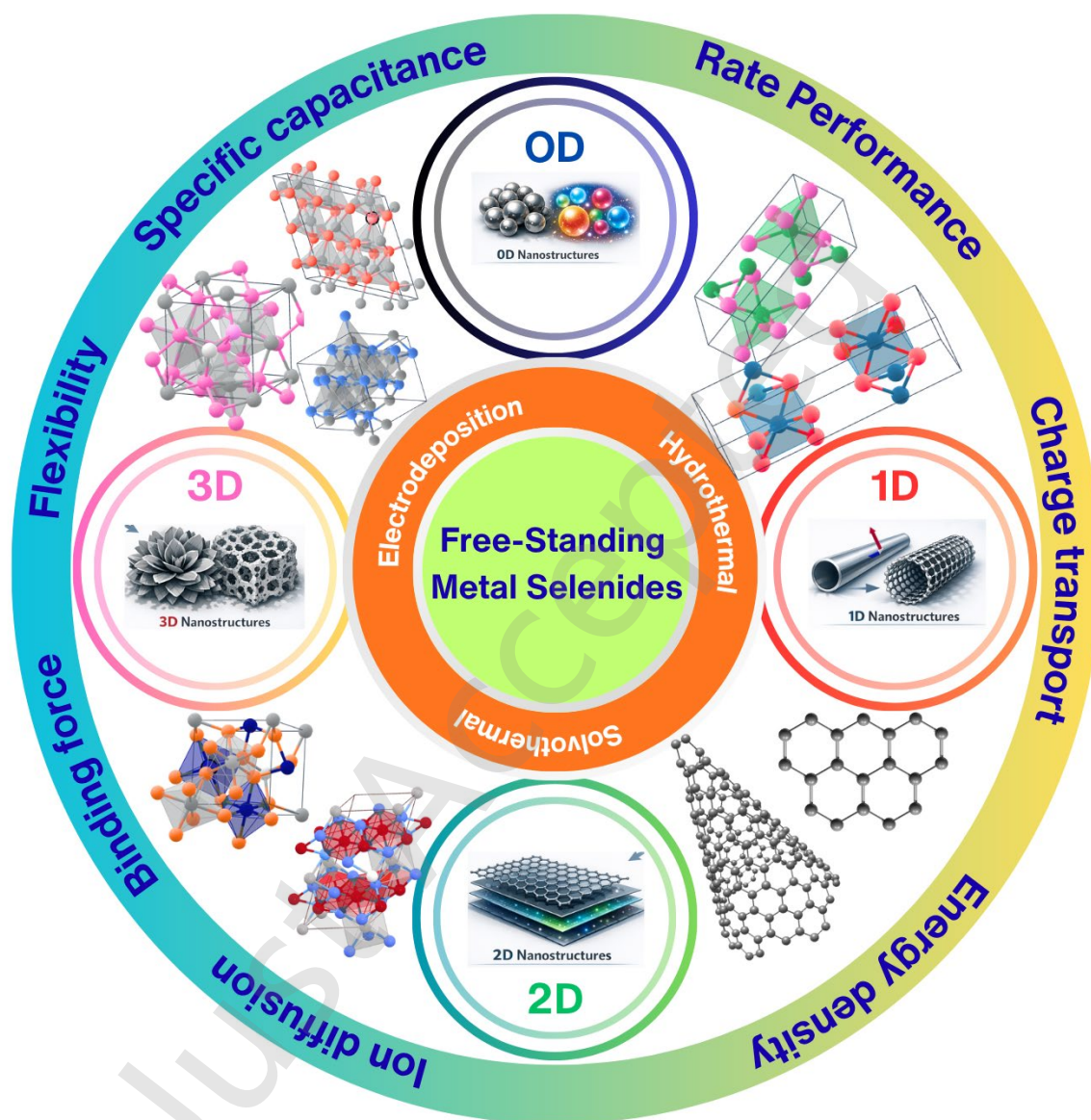


GRAPHICAL ABSTRACT



Self-supported metal selenide architectures provide an effective platform for high-energy supercapacitors by integrating binder-free electron pathways with abundant redox-active sites. This study establishes a structure–property relationship across one-dimensional to three-dimensional architectures, demonstrating that dimensional engineering regulates ion transport, charge-transfer kinetics, and electrochemical energy-storage performance.

Review Article

Engineering self-supported metal selenide architectures for high-energy supercapacitors

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ABSTRACT

Transition-metal selenides are considered emerging electroactive materials for high-performance supercapacitors owing to their high electrical conductivity, multivalent redox chemistry, and tunable electronic structures. The distinct features facilitate rapid charge-transfer kinetics and higher charge-storage efficiency, demonstrating metal selenides as potential pseudocapacitive and battery-type electrodes. Moreover, Electrode design is a key factor for assessing the electrochemical performance of supercapacitors. Notably, developing self-supported metal selenide-based electrode formations effectively boosts the electroactive material-current collector interface and improves charge transfer kinetics, resulting in improved durability of electrodes. Recent advances in materials synthesis and interfacial engineering have enabled the direct integration of metal selenide nanostructures onto conductive frameworks. Controlled fabrication techniques facilitate the production of ordered nanoarrays and hierarchical designs. The unique nanostructures offer consistent electron transport channels, reduced ion diffusion lengths, and increased mechanical flexibility, ultimately resulting in improved rate capability and cycling stability. This review explains the charge-storage mechanism in metal selenides and the rationale for selecting self-supported electrode configurations. The structural dependence of the electrochemical performance of metal selenide-based electroactive materials, such as hybrid materials and multicomponent heterostructures, is systematically analyzed. Finally, the challenges and future perspectives of metal-selenide-based materials for the development of self-supported supercapacitors in next-generation sustainable clean energy storage technologies.

KEYWORDS: supercapacitors, self-supported electrodes, metal selenides, enhanced electrochemical performance

1 Introduction

The rapid transition toward electric vehicles, wearable electronics, and sustainable power systems demands advanced energy-storage technologies with high energy and power densities, rapid charge kinetics, and highly stable devices [1–6]. Rechargeable batteries, particularly lithium-ion batteries (LIBs), and dielectric capacitors are two foremost electrical energy storage technologies, as shown in the Ragone plot (Fig. 1a) [7,8]. Dielectric capacitors store charge based on the physical adsorption of charges through electric dipole polarization (Fig. 1b), exhibit ultrahigh power, rapid response times, high operating electric fields ($\sim \text{MV m}^{-1}$), and excellent cycling stability ($>10^5$ cycles) [9–11]. Nevertheless, their relatively low energy density ($<0.1 \text{ Wh kg}^{-1}$) significantly restricts their broader real-time applications. In contrast, LIBs rely on electrochemical energy storage mechanisms (Fig. 1c) and have delivered gravimetric energy densities approaching 300 Wh kg^{-1} . Due to their high energy density, LIBs are widely used in hybrid and fully electric vehicles, portable electronics, and portable energy storage systems [12]. Despite these advantages, LIBs are limited in their applicability due to poor operational lifespan, low power density, and the leakage of toxic components, which hinder their use in next-generation applications [13]. Supercapacitors have emerged as key components in this context, offering improved rate capability and long cycle life compared to batteries, while producing significantly larger energy densities than traditional dielectric capacitors [14,15]. Supercapacitors address the gap between dielectric capacitors and batteries by providing rapid charge–discharge capability, superior power density, and exceptional cycling stability. However, their relatively poor energy density compared to batteries remains a significant challenge for real-time applications. The electrochemical properties of supercapacitors are typically characterized by specific capacitance, energy density, and power density; the energy density ($E = 1/2(CV^2)$) and power density ($P = V^2/4Rm$) are derived from charge–discharge data [16].

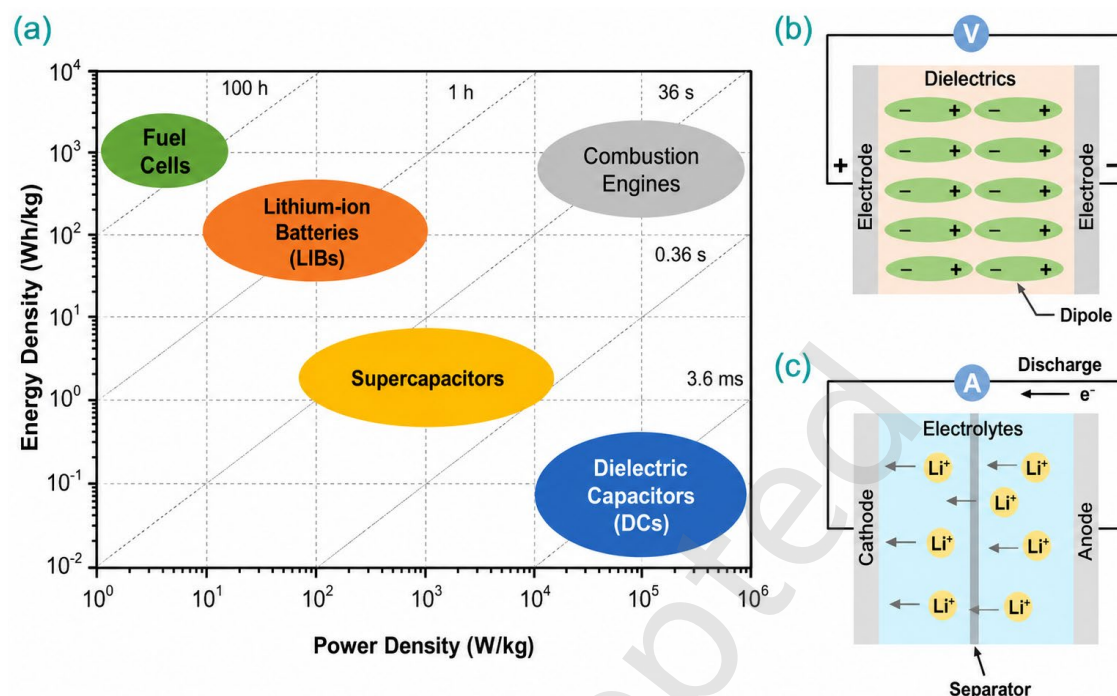


Figure 1 (a) Ragone plot of electrical energy-storage and conversion technologies performance. (b) Graphical representation of a dielectric capacitor in the charge storage process. (c) Schematic illustration of the discharge process. Reproduced under the CC BY license from Ref. [17] © 2022, Wiley-VCH GmbH.

Based on the charge storage mechanisms in electrode materials, supercapacitors have been categorized into three types: electric double-layer capacitive (EDLC), pseudocapacitive, and battery-type [14]. In EDLC (carbon nanotubes (CNT), activated carbon (AC), reduced graphene oxide, graphene), charge is stored through the physical electrostatic adsorption/desorption of ions on the electrode surface. In contrast, pseudocapacitive materials (metal oxides, metal sulfides, conductive polymers, etc.) store energy via surface-dominated reverse redox reactions on/near the electrode surface, whereas battery-type materials store charge via diffusion-dominated Faradaic redox reactions on the electroactive materials. Despite tremendous advances, commercial carbon-based EDLC supercapacitors remain impeded by low energy density. On the other hand, transition metal chalcogenides have been widely studied for electrochemical applications because of their notable physicochemical and electrical properties [18–20]. In particular, transition-metal chalcogenides enable rapid, reversible faradaic redox processes, significantly boosting charge storage capacity. Metal selenides have emerged as notable electrode materials due to their higher electrical conductivity, multiple valence states, and high theoretical capacitance compared with transition-metal oxides/sulfides [21,22]. Notably, selenium has significantly higher intrinsic electrical conductivity than sulfur ($1 \times 10^{-3} \text{ S m}^{-1}$ for Se vs. $5 \times 10^{-28} \text{ S m}^{-1}$ for S) [23,24] due shows lower electronegativity of Se and weaker binding than those of S and O atoms [25] and an analogous theoretical volumetric capacity with sulfur (3253 mAh cm^{-3} for Se, 3467 mAh cm^{-3} for S) [26]. Moreover, the rich reversible redox properties facilitate rapid charge transfer and provide more electroactive sites, confirming their potential as electrode materials for high-energy-density supercapacitors. In addition, transition metal selenides

exhibited superior electrochemical performance compared to metal sulfides and metal phosphides due to the more covalent and polarizable M–Se bonds, which resulted in significantly higher intrinsic electrical conductivity and lower charge-transfer resistance, thereby enabling rapid and reversible Faradaic reactions even at high scan rates [27–29]. In supercapacitor applications, both single- and multi-metal selenides predominantly exhibit pseudocapacitive behavior via fast, reversible Faradaic redox reactions at the electrode–electrolyte interface, enabling high specific capacitance, excellent rate performance, and long-term stability [28,30]. For alkali-ion batteries, metal selenides typically store charge through a synergistic intercalation–conversion process, resulting in high theoretical capacities and accelerated reaction kinetics by tuning their structures into nanoscale, layered, or heterointerface-rich architectures [31,32]. For instance, MoSe₂ with a Se vacancy showed a specific capacitance of 754.54 F g^{−1} at 1 A g^{−1}. The MoSe₂-2//AC asymmetric device delivered an energy density of 42.13 Wh kg^{−1} at a power density of 417.72 W kg^{−1}, demonstrating that vacancy engineering enhances the electrochemical performance of metal selenide materials [33]. The above examples show that the introduction of Se vacancy engineering plays a vital role in improving supercapacitor performance. The hexagonal MnCo-selenide exhibited an excellent specific capacity of 270 mAh g^{−1} at 2 A g^{−1}, with excellent cyclic stability, retaining 91% of its capacity after 10,000 cycles [34]. Besides, a hybrid MCS HS-150-6 h//AC supercapacitor exhibited an energy density of 29.39 Wh kg^{−1} at a power density of 399.9 W kg^{−1}, and the fabricated device proved superior capacitance retention of 97 % after 25,000 cycles. Recently, the CoNiMnSe/PBA composite electrode displays a specific capacity of 1360 C g^{−1} at 1 A g^{−1} and maintains 1024 C g^{−1} at 10 A g^{−1}. The asymmetric supercapacitor (CoNiMnSe/PBA//AC) presented a specific capacity of 384 C g^{−1} (1 A g^{−1}), a power density of 800 W kg^{−1}, and a maximum energy density of 85.3 Wh kg^{−1} [35]. The aforementioned studies clearly indicate that the metal selenides could be a potential electrode material for supercapacitor applications. In addition, the morphology of electroactive materials meaningfully influences the electrochemical performance of the supercapacitor. The unique structural properties significantly control charge-transfer behavior and ion-transport paths. For instance, Co-MoSe_x double-shelled hollow nanocages exhibited exceptional electrochemical performance due to their hierarchical hollow architecture [36]. It is clear that the morphology significantly influences charge-transfer efficiency, ion diffusion kinetics, interfacial resistance, and reaction rates in electrode materials.

Nevertheless, the practical performance of metal selenides is significantly controlled by electrode architecture [37]. Conventional slurry-cast electrodes comprise polymeric binders and conductive carbon additives that restrict ion pathways, increase the dead mass, resulting in increased interfacial resistance, and lead to structural degradation during long-term cycling [38]. Instead, self-supported electrode architectures developed directly on current collectors (Nickel foam, Copper foam, stainless steel sheet, carbon cloth, etc.) significantly boost electrochemical performance by removing dead and inactive constituents. Moreover, self-supported configurations enhance the electrical contact between the current collector and the electroactive materials, facilitate multidirectional charge diffusion, and provide robust mechanical stability [39], which are vital for real-world volumetric and areal energy storage devices. Hence, self-supported electrodes could be a promising approach for realizing next-generation

supercapacitors with superior performance, lightweight architecture, flexibility, and scalable manufacturing potential. Various studies confirm that self-supported metal selenides or metal selenide-based hybrid materials exhibit enhanced electrochemical performance due to improved electrical conductivity, rapid ion diffusion, and an excellent electrode-electrolyte interface [40–42]. This review highlights the recent advancement in self-supported transition-metal selenide electrodes for high-performance supercapacitors. It first discusses the fundamental properties and charge-storage mechanisms of metal selenides, then provides an overview of the major synthesis strategies for fabricating self-supported electrodes. Then, it critically examines the electrochemical performance of one-dimensional (1D), two-dimensional (2D), and three-dimensional (3D) metal selenide architectures, emphasizing the relationship between structure and energy storage performance. Finally, the key challenges, emerging opportunities, and future research directions for developing practical, high-performance, and sustainable metal selenide-based supercapacitors are discussed.

2 Structure and properties of metal selenides

Metal selenides display a rich diversity of crystal structures and play a key role in determining the electrochemical performance of supercapacitors. Broadly, Metal selenides can be classified into layered and non-layered structures, each having distinct advantages for charge storage [37]. As illustrated in Fig. 2a, layered metal selenides with the general formula MSe_2 ($M = Ti, Zr, Hf, V, Nb, Ta, Cr, Mo, W, Mn, Re, Ru, Ni, Pd, \text{ and } Sn$) crystallize in multiple polymorphs, including 1T, 2H, and 3R phases. These phases vary in the coordination geometry of the metal center and selenides, with the 1T phase exhibiting octahedral coordination, whereas the 2H and 3R phases are trigonal prismatic [43]. Different coordination modes of atoms will influence the stacking arrangements of adjacent layers, as depicted in Fig. 2b. The 1T polymorph exhibits tetragonal symmetry, with a single layer in each repeat unit. In contrast, the 2H phase shows hexagonal symmetry with a two-layer stacking arrangement, whereas the 3R phase exhibits rhombohedral symmetry with a three-layer periodic stacking pattern [37]. Among them, the 1T phase is suitable for supercapacitor applications owing to its metallic nature and intrinsic ionic conductivity, which facilitates fast ion transport during charge–discharge processes. The layered Se–M–Se sandwich structure with weak van der Waals interactions enables effective ion intercalation, and interlayer spacing can be adjusted via structural modifications such as doping or defect engineering, thereby significantly affecting ion diffusion kinetics [37].

In addition to MSe_2 systems, metal selenides exhibit a wide range of stoichiometries (MSe, MSe_3, M_xSe_y) and diverse crystal structures, as summarized in Fig. 2c. Metal selenides with formula MSe display layered architectures distinct from those of typical MSe_2 systems. The layered MSe -type materials, including $FeSe, NiSe, CuSe, SnSe, \text{ and } InSe$, possess unique crystal structures that influence their electrochemical performance. For instance, $FeSe, NiSe, \text{ and } CuSe$ exhibit a tetragonal phase ($P4/nmm$), whereas $SnSe$ has an orthorhombic ($Pnma$) structure. $InSe$ exhibits polymorphism, primarily in the β and γ phases, which belong to a hexagonal lattice ($P6_3/mmc$) due to different monolayer stacking orders. Another indium-rich

phase, layered In_2Se_3 , with the α -phase (R_{3m}), is thermally stable at room temperature. Furthermore, layered MSe_3 -type compounds ($M = \text{Ti, Zr, Hf}$) crystallize in the monoclinic system, and the layered MSe_3 structure is constructed from prismatic units with transition-metal atoms at the centers and selenium atoms at the corners. The layered MSe_3 architectures provide enlarged interlayer spacing, which is beneficial for ion intercalation and charge transport in supercapacitor applications.

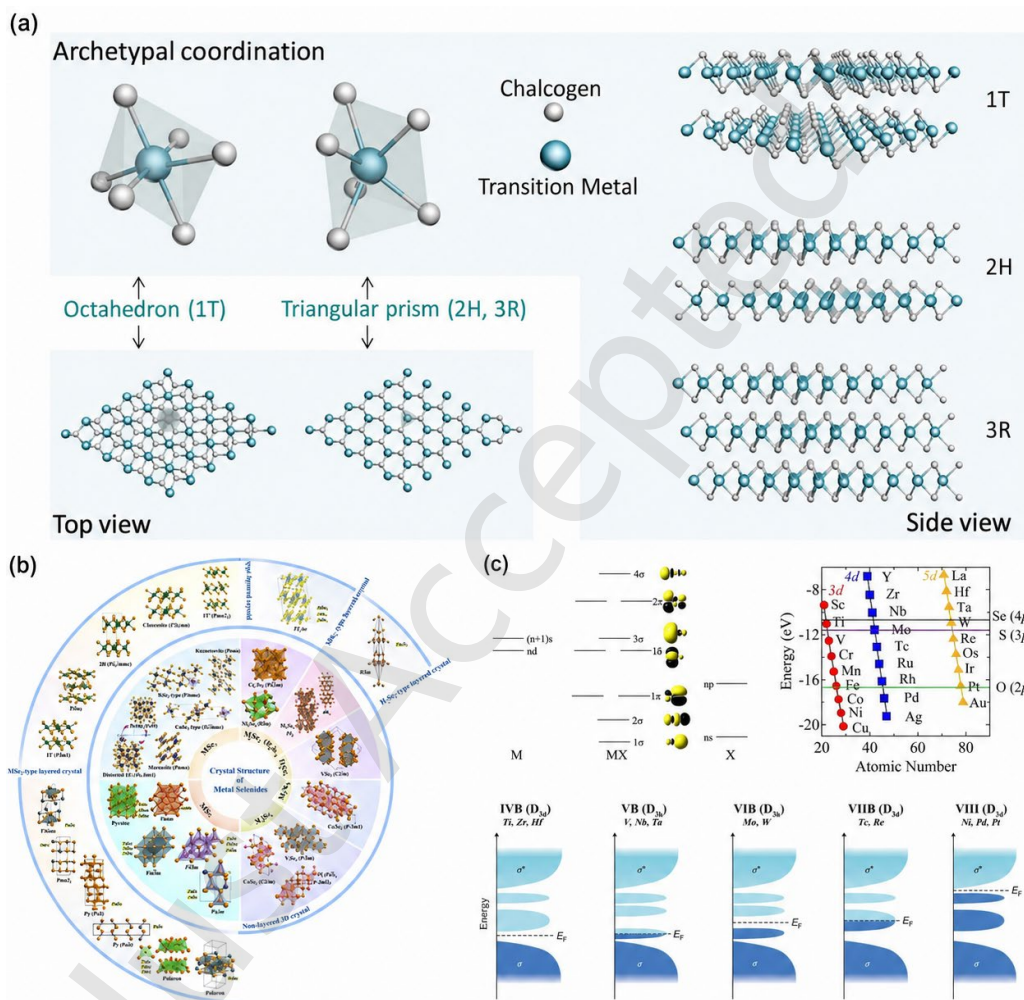


Figure 2 (a) Schematic of MSe_2 and its typical 1T, 2H, and 3R structures (top and side view). (b) Illustration of the crystal structure of metal selenides, and the non-layered 3D crystals are shown in the inner ring while the layered crystals are in the outer ring. (c) Schematic representations illustrating the qualitative molecular orbital framework of transition metal selenides, including atomic orbital energy levels (3d, 4d, and 5d) and the progressive filling of d orbitals within the bandgap between bonding (σ) and antibonding (σ^*) states across group IVB–VIII transition metals. Reproduced with permission from Ref. [37] © 2022, Elsevier Ltd.

In contrast to layered metal selenide structures, certain MSe₂ compositions such as FeSe₂, RuSe₂, CoSe₂, RhSe₂, IrSe₂, CuSe₂, AgSe₂, and AuSe₂ exhibit non-layered structures. Many of the MSe₂ compounds, particularly RuSe₂, CuSe₂, and AgSe₂, exhibit pyrite-type cubic structures (Pa $\bar{3}$), whereas FeSe₂ crystallizes in an orthorhombic marcasite structure (Pnma), forming a fully three-dimensional network. CoSe₂ and RhSe₂ consist of uniform metal–selenium octahedral coordination units, resulting in dense three-dimensional lattices. Most of the metal selenides (MSe, MSe₂, M₃Se₂, M₂Se₃, M₃Se₄, M₂Se₉, and M₉Se₈) belong to non-layered 3D crystal structures, as shown in the inner ring of Fig. 2c, and their anisotropies can promote adsorption/desorption reactions on specific crystal planes.

