Selenium-Based Cathode Materials for Lithium Batteries

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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.

Li-Se batteries cathode interlayers electrochemical performance

1. 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_2Se in a single step without the creation of unstable intermediates that occur in ether-based electrolytes [1][2][3][4][5][6]. The loss of active material caused by the development of intermediate polyselenides results in poor cycling performance of Li-Se batteries. Amine et al. [7][8] 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 ration), 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_2Se without creating intermediate polyselenides that dissolve in electrolytes and cause capacity fading.

Nonetheless, Yi's group [9] showed that direct conversion of Se to Li_2Se 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 [10]. 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.

2. 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.

2.1. 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.

2.1.1. 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. [11] 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 $m^2 g^{-1}$, 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 Li_2Se_2/Li_2Se, respectively. At 2.2 V, there is a single anodic peak, showing the process of overlap delithiation [8]. All consecutive scans overlap, indicating that the MCM-3.8/Se cathodes are stable during the repeated lithiation/delithiation process.

2.1.2. 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 [5] 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/MCPS 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. [12] 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.

2.1.3. Hierarchical Porous Carbon/Se Composites

Hierarchical porous carbon contains pores of different sizes, including microspores that help stabilize polyselenides and accommodate active materials, macrospores 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. [13] 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_2Se, 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.

2.2. 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. [14] 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 [15] 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. [16] 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.

2.3. 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 [17] 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. [18] 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.

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. [19] 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}.

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. [20] 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. [21] 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 [22] 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.

3. Li_2Se 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_2Se cathode with a Li-free anode is predicted to circumvent the problems associated with Li anodes [23]. As the utterly lithiated phase of Se, Li_2Se has the highest volume of any lithiated Se species, dispelling worries about electrode pulverization due to volumetric growth [24]. Compared to its Li_2S sibling, research on Li_2Se-based cathodes is currently scarce [25]. Li_2Se-based cathodes have several of the same issues as Li_2S 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_2Se's excellent thermal stability, the standard melt-diffusion method cannot be used to fabricate composite materials. Unsatisfactory connections between Li_2Se particles and hosts, on the other hand, typically lead to high interface resistance and undesirable electrochemical processes. Wu's group [26] presented a carbon-coated Li_2Se 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_2Se@NC) and a CVD-induced carbon layer (labeled Li_2Se@NC@C). Compared to pure Li_2Se (3.0 V), the first charge curves of Li_2Se@NC and Li_2Se@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_2Se ensures adequate interaction with the carbon layer, resulting in an enticing electrochemical performance.

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 [27], 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 [28]. 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 1**.

Table 1. 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)	[29]
Se@rGO	80	1 C	~270 (500 cycles)	[30]
Se/C@PANI	66	0.2 C	~520 (200 cycles)	[31]
Se@C	54	0.15 C	~430 (250 cycles)	[32]
Se@N-C	41.2	0.5 C	~300 (100 cycles)	[33]
Se/MiPCs	1.4	5 C	~511 (1000 cycles)	[34]
Se/MePCs	48	0.5 C	~310 (100 cycles)	[35]
Se/HPCs	53	2 C	~320 (900 cycles)	[36]
Se/G	80	0.1 C	~970 (500 cycles)	[37]
Se/TiO_2	70.8	0.1 C	~155 (50 cycles)	[38]
Se/CNTs	85	0.1 C	~350 (100 cycles)	[39]
Se/CNFs	75	0.1 C	~380 (350 cycles)	[40]
Se/CNTs/rGO	70	0.74 C	~535 (80 cycles)	[41]
Se/CMK-3	49	0.1 C	~590 (50 cycles)	[42]
Se@rGO	80	0.1 C	~530 (100 cycles)	[43]
Se/HPCFs/rGO	57	0.5 C	~520 (200 cycles)	[44]
SePAN	20	0.3 C	~160 (3400 cycles)	[45]
Li_2Se@C	59	0.17 C	~300 (100 cycles)	[26]
Li_2Se@NC@C	63	0.17 C	~290 (100 cycles)	[46]
Se@COF	40	0.1 C	~950 (100 cycles)	[47]

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