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A comprehensive review on MoSe₂ nanostructures with an overview of machine learning techniques for supercapacitor applications

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In the past few decades, supercapacitors (SCs) have emerged as good and reliable energy storage devices due to their impressive power density, better charge–discharge rates, and high cycling stability. The main components of a supercapacitor are its electrode design and composition. Many compositions are tested for electrode preparations, which can provide good performance. Still, research is widely progressing in developing optimum high-performance electrodes. Metal chalcogenides have recently gained a lot of interest for application in supercapacitors due to their intriguing physical and chemical properties, unique crystal structures, tuneable interlayer spacings, broad oxidation states, etc. MoSe₂, belonging to the family of Transition Metal Dichalcogenides (TMDs), has also been well explored recently for application in supercapacitors due to its similar properties to 2D materials. In this review, we briefly discuss supercapacitors and their classification. Various available synthesis routes for MoSe₂ preparation are summarized. A detailed assessment of the electrochemical performances of different MoSe₂ composites, including cyclic voltammetry (CV) analysis and galvanostatic charge–discharge (GCD) analysis, is given for symmetric and asymmetric supercapacitors. The limitations of MoSe₂ and its composites are mentioned briefly. The use of machine learning methods and algorithms for supercapacitor applications is discussed for forecasting valuable details. Finally, a summary is provided, along with conclusions.

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1. Introduction

The energy demands have been unboundedly increasing recently due to the ever-growing population, urbanization, industrialization and technological advancements. As a result, fossil fuels, the primary source of energy, are being depleted


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rapidly. Excessive use of fossil fuels in automobiles has caused many ecological problems such as air pollution. Other conventional energy sources such as wind energy, solar energy, and hydrothermal energy cannot provide continuous and uninterrupted steady supply of energy. Therefore, there is a need for the search of an alternative energy storage device that is clean, efficient, and sustainable and that can store a considerable amount of energy and provide this energy when required.^{1–5} In this context, the development of electrochemical energy storage devices such as capacitors, batteries, fuel cells, and supercapacitors has gained a lot of interest from researchers. Batteries have the disadvantages of poor power density, shorter cycle life, poor power performance, and overheating problems. Conventional capacitors have limited energy density, making them unsuitable for applications requiring long-term energy. SCs, as modern energy storage devices, have been developed to overcome these shortcomings. They are filling the gap between batteries and capacitors because SCs have 100 times better capacitance than normal capacitors. The SCs have impressive power density, quick charging and discharging rates, good cycling stability, and eco-friendliness.^{6–9} The development of SCs is important since they can address the future energy storage demands of small portable devices, electric vehicles, smart electronic equipment, hybrid systems, and flexible and wearable device applications.^{10–13} A supercapacitor's performance is majorly dependent on the materials of electrodes. Hence, researchers are highly focusing on developing new high-performance electrode materials for SCs for energy storage applications.

Two-dimensional (2D) materials are highly explored for use in supercapacitor electrodes owing to their large surface areas and sheet-like morphologies, which enable the maximum exposure of active sites.^{14–18} The 2D material family comprises graphenes, silicene, germanene, borophene, black-phosphorous, hexagonal boron-nitride, MXenes, TMDs, and many others.¹⁹ The evolution of 2D material-based supercapacitor electrodes started from graphene, and later, many

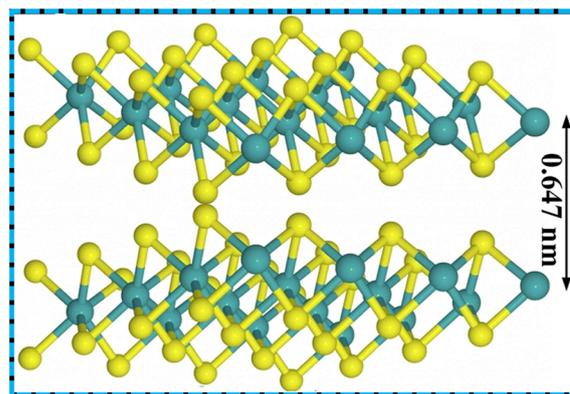


Fig. 1 Three-dimensional view of layers of MoSe₂ (Green: Mo, Yellow: Se). This figure has been reproduced from ref. 23 with permission from Springer Nature, Copyright 2018.

other materials in this class were explored further. Recently, TMDs (MoS₂, MoSe₂, WS₂, WSe₂, etc.) have attracted considerable attention from researchers to investigate as pseudocapacitive materials for SCs in energy storage and conversion fields.¹⁷ In 2D TMDs, two chalcogen layers (X = S and Se) sandwich a metal layer (M = Mo and W) creating an MX₂ structure.^{18,20} Among them, MoSe₂ has received considerable attention from researchers owing to its graphene-like layered structure, weak van der Waals interactions between the layers, and many other interesting properties. An analog of MoS₂, MoSe₂, has a layered structure with two Se layers sandwiched by a Mo layer at an interlayer spacing of about 6.5 Å. MoSe₂ layers are created by covalently joining the nearby atomic layers, and van der Waals forces stack the adjacent MoSe₂ layers into bulk structures.^{21,22} Ion intercalation and de-intercalation can occur easily due to the wide interlayer spacing between the layers of MoSe₂. Fig. 1 shows the three-dimensional view of MoSe₂ layers with a gap of 0.647 nm. Fig. 2 gives the number of research publications on MoSe₂ over the past few years. MoSe₂ has



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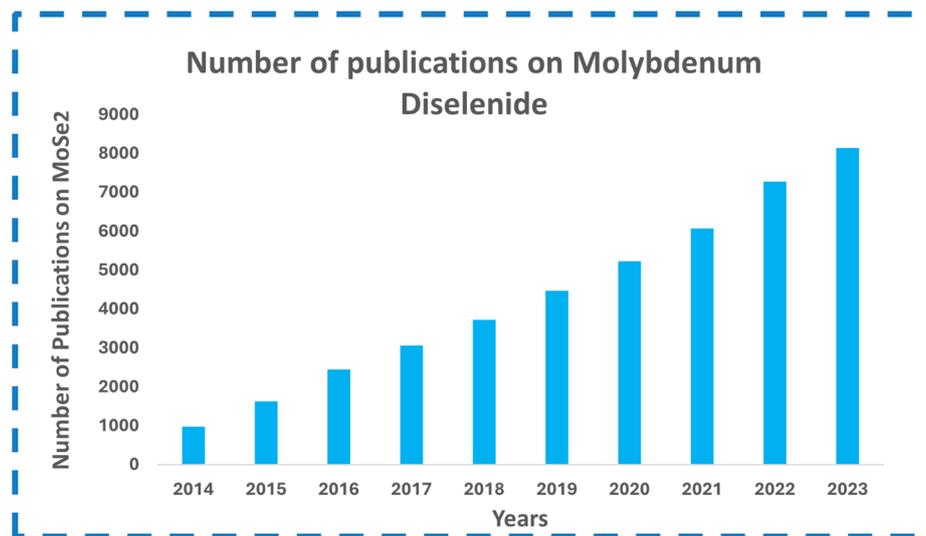


Fig. 2 Number of publications on MoSe₂ electrodes. Compiled from Google Scholar with search "MoSe₂".

a better metallic character than MoS₂, and its lower Gibbs free energy promotes its electrochemical activity in catalytic applications.²⁴ MoSe₂ has a better intrinsic electrical conductivity than that of MoS₂ due to the better metallic nature of Se than that of S, making it an intriguing narrow-band-gap semiconductor with comparable layered structures. Electrolyte ion intercalation is made simple by the wider layer gap (0.647 nm) between two MoSe₂ layers compared to other counterparts (e.g., graphite has 0.335 nm, and for MoS₂, its 0.615 nm). Low internal resistance and high faradaic capacitance are made possible by the unsaturated Se stable edge structure and the ability to provide many electrochemically active sites.²⁵ Two distinct symmetry types are present in MoSe₂ (1T octahedral and 2H trigonal prismatic). The 1T phase is metallic in contrast to the semiconducting 2H phase. Because of its thermodynamic instability, the 1T phase of MoSe₂ eventually changes into the 2H phase.^{22,26} All these interesting properties have made MoSe₂ gain research interests for SCs. Nevertheless, the specific capacitance of MoSe₂ is still too low to be utilized in its pure form for energy storage applications, and its electrical conductivity is still lower than that of carbon materials such as graphenes. Numerous researchers have created a variety of MoSe₂-based nanocomposites including graphenes, carbon aerogels, carbon nanotubes, and polyaniline, to address these shortcomings. Compared to pure MoSe₂, electrochemical experiments have shown that the MoSe₂-based composites provide outstanding cycling stability and a high specific capacity.

Recently, with the advancement of Machine Learning (ML) in the area of Artificial Intelligence (AI), it has become possible to develop better high-performance electrodes for SCs.²⁷ Specifically, by using the ML methods, it is now possible to accurately predict the capacitance of the electrode materials,²⁸ forecast the results without experimentations,²⁹ provide optimized reaction parameters,³⁰ estimate the reaction rate,³¹ explore new electrode materials,³² determine remaining useful

life,³³ analyze the state of health,³⁴ estimate the state of charge³³ and predict the cycling stability³⁵ as well. This will save a lot of time and effort for the researchers since a lot of details can be gathered from the ML models without performing trial-and-error experiments. For SCs, ML algorithms work on a large set of input data from already published works and results. Using suitable algorithms, the ML models are trained to gather and analyze the extensive input data. The ML models learn to map inputs to outputs by adjusting their parameters based on the training data. After this, the ML model can make decisions based on unseen future data. The ML models can update the new data and work in real time. Various ML models such as Neural Networks, Random Forest, XGBoost, Support Vector Machines, Decision Trees, and Linear Regression are available. By selecting an appropriate model, useful details can be accurately derived for developing supercapacitor electrodes as per the requirements.³⁶

Herein, we comprehensively review the MoSe₂ and MoSe₂-based composite electrodes for supercapacitor applications. The different synthesis routes adopted for the development of MoSe₂ are provided. Brief workings of implementing ML models in the field of SC applications for capacitance prediction, remaining useful life determination, *etc.*, are also discussed. Finally, challenges faced by MoSe₂-based electrodes and future perspectives are provided along with conclusions.

1.1 Supercapacitor fundamentals

SCs primarily take the form of Electric Double Layer Capacitors (EDLCs), pseudocapacitors and hybrids depending on energy storage techniques. EDLC electrodes store charges electrostatically and not *via* faradaic reactions or charge transfer. A double layer of charges is accumulated on the electrode when voltage is supplied. The diffusion of electrolyte ions occurs towards oppositely charged electrodes. When double layers are formed, this reduces the inter-separation distance, which contributes directly to an increase in capacitance. Here, the recombination



of ions does not take place during double-layer formation. EDLCs have good energy density due to the use of carbon-based electrodes, which have large surface areas, and due to reduction in charge separation distance. EDLCs have good power outputs, fast and quick energy intake, and better energy distribution. Moreover, energy storage mechanisms do not involve any chemical reactions, and they are non-faradaic in nature. Compared with batteries, which have the ability to withstand only a few thousand cycles, EDLCs have the ability to continue for millions of cycles. They have drawbacks of poor power density. Conducting polymers, metal oxides and other pseudocapacitive materials are selected in the case of pseudocapacitors. Here, energy storage is mainly electrochemical in nature. The mechanism of energy storage is dependent on faradaic reactions that occur between electrodes and electrolytes. Faradaic reactions involve quick oxidation and reduction reactions. In EDLCs, no charge transfer occurs, but in this case, charge transfer occurs between electrolytes and electrodes. When a potential is supplied, a faradaic current passes through the SC cell. Compared to EDLCs, pseudocapacitors can store more charges, provide better energy densities, and show increased specific capacitances. They have the disadvantage of low cyclability issues. Recently, hybrid capacitors have become a major class of supercapacitors which include both EDLCs and pseudocapacitors. Here energy is stored both electrostatically (non-faradaic) and electrochemically (faradaic). It combines the best features of EDLCs and pseudocapacitors, overcoming the drawbacks of both types, thereby providing more energy density and better life cycles. Hybrid capacitors can further be grouped into three types depending on the electrode materials and

configuration: asymmetric hybrids, composite hybrids, and battery-type hybrids. Composite hybrids consist of two or more materials that are utilized for electrodes. Composites can be binary, ternary, and quaternary. Several examples of composites such as carbon-carbon composites, conducting polymer composites, and carbon-metal oxide composites are available. Utilizing two or more materials may produce a synergistic effect that assists in charge transfer and energy storage. In asymmetric hybrids, both EDLCs and pseudocapacitor materials are used in combination. One electrode will be EDLC in nature, and the other electrode will be pseudocapacitive in nature, thereby utilizing both faradaic and non-faradaic processes. From the CV curves, distinction can be made between capacitive-type, pseudocapacitive-type, EDLC-type, and battery-type behaviors of electrodes. In the battery-type hybrid group, battery-type and SC-type electrodes are used together, thereby bridging the gap between the power density and the energy density.³⁷⁻⁴⁶ Fig. 3 gives the classification of SCs depending on possible materials used. Fig. 4 shows the stages of development of SCs, models for describing EDLC mechanisms, and Ragone plot for comparing available energy storage devices.

The performance of lab-scale prepared electrodes can be tested in three designs, namely three-electrode set-up, symmetric set-up, and asymmetric set-up. The three-electrode system comprises electrolyte, working electrode, reference electrode, and standard electrode. The two-electrode setup (symmetric and asymmetric) has two electrodes made up of highly supercapacitive materials. For symmetric, both electrodes will be based on the same material. For asymmetric, the anode material will be different from the cathode material.

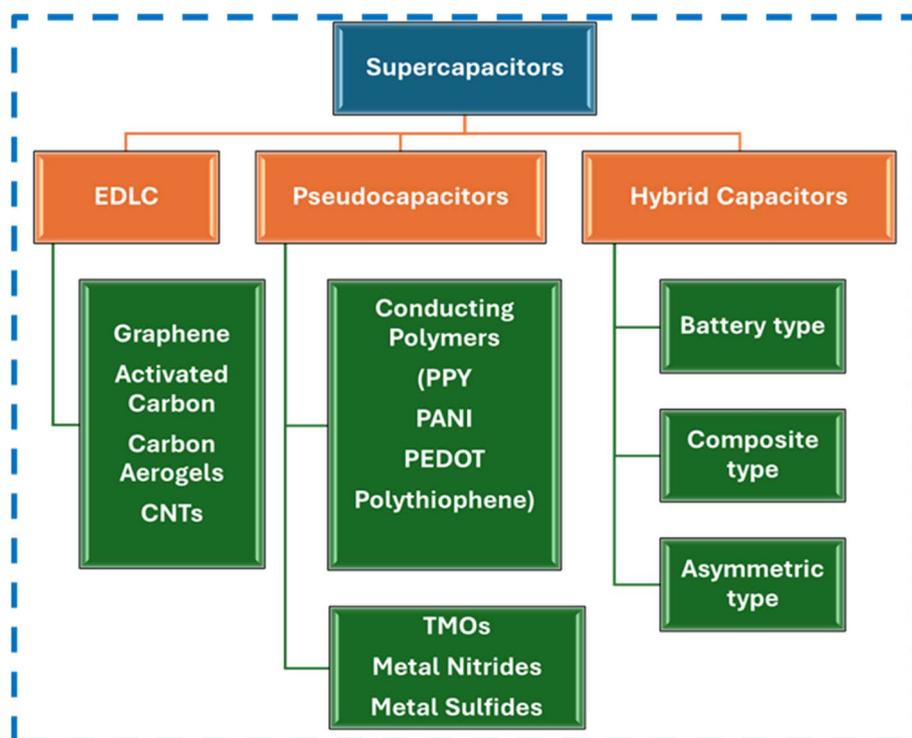


Fig. 3 Supercapacitor classification with the possible materials used.



