se batteries, it had a discharge capacity of 588 mAh g\(^{-1}\) after 80 cycles at 0.5 C. The outstanding performance results from the high electronic conductivity and the peculiar structure of the Se/C composite, which effectively confines ployselnides and Se (Figure 7a,b). Qian et al. [62] manufactured a porous hollow carbon bubbles/Se composite through melt diffusion; when used as the cathode for Li-Se batteries, this composite demonstrated a reversible capacity of 606 mAh g\(^{-1}\) after 120 cycles. Peng et al. [63] synthesized Se @RGO by a self-assembly technique. The resulting material comprises 80% Se. It had a capacity of 533 mAh g\(^{-1}\) at 0.2 C. The high performance of the composite indicates that the Se@RGO composite is an exciting development for Li-Se batteries.

Conductive carbon and binders have been commonly added to many previously reported Se-based electrodes to improve their mechanical integrity and electrical conductivity while decreasing the relative proportion of Se, resulting in a lower energy density. As a result, a flexible, self-supporting, and binder-free Se/C composite electrode is required, in which all electrode components contribute to lithium storage. Additionally, the technique for packing cells can be significantly simplified by employing this structure. Ding et al. [64] developed monolithic and self-supporting selenium (Se) lithium metal cathodes with a record Se loading of 70% wt. The carbon host is produced from nanocellulose, a great and environmentally friendly forestry product. Due to the composite’s density (2.37 g cm\(^{-3}\)), it has a theoretical volumetric capacity of 1120 mAh cm\(^{-3}\). This design is qualitatively distinct from earlier Se–carbon nanopowders or micro powders, providing up to a twofold increase in energy density. The cathode provides a reversible capacity
of 1028 mAh cm$^{-3}$ (620 mAh g$^{-1}$) and 82 percent retention over 300 cycles for Li storage (Figure 7c). Han et al. [65] synthesized a unique free-standing Se/MCN-RGO paper composite with a unique three-dimensional hierarchical structure that may be used as a cathode for Li-Se batteries. With an initial capacity of 655 and 568 mAh g$^{-1}$ after 100 cycles, the composite displayed excellent capacity utilization, increased rate capability, and ultra-long cycling stability.

![Figure 7](image-url)

**Figure 7.** (a) Cyclability and coulombic efficiency of the Se/carbon-rich core–shell composites and the pristine selenium at the current density of 337.5 mA g$^{-1}$, (b) Impedance plots and equivalent circuit for the Se/carbon-rich core–shell composites and the pristine selenium before cycling. Reproduced with permission from [61], Copyright 2015, Elsevier BV. (c) Cyclability of Se–NCMC at 135 mA g$^{-1}$ (0.2 C). Reproduced with permission from [64], Copyright 2018, Wiley Online Library.

Additionally, it has an incredibly long cycle life, with a capacity of around 400 mAh g$^{-1}$ at 1 C and a 1300 cycle life. Due to its high capacity and long cycle life, these findings imply that the Se/MCN-RGO composite may be a suitable cathode material for rechargeable Li-Se batteries in practical applications. Huang et al. [66] developed a binder-free Se@Ni electrode that may be used directly as a cathode for Li-Se batteries without binders or conductive additives. The resultant composite delivered 554 mAh g$^{-1}$ after 100 cycles and maintained a capacity of 137 mAh g$^{-1}$ at a rate of 0.1 C. Coating the Se surface with graphene oxide significantly enhances the electrode’s performance, achieving 665 mAh g$^{-1}$ in the first cycle and remaining at 266 mAh g$^{-1}$ after 100 cycles at 0.1 C. This work substantially contributes to the development of high-capacity cathode materials for Li-Se batteries. Han et al. [67] developed a self-standing graphene-Se@CNT composite sheet as a binder-free cathode for Li-Se batteries using a two-step construction approach. After 100 cycles, the composite preserved 315 mAh g$^{-1}$ of capacity and exhibited a coulombic efficiency of better than 96%. The high selenium utilization in composites is mainly owed to graphene and carbon nanotubes, which form a three-dimensional conductive network with open routes for electron transfer and lithium-ion diffusion and confinement of polyselenide. He and colleagues [68] created a 3DG-CNT@Se aerogel with CNT/Se sandwiched between graphene nanosheets using a solvothermal and freeze-drying process. At 1 C, the 3DG-CNT@Se cathode demonstrated a high reversible capacity of 558 mAh g$^{-1}$. The unique
three-dimensional mesoporous, conductive network provides exceptionally efficient charge transport channels, inhibits polyselenide rapid dissolution, and limits the volume expansion of Se throughout the charging/discharging process.