The electrochemical performance of metal selenides predominantly depends on their electronic structure. The electronic configuration arises from strong interactions between the transition-metal d orbitals and the selenium p orbitals. The interaction between transition metals and chalcogenides results in delocalized electronic states, significantly enhancing electrical conductivity compared to oxide- or sulfide-based materials. The presence of partially filled d-orbitals enables flexible redox chemistry, allowing multiple oxidation states of the metal centers to contribute to charge storage, leading to pronounced pseudocapacitive behavior. Furthermore, the electronic structure of the metal selenides can be successfully tuned through doping, defect introduction, and heterostructure formation. These modifications alter the density of states near the Fermi level, thereby enhancing charge-transfer kinetics and reducing internal resistance. As a result, metal selenides improve rate performance and coulombic efficiency in supercapacitor devices. Therefore, the structural diversity of metal selenides offers a versatile platform for tailoring charge-storage behavior, enabling a balance between energy density (battery-like behavior) and power density (capacitor-like behavior).

3 Charge storage mechanisms of metal selenides

As we all know, the charge stored in a supercapacitor arises from EDLC and pseudocapacitive mechanisms. Transition metal selenides store charge through EDLC, pseudocapacitive, or battery-type Faradaic processes, which predominantly depend on electrode composition, structural engineering, and electrolytes [28,44].

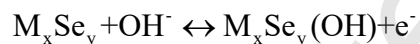
3.1 Electric double-layer capacitance (EDLC)

Metal selenides primarily store energy via Faradaic mechanisms; nevertheless, a significant non-Faradaic or EDLC contribution can arise from ion adsorption at the electrode-electrolyte interface (Fig. 3). The EDLC behavior is particularly pronounced in porous nanostructured electroactive materials or in materials integrated with carbonaceous materials [45]. In particular, the carbonaceous materials integrated with metal selenides facilitate double-layer charge accumulation with intrinsic redox Faradaic reactions [46]. Besides, carbon-integrated transition-metal selenide-based electrodes exhibit dual EDLC–

pseudocapacitive behaviors in cyclic voltammetry (CV) and galvanostatic charge–discharge (GCD) analyses, with the EDLC contribution becoming more prominent at high scan rates [47].

3.2 Pseudocapacitive Faradaic reactions

Metal selenides exhibit pseudocapacitive behavior primarily via rapid, reversible near-surface or surface Faradaic redox reactions. Metal oxides, metal sulfides, metal phosphides, and polymers are typical examples of pseudocapacitive materials [48,49] (Fig. 3). The higher electrical conductivity of metal selenides facilitates rapid electron transport and enhances charge storage kinetics [50]. In alkaline electrolytes (KOH, NaOH, etc.), charge storage originates from the redox transition of metal cations, associated with OH[−] insertion/extraction at the electrode/electrolyte interface:



3.3 Battery-type conversion and intercalation processes

In addition to near-surface PC contributions, bulk conversion reactions may also occur during the prolonged charge–discharge process, predominantly at low scan rates and high current densities [51]. In alkaline electrolytes, metal selenides can partially convert into metal hydroxides/oxyhydroxides and selenium or polyselenide species and act as electroactive redox sites, providing additional charge storage during the cycling process:



Layered or defect-rich selenides operating in neutral or multivalent electrolytes can additionally store charge through Na⁺, K⁺, or Zn²⁺ intercalation into the selenide lattice, thereby allowing a hybrid mechanism that combines intercalation-driven PC with conversion-type Faradaic reactions [37] (Fig. 3).

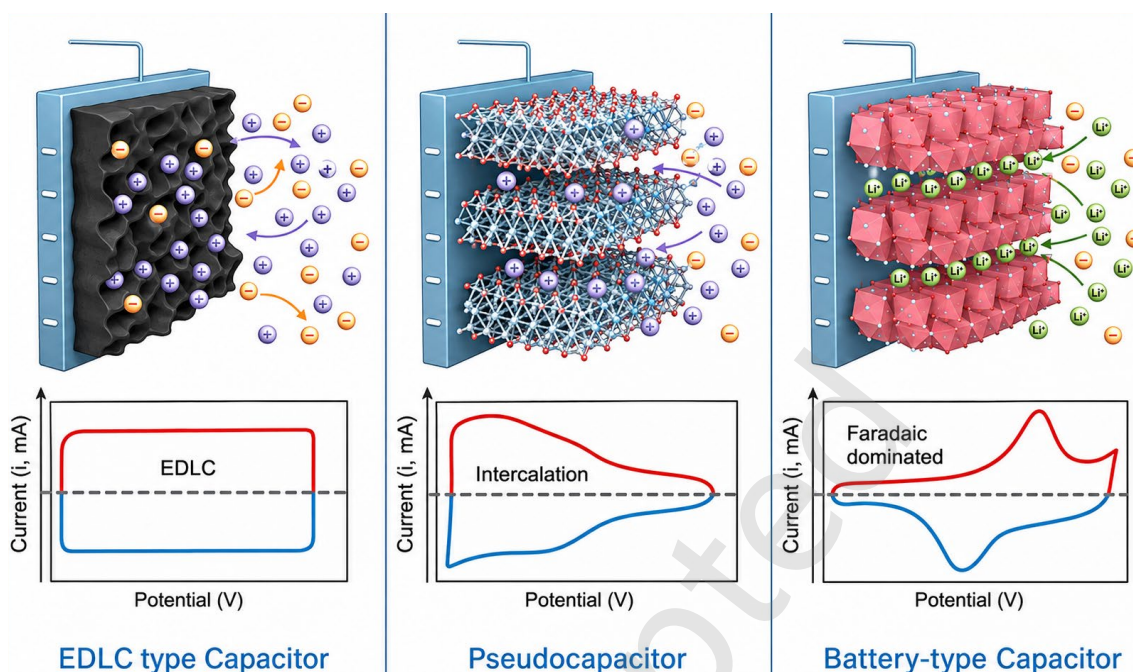


Figure 3 Graphic illustrations of supercapacitor electrodes based on diverse charge-storage mechanisms. Reproduced with permission from Ref. [52] © 2023, Elsevier Ltd.

The charge-storage mechanisms in supercapacitors provide the basis for designing self-supported metal selenide electrodes for high-performance supercapacitor devices. The electrochemical performance depends not only on redox reactions and electrolytes but also significantly on the electrode-current interface and ion transport through the microstructure of the electrode materials [53]. Therefore, it is important to understand the synthesis methods, morphological features, ion-transport channels, and charge-transfer resistance to achieve self-supported transition-metal selenide electrodes for next-generation supercapacitors.

4 Rationale for the selection of self-supported electrodes

As we all know, the electrode plays a vital role in the electrochemical performance of a supercapacitor. In conventional slurry-based electrodes, the active materials, polymeric binders, and conductive additives are mixed to form a slurry, which is further coated over the current collector [54]. However, the polymeric binders are electrically and chemically inactive, resulting in internal interfacial resistance, hindering access to electroactive sites, and reducing the overall performance of electrode materials [55]. In contrast, self-supported architectures enable the direct growth or self-support of active materials on conductive substrates, thereby maintaining continuous electron pathways, increasing the number of electroactive sites, enhancing redox kinetics, and improving cyclic performance [55]. Moreover, mechanically stable electrodes can be achieved, significantly improving cycling stability by preventing electrode damage during charge-discharge [15]. Additionally, the removal of dead mass further enhances gravimetric and

volumetric energy densities [56]. Moreover, the weak binding between active materials and current collectors can severely limit their cycling stability [57]. In contrast, the development of mechanically robust self-supported electrodes offers a feasible approach to overcome the restrictions associated with polymeric binders [55,58]. Fig. 4 compares conventional slurry-based electrodes with self-supported electrode materials. Fig. 5 shows the advantages of self-supported electrodes across various electrochemical behaviors. The engineered nanoarray morphology suppresses electrode agglomeration, shortens ion diffusion pathways, and shields against volumetric degradation during charging and discharging. Moreover, the continuous interface between the electroactive materials and the current collector confirms efficient charge-transfer kinetics and steady electrochemical reactions of the electrodes. Nanoarray architectures greatly enhance electrochemical performance; however, the increased surface area in the nanoarrays also increases the electrode–electrolyte interfacial area, thereby promoting undesired side reactions such as electrolyte decomposition, surface oxidation, irreversible ion adsorption, and the formation of resistive interfacial layers during long-term cycling [59]. The undesired side reactions increase charge-transfer resistance, decrease coulombic efficiency, and cause structural damage, thereby reducing electrochemical performance. Superior electrochemical behavior depends on both optimal electroactive sites and structural architecture, which balance ion transport and structural stability to achieve high-performance supercapacitors [15]. Rational design of nanoarray density, nanosheet thickness, and hierarchical porosity enable rapid ion transport and limited electrolyte exposure, while a suitable current collector effectively suppresses parasitic reactions. Therefore, the remarkable cycling stability of self-supported metal selenide nanoarrays predominantly depends on morphology, interfacial chemistry, and electron/ion transport, rather than on high surface area alone. Compared with slurry-based powder-form electrodes, self-supported electrodes offer several advantages due to their unique features, including flexibility, light weight, and stretchability [60]. The unique advantages of self-supported electrodes make them highly promising electrode materials for next-generation supercapacitors, offering superior electrochemical performance, lightweight configuration, and stretchable devices. For instance, Se-deficient vanadium diselenide (VSe_2) nanosheets were fabricated on stainless steel as a current collector for aqueous zinc-ion battery applications, and the $\text{VSe}_{2-x}\text{-SS}$ electrode shows a high specific capacity of $265.2 \text{ mA h g}^{-1}$ at 0.2 A g^{-1} [61]. Binary nickel-cobalt selenide (NCSe) and iron-cobalt selenide (FCSe) electrodes with high surface area were developed directly on carbon fibers and utilized as an anode for SIBs [62]. The self-supported electrodes show superior electrochemical performance, with energy capacities of 665 mA h g^{-1} and 589 mA h g^{-1} for FCSe/CFs and NCSe/CFs at 0.1 A g^{-1} , respectively, and exceptional rate capabilities. Additionally, the NCSe/CFs electrode demonstrates outstanding stability during long-term cyclic performance, with over 1000 cycles at 1 A g^{-1} . Overall, $\text{VSe}_{2-x}\text{-SS}$ with defects, NCSe/CFs, and FCSe/CFs were developed to improve charge-transfer kinetics. Notwithstanding differences in composition and application, all three approaches reveal that direct growth on conductive substrates eliminates the binder, thereby reducing resistance, strengthening interfacial electrical contact, shortening ion diffusion pathways, and improving cycling durability. The findings suggest that electrode architecture often contributes as significantly as intrinsic material chemistry toward electrochemical performance.

In self-supported electrode configurations, current collectors with electroactive materials are key components (~15–20% of a device's total weight) that significantly influence the gravimetric/volumetric energy density, power density, and long-term cycle stability of the supercapacitor cell [63]. Suitable current collectors are imperative for achieving high-energy-density devices with excellent electrical conductivity and rapid charging and discharging behavior [64]. The ideal current collector must have high electrical conductivity, low contact resistance with the electroactive material, stable, strong bonding with the active electrode material, high specific surface area, high mechanical modulus, low weight, high thermal stability, high electrochemical stability, and be eco-friendly [63]. Metal current collectors (foil, porous metal) and carbon-based current collectors have been widely used in supercapacitor devices. Yadav et al. studied the effect of the current collector on supercapacitor electrodes with NiCo_2O_4 nanospheres [65]. The NiCo_2O_4 nanosphere slurry was coated onto nickel foam, flexible graphite sheet, copper foil, and stainless steel foil. Notably, the nickel foam exhibits a specific capacitance of 128 Fg^{-1} , and $\text{NiCo}_2\text{O}_4\text{-Ni}$ delivers the highest specific capacitance of 960 Fg^{-1} at 1 Ag^{-1} , compared with the NCO-FG (237 Fg^{-1}), NCO-Cu (169 Fg^{-1}), and NCO-SS (64 Fg^{-1}) electrodes. Zhu and Lu developed activated carbon-based CR2032 coin cells. They investigated the electrochemical performance of activated carbon on titanium foil, nickel foil, 316 stainless steel foil, and graphite paper in an aqueous electrolyte [66]. Among them, stainless steel-based supercapacitors exhibit significantly faster self-discharge at a wide potential window (1.6 V) compared to titanium foil, nickel foil, and graphite paper. The selection of current collectors and electroactive materials is crucial to achieving advanced self-supported, high-performance supercapacitor devices.

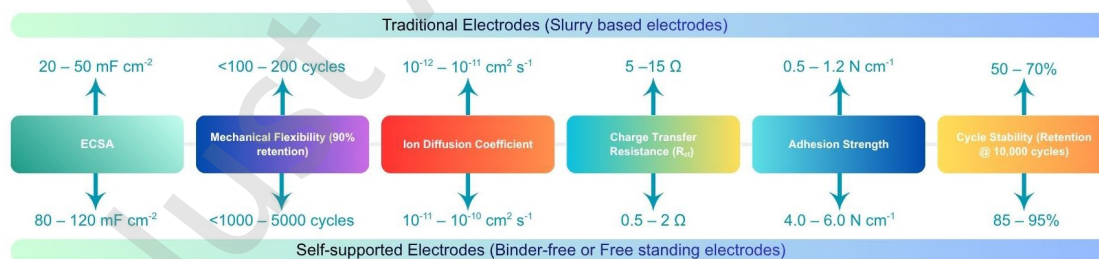


Figure 4 Performance comparison between traditional and self-supported electrodes.

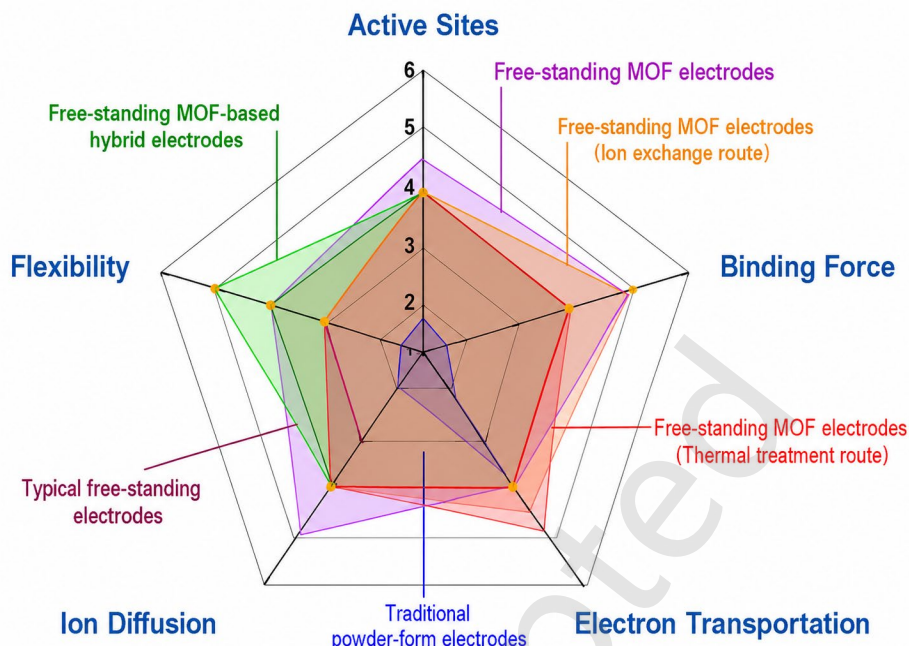


Figure 5 Feature comparison of traditional powder-form electrodes and typical self-supported electrodes in terms of active sites, binding force, electron transport, ion diffusion, and flexibility. Adopted under the CC BY 4.0 license from Ref. [60] © 2022, American Chemical Society.

5 Role of current collectors in self-supported electrodes

Current collectors are vital components of self-supported electrodes, serving not only as current-conducting substrates but also as a matrix for the growth of electroactive materials. In conventional slurry-coated electrodes, polymer binders and conductive carbon additives are introduced in addition to electroactive materials. Still, self-supported electrodes rely on the direct integration of active materials with current collectors. The self-supported electrode configuration meaningfully reduces charge-transfer resistance, improves ion-transfer kinetics, and enhances the utilization of electrochemically active sites without compromising mechanical integrity [15]. The suitable current collector should possess high electrical conductivity, low contact resistance, excellent corrosion resistance, mechanical robustness, thermal stability, and strong adhesion with the electroactive material.

Various current collectors, such as nickel foam, copper foam, carbon cloth, carbon paper, graphite foil, stainless steel mesh, and titanium mesh, are used to construct supercapacitor devices [67]. Transition metal oxides, hydroxides, sulfides, phosphides, and selenides have been successfully developed and have shown excellent electrochemical performance. Among them, nickel foam is widely used as a substrate for self-supported electrode fabrication owing to its outstanding electrical conductivity, high mechanical strength, large surface area, and 3D porous network, which promote efficient electrolyte penetration and shorten ion diffusion pathways, thereby improving specific capacitance and rate performance [68]. However,

nickel foam is relatively heavy and prone to corrosion under highly acidic or alkaline conditions, which affects long-term stability.

Carbon-based substrates such as carbon cloth and carbon paper have emerged as promising flexible current collectors due to their lightweight nature, excellent flexibility, chemical stability, and exceptional electrical conductivity [69,70]. The exceptional flexibility and flat, uniform surface could be well-suited to the uniform development of electroactive materials. The chemical stability and excellent interfacial contact between electroactive materials and current collectors resulted in greater cyclic stability during long-term charging and discharging. Nevertheless, the brittle nature of carbon paper limits its application in deformable or wearable devices. Another carbon-based substrate, namely graphite foil, and graphene-based substrates also offer excellent electrical conductivity, good corrosion resistance, low density, and chemically stable surfaces that minimize undesirable reactions and improve cycling stability [63]. However, the smooth surface and low surface roughness often result in weak adhesion of active materials, which affects their stability in long-term applications.

Instead, metal mesh substrates, such as stainless steel and titanium meshes, are widely investigated as current collectors [66,71]. Stainless steel mesh provides excellent mechanical strength, low cost, and ease of fabrication. Still, it has been shown to exhibit increased undesired Faradaic reactions and corrosion at higher operating potentials, resulting in faster self-discharge and reduced long-term stability compared with titanium-, nickel-, or graphite-based current collectors [66,71]. In contrast, titanium mesh exhibits outstanding corrosion resistance, excellent electrochemical stability, good mechanical strength, and is suited for wide-potential-window supercapacitors. Unfortunately, the higher cost and lower electrical conductivity relative to nickel-based substrates limit the use of titanium mesh.

Table 1 Comparative overview of commonly used current collectors for self-supported supercapacitor electrodes.

Current collector	Advantages	Limitations	Typical application
Nickel foam	High conductivity, porous 3D structure, high active material loading	Heavy; limited acid resistance	High-capacitance self-supported electrodes
Carbon cloth	Lightweight, flexible, chemically stable	Moderate surface area and loading	Flexible and wearable supercapacitors
Conductive textiles	Stretchable, lightweight, wearable	Lower conductivity, durability concerns	Smart textiles and wearable devices
Titanium mesh	Excellent corrosion resistance, high stability	Expensive; lower conductivity than nickel foam	Corrosive electrolytes, high-voltage systems

Graphite/Graphene	Lightweight, conductive, chemically stable	Brittle (graphite); costly (graphene)	Lightweight and flexible supercapacitors
MXene	Metallic conductivity, fast ion transport	Oxidation, restacking, expensive	Next-generation high- power supercapacitors

Recently, MXene films, graphene foams, carbon aerogels, and conductive textiles have gained extensive attention as next-generation current collectors for high-performance supercapacitors, due to their ultrahigh electrical conductivity, lightweight, excellent flexibility, and porous nature [71]. The unique characteristics of the aforementioned current collectors significantly promote rapid electron transport and electrolyte diffusion, thereby enhancing electrochemical performance. Despite their outstanding electrochemical performance, high cost, challenges in large-scale fabrication, and limited mechanical durability and redox stability limit their real-time applications. The discussion makes it clear that the choice of current collector primarily depends on the end application rather than on the electrical conductivity of current collectors. The comparative overview of widely used current collectors for self-supported supercapacitor electrodes is presented in Table 1. In conclusion, rational selection of current collectors based on conductivity, porosity, mechanical strength, corrosion resistance, and compatibility with electroactive materials is critical for achieving high-performance self-supported supercapacitor electrodes.

6 Influence of current collector properties on in-situ growth of metal selenides

The electrochemical performance of self-supported metal selenide electrodes depends not only on the intrinsic properties of the electroactive material but also on the physicochemical characteristics of the current collector during materials synthesis. In self-supported electrodes, the electroactive materials are developed directly on current collectors that regulate heterogeneous nucleation, adhesion, ion transport, and structural stability. Therefore, the surface roughness, electrical conductivity, microstructure, and corrosion resistance of the current collector confirm the mechanistic connection between synthesis route and electrochemical performance of the supercapacitor device. The nickel foam is a widely used current collector in supercapacitors due to its rough microstructure; the porous surface provides ample nucleation sites that promote heterogeneous nucleation, and interconnected macropores offer precursor diffusion during electrode material synthesis, resulting in uniform hierarchical nanosheets, nanowires, and nanoarrays. For example, Kareem et al. [72] synthesized trimetallic FeCoNiSe₂ nanosheets directly on nickel foam via a hydrothermal process and demonstrated that the substrate served as the nickel source and conductive framework. The developed nanosheet electrode exhibits excellent mechanical adhesion, thereby reducing the overall water-splitting voltage and demonstrating notable operational stability over 40 h. Similarly, Lee et al. [73] reported the development of Ni_{0.95}Se nanoassemblies on nickel foam, which served as both the nickel source and current collector. The developed self-supported electrode exhibits

excellent electrocatalytic performance for overall water splitting, with notable structural stability after 24 h of electrolysis, demonstrating that strong adhesion between the substrate and the active material significantly reduces interfacial resistance and enhances long-term stability.

In contrast, carbon cloth can be utilized for flexible supercapacitors due to its excellent flexibility, low weight, and high chemical stability. However, carbon cloth offers fewer nucleation sites than nickel foam due to its woven two-dimensional fibrous architecture. In addition, carbon cloth requires surface activation treatment to enhance wettability and nucleation sites. Ghosh et al. [74] synthesized hexagonal NiSe₂ nanoflakes on carbon cloth via a hydrothermal route, which demonstrated excellent electrochemical performance. Notably, mass loading of electroactive materials on the carbon cloth is lower than that of porous nickel foam, demonstrating that substrate architecture significantly influences crystal growth and electrochemical utilization. In another study, Hu et al. [75] systematically compared the electrochemical performance of nickel foam, nickel mesh, copper foam, and stainless steel mesh [10.1039/C9RA07258F], and found that nickel foam exhibited superior electrocatalytic performance owing to its porous 3D architecture.

Titanium mesh is known for its excellent corrosion resistance owing to its stable TiO₂ passivation layer, thereby maintaining structural stability during hydrothermal selenization [76]. However, its relatively low electrical conductivity and higher cost compared with nickel foam limit its use in high-power supercapacitors. Overall, the current collector not only directly influences the charge storage of active materials but also meaningfully affects selenide nucleation, morphological changes, interfacial charge-transfer resistance, and cycling stability in self-supported metal selenide electrodes. Therefore, the rational design of a substrate with adequate surface roughness, high conductivity, and corrosion resistance offers an effective approach to achieving self-supported metal selenides for next-generation high-performance supercapacitors.

7 Synthetic strategies for self-supported metal selenide electrodes

The electrochemical performance of supercapacitors primarily depends on the structural engineering of electrode surfaces, as morphology critically influences charge storage capacity and charge transport kinetics. The optimized morphology with controlled porosity promotes effective ion diffusion, increases the number of accessible active sites, and enhances electronic conduction during charge–discharge cycles. Various synthesis methods have been developed to achieve the required structural engineering to improve capacitive behavior and cyclic stability [77]. Diverse synthesis routes adjust physical properties, enhance electrical contact between current collectors and electroactive materials, and tune the pore size, thereby inducing electron transport, ion diffusion, and interfacial charge-transfer kinetics. Therefore, synthesis methods play a vital role in the electrochemical performance of self-supported metal selenide electrodes.