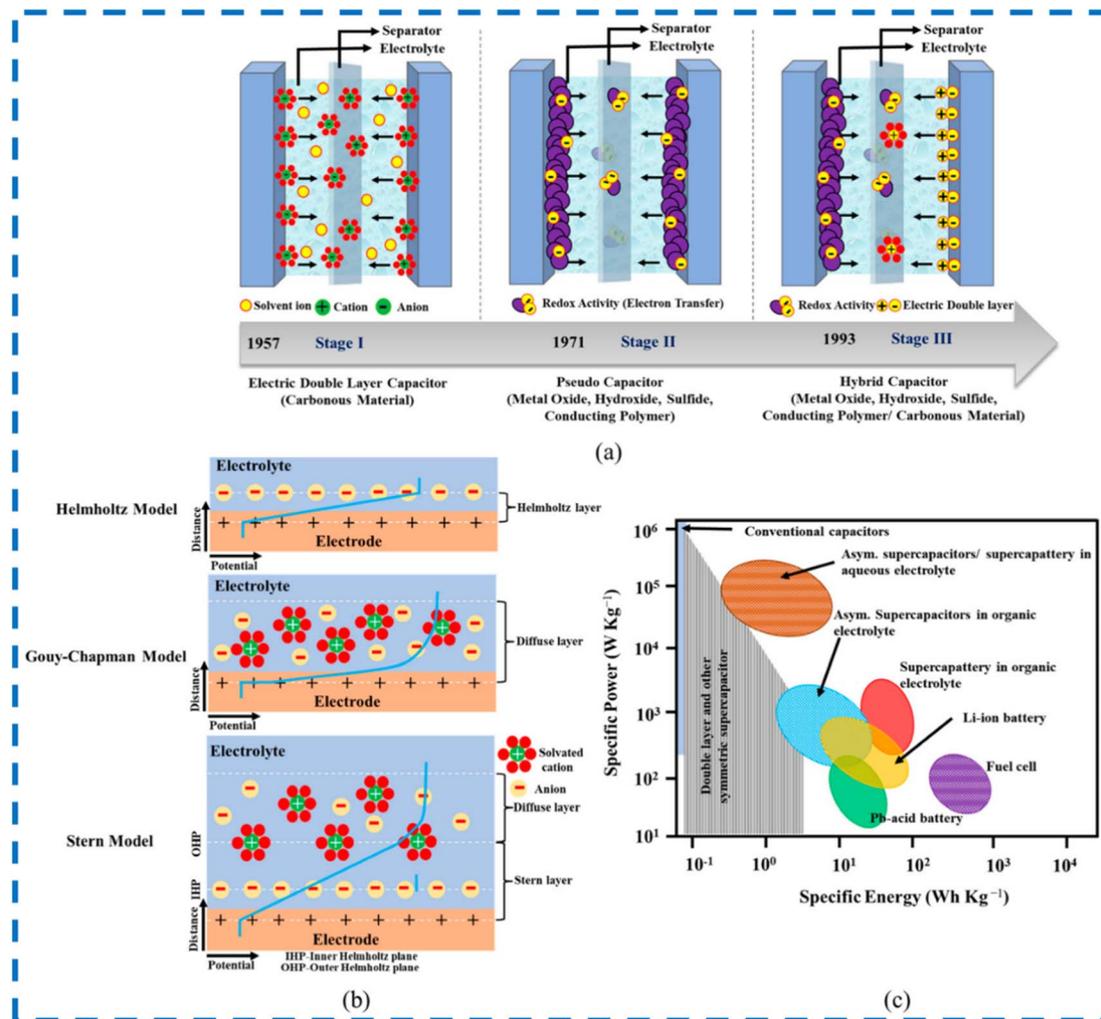


Fig. 4 (a) Classification of supercapacitors. (b) Models for EDLCs. (c) Comparing available energy storage devices. This figure has been reproduced from ref. 47 with permission from MDPI, copyright 2024.

Activated carbon (AC) has been majorly used as one of the asymmetric electrodes in several works. The two-electrode setup provides a better representation of the charge storage performance of the material compared to the three-electrode setup. The most frequently used one is the asymmetric and three-electrode set-up. From the CV curves, for the asymmetric set-up, the specific capacitance can be found using the following equation:

$$C_s = \frac{\int I(V)dV}{vm\Delta V} (\text{F g}^{-1})$$

where v stands for the scanning rate, m stands for the mass of electroactive sample, ΔV stands for the voltage window and integral represents the CV curve area.

From the GCD curves, for the asymmetric setup, the specific capacitances can be determined using the following equation:

$$C_s = \frac{I\Delta t}{m\Delta V} (\text{F g}^{-1})$$

where Δt stands for the discharging time, ΔV stands for the voltage window, and I/m stands for the current density.⁴⁸

2. Development of MoSe₂ nanostructures for supercapacitor applications

2.1 Liquid-phase exfoliation method

Liquid-phase exfoliation is a solution-processing method used to produce two-dimensional nanosheets from layered materials such as graphite in large quantities. This technique involves dispersing layered crystals in a suitable solvent and applying energy to separate the layers into individual nanosheets. The most common energy sources include ultrasonication, high shear mixing, and micro fluidization, which help overcome the van der Waals forces holding the layers together. The exfoliation of MoSe₂ into single-layer or few-layer structures using this technique is made easier by the weak van der Waals forces between MoSe₂ layers in bulk MoSe₂, which are comparable to those between the MoS₂ layers in bulk MoS₂.^{24,49} Jiang *et al.* employed a liquid-phase exfoliation technique to produce 2H MoSe₂ nanosheets using a precursor of MoSe₂ powder and a mixed solvent of water and ethanol.⁵⁰ Bath sonicator was used



for the exfoliation process, and a centrifuge was used to separate unexfoliated powders. The obtained products were further processed to prepare a thin film and an electrode to investigate their performance in SCs. In one of the reports, Rahul and Arora used the same method to produce MoSe₂ nanosheets using bulk MoSe₂ powder and isopropyl alcohol (IPA) as solvents. Sonication was done for 5 h at room temperature. The authors used a temperature-controlled water bath during the sonication process. The formation of exfoliated MoSe₂ nanosheets was validated by peaks in the Raman measurements. According to the FESEM result, the sample is made up of both large and small flakes and sheets that are evenly dispersed throughout the substrate surface. The structure displayed distinct crystal lattice fringes of around 0.64 nm, which were attributed to *d*-spacing for the (002) basal plane of 2H-MoSe₂, according to the TEM results. The electrode made from the obtained nanosheets was further investigated by the authors for further electrochemical analyses.⁵¹ In another study, Mendoza-Sánchez *et al.* reported MoSe₂ nanoplatelet preparation using the same method and studied their charge storage properties (Fig. 5).⁵²

2.2 Microwave-assisted synthesis method

Microwave synthesis is an innovative method that utilizes microwave radiation to accelerate chemical reactions,

significantly enhancing the efficiency and speed of various synthesis processes. This technique allows for rapid heating of reaction mixtures, often reaching temperatures above the boiling point of solvents, which can lead to improved reaction kinetics and higher yields compared to conventional heating methods. Leaf-like MoSe₂ with mesoporous structures was synthesized by Vattikuti and co-workers using a microwave irradiation technique. A solution containing a known amount of sodium molybdate, Se powder, and PVP as surfactant was vigorously stirred for 1 h in DI water and hydrazine hydrate. Then, the solution was exposed to microwave irradiation at 160 °C for 25 min, and then finally, the precipitates were collected for further studies. The Raman and XRD images confirmed the formation of MoSe₂. The authors reported that the as-prepared MoSe₂ had a surface area of 64.63 m² g⁻¹ and mesopore sizes of 8 nm. Mesoporous MoSe₂ nanostructures that resemble dry leaves are formed, as shown in SEM and HRTEM images. For 5000 cycles, the mesoporous MoSe₂ electrode demonstrated exceptional cycling stability while achieving a capacitance of 257.38 F g⁻¹ at 1 A g⁻¹.⁵³

2.3 Sonochemical method

The sonochemical method basically involves exfoliating the precursor material by applying high-intensity ultrasound energy

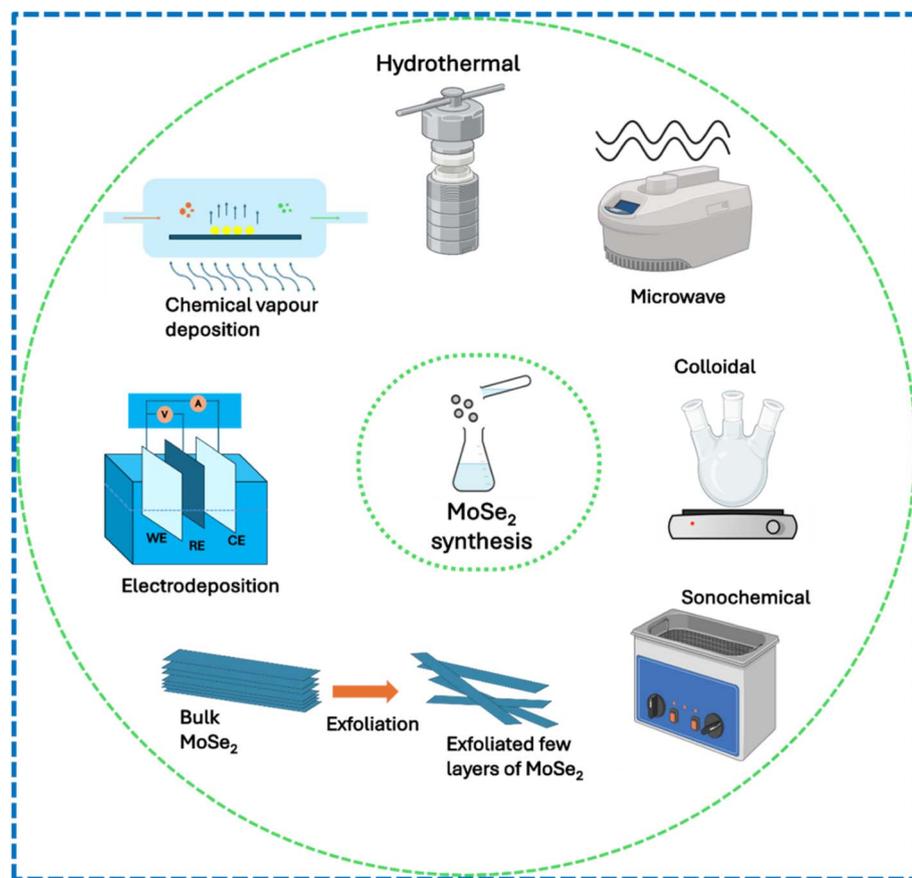


Fig. 5 Frequently used methods for MoSe₂ synthesis. Created in BioRender. FRANCIS, M. (2024) Colloidal icon: <https://BioRender.com/r76t891>, Microwave icon: <https://BioRender.com/o73b224>, Sonochemical icon: <https://BioRender.com/t91j774>, Hydrothermal icon: <https://BioRender.com/j53g540>.



for a certain period. The solvents used, sonication power and sonication time play crucial roles in obtaining a necessary number of layers. Fig. 11 shows the schematic of a typical sonochemical synthesis route. Using H_2O_2 as an intercalating agent in isopropyl alcohol, Kirubasankar and colleagues developed a simple and efficient sonochemical method for fabricating a few layers of MoSe_2 nanosheets. The optimization of the synthesis process was done by testing the probe-sonicator under ice-cold conditions for three distinct periods (15, 30, and 45 minutes) at a constant power.⁵⁴ The suspensions of MoSe_2 were collected after centrifugation and dried to obtain final products. The authors reported that the sonication time played a crucial role in determining the number of layers and yield of the products. A distinct hexagonal phase (2H- MoSe_2) with a space group (P_{63}/mmc) was confirmed from the XRD patterns, and the absence of any impurity peaks suggests that a pure hexagonal phase has formed. For the exfoliated MoSe_2 , the surface areas were determined to be $68 \text{ m}^2 \text{ g}^{-1}$. Further other characterizations were also carried out to confirm the desired material formation. The authors examined the synthesized nanosheets for further electrode performance analysis.

2.4 Electrodeposition method

Electrodeposition, also known as electroplating or electrochemical deposition, is a process used to deposit a layer of materials, typically metal, onto a substrate through the reduction of cations from an electrolyte solution. This technique involves immersing the substrate (acting as the cathode) in an electrolyte containing metal salts. When a current is applied, metal ions migrate to the cathode, where they gain electrons and are reduced to form a solid metal layer.

A simple electrochemical deposition (ECD) method was used by Mariappan *et al.* to create a binder-free electrode made of MoSe_2 nanosheets directly grown on a Ni foam. Ammonium molybdate and selenium dioxide were added to DI water to make the growth solution. In this work, the ECD process was carried out in a three-electrode configuration with Ag/AgCl as the reference electrode, a Pt sheet as the counter electrode, and a Ni foam as the working electrode. With varying deposition times, the procedure was carried out using an applied voltage of -1.1 V (against Ag/AgCl).⁵⁵ More MoSe_2 nanosheets that were arbitrarily orientated and aligned on the surface of Ni foam grew as the deposition time increased. Raman characterization was done to examine the vibration bands for the prepared nanosheets. The authors concluded that the vibration bands seen in the MoSe_2 nanosheets generated using ECD were in good agreement with those found in earlier MoSe_2 research. The nanosheets were further examined in electrochemical tests for supercapacitor performance analysis.

2.5 Colloidal method

The colloidal technique involves the formation of nanoparticles through a solution-based process, where precursor materials undergo nucleation and growth in a colloidal medium. The process typically follows the classical nucleation and growth model, where the rapid formation of nuclei occurs, followed by

their growth into larger particles. Colloidal synthesis helps for precise control over particle size, shape, and composition by manipulating various parameters such as precursor concentration, temperature, and the use of stabilizing ligands. Colloidal routes to MoSe_2 need suitable reactive precursors to accommodate the highly anisotropic crystal growth. This method has been instrumental in producing various nanomaterials including metals and semiconductors, which find applications in energy storage, catalysis, electronics, medical diagnostics, and photovoltaics. Substrate-free MoSe_2 nanostructures with a consistent flower-like shape and tuneable average diameters ranging from roughly 50 to 250 nm were synthesized colloiddally by Sun and coworkers. Precursors included 1-octylamine, oleic acid, and sodium molybdate. For the nanoflowers, a number of highly crystalline few-layer nanosheets protruded from the central core. The authors showed that by varying the reaction time, the nanoflower sizes could be adjusted. Using XRD and TEM, the authors highlighted the formation of nanoflowers contained with nanosheets. The average crystallite size by Scherrer analysis (10 nm) was found to be smaller than that observed by TEM (50–150 nm), due to which the authors emphasized that the nanosheets are laterally polycrystalline. The distinctive first-order A_{1g} -like, and E_{2g} -like Raman active modes are detected at around 241 and 285 cm^{-1} , respectively, based on the Raman analysis. The authors confirmed that these are in perfect agreement with previous Raman experiments in MoSe_2 .⁵⁶ Guo *et al.* utilized colloidal synthesis to produce MoSe_2 nanonetworks and nanoflowers such as morphologies. $\text{Mo}(\text{CO})_6$, selenium (Se), 1-octadecene (ODE), oleylamine (OAm) and oleic acid (OA) were used as reactants. The authors emphasized that the ratio of OAm to OA determines the shape of MoSe_2 nanostructures. When OAm is not present, the synthesized products are nano-networks made up of ultrathin nanosheets that are continuously coupled in a comparatively flat pattern. The MoSe_2 ultrathin nanosheets tend to create random configurations in various directions as the amount of OA in the reaction increases. They can also form porous nanonetworks or discrete nanoflowers. Such morphology tuning can be further investigated for developing better surface for electrodes.⁵⁷ Wang *et al.* synthesized high-quality MoSe_2 nanospheres from the same route.⁵⁸ The microstructure consisted of uniform nanosphere agglomerates in the range of 200 to 450 nm. BET surface areas for the nanospheres were recorded to be $62.3 \text{ m}^2 \text{ g}^{-1}$. This can be beneficial for developing high-surface area electrodes for SCs.