4. Metal Selenide-Based Electrodes

Metal selenides have been examined as a possible improved-capacity cathode material. MoSe₂, a transition metal dichalcogenide, is one of these materials with a high capacity for lithium-ion storage. Jiang et al. [69] synthesized MoSe₂ nanocrystals through a simple thermal deposition technique, revealing a 782 mAh g⁻¹ initial discharge capacity at 0.1 C and a voltage range of 0.1–3 V. Additional analysis indicated a phase transition between 2H-MoSe₂ and O-MoSe₂, followed by the creation of Mo clusters as more lithium atoms are bonded into MoSe₂, resulting in the formation of Mo and Li₂Se. Using KIT-6 as a template, Lee’s group [70] produced mesoporous MoSe₂ with a two-dimensional layered crystalline framework and a three-dimensional porous structure. The resulting structure has a large surface area of 96 m² g⁻¹ and mesopores with a diameter of 16 nm, exhibiting a high reversible capacity of 728 mAh g⁻¹ after 30 cycles. Due to its low electrical conductivity, MoSe₂ maintains its poor cycle stability. By integrating a conductor into MoSe₂, we can overcome the material’s limited electrical conductivity and increase cycling stability. Jin et al. [71] synthesized multifunctional selenium-doped carbon nanotubes/nickel selenide coaxial nanocables with chemical bonding interfaces via electroless deposition in conjunction with the succeeding chemical vapor reaction mechanism, in which the nickel layer is first accumulated onto the carbon nanotube core and is then transmogrified in situ into the nickel selenide sheath via a controlled selenization reaction at an elevated temperature. Due to the wide active material/electrolyte area of contact and the rapid diffusion and transfer of mass and charge during the electrochemical process, this coaxial structure exhibits excellent lithium storage performance. As an anode for lithium-ion batteries, the coaxial nanocables have a lithium storage capacity of 600.0 mAh g⁻¹ at 0.1 A g⁻¹.

The SEM images and electrochemical performance are shown in Figure 8a–c. Liu and colleagues [72] employed a simple hydrothermal and annealing technique to generate a MoSe₂/C composite that displayed a capacity of 577 mAh g⁻¹ after 50 cycles when used as a cathode for Li-Se batteries. Sánchez’s group used a spray deposition technique to create MoSe₂/SWCNT, resulting in a structure with a capacity of 798 mAh g⁻¹. Additionally, molybdenum diselenide’s cycle stability is improved by mixing it with metal oxides. Zhao’s group [73] synthesized MoSe₂@MoO₂, which exhibited a cycle stability of 520 mAh g⁻¹ after 500 cycles. Wu’s group [74] synthesized MoSe₂@C/TiO₂, which after 100 cycles, exhibited a capacity of 987 mAh g⁻¹. Bi₂Se₃ proved to be a good candidate for Li-Se battery cathode materials as a metal selenide. Xu’s group [75] developed Bi₂Se₃ nanosheets with a capacity of 715 mAh g⁻¹ using a green technique. Jin et al. [76] synthesized Bi₂Se₃−ₓSₓ through a solvothermal process, demonstrating a 920 mAh g⁻¹ initial capacity. Han’s group [77] developed indium-doped Bi₂Se₃ with a 998 mAh g⁻¹ discharge capacity. Additionally, it appears as though the ZnSe cathode is a viable alternative for Li-Se batteries. Zhang’s group [78] established a hydrothermal technique for synthesizing ZnSe using zinc (NO₃)₂, 6H₂O and sodium selenide. The ZnSe generated after 50 cycles has a reversible capacity of 433 mAh g⁻¹ at 100 mA g⁻¹. Combining ZnSe with carbonaceous components increases the material’s electrical conductivity, increasing its electrochemical performance. Park’s group [79] synthesized ZnSe/C composites by solid-state synthesis. This indicated a capacity of 705 mAh g⁻¹ after 300 cycles. Using a simple hydrothermal technique, Xu and colleagues [80] synthesized ZnSe@C, which displayed a high reversible capacity of 960 mAh g⁻¹ after 400 cycles. Other metal selenides, including WSe₂ [70], CuSe [81], and SnSe [82], have also shown excellent electrochemical properties for LIBs.
5. Interlayers and Separators