7.1 Hydrothermal and solvothermal methods

The hydrothermal/solvothermal route is an easy and effective method for preparing highly crystalline metal chalcogenides and transition metal oxide/sulfide-based materials [78,79]. Hydrothermal and solvothermal routes enable the in-situ growth of nanoarchitectural metal selenides on current collectors. For instance, Ni-Co-Se (NCSe) and Fe-Co-Se (FCSe) hierarchical nanowires, developed on carbon fiber composites via the hydrothermal method (Fig. 6a), serve as freestanding anodes for SIBs [62]. The superior electrochemical performance observed for the developed electrodes can be attributed to their engineered nanowire architecture integrated with carbon cloth. The carbon cloth not only serves as an efficient current collector that facilitates rapid electron transport but also exhibits robust mechanical stability. Recently, Che et al. developed Ni–Cu–Fe trimetallic selenide (FeCuNi-Se) nanosheets on nickel foam (FeCuNi-Se/NF) using a two-step hydrothermal and electrodeposition approach, as shown in Fig. 6b [80]. Firstly, CuNi-LDH nanoflowers with smooth nanosheets are hydrothermally grown on the nickel foam, forming a densely stacked flower-like morphology. Subsequently, Fe is incorporated via electrodeposition, resulting in FeCuNi-LDH with tightly packed nanosheets. Later, FeCuNi-Se nanosheets with a 3D nanoflower structure were developed via selenization of FeCuNi using selenium powder in the presence of NaBH₄. The FeCuNi-Se/NF hierarchical architecture offers abundant electrochemically accessible active sites and robust inter-nanosheet contact, enabling efficient electron/ion transport pathways and accelerating Faradaic reaction kinetics. Hydrothermally ternary (Ni, Co, Cu)Se₂ nanosheet arrays on CC (Fig. 6c) were developed for the detection of glucose [81]. The (Ni, Co, Cu)Se₂ ternary nanosheet arrays exhibit outstanding glucose sensing, with superior sensitivity and a low detection limit.

Instead of hydrothermal processing, as depicted in Fig. 6d, cerium-doped WSe₂ (Ce-WSe₂) was developed on nickel foam by the solvothermal method, exhibiting enhanced electrical conductivity [82]. Evidently, cerium incorporation significantly alters the morphology and microstructure of WSe₂, resulting in a hierarchical flower-like architecture, and the Ce-WSe₂/NF electrode shows superior HER performance, achieving a low overpotential of 82 mV at 10 mA cm⁻² along with a reduced Tafel slope of 88.2 mV dec⁻¹. Recently, Ma et al. synthesized hierarchical NiSe/Ni₃Se₂ on Ni foam with various Se concentrations (1, 2, 4, or 8 M) by a solvothermal approach at different temperatures (100, 140, 180, or 220 °C) for 6 h and showed a higher catalytic performance towards Cr(VI) reduction through FA (Fig. 6e) [83]. Bai et al. [61] studied Se defect VSe₂ nanosheets developed on a stainless steel battery (VSe_{2-x}-SS) serving as electrode material for the Zn//VSe_{2-x}-SS battery by solvo/hydrothermal method (Fig. 6f), and results show that the battery VSe_{2-x}-SS outperformed with a specific capacity of 265.2 mAh g⁻¹ (0.2 A g⁻¹ after 150 cycles) than Zn//VSe₂-SS. The studies indicate that synthesis parameters affect the electrochemical performance of electrode materials. Overall, the hydro/solvothermal method yields hierarchical architectures with abundant electroactive sites and excellent contact with the current collector, thereby reducing charge-transfer resistance (R_{ct}), enhancing ion-electron transport pathways, and accelerating charge-transfer kinetics.

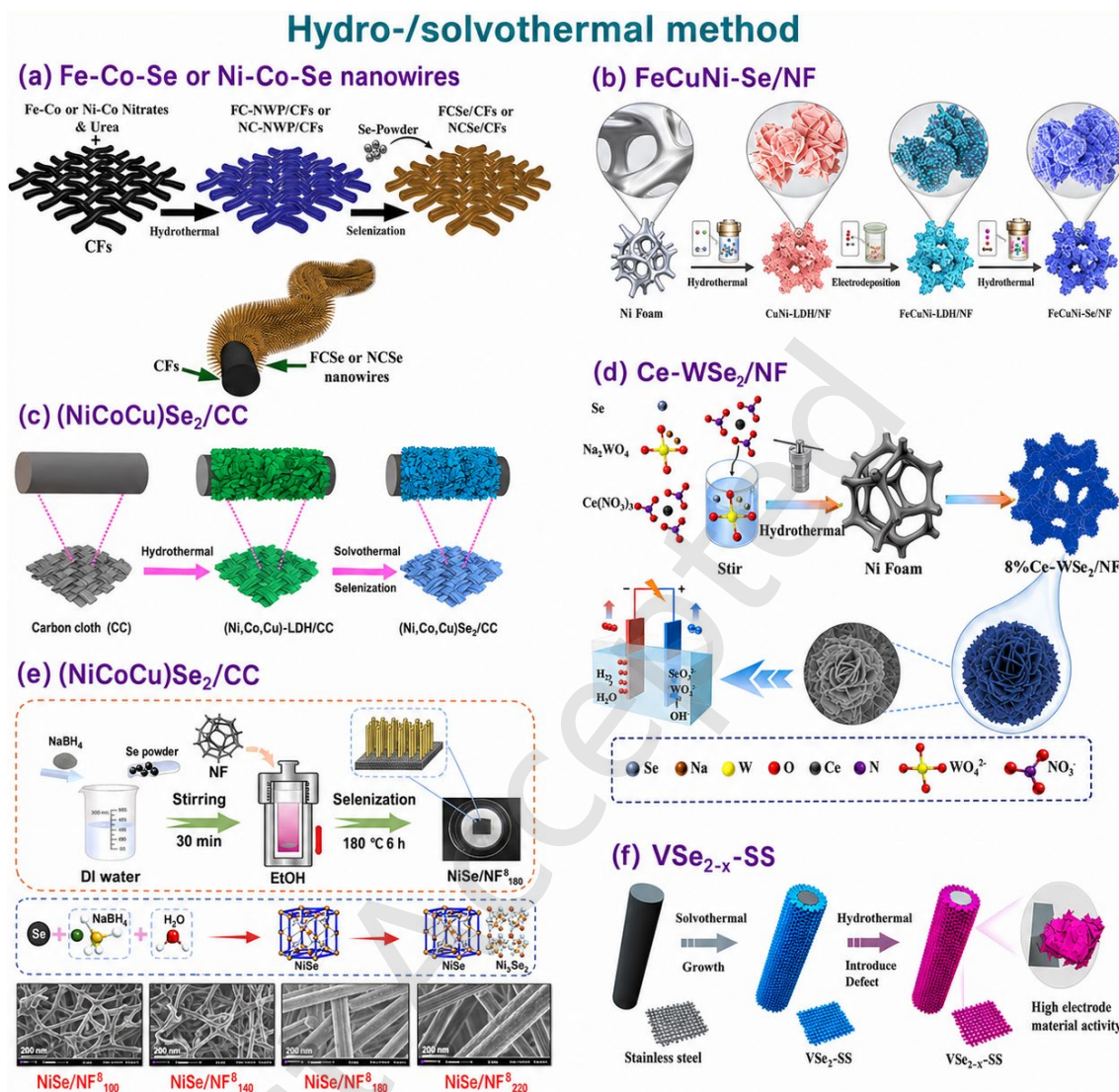


Figure 6 (a) Schematic illustration of the synthesis of FCSe/CFs and NCSe/CFs composites. Reproduced with permission from Ref. [62] © 2022, Elsevier Ltd. (b) Schematic representation of preparation of FeCuNi-Se/NF electrocatalytic materials. Reproduced with permission from Ref. [80] © 2025, Elsevier Ltd. (c) Schematic diagram of the synthesis process of (Ni, Co, Cu)Se₂/CC. Reproduced under the BB CY license from Ref. [81] © 2023, MDPI. (d) Graphical illustration of synthesis of 8 %Ce-WSe₂/NF flower-like nanoflakes. Reproduced with permission from Ref. [82] © 2025, Elsevier Ltd. (e) Illustration of *in situ* hydrothermal synthesis of NiSe/Ni₃Se₂ nanowire films on nickel foam. Reproduced with permission from Ref. [83] © 2025, Elsevier Ltd. (f) Synthesis process of VSe_{2-x}-SS. Reproduced with permission from Ref. [61] © 2021, American Chemical Society.

7.2. Electrodeposition method

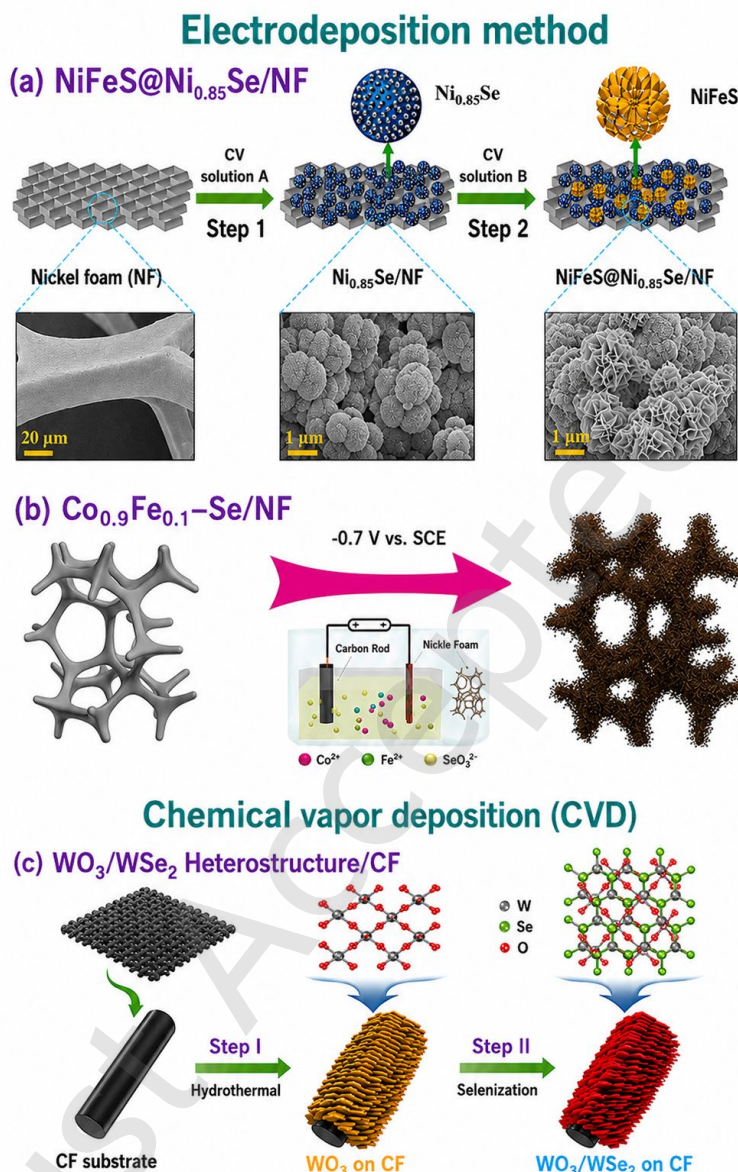


Figure 7 (a) Schematic representation of NiFeS@Ni_{0.85}Se/NF. Reproduced with permission from Ref. [84] © 2025, Elsevier Ltd. (b) Synthesis of Co_{0.9}Fe_{0.1}-Se/NF via electrochemical deposition. (c) Synthesis route of the WO₃/WSe₂ heterostructures on carbon fiber (CF). Reproduced under the CC-BY-NC-ND license from Ref. [85] © 2024, Wiley-VCH GmbH.

Electrodeposition offers precise control over loading and morphology at relatively low temperatures and is compatible with complex-shaped substrates. NiFeS nanosheets attached to Ni_{0.85}Se microspheres (NiFeS@Ni_{0.85}Se) were constructed on a nickel foam skeleton by a two-step electrodeposition method (Fig. 7a) [84]. The NiFeS@Ni_{0.85}Se/NF shows a low overpotential of 63 mV at 10 mA cm⁻² for HER and 311 mV at 100 mA cm⁻² for OER, which is due to the porous nanoflake and microsphere structures that improve the contact between NiFeS@Ni_{0.85}Se/NF and the electrolyte, provide abundant electrochemically active sites, and enhance the electron transfer rate, resulting in improved electrocatalytic reaction. Fe-

incorporated cobalt selenide on nickel foam ($\text{Co}_{0.9}\text{Fe}_{0.1}\text{-Se/NF}$) is fabricated by a simple electrodeposition method, and the device achieves a water-splitting current density of 10 mA cm^{-2} at a cell voltage as low as 1.55 V (Fig. 7b) [86]. Furthermore, it exhibits exceptional long-term stability, sustaining its activity for approximately 36 hrs without visible degradation. Electrodeposition provides precisely controlled thickness with an excellent electrode-collector interface, which significantly minimizes interfacial resistance and facilitates rapid electron transfer between the electroactive material and the current collector.

7.3 Chemical vapor deposition (CVD)

Chemical vapor deposition (CVD) is a controllable and simple method for synthesizing transition-metal selenides that controls the crystalline structure, atomic-layer thickness, microstructure, and defects. In the CVD process, transition-metal selenides can be achieved through the vapor-phase reaction of selenium with transition-metal or transition-metal oxide precursors on conducting substrates in a high-vacuum quartz tube. It is noteworthy that CVD produces uniform, layered transition-metal selenides with excellent crystallinity, which are highly suitable properties for electrochemical energy storage applications [87]. Moreover, the microstructure, number of layers, and thickness can be controlled by the precursor concentration, reaction temperature, gas flow, substrate type, and growth duration. For example, Shinde et al. [85] developed WO_3/WSe_2 heterostructured thin films on a carbon fiber substrate via CVD, as illustrated in Fig. 7c. The heterointerface between WO_3 and WSe_2 facilitates rapid ionic diffusion and efficient electron transfer. Recently, Wu et al. [88] reported the preparation of a CoSe_2 nanosheet on 3D graphene-decorated nickel foam ($\text{CoSe}_2/3\text{DG}$) using ZIF-67 nanosheets as a precursor. In the typical synthesis, firstly, graphene was deposited on nickel foam, then ZIF-67-MOF nanosheets were grown on the graphene-coated nickel foam, and finally, CoSe_2 was obtained by reacting selenium powder at 250°C under an argon atmosphere. The direct growth approach creates a porous microstructure that suppresses aggregation and enhances charge diffusion, thereby improving electrochemical performance. It is noteworthy that CVD enables high purity, controlled layer thickness, and defect engineering, which significantly enhance the electrochemical performance of transition metal selenides. In conclusion, they firmly believe that suitable synthesis methods significantly influence the structural engineering of electroactive materials, thereby enhancing the electrochemical performance of supercapacitor devices.

8 Electrochemical performances of self-supported metal selenide-based electrode materials

Transition metal selenides have attracted considerable attention as high-performance electrode materials for supercapacitors, owing to their high intrinsic electrical conductivity, tunable electronic structures, and distinct faradaic charge-storage mechanism. The crystallographic configurations, generally comprising Se–M–Se layered arrangements or interconnected three-dimensional frameworks, provide an abundant number of electroactive sites [89]. Morphological variation can be an effective means of enhancing

electrochemical performance, since one-dimensional nanowires, two-dimensional nanosheets, and hollow architectures significantly shorten ion-transport pathways and maximize the accessible surface area [90–92]. The incorporation of multiple metals within selenide lattices is considered a vital approach to improve electrochemical performance. Bimetallic electrode materials such as $\text{Ni}_{1-x}\text{Co}_x\text{Se}_2$, MnCoSe_2 , FeCoSe_2 , etc., offer heterometallic reactive centers, resulting in enhanced redox kinetics, improved electrical conductivity, and greater structural stability [93–95]. In addition, bimetallic electrode materials usually exhibit higher specific capacitance with improved cycling stability than single-metal electrodes. Moreover, integration with conductive carbon-based frameworks has been widely shown to improve the cyclic stability of metal selenide-based electrode materials. The combination of transition-metal selenides with graphene, carbon nanotubes, MXenes, and mesoporous carbons establishes efficient charge-transport pathways and conserves the structural stability of electrodes [45,96,97]. The hybrid nanostructures show high-rate capability and prolonged cycling stability, supporting their application in high-performance energy storage devices. Obviously, scientists primarily focus on designing high-performance, self-supported metal selenide-based electrodes to extend cycling stability and significantly enhance energy density.

The morphology of electroactive materials plays a vital role in charge storage mechanisms, charge transport kinetics, and the long-term stability of supercapacitors [98]. 0D nanomaterials, including nanoparticles and quantum dots, offer extremely high surface-to-volume ratios and abundant electroactive sites, enabling rapid redox reactions and high power density [99]. 1D nanostructures, including nanowires, nanotubes, and nanorods, provide constant, directional electron transport pathways with shorter ion-diffusion lengths, thereby significantly reducing internal resistance and enhancing rate capability and cycling stability, particularly in self-supported electrode configurations [100,101]. 2D materials such as graphene, MXenes, and layered transition-metal chalcogenides are thin, exhibit high electrical conductivity, and have tunable interlayer spacing, facilitating rapid ion adsorption/desorption and intercalation-mediated pseudocapacitance [102,103]. 3D nanoarchitectures, including hierarchical porous carbons and flower-like morphologies with macro-, meso-, and microporous networks that maximize electrolyte diffusion, ion transport, and mechanical robustness, thereby delivering high energy density and long-term cycling durability [104,105]. In self-supported configurations, tailoring the active material into nanoarrays significantly enhances the electrochemical performance of the supercapacitor. In contrast, nanosheet-like structures provide large electroactive sites and expose ion-transport networks [106]. Therefore, self-supported nanoarchitectures reduce charge-transfer resistance, enable high active-mass loading, and deliver exceptional rate capability and long-term cycling stability compared with conventional slurry-cast electrodes [55,107].

Obviously, researchers mainly focus on metal selenide-based self-supported electrode materials with suitable structural engineering to achieve a noteworthy extension of cyclic stability and a significant enhancement of energy density. Therefore, we emphasize that various self-supported metal selenide-based electrode materials will be discussed in the following section for high-performance supercapacitors.

8.1 Zero-Dimensional Architectures (0D)

Typically, 0D materials with <100 nm size such as nanoparticles, nanospheres, quantum dots, and carbon nanodots, have been utilized as electrode materials in supercapacitor applications due to their unique advantages in supercapacitor applications due to their size, high surface-to-volume ratio, reduced volume expansion, enhanced solid-liquid interface, reduced ion diffusion pathway, and ample active edge sites, resulting in higher charge storage capability [108]. However, 0D nanostructures also face some challenges, including agglomeration, inadequate porosity, and high surface energy, which significantly reduce the electrochemical performance of supercapacitors [27]. Self-supported nanoparticle electrodes have emerged as an effective strategy for supercapacitor applications, as the direct growth maximizes utilization of active sites, thereby enabling rapid charge-transfer kinetics and improving electrochemical stability [102]. For instance, NiFe₂Se₄ (NFSe) nanostructures were developed on nickel foam by a one-step electrodeposition route with a potential range between -1.6 to 0 V at different scan rates (5, 20, and 50 mV s⁻¹) [109]. The formation of NiFe₂Se₄, NiSe, and FeSe₂ phases in the XRD patterns observed in Fig. 8a. An optimized scan rate of 20 mV s⁻¹, NFSe show the interconnected nanoparticles with high BET surface area (40.5 m² g⁻¹) and pore volume (0.15 cm³ g⁻¹) compared to pristine NiSe (19.1 m² g⁻¹ and 0.09 cm³ g⁻¹) and FeSe₂ (9.2 m² g⁻¹ and 0.04 cm³ g⁻¹) electrodes. Scanning electron microscope (SEM) (Fig. 8b) analysis shows that the NFSe morphology forms a continuous network with nanosized particles that reduce inactive sites and offer abundant electroactive sites for the redox reaction. Among them, as shown in Fig. 8c, the NFSe-20 electrode exhibits distinct redox peaks with a large CV area. It delivers an ultrahigh specific capacity of 372.2 mAh g⁻¹ at 1 A g⁻¹, with extraordinary cyclic stability, retaining 98.3% capacity after 5000 cycles, revealing rapid ion/electron transport kinetics and long-term stability of the NFSe-20 electrode. In addition, NFSe-20 show low R_{ct} value of 0.51 Ω m², which lower than NiSe (0.76 Ω m²), FeSe₂ (0.91 Ω m²), NFSe-5 (0.63 Ω m²), and NFSe-50 (0.60 Ω m²) (Fig. 8d). The assembled NFSe-20//AC asymmetric device delivers a high energy density of 45.6 Wh kg⁻¹ at power density of 800 W kg⁻¹ and device exhibit outstanding long-term cycling stability with 101.4% retention after 10000 cycles (Fig. 8e). Fig. 8f show the practical feasibility, which is demonstrated by powering an LED for prolonged time period. The improved electrochemical performance of self-supported NiFe₂Se₄ is due to synergistic bimetallic redox activity, a conductive nickel foam skeleton, a hierarchical porous structure, and interconnected nanostructured architectures, which together facilitate abundant electroactive sites, reduced ion-diffusion pathways, and an excellent electrode-electrolyte interface.

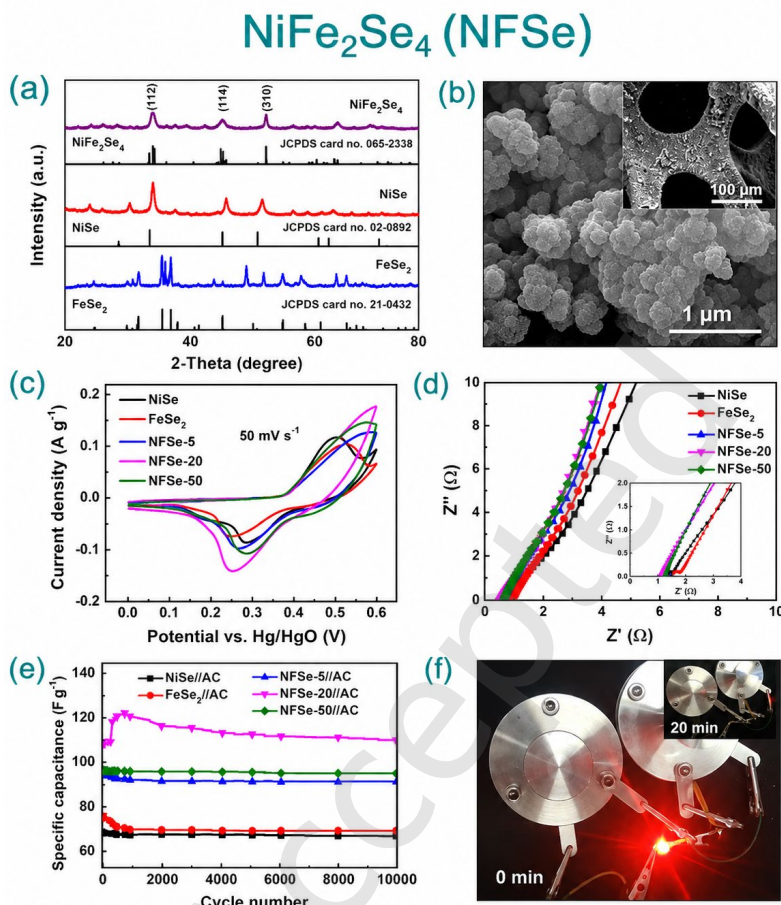


Figure 8 (a) XRD patterns of NiFe₂Se₄ (NFSe-20), NiSe, and FeSe₂. (b) SEM image of NFSe-20. (c) Electrochemical performances of NiSe, FeSe₂, NFSe-5, NFSe-20, and NFSe-50 electrodes. (d) Nyquist plots of NiSe, FeSe₂, NFSe-5, NFSe-20, and NFSe-50 electrodes. (e) Cyclic stability of NiSe//AC, FeSe₂//AC, NFSe-5//AC, NFSe-20//AC, and NFSe-50//AC asymmetric supercapacitors (ASCs). (f) Photographs of a red LED powered by two tandem NFSe-20//AC ASCs (inset displays the lighted LED lasting for 20 min). Reproduced with permission from Ref. [109] @ 2020, American Chemical Society.