2.6 Hydrothermal/solvothermal method

The hydrothermal process refers to a chemical synthesis technique that employs aqueous solutions or organic-based solvents. In the hydrothermal/solvothermal process, a prepared and agitated solution is poured into an autoclave and heated in a high-pressure reactor for a predetermined amount of time at a set temperature. The desired sample is eventually produced by this process, which takes place in a high-temperature sealed enclosure within the hydrothermal/solvent system.⁵⁹ This approach provides cost-effectiveness,



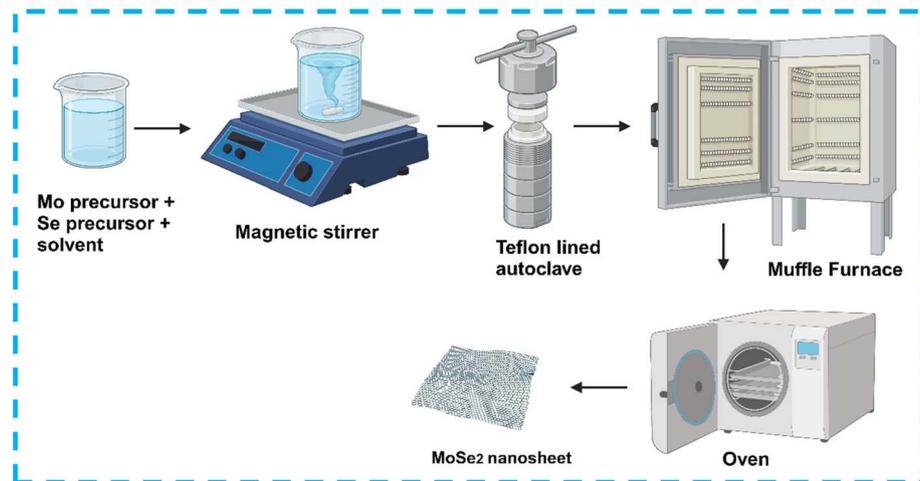


Fig. 6 Hydrothermal/solvothermal synthesis setup. Created in BioRender. FRANCIS, M. (2024) <https://BioRender.com/l67e934>.

ease of control over response conditions, and simplicity in operation. The size and morphology of 2D materials can be efficiently controlled by varying the hydrothermal process parameters, including substrate selection, temperature, additives, and precursor choice.⁶⁰ A schematic of hydrothermal preparation is shown in Fig. 6. Balasingam and coworkers followed a hydrothermal route to prepare a few-layered MoSe₂ nanosheet as an electrode material for SCs.⁶¹ The precursors used for the hydrothermal reaction were sodium molybdate, selenium powder (Se), and NaBH₄. XRD and Raman results confirmed the successful synthesis of MoSe₂ nanosheets. The HRTEM images and Raman spectrum confirmed the formation of few-layered nanosheets. The HR-TEM pictures showed a layered crystal structure with distinct crystal lattice fringes that measure roughly 0.62 nm, which is the usual *d*-spacing for the structure (002) basal plane of the hexagonal crystal structure. Notably, a large surface area for the electrochemical processes is provided by the porous nature of the MoSe₂ nanosheet. Using an aqueous electrolyte, the authors tested the supercapacitive performance of the prepared nanosheets in a two-electrode configuration. Using a simple hydrothermal method, Qiu *et al.* synthesized MoSe₂ nanoflakes and nanorods.⁶² The desired material formation was confirmed by XRD, TEM, and SEM results. According to the authors, the mesoporous structure of the nanoflakes resulted in a large BET specific surface area of 46 m² g⁻¹ and a high porosity of 0.06

cm³ g⁻¹. The authors further developed a solid-state symmetrical SC device of MoSe₂ nanoflakes/MoSe₂ nanorods, showing excellent energy storage capabilities. Ultrathin 1T MoSe₂ nanosheets with an impressive interlayer spacing of ≈1.17 nm were prepared by Jiang *et al.* using a straightforward solvothermal technique.⁶³ The spacing reported was ≈81% wider than that of pure MoSe₂ (0.65 nm). Xia *et al.* prepared nanosheets of 1T@2H MoSe₂ via a two-step solvothermal process, as described in Fig. 7. The crystallinity of the samples was confirmed by various structural characterizations. This work incorporated the 1T phase with 2H MoSe₂ nanosheets.⁶⁴

2.7 Chemical vapor deposition method

The CVD process produces pure and high-crystallinity solid films by depositing gaseous precursors on the substrates. The precursor reacts or decomposes on the surface of the substrate material under high-temperature and -vacuum conditions. To prepare a high-quality film having an adjustable thickness, deposition parameters such as temperature, gas flow, and pressure can be changed.⁵⁹ The precursor selection also plays a vital role in the CVD growth process. The shortcomings of this method are low yield and requirement for complex instruments.

Using Se and MoO₃ as the chemical vapor supply in a mixture of H₂ and Ar gases in a horizontal tube furnace, Shaw *et al.* reported the first CVD development of single- and few-layer

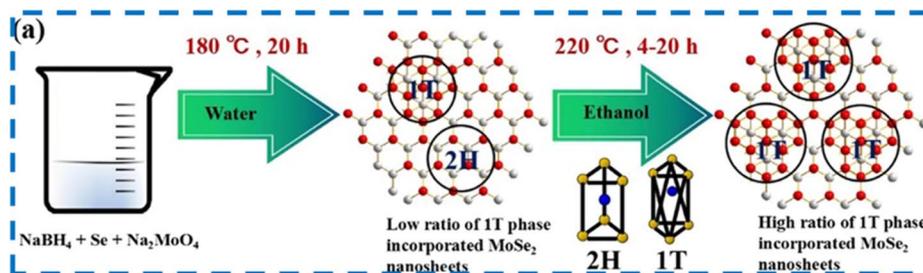


Fig. 7 Solvothermal synthesis of MoSe₂. This figure has been reproduced from ref. 64 with the permission from Nature, copyright 2017.



MoSe₂ nanosheets. It was discovered that H₂ was essential to the reaction because it promoted the reduction of MoO₃ with Se to form MoSe₂. The authors showed that the SiO₂/Si substrates may be used to directly generate extremely crystalline MoSe₂.⁶⁵ A lot of reports are available for the CVD growth of MoSe₂ films and nanosheets. CVD-grown MoSe₂ finds applications in photodetectors, flexible electronics and electrocatalysis. However, the preparation of supercapacitor electrodes from CVD-grown MoSe₂ has not yet been reported. This can be investigated in future to develop pure and crystalline MoSe₂ for energy storage applications.

2.8 Other methods

Upadhyay and Pandey successfully prepared layered 2H-MoSe₂ nanosheets with a stacked nanoplate morphology having a hexagonal crystal structure using an *in situ* selenization process for the first time. Reaction parameters had a significant effect on the phase formation. Stacked plates with porous nanostructures were reported.⁶⁶ Various characterization results for the structural analysis of the prepared nanosheets are shown in Fig. (8–10).

3. SC performance of various MoSe₂ composites

3.1 Asymmetric set-up

MoSe₂/rGO composites for high-performance SCs were developed employing a hydrothermal route. The effect of graphene content was studied by preparing different samples with different rGO concentrations. With the increase in rGO content, MoSe₂ nanospheres were uniformly dispersed on rGO

nanosheets. The composites were checked in a three-electrode setup showing optimal electrode MoSe₂-rGO-25, which was prepared keeping 25 mg GO initially. The rGO nanosheets helped in increasing the electrical conductivity of the sample. Moreover, by using rGO, we get contributions both from EDLC and pseudocapacitive processes. When the rGO content was increased, the diffusion of ions became slow, thereby decreasing performance.⁶⁷ Three-dimensional CoNi₂S₄-graphene-2D-MoSe₂ composites having sphere-sheet-like structures were produced by combining hydrothermal and simple ultrasonication for SC applications. Large amounts of MoSe₂ nanosheets and graphenes were first prepared using a liquid exfoliation technique. For comparison, various composites of CoNi₂S₄ such as CoNi₂S₄-MoSe₂, CoNi₂S₄-G, and CoNi₂S₄-G-MoSe₂ were further synthesized by the same method. The energy storage analysis revealed that the composite exhibited a maximum C_{sp} of 1141 F g⁻¹ with a mass loading of 5 mg cm⁻². The retention of 108% was found after completion of 2000 cycles during the CV and GCD tests. Further, a symmetric device is also built showing a specific capacitance of 109 F g⁻¹.⁶⁸ Vidhya and coworkers synthesized ZnSe@MoSe₂ composites *via* a hydrothermal method having a flower-like morphology. The authors concluded that the flower-like structure increases electrochemical performance by offering electron mobility and additional electrolyte ions. During the CV and GCD tests, the binary electrode delivered high C_{sp} of 450 F g⁻¹ at 1 A g⁻¹ and retained 99.6% after completion of 2000 cycles.⁶⁹ The Ni_{0.85}-Se@MoSe₂ nanosheets were produced by the hydrothermal method on the framework of MoSe₂ using Nickel foam precursors. The Ni_{0.85}Se@MoSe₂ nanosheets exhibited C_{sp} of 774 F g⁻¹, which is twice that of Ni_{0.85}Se and seven times more than

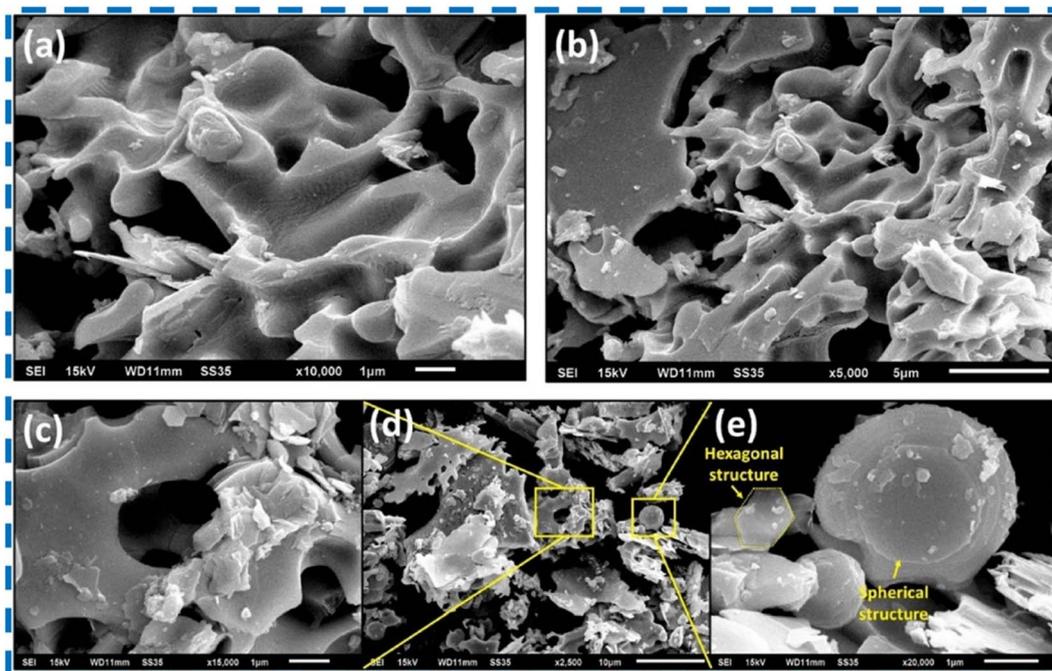


Fig. 8 (a–e) SEM results of the synthesized MoSe₂ powder. This figure has been reproduced from ref. 66 with permission from Elsevier, Copyright 2020.



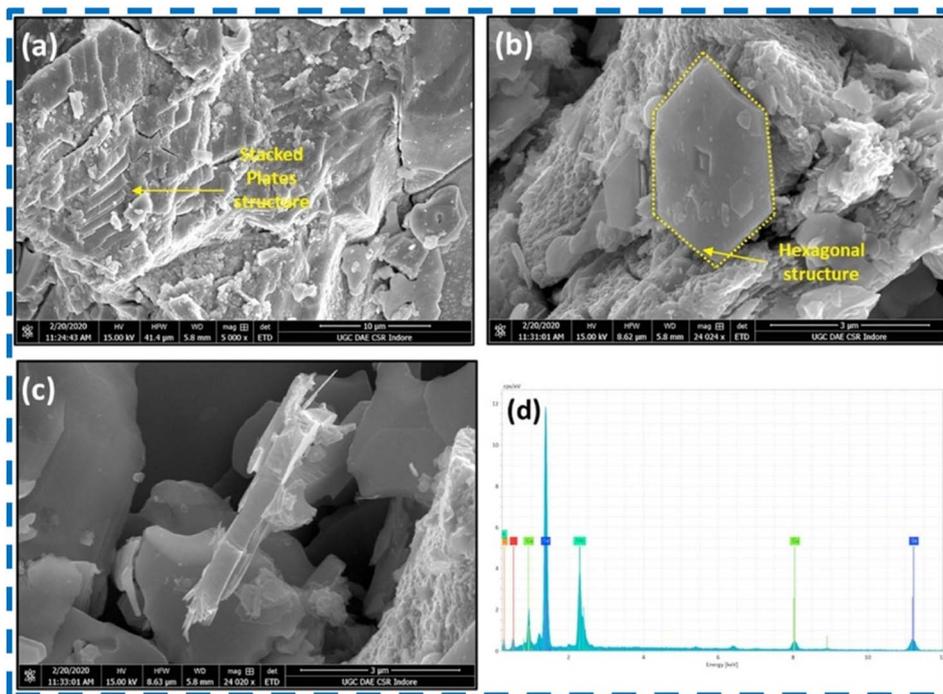


Fig. 9 (a–c) FESEM images (d) EDS of the synthesized MoSe₂ powder. This figure has been reproduced from ref. 66 with permission from Elsevier, Copyright 2020.

that of MoSe₂ nanoparticles. This was associated due to hierarchical and interconnected porous nanosheet array structures. An ASC is fabricated by the Ni_{0.85}Se@MoSe₂ nanosheet electrode, which showed an output of 1.6 V, an E_d of 25.5 W h kg⁻¹ at P_d of 420 W kg⁻¹, and after completion of 5000 cycles it showed good cycling stability having 88% capacitance retention.⁷⁰ Peng and coworkers reported NiSe@MoSe₂ unique nanosheet arrays prepared by a hydrothermal growth method without any surfactants for high-performance ASC applications. Nickel selenide and MoSe₂ are similar in terms of their crystallographic structure due to the same space group. This can be useful for developing a good heterogeneous structure.⁶⁴ NiSe/MoSe₂/MoO₂ ternary composites were reported using a growth-annealing approach. The synergistic effects of MoSe₂, NiSe and MoO₂ worked well to gain good performance of the SC.⁷¹ Gao *et al.* reported NiCoP nanowires decorated with few-layer MoSe₂ nanosheets grown on CC for the first time, employing a hydrothermal method. NiCoOH nanowires on carbon cloth were prepared *via* a hydrothermal reaction followed by low-temperature solid-state phosphorization to form NiCoP. A few layers with an expanded interlayer (from 0.65 nm to 0.76 nm) were seen for the composite. The composite exhibited appreciable long-term durability and at 1 mA cm⁻² gravimetric capacitance of 5613.5 mF cm⁻² (2245.4 F g⁻¹) was recorded. A flexible ASC was constituted using the composite as the positive electrode showing E_d of 55.1 W h kg⁻¹ at P_d of 799.8 W kg⁻¹. After completion of 8000 cycles, it showed 95.8% capacitance retention revealing good cycling stability.⁷² Nanosheets of layered Ti₃C₂T_x MXene/MoSe₂ nanohybrids were hydrothermally synthesized. The authors found out that MXene flakes were enveloped inside MoSe₂ nanosheets. CV and GCD

measurements showed that they possess faradaic pseudocapacitive behavior. The Ti₃C₂T_x/MoSe₂ composite revealed enhanced cycling rate and specific capacitance which were better than those of MXenes and MoSe₂. The authors demonstrated that the observed better performance was due to expanded interlayer spacing of Ti₃C₂T_x by addition of MoSe₂, which enhances the surface area in the electrochemical processes. High cycling ability was associated with the layered structure formation of the composite, preventing agglomeration and stacking issues. After completion of 10 000 cycles, 94.1% capacitance was retained showing good cycling resiliency.⁷³ Using the hydrothermal method, Guo *et al.* demonstrated nanoflower preparation of Na intercalated MoSe₂ based on mixed-phase (1T/2H) MoSe₂/graphene nanocomposites. The transition of MoSe₂ from the semiconductor to the metallic structure was observed after the addition of sodium ions. Na ion insertion causes electrolyte ions to enter the electrode, thereby enhancing the capacitance. The improved conductivity of MoSe₂ was due to the presence of graphene and 1T phase which enhances electrochemical activity. The authors demonstrated that the nanoflower shape, which increased the contact between electrolyte ions and the electrode, were responsible for the composite's increased electroactive area. During the CV and GCD analysis, a specific capacitance of 1407.5 F g⁻¹ was recorded at 1 mV s⁻¹. An all-solid-state SC based on the prepared composite showed a specific capacitance of 244.8 F g⁻¹.⁷⁴ A hydrothermal approach was employed to develop nanohybrids of 2D MoSe₂/Ni(OH)₂, which had superb electrochemical property due to the synergistic effects. The 2D/2D hybrid displayed C_{sp} of 1175 F g⁻¹, which was better than Ni(OH)₂ (933 F g⁻¹) maintaining 85.6% rate capability. The nanohybrid-based