In 2012, Manthiram’s team introduced the notion of an interlayer for a Li-S cell [83]. The authors introduced a multi-walled carbon nanotube (MWCNT) interlayer between the separator and cathode, preventing lithium polysulfides from crossing across the anode. Later, the same group produced a functional separator based on a lightweight carbon covering, further enhancing the Li-S cell’s electrochemical performance [84]. Incorporating conductive interlayers between the electrode and separator is another technique for boosting specific capacity and cycle stability. Throughout discharge–charge processes, the conductive carbon interlayer drastically reduces the charge transfer resistance of cathodes and traps the migrating active material. Increased electrical conductivity permits the practical application of active material and consistent, high-rate battery performance with extended cycle life.

Additionally, the interlayer of porous carbon serves as an exceptional matrix for holding and tolerating dissolved intermediate polyselenides. By introducing interlayers or functionalizing the separators, it is possible to reduce the polyselenide shuttle effect and the polarization effect. Interlayers and separators are frequently incredibly light, contributing to the cell’s diminutive mass. Thus, high Se loading is possible, which is advantageous for Li-Se batteries with a high energy density. Yang et al. [85] used a calcination technique to produce a self-standing Se/PCC cathode containing 46.58% Se. Additionally, electrospun PAN interlayers are introduced between the separator and cathode. It has a reversible capacity of 590 mAh g⁻¹ after 100 cycles at 0.5 C when used as the cathode for Li-Se batteries (Figure 9a–d). Additionally, due to the insertion of PAN interlayers, no polyselenides are found throughout the charging/discharging process. Zhao’s group [86] synthesized Se-TiO₂ composites by melt diffusion and then added nickel foam as an interlayer between the...
separator and the cathode. The synthesized Se-TiO\(_2\)/NFF displayed a discharge capacity of 600 mAh g\(^{-1}\) after 200 cycles. The inclusion of a nickel foam file is credited with improving performance. Zhang et al. [87] studied the performance of Se/TiO\(_2\) composites with free-standing carbon interlayers. Prior to the inclusion of the interlayers, the composite had a reversible capacity of 100 mAh g\(^{-1}\) after 100 cycles. However, after adding the interlayers, it had a capacity of 370 mAh g\(^{-1}\) after 100 cycles at C/5. Carbon interlayers facilitate charge transfer and capture selenium that migrates during the lithiation/de-lithiation processes. Furthermore, carbon interlayers act as an efficient matrix for polyselenide intermediates. Table 2 gives the electrochemical properties of some separators used for Li-Se batteries.