8.2 One-Dimensional Architectures (1D)

1D nanostructures such as nanowires, nanorods, and nanoneedles provide continuous pathways for electron transport and reduced charge-transfer resistance, making them as potential electrode materials for charge-storage applications [110]. In addition, their high porosity and voids reduce volume changes during charging and discharging, thereby promoting ion transport in a supercapacitor. Nevertheless, the low packing density, limited lateral surface area, and bundled or intertwined structures restrict electrolyte penetration and the availability of electrochemically active sites, thereby limiting the electrochemical performance of the supercapacitors [111]. 1D metal selenide nanowires have gained prominence as potential supercapacitor electrodes owing to their high intrinsic conductivity and favorable redox activity compared to oxide and hydroxide-based materials [112]. The comparatively low electronegativity of

selenium enhances charge delocalization, while the nanowire morphology provides continuous electron-transport pathways and short ion-diffusion lengths [113]. Moreover, Interconnected nanowire networks facilitate electrolyte diffusion and maximize exposure of electroactive sites, leading to improved pseudocapacitive performance. The 1D framework efficiently buffers volume changes during continuous reversible redox reactions, thus enhancing structural stability [114]. For instance, $\text{Ni}_{4.5}\text{Co}_{4.5}\text{-Se}$ nanowires were developed on nickel-plated cotton cloth (NPCC) by hydrothermal growth of Ni–Co hydroxycarbonate arrays and subsequent ion-exchange with various Ni: Co precursor ratios (Ni: Co = 0:9, 1.5:7.5, 3:6, 4.5:4.5, 6:3, 7.5:1.5, and 9:0) (Fig. 9a) [115]. Fig. 9b-c reveal typical battery-type Faradaic behavior of Ni–Co selenide nanowires, with distinct redox peaks due to the reversible $\text{Ni}^{2+}/\text{Ni}^{3+}$ and $\text{Co}^{2+}/\text{Co}^{3+}$ and non-linear GCD profiles highlighting clear voltage plateaus. Notably, $\text{Ni}_{4.5}\text{Co}_{4.5}\text{-Se/NPCC}$ nanowires exhibited a higher CV integral area and a longer discharge time than other electrode materials. The $\text{Ni}_{4.5}\text{Co}_{4.5}\text{-Se/NPCC}$ nanowires present a high specific capacity of 1333 C g^{-1} at 1.2 A g^{-1} with a remarkable rate capability of 60% capacity retention at 6 A g^{-1} , indicating favorable redox kinetics and ion diffusion, and show outstanding cyclic stability with 120% retention after 10000 cycles. As illustrated in Fig. 9d, the battery-supercapacitor hybrid supercapacitor was assembled with $\text{Ni}_{4.5}\text{Co}_{4.5}\text{-Se}$ nanowires and Fe_3C as cathode and anode, respectively, and the assembled device achieved a high energy density of 47.4 Wh kg^{-1} at 1.5 kW kg^{-1} . In particular, Fig. 9e demonstrates excellent cyclic stability with 80% capacitance retention after 4000 cycles, highlighting the suitability of Ni–Co selenide nanowire architectures for flexible and wearable energy storage systems. The practical applications of the assembled device were studied with LED and small fan (Fig. 9f). To further explain the role of nanowires in enhancing redox kinetics and charge transport, highly conductive NiSe nanowires were solvothermally grown as vertically, followed by hydrothermal nucleation of $\text{Co}_2(\text{CO}_3)(\text{OH})_2$ at different reaction time (NSCCH/NF-1h, NSCCH/NF-4h and NSCCH/NF-7h) resulting 3D column–mesh framework [116]. Among them, NSCCH/NF-7h exhibited a large CV integral area and enhanced pseudocapacitive behavior, delivering a high areal capacitance of 9.56 F cm^2 at 4 mA cm^{-2} and remarkable rate capability, with 68.1% retention at 80 mA cm^{-2} . The assembled asymmetric ($\text{NiSe@Co}_2(\text{CO}_3)(\text{OH})_2//\text{AC}$) device delivers an energy density of 0.89 mWh cm^{-2} at 3.2 mW cm^{-2} . The enhanced electrochemical performance of NSCCH/NF-4h originates from a synergistic 1D NiSe/ $\text{Co}_2(\text{CO}_3)(\text{OH})_2$ hetero-nanowire architecture. The direct contact between NiSe nanowires and nickel foam confirms fast electron transport, whereas horizontally anchored $\text{Co}_2(\text{CO}_3)(\text{OH})_2$ nanowires increase accessible redox sites and suppress agglomeration.

In addition to nanowires, metal selenide nanorods have attracted significant attention as supercapacitor electrodes due to their anisotropic geometry, which provides continuous electron-transport pathways, short ion-diffusion lengths, and high utilization of redox-active sites [97]. The hierarchical NiCo LDH@NiSe/NF electrode was grown by in situ hydrothermal selenization of nickel foam to produce vertically aligned NiSe nanorods with a diameter of 50 nm and a length of 3 μm , followed by electrodeposition of ultrathin NiCo LDH nanosheets (Fig. 9g) [117]. The CV (Fig. 9h) of NiCo

LDH@NiSe/NF shows broader redox peaks and a higher current response intensity than those of NiCo LDH and NiSe. In addition, the GCD (Fig. 9i) reveals that NiCo LDH@NiSe/NF exhibited a much longer discharge time (815 s), confirming a higher specific capacitance than those of the other samples. Owing to its unique architecture, the NiCo LDH@NiSe/NF exhibited an ultrahigh areal capacity of 1131 $\mu\text{Ah cm}^{-2}$ at 5 mA cm^{-2} , which is higher than those of NiSe/NF (522 $\mu\text{Ah cm}^{-2}$) and NiCo LDH/NF (189 $\mu\text{Ah cm}^{-2}$). In addition, NiCo LDH@NiSe/NF electrode showed excellent rate capability (72% retention from 5 to 50 mA cm^{-2}). The assembled NiCo LDH@NiSe/NF//AC asymmetric device delivers a high areal energy of 0.454 mWh cm^{-2} at a power density of 80 mW cm^{-2} with excellent long-term cycling stability with 91% of capacity retention after 2000 cycles (Fig. 9j). The exceptional electrical conductivity (2630 S cm^{-1} for NiSe/NF) and the nanoscale LDH covering enhance Faradaic activity, resulting in improved electrochemical performance.

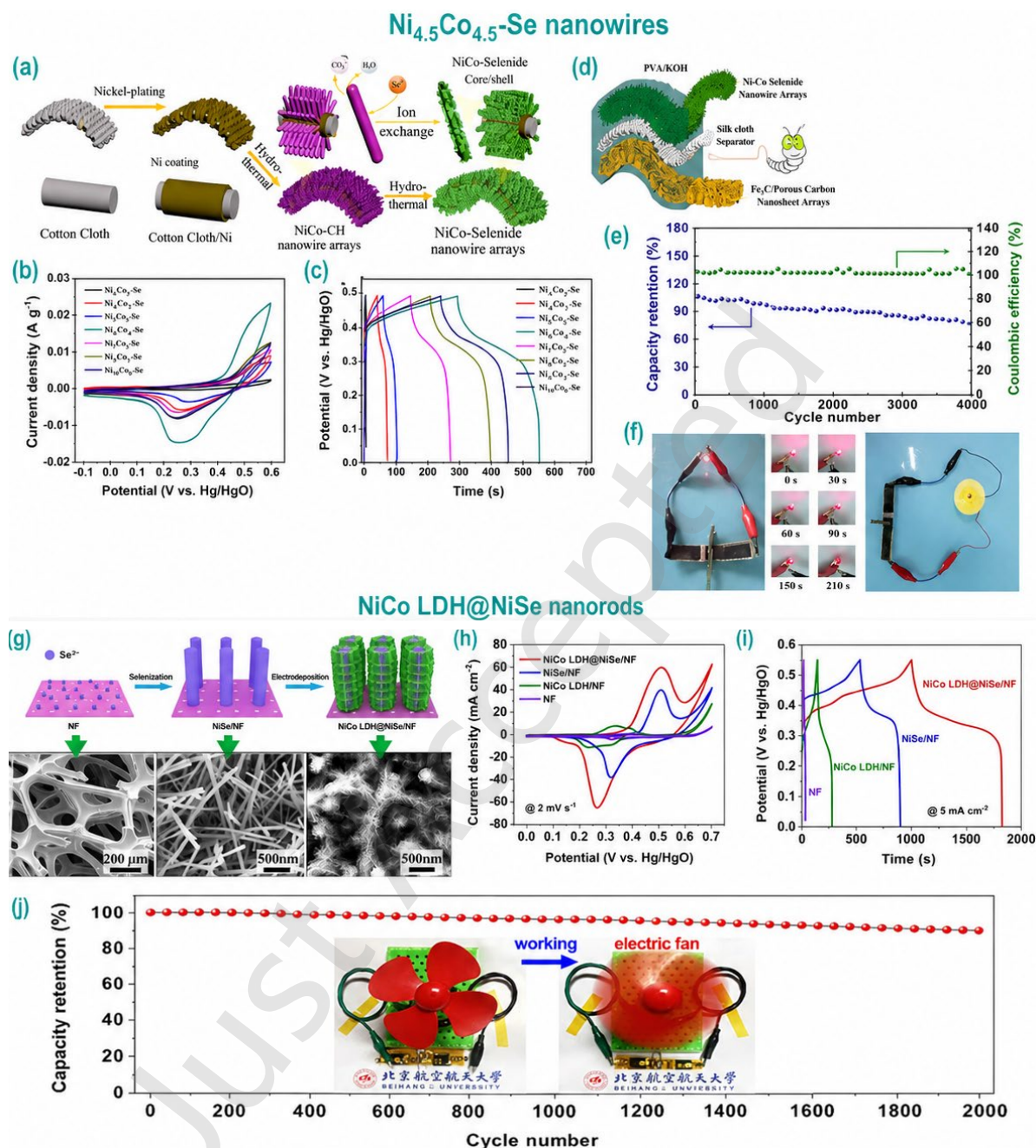


Figure 9 (a) The schematic representation of the synthesis process of Ni-Co selenide nanowires/NPCC. (b) CV curves of Ni-Co selenide nanowires/NPCC electrodes with different molar mass ratios of $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ and $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$. (c) GCD curves of Ni-Co selenide nanowires/NPCC electrodes with different molar mass ratios of $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ and $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$. (d) Schematic diagram of the $\text{Ni}_{4.5}\text{Co}_{4.5}\text{-Se}$ nanowires/NPCC//PVA/KOH// Fe_3C /CF BSH device. (e) The cyclic stability of the BSH device. (f) The BSH drives LED lights and a small yellow fan. Reproduced under CC-BY-NC-ND license from Ref. [115] © 2020, Elsevier Ltd. 2020. (g) Schematic illustration of the preparation of NiCo LDH@NiSe on nickel foam and SEM images of the Ni foam, NiSe/NF, and NiCo LDH@NiSe/NF. (h, i) Comparative CV curves

and GCD curves of the NiCo LDH/NF, NiSe/NF, and NiCo LDH@NiSe/NF. (j) Cycling stability of the NiCo LDH@NiSe//AC hybrid supercapacitor (HSC) device at 20 mA cm^{-2} . Reproduced with permission from Ref. [117] © 2020, Wiley-VCH GmbH.

The engineering of electrode materials significantly influences the electrochemical performance of the supercapacitor. In particular, nanoneedle arrays showed improved electrochemical performance due to their large surface area and greater number of electroactive sites, resulting in high specific capacitance and cycling stability [118]. The hierarchical CuSe₂ nanoneedles were developed on Cu foil using 1 M sodium selenite as a source of copper and selenium [119]. The dense, vertically aligned nanoneedle arrays with a hierarchical porous network deliver a remarkable specific capacitance of 1037.5 F g^{-1} at 0.25 mA cm^{-2} . The CuSe₂/Cu electrode also exhibits outstanding capacitance retention of 118% after 1000 cycles at 100 mV s^{-1} , indicating improved electrolyte penetration and cyclic stability. Notably, the CuSe₂/Cu nanoneedle electrode delivers an energy density of 51.85 Wh kg^{-1} at a power density of 250 W kg^{-1} . The self-supported electrodes, including Ni_{4.5}Co_{4.5}Se nanowires, hierarchical NiSe@Co₂(CO₃)(OH)₂ nanowire arrays, and CuSe₂ nanoneedles, demonstrate that 1D metal selenide nanoarchitectures, which provide high intrinsic electrical conductivity, rich electroactive sites, and robust interconnected networks, promote fast ion transport and efficient redox kinetics while maintaining structural integrity during long-term cycling, which significantly enhances the electrochemical performance [120].

8.3 Two-dimensional (2D) nanosheet architectures

2D nanostructures, including nanosheets, nanoplates, nanowalls, and nanodisks, have been widely used as electrode materials for supercapacitors due to their exceptional lateral structure and layered architecture, which provide more electroactive sites for charge storage, thereby improving electrochemical performance [27]. Nevertheless, the 2D structure of materials limits their electrochemical performance due to van der Waals restacking during charging and discharging, thereby decreasing the active surface area and constraining ion transport pathways [121,122]. Therefore, ultrathin nanosheets have been studied as an ideal electrode material for a supercapacitor due to their high surface area/plane, which shortens the ion-diffusion path and provides enormous redox-active sites, thereby allowing fast kinetics, high specific capacitance, and long-term cyclic stability [123].

8.3.1 Single-metal nanosheets

Single-metal selenide nanosheets exhibit greater electrochemical activity, attributable to their sheet-like morphology. As an example, Ni_{0.85}Se nanosheet arrays were homogeneously developed on carbon fiber cloth using Ni-carbonate hydroxide precursors by a two-step hydrothermal-selenization method [124]. The hierarchical three-dimensional network of overlying nanosheets with high surface area significantly accelerates redox kinetics, resulting in a high specific capacitance of 820 F g^{-1} at 1 A g^{-1} , excellent rate

capability with 83.17% retention from 1 to 10 A g⁻¹, and long-term cyclic stability with 82.63% capacitance retention after 10000 cycles. An assembled Ni_{0.85}Se//AC asymmetric supercapacitor with a PVA/KOH gel electrolyte displays a high energy density of 29 Wh kg⁻¹ at a power density of 779 W kg⁻¹, with notable long-term stability, achieving 81.25% capacitance retention after 5000 cycles. Xu et al. constructed hierarchical MoSe₂ nanosheets on macroporous Ti mesh to demonstrate that substrate-enabled 3D architectures effectively enhance electrolyte permeation and rate capability [125]. Hierarchical MoSe₂ nanosheets show a high specific capacitance of 525.5 F g⁻¹ at 1 A g⁻¹, with excellent rate capability, retaining 230.1 F g⁻¹ at 10 A g⁻¹. The hierarchical network architecture boosts electrolyte and ion diffusion and reduces stacking to 3–4 layers, improving conductivity and charge transfer between the electrode and the electrolyte. Likewise, the hierarchical sheet-like SnSe electrodes displayed a specific capacitance of 794 F g⁻¹ (155 mF cm⁻²) at 5 mV s⁻¹ in 1.5 M KOH and 645 F g⁻¹ in 3.0 M LiClO₄, and the electrode materials exhibit excellent long-term cyclic stability with 97% capacitance retention over 4500 cycles [126]. The assembled SnSe//AC solid-state device with PVA-based gel electrolytes demonstrates a high energy density of 8.88 Wh kg⁻¹ at a power density of 2.7 kW kg⁻¹ with insignificant charge-transfer resistance. Miao et al. successfully grew ultrathin α-MnSe-x nanosheets directly on nickel foam via a one-step hydrothermal strategy with different reaction times (x = 8, 12, 16, and 20 h) [127]. As depicted in Fig. 10a, a well-defined cubic rock-salt crystal structure with a highly open, mesoporous 3D network morphology is observed, which facilitates more electroactive sites, electrolyte diffusion, and rapid ion/electron transport. The α-MnSe-16 electrodes deliver high pseudocapacitive performance with a specific capacity of 88.3 mAh g⁻¹ at 1 A g⁻¹, which is higher than that of the α-MnSe-8, α-MnSe-12, and α-MnSe-20 samples. In addition, α-MnSe-8 samples exhibit a low R_{ct} (0.47 Ω) and impressive cycling durability, with 91% retention over 5000 cycles (Figs. 10b-10c). The assembled α-MnSe//AC asymmetric supercapacitor delivers a high energy density of 39.3 Wh kg⁻¹ at a power density of 0.92 kW kg⁻¹, with long-term stability, retaining 94% of its capacitance after 5000 cycles. The improved electrochemical performance of 2D self-supported metal selenide electrodes is attributed to the binder-free configuration and hierarchical nanoarchitectures, which significantly increase the number of electroactive sites, promote rapid electron transport, enhance electrolyte accessibility, and minimize R_{ct}. In addition, an exceptional interface between metal selenides and the current collector retained the metal selenide architecture, resulting in prolonged cycling and displaying outstanding capacitance, high-rate capability, and excellent cycling stability.

8.3.2 Binary and multi-metallic nanosheets

Single metal selenide-based electrodes have demonstrated high electrochemical activity due to their nanoscale, sheet-like structure. Several studies have shown that the binary and multi-metallic selenides offer superior electrochemical performance due to multi-metallic synergy, tunable electronic structures, and abundant redox-active sites, enabling accelerated charge-transfer kinetics and enhanced charge storage. For instance, Min and associates [128] have demonstrated that MnSe₂/Co_{0.85}Se (MCSe)

nanosheets are a promising electrode for high-performance supercapacitors. The MCSe developed at a scan rate of 15 mV s^{-1} (MCSe-15) displays high electrochemical performance, with a specific capacity of 1380 C g^{-1} at 1 A g^{-1} and robust cycling stability, with 81.1% capacity retention over 10000 cycles, indicating its potential as a battery-type cathode. The assembled MCSe-15//AC asymmetric supercapacitor in 1 M KOH delivers a high specific capacity of 238.2 C g^{-1} at 1 A g^{-1} , and the corresponding Ragone plot displays a remarkable energy density of 49.63 Wh kg^{-1} at a power density of 750 W kg^{-1} . The improved electrochemical performance is due to the synergistic effect of MnSe_2 and $\text{Co}_{0.85}\text{Se}$, which shortens the ion diffusion pathway, thereby reducing charge transfer. In 2020, Wang et al. [129] prepared porous NiCoSe_2 nanosheet arrays on nickel foam via one-step electrodeposition for high-performance asymmetric supercapacitors. Notably, NiCoSe_2 shows superior electrochemical performance, with a high specific capacity of 520 C g^{-1} at 1 A g^{-1} , a low R_{ct} (0.42Ω), excellent cyclic stability with 85% capacity retention over 5000 cycles, and strong cycling stability, due to efficient charge transfer and rapid ion transport within the nanosheet framework. The assembled asymmetric (NiCoSe_2 //AC) supercapacitor, operating within a 1.6 V window, delivers a high energy density of 44.4 Wh kg^{-1} at a power density of 776.7 W kg^{-1} and, remarkably, exhibits extraordinary cycling stability with 133% capacity retention after 10,000 cycles. The 3D porous NiCoSe_2 nanosheet array provides a large surface area, short ion diffusion paths, and superior electron-transfer properties. In parallel, Liu et al. [130] developed a NiCo-LDH -derived self-supported $(\text{Ni, Co})\text{Se}_2$ nanosheet array electrode on flexible carbon cloth (Fig. 11a). As shown in Fig. 11a, NiCo-LDH nanosheets are uniformly and vertically anchored on the carbon fibers, forming a dense and open array that enhances surface accessibility. After selenization, the $(\text{Ni, Co})\text{Se}_2$ /CC electrode retains its nanosheet architecture, which offers a larger surface area ($18.845 \text{ m}^2 \text{ g}^{-1}$), and the porous surface increases the number of electrochemically active sites and facilitates rapid ion transport. CV at 10 mV s^{-1} (Fig. 11b) and GCD at 1 A g^{-1} (Fig. 11c) of $(\text{Ni, Co})\text{Se}_2$ /CC reveal a high specific capacitance of 2054 F g^{-1} , low charge-transfer resistance of 0.6Ω (Fig. 11d), and remarkable rate stability with 90% capacitance retention from 1 to 10 A g^{-1} , confirming faster faradaic kinetics and efficient charge transport compared with NiCo-LDH and NiCo-LDH/CC . An assembled flexible $(\text{Ni, Co})\text{Se}_2$ //AC asymmetric supercapacitor with PVA/KOH gel electrolyte delivers a high energy density of 44.6 Wh kg^{-1} at 976 W kg^{-1} . Robust cycling stability with 85.2% capacitance retention after 5000 cycles is achieved under repeated bending, demonstrating great promise for high-performance, wearable energy-storage systems.

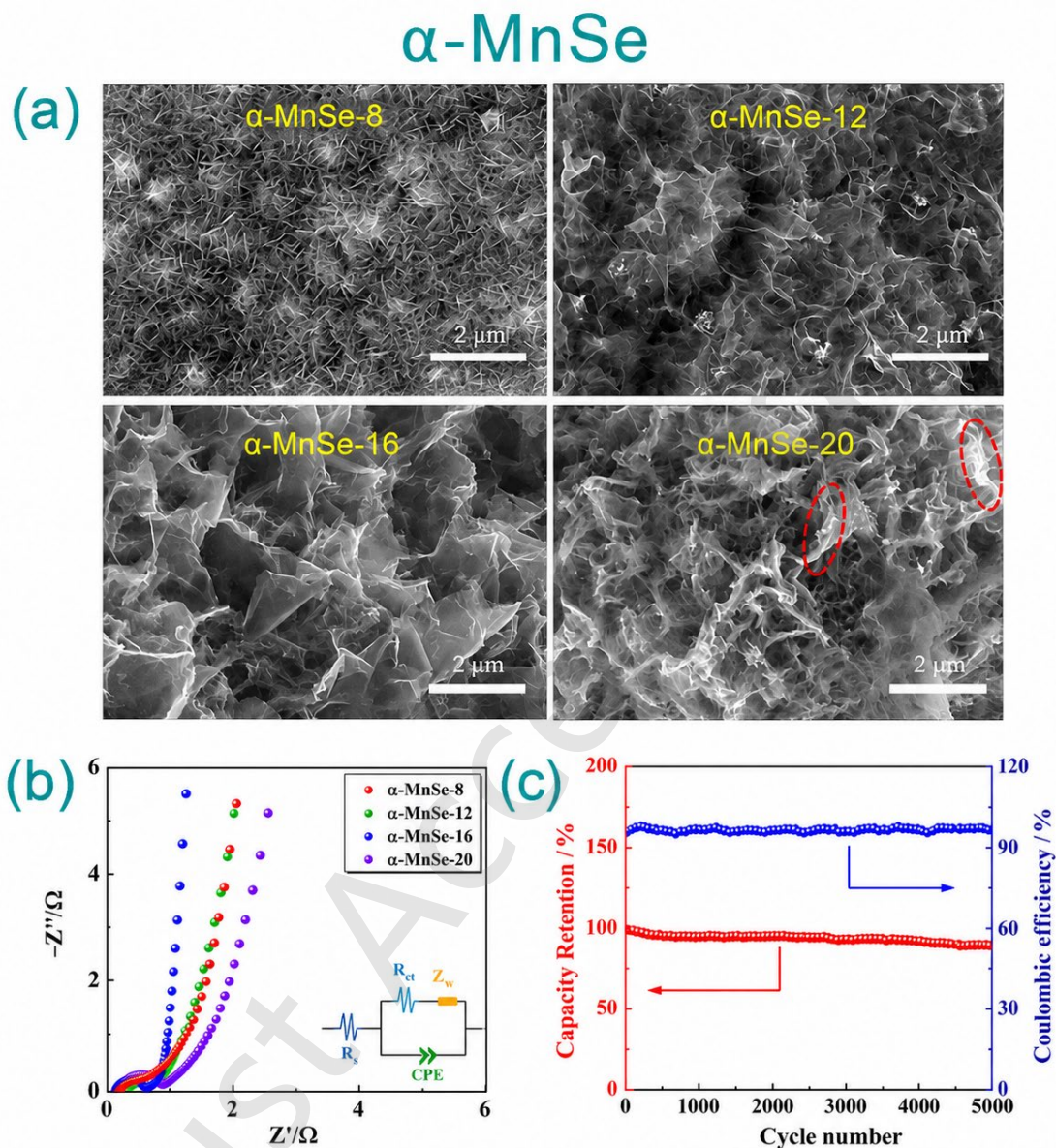


Figure 10 (a) SEM images of α -MnSe-8, α -MnSe-12, α -MnSe-16, and α -MnSe-20; (b) EIS spectra of α -MnSe samples; (c) Cycling stability of α -MnSe-16. Reproduced with permission from Ref. [127] © 2021, Elsevier Ltd.