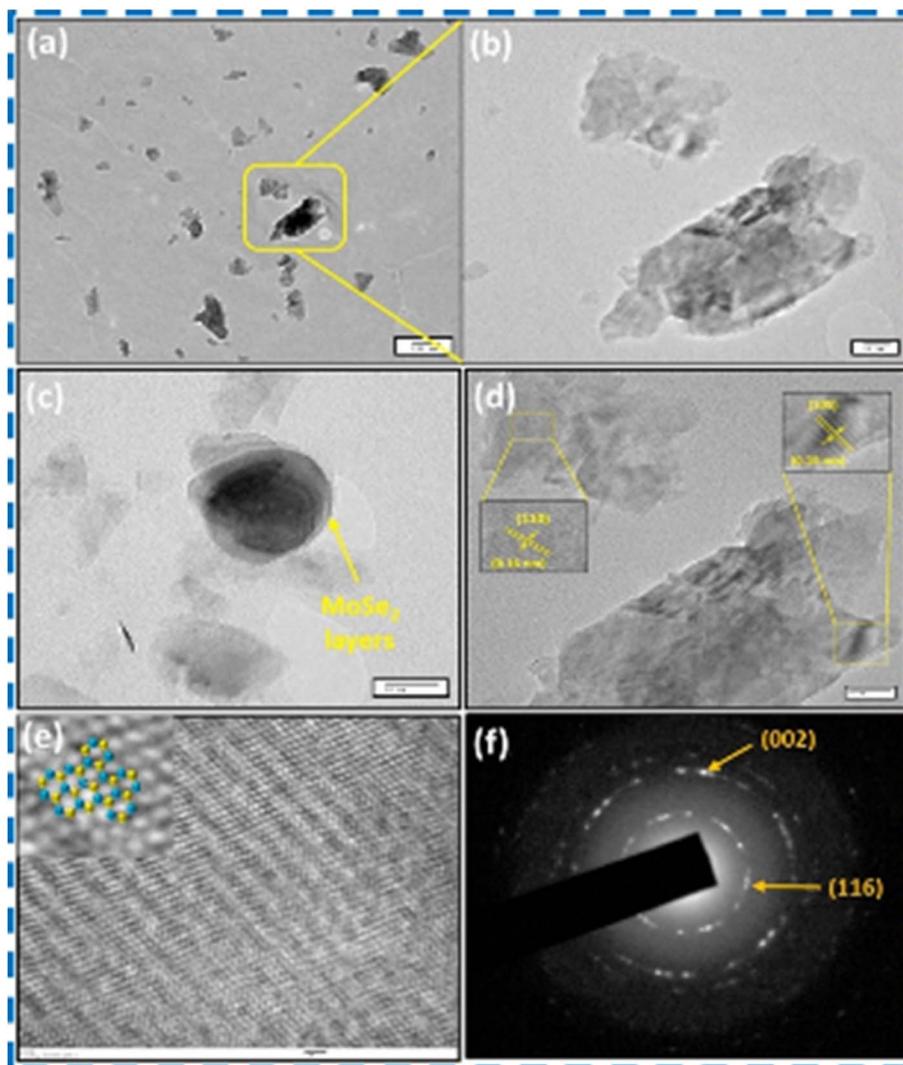


Fig. 10 (a–c) Result of TEM scan, (d) HR-TEM measurements, (e) TEM results (plan view) (f) SEAD measurements of MoSe₂ powder. This figure has been reproduced from ref. 66 with permission from Elsevier, Copyright 2020.

ASC recorded a high E_d value of 43 W h kg⁻¹ and a high P_d value of 8181 W kg⁻¹. Even after the completion of 5000 cycles, 85% of capacitance was retained showing appreciable resiliency. It was used to light small LEDs for few minutes.⁷⁵ Karade and coworkers demonstrated the synthesis of MoSe₂/MWCNT(MSMC) hybrids employing a combination of dip and dry along with chemical bath deposition. The authors in this work compared the energy storage performances between MoSe₂, MWCNTs and MSMC using a 1 M KOH electrolyte.⁷⁶ Yu *et al.* reported Ni_{0.85}Se/N–MoSe₂ hybrids for asymmetric supercapacitors *via* a hydrothermal route, using water and formamide as a mixed solvent. The hybrid had a better specific capacity than that of the individual units, which was due to the cooperative effect and mesoporous structure. The electrochemical performance of the hybrid demonstrated good cyclability, retaining 105.1% of capacitance after completion of 15 000 cycles.⁷⁷ Wei *et al.*, for the first time, successfully designed 3D MoSe₂ nanoflowers on an anisotropic carbon architecture. Most of the studies on MoSe₂ are focused on cathodes. Here,

they prepared composites for anodes. High pseudo capacitance was observed due to good specific surface area of 83.5 m² g⁻¹ and high electrochemical activity of the nanoflowers. Contribution from both EDLC and pseudocapacitance is observed, which produces synergistic effects and helps to provide good areal capacitance.⁷⁸ A hydrothermally prepared MoSe₂/Ni foam showed a specific capacitance of 1114 F g⁻¹ in the works of Huang and team.⁷⁹ Improvement in the specific capacitance was observed when composites are formed with carbon materials such as graphene and acetylene black. MoSe₂/graphene showed a specific capacitance of 1422 F g⁻¹ and MoSe₂/acetylene black showed 2020 F g⁻¹.^{80,81} A self-charging power cell was fabricated making use of MoSe₂ electrodes. The reported better performance was due to the use of ionogel electrolytes and intercalative-type MoSe₂ energy storing electrodes. The cell exhibited 18.93 mF cm⁻² specific capacitance, 37.90 mJ cm⁻² energy density and 268.91 μW cm⁻² power density.⁸² Arulkumar *et al.* reported Ti₃C₂T_x MXene/MoSe₂ nanocomposites for ASC prepared *via* a hydrothermal approach. A bare Ni foam has little



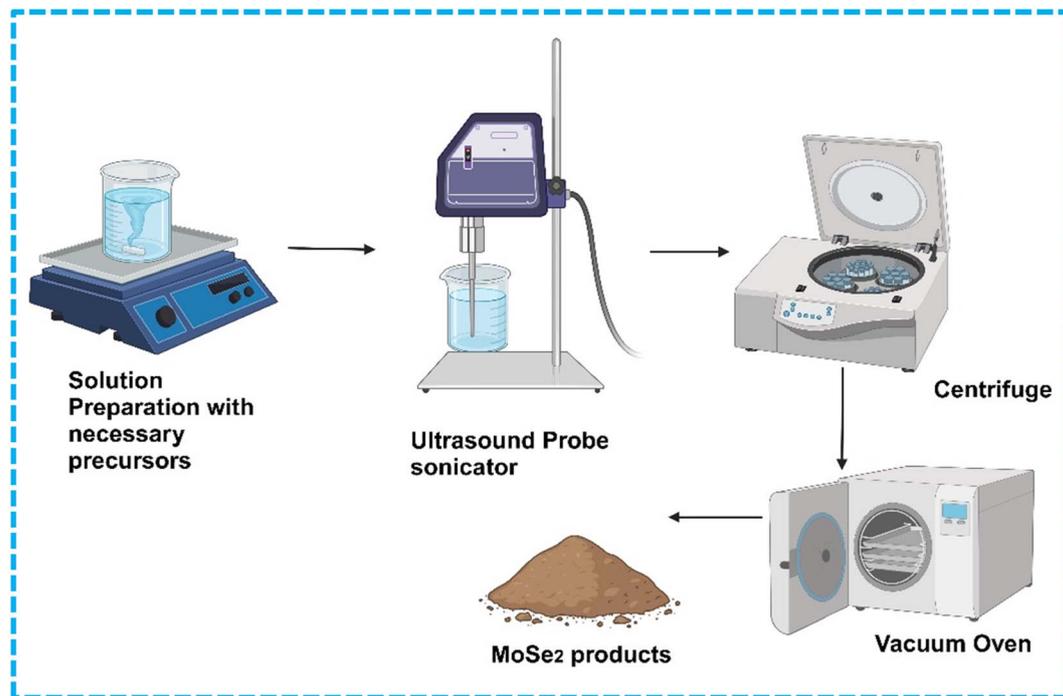


Fig. 11 Sonochemical method for MoSe₂ preparation. Created in BioRender. FRANCIS, M. (2024) <https://BioRender.com/c76w095>.

capacitance, as indicated by a flat line on the CV graph. The composite has better capacity than that of individual elements, which can be inferred from the larger area coverage in the CV plots. The diffusion control contribution slowly declines and pseudocapacitive contribution slowly increases with the increase in scan speeds. The GCD plots are non-linear and symmetrical in nature supporting the redox peaks shown in the CV plots, thereby confirming the pseudocapacitance nature. The composite exhibits the highest specific capacitance of 1531.2 F g⁻¹ at 1 A g⁻¹. An ASC device was built using the composites as one of the electrodes. Good efficiency was reported which can be ascertained from the symmetric GCD profile and very negligible IR drops even when increasing current densities. The authors concluded that MXenes and MoSe₂ work harmoniously to produce good performance. All test results are shown in Fig. 12–14.⁸³

Studies on the effect of alkaline electrolyte concentration (0.5 M, 1 M, 3 M KOH) on energy storage of MoSe₂/PANI were reported by Zheng and team. The samples were hydrothermally prepared using silica templates. MoSe₂ hollow microspheres with rough morphology were formed, which were in sizes of 740 nm contributing to the improvement in surface area. From BET analysis, it was found that the surface areas increase in the order of PANI > MoSe₂/PANI > MoSe₂. MoSe₂/PANI with a mass ratio of 1/1 recorded 146.5 F g⁻¹ as the highest specific capacitance at 0.3 A g⁻¹. It also showed 22.16 W h kg⁻¹ energy density with 198 W kg⁻¹ power density. The authors concluded that good performance of MoSe₂/PANI was attributed to the synergistic effect of PANI and MoSe₂ hollow microspheres, improved conductive path, suitable electrolyte concentration, variable oxidation state, and large surface area of the material.⁸⁴ Mittal

and co-workers for the first time prepared MoSe₂/PANI nanocomposites and studied its electrochemical performances for supercapacitor applications. The optimized MoSe₂/PANI having a ratio of 1/2 showed 96% enhancement in capacitance. Testing for 3000 cycles, the composite showed capacitance retention of 72% showing good cycling stability. MoSe₂ performs like an EDLC material and PANI falls under a pseudo capacitance material. Forming their composite will utilize the benefits of both by synergistic effects, thereby improving the performance. Moreover, other benefits such as improved electron and ion transfer, and improved wettability of electrode were observed.⁸⁵ Zhang *et al.* prepared MoSe₂ porous nanospheres by a hydrothermal method. Nanosheets of PANI were grown *in situ*, and thereafter, MoSe₂/PANI capsule nanospheres (CNs) were synthesized. The authors noted that the composite's performance was dependent on the quantity of PANI. The best composition was found to be MoSe₂/PANI-16 CNs, which recorded a specific capacitance of 753.2 F g⁻¹. It was found that pseudocapacitance contribution dominated more than diffusion capacitance. Further the ASC device fabricated using the composites showed energy density of 20.1 W h kg⁻¹ at 650 W kg⁻¹ power density. Surface areas of MoSe₂ and MoSe₂/PANI CNs were noted to be 5.94 m² g⁻¹ and 44.49 m² g⁻¹ respectively. A seven-fold increment in specific surface area was seen due to the formation of composites. The composites had a good mesoporous structure, which can be confirmed by observing the pore size of MoSe₂ (21.15 nm) and MoSe₂/PANI (8.66 nm). The authors reported that the pore size decreased after adding PANI.⁸⁶ Sukanya *et al.* constructed an asymmetric supercapacitor based on the ternary heterostructure composite of YSe₂/MoSe₂/Ni_xB.⁸⁷ The electrode made from the prepared



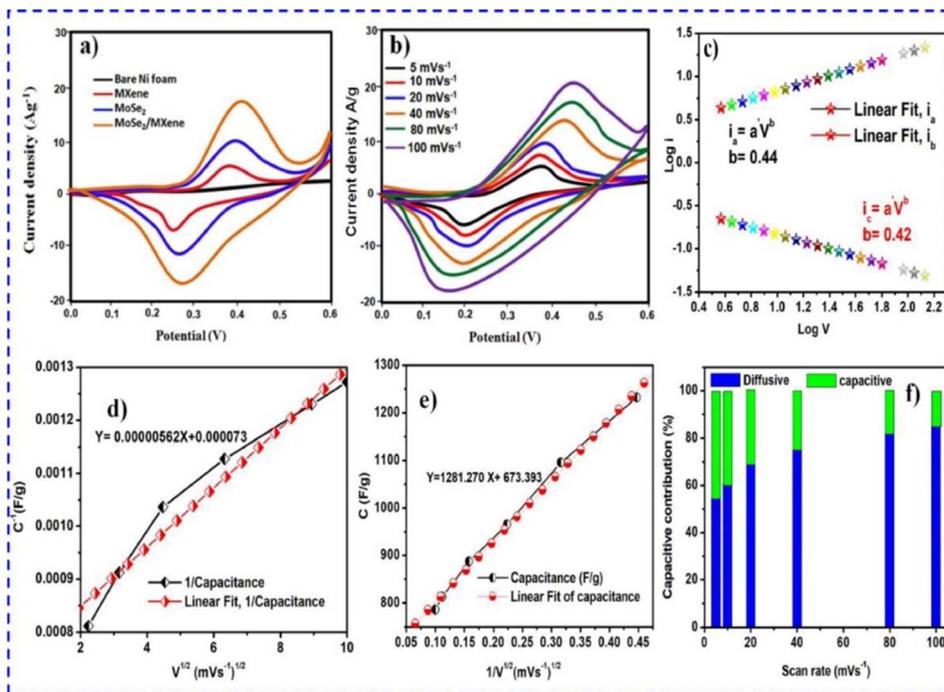


Fig. 12 (a) Cyclic voltammetry results of $\text{Ti}_3\text{C}_2\text{T}_x$ (MXene), MoSe_2 , and composite of MXene/ MoSe_2 . (b) Cyclic voltammetry results of the composite at different scanning rates. (c) Display of the b -value of the composite. (d) Illustration of the total capacitance of the composite. (e) Capacitive contribution of the composite. (f) Diffusive and capacitive contributions of the composite. This figure has been reproduced from ref. 83 with permission from Elsevier, Copyright 2023.

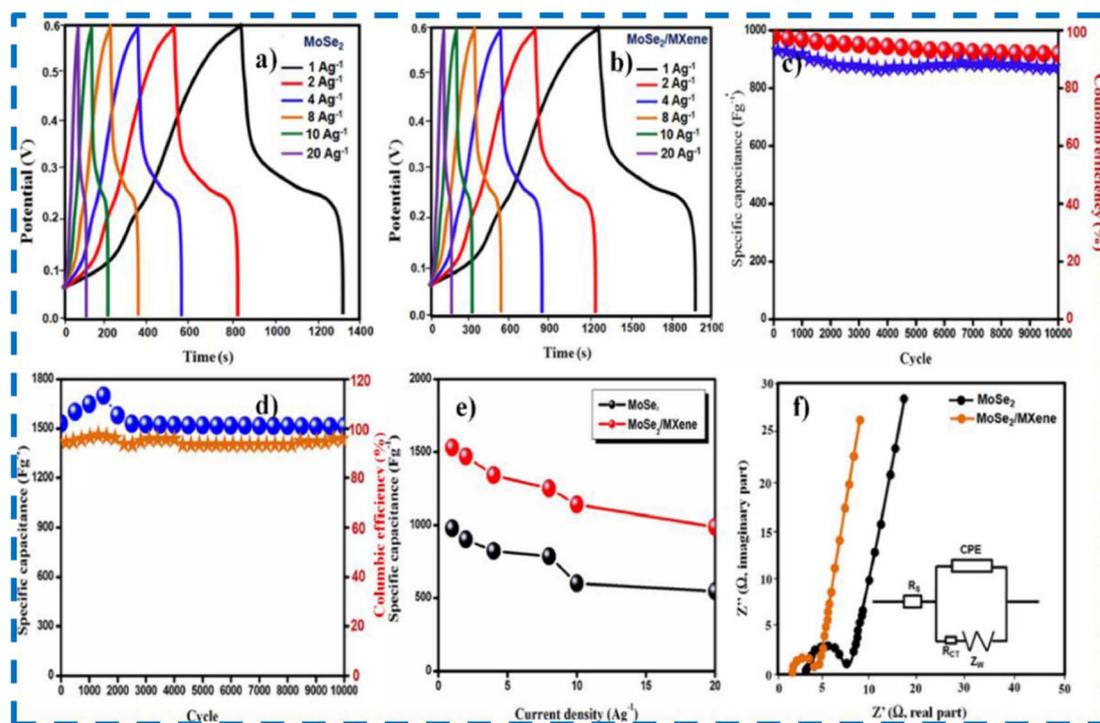


Fig. 13 (a and b) Galvanostatic charging and discharging profile of MoSe_2 and the composite of MXene/ MoSe_2 at 1–20 A g^{-1} (c and d) Display of stability and efficiency results for MoSe_2 and the composite. (e) Display of specific capacitance of MoSe_2 and composite at 5 mV s^{-1} to 100 mV s^{-1} (f) EIS measurements of MoSe_2 and the composite with the corresponding equivalent circuit. This figure has been reproduced from ref. 83 with permission from Elsevier, Copyright 2023.