![Figure 9](image-url)

**Figure 9.** (a) The preparation process for the Se/PCC cathode and PAN interlayer, and the brief model of the assembled Li-Se battery (b,c) CV curves at 0.1 mV s\(^{-1}\) and (d) cycling performances of the Se/PCC-2PAN and Se/PCC-0PAN systems at 0.5 C. Reproduced with permission from [85]. Copyright 2019, American Chemical Society.
Table 2. Some separators/interlayers designed to immobilize polyselenides and their performance properties.

| Separator                          | Film Mass Loading (mg cm⁻²) | Active Material Loading (mg cm⁻²) | Specific Capacity (mAh g⁻¹) | Ref. |
|-----------------------------------|----------------------------|---------------------------------|-----------------------------|-----|
| DMTA-COF + ceramic                | NA                         | 3                               | ~125 (100th cycles)         | [88]|
| PP/CTAB-CNTs-Ti₃C₂Tx              | 0.09/0.27                  | 2.1                             | ~480 (100th cycles)         | [89]|
| PP/Graphene                       | 0.3/5                      | 4                               | ~330 (500th cycles)         | [90]|
| PP + N,S-doped Graphene           | 0.65/32                    | 5                               | ~330 (1000th cycles)        | [91]|

Separators are polymeric membranes that serve as a barrier between the battery’s cathode and anode, preventing short circuits. Traditional separators are inadequate with Li-Se batteries because they cannot prevent the shuttling of the polyselenides. Separator remodeling is a highly effective method for resolving polyselenide dissolution issues. Fang et al. [90] developed a highly lightweight, permeable graphene-coated polymer separator with a vast surface area. This separator is exceptionally effective at inhibiting polyselenid shuttling and promoting redox kinetics. Yang et al. [92] synthesized TPB-DMTP-COF as a coating layer for Celgard separators. Due to the hydrogen bonding between TPB-DMTP-COF and LiTFSI in electrolytes, TPB-DMTP-COF may absorb LiTFSI and accelerate Li-ion transport, resulting in an 800-cycle life at a 4 mg cm⁻² area loading. Coating metal oxides on the separator is a great way to improve the performance of Li-Se batteries. Mukkabala et al. [93] used tungsten oxide as a separator layer to build a Li-Se alkali-activated carbon hybrid cell. The Se loading is 70%, resulting in a capacity of 525 mAh g⁻¹ prior to the oxide layers and a reversible capacity of 808 mAh g⁻¹ with the addition of the tungsten oxide layer, which blocks the polyselenides and increases performance. After 100 cycles, 2D MXenes nanosheets and 1D cetrimonium bromide were electrostatically produced into an ultrathin hybrid film atop a PP separator and covered with carbon black/Se composites [94]; the resulting film displayed a remarkable capacity of 554 mAh g⁻¹. This is because CCNT and MXenes have a synergistic effect on polyselenide suppression. Due to their anisotropic structure and high 2D dimensions, which boost the polyselenide diffusion channel [95,96], MXenes-modified separators have been demonstrated to be beneficial in reducing the shuttle effect. Additionally, MXene exhibits high adsorption to polysulfides owing to the Lewis acid–base interaction involving polysulfides and Ti sites [97], which may also apply to polyselenides. Nazar et al. [98] have demonstrated that the surface environment of MXene increases Lewis acid–base chemisorption through thiosulfate production. Wang et al. [89] constructed an ultrathin CCNT/MXene/PP composite separator via self-assembly and filtration, retaining the benefits of MXenes. When utilized in Li-Se and Na-Se batteries, the as-prepared CCNT/MXene/PP separators exhibit an amplified Lewis acid–base action that prevents the shuttle effect. In the meantime, the interconnected CNT networks and increased interlayer spacing of MXene can enhance electrolyte wettability and enable ionic transport. To explore the electrochemical behavior of Se during cycling, an in situ permeation study was conducted, demonstrating a diminished shuttle effect and a protected Li anode by a CCNT/MXene/PP separator. Consequently, the performance of both Li-Se and Na-Se batteries is significantly boosted in cells using CCNT/MXene/PP separators.