Although binary metal selenides exhibit higher electrical conductivity and intrinsic redox activity, their practical performance is often limited due to insufficient electroactive sites and structural instability [27]. To address these challenges, Salehi Panah et al. [131] developed $\text{NiCo}_2\text{Se}_4/\text{Fe}_2\text{CoSe}_4$ (NCS/FCS) heterostructures on nickel foam by the hydrothermal method. As-prepared NiCo-LDH shows a well-defined chrysanthemum-like microflower composed of radially aligned nanosheets. After Fe incorporation, NiCo-LDH/FeCo-LDH (Fig. 11e) exhibits a broccoli-like, roughened surface with

increased surface density, indicating secondary nucleation on the LDH framework. After selenization, the NCS/FCS display a densely nanosheet-decorated 3D architecture, creating abundant mesoporous networks and exposed active sites favorable for quick ion transport and redox reactions. The CV curves of the NCS/FCS (Fig. 11f) show a larger CV-integrated area and peak current than those of NiCo-LDH@NiF and NiCo-LDH/FeCo-LDH@NiF. Moreover, the GCD (Fig. 11g) clearly demonstrates that the NCS/FCS exhibits a longer discharge time than other electrodes. As a result, the NCS/FCS electrode delivers a high specific capacity of $326.66 \text{ mAh g}^{-1}$ at 1 A g^{-1} and exhibits improved rate performance. It reveals a remarkable cyclic stability with 91% retention after 10000 cycles even at high current density of 20 A g^{-1} (Fig. 11h). Moreover, a hybrid supercapacitor (NCS/FCS//AC) assembled and display a high energy density of 51.83 Wh kg^{-1} at 749.83 W kg^{-1} , with superior long-term stability with 7.67% of capacitance loss, confirming the efficacy of rational nanosheet-assembled flower-like architectures for high-performance supercapacitor applications.

Although mixed-metal selenides exhibit excellent conductivity, their poor stability limits their real-time applications. Therefore, scientists are shifting toward mixed-metal selenides/sulfides to achieve improved stability and overall electrochemical performance. As an example, $\text{CuNi}_2\text{S}_4@\text{NiCoSe}_2$ was grown directly on carbon cloth via a two-step electrodeposition approach [132]. In the first step, CuNi_2S_4 nanospheres were electrodeposited using $\text{Cu}^{2+}/\text{Ni}^{2+}$ /thiourea electrolyte on the carbon cloth, followed by in situ growth of NiCoSe_2 from $\text{Ni}^{2+}/\text{Co}^{2+}/\text{SeO}_2$ precursors, resulting integrated 0D/2D heterostructure CuNi_2S_4 spheres/ultrathin NiCoSe_2 nanosheets (Fig. 11i). The unique morphology $\text{CuNi}_2\text{S}_4@\text{NiCoSe}_2$ electrode delivers a superior electrochemical performance with a high specific capacity of 979.2 C g^{-1} (3.43 C cm^{-2}) at 1 A g^{-1} , notable rate constant (68.1 % at 20 A g^{-1}), low R_{ct} (0.863Ω) compared to CuNi_2S_4 (1.281Ω) and NiCoSe_2 (0.897Ω) and remarkable cyclic stability with 86.5 % capacity retention after 5000 cycles. An assembled ($\text{CuNi}_2\text{S}_4@\text{NiCoSe}_2$ //PPC) asymmetric device results in a gravimetric capacity of 162.1 C g^{-1} at 1 A g^{-1} , a high energy density of 78.9 Wh kg^{-1} at 720.2 W kg^{-1} , and retains 91.4% of its capacity over 50000 cycles (Fig. 11j).

In parallel, Wan et al. [133] constructed hierarchical $\text{FeNiSe}_2@\text{Ni}_{4.5}\text{Co}_{4.5}\text{S}_8$ nanosheet arrays on carbon cloth via an electrodeposition route followed by selenization, as depicted in Fig. 11k. The results show that the metal-like FeNiSe_2 core and the electroactive $\text{Ni}_{4.5}\text{Co}_{4.5}\text{S}_8$ shell alter the electronic structure, resulting in superior electrochemical behavior. Obviously, the CV analysis of $\text{FeNiSe}_2@\text{Ni}_{4.5}\text{Co}_{4.5}\text{S}_8$ shows a larger CV area (Fig. 11l) than those of FeNiSe_2 and $\text{Ni}_{4.5}\text{Co}_{4.5}\text{S}_8$, indicating synergistic heterointerface effects that enhance multi-redox activity and accelerate charge-transfer kinetics. Besides, $\text{FeNiSe}_2@\text{Ni}_{4.5}\text{Co}_{4.5}\text{S}_8$ delivers a significantly longer and more symmetric discharge profile at 1 A g^{-1} than $\text{FeNiSe}_2@\text{Ni}_{4.5}\text{Co}_{4.5}\text{S}_8$, confirming higher reversible capacity and lower internal resistance (Fig. 11m). Notably, $\text{FeNiSe}_2@\text{Ni}_{4.5}\text{Co}_{4.5}\text{S}_8$ display a high specific capacity of 236.9 mAh g^{-1} at 1 A g^{-1} , and remarkable cyclic stability with 77.5% capacitance retention after 5000 cycles (Fig. 11n). The assembled asymmetric ($\text{FeNiSe}_2@\text{Ni}_{4.5}\text{Co}_{4.5}\text{S}_8$ //PC) device delivers a notable energy density of 69.0 Wh kg^{-1} at 799.2

W kg⁻¹. The improved electrochemical performance is mainly due to the vertically aligned 3D nanosheet framework, which exposes ample redox-active sites, and the hierarchical porosity, which facilitates rapid charge transfer at the electrode–electrolyte interface. In addition to binary metal selenides, ternary metal selenides have proven to be promising electrode materials for supercapacitor applications. To confirm, a hollow and porous trimetallic Ni-Zn-Co selenide (Ni-Zn-Co-Se) nanosheet architecture was rationally engineered on reduced graphene oxide–modified nickel foam via a stepwise in situ strategy involving the growth of Ni-Zn-Co ZIF precursors [134]. Owing to the synergistic redox activity of Ni, Zn, and Co species, the intrinsically high conductivity of metal selenides, and the conductive reduced graphene oxide–modified nickel foam substrate, the electrode exhibits a high specific capacity of 464.4 mA h g⁻¹ at 1 A g⁻¹, retains 320 mA h g⁻¹ at 20 A g⁻¹ (68.9% retention), and delivers a low charge-transfer resistance of 0.59 Ω in 2 M KOH. Moreover, the hollow nanoarchitecture showed excellent cycling stability, maintaining 93.24% of its initial capacity after 7000 cycles. The Ni-Zn-Co-Se//PC (porous carbon) asymmetric supercapacitor device achieves a high energy density of 74.6 W h kg⁻¹ at 796.9 W kg⁻¹, with a power density of 96.4% and capacitance retention over 7000 cycles, highlighting the promise of MOF-derived multi-metallic selenide nanoarrays for high-energy supercapacitor applications. The outstanding electrochemical performance of self-supported 2D metal selenide electrodes is predominantly due to the existence of bimetallic/heterostructured selenides with hierarchical architectures (porous nanosheet, core–shell, and flower-like) with a binder-free configuration provide ample redox-active sites, improved electrical conductivity, and high surface to volume ratio, effectively shorten ion diffusion pathways and maintain the structural integrity during repeated cycling, and facilitate fast electrolyte penetration. Besides, the in-situ growth on a conductive current collector eliminates polymer binders (dead mass), reduces interfacial resistance, improves electrical contact, and enhances structural stability, thereby delivering exceptional capacitance, rate capability, energy density, and long-time cycling stability. In conclusion, we firmly believe that binary and multi-metallic selenide-based electrode materials are a great strategy for enhancing the electrochemical performance of supercapacitors.

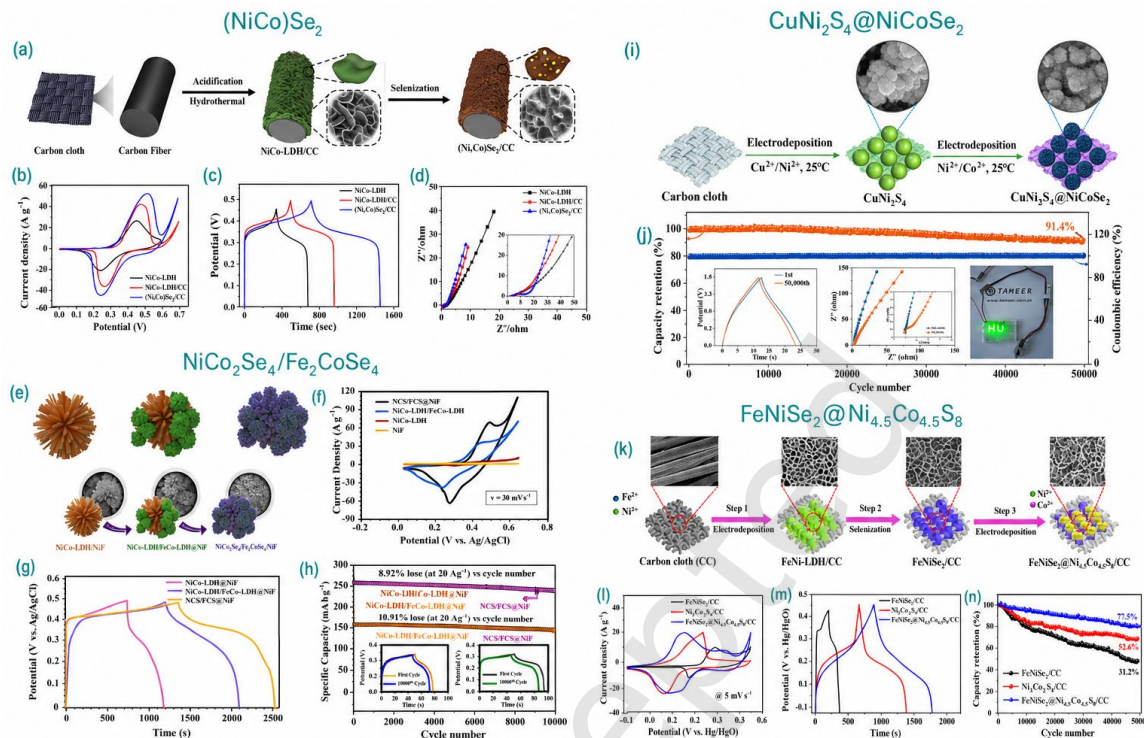
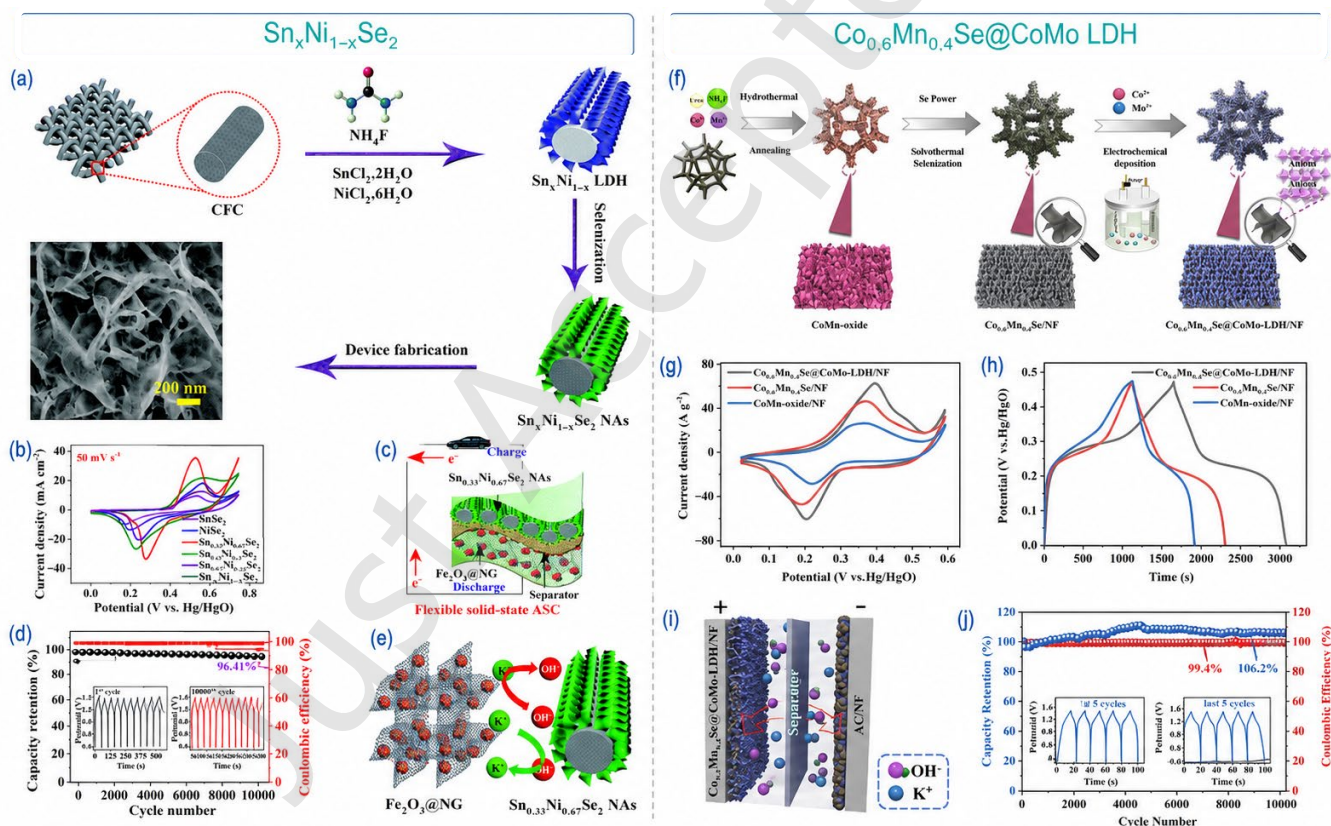


Figure 11 (a) Schematic illustration for the synthesis process of (Ni, Co)Se₂/CC nanoarrays and the assembly of (Ni, Co)Se₂/CC//AC/CC asymmetric supercapacitor. (b) CV curves (10 mV s⁻¹) and (c) GCD curves (1 A g⁻¹) of NiCo-LDH, NiCo-LDH/CC and (Ni, Co)Se₂/CC. (d) EIS plots of NiCo-LDH, NiCo-LDH/CC and (Ni, Co)Se₂/CC. Reproduced with permission from Ref. [130] © 2023, Elsevier Ltd. (e) Schematic sketch of synthesis of NCS/FCS@NiF. (f) CVs of the pure nickel foam, NiCo-LDH@NiF, NiCo-LDH/FeCo-LDH@NiF, and NCS/FCS@NiF electrodes at 30 mVs⁻¹. (g) GCD curves of the NiCo-LDH@NiF, NiCo-LDH/FeCo-LDH@NiF, and NCS/FCS@NiF electrodes at 1 A g⁻¹. (h) Cyclic stability of the NCS/FCS@NiF and NiCo-LDH/FeCo-LDH@NiF electrodes. Reproduced with permission from Ref. [131] © 2024, Elsevier Ltd. (i) Synthetic illustration of CuNi₂S₄@NiCoSe₂ heterostructure. (j) Capacity retention and coulombic efficiency of the HSC device at 20 A g⁻¹ over 50,000 cycles (inset exhibits the first and last GCD profiles, Nyquist profiles before and after cycling, and seventeen green LEDs lit by two serial devices). Reproduced with permission from Ref. [132] © 2025, Elsevier Ltd. (k) Schematic illustration for the preparation process of FeNiSe₂@Ni_{4.5}Co_{4.5}S₈ core-shell nanosheet arrays. (l) CV profiles at 5 mV s⁻¹ of FeNiSe₂/CC, Ni_{4.5}Co_{4.5}S₈/CC, and FeNiSe₂@Ni_{4.5}Co_{4.5}S₈/CC electrodes. (m) GCD profiles at a current density of 1 A g⁻¹ of FeNiSe₂/CC, Ni_{4.5}Co_{4.5}S₈/CC, and FeNiSe₂@Ni_{4.5}Co_{4.5}S₈/CC electrodes. (n) cycling performance at 5 A g⁻¹ over 5000 cycles of FeNiSe₂/CC, Ni_{4.5}Co_{4.5}S₈/CC, and FeNiSe₂@Ni_{4.5}Co_{4.5}S₈/CC electrodes. Reproduced with permission from Ref. [133] © 2022, Elsevier Ltd.

8.3.3 Metal-doped and cation-engineered metal selenide nanosheets

Metal substitution and cation-exchange approaches have demonstrated effectiveness in modifying the electronic structure, reversible redox activity, and charge-transfer kinetics of transition-metal selenides, thus overcoming the constraints of single-metal selenides and enabling superior energy-storage performance, including enhanced capacity, rate capability, and cycling stability in supercapacitors [135,136]. As an example, Deepalakshmi et al. [137] developed interconnected $\text{Sn}_x\text{Ni}_{1-x}\text{Se}_2$ nanosheet arrays on carbon cloth by a two-step in situ strategy and studied the effect of incorporation of tin in the nickel selenide skeleton (Fig. 12a). The $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$ electrode shows vertically aligned ultrathin nanosheets with a 3.3 nm diameter thickness and ample nanopores. Also, it exhibits a high specific surface area ($235.38 \text{ m}^2 \text{ g}^{-1}$). As depicted in Fig. 12b, the $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$ electrode delivers an ultrahigh specific capacity of 346 mA h g^{-1} at 1 mA cm^{-2} in a three-electrode cell configuration, with excellent rate capability (82.37% retention at 50 mA cm^{-2}). Moreover, the $\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2$ electrode demonstrates an outstanding cycling stability with a capacitance loss of 1.78% after 10000 cycles. The assembled flexible solid-state asymmetric supercapacitor ($\text{Sn}_{0.33}\text{Ni}_{0.67}\text{Se}_2/\text{Fe}_2\text{O}_3@\text{N-doped graphene}$) device (Fig. 12c) deliver an excellent energy density of 90.3 Wh kg^{-1} at 0.631 kW kg^{-1} with 96.41% capacity retention over 10000 GCD cycles (Fig. 12d). The hierarchical architectures with porous nature and 3D network effectively improve the electroactive surface area and enhance the electrical conductivity between the active materials and CFC support, which enhance the ion transport kinetics resulting in improved electrochemical performance, as illustrated in Fig. 12e. In another instance, the $\text{Co}_{0.6}\text{Mn}_{0.4}\text{Se}@\text{CoMo-LDH}$ hybrid have developed on nickel foam by three step approaches namely, hydrothermal, solvothermal selenization, and electrodeposition [138]. Fig. 12f reveals that the $\text{Co}_{0.6}\text{Mn}_{0.4}\text{Se}@\text{CoMo-LDH}$ are ultrathin, vertically oriented nanosheets on the nickel foam. As shown in Figs. 12g and 12h, the CV and GCD results indicate that the $\text{Co}_{0.6}\text{Mn}_{0.4}\text{Se}@\text{CoMo-LDH}$ electrode has a larger integral area and a longer discharge time than those of CoMn-oxide and $\text{Co}_{0.6}\text{Mn}_{0.4}\text{Se}$, confirming the high specific capacity. The $\text{Co}_{0.6}\text{Mn}_{0.4}\text{Se}@\text{CoMo-LDH}$ nanostructures display superior electrochemical performance, with a specific capacity of 1405.2 C g^{-1} at 1 A g^{-1} , excellent rate capability (87.67%), and remarkable cycling stability, with 88.2% capacity retention after 10,000 cycles. The $\text{Co}_{0.6}\text{Mn}_{0.4}\text{Se}@\text{CoMo-LDH}/\text{AC}$ hybrid supercapacitor device (Fig. 12i) presents a high energy density of 71.4 Wh kg^{-1} at 1.13 kW kg^{-1} . Furthermore, the device exhibits exceptional cyclic stability, with 106.2% capacitance retention after 1000 cycles (Fig. 12j). The improved electrochemical performance can be attributed to selenium-enhanced electrical conductivity and the rich OH diffusion channels. In contrast, the CoMo-LDH shell provides more open redox-active sites and enhances cobalt consumption through Mo^{6+} -induced electronic variation. Instead of manganese substitution, Du et al. [117] substituted cobalt for nickel and synthesized nickel-cobalt selenide ($(\text{Ni}_x\text{Co}_{1-x})_{0.85}\text{Se}$) nanosheet arrays on nickel foam via hydrothermal growth followed by a cation-exchange process. The $(\text{Ni}_x\text{Co}_{1-x})_{0.85}\text{Se}$ demonstrates honeycomb-like nanoarrays with $\sim 20 \text{ nm}$ -thick interconnected sheets. The $(\text{Ni}_x\text{Co}_{1-x})_{0.85}\text{Se}$ delivers a high specific capacity of $430.87 \text{ mAh g}^{-1}$ at 1 A g^{-1} and long-term durability with 85.25% capacity retention over 3000 cycles. The asymmetric $(\text{Ni}_x\text{Co}_{1-x})_{0.85}\text{Se}/\text{NC}$ hybrid supercapacitor achieves an excellent energy density of 70.58 Wh kg^{-1} at a power density of 320.02 W kg^{-1} , with $\sim 92\%$ capacity retention after 8000 cycles, due to the high surface area

and unique morphology of $(\text{Ni}_x\text{Co}_{1-x})_{0.85}\text{Se}$. Shankar et al. developed $\text{Co}_x\text{Fe}_{3-x}\text{Se}_4$ ultrathin nanosheet arrays on nickel foam by electrodeposition, which show a high areal capacitance of 3393 mF cm^{-2} at 3 mA cm^{-2} and remarkable long-term stability, with 96.4% capacitance retention after 10000 cycles [139]. The quasi-solid-state $\text{Co}_2\text{FeSe}_4//\text{AC}$ asymmetric supercapacitor device with PVA-KOH gel electrolyte achieves an energy density of $157.3 \text{ mWh cm}^{-2}$ and a power density of 2250 mW cm^{-2} , and exceptional cyclic stability over 10,000 cycles. The uniformly aligned 2D Co_2FeSe_4 nanosheet arrays with a porous, interconnected structure provide more redox-active surface area and facilitate effective electrolyte diffusion, resulting in improved charge-transfer behavior. The above examples indicate that metal substitutions in metal selenide networks are a promising approach to enhancing the electroactive sites, achieving superior rate capability, reducing R_{ct} , and achieving exceptional cycling stability in electrode materials, thereby enabling high-performance supercapacitor devices.



CV curves for CoMn-oxide/NF, Co_{0.6}Mn_{0.4}Se/NF, Co_{0.6}Mn_{0.4}Se@CoMo-LDH/NF at 10 mV s⁻¹. (h) GCD curves for various electrodes at 1 A g⁻¹. (i) Schematic of assembled Co_{0.6}Mn_{0.4}Se@CoMo-LDH/NF//AC HSC device. (j) Cycling performance of HSC device. Reproduced with permission from Ref. [138] © Elsevier Ltd. 2023.