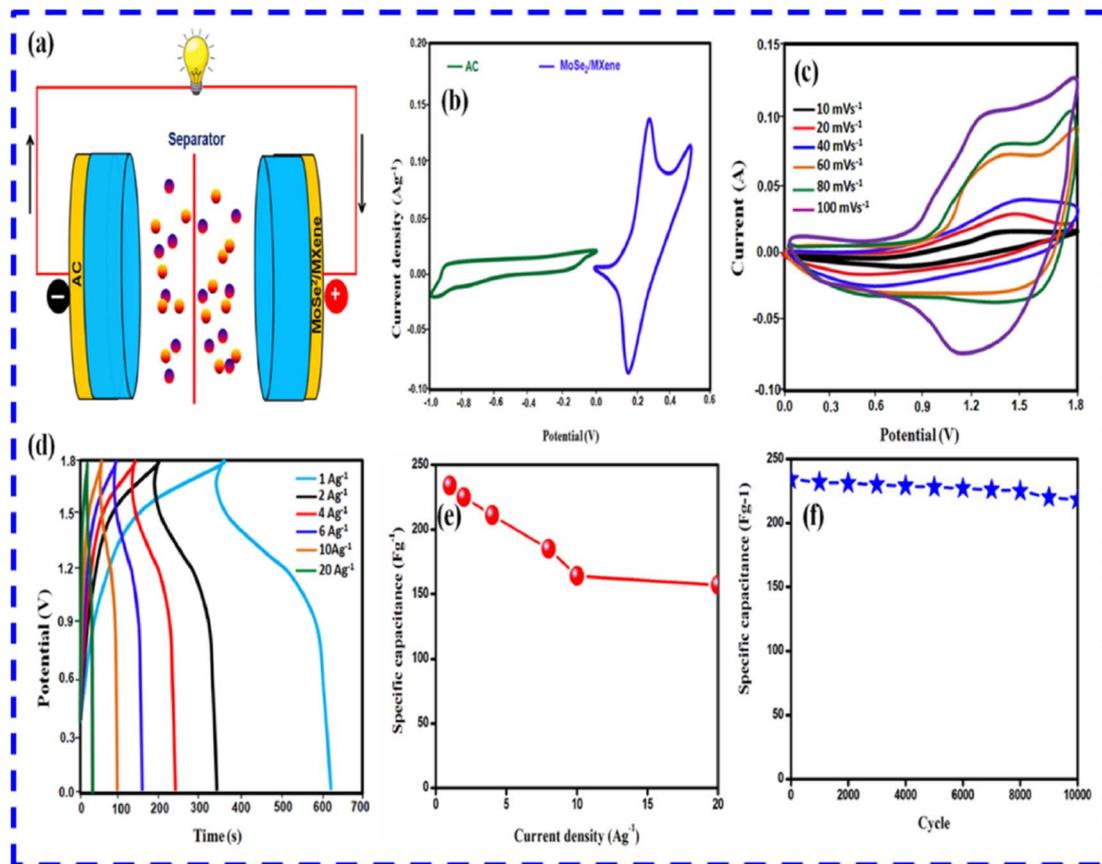


Fig. 14 (a) Illustration of the assembled asymmetric device. (b) Cyclic Voltammetry (CV) results of the composite and AC. (c) Cyclic Voltammetry (CV) results of the device. (d) Display of GCD results of the device. (e) Specific capacitance versus current density plots for the device. (f) Stability analysis of device at 20 A g⁻¹. This figure has been reproduced from ref. 83 with permission from Elsevier, Copyright 2023.

composite showed 893.3 F g⁻¹ as the specific capacitance and 128.17% retention after the completion of more than 5000 cycles. At 800 W kg⁻¹, the ASC device exhibited 39.5 W h kg⁻¹. Masanta and coworkers studied Mn doping on MoSe₂ nanostructures. Compared to pristine MoSe₂, 313% increment in specific capacitance was recorded when 6.2% Mn was doped.⁸⁸ Tanwar *et al.* fabricated activated carbon-coated MoSe₂ nanocomposites prepared *via* a hydrothermal method for high-performance aqueous supercapacitors of asymmetric type. Around 1 mg of active material was coated on the Ni foam during the tests.⁸⁹ Using a hydrothermal method, different noble metals were also explored for MoSe₂ composites. The authors concluded that, among them, MoSe₂-Au showed the highest specific capacitance. It was 1.5 times larger in comparison to bare MoSe₂ nanosheets.⁹⁰ Du *et al.* employing a hydrothermal method prepared nanoflowered porous MoSe₂/N-doped carbon, which was utilized as the anode material in potassium-ion capacitors.⁹¹ Lattice plane expansion is achieved by the authors. From BET analysis, it was found that the specific surface area was 228.85 m² g⁻¹ and the pore size was 3.91 nm. Nitrogen doping and porous carbon enhanced the structural solidity and conductivity of MoSe₂, accelerating the reaction kinetics and promoting the transmission of electrons. Recently, Zhu *et al.* have studied the variation of morphologies of Molybdenum selenides by controlling the Mo : Se ratio.⁹² MoSe_x

was developed on a Ni foam by an electrodeposition process and tests were done in a three-electrode setup. SEM characterization showed that the morphologies were influenced by a Mo/Se molar ratio. The authors deduced that compared to MoSe-12, the MoSe-21 and MoSe-11 compositions had better ion storage capacity. An annealing strategy was employed to improve the overall performance. He *et al.* achieved mixed-phase 3D MoSe₂-NiSe/NF electrodes by controlling the hydrothermal temperature. An electroactive area of 50.84 m² g⁻¹ with a mean pore size of 10.7 nm was found using BET analysis. 2H and (2H-1T) mixed phases were observed, which had interconnected wrinkled nanosheets. Compared to individual MoSe₂ and NiSe, the composite had a good 3D network structure on the surface and all the available active materials were utilized to the full extent.⁹³ Wang and team achieved 2H semiconducting to 1T metallic phase transition in MoSe₂ by tungsten doping at a high temperature.⁹⁴ Yang *et al.* investigated heterostructures of MoSe₂/Bi₂Se₃ for supercapacitive performances. The MoSe₂/Bi₂Se₃ hybrids possess enhanced electrochemical properties due to large surface areas, more redox active sites, and better electron transportation. The authors showed that the hybrids demonstrated better capacitive performances than those of individual Bi₂Se₃ and MoSe₂.⁹⁵ Balasingam *et al.* developed MoSe₂/rGO nanosheets for supercapacitor applications. The nanosheet composites were



developed *via* a hydrothermal route. During CV and GCD tests, a specific capacitance of 211 F g^{-1} was recorded, and 180% capacitance retention was noted after the completion of 10 000 cycles. The authors concluded that these results are better relative to pristine MoSe_2 . This may be due to contributions from EDLC and pseudo capacitances. The hybrid nanostructure has excellent electrochemical performance due to more electroactive sites, better electrical conductivity, small diffusion path length and fast charge transport.⁹⁶ Vidhya *et al.* reported $\text{CuSe}_2@/\text{MoSe}_2$ (CMS) binary composite nanorods for ASC applications, which showed an improved energy density of 113 W h kg^{-1} and showed 98.1% retention after completion of 2000 cycles.⁹⁷

3.2 Symmetric set-up

Upadhyay and Pandey fabricated a symmetric supercapacitor (SSC) based on 2H MoSe_2 nanosheets using Whatman paper/KOH as a solid electrolyte. The Cyclic voltammetry tests were conducted at a scanning speed of 100 mV s^{-1} . GCD tests were conducted at 1 A g^{-1} . The authors revealed that the SSC can operate in the 0–0.6 V voltage window. Above 0.6 V, the CV curves no longer maintained rectangular shapes. The SSC had good capacitive property, which can be ascertained from the CV

curves which do not show many variations when the scanning rates are slowly increased. During higher scan rates around 0.3–0.4 V redox peaks were seen in the CV curves (Fig. 15), which is an indication of pseudocapacitive nature. The GCD curves were almost symmetrical throughout the tests, indicating that SSC has high reversibility. High E_d of $184.5 \text{ mW h kg}^{-1}$ can be achieved at P_d of 74.6 mW kg^{-1} at 0.5 A g^{-1} . Then $22.6 \text{ mW h kg}^{-1}$ of energy density was maintained at high power density (155.6 mW kg^{-1}), which was a proof that the SSC has a good rate capability. The highest specific capacitance was found to be equal to 4.1 F g^{-1} at 0.5 A g^{-1} . High cyclability with a retention of 105% even after completing 10 000 cycles and 98% of coulombic efficiency were shown after the first cycle. Then 100% increment in efficiency was seen after 10 000 cycles. Good structural stability can be confirmed due to small increment of 0.05Ω in the ESR after 10 000 cycles. The charge transfer resistance also got decreased by 0.8Ω , indicating good transportation of the electrolyte ions. The authors concluded that the results support the suitability of MoSe_2 for supercapacitors.⁶⁶ All-solid-state symmetrical SCs based on MoSe_2 /graphene nanocomposites had modification in the lattice structure when a small amount of W ($\approx 5\%$) was inserted into the composite. The characteristics of W-doped MoSe_2 /graphene were evaluated using DFT calculations. Low K^+ adsorption energy and a large

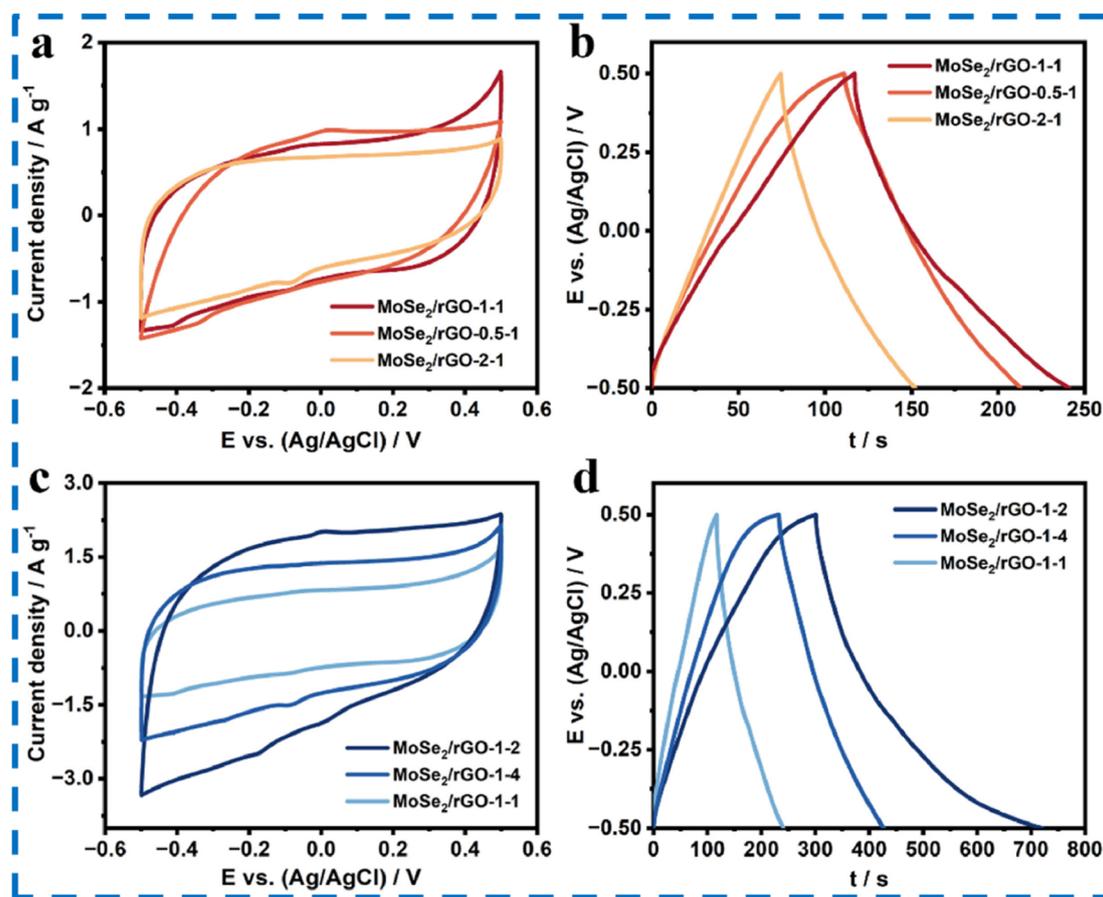


Fig. 15 (a and b) CV and GCD plots of MoSe_2/rGO at different concentrations of graphene oxide. (c and d) CV and GCD plots of MoSe_2/rGO at different Se/Mo ratios keeping the optimum graphene oxide concentration as 1 mg mL^{-1} . This figure has been reproduced from ref. 98 with permission from Elsevier, Copyright 2023.



interlayer distance were reported by the authors. The 444.4 mF cm^{-2} as specific capacity was achieved, and after 5000 cycles, 81.3% of capacitance was retained, thereby showing good stability.⁹⁹ Tanwar *et al.* reported MoSe_2/AC nanocomposites for SC applications *via* a hydrothermal strategy. A surface area of $522 \text{ m}^2 \text{ g}^{-1}$ with 4.6 nm mean pore size was found using BET analysis. Highest specific capacity was shown by the M@AC 1 : 5 electrode when compared with all samples. An aqueous symmetric cell using the electrode was fabricated showing 39.4 W h kg^{-1} energy density and 704.5 W kg^{-1} power density. Testing for 10 000 cycles the composite showed appreciable cyclability, and the coulombic efficiency was nearly 100%. When energy from this symmetric cell was applied for a small LED device, it could glow for around 22 min.¹⁰⁰ Kichi *et al.* studied electrochemical performances of layered MoSe_2 and selenium-rich MoSe_2 by the optimization of reaction parameters such as molar ratio of precursors, temperature and duration of reaction. The supercapacitor based on the MoSe_2 electrodes recorded C_{sp} of 122.66 F g^{-1} . The Se-rich MoSe_2 -based supercapacitor recorded C_{sp} as 19 F g^{-1} , which was less than the previous MoSe_2 sample. The authors concluded that this decrease can be due to unreacted selenium, which reduces the surface area.¹⁰¹ Tanwar and coworkers investigated electrochemical performances of different shaped transition metal selenides. They prepared nanoflowers of MoSe_2 *via* a hydrothermal method and found out that the nanoflower morphology showed better performance than CoSe_2 nanoneedles and NiSe_2 nanospheres. A symmetric supercapacitor was constructed using prepared MoSe_2 , which displayed a maximum specific capacitance of 154 F g^{-1} during the energy storage tests.¹⁰² $\text{MoSe}_2/\text{FeOOH}$ composite formation between nanorods of iron oxide-hydroxide and nanoflowers of molybdenum diselenide

using a chemical blending technique *via* a hydrothermal route was reported by Tanwar and team. A symmetric cell was constructed recording a specific capacitance of 132 F g^{-1} keeping the voltage as 1 V. It shows capacitance retention of 100% after completion of 3000 cycles and after 10 000 cycles 100% efficiency was maintained. It provides 18.3 W h kg^{-1} energy density at 1174 W kg^{-1} power density. The average length of 546 nm and diameter of 80 nm was estimated for the FeOOH nanorods. The Ostwald ripening process was carried out, which influenced the rod and nanoflower morphology of the grown sample. The authors revealed that nanoflowers covering nanorods can be a suitable morphology for effective charge storage.¹⁰³ Shui *et al.* combined phase and interface engineering approaches to improve the energy storage ability of heterogeneous MoSe_2/rGO composites. The authors showed that the intercalation of Na^+ ions can cause phase conversion from 2H (hexagonal) semi-conducting to 1T (octahedral) metallic. The authors studied the effect of the molar ratio (Se : Mo) and GO concentration on Na^+ intercalation. The authors found out that among all compositions the best was $\text{MoSe}_2/\text{rGO-1-2}$. The CV and GCD test results of $\text{MoSe}_2/\text{rGO-1-2}$ electrode are provided in Fig. 16. An ASSC (all solid-state symmetric SC) was developed based on the prepared composite. The prepared composite electrode revealed a high C_{sp} of 169.3 F g^{-1} at 0.5 A g^{-1} . The assembled ASSC device had energy density (4.88 W h kg^{-1}) and good cyclability (83.1% over 10 000 cycles). The device can also be a power source to glowing small LEDs for 70 s.⁹⁸ The various test results for the ASSC are shown in Fig. 17.