Incorporating heteroatoms (such as nitrogen, oxygen, boron, phosphorus, sulfur, or codoping) into carbonaceous materials can provide customized polarity and powerful polar–polar or Lewis acid–base couplings with soluble polyselenides species [99]. It has been established that carbon interlayers or coatings doped with heteroatoms effectively trap active species [100]. The difference in electronegativities between carbon and heteroatoms can relocate electrons and enhance the polarity of carbon at the atomic scale. Moreover,
a codoping technique, such as N and S, N and P, has been employed to alter separators, resulting in a synergistic improvement for Polyselenides trapping over singly doped carbons [101,102]. Based on the solid chemical entrapment of polyselenides by nitrogen and sulfur atoms, a lightweight N, S codoped graphene blocking layer was used for Li-Se cells [91]. The interlayer provided a reversible capacity of 330.7 mAh g$^{-1}$ at 1 C after 500 cycles, but there was minimal capacity retention in the absence of the interlayer. Polyselenides can form strong bonds with polar functional groups such as hydroxyl, epoxy, ether, ester, carboxyl, and amino groups. Carbon-containing polar group functionalities were coated to improve the capture of polyselenides on the separator. These carbons are exemplified by hydroxyl-functionalized carbon nanotubes (CNTOHs) [103] and carbon quantum dots with oxygen-functional groups [104].

6. Li$_2$Se Cathode and Se Toxicity

In addition to the insurmountable concerns with cathodes, Li-Se batteries encounter substantial challenges with the lithium metal anode due to the extreme dendritic development. Dendrite growth invariably results in abrupt battery failure or perhaps a security risk. Combining a Li$_2$Se cathode with a Li-free anode is predicted to circumvent the problems associated with Li anodes [105]. As the utterly lithiated phase of Se, Li$_2$Se has the highest volume of any lithiated Se species, dispelling worries about electrode pulverization due to volumetric growth [106]. Compared to its Li$_2$S sibling, research on Li$_2$Se-based cathodes is currently scarce [107]. Li$_2$Se-based cathodes have several of the same issues as Li$_2$S cathodes. The very first is that it is insulating, resulting in slow electrode dynamics. Conducting hosts, on the other hand, is critical.

Nevertheless, because of Li$_2$Se’s excellent thermal stability, the standard melt-diffusion method cannot be used to fabricate composite materials. Unsatisfactory connections between Li$_2$Se particles and hosts, on the other hand, typically lead to high interface resistance and undesirable electrochemical processes. Wu’s group [86] presented a carbon-coated Li$_2$Se composite via a solution-based synthesis combining commercialized Se and Li triethylborohydride, followed by sequential coating with a PAN-derived N-doped carbon layer (labeled Li$_2$Se@NC) and a CVD-induced carbon layer (labeled Li$_2$Se@NC@C). Compared to pure Li$_2$Se (3.0 V), the first charge curves of Li$_2$Se@NC and Li$_2$Se@NC@C showed a significantly lower overpotential of 2.4 V, demonstrating a significantly reduced interface resistance due to the carbon covering. Additionally, the tiny particle dimension of Li$_2$Se ensures adequate interaction with the carbon layer, resulting in an enticing electrochemical performance. Additionally, they discovered that Li$_2$Se@NC@C with two coating layers exhibited superior cycling stability but lower rate performance (Figure 10a–d). This has been credited to the outer carbon layer, which hinders the diffusion of polyselenides while partially blocking the transfer of lithium ions.