8.4 3D hierarchical architectures

The 3D porous electroactive nanostructured material provides a larger specific surface area, improved pathways for electrolyte diffusion, and robust mechanical support, thereby enhancing the overall performance of Supercapacitor electrodes [11]. Nevertheless, the fabrication of well-defined 3D architectures remains challenging because precise control over pore size, morphology, and hierarchical organization is required. Moreover, scalable, cost-effective synthesis that retains structural uniformity remains a major challenge for practical implementation. To demonstrate the electrochemical performance of 3D nanostructures, Bekhit and colleagues [140] developed snow-crystal-like NiSe on nickel foam, which shows battery-type faradaic behavior in an alkaline electrolyte, delivers a high specific capacity of 763.5 C g⁻¹ at 1.5 A g⁻¹, and exhibits remarkable cyclic stability with 83% capacitance retention after 3000 cycles. The assembled NiSe//AC hybrid asymmetric supercapacitor delivers a high energy density of 32.0 Wh kg⁻¹ at a power density of 1112 W kg⁻¹ and outstanding cycling stability, with 99.95% capacitance retention after 10000 cycles. The same research group electrochemically synthesized sponge-like NiSe electrodes on nickel foam, which demonstrate remarkable electrochemical performance with a high specific capacity of 751 C g⁻¹ at 1 A g⁻¹ and 80 % capacity retention after 1500 charge-discharge cycles [141]. The sponge-like NiSe//AC HSC with a wide voltage window of 1.7 V delivers a high energy density of 42.2 Wh kg⁻¹ at a power density of 958.3 W kg⁻¹, and the assembled device shows outstanding cyclic stability with 99% capacity retention after 9000 GCD cycles. The highly porous sponge-like morphology provides abundant electroactive sites and effectively shortens ion-diffusion pathways, resulting in a highly effective battery-type electrode material for high-performance hybrid supercapacitors. To understand the selenium-rich materials, Subhadarshini et al. developed selenium-rich nickel selenide (NiSe-Se) hollow nanotubes on nickel foam (Fig. 13a) and studied supercapacitive performance [142]. Obviously, the formation of tuberos-like, uniformly aligned, one-dimensional porous nanotubes on the nickel foam is observed in Fig. 13b, which minimizes interfacial resistance due to the high surface area and excellent contact with the current collector. The hollow nanotube shows a battery-type pseudocapacitive behavior. It delivers an ultrahigh specific capacitance of 2447.46 F g⁻¹ at 1 A g⁻¹, with low R_{ct} (0.0375 Ω) and the remarkable capacitance retention (76.8% after 2000 cycles) (Fig. 13c). The NiSe-Se//AC asymmetric hybrid supercapacitor reaches an energy density of 29.9 Wh kg⁻¹ at a power density of 594 W kg⁻¹ with excellent cyclic stability of 95% capacitance retention after 10000 cycles. The improved performance is due to the hollow interior and inter-nanotube voids, which provide more active sites and shorten OH⁻ diffusion pathways, resulting in a high-performance device.

As we all know, the crystalline phase of electrode materials significantly influences their overall performance. As an example, Nazari et al. [143] electrochemically synthesized Ni_xSe_y polymorph nanostructures ($0.5 \leq x/y \leq 1.5$) on nickel foam, as depicted in Fig. 13d. The polymorphs of nickel selenide, including orthorhombic NiSe_2 , trigonal Ni_3Se_2 , hexagonal NiSe , and orthorhombic Ni_6Se_5 , are obtained by varying experimental conditions. Notably, the NiSe_2 and Ni_3Se_2 polymorphs display superior electrochemical performance due to their metallic character. The $\text{NiSe}/\text{Ni}_3\text{Se}_2$ (NS) delivers a high specific capacity of $180.3 \text{ mA h g}^{-1}$ at 1 A g^{-1} along with a superior rate capability (77.1 mA h g^{-1} at 100 A g^{-1}). The AC||NS superbattery device displays a high energy density of 71 W h kg^{-1} at an outstanding power density of 31400 W kg^{-1} , and remarkable cyclic stability with 80% of capacitance retention after 20000 GCD cycles (Fig. 13e). To understand the electrochemical performance of specific phases, density-functional theory calculations are utilized. The superior performance is due to the existence of a higher density of states near the Fermi level and greater quantum capacitance of $\text{NiSe}_2 > \text{Ni}_3\text{Se}_2 > \text{NiSe} > \text{Ni}_6\text{Se}_5$, as presented in Fig. 13f. It is clear that the controlled electrosynthesis with a suitable phase is an effective approach to achieve highly conductive, durable, and high-rate nickel selenide electrodes for supercapacitor–battery hybrid energy storage devices.

To enhance the electrochemical performance of metal selenides, the mixed flexible $(\text{Ni}, \text{Co})_{0.85}\text{Se}$ nanotube arrays were fabricated hydrothermally on carbon fabric with the metallic conductivity of $1.67 \times 10^6 \text{ S m}^{-1}$ and high mass loading (5.63 mg cm^{-2}) [144]. The $(\text{Ni}, \text{Co})_{0.85}\text{Se}$ exhibits a robust faradaic reaction with distinct $\text{Ni}^{2+}/\text{Ni}^{3+}$ and $\text{Co}^{2+}/\text{Co}^{3+}/\text{Co}^{4+}$ redox peaks, resulting in a high areal capacitance of 2.33 F cm^{-2} at 4 mA cm^{-2} , with 80% capacitance retention at 20 mA cm^{-2} . The assembled $(\text{Ni}, \text{Co})_{0.85}\text{Se}$ /porous graphene film asymmetric supercapacitor device, operating at 1.8 V , delivers an areal capacitance of 529.3 mF cm^{-2} and a volumetric capacitance of 6330 mF cm^{-3} at 1 mA cm^{-2} . In addition, the device shows remarkable cyclic stability, with 85% capacitance retention over 10,000 cycles at 20 mA cm^{-2} , and delivers a high energy density of 2.85 mWh cm^{-3} at a power density of 10.76 mW cm^{-3} .

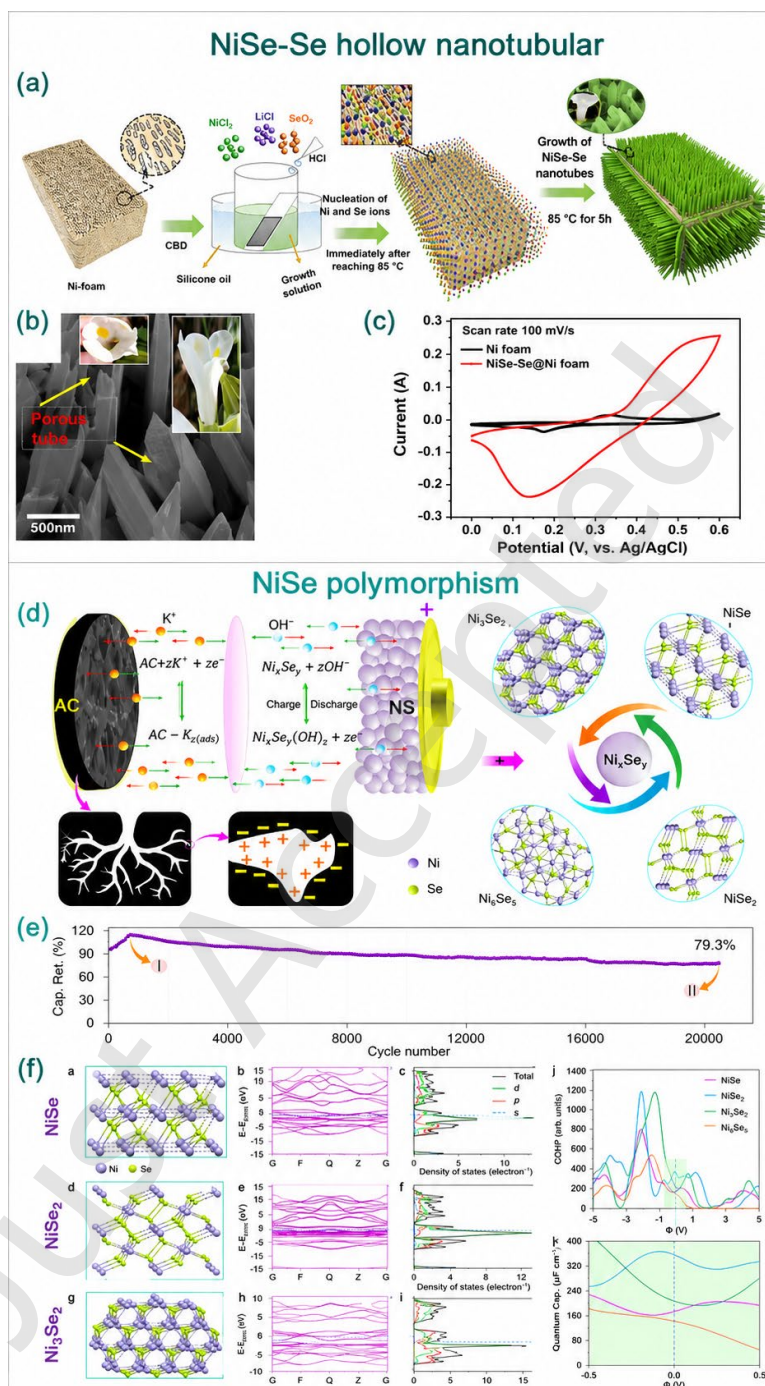


Figure 13 (a) Schematic diagram of the synthesis method for NiSe-Se nanotubes. (b) Tubular and porous morphology of the fabricated NiSe-Se nanotubes showing close resemblance to the tuberos flower (inset digital image). (c) CV curves for pristine Ni foam and the NiSe-Se@Ni foam electrode at a scan rate of 100 mV s^{-1} . Reproduced with permission from Ref. [142] © 2020, American Chemical Society. (d) Graphical representation of the AC||NS device and interconversion of the NS polymorphs. (e) Interconversion of the NS polymorphs (trigonal Ni_3Se_2 , hexagonal NiSe , orthorhombic NiSe_2 , and orthorhombic Ni_6Se_5) in the charge-discharge process. (f) Cycling stability of the AC||NS device at 5 A

g⁻¹; (g) DFT analysis: Crystal structure, band structure, and density of states of NiSe, NiSe₂, and Ni₃Se₂ polymorphs. (h) The calculated quantum capacitance as a function of the local electrode potential (Φ) for NiSe, NiSe₂, Ni₃Se₂, and Ni₆Se₅ polymorphs. Reproduced with permission from Ref. [143] © 2022, American Chemical Society.

Further, the electrochemical activity can be improved through defect engineering and transition-metal doping on the flexible substrate [145]. For instance, nanograss-flower-like Ag-decorated NiV₂Se₄ material grown on Ni–Cu–Co conductive fabric by a simple oil bath approach, as illustrated in Fig. 14a [146]. The SEM result shows the 3D NiV₂Se₄ nanograss flowers on the Ni–Cu–Co fabric, whereas, NiV₂Se₄–Ag is also 3D nanograss flower morphology, where Ag NPs uniformly distributed on the NiV₂Se₄ nanograss fibers (Fig. 14b). The CV curves (Fig. 14c) of NiV₂Se₄ and NiV₂Se₄–Ag confirm the predominant Faradaic in charge storage mechanisms and the CV integral area of the NiV₂Se₄–Ag electrode show a much greater than that of the NiV₂Se₄ electrode, indicating the higher specific capacitance of the NiV₂Se₄–Ag electrode. Likewise, GCD (Fig. 14d) further confirms the enhanced electrochemical performance of the NiV₂Se₄–Ag electrode, with a longer discharge time than that of the NiV₂Se₄ electrode. The NiV₂Se₄–Ag electrode delivers an areal capacity of 424 $\mu\text{Ah cm}^{-2}$ at 1 mA cm^{-2} and exhibits superior cycling performance, with capacity retention of 97% after 10000 GCD at 9 mA cm^{-2} (Fig. 14e). The assembled NiV₂Se₄–Ag//AC pouch cell HSC deliver a maximum energy density of 77 Wh kg^{-1} at a power density of 749 W kg^{-1} at 1 A g^{-1} with capacitance retention of 95% of its capacitance after 10000 cycles.

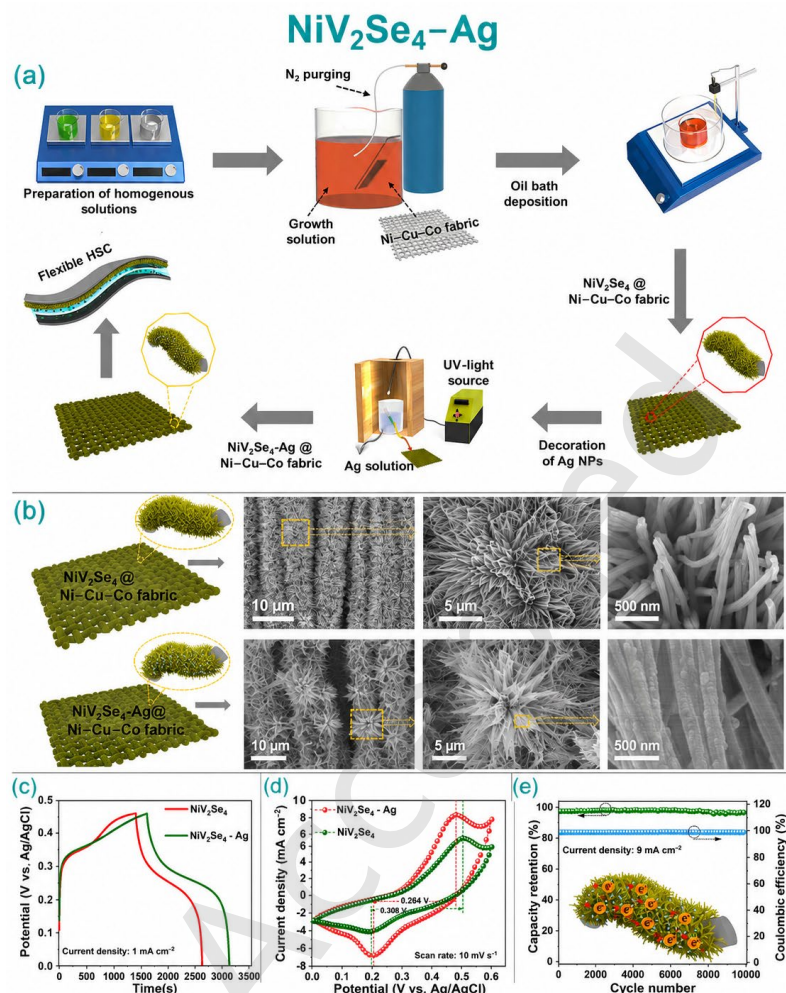


Figure 14 (a) Schematic illustration of the development of NiV₂Se₄ and NiV₂Se₄-Ag nanograin flowers. (b) HRSEM images of NiV₂Se₄ and NiV₂Se₄-Ag. (c) CV curves of NiV₂Se₄ and NiV₂Se₄-Ag. (d) GCD curves of NiV₂Se₄ and NiV₂Se₄-Ag. (e) Cycling stability of NiV₂Se₄-Ag (inset: schematic diagram of the charge-transfer pathways). Reproduced with permission from Ref. [146] © 2024, Springer Nature Ltd.

Similarly, Miao and co-workers [147] developed hierarchical urchin-like Mn-Co selenide (Mn_{0.32}Co_{0.68}Se₂) architecture on nickel foam via a two-step hydrothermal approach (Fig. 15a). The Mn_{0.32}Co_{0.68}Se₂ exhibits 3D microspherical architecture composed with radially aligned nanowires, which offer rich electrochemically active sites, shorten ion-diffusion pathways, and strong electrical contact with current collector (Fig. 15b). The Mn_{0.32}Co_{0.68}Se₂ electrode exhibits obvious Faradaic charge storage, deliver a high specific capacitance of 1656 F g⁻¹ (161 mAh g⁻¹) at 1 A g⁻¹, and show an excellent cycling stability with 91.8% capacitance retention after 8000 cycles (Fig. 15c). The Mn_{0.32}Co_{0.68}Se₂//AC asymmetric supercapacitor with the potential window of 1.4 V, achieve a high energy density of 55.1 Wh kg⁻¹ at power density of 880 W kg⁻¹. The hierarchical Mn-Co selenide nanoarchitectures provide mechanical stability, high conductivity, metal-selenium bonding, and multivalent redox chemistry (Co²⁺/Mn²⁺), thereby enhancing the electrochemical performance of the supercapacitor device. The

annealing condition significantly influences the performance of supercapacitors. In parallel, the MnCo-Se electrodes were fabricated by controllable electrodeposition on nickel foam at different annealing temperatures [148]. The MnCo-Se shows a coral-like morphology composed of interwoven nanowire clusters. Annealing at 400 °C induces a mixed orthorhombic and cubic CoSe₂ phase with homogeneous Mn incorporation (MCSe-400). The MCSe-400 electrode exhibits a larger CV integral area (Fig. 15d) and a longer discharge time (Fig. 15e) than the MCSe-0, MCSe-300, and MCSe-500 electrodes, confirming its superior electrochemical performance. The MCSe-400 delivers a high specific capacity of 509 C g⁻¹ at 1 A g⁻¹, maintains 68.6% of its capacity at 10 A g⁻¹, and shows a low charge-transfer resistance of 0.53 Ω, lower than that of MCSe-0 (0.76 Ω), MCSe-300 (0.89 Ω), and MCSe-500 (0.98 Ω) (Fig. 15f), demonstrating rapid interfacial kinetics. Moreover, MCSe-400 shows excellent cyclic stability, with 94.3% capacity retention after 5000 cycles. The asymmetric MCSe-400//AC supercapacitor delivers a high energy density of 46.2 Wh kg⁻¹ at a power density of 800 W kg⁻¹, with 89.2% capacitance retention over 5000 cycles (Fig. 15g), highlighting the effectiveness of coral-like MnCo selenide nanoarchitectures for high-performance aqueous supercapacitor applications.

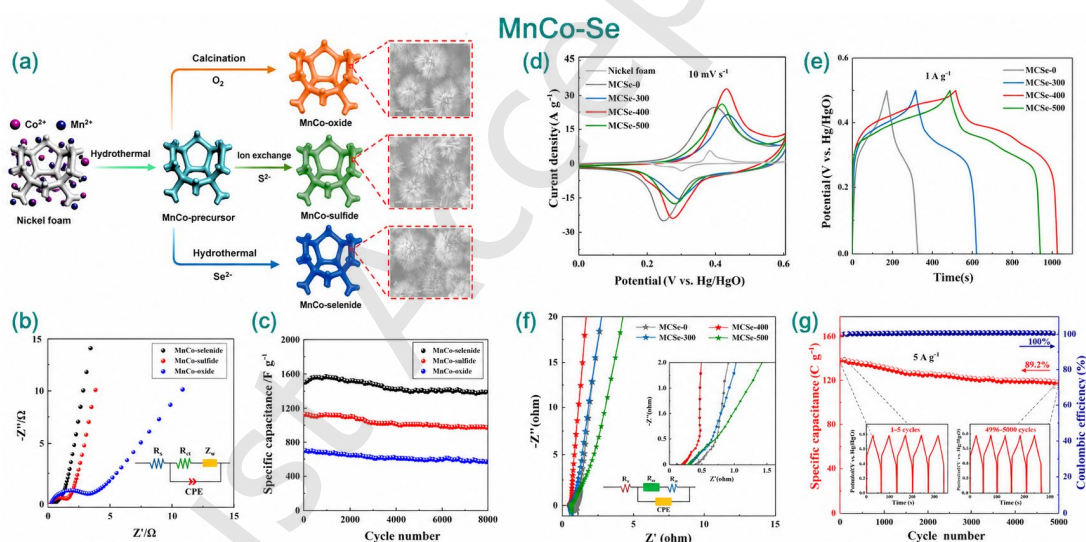


Figure 15 (a) Graphical representation for the construction of hierarchical urchin-like MnCo-oxide/sulfide/selenide on nickel foam. (b,c) EIS and cycling performance analysis of hierarchical urchin-like MnCo-oxide/sulfide/selenide on nickel foam. (Reproduced with permission from Ref. [147] © 2019, American Chemical Society. (d) CV curves of MCSe-X (X = 0, 300, 400 and 500 °C) samples. (e) GCD curves of MCSe-X (X = 0, 300, 400 and 500 °C) samples. (f) EIS curves of MCSe-X (X = 0, 300, 400 and 500 °C) samples (inset: Equivalent circuit). (g) Cyclic stability and Coulombic efficiency of MCSe-400//AC ASC after 5000 GCD cycles at 5 A g⁻¹ (inset: the GCD curves of the first five and the last five cycles). Reproduced under the BB CY license from Ref. [148] © 2023, MDPI.

Metal–organic framework (MOF)-derived metal selenides have emerged as promising electrode materials due to their high electrical conductivity, rich redox activity, and tunable porous structures. The unique morphology provides a large surface area and abundant active sites, facilitating efficient ion diffusion and charge storage. As an example, Ameri et al. [149] developed MOF-derived Mn–Ni–Se (MNSe) nanoarchitectures with hollow spherical morphology, which deliver a high specific capacity of 325.6 mA h g⁻¹ (1172.16 C g⁻¹) at 2 A g⁻¹ and improved R_{ct} (0.25 Ω), outperforming MNS@NF due to the higher intrinsic electrical conductivity of selenium. In addition, the MNSe exhibited excellent cyclic stability, retaining 96.8% of its capacity after 15000 GCD cycles. The asymmetric device (Mn–Ni–Se//AC) achieves a high energy density of 66.1 Wh kg⁻¹ at a power density of 858.45 W kg⁻¹ with 94.1% cycling stability over 15000 cycles, indicating that MOF-derived hollow mixed-metal selenide nanoarrays effectively enhanced conductivity, and a self-supported approach provides robust mechanical stability to meet next-generation supercapacitor devices. In another study, a MOF-derived bimetallic CoMnSe architecture developed on nickel foam (CoMnSe/NF) with optimized reaction time (2 h) [150]. The CoMnSe@NF-2h electrode delivers a high areal capacity of 1714 mC cm⁻² at 1 mA cm⁻² and a specific capacity of 519 C g⁻¹ at 1 A g⁻¹, with a low R_{ct} (3.6 Ω cm⁻²) and remarkable cycling stability with 87% capacity retention after 5000 cycles. Furthermore, an asymmetric supercapacitor CoMnSe@NF-2h//O, N, S-doped AC achieves a high specific capacity of 139 C g⁻¹, an energy density of 35.47 Wh kg⁻¹ at a power density of 459.7 W kg⁻¹, and maintains 82.6% capacitance retention after 10,000 cycles. The mixed-metal centers and selenium's good electrical conductivity significantly enhance charge-transport kinetics, providing a high electrochemically active surface area and shorter ion-diffusion pathways, resulting in improved electrochemical performance.

Furthermore, the electrochemical performance can be enhanced by introducing a metal dopant into the host electrode materials. As an example, vanadium-doped bimetallic Ni–Co–Se (NCSE) was derived from ZIF-67, and its electrochemical performance was studied [151]. The controlled integration of vanadium induces lattice distortion, provides multivalent redox sites (Ni²⁺/Ni³⁺, Co²⁺/Co³⁺, V⁴⁺/V⁵⁺), and enhances electrical conductivity without disrupting the host crystalline structure. Owing to excellent specific surface area (120 m² g⁻¹), the NCSE sample delivers an extraordinary specific capacitance of 2070 F g⁻¹ (828 C g⁻¹) at 1 A g⁻¹. Moreover, the NCSE possesses a low R_{ct} (4 Ω) and the lowest Ohmic resistance (14.55 Ω). The assembled NCSE//MPC asymmetric supercapacitor delivers a high specific capacitance of 326.8 F g⁻¹ and an outstanding energy density of 116.2 Wh kg⁻¹ at a power density of 799.8 W kg⁻¹. Notably, the ASC exhibits remarkable cycle stability with 83.7% capacitance retention after 10,000 cycles, confirming that the vanadium dopant meaningfully improves the electrochemical performance of the supercapacitors. In another approach, Ni_xCo_{3-x}Se₄ electrodes were synthesized by a one-step solvothermal method and evaluated for their electrochemical performance [152]. The Ni_{0.95}Co_{2.05}Se₄ delivers an ultra-high specific capacitance of 1038.75 F g⁻¹ at 1 A g⁻¹ with excellent rate capability (94.6% retention at 10 A g⁻¹), which is attributed to reduced charge-transfer resistance and enhanced OH⁻ diffusion kinetics. The asymmetric Ni_{0.95}Co_{2.05}Se₄//AC supercapacitor device delivers a high energy density of 37.22 Wh kg⁻¹ at a power

density of 800.9 W kg^{-1} and outstanding cycling stability, with 95.21% capacitance retention after 4000 cycles, highlighting the synergistic advantages of the $\text{Ni}^{2+}/\text{Ni}^{3+}$ and $\text{Co}^{2+}/\text{Co}^{3+}$ redox couples in the hierarchical morphology of the electrode materials.