Tanwar and team studied aging impact of Se powders on morphology and electrochemical performances of MoSe_2 composites. Se powder is subjected to aging for several days in hydrazine hydrate as a reducing agent. The MoSe_2/AC

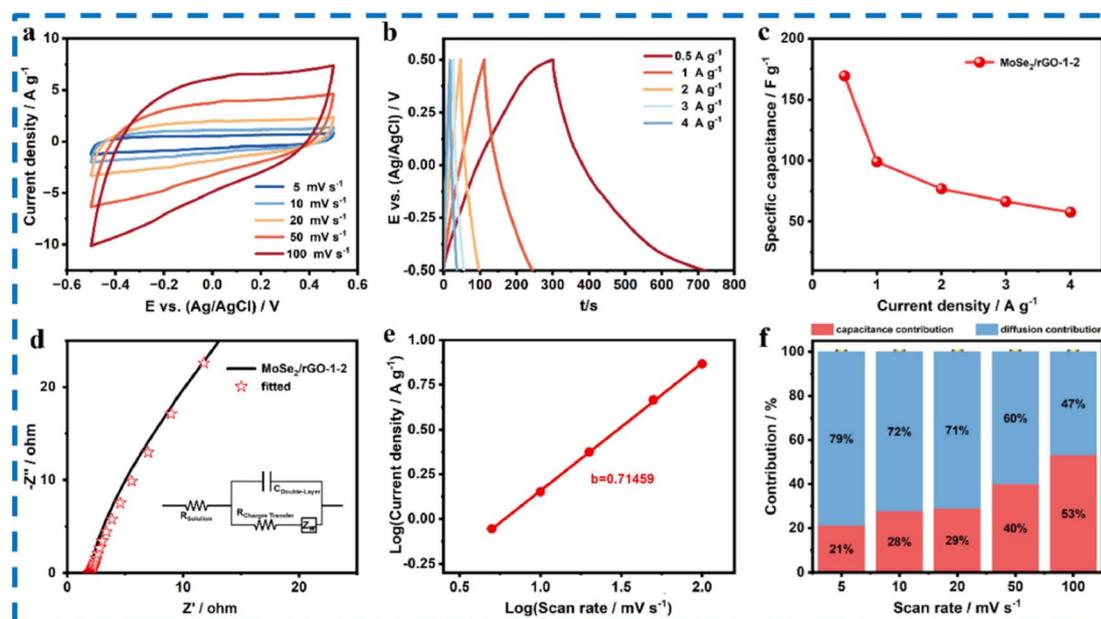


Fig. 16 (a) CV plot of the $\text{MoSe}_2/\text{rGO-1-2}$ electrode at different scanning speeds. (b) CV plots of the $\text{MoSe}_2/\text{rGO-1-2}$ electrode at different current densities. (c) Specific capacitance measurements. (d) Nyquist plots along with fitted curve. (e) Log peak current versus log scan rate. (f) Diffusion and capacitance contribution at different scanning speeds. This figure has been reproduced from ref. 98 with permission from Elsevier, Copyright 2023.



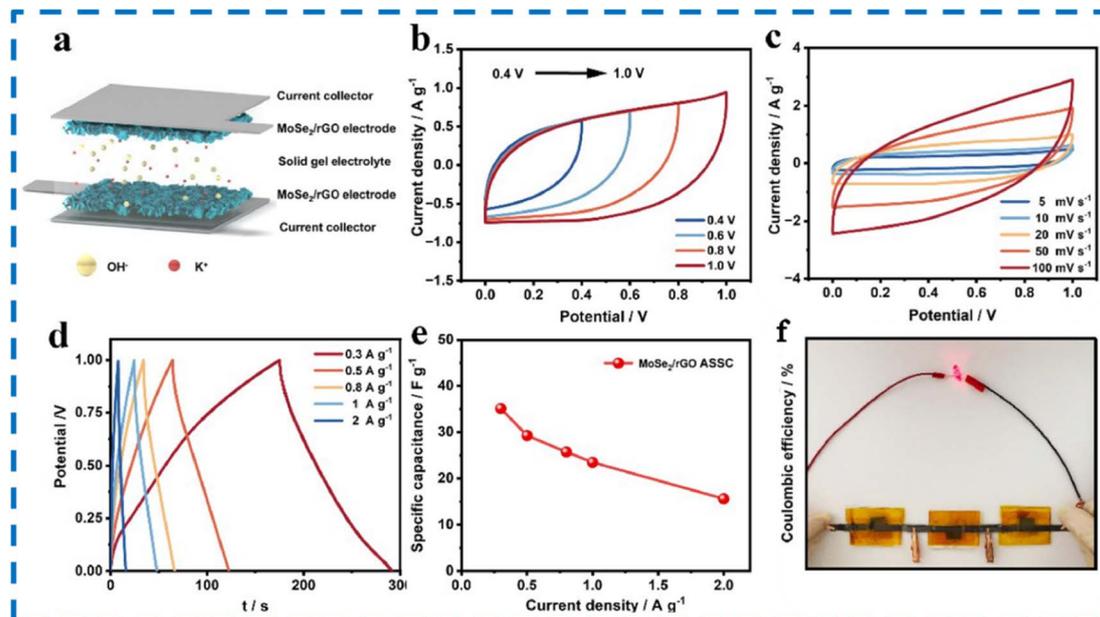


Fig. 17 (a) Schematic of the assembled MoSe_2/rGO ASSC device (b) CV plots with varying potential windows (c) CV plots at different scanning speeds. (d) GCD plots at different current densities. (e) Specific capacitance evaluated from GCD (f) few serially connected ASSC devices to glow a small LED. This figure has been reproduced from ref. 98 with permission from Elsevier, Copyright 2023.

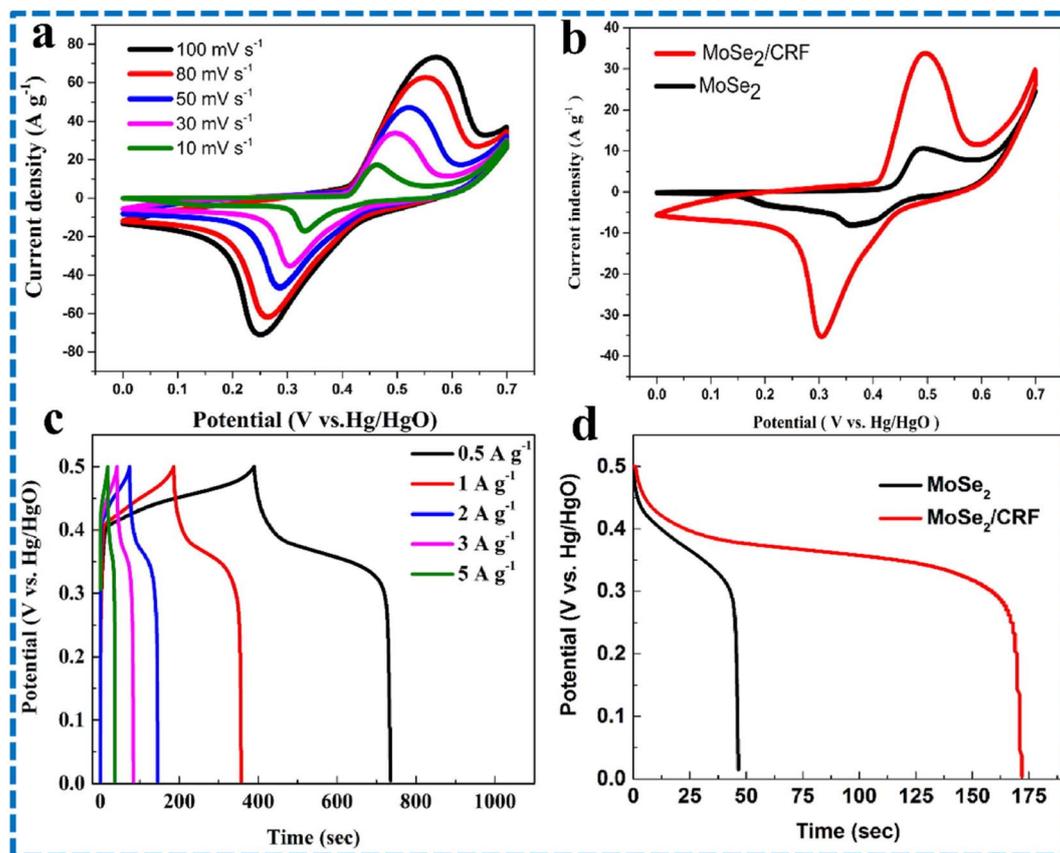


Fig. 18 (a) CV plot of the MoSe_2/CRF composite at different scan speeds, (b) CV plot of MoSe_2 and the composite at 30 mV s^{-1} , (c) GCD curves of the composite at different current densities, (d) discharging profiles for MoSe_2 and the composite electrode at 1 A g^{-1} . This figure has been reproduced from ref. 105 with permission from Elsevier, Copyright 2021.



composite was prepared *via* a hydrothermal route. Compared to the remaining samples, the six days aged sample showed superior performance. With the help of the fabricated symmetric cell, a few small LEDs can be illuminated.¹⁰⁴ The synthesis of ultrathin MoSe₂@carbon aerogel microspheres employing a solvent thermal method was reported by Han and coworkers. Carbon aerogels were derived from resorcinol-formaldehyde (RF) aerogels. Due to composite formation with RF, the surface area and the conductivity were increased. Various test results of the MoSe₂/CRF composite and MoSe₂ are shown in Fig. 18. CV tests were conducted at different scanning speeds employing a three-electrode system. The occurrence of faradaic reactions and the pseudocapacitive nature of the composite were confirmed by looking into the peaks in the CV plots. The peaks become bigger on increasing the scan speeds. Compared to MoSe₂, the composite MoSe₂/CRF electrode demonstrated larger area in the CV plot, showing that it has more capacitive properties. The pseudocapacitive characteristics of the composite were further supported by the appearance of peaks in the GCD plots. The charging and discharging profiles are almost symmetric at different current densities, confirming that the redox reactions had excellent reversibility. The composite electrode outlasted the MoSe₂ electrode in terms of discharge time, aligning with the trends observed in the CV curves, thereby demonstrating that the composite has better specific capacitance as shown in Fig. 18. The electrode's specific

capacitance diminished as the scanning speed increased. A symmetric supercapacitor is also developed using the composites and its performances are checked. Different test results for symmetric SCs based on MoSe₂/CRF are shown in Fig. 19. The current density peaks increased with the increase in scanning speeds revealing the good capacitive behaviour of the symmetric SC. Linear and triangular responses in GCD plots for different current densities indicate high-rate capability and superb charge transportation. After the completion of 2500 GCD cycles, the symmetric SC retained 120.8% capacitance, indicating the excellent cyclability and stability. The authors demonstrated that the primary cause of the composite's overall enhanced performance was the presence of porous CRF microspheres.¹⁰⁵ Various MoSe₂ composite electrodes which are reported until now are shown in Table 1 along with their performances (Fig. 20).

4. Machine learning for supercapacitors

Machine Learning (ML) algorithms can realize their potential in designing and optimizing electrode materials for supercapacitors. Deep learning, artificial intelligence, and data science methodologies will be able to identify the optimal electrode material specific to a particular application with

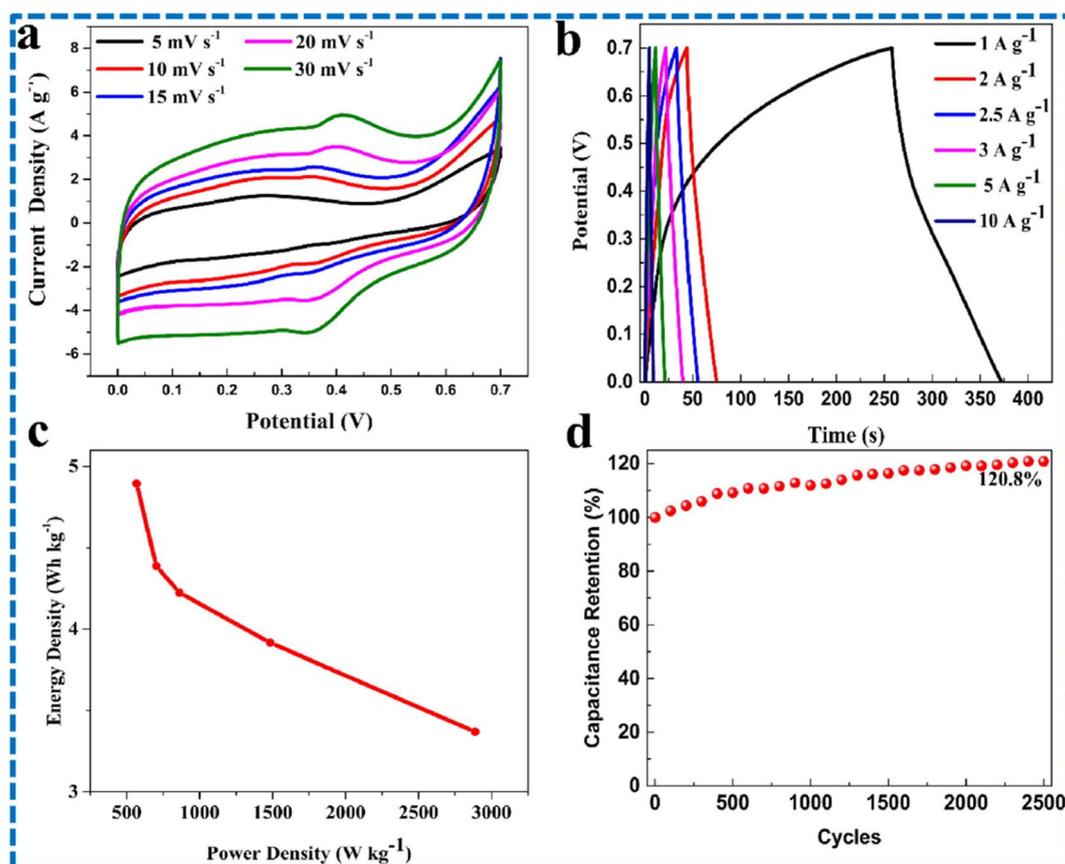


Fig. 19 Symmetric SC performance of MoSe₂/CRF: (a) CV plots at 5–30 mV s⁻¹, (b) GCD plots at 1–10 A g⁻¹, (c) Ragone plots and (d) stability analysis. This figure has been reproduced from ref. 105 with permission from Elsevier, Copyright 2021.