Se is one of the essential elements necessary for the human body’s metabolism, although its abundance is well-known to be harmful. On the other hand, Se is not regarded as a very harmful substance. This is why selenium has garnered significant interest for medical uses [108], with Se nanoparticles being one of the most promising possibilities for radiation therapy. Although Se is more hazardous than its equivalent S, it is less hazardous than accessible electrolytes. As a result, Se toxicity cannot be regarded as a significant impediment to the development of Li-Se batteries. A critical factor is that elemental selenium is less hazardous than its soluble derivatives [109]. This is consistent with the necessity for Li-Se battery development, as Se solubility is associated with undesirable capacity fading. Thus, by minimizing the unwanted shuttle effect, the selenium cell’s toxicity is reduced compared to the toxic effects of Se nanoparticles. The electrochemical performance of several Se-based composites for lithium-Se batteries is compared in Table 3.
Figure 10. (a) Li$_2$Se nanoparticles synthesized by a chemical reaction between Se powder and Li reducing agent (such as a lithium hydride solution), (b) C-Li$_2$Se nanocomposite with Li$_2$Se nanocrystals dispersed in an elastic matrix of conductive carbon and (c) C-Li$_2$Se@C nanocomposite with C-Li$_2$Se composite particles further enclosed in protective carbon shells. SEM micrographs of (a) pure Li$_2$Se nanoparticles, (b) C-Li$_2$Se nanocomposite, and (c) C-Li$_2$Se@C nanocomposite illustrate experimental structures produced corresponding to each concept. (d) Retention of specific capacity and coulombic efficiency during 100 galvanostatic cycles at the C/2 rate. Reproduced with permission from [86], Copyright 2016, Elsevier BV.

Table 3. Electrochemical performance of different Se-based composites for lithium-Se batteries.

| Sample      | Se (wt.%) | Current Rate | Specific Capacity (mAh g$^{-1}$) | Ref.    |
|-------------|-----------|--------------|----------------------------------|---------|
| Se@PPy      | 90        | 0.07 C       | ~60 (50 cycles)                 | [110]   |
| Se@rGO      | 80        | 1 C          | ~270 (500 cycles)               | [63]    |
| Se/C@PANI   | 66        | 0.2 C        | ~520 (200 cycles)               | [111]   |
| Se@C        | 54        | 0.15 C       | ~430 (250 cycles)               | [112]   |
| Se@N-C      | 41.2      | 0.5 C        | ~300 (100 cycles)               | [113]   |
| Se/MiPCs    | 1.4       | 5 C          | ~511 (1000 cycles)              | [50]    |
| Se/MePCs    | 48        | 0.5 C        | ~310 (100 cycles)               | [45]    |
### Table 3. Cont.

| Sample             | Se (wt.%) | Current Rate | Specific Capacity (mAh g\(^{-1}\)) | Ref.  |
|--------------------|-----------|--------------|----------------------------------|-------|
| Se/HPCs           | 53        | 2 C          | ~320 (900 cycles)                | [114] |
| Se/G              | 80        | 0.1 C        | ~970 (500 cycles)                | [115] |
| Se/TiO\(_2\)      | 70.8      | 0.1 C        | ~155 (50 cycles)                 | [116] |
| Se/CNTs           | 85        | 0.1 C        | ~350 (100 cycles)                | [117] |
| Se/CNFs           | 75        | 0.1 C        | ~380 (350 cycles)                | [118] |
| Se/CNTs/rGO       | 70        | 0.74 C       | ~535 (80 cycles)                 | [119] |
| Se/CMK-3          | 49        | 0.1 C        | ~590 (50 cycles)                 | [43]  |
| Se@rGO            | 80        | 0.1 C        | ~530 (100 cycles)                | [120] |
| Se/HPCFs/rGO      | 57        | 0.5 C        | ~520 (200 cycles)                | [121] |
| SePAN             | 20        | 0.3 C        | ~160 (3400 cycles)               | [122] |
| Li\(_2\)Se@C      | 59        | 0.17 C       | ~300 (100 cycles)                | [86]  |
| Li\(_2\)Se@NCC@C  | 63        | 0.17 C       | ~290 (100 cycles)                | [123] |
| Se@COF            | 40        | 0.1 C        | ~950 (100 cycles)                | [124] |

### 7. Conclusions and Future Outlook

Se has gained popularity as a cathode material for Li-Se batteries due to its increased capacity density. Additionally, Se’s strong electronic conductivity permits fast electrode kinetics, efficient active material consumption, and the creation of universal host-free cathodes. Thus far, it has been demonstrated that the electrochemistry of Li-Se batteries is electrolyte-dependent. The redox transition is a single-phase for carbonate electrolytes, revealing the absence of intermediates in the lithiation/delithiation processes. However, powerful side reactions between Se anions and the electrolyte’s carboxyl groups continue to be a significant hindrance. In ether-based electrolytes, multistep lithiation is associated with the production of intermediates, and the remarkable shuttle effect is observed. As a result, hosts capable of immobilizing Se species are commonly required to reduce both side reactions and polyselenide shuttling, two phenomena that have attracted significant attention in recent years.