Mixed metal oxides/selenides are considered potential electroactive materials for supercapacitors owing to their various oxidation states, which enhance redox activity [28]. As an example, Gopalakrishnan, and Badhulika [153] fabricated dahlia flower-like 3D core-shell structured $\text{NiCo}_2\text{O}_4/\text{NiCoSe}_2$ (NCO/NCS) electrodes on nickel foam via hydrothermal and controlled electrodeposition methods, as illustrated in Fig. 16a. Firstly, the dahlia flower-like morphology of NCO (Fig. 16b) developed on nickel foam, and then crumpled nanosheets are decorated over NCO, resulting in a core-shell nanostructure of NCO/NCS (Fig. 16c). The NCO/NCS electrode offers ample electroactive sites and continuous electron transport pathways due to the unique core-shell architecture. The NCO/NCS/NF displays higher integral areas under the CV curve and a longer discharge time than nickel foam and NCO/NF electrodes, confirming its higher specific capacitance. The NCO/NCS/NF delivers a high specific capacitance of 2045.5 F g^{-1} (227 mAh g^{-1}) at 1.8 A g^{-1} with a very low R_{ct} of 0.08Ω , revealing efficient charge transport. The NCO/NCS/NF//AC asymmetric hybrid supercapacitor delivers a high energy density of 54.5 Wh kg^{-1} at a power density of 350 W kg^{-1} and maintains 82.5% capacitance retention after 10,000 cycles. The superior electrochemical performance is ascribed to the hierarchical 3D framework that provides more voids to access electrolyte, promotes rapid ion/electron transport, and presents multivalent Ni/Co redox sites, emphasizing the effectiveness of rationally engineered bimetallic core-shell nanoarchitectures for high-energy supercapacitor applications (Fig. 16d). In a parallel, Rahmati et al., [154] developed NiCoSe_2 nanosheets covered urchin-like NiCo_2O_4 ($\text{NiCo}_2\text{O}_4/\text{NiCoSe}_2$) on nickel foam via hydrothermal followed by electrochemical electrodeposition method (Fig. 16e). The vertically aligned NiCoSe_2 nanowire arrays decorated ultrathin NiCoSe_2 nanosheets results a rough core-shell morphology with rich electroactive sites. In Fig. 16f, the integrated area of the CV curve for the $\text{NiCo}_2\text{O}_4/\text{NiCoSe}_2$ electrode is higher than that of the NiCo_2O_4 electrode. Moreover, the discharge time of the $\text{NiCo}_2\text{O}_4/\text{NiCoSe}_2$ electrode is longer than that of the NiCo_2O_4 electrode (Fig. 16g), further confirming the higher specific capacitance of the electrode material. The $\text{NiCo}_2\text{O}_4/\text{NiCoSe}_2$ hybrid electrode delivers a high areal capacitance of 12.09 F cm^{-2} at 5 mA cm^{-2} , shows a low electrical resistance of 1.18Ω , and retains 87.6% of its original capacitance after 3000 cycles, confirming fast charge kinetics and structural stability. The $\text{NiCo}_2\text{O}_4/\text{NiCoSe}_2$ //AC asymmetric supercapacitor (Fig. 16h) reaches an energy density of 66.13 Wh kg^{-1} at a power density of 233 W kg^{-1} , with remarkable cyclic stability of 90.9% capacitance after 2000 cycles (Fig. 16i), demonstrating that hierarchical nanoarchitectures significantly enhance specific capacitance, rate capability, and cyclic stability of the supercapacitor device.

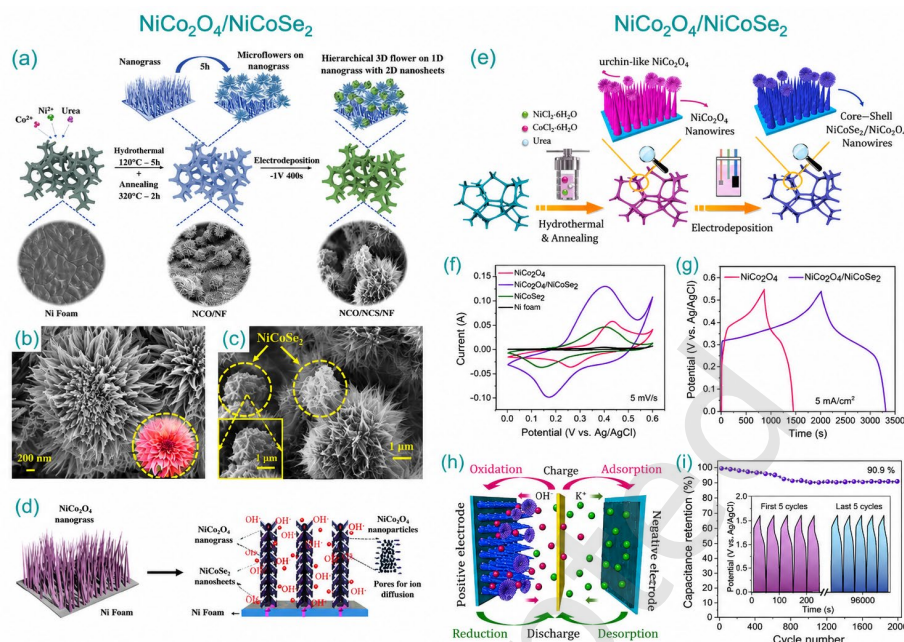


Figure 16 (a) Schematic of synthesis of self-supported NiCo₂O₄/NiCoSe₂. (b) SEM image of microflower-like NiCo₂O₄. (c) SEM image of NiCo₂O₄/NiCoSe₂. (d) Illustration of the mechanism of NCO/NCS/NF for a supercapacitor. Reproduced with permission from Ref. [153] © 2021, American Chemical Society 2021. (e) Schematic diagram of the fabrication process of NiCo₂O₄/NiCoSe₂ on nickel foam. (f) The comparison of the CV curves of the developed electrodes at a scan rate of 5 mV/s. (g) The comparison of the GCD curves of the NiCo₂O₄ and NiCo₂O₄/NiCoSe₂ electrodes at 5 mA/cm². (h) Schematic of the charge and discharge process in the assembled NiCo₂O₄/NiCoSe₂//AC ASC device. (i) Cycling performance of the NiCo₂O₄/NiCoSe₂//AC device (the inset shows the first and last 5 GCD cycles). Reproduced with permission from Ref. [154] © 2023, Elsevier Ltd.

Compared to binary metal selenides, ternary metal selenides exhibit improved charge storage properties due to the presence of multivalent redox sites. A hierarchical ternary Co–Mn–Fe selenide electrode was developed on nickel foam using Co–Mn–Fe layered double hydroxide nanoflowers with different concentrations of SeO₂ (1, 2, and 3 mmol, named CMFSe/NF-1, CMFSe/NF-2, CMFSe/NF-3, and CMFSe/NF-4) [155]. The CMFSe/NF-3 exhibits a three-dimensional interconnected nano-flower morphology composed of ultrathin, porous nanosheet petals, with a high specific surface area (149.5 m² g⁻¹) that synergistically promotes rapid electrolyte diffusion, efficient ion buffering, and continuous electron transport pathways. The CMFSe/NF-3 electrode delivers a high specific capacity of 883.1 C g⁻¹ at 1 A g⁻¹, which outperforms those of CoMnFeLDH (502.1 C g⁻¹), CMFSe/NF-1 (516.3 C g⁻¹), CMFSe/NF-2 (613.3 C g⁻¹), and CMFSe/NF-4 (689.9 C g⁻¹). Moreover, the CMFSe/NF-3 electrode exhibits remarkable cycling stability, with 94.9% capacity retention after 9000 cycles. The asymmetric CMFSe//AC hybrid supercapacitor delivers a high energy density of 62.8 Wh kg⁻¹ at a power density of 840.6 W kg⁻¹, with excellent cyclic stability, retaining 91.6% of its capacitance, demonstrating that rationally designed 3D porous multi-metal selenide materials are highly effective for high-energy and

high-power supercapacitor applications. The enhanced electrochemical performance of 3D transition-metal selenide electrodes is attributed to the synergistic effects of a hierarchical porous structure, multimetallic redox activity, binder-free assembly, and an improved electronic structure. In addition, 3D architectures offer high surface area, abundant active sites, and shorter ion-diffusion pathways, minimizing resistance, enhancing electron transport, and improving structural integrity, thereby delivering high capacitance/capacity, excellent rate capability, elevated energy density, and excellent cycling durability.

8.5 Core–Shell and heterostructured selenide architectures

In recent years, the development of core–shell and heterostructured architectures has emerged as an approach in supercapacitor research, which enhances charge storage, enables rapid ion diffusion, and improves structural robustness under long-term cycling [156]. However, the limited surface area, complex synthesis route, and significant pore damage restrict electrolyte access, increasing internal resistance and reducing cyclic stability, thereby decreasing the electrochemical performance of the supercapacitors. Core-shell layered metal hydroxides have demonstrated superior electrochemical performance due to their unique layered structures, which facilitate efficient ion intercalation/deintercalation and offer abundant electroactive sites. For example, a hierarchical core–shell NiSe/Ni(OH)₂ nanoarchitecture was synthesized via a hydrothermal method followed by electrodeposition, delivering a high areal capacity of 1.89 mAh cm⁻² [157]. The assembled NiSe/Ni(OH)₂//AC asymmetric device presents an energy density of 72.13 Wh kg⁻¹, confirming that the core-shell structure enhances electrochemical performance. In another approach, Liu et al. developed core–shell CoFe₂Se₄@CoNi carbonate hydroxide (CoFe₂Se₄@CoNi-CH) on carbon cloth via two-step electrodeposition and hydrothermal method, as depicted in Fig. 17a [158]. As shown in Fig. 17b, the CoFe₂Se₄@CoNi-CH/CC electrode exhibits the largest integral CV area compared with the pristine samples (CoFe₂Se₄/CC and CoNi-CH/CC), indicating improved charge storage properties. In addition, CoFe₂Se₄@CoNi-CH electrode exhibits the longest discharge time compared to CoFe₂Se₄/CC and CoNi-CH/CC (Fig. 17c). The CoFe₂Se₄@CoNi-CH/CC presents a high specific capacity of 218.6 mAh g⁻¹, which is higher than CoFe₂Se₄/CC (20.8 mAh g⁻¹) and CoNi-CH/CC (164.2 mAh g⁻¹) electrodes, with higher rate performance of 65.5% capacity retention at 20 A g⁻¹. The CoFe₂Se₄@CoNi-CH electrode reveals a lower R_{ct} value of 0.255 Ω than those of CoFe₂Se₄/CC (0.282 Ω) and CoNi-CH/CC (0.493 Ω) electrodes (Fig. 17d), indicate the faster ion diffusion rate and enhanced ion transport kinetics with remarkable capacity retention of 83.1% after 5000 cycles (Fig. 17e). The assembled hybrid supercapacitor (CoFe₂Se₄@CoNi-CH/CC//BPC-800 HSC) device delivers an energy density of 67.3 Wh kg⁻¹ at a power density of 765.9 W kg⁻¹. The HSC device shows excellent cyclic stability, retaining 85.4% of its initial capacity after 20000 cycles. The large surface area and core-shell architecture with a porous framework of CoFe₂Se₄@CoNi-CH/CC provide abundant active sites and rapid ion-diffusion pathways, resulting in enhanced reaction kinetics and charge-storage capability.

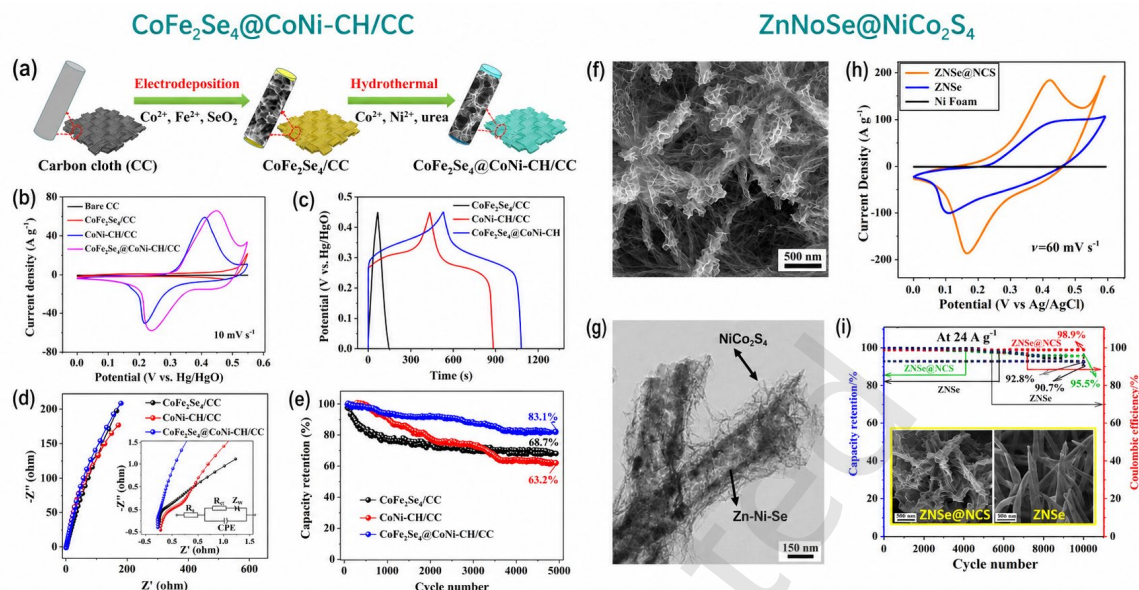


Figure 17 (a) Schematic diagram of the preparation process of $\text{CoFe}_2\text{Se}_4@\text{CoNi-CH}$ hybrid nanowires on carbon cloth. (b) CV profile comparisons of three electrodes at 5 mV s^{-1} . (c) GCD profile comparisons at 1 A g^{-1} . (d) Nyquist plot and (inset: magnified plot and fitted equivalent circuit). (e) Capacity retention versus cycle-number profile after 5000 cycles at 7 A g^{-1} . Reproduced with permission from Ref. [158] © 2022, Elsevier Ltd. (f) FE-SEM images of the $\text{ZnSe}@\text{NCS}@\text{NF}$. (g) TEM images of the $\text{ZnSe}@\text{NCS}$. (h) CVs of the $\text{ZnSe}@\text{NCS}@\text{NF}$, $\text{ZnSe}@\text{NF}$, and bare NF electrodes at a scan rate of 60 mV s^{-1} . (i) Cycle stability and coulombic efficiency of the $\text{ZnSe}@\text{NCS}@\text{NF}$ and $\text{ZnSe}@\text{NF}$ electrodes at 24 A g^{-1} (inset: FE-SEM images of the $\text{ZnSe}@\text{NCS}@\text{NF}$ and $\text{ZnSe}@\text{NF}$ electrodes after the cycling test). Reproduced with permission from Ref. [159] © 2020, American Chemical Society.

In parallel, a bimetallic (Zn-Ni) selenide integrated bimetallic (Ni-Co) sulfur ($\text{Zn-Ni-Se}@\text{NiCo}_2\text{S}_4@\text{ZnSe}@\text{NCS}$) core-shell electrode was fabricated on nickel foam through a series of hydrothermal methods, selenization, and electrodeposition methods [159]. As shown in Fig. 17f-g, the SEM and TEM analyses confirm that the $\text{ZnSe}@\text{NCS}$ hybrid structures comprise a vertical NiCo_2S_4 nanosheet shell and a ZnNi-Se nanowire core, resulting in a core-shell structure. The $\text{ZnSe}@\text{NCS}$ has a higher surface area of $96.5 \text{ m}^2 \text{ g}^{-1}$ than ZnSe ($66.6 \text{ m}^2 \text{ g}^{-1}$), which provides sufficient active sites. The $\text{ZnSe}@\text{NCS}@\text{NF}$ displays a larger CV integrated area and high redox current than the $\text{ZnSe}@\text{NF}$ electrode, suggesting the excellent charge-storage ability and better capacity of the $\text{ZnSe}@\text{NCS}@\text{NF}$ electrode (Fig. 17h). The $\text{ZnSe}@\text{NCS}@\text{NF}$ electrode shows a high specific capacity of 393.7 mAh g^{-1} (1417.3 C g^{-1}) at 1 A g^{-1} , which is higher than the $\text{ZnSe}@\text{NF}$ electrode ($304.75 \text{ mAh g}^{-1}$ (1097.1 C g^{-1})). Moreover, the $\text{ZnSe}@\text{NCS}@\text{NF}$ has lower R_s (0.45Ω) and R_{ct} (0.35Ω) than $\text{ZnSe}@\text{NF}$ ($R_s = 0.68 \Omega$ and $R_{ct} = 0.48 \Omega$), demonstrating improved ionic conductivity. Notably, $\text{ZnSe}@\text{NCS}@\text{NF}$ results in a high-rate capacity (394 mAh g^{-1}), and outstanding cycling stability (95.5% retention over 10,000 cycles) (Fig. 17i). The $\text{ZnSe}@\text{NCS}@\text{NF}@\text{AC}@\text{NF}$ asymmetric device achieves an energy density of 52.37 Wh kg^{-1} at a power density of 800.5 W kg^{-1} , with 94.9% of capacity retention after 10000 GCD cycles. The

enhanced electrochemical performance of the ZNSe@NCS@NF electrode is due to its higher surface area and unique core-shell morphology, which synergistically provide more active electroactive sites, facilitate electrolyte ion infiltration, and enable better contact between the electrode and the electrolyte, resulting in fast charge transfer. The above examples demonstrate that the enhanced electrochemical performance of self-supported core-shell metal selenide electrodes is related to the synergetic interaction between selenide cores and electroactive shell materials; porous architectures, rich heterointerfaces, and direct binder-free configuration facilitate fast ion transport, increase electroactive sites, reduce volume expansion, and ensure excellent structural stability, eventually delivering high capacity, outstanding rate capability, and prolonged cycling durability.

8.6 Selenide/carbonaceous hybrid materials

As we all know, carbonaceous materials have been used in energy storage via the EDLC mechanism due to their exceptional surface area and electrical conductivity, thereby enhancing electrochemical performance. However, a limited operating potential window of carbonaceous materials limits the specific capacitance and energy density of the supercapacitors [160]. Metal selenides possess high electrical conductivity and rich redox activity; nevertheless, they suffer from structural instability during prolonged cycling. Therefore, incorporation of carbon materials with metal selenides improves surface area and stability, resulting in enhanced electron transport and cycling stability of the supercapacitor device. NiSe nanostructures anchored on carbon nanotube (CNT) architectures were electrodeposited onto a stainless steel substrate (NiSe@CNT/SS) with different pulse-reversal cycles (100, 200, 300, 400, and 500) [161]. The pulse-reversal electrodeposition cycles significantly influence the electrochemical performance of NiSe@CNT. The NiSe@CNT/SS-100 electrode displays a greater electrochemical performance with a specific capacity of 126 mAh g^{-1} (1007 F g^{-1}) than NT@NiSe/SS-200 (84 mAh g^{-1} ; 673 F g^{-1}), CNT@NiSe/SS-300 (60 mAh g^{-1} , 479 F g^{-1}), and CNT@NiSe/SS-400 (21 mAh g^{-1} , 166 F g^{-1}). The hybrid CNT@NiSe/SS//rGO/NF supercapacitors deliver an energy density of 32.1 Wh kg^{-1} at a power density of 823 W kg^{-1} with robust floating-test stability over 50 hrs. The intertwined, porous CNT network provides greater surface area. It minimizes charge-transfer resistance by enabling continuous electron pathways and strong interfacial contact. In contrast, the NiSe nanotube architecture prevents particle agglomeration, thereby directly enhancing the electrochemically active surface area and ionic diffusion. As another example, Miao et al. developed MOF-derived CoSe₂ nanoparticles embedded in N-doped carbon nanosheet arrays (CoSe₂/NC) on nickel foam at different annealing temperatures (300 °C (CoSe₂/NC-300), 350 °C (CoSe₂/NC-350), 400 °C (CoSe₂/NC-400), and 450 °C (CoSe₂/NC-450)) [162]. Initially, triangular-shaped Co-MOF nanosheets were developed on nickel foam. Then, CoSe₂/NC-NF with nanosheet arrays was obtained in the presence of selenium at 400 °C (CoSe₂/NC), as shown in Fig. 18a. The CoSe₂/NC has a BET surface area of $74.6 \text{ m}^2 \text{ g}^{-1}$, providing abundant active sites and rapid ion-diffusion channels. The CoSe₂/NC-400 electrode exhibits prominent pseudocapacitive behavior with large CV integral area (Fig. 18b-c) and CoSe₂/NC-400 presented high specific capacity of 120.2 mAh g^{-1} at 1 A g^{-1} , superior rate capability (retaining 61.2% from 1 to 20 A g^{-1}), very low R_{ct} ($0.11 \text{ }\Omega$), and exceptional

cycling durability with 92% of capacitance retention after 10000 cycles (Fig. 18d). The asymmetric (CoSe₂/NC//AC) supercapacitor delivers a high energy density of 40.9 Wh kg⁻¹ at power density 980 W kg⁻¹, maintains 90.2% capacitance after 10,000 cycles. In parallel, MoSe₂/MWCNT (MSNT) composite thin films were developed onto SS using successive ionic layer adsorption and reaction (SILAR) [163]. The MSNT/SS hierarchical structure delivers a high specific capacity (gravimetric capacitance) of 192 mAh g⁻¹ (690 F g⁻¹) at 5 mV s⁻¹ with a low R_{ct} (2.01 Ω). It shows excellent cyclic stability with 88% capacity retention after 2000 cycles. The asymmetric MSNT//MnO₂ cell (1.6 V) achieves 112 F g⁻¹, an energy density of 35.6 Wh kg⁻¹, and 80% capacitance retention over 2000 cycles, confirming efficient charge storage enabled by the MSNT nanoarchitecture. Another report by Pandit and Sankapal [141] synthesized a CeSe₂ nanopebble-anchored MWCNT (CeSe₂/MWCNT) composite on an SS substrate (Fig. 18e). The CeSe₂/MWCNT exhibits both electric double-layer capacitance and Faraday charge-storage mechanisms. Notably, the CeSe₂/MWCNT exhibits the maximum oxidation current and large CV area compared to the MWCNTs and CeSe₂ (Fig. 18f). The CeSe₂/MWCNT delivers a high specific capacitance of 451.4 F g⁻¹ at 2 mA cm⁻² with a low R_{ct} (5.5 Ω cm²) and stable capacitance retention of 70.7% over 4000 cycles, representing efficient ion diffusion and interfacial charge transfer. The flexible solid-state symmetric supercapacitor with a PVA–LiClO₄ gel as electrolyte, present a specific capacitance of 128.8 F g⁻¹, a high energy density of 36.3 Wh kg⁻¹ at power density of 2.8 kW kg⁻¹, and excellent cycling stability with 84.1% capacitance retention over 4000 cycles (Fig. 18g). In addition, the symmetric device maintaining 86.9% capacitance retention under mechanical bending (180°) condition (Fig. 18h).