Table 1 MoSe₂ composites with supercapacitor performances

Sl. no	Electrode	Synthesis	Morphology/structure	Electrolyte	Specific capacitance	Retention (%) after (cycles)	Ref.
1	MoSe ₂	Liquid phase exfoliation	Nanosheets	3 M KCL	15 F g ⁻¹ at 0.1 A g ⁻¹	95% (12)	51
2	2H-MoSe ₂	<i>In situ</i> selenization method	MoSe ₂ nanosheets	2 M KOH	46.22 mA h g ⁻¹ at 2 A g ⁻¹	64% (2000)	66
3	MoSe ₂	Microwave irradiation process	Dry leaf like mesoporous MoSe ₂	0.5 M H ₂ SO ₄	257.3 F g ⁻¹ at 1 A g ⁻¹	95% (5000)	53
4	MoSe ₂	Electrodeposition method	MoSe ₂ sheets on Ni foam	1 M Na ₂ SO ₄	325.92 mA h g ⁻¹ at 4 mA cm ⁻²	80% (1000)	55
5	MoSe ₂	Solvothermal	Nanosheets	3 M KOH	272 F g ⁻¹ at 1 A g ⁻¹	—	106
6	MoSe ₂	Hydrothermal	Few-layered MoSe ₂ nanosheets	0.5 M H ₂ SO ₄	198.9 F g ⁻¹ at 2 mV s ⁻¹	75% (10 000)	61
7	MoSe ₂	Solid reaction, <i>in situ</i> selenization	Nanoflakes	1 M NaOH	25 mA h g ⁻¹ at 1 A g ⁻¹	—	107
8	MoSe ₂ @AC symmetric cell	Hydrothermal	MoSe ₂ nanosheets coated with activated carbon sphere	6 M KOH	394 F g ⁻¹ at 1 A g ⁻¹	85% (3000)	104
9	MoSe ₂ @AC	Hydrothermal	Nanoflower	6 M KOH	514 F g ⁻¹ at 10 mV s ⁻¹	68% (2500)	100
10	MoSe ₂ /C	Solvothermal	Interconnected MoSe ₂ nanosheets and amorphous carbon	3 M KOH	878 F g ⁻¹ at 1 A g ⁻¹	98% (2000)	108
11	Bare MoSe ₂	Solvothermal	MoSe ₂ nanosheets	3 M KOH	436.7 F g ⁻¹ at 1 A g ⁻¹	72% (2000)	67
12	MoSe ₂ /rGO	Hydrothermal	MoSe ₂ nanosheets spheres on rGO nanosheets	2 M KOH	814.4 F g ⁻¹ at 1 A g ⁻¹	—	96
13	MoSe ₂ /rGO symmetric set-up	Hydrothermal	MoSe ₂ nanosheets with rGO nanosheets	0.5 M H ₂ SO ₄	211 F g ⁻¹ at 5 mV s ⁻¹	180% (10 000)	98
14	MoSe ₂ /carbon aerogel	Solvent-thermal method	MoSe ₂ nanosheets grown on rGO into carbon aerogel microspheres	1 M KOH	35.1 F g ⁻¹ at 0.3 A g ⁻¹	83.1% (10 000)	105
15	MoSe ₂ /CA	Hydrothermal	Carbon aerogel embedded in ultrathin MoSe ₂ nanosheets	6 M KOH	498 F g ⁻¹ at 1 A g ⁻¹	127.2% (2500)	109
16	Ni _{0.85} Se@MoSe ₂	Hydrothermal	Carbon aerogel embedded in ultrathin MoSe ₂ nanosheets	6 M KOH	775 C g ⁻¹ at 1 A g ⁻¹	98% (1500)	70
17	Ni _{0.85} Se/N-MoSe ₂ //AC	Hydrothermal	Ulathin nanosheet arrays	2 M KOH	774 F g ⁻¹ at 1 A g ⁻¹	95% (1000)	77
18	NiSe ₂ @MoSe ₂	Hydrothermal	Ni _{0.85} Se nanosheets on N-MoSe ₂	4 M KOH	58.4 F g ⁻¹ at 500 mA g ⁻¹	105.1% (15 000)	110
19	NiSe@MoSe ₂	Hydrothermal	Polyhedral structure of composite	1 M KOH	1000 F g ⁻¹ at 1 A g ⁻¹	77.67% (5000)	111
20	MoSe ₂ -Ni	Hydrothermal	Nanosheet arrays	2 M KOH	223 F g ⁻¹ at 1 A g ⁻¹	93.7% (1000)	79
21	MoSe ₂ /G	Solvothermal	MoSe ₂ nanosheets on Ni foam	6 M KOH	1114 F g ⁻¹ at 1 A g ⁻¹	104.7% (1500)	54
22	MoSe ₂	Sonochemical exfoliation	MoSe ₂ nanosheets	6 M KOH	945 F g ⁻¹ at 1 A g ⁻¹	92% (3000)	80
23	MoSe ₂ -graphene/Ni	Hydrothermal	Graphene nanosheets covered on surface and interspace of MoSe ₂ bars	6 M KOH	576 F g ⁻¹ at 1 A g ⁻¹	84% (3000)	112
24	2H-MoSe ₂ /MoSe ₂	Hydrothermal method	MoSe ₂ nanosheets	0.5 M TEABF ₄ /AN	1422 F g ⁻¹ at 1.5 A g ⁻¹	100.7% (1500)	113
25	MoSe ₂ /MoSe ₂	Epitaxial growth process	MoSe ₂ thin flakes and MoSe ₂ nanosheets	0.5 M H ₂ SO ₄	16.25 F g ⁻¹ at 0.75 A g ⁻¹	51.26% (10 000)	102
26	NiMoO ₄ /NiSe ₂ /MoSe ₂	Hydrothermal	Nanoflower MoSe ₂	6 M KOH	1229.6 F g ⁻¹ at 1 A g ⁻¹	92.8% (2000)	114
27	NiSe/MoSe ₂ /MoO ₂	Hydrothermal method followed by selenylation	Nanowires	2 M KOH	154 F g ⁻¹ at 10 mV s ⁻¹	16.6% (1000)	71
28	MoSe ₂ -Mo ₂ C	Two-step growth annealing strategy	3D hierarchical hollow microspheres with 2D thin nanoflakes	3 M KOH	955 F g ⁻¹ at 1 A g ⁻¹	86.1% (5000)	115
29	NiCoP@MoSe ₂	One-pot chemical synthesis	MoSe ₂ -Mo ₂ C hybrid nanoarrays	6 M KOH	1061 F g ⁻¹ at 2 A g ⁻¹	93.9% (10 000)	72



Table 1 (Contd.)

Sl. no	Electrode	Synthesis	Morphology/structure	Electrolyte	Specific capacitance	Retention (%) after (cycles)	Ref.
30	MoSe ₂ /Bi ₂ Se ₃	Hydrothermal, phosphorization, chemical bath deposition	NiCoP nanowires on flexible CC decorated with MoSe ₂ nanosheets	3 M KOH	1451.8 F g ⁻¹ at 1 A g ⁻¹	—	95
31	MoSe ₂ -Ni(OH) ₂	Hot injection in colloidal system	Bi ₂ Se ₃ hexagonal nanoplates enclosed by MoSe ₂ nanosheets	6 M KOH	1175 F g ⁻¹ at 1 A g ⁻¹	90% (3000)	75
32	Co(OH) ₂ -MoSe ₂	Hydrothermal	MoSe ₂ nanoflakes on Ni(OH) ₂ nanosheets	6 M KOH	541.55 F g ⁻¹ at 1 A g ⁻¹	91.4% (3000)	116
33	MnSe ₂ @MoSe ₂	Hydrothermal	Flowerlike morphology with nanospikes	1 M KOH	719 F g ⁻¹ at 1 A g ⁻¹	99.78% (5000)	117
34	CuSe ₂ @MoSe ₂	Hydrothermal	Nanorods	1 M KOH	509 F g ⁻¹ at 1 A g ⁻¹	99.82% (2000)	97
35	MoSe ₂ /Ti ₃ C ₂ //MoSe ₂ /Ti ₃ C ₂	Ultrasonic reduction process	Nanosheets	6 M KOH	350 F g ⁻¹ at 1 A g ⁻¹	89% (5000)	118
36	Ti ₃ C ₂ T _x /MoSe ₂	Hydrothermal	MoSe ₂ nanosheets incorporated in organ like MXene	6 M KOH	Pure MXene 107 F g ⁻¹ 1358.5 F g ⁻¹ at 1 A g ⁻¹	96.3% (10 000)	73
37	Ti ₃ C ₂ T _x /MoSe ₂	Hydrothermal	MXene flakes with MoSe ₂ nanosheets	3 M KOH	Pure MoSe ₂ 780.7 F g ⁻¹ Pure MXene 169.8 F g ⁻¹	96.3% (10 000)	83
38	MoSe ₂ @CN//CN	Hydrothermal	CN decorated on MoSe ₂ nanotubes	1 M LiClO ₄	101.3 mF cm ⁻² at 5 mV s ⁻¹	—	119
39	3D-CoNi ₂ S ₄ -G-MoSe ₂	Liquid phase exfoliation, hydrothermal	CoNi ₂ S ₄ nanoparticles on G and MoSe ₂ nanosheets	6 M KOH	1141 F g ⁻¹ at 1 A g ⁻¹	108% (2000)	68
40	FeNi ₂ S ₄ -G-MoSe ₂	<i>In situ</i> hydrothermal, liquid phase exfoliation, ultrasonication	FeNi ₂ S ₄ nanoparticles on G-MoSe ₂ nanosheets	6 M KOH	1700 F g ⁻¹ at 2 A g ⁻¹	106% (4000)	120
41	Na ion intercalated 1T-2H MoSe ₂ -G	Hydrothermal	Nanoflowers of MoSe ₂ -G composites	1 M PVA/KOH	143.6 mA hg ⁻¹ at 0.5 A g ⁻¹	—	74
42	MoSe ₂ -VACNTF/NF	Chemical vapor deposition combined with solvothermal technique	MoSe ₂ nanoflakes	2 M KOH	435 F g ⁻¹ at 1 A g ⁻¹	92% (5000)	121
43	MoSe ₂ -G	Hydrothermal and thermal annealing	MoSe ₂ nanosheets	1 M H ₂ SO ₄	148.6 F cm ⁻³ at 0.2 A cm ⁻³	—	122
44	W-MoSe ₂ /G	Hydrothermal method	W Doped MoSe ₂ nanoflowers grown on top of graphene sheets	0.5 M H ₂ SO ₄	248 F g ⁻¹ at 2 mV s ⁻¹	102% (20 000)	48
45	MoSe ₂ /acetylene black	Hydrothermal	Net like MoSe ₂ /AB composites with MoSe ₂ nanosheets	6 M KOH	2020 F g ⁻¹ at 1 A g ⁻¹	107.5% (1500)	81
46	MoSe ₂ /FeOOH	Hydrothermal	MoSe ₂ nanoflowers on FeOOH nanorods	6 M KOH	132 F g ⁻¹ at 1 A g ⁻¹	100% (3000)	103
47	MoSe ₂ /PANI	Hydrothermal	MoSe ₂ hollow microspheres and PANI rods	1 M KOH	146.5 F g ⁻¹ at 0.3 A g ⁻¹	54.5% (1000)	84
48	MoSe ₂ /PANI//AC	Hydrothermal	MoSe ₂ nanospheres on PANI nanosheets	3 M KOH	85.7 F g ⁻¹ at 1 A g ⁻¹	59% (1000)	86
49	MoSe ₂ /PANI	Hydrothermal, in-situ polymerization	Nanosheets and nanofibers	—	463 F g ⁻¹ at 5 mV s ⁻¹	72% (3000)	85
50	CoB//gCN/MoSe ₂	Hydrothermal	MoSe ₂ nanosheets in gCN sheets	1 M KOH	356.4 F g ⁻¹ at 1 A g ⁻¹	93% (10 000)	123
51	60-MoSe ₂ /(Ni, Co)Se ₂	Synthetic encapsulation	Hollow nanospherical structure	KOH	359.9 mA h g ⁻¹ at 1 A g ⁻¹	83.7% (10 000)	124



Table 1 (Contd.)

Sl. no	Electrode	Synthesis	Morphology/structure	Electrolyte	Specific capacitance	Retention (%) after (cycles)	Ref.
52	YSe ₂ /MoSe ₂ /Ni _x B	Hydrothermal	Lamellar structure of MoSe ₂ layers	3 M KOH	893.3 F g ⁻¹ at 1 A g ⁻¹	128.17% (5000)	87
53	6.2% Mn doped MoSe ₂	Hydrothermal	Mn doped MoSe ₂ nanosheets	3 M KOH	1116 F g ⁻¹ at 5 mV s ⁻¹	74%	88
54	(MoSe ₂)AC//6KOH//AC	Hydrothermal	MoSe ₂ nanoflowers coated with AC	6 M KOH	836 F g ⁻¹ at 10 mV s ⁻¹	76% (3000)	89
55	MoSe ₂ -Pt MoSe ₂ -Ag MoSe ₂ -Au	Hydrothermal	MoSe ₂ nanosheets	1 M H ₂ SO ₄	1178 F g ⁻¹ at 1 A g ⁻¹ 1255 F g ⁻¹ at 1 A g ⁻¹	93% (500) 88% (500)	90
56	MoSe ₂ /NC	Hydrothermal	MoSe ₂ nanosheets into a 3D N doped porous carbon skeleton	1 M KFSI in combination with EC and DEC	1338 F g ⁻¹ at 1 A g ⁻¹ 415.1 mA h g ⁻¹ at 100 mA g ⁻¹	95% (500) 71.3% (7000)	91
57	(2H-1T)MoSe ₂ -NiSe//NF//AC	Hydrothermal	Thin nanosheets	2 M KOH	150.9 F g ⁻¹ at 1 A g ⁻¹	92.8% (50 000)	93
58	MoSe ₂ NS-Ti	Hydrothermal	Nanosheets	2 M Na ₂ SO ₄	525.5 F g ⁻¹ at 1 A g ⁻¹	100% (1700)	125
59	W doped MoSe ₂ /graphene symmetric set-up	Hydrothermal	Nanoballs and nanoflowers	PVA-KOH	444.4 mF cm ⁻² at 1 mV s ⁻¹	81.3% (5000)	99
60	MoSe ₂ /MWCNT-2	Hydrothermal	Ultrathin MoSe ₂ nanosheets grown on MWCNTs	3 M KOH	927.25 F g ⁻¹ at 1 A g ⁻¹	70.08% (3000)	126
61	MoSe ₂ /MWCNT	CBD	2D cryptomelane like MoSe ₂ growth over MWCNTs	2 M KOH	232 F g ⁻¹ at 1.4 A g ⁻¹	93% (1000)	76

improved efficiency. Very large datasets are used in ML-based methods, often generated through prolonged experimental research or existing literature, which train models for predicting quantities such as specific capacitance, efficiency, cyclic stability, charge-discharge times, and capacity retention.¹²⁷⁻¹³¹ It typically involves aggregating a plentiful amount of research data and then feeding it into sophisticated algorithms that analyze the information to generate insights relevant to the research objectives. Then, these ML models are used for projecting the performance of electrode materials and estimating parameters such as capacitance, remaining useful life, lifetime, and state of health. Moreover, ML tools allow for investigating complex correlations and interdependencies of various factors influencing electrode properties, facilitating an enhanced understanding of material behavior under different conditions.¹³²⁻¹⁴² ML applied in this context fundamentally quickens the identification of optimal electrode compositions, reducing thermal energy storage time and resources usually spent on experimental trial and error. Synthesis conditions also include the selection of an electrolyte and voltage window specifications that can, additionally, be fed into the machine learning software.¹⁴³⁻¹⁴⁹ Machine learning workflow diagram for supercapacitors is shown in Fig. 21.

If blueprinting performance and accurately predicting enhancement strategies of MoSe₂ nanostructures for supercapacitor applications are feasible, machine learning algorithms provide an extraordinarily robust toolset for doing so. Not only algorithms like multiclass classifiers or random forest but also support vector machines contained in a class may classify nanostructures by either morphology or electrochemical properties. Following the modeling of synthesis parameters and performance metrics, regression techniques are then applied for means such as linear and support vector regression to understand how changes in production can lead to better results. Long-term stability in MoSe₂ electrodes was predicted with time series models such as RNNs and LSTM networks. Deep learning methods within multi-layer perceptron and deep belief networks will allow uncovering complex nonlinear dependencies between variables. Techniques like random subspace and random committee further increase predictive accuracy and robustness, thereby rendering these algorithms indispensable in stepping up research and development in supercapacitor technologies. Many research articles have used machine learning approaches that predict the capacitance of materials. The remaining useful life of SCs were predicted with the aid of machine learning techniques.¹⁵⁰⁻¹⁵⁴ Some hybrid machine learning models have also been formulated by integrating two or more different algorithms to enhance accurate prediction; it becomes quite useful during material science and supercapacitor research. Performance assessments for these machine learning models is typically considered by different metrics as follows: Root Mean Square Error (RMSE), Mean Absolute Percentage Error (MAPE), Correlation coefficient (R^2), Mean Absolute Error (MAE), Accuracy score and error rate. These give a sense of accuracy and effectiveness of the models within their applicability domains. This

ensures that the most suitable electrode materials for any supercapacitor application are found.

Using a machine learning technique, Mishra and colleagues investigated how the physicochemical features of carbon electrodes affected the performance of SCs. The ML models were trained and tested using extracted published experimental datasets having 4899 data entries to ascertain the relative significance of electrode material properties on specific capacitance. These characteristics include the carbon-based electrode material's oxygen and nitrogen content, potential window, specific surface area, pore volume and size, presence of defects and current density. Furthermore, testing procedure, electrolyte and electrode carbon structure are taken as categorical variables. Four ML techniques SVR, DT, RF, XGBoost and one Ordinary Least Square Regression (OLS) method were used to perform the regression of the target capacitance from the input

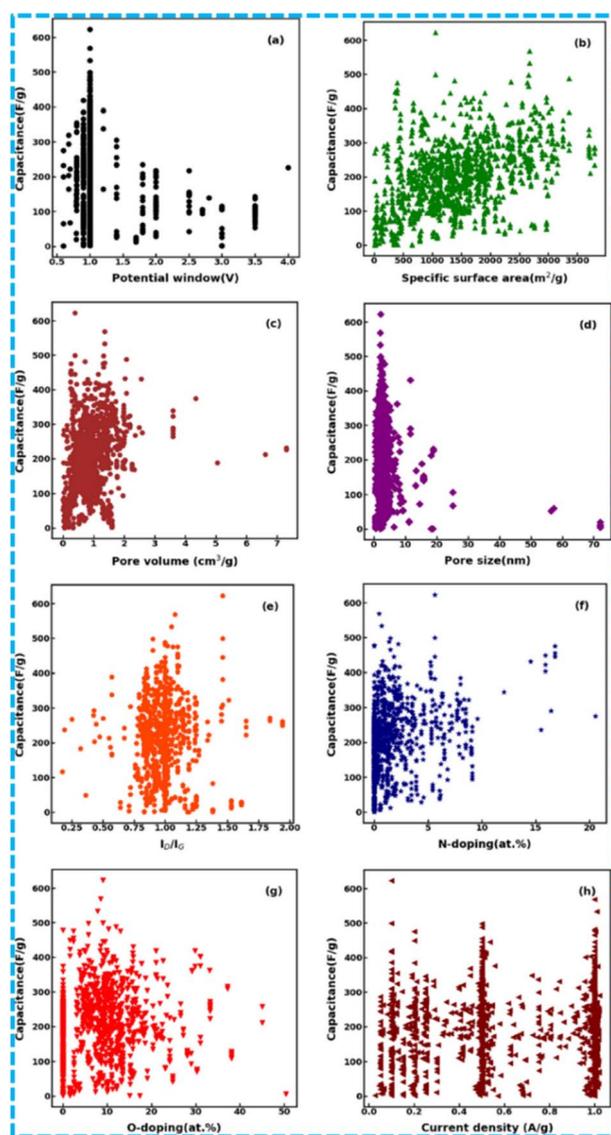


Fig. 22 (a–h) Illustration of the relationship between various input features and specific capacitance. This figure has been reproduced from ref. 155 with permission from Nature, copyright 2023.

features of the SCs. MAPE, bias factor (b'), R^2 , and RMSE were used as performance metrics. The best RMSE and R^2 values were displayed by the XGBoost model. The XGBoost model was reported to be best to correlate the input features with the capacitive performance. This indicates that the most important descriptors among the features chosen for the specific capacitance are potential window, specific surface area and the presence of N doping.¹⁵⁵ Fig. 22 shows the relationship between specific capacitance and various input features for the model. The comparison between actual capacitance and predicted capacitance for various models is shown in Fig. 23.