Se electrodes have various problems in Li-Se batteries, including insufficient Se loading, low Se utilization, excessive volume expansion, low electrical conductivity, low coulombic efficiency, and poor cycle performance. This article briefly summarizes current advancements in Li-Se batteries. We concentrated on ways of significantly enhancing Li-Se battery electrochemical performance, such as electrolyte selection. We discovered that optimizing electrode material design and creating a novel battery structure are critical factors in building Li-Se batteries with excellent electrochemical performance. Numerous approaches have been taken to address these issues, including the use of micro/nanostructured carbon materials (for example, porous carbon, carbon nanotubes, and carbon nanofibers), free-standing conductive matrices, transition metal compounds (for example, ZnO, TiO\(_2\), ZnSe, and MoSe\(_2\)), and Se alloys. Micro/nanostructured carbon with suitable porous structures, high specific surface areas, dimensionality, and surface functionalization can increase the conductivity and loading of active Se in the electrode, absorb polyselenide intermediates, buffer volume change, and thus improve overall electrochemical performance.

Further extensions of the Se host materials include polarized, heteroatom-doped, and hybrid multifunctional materials. Additionally, by using interlayers and separators, the electrochemical performance of Li-Se batteries might be enhanced. Even though significant advances in Li-Se cells have been made, the practical implementation of Li-Se cells still faces significant difficulties and obstacles, and some ideas are described below:
(1) Cost-effective carbon-based materials should be chosen. Se cathodes have limited applications despite their high adsorptive and catalytic activity due to the exorbitant expense of their constituent elements.

(2) The commercial implementations of Li-Se batteries are hindered by the instability of electrodes and the suppression of the shuttling effect during the charge-discharge process. Constructing a stable, effective solid electrolyte interphase (SEI) layer could restrict the dissolution of polyselenide and improve cycle performance.

(3) The As-Se cathode is a dynamic electrochemical system whose electrochemical characteristics are intimately connected to the arrangements of the entities. Thus, the effectiveness of any single carbon material is highly dependent on the cathode’s architecture and its ability to respond to minor adjustments. In light of the vast amount of research data reported, the screening of numerous outcomes, and the in-depth data analysis, it is highly suggested to use machine learning to determine that the incorporation is not only a simple material but also an integral component and arrangement of the entire network.

(4) The thickness of interlayers typically spans from a few tens to hundreds of microns. The substantial thickness and high areal mass loading can lead to a significant increase in the weight of the interlayers, hence decreasing the practical, specific energy. Therefore, prospective interlayer technology research should construct lightweight interlayers employing various techniques. Undoubtedly, the destiny of interlayer technology for high-energy Li-Se batteries depends on the development of ultralight interlayers that can still perform their role in narrower arrangements.

(5) The interlayers have been converted from a carbon-based substance to oxides and other chemicals. The interlayer structure of the future will presumably be considerably more diverse.

(6) The usage of interlayers is hampered by the potential for high costs or a dearth of raw materials. To achieve the industrialization of Li-Se batteries, they must contend with established LIBs and other energy storage devices. Therefore, we must spare no effort in the development of functional systems.

Despite the significant advances in scientific investigations on Li-Se batteries, there are still significant difficulties and possibilities that require more fundamental comprehension. We anticipate that a promising future for Li-Se cells with high energy storage network cell technology is likely to be realized due to the combined achievements of researchers from several related disciplines.

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