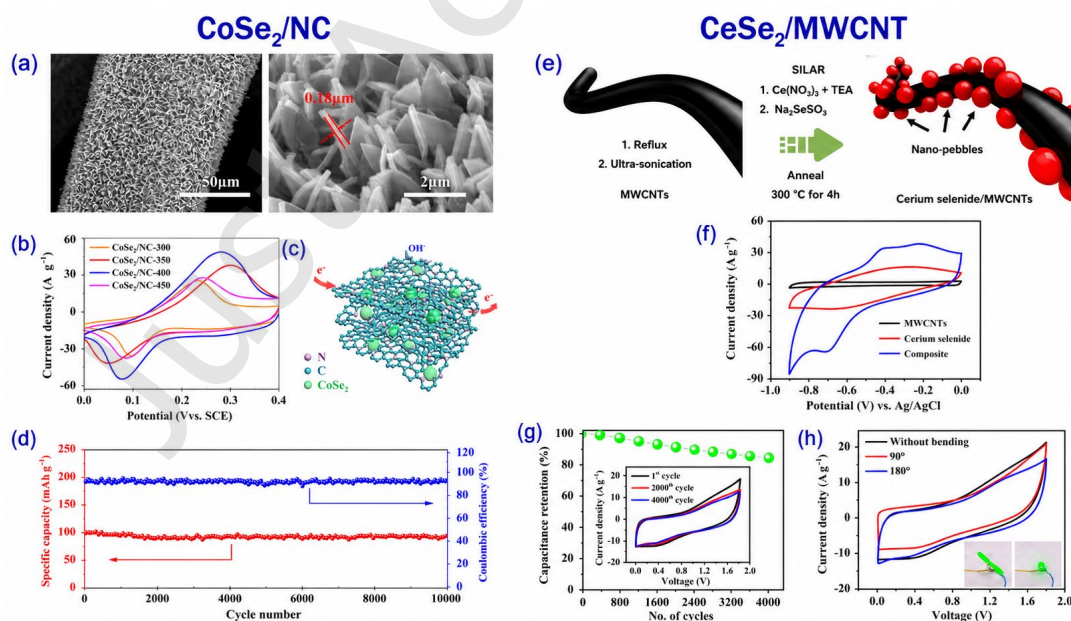


Figure 18 (a) SEM images of the CoSe₂/NC-400 electrode. (b) Comparison of CV curves of CoSe₂/NC-300, CoSe₂/NC-350, CoSe₂/NC-400, and CoSe₂/NC-450 electrodes. (c) Schematic illustration of CoSe₂/NC nanosheets with outstanding electrochemical performance. (d) Cycling performance of the CoSe₂/NC-400 electrode. Reproduced with permission from Ref. [162] © 2020, American Chemical

Society. (e) Preparation of cerium selenide on MWCNTs using a simple chemical process. (f) CV plots for MWCNTs, cerium selenide, and composite electrodes at 100 mV/s. (g) Cyclic stability of the cell for 4000 cycles at 100 mV/s (the inset displays CV plots with cycle numbers). (h) CV with different bending angles at 100 mV/s (inset shows the operation of the mini motor fan). Reproduced with permission from Ref. [164] © 2022, American Chemical Society.

Trimetallic Co–Mn–Fe selenide nanoflowers encapsulated within a reduced graphene oxide (RGO) network (CMFS-RGO) synthesized on nickel foam [165]. The resulting CMFS–RGO architecture displays a hierarchical 3D nanoflower morphology with interconnected porous frameworks, and a large specific surface area ($288.6 \text{ m}^2 \text{ g}^{-1}$), offering abundant electroactive sites and efficient electrolyte diffusion. The CMFS-RGO/NF delivers a high specific capacity of 1112.5 C g^{-1} at 1 A g^{-1} , excellent rate performance with 79.4% capacity retention at 25 A g^{-1} , and superior cycling stability with 94.6% capacity retention after 10000 cycles. The CMFS-RGO/NF//AC hybrid supercapacitor operates over a wide voltage window of 1.6 V, achieving a high energy density of 65 Wh kg^{-1} at a power density of 802.5 W kg^{-1} and maintaining 91% capacity retention over prolonged cycling, highlighting the effectiveness of trimetallic selenide–graphene nanoarchitectures for high-performance supercapacitor applications. As we all know, the enhanced electrochemical performance of nanostructured metal selenide/carbon composites is due to the presence of a conductive carbon skeleton, which maximizes exposure of redox-active sites, facilitates rapid ion transport, and maintains structural integrity. Besides, the self-supported binder-free architecture allows efficient electrical contact and continuous conductive pathways, thereby delivering high capacity, excellent rate performance, long-term cycling stability, and superior energy density. In summary, it is believed that carbon materials with metal selenides are highly meaningful for improving the energy storage of supercapacitor devices. Table 2 summarizes the synthesis, morphology, electrochemical performance, and cycling stability of transition metal selenide-based electrodes for supercapacitors.

Table 2 Summary of the synthesis, morphology, electrochemical performance, and cycling stability of transition metal selenide-based electrodes for supercapacitors.

Electrode	Synthesis method	Morphology	Current collector	Three-electrode system (Electrolyte)	Potential window (V)	Specific capacitance @ Current density	Two-electrode system (Electrolyte)	Potential window (V)	Energy density (Wh kg ⁻¹)	Power density (W kg ⁻¹)	Cyclic stability	Ref.
0D metal selenide Architectures												
NiFe ₂ Se ₄ /NF	Electrodeposition + Selenization	Nanoparticles	NF	3 M KOH	0–0.6	2679.8 F g ⁻¹ @ 1 A g ⁻¹	ASC (NiFe ₂ Se ₄ //AC, 3 M KOH)	0–1.6	45.6	800	101.4% after 10,000 cycles	[109]
1D												
Ni _{4.5} Co _{4.5} Se/NPCC	Hydrothermal + Selenization	Core-shell nanowires	NPCC	6 M KOH	–0.1–0.6	2660 F g ⁻¹ @ 1.2 A g ⁻¹	ASC (Ni _{4.5} Co _{4.5} Se//Fe ₃ C/CF, PVA/KOH gel)	0–1.5	47.4	1500	80% after 4,000 cycles	[115]
NSCCH/NF	Hydrothermal	Nanowire arrays	NF	3 M KOH	0–0.6	9.56 F cm ⁻² @ 4 mA cm ⁻² 1037.5 F g ⁻¹ @ 0.25 mA cm ⁻²	ASC (NSCCH//AC, 3 M KOH)	0–1.6	35.5	128	85.3% after 3,000 cycles	[116]
CuSe ₂ /Cu	Hydrothermal	Nanoneedles	Cu foil	1 M NaOH	0–0.65		NA	—	—	—	118% after 1,000 cycles	[119]
2D – Single metal selenide Architectures												
Ni _{0.85} Se/CFC	Hydrothermal + Selenization	Nanosheet arrays	CFC	1 M KOH	0–0.6	820 F g ⁻¹ @ 1 A g ⁻¹	ASC (Ni _{0.85} Se//AC, PVA/KOH gel)	0–1.6	29.0	5512	81.3% after 5,000 cycles	[124]
MoSe ₂ /Ti mesh	Hydrothermal	Nanosheets	Ti mesh	2 M Na ₂ SO ₄	–0.8–0.1	525.5 F g ⁻¹ @ 1 A g ⁻¹	NA	—	—	—	Excellent after 1,700 cycles	[125]
SnSe/SS	Solution heat-up	Nanosheets	SS	1.5 M KOH	–1.2–0.0	794 F g ⁻¹ @ 5 mV s ⁻¹	SSC (PVA/KOH gel)	0–1.0	8.9	2700	96% after 5,000 cycles	[126]
SnSe/SS	Solution heat-up	Nanosheets	SS	3 M LiClO ₄	–1.0–0.0	645 F g ⁻¹ @ 2 mV s ⁻¹	SSC (PVA/LiClO ₄ gel)	0–1.0	8.9	2700	96% after 5,000 cycles	[126]
α-MnSe-16/NF	Hydrothermal	Nanosheets	NF	6 M KOH	0–0.45	88.3 mAh g ⁻¹ @ 1 A g ⁻¹	ASC (α-MnSe-16//AC, 6 M KOH)	0–1.45	39.3	920	94% after 5,000 cycles	[127]
2D Binary metal selenide Architectures												
MnSe ₂ /Co _{0.85} Se/NF	Electrodeposition	Nanosheets	NF	1 M KOH	0–0.55	1380 C g ⁻¹ @ 1 A g ⁻¹	ASC (MnSe ₂ /Co _{0.85} Se//AC, 1 M KOH)	0–1.6	49.6	750	90% after 10,000 cycles	[128]
NiCoSe/NF	Electrodeposition	Nanosheet arrays	NF	2 M KOH	0–0.50	2054 F g ⁻¹ @ 1 A g ⁻¹	ASC (NiCoSe//AC, PVA/KOH gel)	0–1.6	44.6	976	85.2% after 5,000 cycles	[129]
(Ni ₁ Co) ₂ Se/CC	Hydrothermal + Selenization	Nanosheet arrays	CC	2 M KOH	0–0.50	2054 F g ⁻¹ @ 1 A g ⁻¹	ASC ((Ni ₁ Co) ₂ Se//AC, PVA/KOH gel)	0–1.6	44.6	976	85.2% after 5,000 cycles	[130]
NiCo ₂ Se ₄ /Fe ₂ CoSe ₄ @NF	Hydrothermal + Selenization	Flower-like nanosheets	NF	3 M KOH	0–0.60	323.7 mAh g ⁻¹ @ 1 A g ⁻¹	HSC (NiCo ₂ Se ₄ /Fe ₂ CoSe ₄ //AC, 3 M KOH)	0–1.6	51.8	749.8	92.3% after 10,000 cycles	[131]
CuNi ₂ S ₄ @NiCoSe ₂ /CC	Electrodeposition	Core-shell nanoflowers	CC	2 M KOH	0–0.60	979.2 C g ⁻¹ @ 1 A g ⁻¹	HSC (CuNi ₂ S ₄ @NiCoSe ₂ //AC, 2 M KOH)	0–1.6	78.9	720.2	91.4% after 50,000 cycles	[132]
FeNiSe ₂ @Ni _{4.5} Co _{4.5} S ₈ /CC	Electrodeposition + Selenization	Core-shell nanosheets	CC	2 M KOH	0–0.55	236.9 mAh g ⁻¹ @ 1 A g ⁻¹	HSC (FeNiSe ₂ @Ni _{4.5} Co _{4.5} S ₈ //AC, 2 M KOH)	0–1.6	69.0	799.2	91.2% after 10,000 cycles	[133]
2D – Metal doped												
Sn _{0.33} Ni _{0.67} Se ₂ /CFC	Hydrothermal + Selenization	Nanosheet arrays	CFC	3 M KOH	0–0.60	346 mAh g ⁻¹ @ 1 mA cm ⁻²	ASC (Sn _{0.33} Ni _{0.67} Se ₂ //AC, PVA/KOH gel)	0–1.6	90.3	631	96.4% after 10,000 cycles	[137]
Co _{0.6} Mn _{0.4} Se@CoMo-LDH/NF	Hydrothermal + Selenization + Electrodeposition	Nanosheets	NF	6 M KOH	0–0.60	1405.2 C g ⁻¹ @ 1 A g ⁻¹	HSC (Co _{0.6} Mn _{0.4} Se@CoMo-LDH//AC, 6 M KOH)	0–1.8	71.4	1130	106.2% after 10,000 cycles	[138]
Co ₂ FeSe ₄ /NF	Electrodeposition	Nanosheet arrays	NF	1 M KOH	0–0.50	3393 mF cm ⁻² @ 3 mA cm ⁻²	ASC (Co ₂ FeSe ₄ //AC, PVA/KOH gel)	0–1.6	157.3 mWh cm ⁻²	2250 mW cm ⁻²	85.8% after 10,000 cycles	[139]
3D metal selenide Architectures												
NiSe	Solvothermal	Snowflake-like	NF	2 M KOH	0–0.55	763.5 C g ⁻¹ @ 1.5 A g ⁻¹	AC//NiSe (2 M KOH)	0–1.6	32.04	1112.44	99.95% after 10,000 cycles	[140]

Sponge-like NiSe	Electrodeposition	Sponge-like	NF	2 M KOH	0–0.55	751 C g ⁻¹ @ 1 A g ⁻¹	AC//NiSe (2 M KOH)	0–1.6	42.2	958.3	99% after 9,000 cycles	[141]
NiSe–Se	Chemical bath deposition	Hollow nanotubes	NF	1 M KOH	0–0.60	2447.46 F g ⁻¹ @ 1 A g ⁻¹	AC//NiSe–Se (1 M KOH)	0–1.6	29.90	594.46	95.09% after 10,000 cycles	[142]
Ni ₃ Se ₂ polymorphs	Electrodeposition	Nanostructures	Conductive substrate	1 M KOH	—	180.3 mAh g ⁻¹ @ 1 A g ⁻¹	AC//NixSe _y	—	71	31,400	80% after 20,000 cycles	[143]
(Ni,Co) _{0.85} Se	Hydrothermal–Selenization	Nanostructures	CF	KOH	—	2.33 F cm ⁻² @ 4 mA cm ⁻²	PG//[(Ni,Co) _{0.85} Se (KOH)]	1.8	2.85 mWh cm ⁻³	10.76 mW cm ⁻³	85% after 10,000 cycles	[144]
Ag-decorated NiV ₂ Se ₄	Oil-bath deposition	Nanoflowers	Ni–Cu–Co fabric	2 M KOH	—	—	AC//NiV ₂ Se ₄ –Ag (2 M KOH)	1.6	77	749	95% after 10,000 cycles	[146]
MnCo selenide	Hydrothermal	Hierarchical urchin-like	NF	KOH	0–0.50	1656 F g ⁻¹ @ 1 A g ⁻¹	MnCo-selenide//AC (KOH)	0–1.60	55.1	880	91.8% after 8000 cycles	[147]
Coral-like MnCo selenide	Electrodeposition	Coral-like dendritic	NF	KOH	0–0.60	509 C g ⁻¹ @ 1 A g ⁻¹	MnCo selenide//Activated carbon (KOH)	0–1.60	46.2	800	94.3% after 5000 cycles	[148]
MOF-derived MnNiSe@NF	Hydrothermal + selenization	Hollow spheres	NF	KOH	0–0.60	1172.16 C ⁻¹ (325.6 mAh g ⁻¹) @ 2 A g ⁻¹	MNSe@NF//AC@NF (KOH)	0–1.60	66.1	858.45	94.1% after 15000 cycles	[149]
MOF-derived CoMnSe@NF	Hydrothermal + selenization	Nanosheets	NF	KOH	0–0.60	519 C g ⁻¹ @ 1 A g ⁻¹	CoMnSe@NF-2h//O,N,S@AC@NF	0–1.60	35.47	11500	82.6% after 10000 cycles	[150]
V-doped NiCo selenide (NCSE4)	Hydrothermal	Crumpled Nanosheets	CC	KOH	0–0.50	2070 F g ⁻¹ (828 C g ⁻¹) @ 1 A g ⁻¹	NCSE4//MOF-derived porous carbon (ASC)	0–1.60	116.19	799.81	83.71% after 10000 cycles	[151]
Ni _{0.95} Co _{2.05} Se ₄	Solvothermal	Spherical particles	NF	2 M KOH	0–0.5	1038.75 F g ⁻¹ @ 1 A g ⁻¹	Ni _{0.95} Co _{2.05} Se ₄ //AC (2 M KOH)	0–1.6	37.22	800.9	95.21% after 4000 cycles	[152]
NiCo ₂ O ₄ /NiCoSe ₂ /NF	Hydrothermal–Electrodeposition	Core–shell nanoflowers	NF	3 M KOH	0–0.6	2045.5 F g ⁻¹ @ 1.8 A g ⁻¹	NiCo ₂ O ₄ /NiCoSe ₂ //AC (3 M KOH)	0–1.6	54.5	350	82.5% after 10,000 cycles	[153]
NiCo ₂ O ₄ /NiCoSe ₂	Hydrothermal–Electrodeposition	Core–shell nanowires	NF	3 M KOH	0–0.55	12.09 F cm ⁻² @ 5 mA cm ⁻²	NiCo ₂ O ₄ /NiCoSe ₂ //AC (3 M KOH)	0–1.6	66.13	233	90.9% after 2000 cycles	[154]
CMFSe/NF	Hydrothermal–Selenization	Porous nanoflowers	NF	6 M KOH	NR	671.2 C g ⁻¹	CMFSe//AC (6 M KOH)	NR	62.8	40,735.1	NR	[155]
Core–Shell metal selenide Architectures												
NiSe/Ni(OH) ₂	Hydrothermal–Electrodeposition	Core–shell nanorods	NF	2 M KOH	0–0.5	1.89 mAh cm ⁻² @ 5 mA cm ⁻²	NiSe/Ni(OH) ₂ //AC (2 M KOH)	0–1.6	46.7	800	90.2% after 10,000 cycles	[157]
CoFe ₂ Se ₄ @CoNi–CH	Electrodeposition–Hydrothermal	Core–shell nanowires	CC	3 M KOH	0–0.55	218.6 mAh g ⁻¹ @ 1 A g ⁻¹	CoFe ₂ Se ₄ @CoNi–CH//BPC (3 M KOH)	0–1.6	67.3	765.9	85.4% after 20,000 cycles	[158]
Zn–Ni–Se@NiCo ₂ S ₄	Hydrothermal–Selenization–Electrodeposition	Core–shell nanowires	NF	6 M KOH	0–0.5	393.7 mAh g ⁻¹ @ 1 A g ⁻¹	Zn–Ni–Se@NiCo ₂ S ₄ //AC (6 M KOH)	0–1.6	52.37	800.5	93.3% after 10,000 cycles	[159]
Carbon/metal selenide composite Architectures												
CNT@NiSe/SS	CVD–Electrodeposition	CNT-supported nanoparticles	SS	6 M KOH	0–0.6	126 mAh g ⁻¹ @ 1 A g ⁻¹	CNT@NiSe//Graphene (3 M KOH)	1.6	32.1	823	Stable for 50 h	[161]
CoSe ₂ /NC–NF	MOF-derived selenization	Carbon nanosheets	NF	6 M KOH	0–0.6	120.2 mAh g ⁻¹ @ 1 A g ⁻¹	CoSe ₂ /NC//AC (6 M KOH)	1.6	40.9	980	90.2% after 10,000 cycles	[162]
MoSe ₂ /MWCNT	SILAR	CNT-supported nanosheets	SS	1 M LiCl	—	192 mAh g ⁻¹	MSNT//MnO ₂ (Aqueous electrolyte)	1.6	35.6	—	88% after 2000 cycles	[163]
Cerium selenide/MWCNT	SILAR	Nanopebbles	SS	Liquid electrolyte	—	451.4 F g ⁻¹ @ 2 mA cm ⁻²	SSC (PVA–LiClO ₄ gel)	1.8	36.3	2800	86.9% after 4000 cycles	[164]
CMFS–RGO/NF	Hydrothermal–Selenization	RGO-wrapped nanoflowers	NF	6 M KOH	0–0.6	1112.5 C g ⁻¹ @ 1 A g ⁻¹	CMFS–RGO//AC (6 M KOH)	1.6	65	802.5	91% retention	[165]

NF, Nickel foam; CF, Carbon faber; SS, Stainless steel; CC, Carbon cloth; CFC, Carbon fibre cloth, NPCC, Nickel-plated cotton cloth

9 Summary and outlook

The self-supported materials are considered emerging electrode materials for high-energy-density supercapacitors. In particular, self-supported metal selenide-based supercapacitors have become a pivotal focus for several research groups due to selenium's excellent conductivity compared with metal oxides/sulfides. Despite remarkable progress, numerous critical challenges hinder the transformation of self-supported metal selenide electrodes from the laboratory to practical applications in supercapacitors. The structural instability and significant volume changes during charging and discharging often lead to cracking, pulverization, and loss of electrical contact, particularly in high-mass-loading electrodes, resulting in rapid capacity fading. In addition, despite the advantages of self-supported architecture, limited interfacial adhesion and mechanical strength can lead to delamination or detachment of active electroactive materials during prolonged cycling in flexible devices, necessitating stronger chemical bonding and hierarchical interlocking designs between the electrode and the electroactive materials. Another key issue lies in the trade-off between high mass loading and rate performance: greater thickness leads to sluggish ion transport and internal stress. Moreover, scalable and reproducible manufacturing remains a challenge, as commonly used lab-scale methods (hydrothermal, solvothermal, and electrodeposition) are difficult to translate to large-area and industrial processes. Finally, environmental and safety concerns related to selenium also warrant attention, and sustainable recycling approaches are needed to enable commercialization.

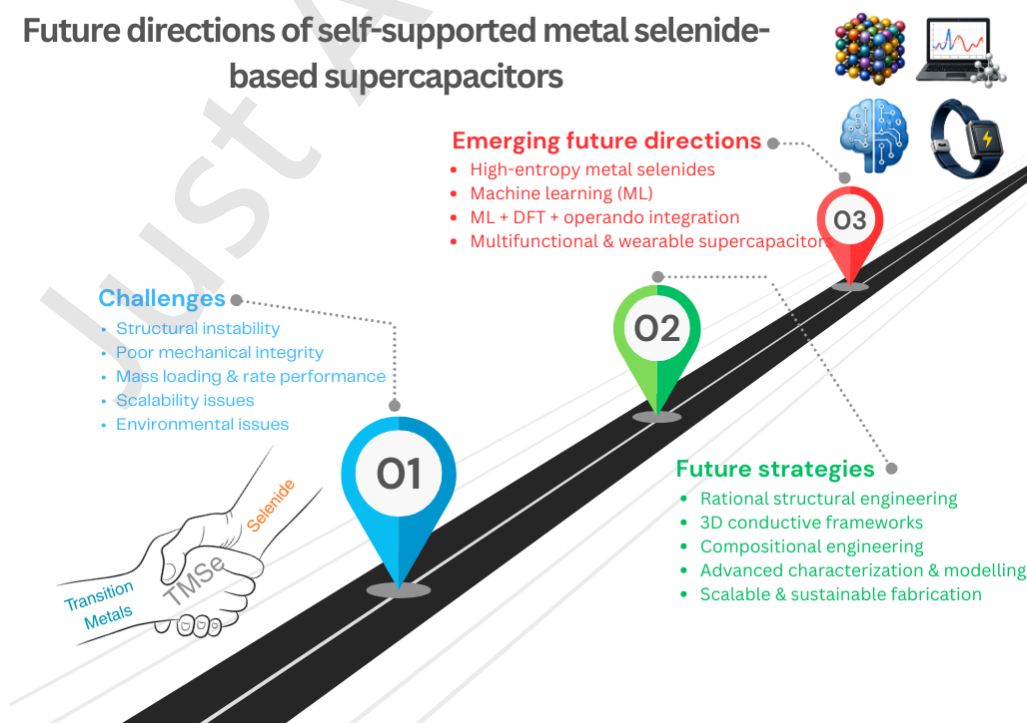


Figure 19 Overview of challenges and future directions of self-supported metal selenide-based electrode materials for supercapacitors.

To address the above-mentioned issues, future research should focus on the rational design of multiscale architectures with meso- and microscale porosity that can be effectively buffered by advanced architectures such as hollow, flower-like, or yolk-shell structures. Developing robust 3D architectures, such as MXene-, graphene-, or foam-based composites, to accommodate selenides without mechanical degradation during prolonged cycling. Hetero-anion/cation (S/Se, O/Se, P/Se, etc.) engineering and high-entropy metal selenides can further improve conductivity, structural stability, and electrochemical activity owing to the presence of multiple redox sites. Advanced characterization techniques, such as in situ or operando measurements with multiscale modeling, will be vital for understanding phase changes, interfacial redox reactions, and ion transport mechanisms, providing insight into the charging-discharging process. Meanwhile, scalable fabrication approaches such as printing methods and CVD require the development of high-performance metal-selenide-based devices with high-rate capability. Incorporating selenide-based electrodes into multifunctional and wearable systems, along with the design of stretchable, self-healable encapsulation materials, can enhance real-world applicability. As we all know, high-entropy metal-based electrode materials have shown potential for energy storage and conversion applications. In particular, slurry-form high-entropy metal/metal oxide/sulfide/phosphite systems are being used as electrode materials, thereby enhancing electrochemical performance. However, the polymer binder in the high-entropy metal-based electrode hinders effective ion transport. As discussed, the self-supported architecture displayed enhanced performance. However, developing high-entropy metal on the current collector is challenging due to the multimetallic nature. The above-mentioned issue can be addressed by developing MOF-derived high-entropy metal selenides on highly conductive substrates. Specifically, ZIF-67 derivatives could be a suitable choice for achieving self-supported, high-entropy metal selenides with enhanced electrochemical performance. Therefore, future research should prioritize developing high-entropy metal selenides on the current collector with suitable microstructure. Furthermore, machine learning is an emerging and powerful tool for the design and discovery of metal selenides, helping overcome the limited energy density of self-supported metal selenide-based supercapacitors. The machine learning approach reduces cost and time, thereby providing precise compositions to achieve high-performance metal-selenide-based electrode materials. Moreover, a machine learning approach integrated with density functional theory calculations and operando characterization can provide the rational design of advanced electrode architectures while minimizing conventional trial-and-error experimentation. In addition, the machine learning approach provides molecular-level information on the influence of structural architecture and charge transfer between electrolyte, electrode, and current collector. The overview of challenges and future directions is illustrated in Fig. 19. Therefore, self-supported metal selenide-based materials could be a promising electrode for next-generation sustainable energy storage technologies.

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Author contribution statement

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Data availability

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Declaration of competing interest

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