A study on a data-driven ML method for forecasting the capacitance of graphene-based SC electrodes was presented by Saad *et al.* Data from over 200 published papers has been obtained and analysed using several ML algorithms. The atomic percentages of C, N and O, I_D/I_G ratio, electrode configuration, pore-volume and size, and specific surface area were among the physicochemical characteristics used in this study. Cell configuration, potential window, ionic conductivity, current density, concentration of electrolyte, equivalent series resistance and charge-transfer resistance were among the electrochemical test characteristics derived from EIS analyses and

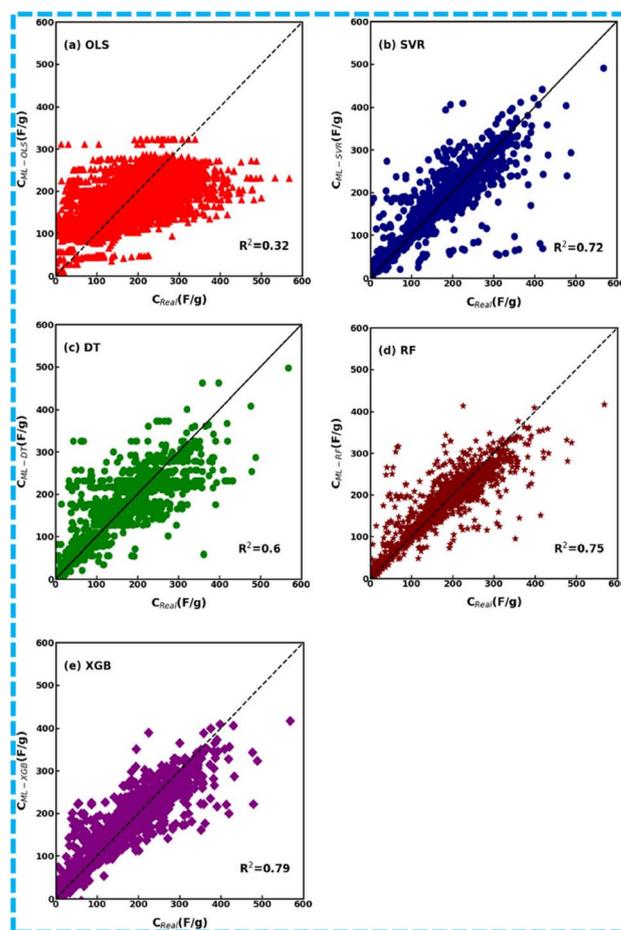


Fig. 23 (a–e) Comparison between predicted specific capacitance and actual reported specific capacitance from various models. This figure has been reproduced from ref. 155 with permission from Nature, copyright 2023.



GCD tests for the same purpose. ANN, k-nearest neighbor regression, decision trees and Bayesian ridge regression were the models used here by the authors. Comparing the constructed ANN model to other models created for this purpose, the former produced incredibly accurate prediction results, with RMSE and R^2 values of 60.42 and 0.88, respectively.²⁸ Deebansok and team performed analysis of various CV and GCD images using a Convolution Neural Network (CNN) model based on image classification. The authors applied supervised ML for image classification to perform electrochemical shape analysis (over 5500 CV plots and 2900 GCD plots). Schematic illustration of the model is shown in Fig. 24. The authors developed this method to address the problem of using simple binary classification to identify which electrode type (pseudocapacitor *vs.* battery) the materials belong to. The CVs and GCDs were analyzed *via* the ML model, trained with datasets obtained from a number of scientific papers. Various CNN architectures are validated and selected based on the evaluations, by applying the theoretical CV and GCD curves. The training of model is based on the classification of the signal, such as the peak *vs.* box shape for CV, and the plateau *vs.* triangular shape for GCD. The study described here has effectively addressed the issue of interpreting signals from CVs and GCDs by utilizing machine learning to categorize materials behaviours as either battery-like or pseudocapacitor-like. Using the supervised machine learning model, the authors also examined many scientific articles that contained the keywords “pseudocapacitor” (1346 articles) or “battery” (2011 articles). This allowed them to statistically analyze the number of articles that contained the keyword that contradicted their signals. In summary, the articles were chosen at random, their pertinent GCD and CV signals were taken out, and they were then simply categorized as either battery or pseudocapacitive. The authors found that 67% of the reports that contain “pseudocapacitor” as keyword align with

their experimental findings. Nevertheless, conflicting signals were found in almost half of the publications that contained “battery” as keyword. These findings support the idea that ML could significantly outperform human-based interpretation.¹⁵⁶

Wang *et al.* used an ANN model to identify oxygen-rich extremely active porous carbon electrodes for aqueous SCs with the aid of machine learning. The information gathered from several literature was used to train the ANN. Surface areas of micropores and mesopores are the structural parameters employed in ANN, whereas the overall percentage of N and O doping is employed as a chemical feature. The SC performance of N/O co-doped activated carbon-based electrodes is gathered in the training database for both 6 M KOH and 1 M H₂SO₄ electrolytes. The electrolyte type is handled as a dummy variable in the ANN model. There are 288 data points in the dataset. Training, validation, and test datasets make up 70%, 15%, and 15% of the total, respectively. According to ANN's prediction, the highest capacitance of a N/O co-doped activated carbon electrode in 1 M H₂SO₄ can be attained with a micropore surface area of 1502 m² g⁻¹, a mesopore surface area of 687 m² g⁻¹, nitrogen doping of 0.5 at%, and oxygen doping of 20 at%. The ANN prediction states that excessive oxygen doping in 1 M H₂SO₄ would provide a notable rise in specific capacitance because it would improve the electrode surface wetting and increase the electronic conductivity.¹⁵⁷ Various ML models are utilized for SC applications such as CNN, SVM, RNN, Regression models, XGBoost, Random Forests, Decision trees, and KNN. The choice of model for specific application depends on the type of data, number of datasets, and type of task (analysis, comparison, prediction, and classification). Each model has its own benefits and disadvantages. If possible, the researcher can apply the data to all models and compare their performances based on accuracy, error and other metrics. The model which shows good performance in metrics can be selected

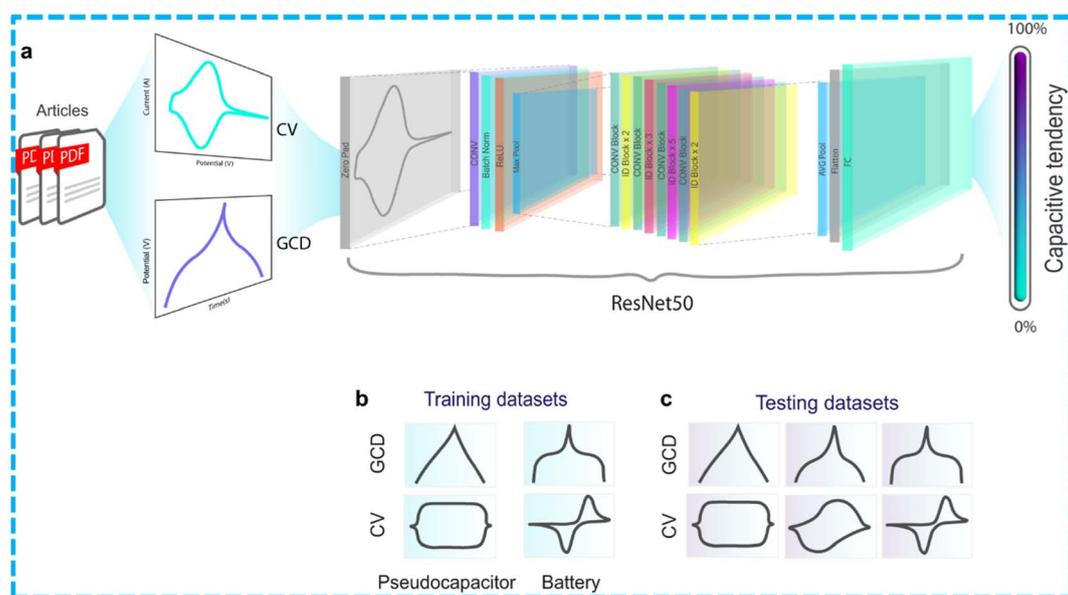


Fig. 24 (a) CV and GCD image extraction and classification from reported papers (b and c) representative of testing and training datasets. This figure has been reproduced from ref. 156 with permission from Nature, copyright 2024.



accordingly. However, this approach is suitable for small sets of data. Several researchers have selected neural network models since they can handle large and complex datasets. Other models are also beneficial depending upon the requirements of the purpose.

5. Challenges and future perspectives

(1) Using the formula $E = 0.5CV^2$, energy density was calculated for a SC electrode. From this, the energy density can be improved either by expanding the voltage window or by increasing the capacitance. The capacitance of MoSe₂ electrodes can be enhanced by creating highly porous structures and making composites or heterostructures with advanced capacitive materials. The voltage window can be expanded by properly selecting suitable electrolytes.

(2) Due to repeated charging/discharging and volume expansion, the MoSe₂ electrodes undergo structural collapse and damage. This affects their capacitance performances to a great extent. New strategies should be identified and adopted to prepare highly efficient MoSe₂ electrodes.

(3) The intrinsic electrical conductivity of MoSe₂ is very less, therefore it is not suitable for practical applications. The electrical conductivity can be improved *via* doping mechanisms, better morphology structures, adopting conductive substrates, nanosheet fabrication and composite formation with highly conductive materials.

(4) Slow ion transport hinders the overall performance of MoSe₂-based electrodes. This can be tackled by selecting proper electrolytes, developing microporous electrodes which can facilitate fast ion diffusion processes.

(5) Researchers have found that the 1T phase of MoSe₂ is metallic in nature, possessing very interesting properties. However, it suffers from structural and thermodynamic instability, and hence, not suitable for practical applications. Only few reports are available for 1T MoSe₂ and 1T/2H mixed MoSe₂-based supercapacitor electrodes. Numerous research studies need to be devoted to the development of stable 1T-phase MoSe₂. Phase engineering can be a modification strategy for MoSe₂ materials, because it can help to gain high intrinsic conductivity, promoting electron transfer and decreasing internal resistance, which are necessary for SCs. Many researchers have reported several methods to obtain phase change from 2H MoSe₂ to 1T MoSe₂ and enrich the energy storage capabilities.²⁴

(6) Interfacial engineering which usually modifies the surface of MoSe₂ with functional materials to enhance or modify its inherent characteristics can be explored for SC applications. There are four uses for interfacial engineering: increasing catalytic performance, modifying the bandgap of MoSe₂, functionalizing the MoSe₂ interface, and promoting electron transfer. Conductive carbon materials and metal compounds are usually employed as backbones in this type of modifications. Interfacial engineering can be made to modify MoSe₂-based materials for the fabrication of SCs because it can create various pathways for electron transmission and distribution of MoSe₂ nanosheets.²⁴

(7) Depending on the doped components, doping engineering can give pure MoSe₂ a variety of advantageous features. To create materials with the best performance, doping techniques can be employed in addition to single-heteroatom doping. To fully realize the potential of the doped heteroatoms, more research should be done to identify the ideal experimental settings for various dopants. The number of active sites can be revealed on the surface of pure materials by introducing defects and vacancies, and the disorganized configuration of active sites encourages reactant adsorption and activation. One effective method for improving the poor conductivity of MoSe₂ is vacancy engineering, and the degree of improvement is directly correlated with the quantity of Se vacancies. Thus, additional research ought to be carried out to investigate the possibilities of vacancy engineering and broaden its uses.²⁴

(8) Inherent drawbacks of MoSe₂ in SC applications can be mitigated by structural engineering, which can change their physical characteristics. The structural changes employed to customize the redox activity and speed up electron transmission in the MoSe₂ interlayers are anticipated to have considerable practicality and application potentials since MoSe₂ has a layered structure with a wide interlayer spacing. This can be an effective method for expanding active sites and speeding up electron transport.²⁴

(9) Recently, a new 2D material borophene has shown good promise to be used in energy storage devices. Still now, no reports are available on MoSe₂/borophene composites. This can be done in future to examine their energy storage abilities for supercapacitor applications.

(10) Integrating MoSe₂ SCs with other devices such as sodium-ion batteries and lithium-ion batteries and other types of batteries can leverage the advantages of both batteries and

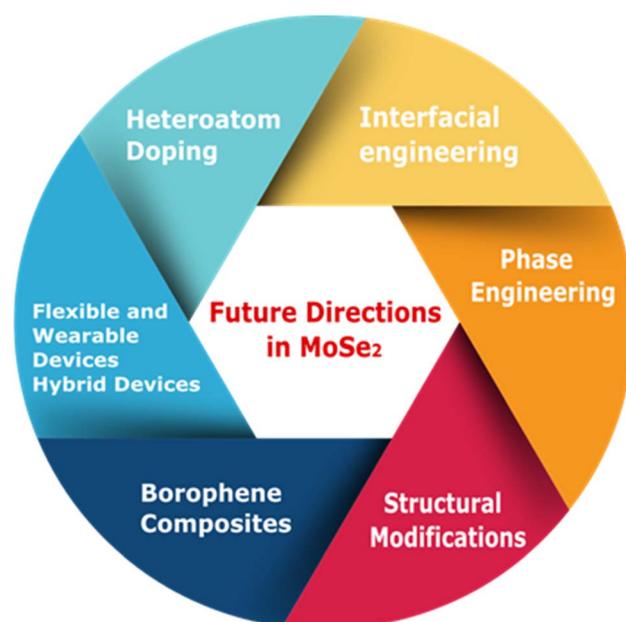


Fig. 25 Future Directions for MoSe₂ Supercapacitors.



supercapacitors, offering high energy densities and power densities.

(11) MoSe₂ SCs can find applications in various flexible and wearable devices, which are in high demand in the future, provided that sufficient research attention is paid in this field. These devices require very flexible small efficient nano electrodes for their operation. MoSe₂ electrodes can be developed accordingly as per requirements. This can be studied and developed in the future (Fig. 25).

6. Summary

Developing optimum electrodes is crucial for obtaining high-performance supercapacitors. Various materials are tested for electrode preparations. MoSe₂-based electrodes attract considerable attention in 2D materials and in the area of supercapacitor applications. MoSe₂ can be a good substitute for MoS₂ and other 2D electrodes. MoSe₂ stands as a good pseudocapacitive material. In this review, we discussed various synthesis approaches utilized for MoSe₂ preparation for supercapacitor applications. Most researchers have used hydrothermal methods frequently. Various structures and morphologies are reported using different synthesis and operating conditions. Each method has its own benefits and shortcomings. A brief discussion about supercapacitor fundamentals was mentioned along with working and classifications. The performance of various MoSe₂ composites was analyzed for asymmetric and symmetric supercapacitors along with a comparison table. Further, machine learning methods for supercapacitors are briefly described for capacitance predictions, remaining useful life predictions, life-time calculations and forecasting useful details, emphasizing that the use of ML is inevitable in the coming days in the area of energy storage. Current challenges and prospects are also provided at the end.

Data availability

No primary research results, software or code have been included and no new data were generated or analyzed as part of this review.

Conflicts of interest

There are no conflicts to declare.

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